Radioactive Waste Management
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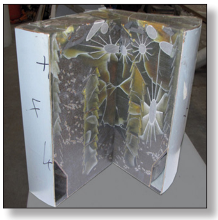
R&D and Innovation Needs for Decommissioning Nuclear Facilities



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Radioactive Waste Management

R&D and Innovation Needs for Decommissioning of Nuclear Facilities

NEA No. 7191

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The mission of the NEA is:

- to assist its member countries in maintaining and further developing, through international co-operation, the scientific, technological and legal bases required for a safe, environmentally friendly and economical use of nuclear energy for peaceful purposes;
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Foreword

The NEA Working Party on Decommissioning and Dismantling (WPDD) brings together senior representatives of national organisations who have a broad overview of decommissioning and dismantling issues through their work as regulators, implementers, research and development experts or policy makers. The WPDD reviews the current views of NEA member countries with the goal of strengthening the overall visibility of decommissioning, an activity that is attracting growing attention.

The current labour-intensive approach to decommissioning and dismantling suggests that research and development (R&D) aimed at more efficient, effective decommissioning could bring significant benefits to current and future R&D projects. However, there is little consensus on where R&D might be best directed. Part of the difficulty in evaluating and co-ordinating R&D efforts is the nature of the decommissioning process itself, which tends to be sporadic and isolated.

The WPDD established the Task Group on Future R&D and Innovation Needs for Decommissioning to provide a forum to discuss these issues among interested specialists from NEA member countries and to report on the outcomes.

This report provides an update on the challenges of current R&D and reports on the WPDD consensus concerning priorities for future R&D and opportunities for collaboration among organisations and NEA member countries.

Acknowledgements

The WPDD wishes to express its particular gratitude to Mr Harvey Farr (Radiation Safety & Control Services, Inc., United States) and Mr Thomas S. LaGuardia (LaGuardia & Associates, LLC, United States) for their important contributions to the drafting of this report.

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Executive summary

The current labour-intensive approach to decontamination and decommissioning (D&D) suggests that research and development (R&D) aimed at more efficient and effective decommissioning technologies and processes could bring significant benefits to current and future D&D projects. Improvements are required to decrease the hundreds of billions of dollars (USD) that will be required to decommission facilities that have shut down or will shut down around the world. This report discusses decommissioning activities and challenges that could benefit from R&D and summarises applicable emergent technologies and research efforts to build a base of knowledge regarding the status of decommissioning technology and R&D. This base knowledge can be used to obtain consensus on future R&D that is worth funding. It can also assist in deciding how to collaborate and optimise the limited pool of financial resources available among NEA member countries for nuclear decommissioning R&D.

This report discusses challenges, current guidance pertaining to the themes listed below, applicable current innovative technologies and R&D being conducted. Whenever possible, the source of the information and its URL are referenced to ease retrieval for more detailed evaluation. It is impossible to delve into the pros, cons and potential future uses and R&D for the wide array of topics and technologies examined in this report. Each theme is concluded with suggestions for future R&D and areas for international collaboration, with broad objectives and deliverables to help focus future R&D on the problems that need to be solved. Specific R&D, technologies or solutions are generally not offered in order to allow member countries and their representatives to make R&D funding decisions based on the unique needs and conditions of their individual countries.

The introduction discusses the backlog of closed facilities awaiting decommissioning and projected closures and decommissioning costs. The decommissioning alternatives, processes, phases and the challenges associated with these are also discussed. Five themes are then examined in this report.

Theme 1, *Characterisation and Survey Prior to Dismantling*, discusses the use of geostatistical software applications, imaging and remote sampling systems, as well as novel detection and sample analysis technologies to provide more accurate, rapid and cost-effective determination of contaminant concentrations and spatial distributions. Modelling of mobile nuclides and the need for an international approach or standard for estimating trace impurity levels in activated reactor alloys and concretes is also discussed.

Theme 2, *Technologies for Segmentation and Dismantling*, discusses experience with technologies commonly used for segmentation and dismantling tasks and evaluates the use of robotics, remote systems and innovative cutting technologies such as arc saws and lasers for system, structure, component (SSC) and reactor vessel and internals segmentation. Robotic and remotely operated technologies and end effectors are also discussed for material handling. The challenges associated with generation, capture and processing of secondary wastes associated with cutting technologies are also discussed in this theme.

Theme 3, *Decontamination and Remediation*, covers a wide array of subjects. Topics explored include achieving a better understanding of the chemical interactions and mobility of contaminants with substrates, and chemical and physical processes that can be used for decontamination of decommissioning materials such as concrete, metals, graphite, tank heels, groundwater and soil. Innovative technologies such as ligands that bind actinides, laser scabbling and decontamination, as well as cryogenic technologies, are also discussed. Prospects for expanded use of robotics and for more automated, modular decontamination processes for processing decommissioning materials and bulk soil and groundwater remediation are also touched upon, as are innovative developments in the use of nanotechnology, bio-remediation and fixatives.

Theme 4, *Materials and Waste Management*, addresses challenges and technologies as well as fundamental research to better understand the long-term interactions between waste, packaging and disposal environs. This includes difficult-to-manage waste forms such as mixed wastes, organic materials, transuranics, depleted uranium and high- and intermediate-level wastes. Optimisation of the waste hierarchy through improved automated handling, segregation, packaging and waste conditioning technologies is also addressed in this theme.

Theme 5, *Site Characterisation and Environmental Monitoring*, focuses on SAFSTOR (or SAFe STORage – a decommissioning method where a nuclear facility is monitored for a period of up to sixty years before complete decontamination and dismantling at the site), end state, and post-decommissioning challenges and technologies. Areas explored include three-dimensional modelling and non-intrusive sampling for characterisation, current and potential future use of robotics including rapidly developing autonomous robotic capabilities, current and potential future application of remote sensing, telemetry and satellite technologies for characterisation and monitoring. The current characterisation, fate and transport, exposure modelling software capabilities and shortcomings are discussed as well. This theme explores the R&D being undertaken to develop more integrated, flexible, modular codes that can better model complex sites with a variety of contaminant sources and hydrogeological conditions. Codes are written for laptops and desktop applications for less complicated models, expanded use of cloud computing capabilities for intermediate models and for super computers at government and research institutions for complicated models. The complex models include those with existing soil and groundwater contamination and multiple sources of end-state sources such as concrete subsurface structures or tanks that are contaminated in addition to the conventionally modelled surface soils and structures.

The report concludes with a focus on the historical reluctance of the nuclear and D&D community to invest in and incorporate new technologies in comparison to other industries such as manufacturing. It explores the obstacles that must be overcome to bring innovative solutions and technologies to bear on nuclear decommissioning. The necessity of breaking the expensive and time-consuming cycle of re-invention that plagues the current decommissioning effort and ideas on how to start and sustain a cycle of deployment and continuous improvement within the supply chain are also discussed. The industry has historically attempted to adapt technologies developed for other industries or military purposes rather than selecting promising fundamental innovations and investing in R&D further down the line to develop technologies specifically tailored for the nuclear industry and decommissioning. Finally, reference is made to the challenges of getting new technologies into the field of decommissioning projects and the risk-averse reluctance of decommissioning managers to adopt new technologies in their projects. Development of innovative technologies and processes will not decrease costs, particularly if they never make it to the field or if they are abandoned and re-invented after each decommissioning project. In short, the report documents the vast array of promising current and future technologies that could be deployed today or developed in the future to accomplish the goal of better, cheaper, faster and safer decommissioning. Targeted, collaborative R&D funding, as well as strategies and funding to deploy, test, improve and redeploy technologies on current large-scale decommissioning projects, will be necessary to achieve this goal.

1. Introduction

Background

The generally accepted purpose of decontamination and decommissioning (D&D) is to allow removal of some or all of the regulatory controls that apply to a nuclear site (buildings and grounds) while protecting the decommissioning workers and securing the long-term safety of the public and the environment. The core objectives of the D&D process are:

- protect and minimise the impact to workers, the public and the environment;
- remove and sentence facility materials to optimise waste minimisation, cost and schedule;
- achieve a property end state that releases the facility from all or some regulatory controls and supports sustainable development and re-use.

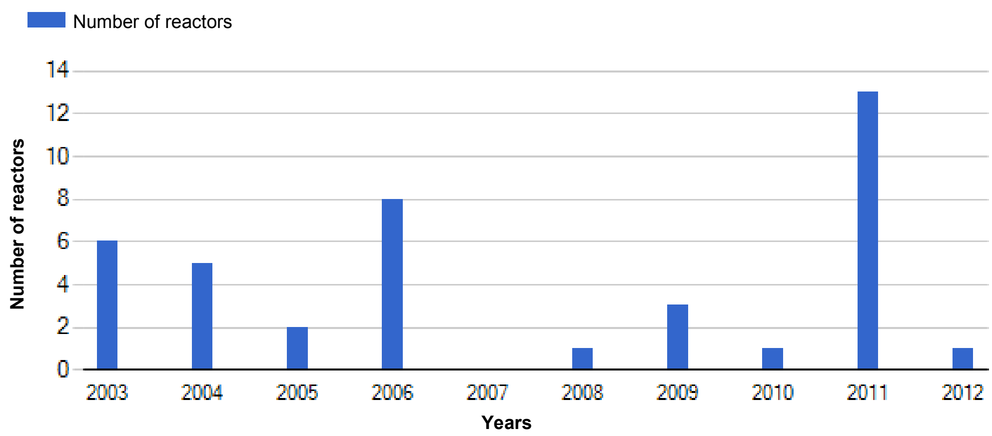
In order to achieve this, the facility systems, structures and components (SSC) must be characterised and sentenced to a final disposition. This may range from unrestricted release of SSC that are un-impacted by hazardous materials and can be left in place, to removal, treatment and stabilisation of high risk materials such as spent nuclear fuel. Different projects face different technical and social challenges, and while experience suggests that the overall purpose of D&D can be achieved, advances in D&D technology, strategies and infrastructure or support systems are required to achieve safer, better, cheaper and faster decommissioning. As stated in a National Academy of Science study:

> *Many current technologies are labor intensive and time consuming. Most current D&D technologies require hands-on contact by workers who must operate powerful equipment (e.g. plasma torches, saws, and lifting devices) while wearing bulky protective clothing. The facilities present hazards to workers that include penetrating irradiative areas, airborne contamination, toxic chemicals, and other industrial hazards*. (NRC, 2001)

The current labour-intensive approach to D&D suggests that research and development (R&D) aimed at more efficient and effective decommissioning could bring significant benefits to current and future D&D projects. There are many D&D activities that might be improved by R&D. The difficulty lies in the lack of consensus concerning where the R&D might be best directed (e.g. towards more effective/efficient remote cutting techniques or towards more efficient characterisation or decontamination). Part of the difficulty in evaluating and co-ordinating R&D efforts is the nature of decommissioning itself, which tend to be sporadic and isolated. The Working Party on Decommissioning and Dismantling (WPDD), operating under the umbrella of the OECD/NEA Radioactive Waste Management Committee (RWMC), established the Task Group on Future R&D and Innovation Needs for Decommissioning in early 2010 to provide a forum where these issues could be discussed among interested specialists from member countries and to report on the outcomes. Based on the very limited and slow adoption and integration of existing technologies such as robotics and telemetry into decommissioning projects, it is also likely that incentives will be necessary to get technologies developed through R&D funding into use in the field.

A substantial investment will be required to decommission existing facilities if progress is not made in developing and adopting innovative technologies and approaches to decommissioning as well as waste conditioning, storage and disposal. In 2011 the number of nuclear power plants in permanent shutdown increased significantly as a result of the Fukushima Daiichi nuclear power plant accident.

Figure 1.1: Number of nuclear power plants in permanent shutdown



Source: IAEA Power Reactor Information System website, accessed 20 February 2014, www.iaea.org/programmes/a2.

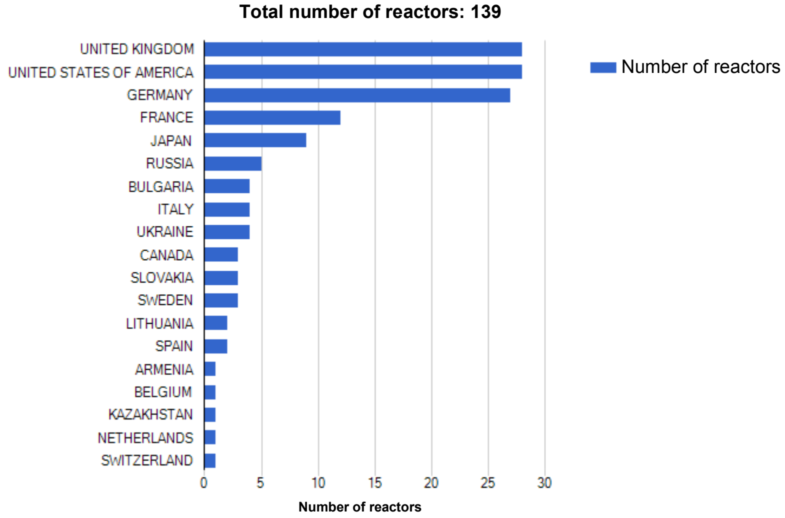
As of May 2012, 139 civilian nuclear power reactors had been shut down in 19 countries, including 28 in the United States, 28 in the United Kingdom, 27 in Germany, 12 in France, 9 in Japan and 5 in the Russian Federation. Three more facilities permanently shut down in the United States in 2013 (Kewaunee, SONGS 1, 2 and 3, and Crystal River). A fourth, Vermont Yankee, announced it will close in 2014. Decommissioning had only been completed for 17 of the world's permanently shut down reactors as of October 2010. Decommissioning is a complex process that takes years. The backlog of civilian nuclear power reactors that have been shut down but not yet decommissioned is expected to grow. There is also a large legacy of military and research reactors requiring decommissioning. The typical design life of a civilian nuclear power reactor is 30 to 40 years. There are currently 436 such reactors in operation worldwide, with a total installed electrical capacity of 370 499 billion watts (GWe). Of these 436 civilian nuclear power reactors, 138 are more than 30 years old and 24 are more than 40 years old. The average age of the civilian nuclear power reactors currently in operation is 27 years (UNEP, 2012).

The largest number of nuclear power plants either already closed or to be closed by 2025 is in Western Europe (>160), with those in North America much lower (>60), followed by Russia (>40). Of the research reactors, 241 are in operation, 165 shut down or under decommissioning and 431 have been fully decommissioned. In this case the number of those either already closed or scheduled to be closed by 2025 is greatest in North America (>100), followed by Western Europe and then Russia. With respect to fuel cycle facilities, on a global basis 198 are shut down or under decommissioning and 172 have been fully decommissioned. The forecast of future decommissioning by 2025 shows Western Europe again with the largest demand with 120 facilities (OECD/NEA, 2012c).

A great deal of capital is estimated to be required to decommission nuclear facilities in the coming decades in order to deal with the backlog of shutdown facilities that have not been decommissioned yet and those that will be shut down in the near future. The United States Department of Energy (US DOE) has nuclear liabilities on the order of USD 35 billion and has been spending around USD 6 billion per year on decommissioning. The current United States operating reactor fleet of 100 reactors represents a future liability of around USD 47 billion if the average decommissioning cost per reactor is maintained at USD 470 million. France has combined future liabilities from EDF, AREVA and the CEA estimated at around USD 80 billion. The Nuclear Decommissioning Authority

(NDA) in England has 2.2% discounted liabilities of USD 80 billion and is spending around USD 5 billion per year. The future liabilities for the United Kingdom operating EDF fleet are estimated at around USD 17 billion (OECD/NEA, 2012c). Thus a great deal of resources and capital will be spent in the coming decades to safely decommission nuclear facilities. It is therefore essential that technologies and innovations are continuously developed and implemented to accomplish this as safely, cost effectively and as expediently as possible.

Figure 1.2: Number of reactors in permanent shutdown as of May 2012



Source: IAEA Power Reactor Information System website, accessed 20 February 2014, www.iaea.org/programmes/a2.

In 2001, the Assistant Secretary for Environmental Management (EM) of the US DOE estimated that use of new technologies could save about half of the USD 30 billion that it estimated at the time as the cost for facility DOE D&D (NRC, 2001). Obviously with the number of permanently shut down facilities increasing dramatically since 2001, this potential cost savings has increased substantially.

This report presents the D&D priorities and examples of current research identified by the working group. In preparation of the report, examples of current R&D and emergent technologies both within and outside the nuclear industry were incorporated into the report. The opportunities to dramatically improve D&D performance and lower costs are numerous if an integrated, prioritised and co-ordinated international approach can be adopted.

Overall scope and objectives

The overall goal of this report is to define the aspects of decommissioning with greatest potential for future improvements through R&D. It is not intended to develop R&D solutions but to assist in reaching a consensus on which items future R&D work should be focused, including identifying potential projects that might best be addressed on a collaborative basis. This goal has two main underlying objectives:

1) to undertake an analysis of R&D needs for decommissioning and to assign broad priorities to these (first phase);
2) to define relevant R&D projects that might be undertaken on a collaborative or jointly funded basis (second phase).

To meet these objectives the report presents the challenges posed by decommissioning that underscore the necessity of R&D in these areas and a summary of current research and emerging technologies that underscore the present state of the art and offer potential solutions to the posed challenges. The examples of current R&D are provided to enable technically grounded and informed consideration of future R&D projects following issuance of this report. The report then makes suggestions for future R&D and provides broadly inclusive descriptions, objectives and desired deliverables.

Method of working

As mentioned above, the OECD/NEA Working Party on Decommissioning and Dismantling established the Task Group on Future R&D and Innovation Needs for Decommissioning in February 2010. The objective of this Task Group was to provide a forum where R&D issues could be discussed among interested specialists and the outcomes reported. It is important to mention that this Task Group is not responsible for the development of the technology; it is responsible for determining in which D&D areas international efforts should be invested. The Task Group's goal was to develop a report that defines where future development work should be focused. This focus includes the following:

- to undertake an analysis of R&D needs for decommissioning;
- to assign broad priorities to these;
- to define relevant R&D projects that might be undertaken in a collaborative or jointly-funded basis.

The Task Group subdivided the R&D tasks into five themes:

- characterisation and survey prior to dismantling;
- technologies for segmentation and dismantling;
- technologies for decontamination and remediation;
- materials and waste management;
- site characterisation and environmental monitoring.

The following membership comprised the Task Group: Gérard Laurent (Chair, EDF); Boby Abu-Eid (US NRC); Melanie Brownridge (NDA); Jean-Marc Idasiak (CEA); James Mac Kinney (NDA); Per Lidar (Studsvik Nuclear AB); Nieves Martin (ENRESA); Harald Maxeiner (NAGRA); Jean-Guy Nokhamzon (CEA); Angelo Paratore (SOGIN); Annika Rüdebusch (NAGRA); Andrew Szilagyi (US DOE).

The participating representatives completed a form to indicate their country's R&D needs for each of the five themes. Each theme's working group then used this input to target R&D challenges, identify examples of current research and prioritise future R&D.

Structure of the report

Chapter 1 of the report consists of the introduction and background information. Each subsequent chapter is written for each theme: Chapter 2 for Theme 1, Chapter 3 for Theme 2, and so on until Chapter 6. At the end of each chapter, suggested areas for future international collaboration for the theme being considered are presented. Chapter 7 provides the overall conclusions of the report.

This report provides the status and challenges of current R&D and reports the WPDD consensus on the priorities for future R&D and opportunities for collaboration among organisations and member countries.

Summary of common decommissioning practices

Applicable strategies, practices and guidance also vary widely due to the diverse nature of the facilities that require decommissioning and release from regulatory control. Decommissioning ranges from small facilities such as research facilities, laboratories, or manufacturing facilities with a history of limited use of radioactive and hazardous material and minimal or limited potential for impacted SSC, to large facilities with an extensive historical use of such materials and broadly impacted SSC and grounds such as large commercial nuclear reactors and national laboratories or fuel reprocessing facilities.

D&D strategies and practices are also driven by the marketplace, waste disposal options and regulatory requirements. As a result, decommissioning strategies and practices are not uniform among member countries. Although waste minimisation and material recycling and reuse are fundamental objectives of most decommissioning, the extent to which they are implemented is often driven by availability of waste disposal options, cost and schedule. When waste disposal options for all but the very high-level waste associated with spent fuel and highly irradiated reactor internals were widely available in the United States, the most cost-effective approach to decommissioning was widely viewed to be the "rip and ship" mentality that focused on expedited removal, packaging and transport of SSC to licensed disposal facilities.

The availability and cost of disposal options set a very low bar for the level of effort that was cost effective for segregation, treatment and processing of SSC materials for optimisation of the waste hierarchy. Thus the technology and strategies employed were not optimised to promote very low-level waste (VLLW) segregation or re-use and recycling of materials. In other countries, such as those in Europe and East Asia with more limited waste disposal options and higher disposal costs, decommissioning experience and technology have placed far greater emphasis on the optimisation of the waste hierarchy. Strategies and technologies used for D&D are continuously shaped by the marketplace. For example, the closure of the Barnwell facility to most of the US facilities left no disposal option for all but the lowest-level radioactive wastes. This required on-site or off-site temporary storage of higher-level wastes that were routinely disposed of at Barnwell and significantly increased the long-term costs associated with such wastes. In response, decommissioning made greater efforts to minimise the generation of the higher-level Class B and C wastes in the United States. The opening of the Waste Control Specialists facility in Texas and the willingness of the compact commissioners to accept waste from non-compact states has changed the market dynamic once again by offering a viable solution for disposal of B and C waste.

Another significant factor that has stifled the adoption of new technologies and development of D&D strategies and technologies is that decommissioning tends to be sporadic and isolated. This lack of sustainability and co-ordination limits the potential "payback" for capital expenditures required to incorporate and develop new technologies and practices. The cost effectiveness of such efforts to date have tended to be evaluated on a project-by-project basis, rather than spreading the investment in a new technology over multiple projects. The "stop and go" nature of decommissioning also hinders the transfer of knowledge and technology among projects.

Despite these variations, there are certain features that D&D efforts tend to have in common. The IAEA DeSa and FaSa projects have focused on harmonising safety cases for decommissioning facilities (IAEA, 2012a, 2013d). The R^2D^2 project (IAEA, 2013e) has implemented a new approach to enhance the exchange of information and the lessons learned between countries that have actual decommissioning experience for research reactors and those whose decommissioning programmes need to be developed. The approach is to hold workshops that provide "hands-on" experience to participants in an international demonstration project. Features that all decommissioning has in common include characterisation and survey, segmentation and dismantling, remediation and decontamination, material and waste management, site de-licensing or clearance, and environmental monitoring.

Characterisation and survey

Characterisation of systems, structures and components (SSC), as well as potentially impacted land areas of the property, must be conducted for all decommissioning facilities regardless of their size and use. The characterisation process' objectives are to provide physical, radiological and hazardous materials inventories to support decommissioning planning and execution. Characterisations occur throughout the decommissioning process and are refined to provide increasing detail and information to support the project as it progresses. Characterisation of contaminated materials is necessary at several stages of D&D. Substantial cost savings may result from basic research toward developing the means, preferably real-time, minimally invasive and field usable, to locate, identify and quantify contaminants. This includes difficult to measure contaminants on SSC, soils and groundwater. It also include demolition debris such as concrete or soils where the dismantlement and remediation method can change the contaminant concentration of the final form material and influence the final disposition of the material (e.g. clearance, recycling or waste) (NRC, 2001).

Segmentation and dismantling

The removal of plant systems, structures and components is also a phased or graded approach. The National Academy of Science (NRC, 2011) described the phases as follows:

- an assessment and decision-making phase;
- a planning phase for the development of the D&D plan;
- the physical decontamination and decommissioning operations phase.

Most often, high risk items such as spent nuclear fuel are targeted early in the decommissioning process in order to achieve a safer configuration. The majority of the dismantling process at nearly all facilities involves demolition and removal of metallic and concrete SSC. Thus, improvements in the demolition and handling technologies for these two classes of materials will benefit decommissioning across the board.

Decontamination and remediation

Most decommissioning requires some degree of decontamination and remediation. Decontamination is often used for industrial and radiological safety purposes in the initial phases of decommissioning and periodically thereafter to reduce worker exposures, risk and the degree of difficulty in executing decommissioning activities (e.g. chemical decontamination of systems, asbestos abatement, sodium passivation, acid/caustic neutralisation). Remediation involves removal of contaminants to lower levels in order to minimise waste disposal costs and to meet facility clearance criteria. For the most part remediation technologies apply to the metal and concrete removed, and to a third, potentially large, contributor to decommission waste volumes – contaminated soils. In regions such as Europe that have constraints on developing suitable shallow land disposal facilities, the decontamination, release, recycling and re-use of materials is extremely important to minimising decommissioning costs. In other areas of the world, where more suitable environmental siting conditions exist for disposal facilities (e.g. low rainfall, low permeability soils and significant groundwater aquitards) limited decontamination with an emphasis on removal and disposal may provide a more cost-effective decommissioning model.

Materials and waste management

All materials at the facility must to some extent be characterised and sentenced. The sentencing process involves determination of the most cost-effective ultimate disposition of the material. This may be to leave the material on site and verify that it meets site clearance criteria. It may involve targeting it for asset recovery to be sold and used at another facility or to be cleared and sold for recycling. It can also mean determining the probable waste classification very low-level radioactive waste (VLLRW), low-level radioactive waste (LLRW), intermediate-level radioactive waste (ILRW) or high-level radioactive waste (HLRW) and methods to optimise disposal options for the materials. Many materials, such as activated graphite, reactor internals and high activity sludges or organic materials such as resins, pose significant challenges to handle, stabilise and package in manners that are suitable for interim storage and long-term disposal, and therefore require significant investments in the development and study of geological repository disposals. More cost-effective material characterisation, segregation, assaying, stabilisation, packaging and disposal options will benefit nearly all decommissioning facilities in the future.

Site characterisation and environmental monitoring

The “end state” of a facility undergoing D&D is a major determinant of the cost, schedule and risk (NRC, 2001). Acceptable contaminant concentrations clearance levels or acceptable future occupant risk levels are typically defined by applicable regulations. However, overall end states are also driven by modelled risks, implementation of the ALARP or the “How clean is clean enough?” principle. In addition, choosing an end state that is compatible with sustainable economic development often requires negotiation by facility owners working with regional and national regulators as well as local stakeholders. Some decommissioning scenarios require long-term monitoring of waste storage facilities, site environmental contaminants or conditions at partially decommissioned facilities in care and maintenance (e.g. SAFSTOR). Facilities whose decommissioning requires long-term monitoring may benefit greatly from emerging telemetry and monitoring technologies. As was mentioned in the discussion on characterisation and survey, better characterisation technologies for SSC and site environs could benefit final status survey efforts which currently rely on direct sampling and laboratory analysis as well as labour intensive survey methods. In addition, end state planning generally requires modelling of future site occupants’ potential exposure to contaminants that remain upon completion of decommissioning. This involves fate and transport modelling of contaminants in the environment and estimates of potential (human and other species) exposures based upon the contaminant pathways. Once acceptable end state contaminant levels are determined to meet the site clearance criteria, surveys and sampling must be conducted and evaluated statistically to demonstrate that the clearance criteria have been met. Nearly all decommissioning facilities could benefit from advances in the ability to model complex sites, evaluate non-uniform and subsurface contaminant distributions, and more efficiently interface measurement data and modelling software.

These common D&D attributes provide a framework with which R&D efforts can be prioritised and targeted for collaboration so that the benefits of such efforts are broadly applicable across decommissioning facilities of all types.

Challenges for decommissioning and dismantling nuclear facilities

The generally agreed upon international objective of nuclear facility decommissioning is to release the land for all foreseeable future uses. There may be practical constraints to reaching this objective depending on the initial conditions, available decommissioning funds, the timing/schedule of the decommissioning, the current status of dismantlement/remediation technologies and waste treatment and disposal options. In some cases sites may require active management for many decades and will only be suitable for restricted re-use until there are solutions for economic and technical challenges at some point in the future.

Decommissioning plans are developed to achieve this outcome but they also need to take cognisance of the health and safety of workers, environmental protection and value for money. Generally, asset management costs of redundant facilities drive early D&D but in some cases potential decommissioning impacts on co-located facilities, technical D&D or waste disposal challenges, or health and safety considerations can introduce long periods of care and maintenance into plans.

Initial decommissioning planning is generally optimistic. Most decommissioning projects encounter schedule delays in one form or another due to unforeseen difficulties with D&D technology performance, risk/incident management, changes in regulatory requirements or waste disposal options. An objective of decommissioning is to maximise re-use of equipment and material, thereby reducing the data gaps in performance of D&D technologies, waste burden and the impact on the environment. Decommissioning approaches will be informed by environmental and health and safety risk analysis, dismantling and waste treatment techniques, site characterisation information, and waste lifecycle impact analyses.

In most countries decommissioning is complicated by legacies of stored historical radioactive wastes and past decisions to temporarily close facilities before they have been adequately prepared for decommissioning. In these cases the challenges can be summarised as follows:

1) retrieval and treatment of radioactive wastes that are often stored in ageing legacy facilities (includes contaminated land) – time critical programmes;
2) stabilisation of redundant nuclear facilities – removal of all the mobile radioactive waste and implementation of asset management designed to allow the facility to be "mothballed" with limited care and maintenance;
3) dismantling of legacy facilities – "construction in reverse" in some cases involving the decontamination and treatment of significant quantities of radioactive waste;
4) remediation and restoration of nuclear sites to allow re-use.

Each of these activities presents unique challenges that will be assisted by research and development within this report's five themes; the broad challenges associated with each theme are discussed herein.

Characterisation and survey

The challenge under this theme is to optimise the use of remote and *in situ* characterisation technologies to ensure more complete and cost-effective characterisation of the facility and legacy wastes are available for decommissioning planning. Another challenge is to increase the reliability and quality of characterisation data collection and measurement data analysis and interpretation. Characterisation of legacy wastes is a challenging activity at many decommissioning facilities because of access limitations and/or high radiation levels and transuranic alpha emitter content. Lessons learned from many major programmes indicate that high quality information on the legacy wastes is necessary to ensure that decommissioning plans have made the correct assumptions for waste retrieval and transfer methods, storage and waste treatment.

In some cases historical records can be relied on to provide information that informs planning for waste clean out as a precursor to dismantling, extended asset management, stabilisation or preparation for entombment. However these records are often missing, unreliable or require confirmation. The level of information required is often different than that needed for dismantling. A key extended management issue is the robustness of the ageing facilities and future waste packaging and stores. In some cases, wastes cannot be retrieved quickly and there is a need to develop better models and monitoring methods to predict the future performance of concrete structures and corrosion of vessels and pipe work. Development of better remote and *in situ* characterisation capabilities would greatly benefit the initial and long-term characterisation of such wastes.

It is important to focus waste management resources on wastes that represent the greatest environmental risk. Early characterisation and record keeping integrated as part of decommissioning planning will assist in decisions on sorting, segregation and decontamination techniques required to support the implementation of the waste management hierarchy. Characterisation plays a crucial role in reducing waste through sorting, separating, decontaminating, recycling and re-use of materials where practicable. Appropriate waste characterisation and understanding waste behaviour is essential to address important issues (e.g. long-term package performance) and to take advantage of alternative treatment and packaging solutions, decay storage, or novel decontamination techniques.

The level of detail and quality of the facility characterisation informs D&D and end state planning as well. Facility characterisation of SSC, working conditions and end state materials can be challenging using existing technologies due to access limitations related to hazardous conditions or physical attributes and operational considerations at the site. Limited on-site analytical capabilities, cost and schedule are also constraints on characterisation survey and sampling efforts. It is important to identify any critical characterisation data gaps to ensure planning includes efforts to close them and to ensure that working condition constraints and high risk evolutions are identified and addressed in decommissioning planning. Identification and closure of characterisation data gaps is also important to ensure the overall decommissioning and individual project plans are viable, and that contingencies are factored into the decommissioning plan where significant uncertainty exists. Technologies that improve remote and *in situ* sampling capabilities as well as those that improve the analysis and interpretation of characterisation data would help minimise and identify characterisation data gaps associated with decommissioning.

Segmentation and dismantling

The challenge under this theme is to adopt new technologies in order to improve efficiency and safety, and reduce cost. Technologies presently in use do not employ the latest cutting methods such as lasers and arc saws and are very labour intensive, requiring manual labour to perform the cutting and handling of the materials. Robotics and remotely operated equipment are rarely used except in under water or extremely hazardous situations. Use of labour intensive methods places humans in the line of fire and requires detailed planning, monitoring and oversight of activities to minimise the risk of accident and injury and to ensure industrial safety and radiological protection regulations are followed. There are opportunities to develop or use existing end effectors for segmentation and handling tasks that will remove personnel from hazardous operations. Segmentation of reactor internals and reactor vessels has not been perfected and projects utilising current technologies have been fraught with problems and delays despite attempts to document and implement lessons learned from previous projects.

The process of handling, segregating and loading materials that have been segmented is also still very labour intensive. Integrated, automated technologies for handling and packaging materials have not been developed or deployed as systems to optimise these tasks. Better end effectors that both cut and handle smaller components such as piping, cable trays and hangers need to be developed. In addition, end effectors with dual capabilities of demolishing concrete and cutting rebar could greatly expedite the demolition process. The challenge is to develop integrated systems that remove and process the materials being dismantled and to take advantage of developments in autonomous and semi-autonomous robotics capabilities.

Decontamination and remediation

Current technologies are very labour intensive for handling, decontaminating and assaying the material. More effective automated processes are required to optimise the ultimate disposition of the materials and to minimise waste volumes. Facilities that have been in SAFSTOR for many years often have components that have not been maintained or are inoperable and therefore are not candidates for *in situ* chemical decontamination. An option for stabilisation is entombment. Linked to the need for characterisation is the need to develop an understanding of the performance of techniques to decontaminate or fix contamination in place and retard transport of contaminants to the environment. More automated, modular handling, sorting and decontamination of materials removed during decommissioning is also needed.

Raw and bulk wastes in historical facilities often carry the greatest level of environmental risk because of the limitations in the design of the facilities and their reliance on active systems to maintain their safety and environmental protection status. In some cases contaminated land or groundwater remediation is a necessary activity. In others, stabilisation or some other intervention may be the only viable option. Large volumes of contaminated land and groundwater may require treatment (preferred to disposal or managed *in situ*). More development of thermal, chemical and biological treatment methods may offer less energy-intensive and less intrusive methods of remediation.

More effective methods of remediation are required, especially for more mobile radionuclides such as tritium, ^{14}C and ^{36}Cl. Developments in these methods may also bring benefits to the protection of the environment from shallow land waste disposal facilities.

Materials and waste management

The priority is to deal with high hazard, high environmental risk facilities to ensure that the wastes are removed from ageing facilities at the earliest safe opportunity. All wastes being conditioned for long-term storage and disposal need to be supported by safety cases underpinned by an understanding of how the conditioned waste and waste containers will evolve over time, recognising that the conditions for acceptance (CFA) for transport and disposal will need to be met. New methods for waste treatment and packaging can improve confidence in meeting CFA requirements by reducing volumes of disposal (meaning less impact on the environment and less cost). In many counties intermediate risk waste will need to be stored for long periods (up to 100 years or more). Periodic monitoring will need to be undertaken to provide confidence that the waste packages are ageing as predicted and within the bounds considered acceptable in the safety cases. Innovation in package monitoring will bring benefits to waste store design and increased confidence in meeting CFA.

For many smaller facilities it is too onerous to develop waste management facilities to support stabilisation and clean-out activities. Mobile or modular waste treatment facilities that can be easily transported and adapted to a number of stabilisation tasks will be invaluable, particularly as it becomes increasingly expensive to maintain large scale site support services as site operations run down at decommissioning sites. In addition, these mobile or modular waste treatment facilities help to provide up-to-date and best-available technologies for long-term dismantling.

Some wastes (organics or metals such as uranium) react with encapsulants, causing problems with waste form integrity (cracking) and transport and disposal (hydrogen generation). Graphite contains long-lived highly mobile radionuclides such as ^{14}C, ^{36}Cl and ^{3}H that pose a challenge for conditioning and long-term sequestration of encapsulated graphite wastes. In addition, conventional cement or grout encapsulants do not readily adhere to graphite or other organic form wastes. Plutonium and high transuranic wastes also have very long half-lives, can generate significant heat in the waste, and are significant internal dose hazards. There are large stockpiles of depleted uranium that must be recycled and re-used within the fuel cycle of next generation nuclear facilities or must be processed and disposed of safely. Other waste forms such as natrium (a sodium bicarbonate product from passivation of sodium-cooled reactors) and organic wastes like ion exchange resins with long-lived radionuclides pose unique processing, packaging and disposal challenges.

Site characterisation and environmental monitoring

In addition to the challenges discussed for characterisation and survey, modelling software for the fate and transport of contaminants in the environment and resultant dose or risk are currently very limited and have not taken advantage of current advances such as geostatistics. Contamination scenarios with multiple sources in complex hydrogeological environments cannot be modelled by current software. Simplifying assumptions or separate runs for each contaminated zone need to be made in order to determine appropriate clearance criteria. In addition, probabilistic analysis run times can be lengthy for some radionuclides, even if the model is very simple (without multiple soil types). There has also been very little bench testing for comparison of various software systems. Modelling contaminated concrete that will remain in subsurface buildings often requires the use of several codes such as DUST MS and RESRAD or spreadsheet calculations (BNL, 2003, 2004).

In some cases, facility or groundwater monitoring will need to continue for many decades after sites are remediated. Remote and automated methods of monitoring will reduce the burden of programmes, meaning better quality data will be collected and bring greater assurance of successful remediations.

2. Characterisation and survey prior to dismantling

Theme overview

Physical, radiological and non-radiological characterisation prior to dismantling is a key element of all decommissioning projects. The information gained through characterisation identifies the hazardous constituents, their concentrations and surface/subsurface distributions. This understanding of the hazards at the facility identifies the systems, structures, components (SSC) and areas that require remediation, identifies the occupational and environmental hazards that need to be controlled, and underpins the sentencing options for the materials to be removed. This information forms the basis of the conceptual site model. Characterisation prior to dismantling impacts directly on all other activities in a decommissioning programme, including the choice of technologies for segmentation/removal, pre- or post-removal decontamination, treatment and processing of the material, as well as the survey, monitoring and assay methods to be used to sentence the material. It also informs the waste optimisation strategy to be employed. The objectives of facility pre-demolition characterisation are to:

- Identify the types, quantities and physical properties of systems, structures and components (SSC) to be removed or dismantled.
- Identify the radiological and non-radiological hazards associated with the SSC. This may be achieved using a combination of historical information on uses and materials in the area, radiological and non-radiological surveys or assays, as well as material sampling and analyses.

This information is then used to plan the material removal strategies, material sentencing, and pre- and post-removal handling and treatment of the material. The pre-dismantling characterisation provides the basis of the monitoring, surveillance and engineering controls necessary to protect workers, the public and the environment during dismantling. It also provides the basis for the pre- and post-removal treatment (e.g. fixing contaminants or decontamination), as well as the material assay and monitoring required for sentencing, packaging and transport of the materials.

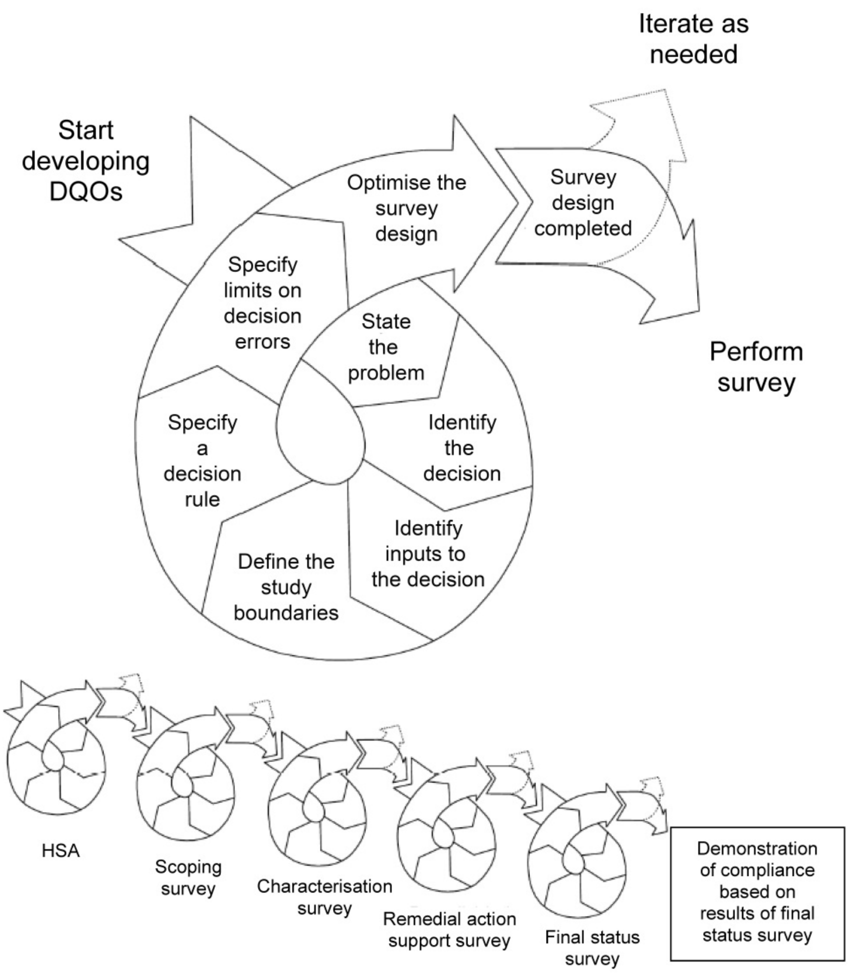
The scope of the pre-dismantling characterisation theme is defined by the working group as including the physical, radiological and non-radiological assessments of SSC prior to dismantling. These measurements and evaluations are undertaken to support planning for dismantling and material management. Characterisation includes: HSA; physical measurements of materials and structures; hazards analysis (which includes industrial safety, radiological and hazardous materials assessments); survey/sampling; non-destructive and destructive laboratory analysis; and end-state modelling to determine acceptable levels of residual contaminants.

The analysis of responses provided by different countries showed a good level of homogeneity in national priorities for research needs within this theme. For certain issues, such as statistical sampling, developing scaling factors and estimating levels of impurities in recycled materials, the need appears to be related more to having an international approach or standard. Other issues of high interest for many programmes include the development of technologies for rapid and non-destructive measurement of alpha and beta emitters (e.g. ^{3}H, ^{90}Sr, ^{14}C, ^{36}Cl, ^{63}Ni) on hard-to-access surfaces.

Summary of current practices and guidance

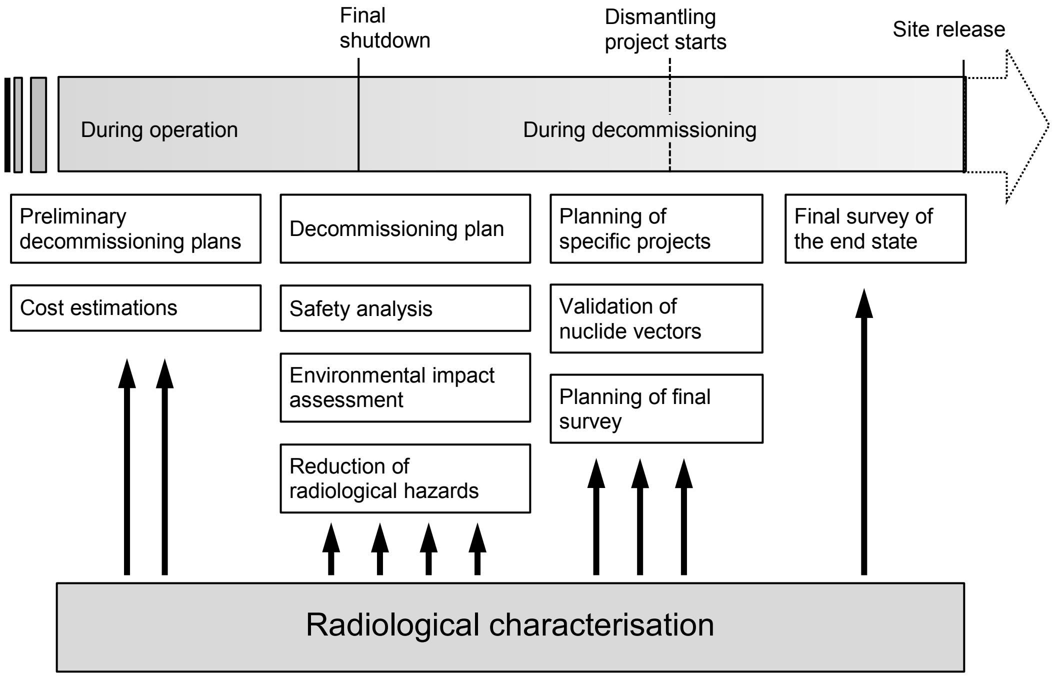
Characterisation of SSC must be conducted for all decommissioning facilities regardless of their size and use. The objectives of the characterisation process are to provide physical, radiological and hazardous materials inventories to support decommissioning planning and execution. The radiological and non-radiological contaminants and their levels and distributions must be identified and understood in the context of a site conceptual model in order to select the proper D&D methods, to implement the proper engineering controls, to select the proper protective equipment for the workers and to perform the proper monitoring and sentencing of the facility materials. As seen in Figure 2.1, characterisations occur throughout the radiation survey and site investigation process and are refined to provide increasing detail and information to support the decommissioning as it progresses.

Figure 2.1: Repeated EURSSEM/MARSSIM applications of the data quality objectives process



Another elegant graphical conceptualisation of the characterisation process was provided at the 2011 Radioactive Waste Management Summer School in Ispara, Italy by S. Wörlen (2011), as shown in Figure 2.2. A nuclide vector is equivalent to waste stream (United States) or fingerprint (United Kingdom) and can be represented as ratios among radionuclides or scaling factors that can be used to infer hard-to-detect nuclide concentrations.

Figure 2.2: Graphical conceptualisation of the characterisation process



Source: H. Efraimsson, OECD/NEA WPDD, accessed from Wörlen (2011).

Thus, characterisation occurs throughout the decommissioning process in major phases with different objectives and methods:

- *Decommissioning cost estimation* – Although often not recognised as part of the characterisation process, characterisation data is an essential part of the decommissioning cost estimating process. Cost estimating requires an inventory of physical attributes and contaminants (radiological and hazardous) as part of the basis for disassembly and material disposal costs (NRC, 2001). The physical inventory is a preliminary estimation of the physical characteristics, quantities and attributes of the facilities systems, structures and components (SSC). Physical characteristics include the types of materials such as the amount of stainless steel, carbon steel, concrete, graphite, insulation and copper cable to be removed, as well as the size, thickness and weights and bulk amounts. These material characteristics are important in gauging the level of effort and equipment required for removal, handling, packaging and transport. In addition, preliminary estimates of the nature and extent of contaminants is also required to incorporate removal, sentencing, transportation and disposal costs into the decommissioning estimate. This may also require an initial estimate or characterisation of non-hazardous contaminants that determine if components or materials will be suitable for re-use (e.g. high value components such as pumps, valves, turbine generators, transformers) at other licensed or unlicensed facilities, or re-use in end-state design of the facility, (such as concrete backfill or paving aggregate). Re-use also entails sentencing materials as scrap for recycling. Characterisation is acknowledged to be an important part of cost estimating accuracy, as it affects costs for system and structure dismantling, decontamination and waste disposal. These estimates are typically made based on a review of plant drawings, survey and sampling records, effluent permits, interviews with facility personnel and site walk downs.
- *Historical site assessment (HSA)* – This phase starts the clearance or license termination process and is included in the license termination plan submitted to

regulatory authorities. It is also a crucial building block for developing an accurate conceptual site model that will underpin the decommissioning planning and execution. The HSA provides a more detailed identification of the hazardous constituents of concern at the time of facility closure or cessation of operations than the basis in the decommissioning cost estimate. It often incorporates and builds on the cost estimate data. The HSA focus is to provide sufficient information to form a conceptual site model and plan scoping, sampling and surveys to confirm model assumptions and fill in data gaps. Hazardous constituents can range from the radionuclides present to hazardous chemicals and substances such as asbestos, PCB and heavy metals. The assessment identifies the hazardous materials present historically, the practices for use and control of the materials and facility employee knowledge of potential areas, systems, structures and components exposed to the hazardous materials and locations of known spills or on-site disposals. Data is collected through interviews with staff, review of records and site walkdowns. Changes to the facility since the decommissioning cost estimate was completed, such as modifications to the facility or the inventory of stored materials at the time of closure, are often factored into the HSA.

- *Scoping characterisations* – Once the list of hazardous constituents is determined along with historical practices and uses, a determination of the degree to which various SSC are potentially contaminated is made. Data gaps requiring evaluation are also identified and assumptions about radionuclide fingerprints, concentrations and distributions are verified. In order to focus, prioritise and implement the appropriate level of effort to the quantification of the hazards, HSA data are used to categorise facility SSC and land areas as impacted, potentially impacted or non-impacted by a hazardous constituent. Survey, assay and sampling plans are then implemented using on- and off-site resources to refine the list of hazardous constituents present, fill in data gaps, verify assigned area/material levels of impact and define extent and levels of contamination. The scoping sampling plans are typically statistically-based, with the highest level of characterisation devoted to materials that are potentially impacted as opposed to those that are known to be highly likely to be impacted or un-impacted.
- *Targeted characterisations* – These efforts are used to provide more detailed knowledge of the contaminants to support planning decommissioning activities for industrial safety and environmental considerations and to plan the removal, treatment (e.g. decontamination and stabilisation), packaging, transport and ultimate disposition of the materials. They are also used to plan clearance or sentencing survey, assay or sampling protocols. High priority or high risk SSC are targeted for more detailed survey sampling and assay efforts. This can involve more rigorous and detailed surveys, accessing system and component interiors for sampling and survey, or coring structures to determine contaminants profiles prior to disassembly. It can also involve detailed analysis using computer models and material properties for activated reactor or accelerator SSC. More detailed evaluations of physical characteristics may also be targeted in order to refine D&D plans and sentencing options. A determination of the nature of the contaminants' distribution may be required (such as levels being uniform and homogenous or intermittent and localised in order to plan sentencing and segregation of materials and ensure that proper monitoring and characterisation meets required statistical confidence levels.
- *Confirmatory characterisations* – Survey, sampling and assays are conducted during and after the dismantling/remediation process to ensure that workers, the public and the environment are adequately protected and to verify that final sentencing, storage and transportation planning are correct and were properly conducted. These surveys and sampling can be performed on the material removed to confirm fingerprints and monitoring and assay plans for material sentencing and waste

classification. They can also include confirmatory surveys to verify that the impacted materials have been removed and that further remediation is not required to meet license termination or clearance criteria. These are also critical surveys since it is costly to demobilise dismantling and remediation resources only to find that further remediation is required after the materials are packaged or upon performance of the final status survey.

- *Final status survey (FSS)* – Surveys and sampling are conducted as the final characterisation demonstrating that the material or area has met the clearance criteria for release of the item, area or facility from regulatory control. FSS of areas, structures or facilities are normally performed in accordance with a license termination plan (LTP) approved by the regulatory agencies. These surveys demonstrate that the concentration or contamination guidelines derived to meet the license termination dose or risk criteria have been met. They involve establishing survey plans and packages with data quality objectives (DQO) identified, and the specification of the required survey methods and instrumentation to meet the DQO. Measurement data undergoes a data quality assessment and statistical evaluation to ensure compliance with the facility clearance criteria has been demonstrated. The survey package/unit is documented in reports submitted to regulators as a basis for authorising the license termination. Confirmatory measurements by independent organisations are often scheduled by the regulator to confirm DQO for the survey package/unit have been met (EURSSEM, 2010; US NRC, 2002a).

  Similar survey constructs and requirements are also being applied to the release of materials and items from regulatory control. These surveys are performed to meet regulatory requirements for re-use, recycling or disposal of SSC as unregulated material. These characterisations are performed to meet acceptable detection thresholds or contaminant levels defined by regulatory agencies for each sentencing option (US NRC, 2009b; CEWG, 2006).

Characterisation is a process that is graded and iterative in nature. It entails significant costs to perform the surveys, sampling and assays both to physically obtain the data and samples and to perform on-site and off-site analyses. This chapter of the report focuses on R&D for characterisations that occur prior to dismantling. Characterisation R&D is inherent to the other themes and is discussed to some extent in each of them.

Typical characterisations conducted prior to dismantling once the facility is permanently shut down take the form of HSA and scoping survey/sampling as part of license termination or facility final clearance planning. They also include investigatory characterisation sampling to determine waste streams or fingerprints in support of material sentencing options and survey planning for decommissioning planning. There are a number of publications that provide guidance for initial facility characterisation, as seen in Table 2.1.

Table 2.1: Guidance documents for pre-dismantling characterisations

Facility type	Phase	Region	Document
Power reactors	Decommissioning cost estimating	United States	*Standard Review Plan for Decommissioning Cost Estimates for Nuclear Power Reactors*, US NRC (2004)
All types	Decommissioning cost estimating	International	*Cost Estimation for Decommissioning, An International Overview of Cost Elements, Estimation Practices and Reporting Requirements*, OECD/NEA (2010)
All types	Decommissioning cost estimating	France	*Safety of Laboratories, Plants, Facilities Being Dismantled, Waste Processing, Interim Storage and Disposal Facilities – Lessons Learned from Events Reported in 2009 and 2010*, IRSN (2012)

Table 2.1: Guidance documents for pre-dismantling characterisations (*cont'd*)

Facility type	Phase	Region	Document
Reactors	Decommissioning surveys and sampling	International	*Radiological Characterization of Shut Down Nuclear Reactors For Decommissioning Purposes*, IAEA (1998b)
Small medical industrial	Decommissioning surveys and sampling	International	*Decommissioning of Small Medical, Industrial and Research Facilities*, IAEA (2003a)
Reactors	Site characterisation	United States	*Standard Review Plan for Evaluating Nuclear Power Reactor License Termination Plans*, US NRC (2003)
All types	Decommissioning surveys and sampling	United States	*Standard Guide for Preparing Characterization Plans for Decommissioning Nuclear Facilities*, ASTM (2009)
All types	Decommissioning surveys and sampling	United States	*A Nonparametric Statistical Methodology for the Design and Analysis of Final Status Decommissioning Surveys*, US NRC (1998)
All types	Decommissioning surveys and sampling	United States	*A Subsurface Decision Model for Supporting Environmental Compliance*, US NRC (2011c)
All types	Decommissioning surveys and sampling	United States	*Environmental Radiation Survey and Site Execution Manual*, EURSSEM (2010)
All types	Decommissioning surveys and sampling	United States	*Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM)*, US NRC (2002a)
All types	Decommissioning surveys and sampling	United States	*Multi-Agency Radiation Survey and Assessment of Materials and Equipment Manual (MARSAME)*, US NRC (2009b)
All types	Decommissioning surveys and sampling	European Union	*Inventory of Best Practices in the Decommissioning of Nuclear Installations: Final Report*, EC (2006)
All types	Decommissioning surveys and sampling	International	*Decommissioning of Facilities Using Radioactive Material*, IAEA (2006c)
Reactors	Decommissioning surveys and sampling	International	*Decommissioning of Nuclear Power Plants and Research Reactors*, IAEA (1999b)
Fuel cycle	Decommissioning surveys and sampling	International	*Decommissioning of Nuclear Fuel Cycle Facilities*, IAEA (2001)
All types	Decommissioning surveys and sampling	International	*Safety Assessment for the Decommissioning of Facilities Using Radioactive Material*, IAEA (2009c)
All types	Decommissioning surveys and sampling	International	*Concrete Characterization and Dose Modeling During Plant Decommissioning*, EPRI (2008a)
All types	Decommissioning surveys and sampling	International	*Characterization and Dose Modeling of Soil, Sediment and Bedrock During Nuclear Power Plant Decommissioning*, EPRI (2009a)
All types	Decommissioning surveys and sampling	International	*Groundwater Monitoring Guidance for Nuclear Power Plants*, EPRI (2005c)

Table 2.1: Guidance documents for pre-dismantling characterisations (*cont'd*)

Facility type	Phase	Region	Document
All types	Decommissioning surveys and sampling	International	*A Practical Guide for the Performance of Combined Risk Assessment at Nuclear Power Plant Decommissioning Sites*, EPRI (2005a)
All types	Decommissioning surveys and sampling	International	*Capturing Undocumented Knowledge for Decommissioning of Nuclear Power Plants: Summary of Historical Site Assessment Documents and Lessons Learned at Eight Decommissioning Plants*, EPRI (2004b)
All types	Decommissioning surveys and sampling	International	*Radiological Characterizations for RPV and Internals Enhanced SAFSTOR*, EPRI (2003)
All types	Decommissioning surveys and sampling	International	*Summary of Utility License Termination Documents and Lessons Learned*, EPRI (2002b)
All types	Decommissioning surveys and sampling	International	*Guide to Assessing Radiological Elements for License Termination of Nuclear Power Plants*, EPRI (2002a)
All types	Decommissioning surveys and sampling	International	*Embedded Pipe Dose Calculation Method*, EPRI (2000a)
All types	Decommissioning surveys and sampling	International	*Use of Probabilistic Methods in Nuclear Power Plant Decommissioning Dose Analysis*, EPRI (2002d)
All types	Decommissioning surveys and sampling	International	*Trojan License Termination Plan Development Project*, EPRI (2002c)
All types	Decommissioning surveys and sampling	International	*Determining Background Radiation Levels in Support of Decommissioning Nuclear Facilities*, EPRI (2001b)

Summary of challenges and R&D needs for characterisation prior to demolition

Characterisation poses many challenges to ensure the hazardous contaminants are properly identified and the levels and distributions are sufficiently understood to provide the information necessary to support upcoming decommissioning activities. Unidentified data gaps or insufficient characterisation can result in unanticipated personnel and environmental exposures to hazardous constituents. They can require costly changes to work execution plans and schedules or impact material sentencing and waste disposal. Thus, the more quantitative and the less qualitative the characterisation data is, the greater the statistical certainty and confidence that the characterisation data has provided the required information to make good decisions.

Radiological and non-radiological hazardous contaminants are equally important for pre-dismantling characterisation. An accurate conceptual site model is required to properly plan work activities, remediation and sentencing of materials. Some hazards are relatively easy to determine using existing technologies and on-site resources such as direct radiation levels from radioactive materials or hazardous atmospheres that are readily detected using hand-held instruments. Others are more labour intensive, such as scans to determine surface contamination levels using hand-held alpha or beta detectors or X-ray fluorescence detectors for certain heavy metal contaminants. While photon-emitting radionuclides can be analysed by gamma spectroscopy systems in the field and at on-site laboratories with quick turnaround times, hard-to-detect radionuclides such as pure beta or alpha emitters and most hazardous materials require sample analysis at off-site laboratories. Characterisation of these constituents typically involves very specific sample preparation, preservation, shipment, processing and analyses requirements that result in higher expense and longer turn-around times. These limits on time and expense are driven by survey/sampling time and analytical costs and tend to limit the available sample and survey data. These limitations effect the confidence or certainty that contaminants have been identified and their concentrations and distributions are known.

The data and information for the various types (e.g. physical, radiological, hazardous waste, industrial hygiene and environmental) and the phases (e.g. decommissioning cost estimation, HSA) of characterisation are often not well integrated and are maintained in decentralised often separate reports and databases. This can result in incomplete or redundant sampling to characterise a material or SSC.

For large decommissionings with a wide array of hazardous constituents, the storage, maintenance and evaluation of data to ensure data quality objectives are met is often a labour-intensive task with potential for significant analytical and human performance errors. In addition, general and analyte-specific processes and procedures are required to support the characterisation process and maintain data integrity from the time the samples and information are collected in the field until the results are delivered from the laboratory.

In 2001 the National Research Council (NRC) made three research recommendations with regard to characterisation:

- basic research leading to ultra-sensitive devices for rapid characterisation and certification of amounts of radionuclides and EPA-listed substances on the surfaces of construction materials and equipment (e.g. pumps, motors);
- basic research leading to development of real-time and minimally invasive methods to characterise radionuclides and EPA-listed substances as a function of depth in construction materials, especially concrete;
- basic research leading to the development of methods for remotely mapping radionuclides and EPA-listed substances.

The labour-intensive sample collection and measurement methods required for radiological and non-radiological hazardous contaminants such as PCB and heavy metals expose workers to radiation and additional risks and contribute to the high costs of characterisation. These costs are presently estimated at 15-25% of the total D&D budget. Characterisation generally requires a detector tailored to the contaminant being measured and its matrix; for example, concrete, metal, liquid or air. In many instances, reliance is placed on characterising and mapping sites by physically removing samples (e.g. wipes, cores, paint scrapings), sending these to an offsite lab and conducting chemical extraction and analyses. When on-site measurements are obtained, use is typically made of handheld monitors or on-site laboratory counting equipment, and the data are recorded manually. Often measurements must be repeated several times at each step of the D&D process. As an example, over 400 000 survey measurements were made in the course of decommissioning the Fort St. Vrain commercial power reactor. Over half (221 000) were required for the final survey, which required 22 months to complete. In addition, the allowable levels of residual contamination had to be reduced by about 25% below the regulatory guide to account for nuclides such as ^{55}Fe and tritium that could not be detected with available field instrumentation (NRC, 2001).

Some of the challenges identified under this theme include:

- statistical and calculation methods for modelling, including validation of methods (e.g. in relation to representativeness; grid density; number of samples; where; point samples/heterogeneity within the grid; defining an acceptable level of uncertainty) and the efficiency and accuracy of non-destructive testing (NDT) methods;
- correlation between contamination measurements from sampling and calculated values from dose rate measurements (gamma emitters) and scaling factors (beta and gamma emitters), including piping; concrete and depth of intrusion of contamination into the concrete, graphite (including alpha contamination);

- modelling the movement of highly mobile nuclides (tritium, ^{14}C in graphite);
- measuring activity of hard-to-detect pure beta and alpha emitters;
- correlation of key radionuclide ratios and scaling factors (between easy to measure and hard to measure nuclides), different solubility of scaling nuclides such as ^{137}Cs, ^{60}Co, ^{241}Am, and hard-to-measure nuclides such as ^{3}H, ^{14}C, ^{36}Cl, ^{90}Sr, ^{237}Np, $^{239/240}Pu$, $^{242/243}Cm$;
- estimating levels of impurities in metals, concrete, etc. for recycling and re-use and for reduction of activation contaminants in new build materials;
- development of remote and non-destructive techniques for rapid characterisation of contaminants to allow segregation and/or changes in the classification of waste;
- *in situ* (rather than off-site) measurements, e.g. use of mobile laboratories;
- characterisation in and around difficult to access structures (e.g. drains).

Suggested additional research and development

Statistical modelling and sampling

Challenges

Information gathered during the cost estimating and HSA phase is used to develop a conceptual site model of what materials and contaminants are present, their amounts and their distributions. This conceptual site model is used as the basis for identifying data gaps, such as further sampling and testing required to determine the extent of asbestos insulation in a given area or the additional sampling required to identify fingerprints or waste streams with potentially high transuranic levels, etc. The initial conceptual site model forms the basis of near-term and long-term decommissioning planning. However, these additional sampling decisions are often based on qualitative or semi-quantitative data without a clear understanding of the potential contaminant surface and subsurface distributions and concentrations or the statistical confidence level of the data being used as the basis for decommissioning planning.

Processes such as those found in MARSSIM (US NRC, 2002a), MARSAME (US NRC, 2009b), EURSSEM (2010) and NICOP (CEWG, 2006) are based on assumptions of homogeneous contaminant distributions within a survey unit and rely on randomly generated survey or sampling points in order to achieve the desired statistical confidence level (e.g. often 95% confidence) that contaminants meet a desired action level. More advanced sample design strategies can be employed, such as two-phase sampling design, when these underlying premises are questionable (US EPA, 2002).

Action levels used to plan pre-demolition characterisation efforts could be for clearance or recycling of the material, a waste classification or a license termination. Because contaminants typically originate and disperse from a source or sources within the survey unit such as spills, leaks or non-uniform neutron fluxes, they are rarely uniformly distributed within a material or area. To compensate for this under the existing survey and sampling guidance, the minimum number of randomly generated survey and sampling points are often supplemented with additional "biased" locations qualitatively added to the survey plan to provide sufficient detail with regard to concentrations and distributions to support the removal and sentencing planning.

This can often result in high costs for sampling, scanning, analysis and significant decommissioning schedule time and effort to complete the surveys/analyses to characterise materials to a high level of confidence in the data. The working group felt that the following areas required additional R&D to refine the statistical and calculation basis of characterisation surveys prior to dismantling:

- Better understanding of the representativeness of the characterisation data to the actual contaminant levels and distributions.

- Better definition of grid density and number of samples required to achieve the desired confidence level of the data and how to factor the degree of heterogeneity within the sample area grid into this determination.
- Better definition of the acceptable levels of uncertainty depending on the use of the characterisation data. For example, clearance of materials for release from regulatory control or to meet license termination criteria typically requires 95% or greater confidence level in the measurement data, whereas far less rigorous overall confidence levels (such as within a factor of ten) are often acceptable for waste classification and occupational safety and health purposes.

Developing a clear methodology to define statistically based survey and sampling characterisation plans that achieve the desired confidence levels and address contaminant heterogeneity in the survey unit is very important in order to optimise characterisation efforts.

In addition, better modelling of activated materials and long-lived highly mobile radioactive contaminants are also necessary to support evaluations of intermediate storage and long-term disposal options and impacts. This is particularly necessary for better modelling and statistical confidence in long-lived radionuclides in irradiated graphite for reactor decommissioning in Europe.

Summary of characterisation R&D on statistical modelling and sampling

- Integration of geostatistics with EURSSEM, MARSSIM and MARSAME guidance

Geostatistical software applications have been developed and are being used to produce 2-D and 3-D maps of contaminant distributions within an area of interest. Many of these applications provide statistical confidence levels and uncertainties associated with the projected contaminant levels within the 2-D or 3-D grid. The 2-D or 3-D grid is obtained from CAD drawings if the area. The available sample data is entered into the software, including the location co-ordinates and contaminant levels or concentrations and measurement statistics such as the standard error of the result. Most geostatic software packages support upload of this data from standard CAD file formats and from spreadsheet files of the sample data that include the grid co-ordinates. The software uses the available known data to interpolate contaminant concentrations at grid locations between input data points through a process known as kriging. Most include selection of several kriging algorithms for interpolation of the data. The results are displayed as maps that show the likely contaminant distributions and statistical confidence levels and uncertainty associated with the data.

Some research has been conducted in using known factors that affect contaminant distributions to weight and inform the interpolation process. These applications hold the promise of developing better conceptual site models of contaminant distributions that provide a quantitative basis for identifying data gaps and targeting and optimising the subsequent survey and sampling required to fill them. This enables users to identify the minimum number of samples at the correct locations required to complete the conceptual model and achieve the desired level of statistical confidence. These models can also be used to plan and visualise the remediation necessary to meet license termination criteria with a high level of confidence. They can be updated as the decommissioning proceeds to plan targeted characterisations as contaminants and materials are removed.

Geostatistical applications are currently being developed, used and tested for decommissioning and environmental monitoring as noted in the references cited in this section. Some survey planning tools used to implement MARSSIM (US NRC, 2002a) have incorporated geostatistical capabilities. NUREG/CR-7021 (US NRC, 2011c), shown in Table 1, provides a framework that incorporates geostatistical methodologies into the United States license termination criteria for sites with subsurface contamination (Stewart, 2011).

Subsurface contaminants are also commonly present in structural materials that sorb radionuclides (e.g. concrete) or are activated.

Comparisons and contrasts with geostatistical and statistical approaches (Desnoyers and Dubot, 2012a; Desnoyers, et al., 2011) are being conducted to optimise the sampling effort and uncertainty quantification of the results. Research indicates that a geostatistical framework is a sound data processing technique and an efficient way to optimise the sampling strategy for the initial radiological and non-radiological characterisation of concrete structures and soils (Desnoyers and Dubot, 2011, 2012a; Desnoyers, et al., 2011; Faucheux and Jeannée, 2011; Aubonnet and Dubot, 2011; Desnoyers and de Moura, 2011; Candeias, et al., 2011). HSA data, core sample data, and surface scan data have been integrated into geostatistical models in order to map concrete structure contaminant concentrations and determine waste classification levels. (Desnoyers and Dubot, 2011; Ramsey and Boon, 2012). They have also been used for shallow and deep subsurface soil contaminations using historical data, sample gamma scan results, and coring data to optimise sampling and evaluate various remediation scenarios, costs and risks (Aubonnet and Dubot, 2011; Desnoyers and de Moura, 2011).

Specialised vehicles outfitted with scanning instrumentation have been developed for surface mapping contaminated areas using geostatistical software (Attiogbe, de Moura and Lavielle, 2011). Comparisons of estimated versus actual contaminated soil removal volumes have shown that geostatistical modelling tended to underestimate the soil volume removed by 10-30% (Faucheux and Jeannée, 2011). It should be noted that estimates of soil volumes requiring remediation are typically low due to the excavation process itself, which often requires ramping, sloping, and results in cross contamination of some clean soil during the remediation process. The technique has been used to identify areas where the confidence interval is too large and additional sampling is required (Faucheux and Jeannée, 2011; Desnoyers, et al., 2011; Desnoyers and de Moura, 2011). There are indications that kriging may be enhanced, depending on the statistical approach used (Desnoyers, et al., 2011; Lin, et al., 2011) and that *in situ* versus *ex situ* measurements may correlate better with actual concentrations (Ramsey and Boon, 2012). Cartographies created through kriging capture the contaminant's spatial concentrations and, according to measurements points, predict a likely value on each map point while also quantifying the associated uncertainty. Geostatistical calculated cartographies have been successfully performed using ISATIS software (Aubonnet and Dubot, 2011). Some studies conclude that conventional statistical (e.g. EURSSEM, MARSSIM) and geostatistical data processing are complementary rather than in opposition to one another when applied to the proper radiological characterisation stage of a decommissioning and dismantling project (Desnoyers and Dubot, 2012a; Attiogbe, de Moura and Lavielle, 2011).

Some limited work has indicated the geostatistical methods in combination with Bayesian statistical weighting may provide a means to optimise surficial scan data as well (Eby, 2010). Such approaches may be of value as alternatives to 100% surface scans currently required for known impacted survey units and materials and provide assurance that the required statistical confidence levels have been met.

- Statistical characterisation and modelling of irradiated graphite

Graphite moderated reactors pose additional decommissioning challenges because of their, design, core size and irradiated graphite disposal issues (MacKerron, 2012). Further research and development is needed to more accurately quantify the radioactive source terms of activated graphite. A better understanding of the variation in trace contaminants that lead to the formation of ^{3}H, ^{14}C and ^{36}Cl is required for nuclear grade graphite used in various countries. Consistent reproducible analytical determinations of trace contaminant quantities and better neutron activation models for graphite cores are required in order to provide accurate estimates of overall source terms within the irradiated graphite. This is necessary in order to design and evaluate long-term storage solutions. The capability to more accurately model activation products in graphite could also be important in evaluating other options for irradiated graphite recycling and re-use (Pappano and Burchell, 2010; Burchell and Pappano, 2012).

To date, the correlation between sampling results and predicted source terms among countries such as France and Britain has not been good. IAEA-TECDOC-1647 (2010b) notes that present work under development at the University of Manchester includes thermal treatment and leaching of activated graphite samples to develop methods to accurately determine the activity of nuclides in nuclear graphite. This work is initially concentrating on the activity of ^{14}C and ^{3}H. The aim is to understand the type and quantity of impurities in both un-irradiated and irradiated graphite.

Future research will focus on questions such as graphite isotopic concentration and differences between the surface and the matrix (IAEA, 2010b). Discussions at the 2010 meeting on Progress in Radioactive Graphite Waste Management (IAEA, 2010b) indicate that there is a lack of confidence in determining the isotopic contents of gas-cooled reactor graphite cores. There are significant differences between French model calculation and measurement results reported for Lithuanian, United Kingdom and French reactors. Measurement results are widely scattered (IAEA, 2010b; Poncet, 2010). At the time of the 2010 meeting, Magnox (United Kingdom) had only one measurement on each of six reactors and some felt far more measurements were needed. Some participants cautioned against "blindly" making measurements; the usefulness and application of each measurement should be evaluated first. At this time large variances in the nitrogen content (parent of ^{14}C) between French EDF/CIDEN and British irradiated graphite results are unresolved for ^{14}C and for ^{36}C content (IAEA, 2010b; Poncet, 2010).

In addition to the intensive work being carried out in the United Kingdom and France, Idaho National Laboratories in the United States is also undertaking a major effort to better characterise un-irradiated and irradiated graphite specimens (Moore, 2011). Research is under way in Lithuania as well to correlate characterisation data with neutron activation modelling results (Remeikis, et al., 2010). As noted later in the section entitled *Develop improved methods for measuring ^{36}Cl in graphite* wherein the need for research and development to better quantify ^{36}Cl concentrations in activated graphite is discussed, analyses using mass spectroscopy technologies, such as accelerator mass spectroscopy (AMS), inductively coupled plasma mass spectroscopy (ICP-MS) and laser ablation mass spectroscopy (LAMS) may provide analytical alternatives to conventional analysis techniques that rely on sample prep and liquid scintillation counting.

Typically, activation analysis models rely on Monte Carlo methods that incorporate the facility's core design, operating history and pre-established neutron spectrums. EDF is using a generalised data assimilation method to determine radioactive inventories for graphite piles at decommissioned French gas-cooled reactors (BUG1, CHA3, SLA1 and SLA2) (Poncet, 2010; 2011). "Data assimilation" or "melding" is a method of identification that gives a general tool to assess scaling factor (SF) values and uncertainties. EDF believes this method is applicable to all activation scenarios. Characterisation is expressed according to three conventional approaches: *i)* an exclusively calculation mode (theoretical estimation); *ii)* an exclusively experimental mode (measurements *in situ* or on samples); *iii)* a mode combining the two approaches by optimising with calculation measurement adjustment (data assimilation) (Poncet, 2011). The main limit to this characterisation is the lack of knowledge of graphite impurities that leads to the activation radionuclides of concern. Data assimilation is based on physical measurements, if there are enough of them, and the laws of nature (Boltzmann's equations for neutron fluxes and Bateman's equations for activation by neutron fluxes).The data assimilation method uses the ratio of the modelled versus measured radionuclide concentration and calculates confidence interval concentrations based on student t-test values for the desired confidence level and the degrees of freedom of the number of samples used (Poncet, 2011).

EDF has used this approach to calculate radionuclide concentrations in irradiated graphite. EDF concludes that this method provides a scientific demonstration that about thirty samples each weighing a few tens of grams (namely less than a kilogram of graphite) can be sufficient to calculate the radioactive inventory of several thousand metric tonnes of graphite in a pile (Poncet, 2011). In order to take uncertainties into account conservatively, the final result corresponds to the upper boundary of the confidence interval of the average specific activity for a given probabilistic uncertainty of underestimating the true concentration.

Similarly, a Monte Carlo code, MCNPX version 2.6, has been combined with sample data for calculation of activation of the graphite stack in the RBMK-1500 reactor (Plukiene, 2011). This method used a simplified 3-D Monte Carlo model of the RBMK-1500 reactor core and ICP-MS mass spectrometry measurements of impurity concentrations in virgin graphite from the RBMK-1500 to calculate the activated radionuclide concentrations. The calculated concentrations were then compared with previous calculations made with different impurity concentrations obtained by neutron activation analysis and glow discharge mass spectroscopy (GDMS) (Plukiene, 2011).

R&D to combine and reconcile estimates based on these approaches by applying geostatistical methods may be of benefit for the estimation of graphite core source terms.

Future suggested R&D for statistical modelling and sampling

- ***Description*** – Develop and integrate geostatistical and conventional statistical models and applications to optimise contaminant characterisation in SSC, the environment and activated materials.
- ***Objectives*** – To develop or test statistical methodologies, software or data collection systems to enhance accuracy and statistical certainty of conceptual site models and activated material characterisation. Develop and refine the use of statistical models to optimise survey and sampling protocols for lands, materials (including activated components) and buildings. The aim should be to research means to achieve desired statistical certainty with the least amount of scanning, surveying or sampling required to meet data quality objectives for a wide variety of decommissioning characterisation objectives. More research validating modelled or calculated values with *in situ* and *ex situ* measured values should also be conducted when new applications are developed.
- ***Desired deliverables*** – More accurate, robust and adaptable methodologies and decommissioning software with geostatistical capabilities that are adaptable to a wide variety of decommissioning facilities and contaminated materials.

Develop a method for characterising concrete contamination at depth

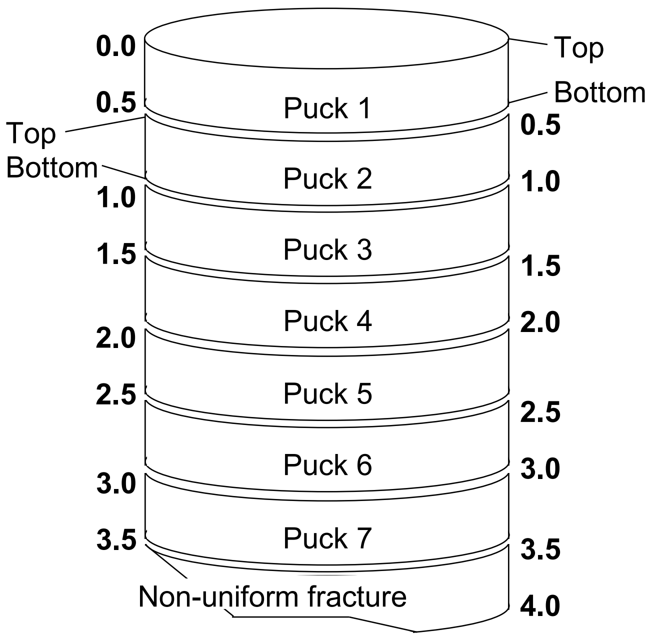
Challenges

A report on recommended decommissioning research and development by the NRC (2001) concluded that there is a need to develop a method for characterising contamination intrusion in concrete at depth and in cracks. Concrete constitutes most of the volume and weight of materials at decommissioning facilities. Because of long-term exposure, the concrete is often contaminated to a depth of several millimetres beneath its surface. In some cases the contaminant intrusion into concrete can be considerably deeper, such as for tritium, activated concrete or facilities with a history of spills and standing water. Characterisation is currently very labour-intensive, requiring sampling by obtaining concrete cores, sectioning the cores, then counting and analysing the sections to determine the contaminant distribution gradient at depth. It is also difficult to distinguish source term that has diffused into the concrete from source term that has travelled along cracks. Diffused versus crack deposited source terms have the potential to release back to groundwater at different rates (EPRI, 2008a; BNL, 2003, 2004).

The development of minimally and non-invasive real-time *in situ* sensing technologies to characterise the concentration of contaminants, as a function of depth within concrete, could eliminate difficulties associated with core sample collection and subsequent analysis and greatly reduce characterisation costs. There are presently no real-time non-invasive means available to adequately determine the concentration depth profile of contaminants in concrete; hence there are good opportunities for research in this area (NRC, 2001; Slaninka and Slávik, 2012).

Figure 2.3: Core sectioning and labelling used for United States decommissioning facility

Each puck is counted by gamma spectroscopy on top and bottom



Gamma-ray spectroscopy has been used with limited success for selected isotopes (Boden, 2012; Siclen, 2011; Bronson, 2011; Dewey, Whetstone and Kearfott, 2011; Oberer, et al. 2011) but is not applicable for all radionuclides of relevance since nuclides such as tritium and ^{14}C do not necessarily scale to gamma-emitting radionuclide concentrations. An additional challenge is the limited range and heavy attenuation of alpha and beta particles in concrete, making the quantification of pure beta- or alpha-emitting radionuclides at depth very difficult. Similarly for non-radiological toxic or hazardous contaminants, X-ray fluorescence is limited to the measurement of surface contaminants.

Summary of characterisation R&D on characterising concrete contamination at depth

- Contaminant imaging gamma camera, alpha camera, software imaging of radiation distribution

No current research specifically related to characterising or modelling contaminant intrusion along cracks in concrete was identified. However, there are current technologies being developed that may be applicable to this recommended R&D effort. When the radionuclides of concern are X-ray or gamma-emitting radionuclides or scaling factors have been established for non-gamma-emitting nuclides, gamma cameras have been used to image radiation levels and pinpoint concentrated localised contamination such as that in cracks (MacGregor, Slater and Mort, 2010; Baek, et al., 2011; Khalil, et al., 2011; Carrel, Gmar and Schoepff, 2011).

Gamma camera imaging systems are used to develop detailed three-dimensional images showing localised areas of radionuclide concentrations in patients undergoing positron emission tomography (PET) scans. Similarly, waste inspection tomography uses externally generated X-ray tomography to produce a 3-D image and gamma-ray emissions to characterise waste. This technology has been available for characterising drums of waste for a number of years (Bernardi, 1995; US DOE, 1999). REACT Engineering Ltd has made improvements to further develop its N-Visage™ gamma imaging and 3-D mapping software system. The N-Visage™ software system takes laser scanning and gamma camera radiation data from a building and constructs a 3-D map of where the radioactive sources are potentially located within the building (NDA, 2010b). A track etch technology RadBall can also be used to construct 3-D images of radiation source locations (Farfán, et al., 2010). Three-dimensional imaging based upon variations in detector positioning for objects such as walls or floors may also be starting to be feasible (Bernardi, 1995; Jaworski and He, 2011; Cattle, Goddard and West, 2005; Cattle and West, 2006). *In situ* object counting systems (ISOCS) utilise an intrinsically calibrated germanium crystal with modelling software to provide highly accurate estimates of activity in volumetric sources (Nuccetelli, et al., 2010). It may also be feasible to estimate contaminant depths based upon the attenuation differences of lower and higher energy photons emitted from a radionuclide such as ^{137}Cs (Siclen, 2011). This technology may be used to characterise gamma-emitting contaminants at depth in structural materials based on collimated measurements at various angles (Siclen, 2011; Shippen and Joyce, 2009). Imaging based on gamma or X-ray detection from multiple fields of view are also applicable to new medically-based gamma camera technologies such as MediPix (CERN, n.d.; Campbell, 2011; Hindorf, et al., 2012).

Various non-destructive testing methodologies are also available to characterise the physical properties of concrete cracks (CERN, n.d.; Aggelis, Shiotani and Polyzos, 2009; Quiviger, et al., 2010; Paris, et al., 2003; Shippen and Joyce, 2011). In the same way that X-ray tomography has been used in combination with gamma spectroscopy and laser scanning has been combined with gamma camera imaging to characterise rooms and waste drums three-dimensionally, it may be feasible to evaluate contamination migration in cracks using a combination of non-destructive testing and gamma camera or gamma spectroscopy techniques.

There are other imaging and characterisation technologies being developed that may be applicable when there are no gamma-emitting nuclides to use for scaling. These technologies use secondary emissions from alpha or beta interactions with matter as the signal input. Photo-stimulated phosphor imaging plates can be used to construct two-dimensional images of beta emitter distributions (BioSpace Lab, n.d.; Ohuchi and Hatano, 2011; Salbu and Lind, 2002; Fichet, et al., 2011) and have been used to characterise tritium in building materials (Ohuchi and Hatano, 2011; Fichet, et al., 2011) and uranium particle distributions in soils (Salbu and Lind, 2002). These phosphor plates have also been used to characterise tritium at depth in materials using bremsstrahlung induced by beta rays with an imaging plate (Ohuchi-Yoshida, et al., 2012). Phosphor plates have also been used for alpha imaging of medical samples (Bäck and Jacobsson, 2010). Improvements are also being made in the sensitivity of phosphor plate imaging algorithms (Zhang, et al., 2008). Similarly, ultraviolet cameras have been used to quantify alpha surface contamination levels based on the UV emission of nitrogen in air when alpha particles interact with it (Ivanov, et al., 2009; Chichester and Watson, 2010; Sand, et al., 2010; Inrig, et al., 2011). Emerging nanotube imaging technologies are likely to allow greater sensitivities to be achieved for characterisation and imaging based on secondary emissions of the interaction of alpha or beta particles with matter (Zou, et al., 2010).

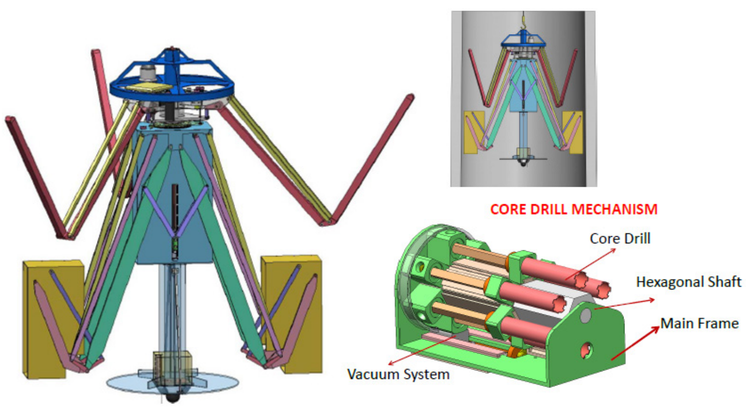
- Remote stack characterisation system

The Stack Characterization System (SCS) is a collaborative project with the Robotics and Energetic Systems Group (RESG) at Oak Ridge National Laboratory (ORNL) and the Applied Research Center (ARC) at Florida International University (FIU). The SCS is a robotic system that will be deployed into off-gas stacks located around the central campus at ORNL (Vargas, 2011; ORNL, 2009).

The SCS is a remotely operated articulated radiological data recovery system designed to deploy down into off-gas stacks from the top via crane. The battery-powered SCS is designed to stabilise itself against the stack walls and move various data recovery systems into areas of interest on the inner stack walls. Stabilisation is provided by a tripod structure; sensors are mounted in a rotatable bipod underneath the tripod. Sensors include a beta/gamma/alpha detector, a removable contaminant multi-sample automated sampler, and a multi-core remote core drill. Multiple cameras provide remote task viewing, support for sampling and video documentation of the process (Vargas and Noakes, 2010).

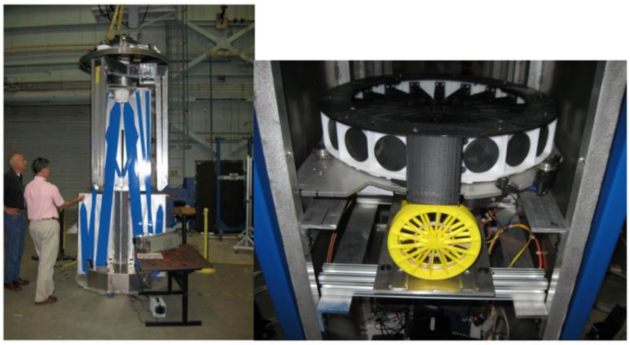
The system consists of surveying equipment capable of taking surface contamination samples, radiation readings and core samples as well as transmitting live video to its operators. Trade studies were conducted on varying concrete materials to determine the best way of retrieving loose contamination from the surface. The studies were performed at the ARC facility by FIU students, where traditional cloth wipes were compared to adhesive material. The adhesive material was tested on the RESG's smear sampler to record how much loose surface material could be retrieved (Vargas, 2011).

Figure 2.4: SCS main structure and core drilling apparatus



On-board controls are managed by networked programmable logic controllers (PLC). The PLC are used for embedded control to provide maximum reliability despite wide ranges in operating temperature, humidity and vibration/impacts. The automated removable contaminant sampler and the remote core drill each have their own PLC and could be used on other remote systems. Multiple PLC are used elsewhere on the SCS to minimise cabling and to modularise function. The operator station uses a two-operator approach to overall control. One operator manages SCS functions. The other operator focuses on data collection. Both operators have access to all video channels. In order to maintain simplicity and to lower cost, each portion of the operator station is based on a commercial desktop computer with two 24-inch flat panel displays (Noakes, et al., 2010).

Figure 2.5: SCS in retracted position and automated sampler assembly



The FIU students completed a summer internship during which conceptual designs were created for a deployable radiation detector and core drill capable of retrieving multiple core samples (Mendez, Vargas and Noakes, 2010).

Future suggested R&D – Modelling characterising concrete contamination at depth

- ***Description*** – Develop and integrate imaging technologies with imaging software applications to characterise contaminant distributions in concrete cracks and at depth in solid materials.
- ***Objectives*** – Develop or test existing and emerging technologies for detecting and imaging beta, gamma and alpha emissions directly or by secondary emissions and potentially coupled with other imaging technologies in order to image contaminant distributions at depth three-dimensionally.
- ***Desired deliverables –*** More accurate and sensitive characterisation of contaminant concentrations and distributions at depth in materials.

Develop ability to detect and quantify hard-to-measure radionuclides in solid samples with no dissolution

Challenges

Quantification of pure beta-emitting radionuclides such as ^{3}H, ^{14}C, ^{36}Cl, ^{90}Sr and alpha emitters such as ^{239}Pu, ^{240}Pu, ^{241}Am, ^{242}Cm and ^{243}Cm is often not possible without sample preparations requiring chemical extractions or separations such as combustion, distillation, precipitation, or resin extraction. Many of these preparations require equipment and expertise that preclude analysis at on-site laboratories. These processes can also introduce uncertainties in the overall contaminant concentrations, depending on the chemical form of the contaminant, due to the potential for varying yields and the applicability of the sample preparation and assay method. When gamma emitters are present at known ratios to the hard-to-detect contaminants, scaling factors such as those based on ^{241}Am for transuranic waste streams can provide a reliable method of quantifying the concentrations of hard-to-detect radionuclides. In many instances gamma-emitting radionuclides are not present, however, or their ratios to other contaminants vary or are unknown. In these situations the concentrations of hard-to-detect radionuclides cannot be scaled reliably based on gamma spectroscopy results. Therefore, development sample analysis technologies capable of quantifying hard-to-detect radionuclide concentrations *in situ* or without dissolution of the sample are desirable.

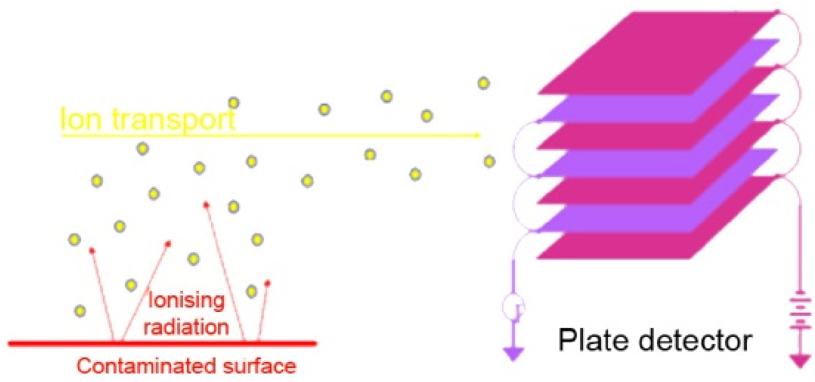
Development of such technologies poses challenges because of the short range of beta particles in solid samples and because there is no discrete beta energy that can be used to distinguish one beta emitter from another. Since beta particles are emitted at a range of energies, the spectrums of various natural and man-made beta-emitting radionuclides overlap in a composite spectrum of observed beta particle energies. Despite these challenges, the goal of this research and development effort is to eliminate chemical separation and extraction methods and allow direct detection of non-gamma emitting radionuclides *in situ* or in solid samples.

Summary of characterisation R&D on hard-to-detect radionuclide characterisation in solid samples without dissolution

- Measurements of gross alpha and beta emissions

In addition to phosphor plate imaging technologies and alpha particle UV imaging technologies discussed earlier in the sub-section entitled *Contaminant imaging gamma camera, alpha camera, software imaging of radiation distribution*, other new technologies that quantify alpha and beta emissions from solid samples or items are in development. As an example, large measurement chambers have been constructed such that the item being assayed is surrounded by the detector, as is the case with liquid scintillation counting. In the large measurement chamber devices, the item is enclosed within the chamber and surrounded by air; air acts as the detectable volume. Ions created in air by the alpha particles are blown to a detector using a fan and the measured charge is used to quantify alpha contamination levels present. An example of the principle for a long-range alpha detection (LRAD) device was provided by Chris Goddard at the RWM 2011 Summer School, as shown in Figure 2.6.

Figure 2.6: LRAD detection principle



Source: Goddard (2011).

LRAD devices were built with 10 cm × 10 cm × 10 cm rectangular chambers and with 5 cm × 9 cm × 32 cm cylindrical chambers. They were able to achieve an alpha detection limit of 60 to 120 Bq on up to 10 kg of waste. If alpha emitters are homogeneously distributed within the solid material being monitored, this equates to a sensitivity of 0.006 to 0.012 Bq/g (Naito, et al., 2010; Hirata, et al., 2008). The Babcock IonSens has chambers of 100 cm × 100 cm × 80 cm or cylindrical 600 cm × 15 cm × 15 cm and achieves MDA of 10-15 Bq (total alpha) for a 3-minute measurement. Longer measurements have achieved detection limits below 5 Bq (Goddard, 2011).

- Measurement of specific radionuclide concentrations

The National Research Council (NRC) report on R&D for D&D (2001) states that laser ablation mass spectroscopy (LA-MS) is an example of a rapid characterisation technique that is suitable for solids such as concrete and requires no sample preparation. An intense pulsed laser is used to vaporise surface material (ablation). An inert carrier gas (argon) transfers the ablated material to an inductively-coupled plasma torch, where the sample plume is disassociated into ionised atomic species. A mass spectrometer subsequently identifies the species and determines its abundance in the sample. Continued ablation of the surface provides the possibility of obtaining a three-dimensional profile of the contamination. Sensitivity and dynamic range are such that constituent concentrations of most elements in the periodic table can be measured from parts-per-billion to tens of per cent with a single analysis. The sensitivity of this analytical chemistry approach is many orders of magnitude better than can be achieved by direct radioactivity measurements for some radionuclides. Typical potential minimum detectable levels are 1×10^{-8} Bq/g for ^{238}U, 10^{-2} Bq/g for ^{239}Pu, 27.7 Bq/g for ^{137}Cs, and 277 Bq/g for ^{60}Co. The technique is applicable to organic and inorganic species. Variations of laser ablation spectroscopies are attractive as well. These include laser ablation, inductively-coupled plasma atomic emission spectrometry (LA-ICP-AES), and laser-induced breakdown spectroscopy (LIBS). Research to adapt LA-MS and related approaches for D&D applications that require ruggedness, portability and high sensitivity would most likely involve basic principles of energy beam-material interactions, including energy coupling, mass removal by vaporisation and ablation, particle generation, gas dynamics, solid vapour entrainment and transport processes (NRC, 2001).

The laser ablation mass spectroscopy (LA-MS) technologies have come to be considered off-the-shelf instruments and allow isotopic analysis of solid samples to be achieved with little or no sample preparation (Koch and Gunther, 2011). Miniaturised systems using time of flight (TOF) mass spectrometers have been developed for interplanetary space exploration (Tulej, et al., 2012). Laser ablation molecular isotopic spectrometry (LAMIS) uses the energy of a high-powered laser beam to ablate a tiny spot on a sample, creating a plasma plume for spectroscopic analysis that reveals chemical elements and their isotopes (Yarris, 2012; Theriault and Lieberman, 1995). Savannah River tank liquid slurry samples have been analysed by inductively coupled plasma mass spectroscopy (ICP-MS) for ^{99}Tc, ^{233}U, ^{234}U, ^{235}U, ^{237}Np, ^{238}U, ^{239}Pu and ^{240}Pu (Hay and Pareizs, 2011). Laser ablation mass spectroscopy (LA-ICP-MS) measurements on uranium oxide were tested against NIST standards and found to provide sufficient precision and accuracy to determine uranium isotopic concentrations (Marin and Sarkis, 2011). Accelerator mass spectroscopy is able to achieve better isotopic ratio resolution and sensitivity and may be feasible for limited material testing such as for non-proliferation verification and activation analysis studies (De Cesare, 2011).

Another possibility that holds promise for transuranic detection without sample dissolution was recently reported (Science, 2012). A nuclear magnetic resonance (NMR) peak for ^{239}Pu was identified. NMR and its spatially sensitive cousin, magnetic resonance imaging (MRI), have found widespread application in chemical and biological characterisation studies. For the most part, these studies take advantage of the energy bifurcation manifested by hydrogen nuclei with oppositely directed spins in a strong magnetic field (Science, 2012). The nuclei in a magnetic field absorb and re-emit electromagnetic radiation. This energy is at a specific resonance frequency that depends on the strength of the magnetic field and the magnetic properties of the isotope of the atoms; in practical applications, the frequency is similar to VHF and UHF television broadcasts (60-1 000 MHz). NMR allows the observation of specific quantum mechanical magnetic properties of the atomic nucleus. Many scientific techniques exploit NMR phenomena to study molecular physics, crystals and non-crystalline materials through NMR spectroscopy. NMR is also routinely used in advanced medical imaging techniques, such as in magnetic resonance imaging.

From the approximately 90 nuclear isotopes that are known to have a non-zero magnetic moment, about 30 feature a nuclear spin of $I = ½$. The study of this class of nuclei is generally advantageous because their NMR properties are dictated solely by magnetic interactions, avoiding effects such as additional shifts or line broadening due to the nuclear quadrupole interaction. The most commonly leveraged spin-½ nucleus in this context is ^1H, the probing of which has had a profound impact on structural analysis and magnetic resonance imaging in physics, chemistry and medicine. More generally, many heavier elements manifest the same effect – including ^{13}C, fluorine and phosphorus (Science, 2012).

In actinide science in general, NMR studies have been limited in scope to nuclei associated with ligand atoms. The only exception is the application of ^{235}U NMR to UO_2 and UF_6. There have been extensive efforts to realise NMR on the actinide nuclei of various compounds because their electronic properties are governed predominantly by the actinide atom itself. The search for NMR in actinides has focused primarily on ^{239}Pu. The spin-½ nucleus of ^{239}Pu with its sizable nuclear moment should, in principle, be an ideal candidate for NMR measurements in Pu-based compounds (Yasuoka, et al., 2012).

In theory, researchers have known for 50 years that plutonium nuclei have a net spin conducive to NMR. ^{239}Pu is the only spin-½ nucleus that has yet to be observed by NMR, in spite of more than 50 years of effort on a range of ^{239}Pu compounds. Yasuoka, et al. have now at last observed the resonance of the ^{239}Pu isotope in a sample of plutonium dioxide (Science, 2012). They observed the ^{239}Pu resonance from a solid sample of plutonium dioxide (PuO_2) subjected to a wide scan of external magnetic field values (3 to 8 tesla) at a temperature of 4 kelvin (-269°C). By mapping the external field dependence of the measured resonance frequency, they determined the nuclear gyromagnetic ratio $^{239}\gamma n(PuO_2)/2\pi$ to be 2.856 ± 0.001 megahertz per tesla (MHz/T). Assuming a free-ion value for the Pu^{4+} hyperfine coupling constant, they estimated a bare $^{239}\gamma n/2\pi$ value of ~2.29 MHz/T, corresponding to a nuclear magnetic moment of $\mu n \approx 0.15\mu N$ (where μN is the nuclear magneton) (Yasuoka, et al., 2012).

Whether or not this new understanding of the ^{239}Pu magnetic moment can be translated into a NMR-based detection and quantification of ^{239}Pu in samples with other lanthanides and actinides present is unknown at this point. There are two main reasons why ^{239}Pu NMR has remained elusive. First, in atoms with unpaired *f* shell electrons (e.g. lanthanides and actinides), an extremely strong hyperfine interaction between electron and nuclear spins gives rise to a large internal magnetic field (~100 T) at the nuclear site. As a consequence, the resonance frequency is shifted by several orders of magnitude, and the nuclear spin-lattice relaxation rate (1/T1; where T1 is the relaxation time) becomes exceedingly fast ($T1 << 1\ \mu s$), rendering any NMR measurement very challenging. The second reason was that there was until this recent discovery no accurate account of the ^{239}Pu nuclear moment μn. Therefore, there is reliable account of the relevant value of γn. Plutonium and its compounds show incredibly complex behaviour, exhibiting a multiplicity of oxidation states and a pervasive tendency to form a range of non-stoichiometric phases in the solid state (Yasuoka, et al., 2012). This experiment determined the magnetic moment in PuO_2. Further research directly accessing the consequences of plutonium's 5f electrons at the atomic and structural unit-cell scales with NMR can provide a valuable tool for the study of plutonium solid-state physics, chemistry and materials science. A better understanding of the NMR spectra associated with ^{239}Pu in various sample forms may lead to an NMR-based assay approach.

Soil vapour extraction monitoring for tritium measurements in soils

In another approach to quantification of radioactivity without dissolution of solid samples, EPRI is investigating the feasibility of monitoring for tritium groundwater contamination by the vapours emitted from the vadose zone (EPRI, 2008b). The distribution and migration of tritium contamination at the decommissioning facilities investigated in the EPRI report suggested that detection of tritium (^3H) in unsaturated soils was a possible technique for detecting leaks to mitigate widespread contamination through early detection. The proposed monitoring approach is an integration of NPP decommissioning findings, current soil vapour extraction (SVE) technologies and United States Geologic Survey studies involving ^3H migration in the vadose zone. This is currently a qualitative monitoring methodology that is used as an indicator of potential groundwater tritium contamination. Other research on vadose zone vapour emission modelling may provide useful methodologies to allow for more quantitative estimates of groundwater tritium concentrations using this technology (Whicker, et al., 2011; Jiménez-Martínez, et al., 2012; Marang, et al., 2011; McAlary, et al., 2010).

Future suggested R&D on hard-to-detect radionuclide characterisation in solid samples without dissolution

- ***Description*** – Develop and test technologies and methodologies/approaches to enable qualitative and quantitative determination of hard-to-detect radionuclide levels in solid samples without sample dissolution.
- ***Objectives*** – Develop/refine equipment and instrumentation capable of quantifying hard-to-detect levels in solid samples using primary or secondary particle or photon emissions. Deploy and test or develop mass spectroscopy-based systems and applications capable of supporting decommissioning characterisation efforts.
- ***Desired deliverables*** – Development of less labour-intensive and more broadly applicable solid sample analysis and monitoring assays.

Develop improved methods for measuring ^{36}Cl in graphite

Challenges

IAEA-TECDOC-1647, *Progress in Radioactive Graphite Waste Management* (2010b) states that radioactive graphite constitutes a major waste stream that arises during the decommissioning of certain types of nuclear installations. Worldwide, a total of around 250 000 tonnes of radioactive graphite, comprised of graphite moderators and reflectors, will require management solutions in the coming years. ^{14}C is the radionuclide of greatest concern in nuclear graphite; it arises principally through the interaction of reactor neutrons with the nitrogen that is present in graphite as an impurity or in the reactor coolant or cover gas. ^{3}H is created by the reactions of neutrons with ^{6}Li impurities in graphite as well as in fission of the fuel. ^{36}Cl is generated in the neutron activation of chlorine impurities in graphite. ^{36}Cl has a 300 000 year half-life. The TECDOC notes that there have been a substantial number of measurements carried out to determine the chlorine levels in Magnox reactors in the United Kingdom. However, it is known that ^{36}Cl is transported around the circuit; hence, the original chlorine level in the un-irradiated graphite may not accurately reflect the actual ^{36}Cl level in the core graphite.

As noted earlier in the subsection entitled *Statistical modelling and sampling*, various approaches to modelling activation product concentrations in reactor graphite have yielded differing estimates of the activation source terms. Modelling concentrations are benchmarked against measured results in samples. Therefore, more reproducible, standardised measurement protocols will assist in the quantification and final determination of activation products in reactor graphite (Vaudey, et al., 2011). Accurate estimates of ^{36}Cl source terms in irradiated graphite are especially important for the development of disposal, treatment and recycling solutions.

Most conventional processes, such as the following, require extraction and chemical separation with liquid scintillation counting and have potential for interferences by ^{3}H, ^{14}C, ^{35}S and ^{129}I:

- alkaline digestion followed by chemical separation;
- acid digestion and volatilisation of ^{36}Cl;
- thermal decomposition of the sample (with or without modifiers) and liberation of Cl species as HCl or Cl_2;
- final measurement of ^{36}Cl by liquid scintillation counting (high counting efficiency).

Summary of characterisation R&D on methods for measuring ^{36}Cl in graphite

- Acid dissolution or pyrolyser furnace with Cl resin chromatography

Radiochemical analysis of ^{90}Sr, ^{41}Ca, ^{129}I and ^{36}Cl in waste samples by Xiaolin Hou is provided in the NKS-218 Workshop Report (2010c). This report notes that beta and alpha emitters (^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{63}Ni, ^{90}Sr, ^{99}Tc, ^{129}I and some transuranics in waste) have to be determined by radiochemical analysis, including a complete separation of individual radionuclides from the sample matrix and other radionuclides before measurement by beta counting, alpha spectrometry or mass spectrometry is performed. ^{36}Cl measurement is normally carried out by liquid scintillation counting (LSC) and accelerator mass spectroscopy (AMS) (Hou, 2010b). In this presentation, issues with ashing and normal dissolution methods of sample preparation for graphite are noted and a recommended method for complete decomposition of graphite using nitric, sulphuric and perchloric acid is recommended. Dissolution heating is performed in a closed system with a condenser and absorption bottles containing sodium hydroxide. First the ^{129}I is removed from distillate solutions using chloroform, then the Cl is precipitated out using silver. The silver chloride precipitate can then be dissolved in ammonium hydroxide and counted using liquid scintillation. An added step of anion exchange chromatography can be used to separate the silver cation and chloride anion. The Cl eluent can then be counted by LSC with a lower quench. Anion exchange chromatography also has a higher decontamination factor for other radionuclides. The detection limit using LSC is 14 mBq.

A similar methodology using a pyrolyser furnace (Warwick, et al., 2010) with Cl-specific anion resin (Triskem, 2012) chromatography can be used. This procedure uses alkaline sodium carbonate bubbler traps to capture combustion products. In this case, bubbler Cl is removed on resins conditioned with silver nitrate and eluted with potassium thiocyanate; the eluent is then mixed with LSC cocktail for counting. The ^{129}I is not eluted and can later be removed using sodium sulphide. The mean Cl recovery in the first bubbler is 86%. A limit of detection of 0.02 Bq/g is achieved. Other applicable eluent and washing procedures for use of the silver-loaded Cl resin were presented at the Eichrom Users' Group Workshop, 57th Annual RRMC (Happel, 2011).

- Use of accelerator mass spectroscopy to determine ^{36}Cl concentrations in graphite

Using a sample preparation method that involves acid digestion and silver precipitation of the sample, accelerator mass spectrometry (AMS) offers a more sensitive alternative to scintillation techniques for the determination of ^{36}Cl in foods. The use of AMS provided a detection limit of 0.0005405 Bq/g (Baxter, et al., 2009; Yin, 2012).

AMS has been used to monitor ^{14}C levels during the dismantling and removal of the ANSTO's reactor structure, and its 12.1 tonnes of graphite reflector. These measurements involved direct measurements on the reactor graphite and concrete bioshield blank targets that were exposed in the building, swipe samples taken inside the tent and around the building and aerosol samples collected inside the building throughout the operation (Smith, Levchenko and Malone, 2012). A new 6 MV electrostatic tandem accelerator has been put into operation at Helmholtz-Zentrum Dresden-Rossendorf (HZDR); the system is equipped for AMS. The Dresden Accelerator Mass Spectrometry (DREAMS) facility, based on a 6 MV Tandetron, will be primarily dedicated to long-lived radioisotopes ^{10}Be, ^{26}Al, ^{36}Cl, ^{41}Ca and ^{129}I (Akhmadaliev, et al., 2012; Alamelu, Choudhary and Aggarwal, 2010).

Future suggested R&D – ^{36}Cl measurement in graphite

- ***Description*** – Develop methodologies and compare results on irradiated graphite samples to improve and validate ^{36}Cl measurements.

- ***Objectives*** – Develop, refine, or compare and validate measurement of ^{36}Cl in irradiated graphite samples using chemical separation and liquid scintillation count, mass spectroscopy or other methods.
- ***Desired deliverables*** – Development of less labour-intensive and more broadly applicable ^{36}Cl measurement techniques and more data and comparison studies between methods on actual irradiated graphite samples.

Develop new methods of alpha measurements on structures before dismantling

Challenges

Typically alpha contamination on structures is evaluated by direct scanning, smears and airborne particulate sampling. These methods require many scans and samples in order to complete a comprehensive assessment of alpha contamination on a structure. Direct scanning for alpha requires the surface to be nearly flat in order to be accurate due to the limited range of alpha emissions in air. There are currently two main ways in which alpha concentrations can be inferred in a more comprehensive manner: by scaling the levels based on gamma-emitting radionuclides or by attempting to infer overall contamination levels based on air sample results. If gamma emitters are not present or scaling factors are unknown or inconsistent, this method can result in high errors. Basing an estimate of alpha contamination levels on air sample results is also prone to higher errors due to uncertainties in re-suspension factors, sample positioning relative to the source(s),and air turn over and filtration rates associated with the ventilation.

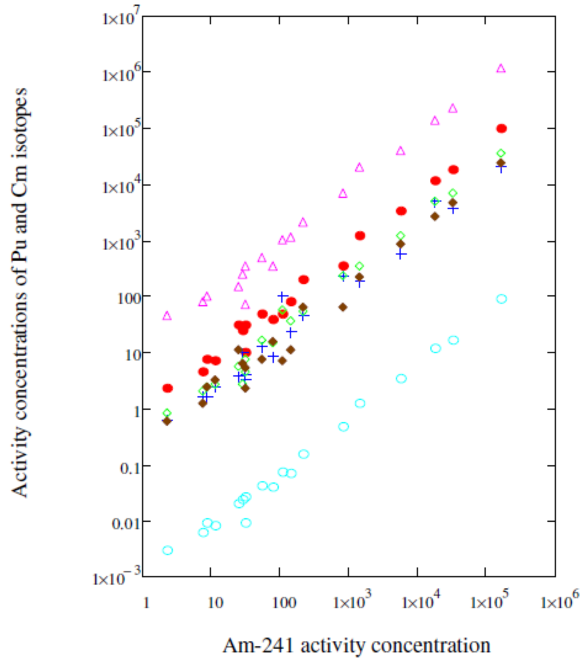
Development of a direct measurement method for alpha emitters seems to be a very important issue among member countries polled. As mentioned above in the section entitled *Develop a method for characterising concrete contamination at depth*, alpha cameras using ultraviolet emissions from air fluorescence caused by alpha particles are able to image surface deposits of alpha emitters, but are only capable of imaging very high alpha contamination levels. So there is a great interest in improving the alpha camera or developing a different technology to fulfil these requests.

Summary of current R&D for alpha detection on structures prior to dismantling

- Scaling transuranics to ^{241}Am

Americium-241 is the daughter of ^{241}Pu, which has a 14.4 year half-life, thus it is the universal scaling nuclide for transuranics (TRU) (ITRC, 2006). In their paper on determination of transuranic source terms, Slavchev, et al. (2010) explain that it is a predominant nuclide for both atomic explosions and spent nuclear fuels. The most abundant Pu nuclide resulting from atomic bomb tests was ^{241}Pu. The ratio of $^{241}Pu/^{239,240}Pu$ was 15 in the atmosphere in 1963. In 1986, the $^{238}Pu/^{239,240}Pu$ ratio in the releases from the core meltdown and fire at the Chernobyl accident was typically about 0.5 and that of $^{241}Pu/^{239,240}Pu$ was 83. Due to the high production of ^{241}Pu in spent fuel, the short half-life of ^{241}Pu (14.4 y), and the comparatively long half-life of ^{241}Am (432 y), this nuclide is normally present in stable ratios to other alpha emitting transuranics. ^{241}Am comprises 40-70% of the transuranic alpha emitters (^{241}Pu not included) at most BWR and PWR power plant decommissioning facilities with a history of failed fuels and significant alpha emitter source terms. At Rocky Flats, a United States weapons production facility where Pu is preferentially extracted, the ratio of ^{241}Am to Pu was 7:1 (14%) in contaminated soils at the facility (Bronson, Booth and Groff, 1999). At Oak Ridge the soil ratio of ^{241}Am to total TRU ranged from 18-74% (Meyer, Remington and Wojtazsek, 2007). ^{241}Am has also been found to be a good scaling nuclide for Pu-based fuels (Aggarwal, et al., 2010). Recent studies of scaling factors for clearance of materials from a hot cell facility at the Risø DTU in Denmark (Søgaard-Hansen, 2013) concluded that ^{241}Am was a good key nuclide and scaled well to other transuranic nuclides, as seen in Figure 2.7.

Figure 2.7: Plot of hot cell facility ratios at the Risø DTU site in Denmark



Source: Søgaard-Hansen (2013).

The half-lives are such that, once scaling factors of other transuranics (e.g. ^{239,240}Pu, ^{242}Cm, ^{243,244}Cm, ^{237}Np) relative to ^{241}Am are established, the ratios are relatively stable over the life of a decommissioning and are easily corrected for decay using the Bateman equations for decay and in-growth from a parent nuclide.

^{241}Am has a broad range of photon and particle emissions of different energies for 800 monoenergetic electrons, 154 photons and 22 alpha particles. As seen in Table 2.2, all of the photon yields of greater than 1% are well within the Compton continuum and very hard to distinguish from background, with the exception of the 59.4 keV gamma emission emitted 36% of the time.

This photo peak is readily detectable on laboratory equipment and has been used to quantify transuranic concentrations in wastes, soils and in rooms. It has also been shown to be a good scaling nuclide for Pu-based fuels (ITRC, 2006). Other laboratory experience quantifying ^{241}Am from ^{137}Cs in high Compton backgrounds were addressed using customised counting geometries, lead shielding and collimators such that it could be used reliably as a scaling nuclide for transuranics at a remediation conducted at Oak Ridge (Meyer, Remington and Wojtazsek, 2007). The 2009 IAEA guidance on scaling factors states:

> *Often, it is convenient to correlate $^{239+240}$Pu with ^{60}Co or ^{137}Cs and mutually correlate the other transuranic nuclides such as ^{241}Am and ^{244}Cm with $^{239+240}$Pu. In this respect, the ratios ^{241}Am/$^{239+240}$Pu and ^{244}Cm/$^{239+240}$Pu do not conform to the typical definition of a scaling factor (SF), the latter being defined earlier as the ratio of a difficult to measure (DTM) nuclide to a key nuclide. $^{239+240}$Pu is used as an auxiliary key nuclide in this case.* (IAEA, 2009c)

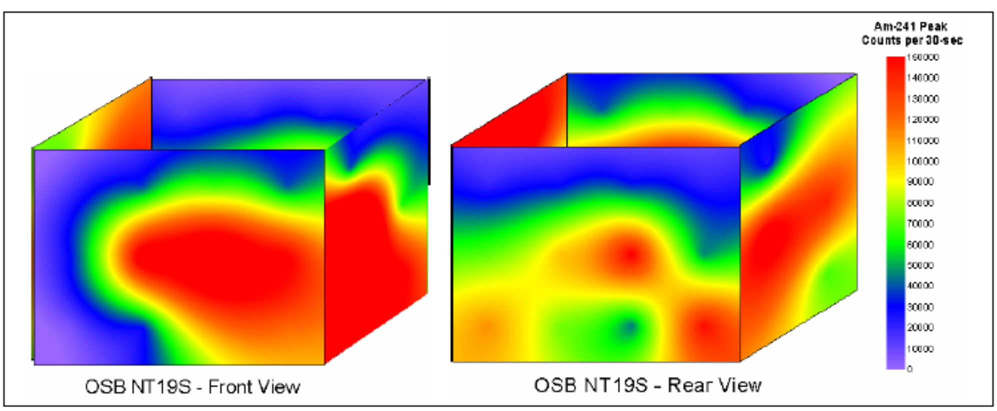
Table 2.2: ^{241}Am photons with 1% or greater yield

Type	% intensity	Energy (keV)
Gamma ray	35.7%	59.54
X-ray	21.9%	13.94
X-ray	18.8%	17.75
X-ray	5.8%	16.84
X-ray	4.6%	20.78
X-ray	2.5%	13.76
Gamma ray	2.4%	26.35
X-ray	2.2%	17.06
X-ray	2.0%	17.99
X-ray	1.3%	17.50
X-ray	1.2%	11.89
X-ray	1.0%	21.49

While the IAEA is correct that ^{241}Am scales well to other transuranic nuclides, it is a little bit backwards in situations where ^{241}Am is readily detectable by gamma spectroscopy and can be used as the key scaling nuclide. ^{241}Am monitoring results using ISOCS measurements on quadrants of transuranic waste boxes at Nevada Test Site were used to construct contour maps of ^{241}Am photon flux that indicate hot spots in the waste (Watters, et al., 2009).

A similar process was evaluated by the US EPA at Savannah River. It recently announced approval of a radioactive, remote-handled (RH), transuranic (TRU) waste characterisation programme implemented by the Central Characterization Project (CCP) at the Savannah River Site. The TRU waste characterisation activities at SRS-CCP uses acceptable or process knowledge, dose-to-curie, in conjunction with radionuclide-specific scaling factors derived in part by measurement with the In Situ Object Counting System (ISOCS) and historical assays of waste streams as well as real-time radiography (RTR) to confirm the physical form and waste material parameters of waste drums. The process uses ^{241}Am as the scaling nuclide for TRU (US EPA, 2012b). Using ^{241}Am as the scaling nuclide for TRU has also been done for TRU characterisation in the United Kingdom (Miller, 2012).

Figure 2.8: Contour maps of ^{241}Am gamma fluence from outer surfaces of OSB NT19S



Source: Watters, et al. (2009).

- TruPro® for destructive samplings in cores of concrete and metals

TruPro® is a patented decommissioning and radwaste management sampling and characterisation technology. It features a drill with a specialised cutting and sampling head, drill bits, a sample collection unit and a vacuum pump. The drill head is used to penetrate hard surfaces, which causes the bulk material to be pulverised as the drill travels through the radioactive media, efficiently transmitting a representative dry sample of bulk material to the specially designed, two-stage vacuum sample retrieval unit that prevents cross contamination of the clean retrieved samples. By using portable calibrated equipment such as liquid scintillation counters and gamma spectrometers, analysis is performed directly on the cored sample material in close proximity to sampling operations. Samples can also be analysed for hazardous materials. The results acquired from the samples using TruPro® are used to develop a more detailed subsurface chemical or radiological contamination profile from which a strategy of clean-up action can be derived. An initial activity report is generated for each sample volume in near real-time, taking from 5 to 60 minutes. Sampling areas determined to be of actionable value and concern are sampled rapidly and incrementally to acquire "dry" representative samples that are then quantitatively analysed in close proximity to the sampling operations (Charters and Aggerwal, 2006, 2011).

- Alpha sensitive scintillation and fluorescent materials

New methods of fabricating ZnS(Ag) alpha scintillators (Lee, Seo and Han, 2010; Lee, et al., 2011) used for alpha scanning may assist in developing other alpha imaging innovations. As was discussed in the subsection entitled *Contaminant imaging gamma camera, alpha camera, software imaging of radiation distribution*, alpha imaging is feasible under certain lighting conditions using the UV fluorescence in air that results from alpha particles interacting with air (Ivanov, et al., 2009; Sand, et al., 2010; Inrig, et al., 2011; Zou, et al., 2010; Hannuksela, et al., 2010). Currently these systems are capable of detecting 40 Bq/cm^2 with a 1-hour exposure and 100 Bq/cm^2 per minute of exposure when using a 10 minute imaging time (Ivanov, et al., 2011).

Zhou, et al. (2011) suggested that deep UV emitting scintillators whose emission falls in the solar blind region of the spectrum (200-280 nm) could be used in powders, paint or gels. Development of products to fix contamination that contain these scintillators could enhance UV imaging of alpha contamination. Similarly, technologies such as pulsed lasers tuned to excite ionised atoms could increase the signal from the ionised air directly above the alpha contamination. A patent filed for such a device states that nitrogen comprises approximately 78% of air but only 0.5% of the excited nitrogen ions generated by radiation emit the UV signal naturally (Rosson, et al., 2012). Alpha, beta and gamma ionising radiation create 200 times more nitrogen ions than those that fluoresce naturally. If the non-fluorescing ions remain in such a state, 95.5% of nitrogen ions will go undetected, thereby limiting the sensitivity of UV imaging systems. A light detection and ranging (LIDAR) system that uses a pulsed laser at a wavelength to be absorbed by the ionised nitrogen was recently patented (Rosson, et al., 2012). LIDAR technology is an optical remote sensing technology that measures properties of scattered light in air. Originally developed for Defense Advanced Research Project (DARPA) dirty bomb applications, LIDAR technology can be used from a safe distance to measure ionisation resulting from alpha and beta particles as well as gamma rays. The recently patented LIDAR system employs a pulsed laser transmitter, a telescope receiver and associated control and acquisition systems. Pulsed light propagates out from the laser transmitter and is directed into the air volume surrounding the radioactive source, or the "ion cloud." The ion cloud absorbs the transmitted light. This absorption induces otherwise undetectable, non-fluorescing ions to fluoresce. Light from the ion cloud is then backscattered and the telescope receiver subsequently collects the photons from the backscattered light. The intensity of the fluorescence (determined by the photon count) is measured. This provides an indication of the density of the ionised atoms and source strength. Advances in photo sensors could also lower the alpha detection threshold by air ionisation methods (R&D, 2013; Gilblom and Yoo, 2004). Technologies such as fluorescing coatings or laser excitation of nitrogen ions could increase the UV camera detection signal and lower the alpha contamination sensitivity of alpha cameras. This would enable surface contaminants to be mapped in a manner similar to a gamma camera (Sand, et al., 2010).

Future suggested R&D – Alpha measurements on structures before dismantling

- ***Description*** – Develop technologies, products and methodologies to enhance characterisation of alpha contamination on structures prior to dismantling.
- ***Objectives*** – Develop innovative technologies, products and methods that can replace or optimise the scanning and sampling required to characterise alpha contamination prior to demolition activities.
- ***Desired deliverables*** – Technologies, products and methods that increase the visibility and detection of alpha contamination or that optimise the alpha characterisation process.

Modelling of mobile nuclide behaviour on different substrates

Challenges

Numerous studies of mobile radionuclides' fate and transport in waste forms, waste disposal facilities and in the environment have allowed sophisticated environmental risk and human exposure models to be developed as discussed earlier. However, mechanisms affecting the dispersal of highly mobile, hard-to-detect radionuclides within operating and decommissioning facilities are not well understood.

In its 2001 study of required R&D for decommissioning, the National Research Council (NRC) noted that scientific understanding of the interactions among contaminants and construction materials is fundamental to developing more effective D&D technologies. Such information includes how contaminants bind to steel and concrete surfaces; how they penetrate into these materials; their migration into pores, fissures and welds; and time-dependent ageing effects (NRC, 2001). They also noted that modelling of radionuclide and chemical contaminant behaviour that is relevant to D&D problems was then almost non-existent. The models available at the time were not adequate for developing improved decontamination, storage or disposal processes. For example, surface oxides are known to sorb metal ions. The sorption has been described by a wide variety of models. Most of these models are based on measurements taken under a specified set of conditions (e.g. sorption isotherms) rather than on fundamental parameters. They are not generally applicable to the variety of conditions encountered in decontamination activities or in waste storage or disposal environments. Neither the surface nor the metal ion is explicitly treated in most studies; often the role of the chemical form of the contaminants (speciation) is neglected (NRC, 2001).

The NRC felt that investigations on radionuclides of particular concern to the DOE, such as actinides, should be stressed. A variety of conditions (pH, temperature, ionic strength) should be examined. The interactions should be kinetically and thermodynamically described to facilitate applying the data to a variety of decontamination, storage and disposal conditions and to ease the incorporation of data into first-principle models. Modelling from first principles provides an opportunity to integrate relevant results of fundamental research in both chemical interactions and biological processes to D&D problems. Such models can be the first step in bringing new knowledge to bear on improving decontamination approaches and processes. Properly integrating chemical and radionuclide speciation into D&D models is likely to be especially informative and add new knowledge in general since the most important species will likely be different from those in high-level waste or in subsurface contamination because of their different chemical environments. Beyond their use in decontamination, the models can help provide a more general scientific basis for predicting behaviour of contaminants in construction materials as a scientific underpinning of facility end states (NRC, 2001). These NRC insights and recommendations made in 2001 remain accurate and relevant a decade later.

Once the structures are decontaminated, the retention and release mechanisms of the radionuclides in the waste materials themselves are not well understood either. Even if the original distribution of mobile radionuclides is well known, it is important to have a clear understanding of their chemical form and speciation after several years on different substrates (like concrete, metals). Mobile radionuclides, as well as highly soluble nuclides such as Sr and Cs, can migrate into pore spaces in concrete and even penetrate stainless steel lattices as evidenced by sorption and weeping of ^{137}Cs on steel casks after being submerged in spent fuel pools. When high concentrations are involved, the materials can continue to sweat or leach radionuclides over time, even after repeated decontaminations (IAEA, 1999c). The behaviour of some mobile radionuclides on different substrates has been modelled in different computer codes. But a rigorous comparison of the underlying assumptions and benchmarking of the results has not been completed.

In a recent review of current models and comparison to field measurements for mineral wastes such as slag and concrete, the US NRC (2010) reports that a large discrepancy exists between: *i)* the mineral dissolution rates that are inferred from mass balance calculations performed for natural systems based on the analysed stream water and ground water compositions from large watersheds, catchments and aquifers; *ii)* the dissolution rates that are measured directly in laboratory experiments conducted with single, pure minerals in a dilute solution at an imposed pH value. Differences of up to five orders of magnitude between the field rates and laboratory rates have been reported for compositionally simple minerals. The calculations currently used to assess long-term waste material behaviour rely on fairly simple models for radionuclide diffusion and leaching, which may or may not be consistent with the degradation mechanisms of the wastes being evaluated. The ability to identify mechanistic bases for the laboratory tests and source term models used in assessment calculations will add credence to site assessments and the evaluations of site remediation plans.

Argonne National Laboratory (ANL) has initiated an NRC-sponsored activity to identify the laboratory and field tests used to characterise waste form degradation and measure the release of radionuclides and how they interface with the models used to predict radiation doses in risk assessment calculations (US NRC, 2010, 2011e). Two groups of waste materials of current interest to the NRC are: *i)* slags produced during ore processing and metal recycling; *ii)* contaminated concrete and metal debris from decommissioning activities. The study consists of four parts:

- *Part I: Conceptual Model of Leaching from Complex Materials and Laboratory Test Methods*. This report (published as NUREG/CR-7025) (US NRC, 2010) provides a summary of the initial review and analysis of existing literature regarding the weathering of various slag and concrete waste materials and waste forms, including experimental results, field measurements and modelling approaches.
- *Part II: Relationship Between Laboratory Tests and Field Leaching.* The relationships between the behaviour measured in laboratory tests and field measurements will be evaluated and the methods used to relate laboratory-measured values to field measurements will be discussed. The US NRC published NUREG/CR-7105 in October of 2011. This study focused on mineral waste forms. The terms “waste material” and “waste form” are used in the report to represent the source material from which the radionuclide is being released and the stabilised material, respectively.
- *Part III: Application of Models to Leaching Data from Slags and Concrete*. Existing source term models that may be useful for calculating weathering behaviours of slags and concretes will be identified, summarised and evaluated. The consistency of these

models with data available for the release of components from slags and concretes will be evaluated with several examples. How well current models represent the measured release behaviour will be assessed with regard to the uncertainty in long-term predictions due to uncertainties in the measured model parameters and uncertainty in how well the model represents the degradation behaviour. Alternative models or modelling approaches will be recommended as appropriate.

- *Part IV: Application of Leaching Model to Dose Assessment Codes*. The source term models for radioactive and hazardous contaminants must be interfaced with a dose assessment code to evaluate the performance of a waste site. Guidance will be developed regarding the appropriate use (or the need for integration) of laboratory data and field test information in dose assessment codes such as DUST MS (Sullivan and de Lemos, 2001; Sullivan, 2006) and RESRAD (Yu, n.d.). This will include propagating uncertainties in the mechanistic and abstracted source term models and in the coefficient values to estimate confidence levels. It is important that the abstracted models capture the environmental and temporal effects on material degradation and the release of hazardous components that are important to performance assessment.

The current use of test methods to parameterise degradation and transport models is being evaluated to better represent the mechanisms of radionuclide release in site assessments. The expected output from this activity is a protocol that can be used by the NRC to integrate the results of short-term laboratory tests and field measurements that address waste material (or waste form) degradation and leaching into the model calculations that are used to assess the stability of wastes at NRC-regulated sites prior to decommissioning. The approach will be to: *i)* develop a mechanistic understanding of the weathering processes of the particular waste material; *ii)* identify the appropriate degradation model(s) to describe the release of radionuclides; *iii)* follow an appropriate testing protocol to measure values of the model parameters to be used in performance assessment calculations. Guidance will also be developed for using leach test data in source term models.

It seems very important to find international agreement on the mechanisms and modelling principles that can be applied to the wide variety of climate, disposal and interim storage issues currently in use and under consideration globally.

Summary of current R&D for modelling mobile nuclide behaviour on different substrates

- Radionuclide release from irradiated graphite

The key radionuclides in irradiated graphite are ^{14}C and ^{36}Cl, both of which can be leached from graphite in geologic disposal installations (Towler, et al., 2011). Uncertainties still exist regarding the fate and transport of these nuclides within the material. As noted in the subsection entitled *Statistical characterisation and modelling of irradiated graphite*, uncertainties remain with regard to the radionuclide inventory of the graphite for key radionuclides such as ^{14}C, ^{36}C and ^{3}H, and there is little information available on the long-term behaviour of irradiated graphite in storage or in the environment (Vaudey, et al., 2010). In particular, little data is available on the leaching characteristics (IAEA, 2010b). Graphite leaching behaviour has been identified by the IAEA as a key issue (Towler, et al., 2011). The kinetics of release and leaching of radionuclides from irradiated graphite have not been described due to the limited amount of empirical data. It is anticipated that following closure of the geologic disposal facility being planned in the United Kingdom the entire inventory of ^{36}Cl will rapidly be leached from the graphite (Towler, et al., 2011). Transfer assessments of radionuclide release fluxes show that the main radionuclide responsible for disposal impact is ^{36}Cl, which is difficult to retain or delay (IAEA, 2010b; Serco, 2011d). A potentially significant proportion of the ^{14}C inventory may be incorporated in the graphite lattice, and may only be released very slowly, or not at all. However, this proportion is unknown, so for current work the base assumption is that ^{14}C is leached at the rates measured in short-term experiments until the entire inventory has been released (Towler, et al., 2011). ^{14}C is also retained and delayed mainly by the concrete of the package and emplacement cells. The scope of research work concerning a better understanding of the ^{36}Cl behaviour in the graphite waste is currently defined and scheduled for the coming years (IAEA, 2010b).

A recent study attempting to establish the ^{14}C release rate in high pH fluid that simulates the cementitious conditions anticipated in the post-closure repository upon groundwater intrusion is illustrative of the difficulties in providing the empirical underpinning upon which kinetics models are established. The report starts by noting that some gaseous species containing ^{14}C, such as $^{14}CH_4$ and ^{14}CO, could migrate with bulk gas and may reach the biosphere as gaseous species or dissolved in groundwater (Serco, 2011e). Previous experimental studies have measured the release of ^{14}C and tritium from samples of irradiated graphite. These have shown that 0.001% to 0.01% of the ^{14}C in graphite was released as volatile species in the short term (possibly within a few days). Another current study measured releases over a longer time scale of 14 months from 59 g of intact BEPO graphite, submerged in a pH 13 solution of sodium hydroxide. The data were also fitted to a first order kinetic function, which appeared to show the cumulative ^{14}C gas release reaching a plateau at around 0.005% of the total inventory. However, it is suspected that this does not necessarily represent an upper bound on the total release. It is possible that the data from this experiment show an "initial" release of relatively labile ^{14}C as volatile species. On much longer time scales, a larger fraction could be released at a slower rate but this could only be measured in very long-term experiments of several years, where the "initial" release has declined and is no longer a significant contribution. Tritium is also present in irradiated BEPO graphite and appears to be released mainly as tritiated water and so the amount collected is predominantly controlled by evaporation from the solution in which the graphite was immersed (Serco, 2011e).

These studies are using relatively young or new graphite samples to model leaching behaviour far in the future when the repository is inundated with groundwater intrusion. These studies do not account for the changes that may have occurred while the graphite is in storage in the intervening time. Radioactive particles and photons emitted can break covalent bonds and alter the nature of the lattice structure retaining the contaminants, contaminants can migrate to the surface through weeping induced by kinetic motion from radioactive decay and heating cooling cycles, or near-field microbial activity while in storage could facilitate transport of radionuclides to the material surface. These potential mechanisms should be well understood since they can affect the total source term available for immediate release upon groundwater intrusion (Lollar, 2011).

In another investigation, the radioactive inventory of graphite samples taken from stacks and sleeves of Russian water-graphite reactors were determined before and after 10 years of storage, as well as after additional mechanical treatment (milling) (Girke, et al., 2011). The comparison of the experimental data for the investigated samples led to the conclusion that the ^{14}C activity level after 10-year storage is consistent in the frame of the experimental error range (< 20%). On the other hand, the ^{14}C activity level in the graphite decreased drastically after milling (15-40%), with an average of 24% when placed in storage 10 years ago (Girke, et al., 2011). There were no comparisons of surface contamination levels on the blocks to evaluate potential desorption and accumulation on the surface with time. Further investigations are necessary to better understand the source terms that will be available at the surface of graphite in interim storage and in repositories and to characterise the desorption and release of radionuclides under the anticipated environmental conditions.

- Radionuclide mobility in concrete and steel

In contrast to graphite, the behaviour of mobile radionuclides in concrete appears to be better understood (BNL, 2003, 2004), although the infiltration and diffusion of contaminants in concrete cracks and fractures seems to be less well understood. The rate of ^{3}H diffusion in uncoated concrete and in concrete sealed with hydrophobic paints was recently studied. After exposure of varying durations under a constant tritiated (HTO) vapour pressure in an acrylic box, the amount of water-soluble HTO on/in the disks is determined using a technique of H_2O dissolution (Fukada, et al., 2012). Similarly, the rates of diffusion of ^{3}H and ^{137}C when contaminated concrete is placed in contact with water have been established for concrete samples with and without aggregate (Bucur, 2010). The ^{3}H diffusion coefficients were usually well correlated with the matrix porosity, with the lowest porosity sample exhibiting the lowest diffusion coefficients. Since tritium is a non-sorbing radionuclide, the small difference between pore-water diffusion coefficients experimentally obtained on two concrete wafers is explained by the higher porosity sample. In contrast, the cement-based materials provide low ^{137}Cs retention. The distribution coefficient obtained for the porous cement matrix (with sand as aggregate) was a little bit higher than that obtained on the non-aggregate cement matrix due to the additional exchange positions brought in the system by the aggregate. Desorption experiments showed that caesium sorption on concrete matrices is reversible. Usually sorption is considered reversible when desorption equilibrium is attained even within twice the time of the sorption equilibrium, and the total desorption is more than 75% of the amount sorbed. It was expected that ^{137}Cs would have smaller diffusion coefficients in concrete than tritium due to the larger size of the caesium cation and the presence of the retardation component, as determined from the sorption experiments. These experimental values obtained for tritium and caesium pore-water diffusion coefficients on concrete agree with values found in the literature. The low values of the diffusion coefficients on concrete matrices indicate that the radionuclide release from a repository would be extremely low in the first years when the concrete is intact (Bucur, 2010). Models are available for comparison to measured data as described above (Jacques, et al., 2010; Hou, 2010a; Dayal, 1994).

The corrosion of steel (Kim, Cho and Choi, 1992) and desorption of radionuclides have also been modelled and are being compared to experimental results (Eurajoki, et al., 2010; Hsieh, 2010).

Future suggested R&D – Modelling of mobile nuclide behaviour on different substrates

- ***Description*** – Provide a more robust and diversified set of laboratory and field measurements and mathematical interpretations that can be used to develop more accurate models of radionuclide interactions with decommissioning waste forms. Evaluate the chemical, physical and environmental mechanisms that influence radionuclide behaviour in waste forms and develop mechanistic first principle and descriptive empirical models that correlate well with field measurement observations.
- ***Objectives*** – To broaden the basis and understanding of the factors that influence radionuclide behaviour in waste forms to support development of decontamination methods and understand behaviour in the wide variety of interim storage and disposal environments that can be encountered internationally. To develop accurate models for use in evaluating decontamination and treatment strategies and the potential impacts and options for storage and disposal of contaminated materials.
- ***Desired deliverables*** – More robust and diverse laboratory and field measurements that better define the chemical, physical and environmental factors/mechanisms that influence the behaviour of radionuclides in decommissioning waste forms. More accurate mechanistic and empirical models whose predicted results correlate with field observations for decommissioning wastes in interim storage and disposal environments.

Developing an international approach or standard for estimating trace impurity levels in activated reactor alloys and concretes

Challenges

Before the development of the American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel Code (2003) trace element requirements specifically applicable to reactor vessels and internals, the design of reactor vessels and internals was based on criteria specific to each nuclear steam supply system (NSSS) vendor. Currently, the United States and others countries regard NUREG/CR-3474, *Long-Lived Activation Products in Reactor Materials*, completed in 1984 (Evans, et al.), as a default basis for evaluating elemental contaminant concentrations in reactor components of the current fleet of reactors. These trace element concentrations evaluated in the report were based on analysis of reactor vessel and core component steels, reactor bioshield concrete and rebar samples solicited from reactor component manufacturers and reactors under construction in 1984. As seen in the report's composition summary tables, the NUREG provided average values, ranges and standard deviations for elemental constituents in different steels. They serve as the basis for activation estimates even though the sample set was very limited. For instance, six nuclear power plants provided seven samples of reactor internals steel. Consequently, the data on cobalt concentrations is based on eight samples, which is not a statistically robust basis for the generation of ^{60}Co, which is the principle scaling nuclide (Evans, et al., 1984). The manufacturer, Westinghouse, provided an additional six samples. The analysis showed significant variation in elemental constituents based on this limited sample set. Without specific elemental composition data for the specific activated component being assessed, employing the range of values in the report can result in inaccuracies in assessed source terms that can result in disposal problems.

As noted in IAEA-TECDOC-1557 (2007a) for new reactors in the United States, the use of Subsection NB of the ASME Section III is required by 10 CFR 50 regulations promulgated by the NRC. Basically the same design basis applies for PWR reactor vessel internals in western countries throughout the world. The applicable standard are RCC-M (AFCEN, 1988; IAEA, 1999a) in France, and KTA 3204 (1998) in Germany. In Japan the applicable standards are the Ministry of Economy, Trade and Industry (METI) notification 501 (1980) and the Japan Society of Mechanical Engineers (JSME) Code on Design and Construction for Nuclear Power Plants, JSME SNC1-2001 (1991), which is based on the ASME Code. In the Russian Federation, the applicable codes and standards are listed later in Section 3.5 of IAEA-TECDOC-1119 (1999a). These codes and standards have been adopted in most other countries operating WWER reactors (IAEA, 1999a). The standards typically include chemical specifications for the elements C, Mn, Si, S, P Ni, Cr, Mo, Nb, Ci, Co, Cu, N and B.

Table 2.3: Other activation products potentially important to waste classification

Radionuclide	Half-life (yr)	Primary production mode(s)	Primary radiations
^{10}Be	1.60E+06	$^{10}B(n,p)^{10}Be$	556 keV beta
^{36}Cl	3.01E+05	$^{35}Cl(n,\gamma)^{36}Cl$	709 keV beta
^{108m}Ag	130	$^{107}Ag(n,\gamma)^{108m}Ag$	434 keV, 614 keV gamma and 723 keV beta
^{113m}Cd	14.1	$^{112}Cd(n,\gamma)^{113m}Cd$	590 keV beta
^{121m}Sn	55	$^{120}Sn(n,\gamma)^{121m}Sn$	354 keV beta; 26.3 keV X-ray

Further investigations on the trace element constituents of stainless steels, such as chlorine, niobium, samarium, technetium and selenium, in current reactor steels and new reactor construction are warranted. This will enable better evaluation of recycling, re-use and disposal scenarios and an understanding of the properties, production pathways, decay processes and relative biological importance of the radioactive activation products.

It will also provide the baseline data necessary to support activation analysis and evaluation of waste classification based on whatever standards are in place at the time of facility decommissioning.

It seems very important to collect a new set of certified material test reports for stainless steel and some nickel alloys (Inconel) used inside reactor core components and to re-assess the disposability of these materials in future reactors. This is especially timely in the United States case with regard to revising waste classification regulations and Greater-Than-Class C low-level waste disposal issues that are currently being considered (US DOE, 2011a). The goal should be to collect the data necessary and establish standards that will ensure that potential issues with long activation products are addressed.

Summary of current R&D developing an international approach or standard for estimating trace impurity levels in activated reactor alloys and concretes

- Activated metal scaling and cobalt concentrations in alloys

NUREG/CR-3474 (Evans, et al., 1984) is used as a basic reference for elemental cobalt in base metals of current reactors. NUREG/CR-4968 (Bedore, Levin and Tuite, 1987) notes that the industry uses two different characterisation methods for activated metals. The first uses a combination of gamma scanning, direct sampling, underwater radiation profiling and radiochemical analysis to determine radionuclide content. This approach is commonly used for ponds or spent fuel pool clean-ups where the materials being characterised are relatively accessible and can be sampled (Cline, 1993). Some direct sampling of reactor vessel internals is also being used for reactor internals characterisation (Oberhaeuser, 2012; EC-CND, n.d.b).

The second method uses activation analysis calculation or modelling based on historical neutron spectra and operating history in conjunction with confirmatory radiation surveys of the activated components. This approach is typically used for reactor internals and vessels, where significant disassembly and movement of large components underwater is required to access the materials, and waste classifications and planning are required far in advance of dismantling to support reactor internals/vessel segmentation and packaging (Cho, et al., 2011; Love, Pauley and Reid, 1995; BNL, 2007; Holden, Reciniello and Hu, 2004; Vinson, et al., 2010). A code such as the ANISN (ORNL, 2007; OECD/NEA, 2003) computer program is used to estimate neutron fluxes at various radial and axial locations in the vessel. Reactor coupon data that are periodically tested to monitor vessel embrittlement during the plant operation as well can be used with cross-section libraries such as BUGLE (ORNL, 1999a) to normalise the calculated fluxes to the specific reactor. A code such as ORIGEN (ORNL, 1999b; OECD/NEA, 2002) is then used to calculate the activation and depletion of radionuclides in components exposed to a neutron flux. Each component is irradiated based on the plant-specific operating histories using the appropriate flux as determined from the normalised ANISN transport models. The ORIGIN calculated radionuclide inventory is then usually normalised based upon radiation survey data on the reactor vessel or internals. MCNP is another code has been used to calculate activation levels in core components of various reactor designs, e.g. PWR, BWR, CANDU, VVER, V-230 and research reactors (Agosteo, *et. al.*, 2005; Tzika, Savidou and Stametelatos, 2007; Henderson, et al., 1997; Marcinkevičius and Plukis, 2012; Bouhaddane and Farkas, 2013; Cho, 2011).

Both the direct sample and the computer activation methods employ two distinct steps: *i)* the determination of ^{60}Co content; *ii)* the determination of scaling factors for hard-to-detect radionuclides. The accurate determination of ^{60}Co concentrations and the ratios of cobalt to hard-to-detect precursor trace constituents is critical since the activation product radionuclides (which affect waste classification) are scaled from ^{60}Co. This method relies on the relative content of elemental cobalt to the content of other elemental materials, such as nickel, niobium, nitrogen and molybdenum, to estimate scaling factors for hard-to-detect radionuclides (e.g. ^{14}C, ^{59}Ni, ^{63}Ni, ^{94}Nb, ^{36}Cl, ^{99}Tc) Except for nickel, all of these metals are usually present in trace quantities in most of the materials used for reactor hardware. Thus, the uncertainties arise from the unavailability of materials composition data for each batch of components and the variability of the data that is available (Kawata, et al., 2010).

The most highly activated components are the reactor internals. These components are constructed almost exclusively from 304 stainless steel and form the greater part of Greater-Than-Class C or intermediate- and high-level waste that is not suitable for shallow land disposal. This is a high nickel steel that owes its corrosion resistance to that element. Consequently, nickel concentrations are tightly controlled in production. In most cases, the activation product ^{63}Ni with a 100-year half-life overwhelmingly controls the waste classification of activated steels. One particularly troubling aspect of using ^{60}Co as a scaling nuclide for hard-to-detect nuclides such as ^{63}Ni is that the range of potential cobalt concentrations is based on NUREG/CR-3474 data (Evans, et al., 1984). Table 2.4 excerpts cobalt and nickel data from NUREG/CR-4968, which evaluated the uncertainties in irradiated hardware characterisation (Bedore, Levin and Tuite, 1987). For cobalt as an example, the Table 2.4 range from maximum to minimum concentrations is three times greater than the potential range of nickel concentrations in 304 stainless steel. The NUREG also contains the ranges of other elements such as Fe, N, Nb and Mo.

Table 2.4: NUREG/CR-4968 comparison of nickel and cobalt ranges in reactor internals alloys

Element	Min	Max	Ratio	Min	Max	Ratio
Stainless steels		**304**			**316**	
Ni (%)	8.8	11	1.25	12.5	13.2	1.06
Co (ppm)	750	2600	3.5	1300	1600	1.2
Inconels		**600**			**700**	
Ni (%)	58	72	1.24	52	72	1.38
Co (ppm)	400	700	1.8	400	700	1.8
Zircaloy						
Ni (%)	0.02	0.5	25.00			
Co (ppm)	10					

In the absence of sample data that provides the radionuclide concentrations on a particular component, activation analyses tend to be conservative, using concentrations at the higher end of the potential range of concentrations. Conservative assumptions can lead to overestimates of the ^{60}Co concentrations based on activation modelling if the actual cobalt concentration in the component under consideration is at the lower end of the range. Activation analyses are typically base-lined against component dose rates, which are driven by ^{60}Co. For cases in the second scenario it would be unwise to scale down hard-to-detect nuclide concentrations, such as ^{63}Ni, based on the lower observed ^{60}Co concentrations, since the range of potential nickel concentrations are much tighter than cobalt and nickel is tightly controlled in production of the alloy. In addition, the neutron flux and core history data are monitored to control fuel burn-up, and it is unlikely that a large discrepancy in calculated versus observed ^{60}Co is due to errors in the activation analyses, which are based on verified computer codes and benchmarked against reactor vessel coupon data. It is much more likely that the ^{63}Ni concentrations which drive the waste classification are accurate and that the ^{60}Co levels calculated were too high. In the absence of direct sampling, the use of chemical constituent concentration ranges has a direct impact on activated waste classifications. Even sampling and measurement of accessible components or the analysis vessel coupons, used to monitor embrittlment, are of limited value in determining the source terms of other components since they are not necessarily representative of the chemical constituents in the batches of steel used to make the other components.

An additional difficulty with using ^{60}Co to scale other hard-to-detect radionuclides in activated metals is the discrepancy in half-lives with other nuclides of concern. Often in the first scenario, the activated materials, such as those in a spent fuel pool, are accessible for sampling but it is not feasible to sample each and every item requiring characterisation. It is also frequently the case that records are not available for each item to document how long the item was irradiated (effective full-power years) in the core or when it was removed from the core (cooling time). It is often necessary to conduct a mini-historical site investigation to base these estimates on records of plant modifications and employee recollections in order to determine the "vintage" of the material being assessed (Cline, 1993). Since ^{60}Co decays with a five-year half-life and ^{63}Ni decays with a 100-year half-life, a sample obtained from one activated item is not necessarily representative of the ^{60}Co to hard-to-detect nuclides in a similar item unless the removal times from the core are known to enable decay corrections of the mix. Determining the removal time from the core for items in a spent fuel pool is often difficult at operating facilities and not possible at those that have been in SAFSTOR for prolonged periods. Previous laboratory analytical work has shown that two samples from the same component in different locations can contain differing quantities of trace elements. Variations by an order of magnitude have been measured. In addition, samples may not be representative of the full volume distribution. Samples may have higher or lower activities if they are taken from surface metal. This must be considered during the sampling process planning, and later during the comparison to the calculated estimates (Cho, et al., 2011). As summarised in the DOE irradiated hardware direct measurement guidance, "*The assumption in selecting a representative sample from a collection of components having the same 'vintage' is that all components of the same type and purchased at the same time were fabricated from the same batch of alloy material*." The accuracy of this assumption is not known and is difficult to ascertain. Decay knowledge is necessary to correct the dose-to-curie factor for the relative contribution of ^{60}Co to the measured dose if the adjustment factor is important. Accuracy is directly related to the size of the adjustment and the radioactive decay (Mancini, et al., 1994).

A better understanding of the range and statistical distributions of cobalt and trace contaminants in irradiated hardware [see Appendix A of (Cho, 2011)] would aid in selecting concentrations at reasonable confidence intervals for Scenarios 1 and 2 and aid in understanding the uncertainties associated with scaled nuclide concentrations. An analysis of the Fugen Nuclear Power Plant chemical composition of construction materials was carried out in Japan and compared to the NUREG/CR 3474 data (Evans, et al., 1984). Translation of this study into other languages would benefit the international community.

- Implications for advanced materials assessments

There appears to be a need for more focus on waste disposal implications for evaluation of advanced reactor materials for fission and fusion reactor designs. The current focus appears to be on material properties related to operational performance without consideration of decommissioning and waste disposal issues (US DOE, 2010b; Nanstad and Odette, 2011). There is the potential to create new alloys with chemical compositions that will create waste disposal issues for radionuclides not considered in current waste classification requirements. NUREG/CR-6567, *Low-Level Radioactive Waste Classification, Characterization, and Assessment: Waste Streams and Neutron-Activated Metals*, was completed in August 2000 (Robertson, et al.). This study focused on identifying and characterising a group of very long-lived radionuclides that are not specified in the United States waste classification regulations (e.g. 10 CFR Part 61) but which are present in significant concentrations in various types of LLW materials generated at commercial nuclear power stations. The concentrations of ^{10}Be, ^{36}Cl, ^{93}Mo, ^{93m}Nb, ^{108m}Ag, ^{113m}Cd and ^{121m}Sn (as well as the specified 10 CFR Part 61 radionuclides) have been measured in a variety of neutron-activated metal and spent primary demineralisation resin LLW samples obtained from United States nuclear power stations. Of this group of radionuclides, the ^{10}Be, ^{36}Cl and ^{108m}Ag appear to be present in some types of LLW materials in sufficient quantities to warrant further investigations to better assess their radiological and environmental impacts associated with LLW disposal. Interestingly, in the context of the previous section, the report recommended that further studies be focused on: *i)* providing an accurate assessment of the total quantities of these radionuclides in LLW from these sources; *ii)* determining the leaching characteristics of these LLW materials; *iii)* determining the migration behaviour and environmental pathways of the radionuclides upon release from LLW disposal facilities; *iv)* providing performance assessment modellers with the necessary radiological/geochemical information to better predict the potential impacts from disposal of this group of radionuclides (Robertson, et al., 2000).

Countries embarking more vigorously on new reactor construction are evaluating and pursuing chemical specification requirements and the production controls to meet them (Narayana, 2011). Evaluation of radionuclides potentially significant to waste disposal could be integrated into ongoing evaluations of new materials (Hoblit, et al., 2011; Herman, 2011; Herman and Trkov, 2009; Lee, et al., 2010; Ren, Naus and Oland, 2010; Mortazavi, et al., 2010).

Future suggested R&D – Developing an international approach or standard for estimating trace impurity levels in activated reactor alloys and concretes

- ***Description –*** Provide a more robust and diversified set of chemical specifications for activated alloys and concretes for reactors. Collect data, sample and measure chemical concentrations of elements that result in long-lived activation products that are of potential importance to classification and evaluations of waste storage and disposal. Integrate assay of important trace elements into the assay and testing of materials for new reactors, and develop a methodology to store or archive the data for use at the time of facility decommissioning.
- ***Objectives –*** To develop chemical specification data for activated materials with a robust statistical basis for use in activation analyses of current and new reactors. The data should be collected and categorised by material/alloy type, manufacturing date, manufacturing country of origin and component country of use in order to aid in activation analysis and safety case evaluations internationally.
- ***Desired deliverables*** – Definition of reactor material trace contaminants and activation nuclides of potential importance to waste classification and storage and disposal evaluations. Collection of historical data for chemical specifications on activated materials. Measurement of important trace contaminants in current reactor and next generation materials. Integration of trace constituent data and radioactive waste implications into next generation material testing in order to develop alloy/material chemical specifications that minimise waste handling and disposal impacts.

Suggested areas of future collaboration

A number of issues and R&D areas appear to be common to national programmes. Potential areas for collaboration could include:

- developing an international approach and/or standard for statistical sampling (representativeness, grid density, defining an acceptable level of uncertainty);
- method and hardware to develop characterisation of contamination intrusion along concrete cracks;

- technologies for rapid alpha and beta non-destructive measurements on structures before dismantling, especially for difficult-to-access structures;
- international approach for scaling factors between easy- and hard-to-measure nuclides;
- developing an international approach or standard for estimating the level of impurities in metals and concretes, especially for new reactors.

3. Technologies for segmentation and dismantling

Theme overview

The objectives under this theme can be summarised as the need to improve efficiency and safety, and to reduce cost. While technologies exist that have been successfully deployed, there is potential for these technologies to be improved or new technologies to be developed – in some instances by using good practices established in other fields. Successful delivery of these technologies could reduce worker exposure and improve efficiency through the deployment of robotics and/or remote technologies, and also potentially reduce the resultant wastes generated by conventional technologies.

The survey responses under this theme showed some commonality and some contrast to issues relating to technologies for segmentation and dismantling. For example, a number of responses indicate the desire to develop thermal techniques for use in nuclear applications in order to benefit from improved cutting efficiencies, while other responses seem to show concern for the safety hazards of using thermal methods in nuclear environments and plan to execute decommissioning using well established mechanical methods.

The majority of the survey responses identified a need to develop remote deployment methods for cutting techniques, as current remote methods do not seem to provide the flexibility required for complex decommissioning operations. Although the field of robotics is established, currently there is little underpinning R&D in the development of robotics and remote autonomous systems relevant to nuclear applications.

A number of responses identified a need to develop processes for addressing secondary decommissioning effluents while other respondents felt this could be addressed using existing facilities or by using techniques that minimise/avoid generating secondary wastes.

Table 3.1: Guidance documents for segmentation and dismantling

Facility type	Phase	Region	Document
All types	Decommissioning remote milling and shearing	International	*Application of Non-Nuclear Robotics to Nuclear Industry Decommissioning*, EPRI (2004a)
All types	Decommissioning remote cutting and handling	International	*Application Procedures for Technical Catalogue Proposals and Workshop for Machine/Equipment Development for Fuel Debris Removal Preparation*, METI (2012a)
Power reactors	Decommissioning reactor internals segmentation	International	*Decommissioning: Reactor Pressure Vessel Internals Segmentation Final Report*, EPRI (2001a)
Power reactors	Decommissioning reactor internals segmentation	International	*Reactor Internals Segmentation Experience Report: Detailed Experiences 1993-2006, Final Report*, EPRI (2007)
Power reactors	Decommissioning reactor internals segmentation	International	*Rancho Seco Reactor Vessel Segmentation Experience Report*, EPRI (2008c)

Table 3.1: Guidance documents for segmentation and dismantling (*cont'd*)

Facility type	Phase	Region	Document
Power reactors	Decommissioning reactor internals segmentation	International	*Experience with Reactor Internals Segmentation at US Power Plants*, Wood and Naughton (2007)
Power reactors	Decommissioning reactor internals segmentation	International	*Trojan Nuclear Power Plant Reactor Vessel and Internals Removal*, EPRI (2000c)
Power reactors	Decommissioning reactor internals segmentation	International	*Decommissioning Reactor Pressure Vessel Internals Segmentation, Final Report*, EPRI (2001a)
Power reactors	Decommissioning reactor internals segmentation	International	*Decommissioning San Onofre Nuclear Generating Station Unit 1 (SONGS-1)*, EPRI (2005b)
Power reactors	Decommissioning reactor internals segmentation	International	*Connecticut Yankee Decommissioning Experience Report*, EPRI (2006)
Power reactors	Decommissioning reactor internals segmentation	International	*San Onofre Nuclear Generating Station – Unit 1 Decommissioning Experience Report*, EPRI (2008d)
Power reactors	Decommissioning reactor internals segmentation	International	*Recent United States and International Experiences in Reactor Vessel and Internals Segmentation*, EPRI (2011)
Power reactors	Decommissioning large component removal	International	*Trojan PWR Decommissioning: Large Component Removal Project*, EPRI (1997a)
Power reactors	Decommissioning dismantlement	International	*Yankee Rowe Decommissioning Experience Record: Volume 1*, EPRI (1997b)
Power reactors	Decommissioning dismantlement	International	*Yankee Rowe Decommissioning Experience Record: Volume 2*, EPRI (1998)
All types	Decommissioning dismantlement	International	*State-of-the-Art Technology for Decontamination and Dismantling of Nuclear Facilities*, IAEA (1999d)
All types	Decommissioning dismantlement	International	*Construction Technologies for Nuclear Power Plants*, IAEA (2011a)
Power reactors	Decommissioning dismantlement	International	*Heavy Component Replacement in Nuclear Power Plants: Experience and Guidelines*, IAEA (2008a)

Summary of current practices and guidance

With respect to segmentation and dismantling technologies specifically for reactor vessels and internals, steam generators, pressurisers and reactor coolant pumps and associated piping, the current practices have by and large been developmental, using off-the-shelf equipment and adapting it to decommissioning. Each application of existing technologies has gained the benefit of previous experience but has uncovered new technological difficulties to be overcome. This evolution of the technologies has built confidence within the specialty contractors to apply their expertise to larger and more difficult configurations and constraints.

Reactor vessel and internals segmentation

The current technologies for segmentation include plasma arc cutting, electric discharge machining (EDM) and metal disintegration machining (MDM), high pressure abrasive grit cutting and mechanical cutting (milling cutters, broaches, end mills), and diamond wire saws. Each of these methods has undergone the development phase of adapting to each new configuration and challenge, but there remain unresolved problems that have reoccurred with each application. The basic characteristics of each of these methods can be summarised in Table 3.2.

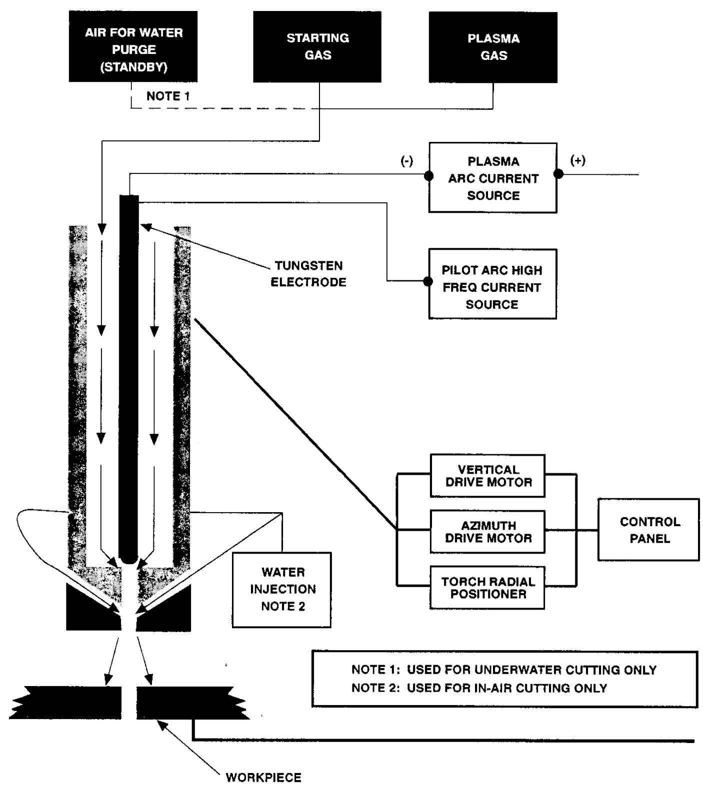
Table 3.2: Comparison of cutting technologies

Technology	Cutting depth, mm	Cutting speed, cm^2/min	Kerf size, mm	Waste generated
Plasma arc	100	129	6-25	Vaporised dross; difficult to control
EDM/MDM	80	0.1	6-8	Fine particles and aerosols
High pressure abrasive grit	160	130	5-10	Large quantity of grit – 2-4 kg/min, large quantity of water – 19-30 litres/min
Milling cutters	200	Slow	6	Swarf is in the form of filings, chips and turnings
Diamond wire saw	200	Slow	8	Swarf is fine filings

Plasma arc torch

The plasma arc cutting technique is based on the establishment of a direct current arc between a tungsten electrode and any conducting metal (the workpiece). The arc is established in a gas, or gas mixture that flows through a constricting orifice in the torch nozzle to the workpiece. The constricting effect of the orifice on both the gas and the arc results in very high current densities and high temperatures in the stream (10 000-24 000 K). Figure 3.1 illustrates the basic components of the plasma arc torch.

Figure 3.1: Schematic of plasma arc torch



The stream of plasma consists of positively charged ions and free electrons. The plasma is ejected from the torch nozzle at a very high velocity and blows the molten metal away. A typical cut starts at the metal edge, and a through cut is made in a single pass by moving the torch at a fixed rate of speed in the direction of the cut and at a fixed nozzle spacing relative to the workpiece.

The plasma arc system requires a direct current power supply of up to 1 000 amperes. An automatic plasma arc system would include torch positioning equipment; torch travel system; starting gas (usually argon) and cutting gases (air, nitrogen or carbon dioxide); pilot arc high-frequency power supply; cutting arc power supply; and associated gas flow, arc and mechanical travel controls.

The plasma arc torch was used most recently at the Yankee Rowe NPP dismantling of the reactor vessel internals. The torch generated fine particulates that were difficult to control and the heat from the torch caused thermal currents in the water to rise, bringing the high dose particulates to the surface and exposing the workers (EPRI, 2007). Thick shielding had to be installed on the cutting platform to protect the workers. In addition gas bubbles in the water created high airborne radioactivity levels (EPRI, 2001a). The dross collection system did not reach the surface, allowing the fine particles to be released to the cavity and thus requiring additional filters to be used to reduce the activity of the cavity water (EPRI, 2001a). The maximum water depth for cutting with plasma arc torches is about 10.5 m (35 ft), as the hydrostatic pressure affects the gas flow. The plasma arc can only cut through a single thickness of material, as it must maintain the arc to sustain cutting (EPRI, 2001a, 2007).

Electric and metal discharge machining

Electric discharge machining (EDM) uses an electrode (typically graphite) positioned at a fixed distance (gap) above the workpiece. The distance controls the energy at the cutting surface and is adjustable by the system operator as a function of the voltage across the gap. The electrode and the workpiece are submerged in a dielectic field. As the electrode is energised, ion columns are established between the electrode and the workpiece and controlled arcing occurs across the gap, resulting in localised heating. The cutting rate is proportional to the amount of energy, and the frequency controls the resulting surface finish. The thermal expansion of the locally heated area causes small molten particles to lift off of the surface. Flushing of the dielectric in the cutting area results in the resolidification of these particles and washes them away from the surface of the workpiece. The EDM method can make penetrations of virtually any shape by using electrodes fabricated in the geometry of the desired hole. Since the electrode never contacts the workpiece, this drastically reduces the cutting equipment's mechanical strength and weight requirements. In addition, although EDM requires a fairly complex electronic package, it allows for finite operator manipulation of electrical parameters, which subsequently control such variables as cut rate, surface finish and electrode wear.

Metal disintegration machining (MDM) is similar to EDM but requires the use of a constant-current power supply and a vibrating electrode to generate the cutting pulses. As the MDM electrode is brought close to the workpiece, an ion column is formed and allows current to pass through the gap. This causes very high energy at the instant before the electrode makes physical contact with the surface. Since constant current is used, the electronics needed are fairly simple compared to EDM, but the degree of control of cut rate, surface finish and electrode wear is substantially less. Yet, since the electrode contact the workpiece, resulting reactionary machining forces require a sturdier mechanical delivery system. In general, MDM is faster and less precise than EDM.

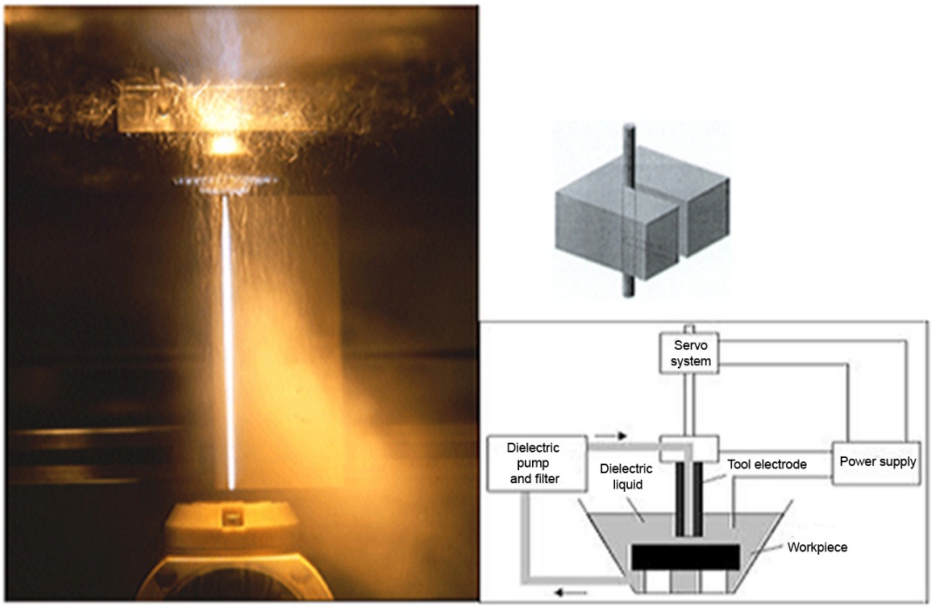
The faster cutting speed of the MDM process (compared to the EDM process) makes it more suitable for reactor vessel internals cutting. With the MDM process, a power supply of up to 20 KVA is used. The electrode in the machine vibrates up and down 3 600 times a minute. Each time the electrode touches the piece to be burned, an arc is struck. The arc has a point of contact temperature of approximately 3 100°C. A constant supply of fresh water is pumped down through the electrode, causing the molten metal to thermally break down; at the same time the water flushes the thermally shocked metal back out the discharge hole. The discharged pieces are normally less than 10 microns in size (less than a grain of sand). The water also acts as a heat exchanger, i.e. the only portion to get hot is the small area that the tip of the electrode contacts, unlike drilling where heat from the bit transfers to the matted portion. The MDM process maintains everything at ambient temperature, except what needs to be removed.

Although the cutting speeds are not as high as other technologies, the minimal generation of secondary waste is an advantage over other technologies. This process is generally used when high precision cuts are needed. Figure 3.2 shows MDM cutting within a gas control hood.

Figure 3.2: MDM cutting under a gas control hood

EDM and MDM were also used at Yankee Rowe (EPRI, 2001a, 2007), but its slow cutting speed limited its application. Figure 3.3 shows the principle of wire EDM, where a wire from a spool is fed through the workpiece submerged in deionised water.

Figure 3.3: EDM cutting

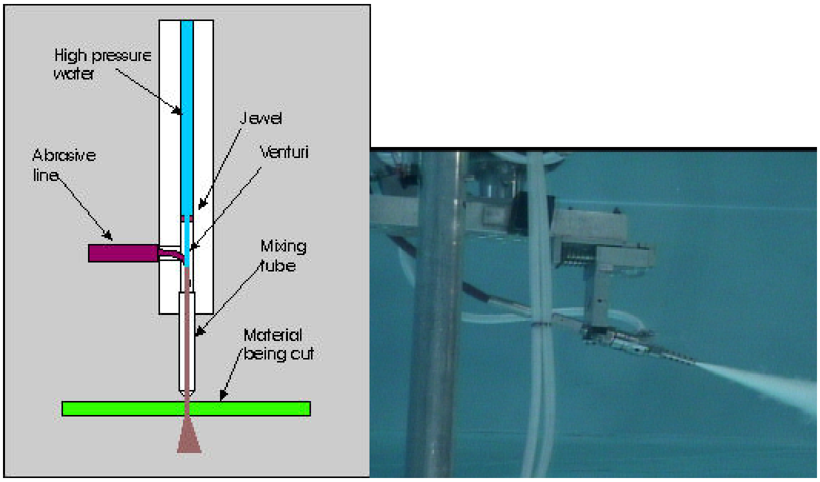


High pressure abrasive grit cutting

The abrasive water jet cutting technique involves the use of highly pressurised water (as high as 3 900 kg/cm^2). The water is pressurised by a hydraulically-driven intensifier pump. The water flows through a chamber where it is mixed with an abrasive, the most common being crushed garnet. Steel shot is also used. This mixture of water and abrasive is then forced through a wear-resistant nozzle with a small orifice that focuses the abrasive jet stream at the workpiece. The pressurised jet stream exits the orifice at extremely high velocities, producing erosion that yields a clean cut with a narrow kerf through thin materials. Thicker materials cause the jet stream to diverge, causing a wider kerf and reducing the effectiveness of the cutting action.

High pressure abrasive grit (HPAG) cutting was used at the Connecticut Yankee NPP (EPRI, 2001a, 2007), but the large quantity of grit was difficult to control. The entire cutting pool was covered with secondary waste grit, and a remotely operated Grant (trade name) machine had to be installed to clean up the pools. Figure 3.4 shows the basic components of an abrasive water jet nozzle and an underwater photograph of an HPAG cutter.

Figure 3.4: HPAG cutter



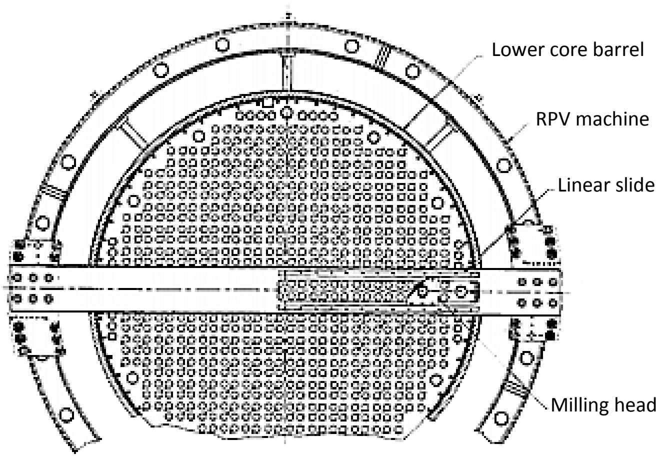
Maine Yankee and San Onofre 1 also used abrasive water jet technology but refined the dross collection system and accomplished the work with greater success than Connecticut Yankee (EPRI, 2001a, 2007). The Rancho Seco reactor vessel was segmented using abrasive water jet after the internals were segmented (EPRI, 2008c).

Milling cutters

A mechanical cutter system consists of a self-propelled circular milling machine cutter mounted on a track attached to a specially designed support fixture. It is powered pneumatically, hydraulically or electrically. The fixture is attached to the component to be cut and the cutter advanced into the workpiece. The very large reaction forces of the cutter require individual support tracks for each type of cut. These fixtures can be horizontal rings, vertical tracks or radial arms. Figure 3.5 shows one of the mechanical cutter tools installed on top of a reactor vessel.

Milling cutters were used at Rancho Seco NPP but their slow cutting speed and difficulty in bit and blade change-out prolonged the cutting programme. Big Rock Point also used these mechanical cutters, although the amount of cutting was limited (EPRI, 2001a, 2007). Millstone Point Unit 1 NPP used milling cutters, but again progress was slow. Zion Station Units 1 and 2 and Humboldt Bay Unit 3 are currently using milling cutters, but it is taking much longer than anticipated. In addition to blade and bit change-outs, difficulties performing minor maintenance such as replacing cables and insufficiently robust cutting platforms and delivery mechanisms are thought to be contributing to the slower-than-expected progress.

Figure 3.5: Circular milling cutter



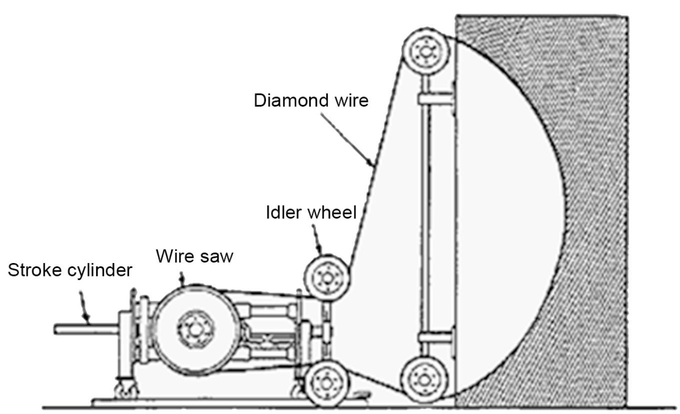
Diamond wire saws

In operation, the diamond-embedded wire is driven and guided by a pulley system. The guide wheels or pulleys are mounted near the structure to be removed and generally are no larger than 4.9 metres in diameter. The power unit can be placed several yards from the work area. The pulley system allows for the removal of heavily reinforced concrete or steel components where the work space is limited or in areas that pose a safety hazard for the operator. The length of the wire is virtually unlimited – any size cut can be made.

The "typical" wall cut is illustrated in Figure 3.6. A small hole is drilled at each end of the cut to be made. The wire is passed through the two holes and then coupled together. It is placed on the drive wheel and around idler wheels that guide the wire. Water is used to cool the wire and to wash away the swarf created by the cutting operation. Wire tension is maintained via a hydraulic "stroke" cylinder that pulls the main drive wheel along its sliding carriage assembly. The main drive assembly is a simple flywheel that is either hydraulically or electrically driven.

Diamond wire saws have not been used on reactor vessel internals, but were used on the Rancho Seco steam generators to size reduce them for transport and disposal.

Figure 3.6: Diamond wire saw

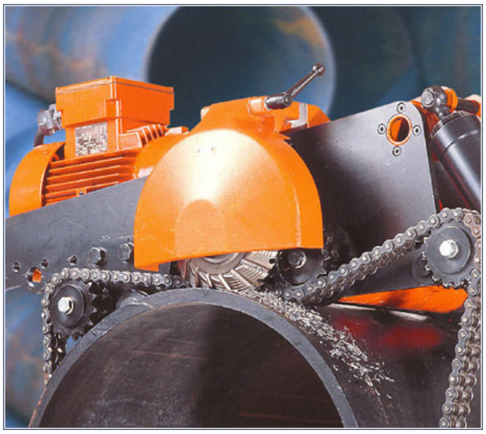


Other large components

Conventional cutting technologies

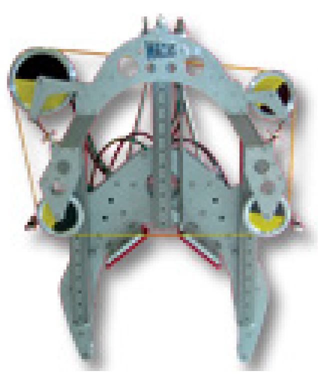
With respect to segmentation of other large components, such as steam generators, pressurisers, reactor coolant pumps and large diameter reactor coolant piping, the technologies can be considered state-of-the-art and mature with little new research or development necessary. For example, in the United States virtually every pressurised water reactor (PWR) experienced steam generator leakage problems, necessitating complete removal and replacement of the generators using off-the-shelf mechanical cutting equipment. The equipment includes milling cutters that attach circumferentially around the pipe and cut and prepare the pipe for re-welding. Figure 3.7 shows a milling cutter for pipe cutting.

Figure 3.7: Pipe milling cutter



Recently, diamond wire saws were adapted from underwater oil drilling rigs that can be attached to the pipe and cut remotely from a safe distance. Figure 3.8 shows a diamond wire saw attachment for a Brokk.

Figure 3.8: Brokk attachment for pipe cutting



Diamond wire sawing was used at the Rancho Seco NPP in California to cut the steam generators in half, facilitating removal and transport to the disposal site. Accordingly, the level of difficulty and challenge is small compared to reactor vessel and internals segmentation. Figure 3.9 shows a steam generator at Rancho Seco being cut with a diamond wire saw.

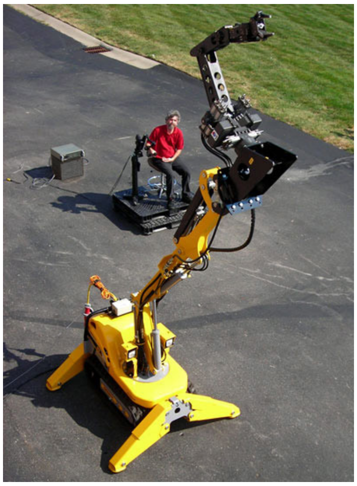
Figure 3.9: Rancho Seco steam generator cutting with a diamond wire saw

Nevertheless, new technologies are being adapted from the non-nuclear industry, such as laser cutting for application to pipe cutting and concrete scabbling. Lasers are now fully capable of cutting through 5 cm (2 inch) thick steel plates and have undergone successful demonstration testing at the West Valley, NY Reprocessing Center.

Robotics

The use of robotics has greatly reduced exposures to workers by having the operator control the device from a safe distance or even another building. Brokk has developed a series of electrically-driven, hydraulic machines that can navigate through narrow doorways, down staircases and into very high dose areas. They are being used at Fukushima to survey, videotape and remove debris from the very high exposure areas of the damaged reactors. Brokk has added manipulator end effectors [similar to a hot cell manipulator with load carrying capacities exceeding 75 kg (165 pounds)]. The application of robotics to cutting pipe, electrical conduit, cable trays, structural steel and scarifying (scabbling) concrete are virtually limitless. Figure 3.10 shows a typical Brokk machine with a manipulator attachment. Other attachments are available for sawing, scabbling, crushing, bucket loading and concrete breaking.

Figure 3.10: Brokk machine



With respect to the use of robotics for reactor vessel and internals segmentation, each cutting technology selected drove the design of the required robot to position and cut the materials. Such considerations as the weight of the cutting head, cutting reaction force, articulation and degrees of freedom, positioning accuracy and repeatability, access limitations, water depth required, corrosivity of the cutting environment, and ease of use/training of operators all effect the design of the required robot. While there may be some advantage in designing an all-purpose robot, the cost may be prohibitive if only a simple manual tool will do the job. Chapter 4 discusses some of the robotic technologies that are currently in use.

Segmentation, secondary waste generation and collection

The current techniques for collection of the secondary wastes generated during underwater segmentation of internals have been a major source for cost over-runs, exposures to workers, waste packaging and disposal. Experience at Yankee Rowe with plasma arc cutting of the internals caused fine particulate (aerosols) to be generated in the cutting pool that were carried upward with the cutting gasses and the thermal currents from the high-temperature torch head. The particulate initially caused excessive personnel exposures until thick shielding could be installed on the cutting platform. The particulate spread over most of the surface of the cutting pool, requiring extensive underwater vacuuming after the cutting operations. The vacuum filters added to the secondary waste volumes that had to be disposed of as Class B, C and Greater-Than-Class C (intermediate) level wastes.

Connecticut Yankee experienced a similar but more extensive problem with the high-pressure abrasive grit (HPAG) cutting technology used for its vessel internals. The HPAG cutting head generated as much as 4 kg (8 lbs) of grit per minute of cutting time, which was mixed with highly activated swarf particulates and spread over the entire cutting pool. Post-cutting clean-up required a robotic arm system to be installed to vacuum the particulate and intermediate-level wastes. The amount of grit required three additional fuel-sized dry storage casks to be purchased to store the wastes until a federal repository was available.

Maine Yankee had much of the same experience as Connecticut Yankee using the HPAG cutter but required the cutting subcontractor to stop work and re-design the grit collection system. The post-cutting clean-up still required underwater vacuuming to clean up the uncaptured wastes.

San Onofre Unit 1 had the same experience as Connecticut Yankee with the HPAG cutting technique.

These experiences point to the need for research and development of cutting technologies that minimise the generation of secondary wastes, and for improved secondary waste collection and filtration methods.

Summary of challenges and R&D needs

Industrial safety is a major issue in D&D projects because many current technologies require hands-on labour in hazardous areas. The Task Group found that most of these technologies are labour intensive and time consuming, and therefore expensive. The hands-on nature of current technologies risks exposing workers to radiation, hazardous materials and industrial hazards. The major opportunities for reducing risks to workers lie in development of intelligent remote systems (robots) that can substitute for human workers in hazardous areas. The Task Group recommends basic research toward creating intelligent remote systems that can adapt to a variety of tasks and be readily assembled from standardised modules, with special emphasis on actuators, universal operational software and virtual presence.

Some of the challenges identified by the WPDD under this theme include:

- Contamination levels dictate that some large components (reactor vessels, steam generators, evaporators, vitrification rigs) will need to be size-reduced to fit into relatively small waste containers for safe storage/disposal.
- The large size and/or thickness, complex construction and inaccessibility of some components will make *in situ* size reduction challenging, while removal off site for size reduction is also challenging for large items.
- Thermal cutting methods provide efficient cutting capability relative to mechanical methods. However, there is a reluctance to use these methods in some countries due to safety concerns (including generation of noxious gases and fire risk). They also place higher demands on filtration systems (generating more used filters).
- Remote/robotic systems improve worker safety (in high radiation environments/ alpha-contaminated environments); however, they take more time to deploy, often need manual set-up, and often do not have the required flexibility for segmentation tasks or the ability to secure the item being cut, and remote underwater cutting needs further development.
- Secondary wastes resulting from decommissioning activities (such as concrete dust, decontamination liquids, resins from decontamination systems) need to be treated for disposal. This can be overcome in some instances where significant secondary waste treatment infrastructure is available on site.

Suggested additional research and development

Improvements in efficiency by use of remote systems and/or innovative technologies

Challenges

Because robotic and intelligent machines (RIM) cross-cut almost all of the US DOE's programmes, the DOE has laid out long-range plans for developing this technology in a RIM roadmap (DeGregory, et al., 2001). Technology roadmaps are planning documents that the DOE uses to call attention to future needs for development in technology, provide a structure for organising technology forecasts and programmes in order to avoid gaps or overlaps, and communicate needs and opportunities throughout the R&D community. For EM activities, the roadmap lays out ambitious goals for RIM, which include:

- increasing productivity by 300%;
- reducing personnel exposure by 90%;
- reducing secondary waste by 75%.

The DOE's Deactivation Decommissioning Focus Area (DDFA) estimates that 30% of its needs include RIM requirements. Facility D&D presents workplace hazards that are unique among EM's challenges. Most D&D baseline technologies require that workers routinely enter areas with radiation and many other industrial safety hazards and perform hands-on work with powerful and heavy equipment, including cutting devices that can instantly penetrate protective clothing. This routine work includes sampling (for characterisation), decontaminating and eventually dismantling the barriers that were originally constructed to protect workers from radioactivity and toxic chemicals. DOE facilities are massive: they are often crowded with complex, heavy equipment and in many cases details of how the equipment was designed and operated have been lost. The reality of nuclear facility D&D is that the physical tasks are unstructured (not repetitive) and involve a wide variety of highly contaminated components (e.g. piping, valves, wiring, tanks). When performed directly by human workers this work represents a significant safety risk and a high cost in terms of resources and time. The first goal of remote systems technology is to remove the workers from harm's way, which dramatically improves safety. The second goal is to increase productivity and reduce costs and project schedules, all of which would make D&D more manageable. Considering the present time line for this D&D work, there is at least a decade available during which an accelerated science-based development programme could be pursued to revolutionise the technology to meet the goals of the RIM roadmap.

Criteria-based decision making is the essence of intelligence in robotic systems. Today's control of robotic devices is derived from techniques developed during World War II in which control is linear (based only on the difference between two measured parameters). A robot capable of mimicking human adaptability, however, would require a non-linear control system in terms of many highly coupled parameters corresponding to the physical features that accurately represent performance of the task. The criteria-based software could be universal in the same sense that operating systems on microcomputers are universal – one system supports many different applications (Sturzenbecker, et al., 2000). In the initial planning and characterisation phases of D&D work, workers often must enter an area of high radiation and contamination that is also congested with left-in-place equipment and materials for which removal inevitably involves physical stress (fatigue) and the potential for personal injury. Virtual reality systems could allow workers to perform essential survey and decision-making functions from a remote location, thus enhancing their safety and productivity. Advances in the state-of-the-art as, for example, in deep sea exploration, could improve overall system performance by providing force feedback, remote vision, collision avoidance and radiation-resistant sensor technology.

Here the need is to make dismantling and segregation faster, either by the adaptation of existing remote systems, development of improved remote systems or by the use of other technologies.

There are a number of facilities where human access will not be possible so the flexibility in operation will be important. The identified needs are:

- segmentation of highly irradiated and thick vessel walls and piping in inaccessible areas of hot cells is a common need (or in some cases a need not to segment);
- how to achieve *in situ* segmentation where the location is difficult to access;
- the need to reduce robot size in order to deploy through small access ports;
- development of multi-purpose, adaptable remote handled arms;
- the adaptability of remote systems to difficult geometries, inaccessible areas and inhospitable environments (high radiation, deteriorated structures);
- development and deployment of novel cutting technologies such as laser and plasma cutting;
- versatility of systems, increase of load factor during operation.

Summary of current R&D projects – Improvements in efficiency through the use of remote systems and/or innovative technologies

- Remote system using force feedback

A remote system using force feedback, and the coupling of a force feedback system with a crane to avoid a need for second robotic arm for cutting operations, is discussed in Chapter 6 in the subsection entitled *CEA LIST computer-aided teleoperation.*

- Remote laser cutting in air/water

After more than 15 years of R&D, the CEA has developed and is now testing a robotic arm, "Maestro", with a high thickness laser cutter capability underwater and in air (EC-CND, 2008a, 2008b; Behar, 2012). To minimise human intervention in hostile environments, this arm is outfitted with hardware for underwater exploration and sampling.

Figure 3.11: CEA robot Maestro with end effectors

In 2010, the CEA signed a licensing agreement with Cybernetix that will allow Maestro to be used for decommissioning projects planned in 2014 at the pilot plant at Marcoule (APM) and in 2015 in the Fontenay-aux-Roses radiochemistry laboratory. An additional innovative technology, an underwater laser cutting head (100 mm) is being developed for the dismantling of large components and thick radiating, very complex geometry, such as a reactor vessel. Another technique used to decontaminate surfaces, the Aspilaser, is being tested by the CEA for paint stripping in Fontenay-aux-Roses and Cadarache (SDMS Technologies, 2011). The hot cell building, located adjacent to the experimental ITER Tokomak fusion reactor, is also equipped with a laser cutter operated robotically from CEA's virtual reality room (Dulon, 2009).

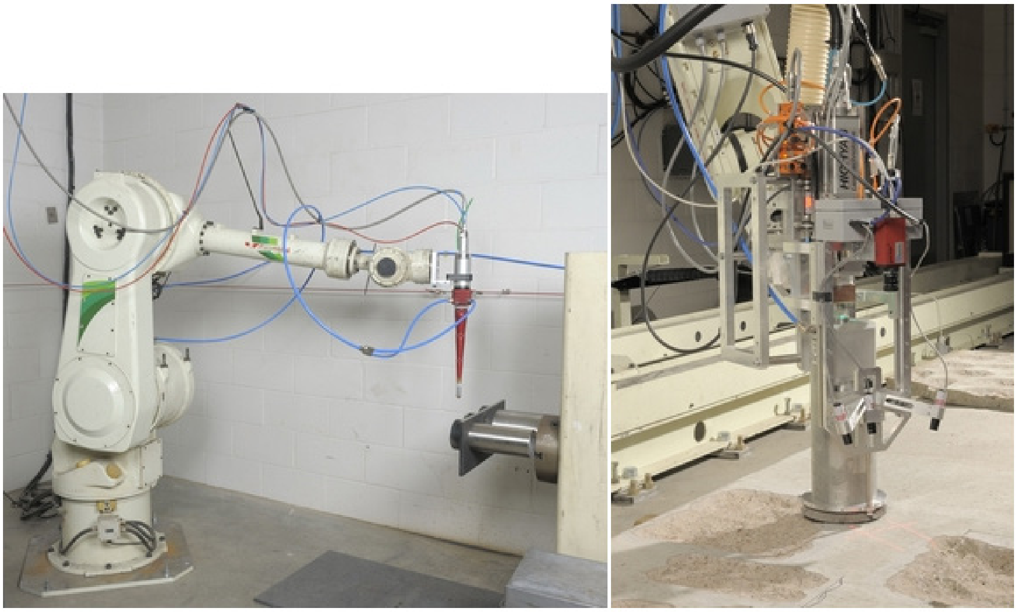
- Technology demonstration project on laser cutting and scabbling

Laser cutting and scabbling

In March 2009, the United Kingdom's Nuclear Decommissioning Authority awarded a contract to develop prototype equipment for demonstrating the twin processes of concrete scabbling and tube cutting, and how these technologies might be implemented for remote use in nuclear decommissioning environments. The goal of the project was to allow site-licensed and supply chain companies to evaluate the technology in terms of both process capability and operating costs, assuming the underlying technologies had been addressed. The results showed a 5 kW pipe cutting laser could cut through a 25 mm diameter (1 in.) tube of 11.1 mm wall thickness at a speed of 110 mm/min. The results also showed that a 5 kW scabbling laser removed an area of one square metre to a depth of 10 mm in 110 minutes. The approximate costs for the systems were: laser scabbling system GBP 411 000; laser pipe cutting system GBP 356 000.

Hilton, Kahn and Walters (2010) provide a full description of the two laser systems. Khan and Hilton (2010), Hilton and Khan (2010), OC Robotics (2011), and Hilton and Waters (2010) provide additional information on the R&D of laser cutting. Figure 3.12 shows photos of the demonstration set-up.

Figure 3.12: IWT pipe laser cutting (left) and laser concrete scabbling (right)



- Monitoring methods for cutting operations

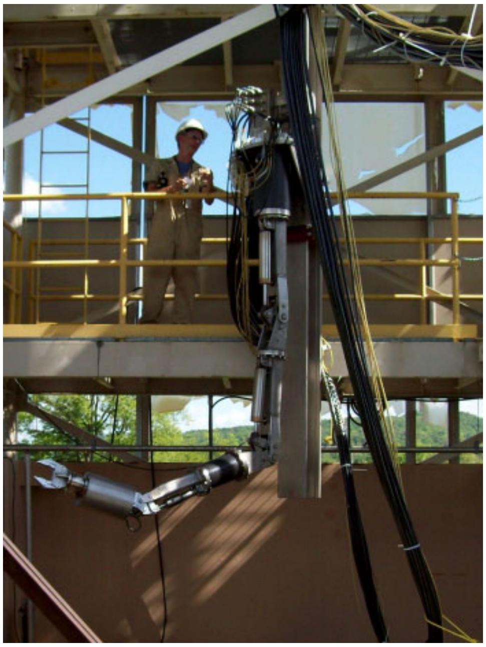
Acoustic emission (AE) monitoring is being used as a method to monitor cutting tool efficiencies. An example of this is work on monitoring titanium machining using AE. The effects of cutting parameters and tool wear on the AE signal in the high-speed turning of Ti-6Al-4V alloy with a new generation of cemented carbide tools was investigated in a recent study. The results demonstrated AE signals as a potential indicator for tool condition monitoring (TCM) in turning of titanium Ti-6Al-4V alloy (Fadare, et al., 2012). Similarly, the AE technique (AET) has been used to monitor the progress of tool wear during the turning of a silicon carbide (20 wt.%) dispersed Al alloy metal matrix composite. Different parameters such as skewness and kurtosis of the statistical distribution, b-parameter of amplitude distribution and uncertainties can be used in a complimentary manner for comprehensive evaluation of tool wear (Mukhopadhyay, 2012). Another study in 2010 used AE from an embedded sensor for the computation of features and prediction of tool wear. A reduced feature subset that is optimal in both estimation and clustering least square errors was then selected using a new dominant feature identification (DFI) algorithm to reduce signal processing and the number of sensors required. Tool wear was then predicted using an ARMAX model based on the reduced features. The experimental results on a ball nose cutter in a high speed milling machine show a reduction in 16.83% in mean relative error when compared to other methods proposed in the literature (Pang, et al., 2010).

- Review of available robotic decommissioning techniques

West Valley Environmental Services (responsible for decommissioning the West Valley Reprocessing Plant in New York) contracted with Nuvision Engineering, Inc., to design a robotic arm (ARTISAN) to reach into the chemical processing cell to cut and remove thousands of feet of piping ranging from 5 cm (2 in.) to 20 cm (8 in.) in diameter, and structural steel up to 20 cm in thickness. The manipulator was fitted with a mechanical shear for 5 cm piping, a band saw for 15 cm piping, and a circular saw for structural steel. The arm was designed to lift 112 kg (250 lbs) at full extension. The arm was also designed to remove three large tanks by unbolting them. A remotely operated wrench was also fitted for the arm. Figure 3.13 shows a photo of the manipulator arm (Judd, 2011).

The ARTISAN arm performed beyond expectations and avoided the need for direct personnel exposure for the dismantling operation. Judd (2011) provides a complete description of the robotic arm.

Figure 3.13: West Valley, NY ARTISAN robotic arm



Hinkley Point A ROV

The fuel ponds at Hinkley Point contained considerable amounts of sludge and debris. The cost of a new remote operated vehicle (ROV) was prohibitive (GBP 175 000), so a suitable used construction tracked micro-excavator was purchased (GBP 3 000) and modified for underwater use. An underwater CCTV was acquired (GBP 15 000) and attached to the ROV. The planned lifetime was three weeks at a depth of 6 m (20 ft), but it actually operated for 326 days without significant problems. All sludge and fuel element debris was removed, with the only problem being visibility through the CCTV from clouds of disturbed sludge. The unit was subsequently sent to Bradwell NPP to perform the same tasks and operated for an additional 253 days. Figure 3.14 shows the ROV after modifications. Pitman (2011) provides a complete description of the ROV modifications and use.

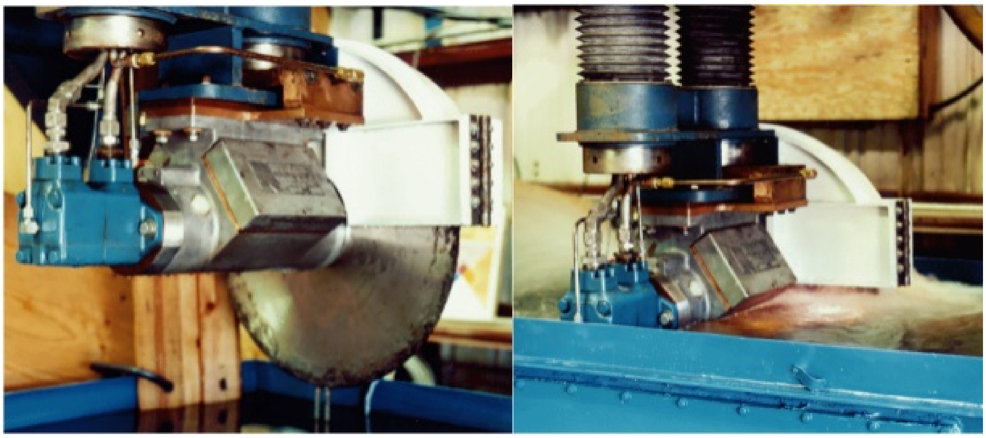
Figure 3.14: Refurbished ROV for Hinkley Point A

- Underwater cutting of reactor vessel and internals using an arc saw

The arc saw is a circular, toothless saw blade that cuts any conducting metal without physical contact with the workpiece, eliminating any reaction forces between the two (Boing, 2006; Deichelbohrer, 1982; JAEA, 2013; Allison, 1980). This means there are no reaction forces between the blade and the workpiece. The cutting action is obtained by maintaining a high current electric arc between the blade and the material being cut while the water (pool or spray) cools the blade and washes out the swarf. The blade, made of any electrical conducting metal such as tool steel, mild steel or copper, rotates at 300-1 800 rpm, causing removal of the molten metal created by the arc in the kerf of the cut. The molten metal then condenses in the form of highly oxidised pellets as it is expelled from the kerf. The depth of the cut, up to 0.9 metres, is determined by the blade diameter and the motor drive head diameter.

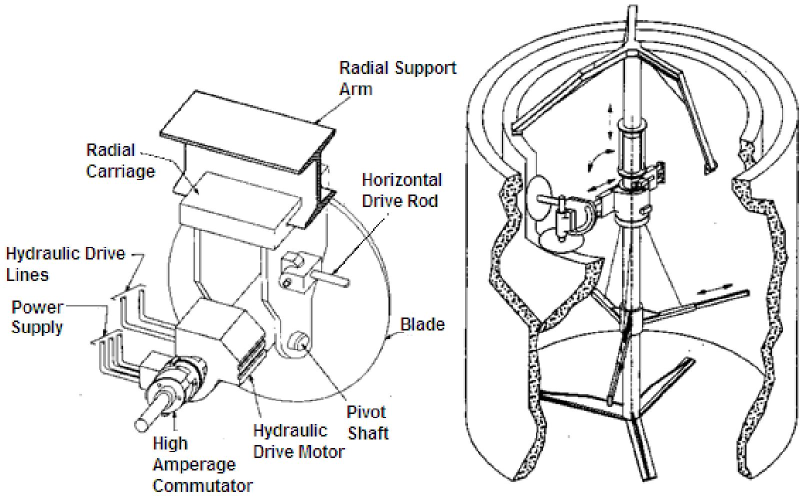
The arc saw is capable of cutting any electrical conducting material. High conductivity materials (e.g. stainless steel, high alloy steels, aluminium, copper and Inconel) produce the best results. Although carbon steel cuts produce slag build-up in the kerf, which impedes the cutting rate of speed, most materials are cut rapidly and cleanly. Other materials, such as magnesium, titanium and zirconium, will produce hydrogen gas when cut, resulting in the possibility of small, localised ignitions.

Figure 3.15: Arc saw cutting head



The arc saw can be operated under water, or in air with water spray. However, under water is the preferred medium since in-air cutting produces significant amounts of smoke, greater noise and a rougher cut. Cutting in air requires adequate ventilation controls to filter the resultant particulates. Underwater cutting produces a small quantity of steam bubbles, which quickly condense as they rise within the pool. Figure 3.15 shows photographs of an arc saw head, and the saw cutting in a cutting tank. Figure 3.16 shows a conceptual drawing of the arc saw in a cutting configuration.

Figure 3.16: Arc saw in cutting configuration



Cutting speed

Retech, Inc., the original designer and supplier of the arc saw (its patent has run out), claimed its maximum cutting speed on stainless steel under water is 1 290 cm^2 of cut surface per minute for its large diameter saw. This was accomplished with a 0.9 metre diameter blade, using a 480-volt AC, three-phase, 750 kVA input power supply, and cutting at up to 40 000 amps and 25 volts DC (Deichelbohrer, 1982; JAEA, 2013).

The Idaho Nuclear Engineering Laboratory (INEL) had used an arc saw for more than 10 years (and may still be using it). The INEL unit was small, using a 5 000 amp power supply cutting at 25 volts DC and less than 900 amps (the lower amperage extends blade life). This saw achieved a cutting speed under water of approximately 38.7 cm^2 per minute through 2.54 cm thick stainless steel tubing. Rockwell Hanford (Deichelbohrer, 1982) performed extensive testing of an arc saw in the early 1980s and recorded cutting speeds in air of approximately 161.3 cm^2 per minute with a small arc saw (7 500 amp capacity at 22 volts DC).

For the large-diameter (0.9 metre diameter blade) high-power saw, the cutting speed under water is approximately ten times faster than plasma arc torches rated for the same service, and 100 times faster than known mechanical cutters (Deichelbohrer, 1982; JAEA, 2013).

Cutting thick cross-section materials

The arc saw cutting capabilities are limited only by the diameters of the blade and the drive head. With a Retech V-8 drive head of approximately 20.3 cm diameter, and a 0.9 metre blade diameter, the maximum thickness of cut is 35.6 cm. The arc saw is especially suited for cutting stainless steels because they are non-magnetic. For under water cutting, no other tool can perform this depth of cut (Deichelbohrer, 1982; JAEA, 2013).

Cutting tool reaction forces

Since the arc saw never touches the workpiece, there are no reaction forces between the two. This means the support system and end-effector (manipulator positioning device) does not have to be built to resist high forces typical of mechanical cutting methods (Deichelbohrer, 1982; JAEA, 2013).

Cutting through multiple thicknesses

One of the distinguishing features of the arc saw is its ability to cut through multiple thicknesses of steel in a single pass. As the blade encounters a new workpiece surface, an arc is automatically struck and melting begins. When it passes through the surface, the arc is extinguished at that location but continues at the original location until the cut is completed. The arc current is used as feedback to automatically control the rate of advance into the workpiece (Deichelbohrer, 1982; JAEA, 2013).

Swarf diameter

The arc saw was used extensively by the Japanese at the Japanese Atomic Energy Research Institute (JAERI) in segmenting the reactor vessel. The swarf particle size and distribution were recorded as 98% of the total, being greater than 37 micrometres. Most of the swarf was 100 μm in diameter and readily removed from the water by gravity and later vacuuming, while the rest of the fines were collected on a 0.9 μm filter and removed. This is significantly larger than the swarf from the plasma arc torch, which will make swarf removal from the pool much easier (Deichelbohrer, 1982; JAEA, 2013).

Gas generation

The arc saw does not use gases for cutting, and therefore should not generate a rising gaseous plume to carry radioactive particulate to the pool surface. Any steam produced by the saw heat should be rapidly condensed in the cutting region, as was observed at an INEL arc saw demonstration. Hydrogen generation, by disassociation of water, recombines under water. Only when cutting in air is there some minor hydrogen generation, which quickly recombines in air with a crackling sound.

Any steam produced can be captured and vacuumed away by an underwater vacuum system provided to collect particulate generation in the vicinity of the saw blade. In addition, to facilitate viewing through the water surface, an acrylic plastic (Plexiglas) or polycarbonate (Lexan) viewing window can be floated over the arc saw and vacuum suction maintained at the plastic-water interface to ensure particulate is continually removed.

Pool heating

The arc saw cutting power requirements of approximately 20 000 amps at 25 volts DC would likely generate some pool heating. Virtually all of this power goes directly into the arc heating the metal in the kerf. Obviously, some of it will also contribute to pool water heating, just as will occur from the hot chips from mechanical cutting. This heat generation should be far less than the 20 000°C (36 000°F) flame of the plasma arc torch. The INEL arc saw did not indicate significant thermal currents from the arc (Deichelbohrer, 1982; JAEA, 2013).

Blade life

Retech, Inc., has done testing on arc saws and determined that the blade consumption for carbon steel blades on stainless steel workpieces is approximately 6.5 cm^2 of blade loss per 71.0 cm^2 of work lost. This relatively low rate of blade consumption will permit cutting virtually all day without a blade change. Blade change-out can be accomplished remotely under water to minimise downtime, and can be accomplished in less than 30 minutes (Deichelbohrer, 1982; JAEA, 2013).

Future suggested R&D for improvements in efficiency by use of remote systems and/or innovative technologies

Because this is such a broad area which includes robotic systems, remote cutting and handling technologies, several separate R&D efforts with different deliverables and objectives are warranted.

- Laser cutting and scabbling
  - ***Description*** – Develop improved laser cutting scabbling capabilities end effectors and delivery systems as well as other supporting technologies to improve cutting speeds and capabilities in air and in water.

- ***Objective*** – The objective of these efforts should be to improve the speed and efficiency of overall cutting and scabbling operations by taking advantage of higher power laser end effectors and through remote delivery systems that remove personnel from the immediate work area simplifying the work planning and execution process.
- ***Desired deliverables*** – Field deployable higher power laser end effectors and remote delivery systems to enhance cutting and scrabbling. Integrated safety interlocks, airborne radioactivity and waste collection systems, as well as slag removal to afford laser cutting of thicker materials should also be part of the technological improvement effort. Remote or robotic delivery systems to enable cutting and scabbling operations that remove personnel from the work area should also be an objective of research and development.

▪ Arc saw underwater cutting

- ***Description*** – Develop improved arc saw cutting capabilities to improve speed and efficiency of underwater cutting and the monitoring of cutting parameters.
- ***Objective*** – To develop an arc saw that will perform more effectively than current technologies for segmenting reactor vessels and internals.
- ***Desired deliverables*** – The following deliverables were demonstrated in earlier versions of the arc saw (Deichelbohrer, 1982; JAEA, 2013) and new R&D is needed to further refine and demonstrate its cutting characteristics for cutting speed through various materials and thicknesses, blade life, reaction forces, swarf characteristics and collection and pool cooling and temperature controls.

▪ Suggested technology for improving efficiency with underwater collection, separation and filtration of swarf and fine particulate from underwater cutting of reactor vessel internals

- ***Description*** – Develop an improved system for collecting swarf at the source, separating it by particle size, filtering the fine swarf and packaging for disposal.
- ***Objective*** – Develop an underwater vacuum system with a collection funnel, cyclone separation unit to separate the particle size, roughing filter, polishing filter and remote packaging system for filter disposal.
- ***Desired deliverables*** – The system needs to be an underwater high-flow (3 785 l/min, 1 000 gal/min) vacuum, with a cyclone separator fitted with a bottom collection container, a roughing filter, a final polishing filter and remotely operated packaging system for disposal. The packaging container should be fitted with valved nozzles and vents to vacuum the residual water from the pool, and of a size to fit in a standard 200 litre drum or equivalent disposal container. Grouting of the 200 litre drum should be a part of the drying system.

▪ Suggested technology for standardising robotic modules, articulated arms, hydraulic or electric power packs

- ***Description*** – Each robotic arm technology developed for a specific purpose appears to have been designed from scratch, with little or no standardisation. This is wasteful, time consuming and expensive. While it surely provides jobs for creative designers, it causes delays to a project when the robot must pass mock-up and verification trials before use.
- ***Objective*** – Develop a series of modular robotic modules on a basic design frame, each with varying capacities for lifting, reach and articulation. Some companies

have developed and successfully marketed standardised units, such as the various models of the Brokk machine.

- ***Desired deliverables*** – A family of robotic modules that can be assembled from a "toolbox" of options for several capacity sizes and range of motion applications. Hydraulic or electric power packs can similarly be developed with varying pressures, hydraulic flow rates and electricity requirements.

Improvements in safety by using remote systems and innovative technologies

Challenges

There is a need to improve the operational flexibility of remote cutting systems by taking the benefits of generic robotic systems from other applications or reducing robot size. Development of technologies that will protect the workforce or minimise man access is a common objective, although more so for those countries with diverse facilities. It is here that the need to better understand the environment in which the work is to be carried out can help.

The robotic and intelligent machines (RIM) roadmap contains a list of applications and a tabulation of the desired technology (DeGregory, et al., 2001). Some of the most significant science opportunities to achieve these technology goals are listed below:

- Sensors for site characterisation and acquisition of performance data are essential to support decision making either through software or by visualisation and human judgment.
- A special need is the kinaesthetic interface to human operators to enhance their motor skills and input commands to the remote system.
- Mobile platforms that in themselves are modular and highly dexterous must be further developed to gain access to the work environment and to transport size-reduced facility components.
- Quick-change end-effector tools based on a science of tools (design, modelling and operation) are needed to perform the in-contact physical tasks for D&D.
- Dexterous high-load robot manipulators capable of tool management, size reduction, parts transport and parts packaging under human supervision will be especially important for D&D tasks.
- Intelligent and standardised actuator modules to build all D&D remote systems on demand from a minimum number of high-performance and low-cost modules just as we now build computers on demand.
- A universal operating software for any intelligent machine used for D&D. Similar to the operating systems in today's micro-computers, this software would support operation of mobile platforms, gantries, small automation subsystems and dexterous manipulators, all under the centralised supervision of operators in a remote position.
- Electronics will be pervasive in a modern remote systems technology. Hence, their hardening against radiation, temperature, shock and particulates is necessary.

Summary of current R&D projects – Improvements in safety by use of remote systems and/or innovative technologies

- Cooled personnel protection equipment

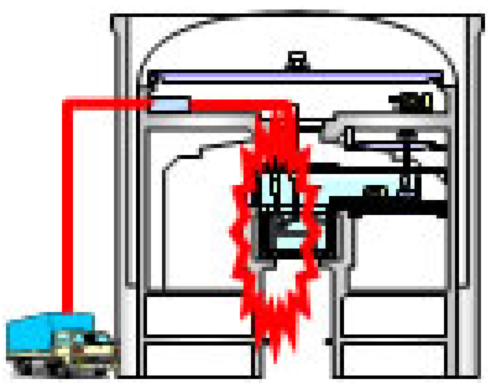
The concept of cooled personal protective equipment (PPE) is not new and in fact the US DOE funded studies and prototypes as early as the mid-1990s (Ebadian, 1999). The use of cooling systems built into PPE with a circulating system greatly improves worker comfort and safety.

Three types of cooling systems were investigated: pre-chilled or forced-air system (PCFA), umbilical fluid-chilled system (UFCS) and passive vest system (PVS). Of these, the UFCS leads the way. The PVS or gel pack vest lagged due to a limited cooling duration and the PCFA or chilled liquid air supply was cumbersome and required an expensive and complex recharge system. The UFCS in the form of the personal ice-cooling system (PICS) performed exceptionally. The technology uses a chilled liquid-circulating undergarment and a PPE external pump and ice reservoir. The system is moderately expensive, but the recharge is low-tech and inexpensive enough to offset the cost.

- Study of cutting methods applicable under water

LaserX is performing R&D on development of an underwater laser processing head for thick sheet steel. The basic data acquisition is for underwater cutting for nuclear reactor dismantling. The underwater thick sheet steel cutting technology was successfully developed. Carbon steel and stainless steel of 50 mm were cut at speeds of 50 to 100 mm/min, using an assist oxygen and air assist gas supplied by tetra nozzle (Ebadian, 1999; WERC, n.d.).

Figure 3.17: Remote cutting by mobility fibre laser



A number of technologies are being trialled for cutting up fuel skips in the United Kingdom, including fibre lasers by Sellafield Ltd, diamond wire by Magnox Ltd and plasma arc by Dounreay Site Restoration Ltd (Sellafield Ltd, 2012c). Sellafield Ltd has already sent Cambridge-based TWI three clean fuel skips for the cutting trials. They have developed a number of cutting plans to size reduce the skips using a 5 kw fibre laser. One plan shows a single 1 m^3 skip reduced to a pile of metal of less than 20 cm in height, which theoretically means five skips can be consolidated into one. The trial should be completed soon, after which work on the concept design will start following a decision on the best cutting method. The process to design, manufacture, install and commission the equipment will take a number of years and should be available to start processing the first redundant fuel skip in 2015.

Future suggested R&D for improvements in safety by use of remote systems and/or innovative technologies

- Suggested technology for underwater cutting of reactor vessel and internals using an arc saw
    - ***Description*** – The arc saw was described earlier in detail.

- ***Objective*** – Development of an arc saw to improve safety when segmenting reactor vessels and internals.
- ***Desired deliverables*** – Demonstration that the arc saw can improve safety when segmenting reactor vessels and internals in six major areas:
  - Reduction in overall cutting time and therefore reduction in radiation exposure to workers.
  - Reduction in fine particulate generation and therefore minimal contamination of the cutting pool and potential exposure to workers.
  - No cutting gases are used so there should be no obstruction of visibility while cutting, and no gaseous effluents to collect. The steam produced is quickly condensed in the cutting pool. The (small) amount of hydrogen produced is dissipated in the cutting pool.
  - There is minimal pool heating from the saw blade as the only heat is from the molten swarf particles and the segmented sections. The fast cutting speed reduces the amount of heat introduced into the cutting pool.
  - The diameter of most of the swarf is large but quickly cools and drops into a collection tray or vacuum and minimises contamination of the cutting pool.
  - Blade change-out is underwater and fast so there is minimal worker exposure.

▪ Improving safety for treatment of swarf and fine particulate during underwater cutting

- ***Description*** – The system description is as described earlier.
- ***Objective*** – To reduce exposure to workers in packaging and handling underwater particulate filters used in reactor vessel internals segmentation.
- ***Desired deliverables*** – A fully remote skid module for separating, filtering and packaging the filter unit for disposal without direct operator contact. This would include a remote method for grouting the final container for disposal to minimise worker radiation exposure.

Reduction in secondary waste generation

Challenges

It must be ensured that effective cutting tools do not generate excessive additional secondary waste. For example, research carried out in Japan has shown that plasma cutting gave good cutting speed but generated more secondary waste (HEPA filters). In contrast, Spain deploys mechanical cutting for this reason. The needs for optimising the segmentation process to minimise waste container filling are summarised as follows:

- reduction of secondary effluent produced from mechanical cutting, in particular where there is no *in situ* treatment infrastructure;
- improvements to water cleaning technologies;
- decontamination systems to reduce waste volume;
- de-watering or concentration of sludge;
- methods to absorb liquids to allow disposal as solid;
- methods to absorb and remove contaminants while releasing clean water.

Summary of current R&D projects – Reduction in secondary waste generation

▪ Modular/mobile effluent and waste retrieval plants

Radioactive waste was brought to Sellafield to be stored in the silos from nuclear sites all across the United Kingdom. The waste was deposited through access charge holes located above the silo compartments. Nuclear Engineering Services (NES) is supplying Sellafield Ltd on behalf of the Nuclear Decommissioning Authority (NDA) with three silo emptying plant (SEP) Mobile Cave waste retrieval machines, for the removal of intermediate-level wastes (ILW) from the 22 silos (Patel, 2012). The three SEP Mobile Caves are deployed to mechanically retrieve and package waste items by size-reducing and consigning the waste from each of the silos into waste skips. The skips have an internal volume capacity of 1.2 m^3. The SEP Mobile Caves are mounted on a rail system, allowing them to be moved from silo to silo as the retrieval operation progresses. The skips are then transferred to the Silo Direct Encapsulation Plant (SDP) for further processing and exporting of the waste. NES is currently in the commissioning phase of SEP 2 Mobile Cave, with completion scheduled in 2014; SEP 1 and SEP 3 Caves will follow. SEP 2 Cave will be used to retrieve waste from 6 of the 22 silos. When assembled, it will weigh approximately 320 tonnes, with dimensions of approximately 12 m long × 5 m wide × 7 m high.

The SEP Mobile Caves house a range of specially designed tools specifically for size reduction and the retrieval process. NES' designated SEP tooling team has developed a range of these tools for size reduction to ensure that large waste items fit inside a waste skip, including:

- water jet cutting (WJC) charge tube size reduction/export skip;
- water jet cutting (WJC) swarf bin skip;
- thermocouple cropper (TCC);
- deformator.

Trials have been completed on a number of size-reducing tools, while others are in final development and currently going through the trial process.

Future suggested R&D for reduction in secondary waste generation

- Suggested technology for underwater cutting of reactor vessel and internals using an arc saw
  - ***Description*** – The arc saw was described earlier in detail.
  - ***Objectives*** – Development of an arc saw to reduce secondary waste generation.
  - ***Desired deliverables*** – Demonstration that the arc saw can reduce secondary waste generation in the following five major areas:
    - Reduction in fine particulate generation and therefore minimal use of fine particulate filters to collect the swarf. The heavy swarf is between 0.037 to 0.1 mm in diameter. The fine swarf can be collected on a 1 micron filter.
    - No cutting gases are used so there should be no obstruction of visibility while cutting, and no gaseous effluents to collect. The steam produced is quickly condensed in the cutting pool. The (small) amount of hydrogen produced is dissipated in the cutting pool.
    - A kerf size essentially equal to the thickness of the blade is between 6-9 mm wide and therefore a minimal amount of swarf needs to be packaged for disposal.
    - Blade usage is low, requiring about one blade for an entire day of cutting resulting in minimal packaging of spent blades for disposal.
    - The cut surface has no back-face slag to interfere with packing segments closely in a disposal liner.
- Underwater treatment of swarf and fine particulate during underwater cutting
  - ***Description*** – The system description is as described earlier.

- ***Objective*** – To reduce the generation of wastes in packaging for disposal, the key being to classify generated wastes by particle size.
- ***Desired deliverables*** – A fully remote skid module for separating, filtering and packaging the filter unit for disposal without direct operator contact. The cyclone separator will remove large, heavy-weight particles that can be packaged efficiently in a liner and grouted for disposal remotely. The roughing filter will remove larger particulates from the waste stream, which can similarly be grouted in a liner for disposal. The sub-micron HEPA filter will collect all the residual particulates and can be grouted remotely for disposal.

4. Decontamination and remediation

Theme overview

This theme focused on the issues surrounding the decontamination of components (including both concrete and metals), remediation of soils and limiting the spread of contamination in groundwater. Nine issues were formulated to assess the rudimentary challenges encountered during the facility decommissioning process.

The issues addressed were new physical processes and chemical processes, surface treatment and removal of contamination and surface polishing; heels and residues (e.g. from process fuels/fuel cycle reprocessing); concrete remediation; optimising the use of robotics; bulk soil remediation; fixing contamination in soil; decontamination of large components; and methods for decontaminating high volumes of water or chemicals contaminated to low levels.

The working group for this theme noted that the R&D requirements and the priority of consequences to the issues varied significantly among the surveyed participants. They also noted that the problematic issues of disposing of contaminated concrete and using robotic technology during facility decommissioning were common concerns to all the national programmes.

Summary of current practices and guidance

Current practices and guidance for decontamination and remediation cover a wide range of technologies and methods, but tend to rely on standard practices such as chemical treatment and abrasive, high pressure, mechanical surface removal that have been in place for many years. The use and efficacy of these various technologies are the subject of many guidance documents and reports published by various organisations, as seen in Table 4.1.

The accelerated retirement of facilities and backlog of decommissioning have resulted in a new urgency to develop more efficient and effective decontamination and remediation methods to minimise the production of wastes and to optimise recycling and reuse of materials. Decontamination and remediation technologies tend to be of three types: *i) in situ* technologies used to decontaminate intact systems and structures in place; *ii)* material and equipment decontamination systems used as part of the handling and processing of materials; *iii)* equipment and environmental decontamination and remediation of contaminated soils and groundwater.

In general the *in situ* decontamination methodologies are the ones that have been developed and used to the greatest extent. The prolonged placement of facilities into SAFSTOR eliminates the use of highly effective low-waste generation options such as full system chemical decontaminations. Since modular chemical decontamination systems for removed materials have not been developed, deployed and integrated into the material removal handling and assay processes at decommissioning facilities more labour-intensive and higher-waste-generating technologies are often the only options. Furthermore, decontamination techniques tend to be labour intensive and performed *in situ* rather than post-removal as part of an automated handling and processing system even though the commodities removed and packaged during decommissioning tend to be a relatively small set of similar materials.

Table 4.1: Guidance documents for decontamination and remediation

Facility type	Phase	Region	Document
All types	Decommissioning and decontamination	International	*Jose Cabrera Nuclear Power Plant Full System Chemical Decontamination Experience Report*, EPRI (2009b)
All types	Decommissioning and decontamination	International	*Remediation of Embedded Piping*, EPRI (2000b)
Power reactors	All phases	United States	*Groundwater Contamination (Tritium) at Nuclear Plants*, US NRC (2013b)
All types	All phases	United States	*20.1406 Minimization of Contamination*, US NRC (2011a)
All types	All phases	United States	*Minimization of Contamination and Radioactive Waste Generation: Life-Cycle Planning*, US NRC (2008a)
All types	All phases	United States	*Release of Radionuclides and Chelating Agents from Full-System Decontamination Ion-Exchange Resins*, US NRC (2002b)
All types	Decommissioning and decontamination	International	*State-of-the-Art Technology for Decontamination and Dismantling of Nuclear Facilities*, IAEA (1999d)
All types	Decommissioning and decontamination	International	*New Methods and Techniques for Decontamination in Maintenance or Decommissioning Operations: Results of a Co-ordinated Research Programme 1994-1998*, IAEA (1998a)
All types	Decommissioning and decontamination	International	*Summary Report of the Preliminary Findings of the IAEA Mission on Remediation of Large Contaminated Areas Off-Site the Fukushima Dai-ichi NPP*, IAEA (2011d)
All types	Decommissioning and decontamination	United States	*Inventory of Radiological Methodologies for Sites Contaminated with Radioactive Materials*, US EPA (2006)
All types	Decommissioning and decontamination	International	*Technologies for Remediation of Radioactively Contaminated Sites*, IAEA (1999e)
All types	All phases	United States	*Contaminated Site Clean-Up Information (CLU-IN)*, US EPA (n.d.)

New robotic and remotely operated technologies as well as decontamination tooling such as laser scabblers and surface cleaning/stripping offer opportunities to improve *in situ* decontamination methods and technologies to make them more effective and less labour intensive.

Although they have been around for many years, *ex situ* technologies are not generally modular, field deployable or integrated into the material removal, handling and assay process. As the use of remote cutting and handling technologies becomes more routine the pre-demolition decontamination of systems and components to minimise personnel exposures will become less of an incentive for early decontamination and the need to process and decontaminate removed materials to enhance recycling, re-use and waste minimisation will gain in importance. Due to the sectioning and sizing of removed materials, internal surfaces of components such as piping and pumps are often more accessible and less labour intensive to reach post-removal. Development of automated, modular, remote assembly-line-type decontamination technologies that are field deployable is likely to become a greater need in future decommissioning as technological developments allow demolition tasks to be accomplished in a more automated and remotely operated manner.

In addition, remediation technologies for contaminated soils and groundwater are currently not well developed, with excavation, pump and treat technologies being the most commonly used practices. These technologies have severe limitations as evidenced by the clean-up challenges for Chernobyl, Fukushima and many defence-related facility decommissioning in various countries. Accidents and operational practices in the early years of nuclear technology development have led to wide-scale soil and groundwater contamination at some facilities. Thus development of options for remediation and treatment of soils and groundwater in place or for decontamination and reuse of soils is of paramount importance to effectively and efficiently complete many decommissioning.

Summary of challenges and R&D needs

A summary of the decontamination and soil/groundwater contamination challenges by the National Research Council in 2001 included items addressed by the OECD Working Group. With regard to decontamination processes, the NRC noted that scientific understanding of the interactions among contaminants and construction materials is required to develop more effective decontamination technologies. Improvements necessary for the fundamental understanding of contaminant interactions with materials are discussed in Chapter 2 of this report, in the section entitled *Modelling of mobile nuclide behaviour on different substrates*. Understanding the fundamental physical and chemical processes, such as sorption, desorption, chemical reactions and chemical bonding of contaminants within waste materials (graphite, steel, concrete) are necessary in order to evaluate and describe the mechanisms by which decontamination techniques work. The National Research Council made two recommendations in the area of decontamination (NRC, 2001):

- Basic research toward fundamental understanding of the chemical and physical interactions of important contaminants with the primary materials of interest in D&D projects, including concrete, stainless steel, paints and strippable coatings. The results should be used to develop first-principle models that describe the interactions and can thus be used to investigate improved approaches to decontamination.
- Basic research on biotechnological means to remove contaminants from surfaces and from within porous materials found in surplus DOE facilities.

With regard to the first recommendation, the need for research on the first principles of understanding contaminant interactions with waste materials is covered in Chapter 2 (*Modelling of mobile nuclide behaviour on different substrates*). Strippable coatings is a decontamination method similar to chemical decontamination, abrasive blasting, etc. and should probably not be lumped into that category. It will be discussed in this section of the report along with the need to better understand the first principles by which the decontamination methods operate. The NRC noted in 2001 that present decontamination approaches are usually based on experience or trial and error, rather than quantitative prediction of how the contaminants are bound to construction materials and how chemical or physical methods can best remove them.

The NRC further remarked that for large-scale applications, almost all current decontamination methods are time consuming, involve risks to workers, produce significant volumes of secondary waste and often leave behind residual contamination, especially actinide contamination. They usually require direct, hands-on work such as concrete spalling work, wiping the surface with cleaners (e.g. detergents, acids, complexants), washing with high-pressure water, abrasive blasting, immersing objects in various cleaners or electro-polishing. Given current technologies, D&D contractors usually choose to send large amounts of contaminated materials (e.g. concrete and steel) to licensed disposal facilities rather than attempt to decontaminate them for possible reuse. However, even for concrete, a relatively cheap raw material, recycling can be economical. The 2001 presentations to the NRC committee indicated a need to improve current technologies for removal of radionuclides and EPA-listed organics and metals from equipment and building structures and metal, concrete and wood debris. Many DOE and fuel reprocessing sites such as Sellafield encounter the problem of actinide-contaminated materials, which include glove boxes, shielded cell liners, concrete, lead bricks, lead glass and plastics.

Radioactively contaminated lead, which is also chemically toxic, is a particular challenge. The DOE has a large inventory of contaminated lead due to its use as shielding material. The ability to efficiently remove actinides from the surface of construction materials will allow recycling or cheaper disposal of the material. The NRC noted that in many instances, paints, sealers and varnishes create a laminate problem, with aged materials being harder to decontaminate than more recent depositions. Deep penetration of contaminants into porous structural material such as concrete also renders decontamination difficult (NRC, 2001).

Not much has changed in the intervening years since the 2001 publication of the NRC report. There has been a greater focus on screening and clearance for re-use, recycling or disposal of very low-level waste, but very little thought or effort is given to decontamination of materials removed in order to further optimise the process in current decommissioning efforts. The challenge is to adapt and develop less hands-on, modular and reusable decontamination systems and methods that can be integrated into the material removal, assay/monitoring and packaging processes (NRC, 2001).

Remediation of soils and limiting the spread of contamination in groundwater pose a separate and distinctive set of challenges. According to the DOE, there are 79 million cubic metres of contaminated solid environmental media associated with nuclear weapons complexes, of which 70% is contaminated with radionuclides. In addition, there are about 1 800 million cubic metres of contaminated soil, of which 57% is contaminated with radionuclides. The first principles and fundamentals of radionuclide interaction in the environment are well known and sophisticated fate and transport models describing the interactions of radionuclides with soils and radionuclide transport with groundwater have been developed and are continuing to be refined. Despite this knowledge there are currently limited options for dealing with soil and groundwater issues. They normally involve *ex situ* remediation like excavations, pump and treat, or *in situ* interventions like barrier installations or plume retardation through pumping to reverse groundwater flow.

Suggested additional research and development

New physical processes and chemical processes for decontamination

Challenges

New physical and chemical processes that have demonstrated effectiveness over the range of radionuclide contaminants and waste forms associated with decommissioning are needed. The decontamination processes need to be based on a better understanding of the contaminant interactions with the waste materials. They also need to provide technologies and applications that integrate well with the decommissioning and material removal and sentencing processes.

Concrete is one of the main waste forms generated from decommissioning facilities and thus has a high priority for development of decontamination processes. *In situ* decontamination using mechanical removal means, chemical applications, gels and strippable coatings are usually only feasible once systems and components have been removed and the surface is free of obstructions. This places this activity in the critical path for release or demolition of the building, requiring decontamination methods to be effective and efficient. Initial phases of electrokinetic treatment that create an electromagnetic gradient to mobilise ionic contaminants to the surface may be able to be used earlier. In general, decontamination applications, *ex situ* or post-removal treatment of contaminated concrete, have not been developed and tested in decommissioning, but could play a role if concrete recycling and reuse becomes more widespread.

Decontamination of irradiated graphite is an inescapable need, regardless of whether the material is disposed of in a geologic repository or recycled and reused. Under any scenario the capture and separation of volatile gaseous phase ^{14}C, ^{36}Cl and ^{3}H will be required. Technologies exist for decontamination and separation of these constituents in off-gases but developing industrial processes that can perform at the scale needed to handle hundreds of thousands of tonnes of irradiated graphite waste are a serious challenge that has been considered for many years with little progress made. In the meantime, new technologies for isotopic separation are emerging.

Transuranic contamination has an extreme impact on decommissioning due to the radiation protection issues associated with preventing personnel intakes from inhalation and injection wounds and the stringent waste classification limits that result in larger amounts of more costly waste forms for disposal. Chemical and physical removal methods that reduce airborne radioactivity potential and can efficiently and effectively remove transuranic contamination to reduce high-level waste volumes are needed. Often, due to the contamination levels present in areas such as hot cells, decontamination must be performed remotely during the initial application phases. Removal of decontamination agents often requires manual vacuuming or removal of strippable coatings. This final step in the decontamination process is often performed *in situ* prior to removal and thus is in the critical path for the decommissioning. Technologies for more automated separation and removal of the decontamination agents after material removal could integrate their use more effectively into decommissioning processes, whether the waste form is concrete, metal or graphite and regardless of the radionuclides involved.

Summary of current R&D on new physical processes and chemical processes for decontamination

- Decontamination of concrete

There are three decommissioning scenarios where concrete decontamination is frequently required: *i)* situations where all or part of the building will remain intact and concrete requires decontamination to levels that meet clearance or license termination criteria; *ii)* where decontamination is required on interior surfaces in order to meet requirements for open air demolition of the structure; *iii)* when all or part of the structure is to be demolished and the concrete needs to be decontaminated to implement the waste hierarchy.

Scenarios *i)* and *ii)* are similar and require similar, often localised decontamination strategies. Direct physical removal methods (gels, coatings, etc.) are all viable options and require more research. These options are also applicable to Scenario *iii)* demolitions since the concrete can be decontaminated prior to demolition. Scenario *iii)* also offers the unique opportunity to develop decontamination methods that integrate with reuse and recycling technologies for the concrete aggregate. Decontamination of concrete is an important issue since a majority of building waste associated with decommissioning demolition is concrete waste.

Physical removal methods using tooling, such as hammering or chiseling, scarifying, needle scaling, scabbling, shaving/milling, spalling through induction heating of rebar, abrasive blasting (with sponge, steel grit or CO_2 ice), liquid nitrogen jetting and hydrolasing decontamination processes are widely available and have been developed, tested and compared (US EPA, 2011a). In addition, thermal treatments, microwave ablation and laser ablation of painted or coated concrete surfaces have been evaluated and are continuing to be developed (OECD/NEA, 2011; O'Sullivan, Nokhamzon and Cantrel, 2010). The EPA has completed comparative testing of wire brushing, diamond flap wheel, sanding, rotating water jet and abrasive blast vacuuming on concrete test coupons contaminated with ^{137}Cs and found removal efficiencies ranging from 38-96% using these methods (Boing, 2011). Continued research on these technologies to increase their efficacy and efficiency is warranted.

Interest in non-destructive concrete decontamination technologies for use in response to the Fukushima Daiichi accident and potential "dirty bomb" attacks is also currently increasing. Many chemical application and strippable coating processes have been developed and used successfully for concrete surface decontamination. Most can be applied like paint, using commercially available, airless paint sprayers, brushes or rollers. The US EPA has evaluated two chemically-based technologies for their ability to remove ^{137}Cs from concrete surfaces and found that removal rates of 70-80% are achievable (US EPA, 2008c). Another recent test of a similar product showed over 80% removal of ^{137}Cs from concrete coupons (US EPA, 2011b, 2011e; Drake, 2011). More in-depth testing of these applications on concrete contaminated with decommissioning facility waste streams is required.

Argonne National Laboratories has developed a spray-on gel called "supergel" for decontamination of porous materials such as brick and concrete. The polymer gel that absorbs the radioactivity is similar to the absorbent material found in disposable diapers. When exposed to a wetting agent, the polymers form something like a structural scaffold that allows the gel to absorb a great amount of liquid. The amount of contamination removed depends on the characteristics of the contaminated structure (e.g. its age, type of material, whether painted or unpainted) and the radioactive isotope involved. Removal rates are reported to range from roughly 80% to nearly 100%. Remote spray washers apply a wetting agent and a super-absorbent gel onto the contaminated surface. The wetting agent causes the bound radioactivity to re-suspend in the pores; the superabsorbent polymer gel then suctions the radioactivity out of the pores and becomes fixed in the engineered nanoparticles that sit in the gel. The gel is vacuumed and recycled, leaving behind only a small amount of radioactive waste for disposal (ANL, 2006).

Idaho National Laboratories developed another concrete decontamination process that uses a foam treatment to decontaminate the surface, followed by a long-term application of clay paste to remove contamination embedded in the concrete. The foam, known as Rad-Release, removed about 30% of the radioactive contaminant and within six weeks after paste application, approximately 89% removal was obtained. Additional optimisation of the decontamination system is expected to be completed in the near future. Patents have been filed on these technologies (Martin and Demmer, 2011; Stricker, 2012; Thompson, 2007).

This process is now commercially available and has been adapted to allow the foam to be sprayed on (Martin and Demmer, 2011). The foam is produced by aspirating a foaming agent and specific affinity-shifting cleaning solutions, mixing them with air to create the foam, and then spraying them onto the surfaces. As the mixture exits the nozzle of the spraying device, the air expands 20 times and creates the foam. The foam contains strong but highly buffered acids, foaming agent, gelatin and chelants or other chemicals. This creates an affinity shifting chemistry that causes the contaminant to be drawn into and held in the foam matrix (Martin and Demmer, 2011).

The foam shifts the affinity of the radionuclide from a bias for the building's surface toward the decontamination solution. Typically, these "affinity shifting" chemicals are simply called chelators, meaning a chemical that binds other chemicals to it. While the acids in the foam make the metals soluble, the chelants in the foam act ionically bond to the contaminants to prevent them from reattaching to the structure's surface. After the foam's allotted residence time (usually 30 minutes to one hour), the contaminants can be removed easily by vacuuming up the foam (Martin and Demmer, 2011).

The foam is then allowed to collapse and return to a liquid state where it can be treated, evaporated, incinerated or solidified using a grouting process. The small amount of waste produced by the process is then packaged and sent to appropriate disposal facilities, such as a low-level waste repository. At this point, at least 50%, and in some cases as much as 95%, of the surface contaminants have been removed. The process can be repeated as many times as deemed necessary if further decontamination is required. Each cycle will continue to remove contamination until the target goal is achieved; in some cases, to eliminate labour and additional exposure to personnel, a clay application step can be used to achieve final decontamination (Martin and Demmer, 2011). Additional coating test results are discussed in the subsection below on transuranic decontamination.

The clay has the consistency of wet paste or mud. It contains water, clay and a proprietary salt. After the surface contaminants and most of the lightly bonded subsurface contaminants are removed, the remaining subsurface contaminants lodged in the pores of the materials, such as cement, may be drawn out and captured using the clay solution. The wet clay is applied to the surface and left for an extended period of time (three days to six weeks), depending on the contaminant's characteristics. Beneath the surface, and at the molecular level, the salts within the clay spread into the surface's pores via the liquid in the paste and the contaminants are diffused from the pores into the clay due to the concentration gradient (Martin and Demmer, 2011).

Essentially, the contaminants diffuse to the surface, exchange places with the salts, and are captured by the clay's unique mineral structure. The major benefit to using the clay paste is that the contaminants are absorbed and bundled into the paste and can be removed easily for disposal. When the clay dries, it shrinks by 80%, producing a drastically smaller waste volume for final disposal. At this point, Rad-Release has removed up to 99% of the contaminant, and verification of the cleaned surface is conducted (Martin and Demmer, 2011).

More recent technologies that warrant further evaluation include electrokinetic decontamination (using gel or paste electrolyte) (Purdue University, 2014). One recent study on radioactive concrete with 1 940 Bq/kg used a prolonged pre-treatment with an electric current (25 days) followed by acid washing. The study reported removal efficiencies for ^{60}Co and ^{137}Cs of 99.8% and 92.3% (Kim, Choi and Lee, 2010). Laboratory experiments indicate that the type of material (brick versus mortar) and the electrolyte selected could affect the electrokinetic decontamination efficiencies (Castellote, Botija and Andrade, 2010).

Recent studies have confirmed that the bulk of ^{60}Co and ^{Nat}U in concrete are in the paste rather than the sand or gravel aggregates and that, with heat treatment, crushed concrete can be pulverised easily and the contaminated paste can be separated from the sand and gravel aggregates by sieving. This process demonstrated that recovered sand and gravel concentrations are approximately an order of magnitude less than the initial concentrations. A volume reduction of the activated heavy-weight concrete waste and uranium-contaminated light-weight concrete waste was achieved by up to about 80% and 75%, respectively (Min, Choi and Lee, 2010b).

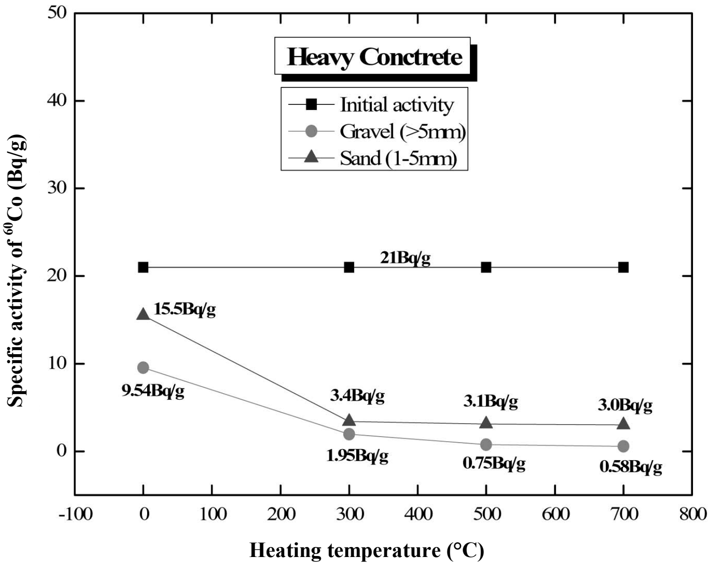
Other studies indicate that ^{137}Cs is also partitioned in the clay like concrete paste and that it may be possible to use chemical reagents rather than heat to separate the aggregate from the paste (Min, Choi and Lee, 2010a, 2010b). More research on the partitioning of other radionuclides and the mechanisms that can create electrochemical or concentration gradients to desorb them should be performed for a comprehensive understanding of the behaviour of radionuclides in a waste stream or fingerprint, and the decontamination strategies that are likely to have the most favourable outcome (Nikolaev, et al. 2012).

- Removing transuranic surface contamination

Decontamination of transuranics is a problem at waste reprocessing facilities and facilities that have experienced significant fuel failures over the course of their operations. *In situ* decontamination is often necessary for health and safety reasons in order to safely conduct the disassembly activities. An *in situ* transuranic chemical decontamination process was used at Rocky Flats to remove high levels of transuranic contamination from glove boxes and tanks (ITRC, 2008). Cerium nitrate [$Ce(NO_3)_3$] was used to decontaminate surfaces with great success. An earlier decontamination process, TechXtract RadPro (2011), applied a complex blend of acids and other chemicals to equipment surfaces in a three-step process. Cerium nitrate is injected with steam into tanks and other equipment, or diluted solutions of cerium nitrate are simply applied to interior surfaces, which are wiped and rinsed with a neutraliser. The extraction solution uses micro-emulsification and chemical ion exchange to bind itself to contaminants.

After 24 hours, surfaces are surveyed to determine whether transportation criteria have been achieved. The life cycle estimates were reduced by nearly 30% by using chemical decontamination technology on TRU waste in projects involving hundreds of contaminated glove boxes and tanks destined for more hazardous and costly size reduction. The most significant benefit of chemical decontamination was thousands of hours of avoided worker exposure to high airborne radioactivity, injection wounds, exertion and several industrial hazards that result from size reduction (ITRC, 2008).

Figure 4.1: Heat treatment removal of ^{60}Co in concrete constituents



Ex situ decontamination is also often required to reduce the amount of high-level waste generated. When transuranics are present in significant quantities in a waste stream, they often drive the waste classification of the contaminated materials. Transuranic decontamination methods have been under development and have been used successfully for the decommissioning of facilities such as Rocky Flats, Paducah Gaseous Diffusion Facility and Savannah River.

A common approach is the use of strippable coatings that can be sprayed on to lock down and control surface TRU contamination during disassembly. They can then be stripped off and disposed of to decontaminate the removed material and reduce the TRU waste generated. Products such as Carboline ALARA 1146, Bartlett TLC Free, Glygel, Cellular Bioengineering Decon Gel 1101 (Agent C) and TechXtract Radpro have been used successfully as strippable coatings for transuranic lockdown and decontamination. TechXtract has been studied extensively and used for transuranic decontamination at many decommissionings (Duncan, et al., 2009; Lear, et al., 2007; US EPA, 2003, 2008a; Holt, 2007; US DOE, 2000). Studies indicate that decontamination efficiency may vary among such products, depending on the material, surface characteristics and contaminant, and that remote use and accessibility may also play a role in the selection of the correct product (Draine, 2009; Farrell, May Howell, 2005).

At West Valley the vitrification cell inside the vitrification facility was dismantled and the equipment was removed after completion of high-level waste solidification. The large facility is now being used as a remote waste processing area for large scale and highly radioactive components. Processing equipment in the cell consists of Brokk® equipment with demolition end effectors, power manipulators, a plasma system and a portable Nitrocision® decontamination system (Blankenhorn, et al., 2011; Vandegrift, et al., 1984).

Decontamination methods also need development for *ex situ* decontamination of materials after they are removed. It has long been known the chemical decontamination techniques are effective at removing transuranic materials. In addition, physical removal processes such as vibratory finishing have been used effectively to decontaminate transuranic contaminated tools, equipment and components (McCoy, Arrowsmith and Allen, 1980). For strippable coatings that remain intact on materials during the removal process, an automated method of removing and recovering the strippable coating on materials prior to waste packaging would increase efficiency (Heshmatpour, Copeland and Heestand, 1983).

- Graphite processing

Storage of graphite in a geologic repository may involve some pre-treatment to remove highly mobile, volatile fractions of ^{3}H, ^{14},C, ^{36}Cl in the graphite matrix. Pre-treatment options being evaluated include treatment of graphite with liquid decontamination agents such as mineral acids, alkaline solutions, dissolved oxidising agents, organic washing detergents or the removal of contamination in the graphite with centrifugation (Jones, 2010).

The potential to recycle irradiated graphite is being investigated by the CARBOWASTE initiative in Europe. Potential products from recycled nuclear graphite include electrodes for use in nuclear waste vitrification, nuclear graphite moderator for gas-cooled reactors, silicon carbide, absorbent material to remove nuclides from liquid/gaseous waste, (e.g. charcoal filters) and the manufacture of nanotubes for use in waste disposal technology (Bradbury, 2010). Methods for decontaminating irradiated graphite and separating ^{14}C are being considered as part of this initiative (Bradbury, 2010). Preliminary heating or "roasting" of activated graphite evokes an early release of ^{14}C due to release from pore surfaces, ability of recoil atoms, etc. Significant parts of the ^{14}C inventory can be selectively extracted because most of the ^{14}C may be adsorbed on the surface of the crystallites in the pore structure and not integrated into the crystal lattice (Mason and Bradbury, 1999).

As an accompaniment to thermal treatments, steam reforming is an alternative method for decontaminating graphite from radionuclides. The decontamination rates are even higher in comparison to pure thermal treatment in an inert atmosphere, as was first evidenced by basic experiments in the HTR-N/N1 project. The early release fraction can be collected and undergo isotope separation or direct recycle of ^{14}C. The remaining fraction may be very useful for recycled products since the ^{14}C levels are lowered.

Pyrolysis or steam reforming of materials in a low oxygen environment is being considered for this preliminary heating step as a decontamination technique and as a waste processing method (Mason and Bradbury, 1999; Fachinger, 2010). Pyrolysis in an inert atmosphere, such as argon or steam reforming, seems to be a possibility to separate large amounts of ^{14}C. Initial experiments performed under an inert atmosphere showed a high selective removal of ^{14}C from graphite. A higher ^{14}C decontamination factor was obtained when pyrolysis was conducted in a steam atmosphere, but selective removal of ^{14}C decreased. Further research will be to use a combination of inert and steam atmosphere processes to optimise the ^{14}C release rates for an industrial process development. Off-gases are treated to remove other radionuclides like ^{36}Cl as well as ^{14}C (Fachinger, 2010). Thermal treatment with reactive gases is also being considered.

Table 4.2: Initial feasibility assessments for gaseous centrifuge ^{14}C separation from off-gas

Process gas	Mean mol. weight	Mass fraction of carbon	Assessment
CF_3J	195.9 g/mol	6.1%	Proven in laboratory
CH_2JF	160 g/mol	7.5%	Very likely to work
CF_4	88 g/mol	13.6%	Lowest limit for UF_6 design
CO_2	44 g/mol	27.3%	Ligand isotopes; low weight
CH_4	16 g/mol	75.0%	Not feasible in UF_6 design

Source: Bradbury (2010).

The ^{14}C separation and capture technologies are not economical for the large amounts of CO_2. Isotopic separation methods are being considered and need further R&D. The leading candidate for separation of ^{14}C from ^{13}C in the off-gas from roasting/pyrolysis is pressure swing absorption. This technology has recently been used successfully in a demonstration project in China for capture and sequestration of CO_2 for a coal fired power plant. The isotopic separation uses CO or CO_2 as the chemical form at a temperature of 30°C (Bradbury, 2010). Gaseous centrifugation separation is being proposed and investigated by AREVA and URENCO, operating uranium enrichment facilities in Germany, the Netherlands, the United Kingdom and the United States (URENCO, 2006).

Remaining considerations to be reviewed include: *i)* a quantitative estimate of the separative work needed as a function of process gas and the degree of ^{14}C enrichment; *ii)* determination of the rudimentary cascade design (number of stages) for a multi-isotope feed with isotopes ^{12}C, ^{13}C, ^{14}C; *iii)* an estimate of the enrichment cost based on the current market price for uranium enrichment; *iv)* the additional cost for the preparation/ conversion of the process gas has to be considered (Bradbury, 2010).

Amine-Carbamate was an unsuccessful proposal in the United Kingdom following lab-scale trials, but a Romanian laboratory claims to have greatly increased success recently, depending on small differences in chemical rates for the different isotopes (Jones, 2010; Takeshita and Ishida, 2006; Dronca, et al., 2011). Cryogenic distillation or separation is also being investigated. Investigation directed at improving ^{13}C separation using this technology may be applicable to ^{14}C off-gas separation (Dulf, Festila and Dulf, 2009; Muntean, Stuckert and Abrudean, 2011; Li, et al., 2010). Similar technologies and issues are being considered for gaseous waste management for the recycling of nuclear fuel and may offer an opportunity for synergy and more targeted R&D expenditures (Paviet-Hartmann, Kerlin and Bakhtiar, 2010; Strachan, et al., 2009). Unique quantum characteristics of carbon nanotubes have been applied for high-efficiency enrichment of deuterium and may provide enhanced isotopic separation options (Wang and Bhatia, 2009a, 2009b). In addition, the enriched ^{13}C and ^{14}C by-products of the separation process may have useful applications in isotopically engineered nanotube construction (Simon, 2009). Laser-based separation and enrichment are also being evaluated; isotope separation by laser ionisation (AVLIS) has been developed in recent years. This approach is highly selective but requires multiple (typically three) high-power pulsed lasers for efficient ionisation. Another laser-based method, SILEX, relies on molecular excitation and similarly requires high-power lasers. A lower power laser isotope enrichment (LIE) method was also proposed for lithium and barium isotope separations, but was not demonstrated to produce sufficient quantities or levels of enrichment. A new approach of magnetically activated and guided isotope separation has been suggested and tested for the production of isotopes such as ^{7}Li for the medical industry (Raizen and Klappauf, 2012). Applications such as Separation of Isotopes by Laser Excitation (SILEX) are already being developed for ^{12}C purification for the semi-conductor industry with plans to sell the ^{13}C by-product for medical imaging uses.

- Decontamination of large components

In general, the use of various chemical decontamination methods has successfully been applied to large component decontaminations at operating and decommissioning facilities; hence the working group did not feel that any further research was necessary (Jamieson, 2001; EC-CND, n.d.a). Improvements to chemical decontamination methods using particulate photocatalysis are being investigated to provide a more specific decontamination technique. The use of photocatalytic, reductive manipulation of metal ion valence states in order to improve heavy metal deposition has already been extensively studied, with reductive manipulation also being achieved with uranium and plutonium simulants (Ce). Recent studies explore the use of photocatalysis in oxidative manipulation of metal valence states. The objective was creating a hole-driven metal dissolution process that would provide improved area specificity with an equal dissolution power and reduced secondary waste production in comparison to current chemical decontamination techniques (Wilbraham, Boxall and Taylor, 2012).

A limitation of full system chemical decontaminations is that they require the facility to be in relatively good working order to prevent leaks and spills from valve packing, pump and flange gaskets, etc. This precludes the application of this decontamination method for facilities that have been shut down for prolonged periods of time. IAEA STL Publication 1502 (2006b) notes that it is estimated by the OECD Nuclear Energy Agency (OECD/NEA) that about 400 commercial nuclear power plants will be decommissioning between now and 2050, which may result in more than 5 million tonnes of scrap metal suitable for recycling. Taking account of all other types of nuclear installations that will also be decommissioned, the amount of scrap metal available from nuclear decommissioning in the coming decades has been estimated to be as high as 30 million tonnes. The IAEA also makes note of the costs for loss of production while the decontamination is in progress. This cost could be minimised by performing decontamination after component removal and off critical path. Both United States and European companies have developed central processing facilities that specialise in processing large components, such as steam generators and reactor heads, for recycling and waste minimisation in response to this need. Either centralised or modular on-site processing capabilities need to be considered and developed for recovery of decommissioning scrap metals, in addition to large components, since shipment of other, less dense scrap metal wastes to a remote centralised processor is often not cost effective. On-site decontamination systems for scrap waste metals also need to be considered for R&D.

The working group felt that two issues related to large component decontamination warranted further research and development:

- gaseous diffusion plants and sodium reactors – treatment of passivated waste such as neutralised sodium waste;
- ice pigging of pipes as a decontamination method.

Both sodium and uranium hexafluoride are typically passivated using steam and hydrogen to convert them to sodium hydroxide and hydrofluoric acid, respectively (Peter, et al., 2006). Detroit Edison's Fermi 1 liquid-metal-cooled fast breeder reactor and Idaho National Laboratory's fast breeder reactor, EBR II, recently had their sodium successfully passivated and the waste treated. Both reactors had been treated with carbon dioxide gas to form a bicarbonate layer over the sodium, creating "passivated sodium". At Fermi 1, superheated steam was injected into a heated, nitrogen inerted system to passivate the sodium left in the system. The by-products of this reaction are caustic sodium hydroxide, hydrogen gas and heat. The waste sodium hydroxide was captured in the off-gas and by flushing the systems. The waste water was incinerated in Tennessee. The AMANDA process using ammonium to remove sodium was also used there.

At EBR II, sodium was passivated using citric acid. The citric acid solution was slowly pumped into closed-loop piping by adding about 1 200 gallons of treatment solution to dissolve sodium bicarbonate and treat elemental sodium. This yields a sodium-citrate product, thus forming a buffered system that is also cleaned up using standard water processing techniques. Passivation of uranium hexafluoride with water or steam, and uranium precipitation by high pH adjustment to separate the uranium from the liquid waste, are standard practices for gaseous diffusion and gaseous centrifuge enrichment facilities. The separated uranium precipitate is removed as filter cake and disposed of (Jones, 2011; Mendiola, 2011; Young, et al., 2011; NDA, 2010c; Peter, et al., 2006).

The United Kingdom has used a similar approach to decommissioning sodium-cooled reactors. Over 1 500 tonnes of sodium used as reactor coolant have already been destroyed during the decommissioning of Dounreay, Britain's centre of fast neutron reactor research until 1994. Of the 57 tonnes of sodium and potassium mix (NaK), which was the coolant for the Dounreay fast reactor, almost 14 tonnes have been destroyed and DSRL is on schedule to destroy all 57 tonnes by 2013. This material is heavily contaminated with radioactive caesium from the fuel used in the core. Its destruction is one of the national priorities of the United Kingdom government's Department of Energy and Climate Change. The plant lifts the highly reactive alkali metal from the reactor system in 200-litre batches.

Each batch is reacted with water in a nitrogen atmosphere to create a hydroxide solution that is neutralised with nitric acid. The process turns it into 2 000 litres of effluent that is about twice the strength of standard household drain cleaner. But it still contains high levels of radioactive caesium, so an ion exchange unit loaded with caesium-specific resin is used to process the neutralised effluent. This has been so successful that the level of caesium in the effluent discharged to sea is barely detectable (NDA, 2010c).

An ammonia-based process called AMANDA (acronym for Alkali Metals AND Ammonia) technology eliminates the pyrophoric hazard of neutralising sodium by quickly converting sodium to sodium hydroxide. The AMANDA process is an inherently safer treatment technology, completely dissolving alkali metals rather than passivating them in place. Using this chemical process in Solvated Electron Technology (SET), a non-thermal alternative to waste incineration, residual and bulk alkali metals can be removed from metal components. SET technology was designed to treat mixed waste and hazardous soils and liquids to enable landfill disposal. It has been deployed commercially at many DOE, DOD and commercial sites throughout the United States since 1995. In these deployments, sodium is injected into the system as a reagent that dissolves in ammonia prior to the addition of waste materials. The solvated ammonia solutions attack the chemical components of the waste materials to destroy the hazardous constituents. The solvated solution is reduced to a suitable waste or product form by addition of water or other established chemical techniques. The primary output is simply sodium hydroxide, NaOH. Neutralisers may include HCl, H_2SO_4, HI, H_3PO_4, HNO_3 and others, converting to chlorides, sulphates, iodides, phosphates or nitrates. Water is evaporated, leaving a dry compound that is collected into 55-gallon drums for disposal as low-level waste (LLW). Note that Na_2SO_4 does not dissolve in water and makes an ideal candidate for landfill disposal, compared to other compounds (Rogers, Foutz and Smart, 2011).

The NDA commissioned Bristol University to investigate using ice to clean pipes on nuclear sites. Sellafield Ltd and Magnox North Ltd also contributed to the work with funding and support of the practical work on sites. Other industries use “pigging” to clean pipe walls by pushing a solid plug-like object (known as a “pig”) through a pipe. What is novel about this new approach to pigging is the innovative use of crushed ice in water, known as “ice pigging” and patented by Bristol University. Crushed ice pigs can negotiate bends or minor changes in pipe work from 1 to 36 inches in diameter. Once a quantity of ice has been pushed into the pipe to form a pig, water pressure then pushes it through the pipeline. It does not lose its shape as the pig is pushed through the pipeline, allowing it to clean the entire pipe surface. This results in a highly fast and efficient system, improving on conventional methods, not just by cleaning the pipes and reducing time taken but also dramatically reducing the cost per metre. Ice pigging has the potential to accelerate work already identified in lifetime plans, while protecting the health and safety of workers and significantly reducing the amount of radioactive effluent that might have been generated by previous pipe-cleaning techniques. It does not require special, purpose-built launch or receive stations, and unlike a fixed size, solid pig it will not get stuck if the pipe is partially obstructed (NDA, 2009b). Inactive trials have been commissioned to provide the technical underpinning for any future active deployment. The areas of examination covered are (Sellafied Ltd, 2012d):

- conditions to create a self-blocking scenario;
- alternatives to “salt” as an additive to sustain the ice pig;
- an understanding of ice pig functional longevity, e.g. volume per time/distance;
- multiple segment ice pig “trains”.

Future suggested R&D for new physical processes and chemical processes for decontamination

- ***Description*** – Develop more effective and efficient *in situ/ex situ* decontamination methods for decontamination of concrete, graphite, transuranic, sodium-cooled reactor and gaseous diffusion passivated wastes.

- ***Objectives*** – Develop *in situ* (prior to disassembly) and *ex situ* (after disassembly or removal) decontamination processes that provide higher decontamination factors and integrate with the decommissioning and material removal and sentencing processes.
- ***Desired deliverables*** – Physical and chemical decontamination technologies, methods and applications that have been tested and compared for a wide range of radionuclides and waste forms and that provide increased decontamination factors and efficiencies.

Concrete remediation and regeneration

Challenges

Chemical and mechanical means of concrete decontamination have already been discussed and, as noted previously, this decontamination theme relates mainly to the *in situ* preparation of concrete prior to demolition. This poses challenges for effectively integrating concrete decontamination into the decommissioning process. Concrete decontamination tasks are difficult to perform in parallel with other work. These shortcomings are an obstacle to optimising the waste hierarchy and maximising the clearance, recycling and reuse of the material. Due to the depth of contaminant absorption in areas like ponds trenches, sumps, etc., substantial concrete and surface areas can require remediation to meet SAFSTOR, open-air demolition or intact license termination criteria. Other issues concerning concrete at decommissioning facilities relate to the demolition and removal processes that require dust suppression that, if significant levels of contamination remain during demolition, can lead to site cross-contamination issues. It is not feasible for highly activated concrete decontamination, and conventional demolition and cutting techniques create a cross-contamination hazard due to dust generation and dispersal. Removal of contaminated concrete dust from piping and equipment can also pose a challenge for decommissioning facilities. The working party recommended the following R&D initiatives to address these issues.

Development of cost-effective ex situ decontamination and paste/aggregate separation methods as part of concrete processing for recycling and reuse would reduce the scope of concrete decontamination necessary prior to structure demolition and still optimise the waste hierarchy. Post-removal, ex situ decontamination of concrete and concrete aggregate will be discussed as part of waste management in Theme 5.

Summary of current R&D for concrete remediation, regeneration

- Laser cleaning process for painted walls

As noted above in the subsection *Decontamination of concrete*, laser ablation of painted or coated concrete surfaces is continuing to be developed and evaluated (Boing, 2011; Daurelio, et al., 2010). It appears that these systems have reached the state where they are ready or nearly ready for deployment and use at decommissioning facilities. The United States Navy has tested laser ablation paint removal systems for paint stripping on steel that could be applicable to nuclear decommissioning applications. The lasers cover a reasonable surface area, have a vacuum system to capture vapours and particulate contaminants, and appear to be adaptable to end effectors on remotely operated equipment (Oller, 2011).

Figure 4.2: OSH 80 laser paint stripper

Source: Jones (2010).

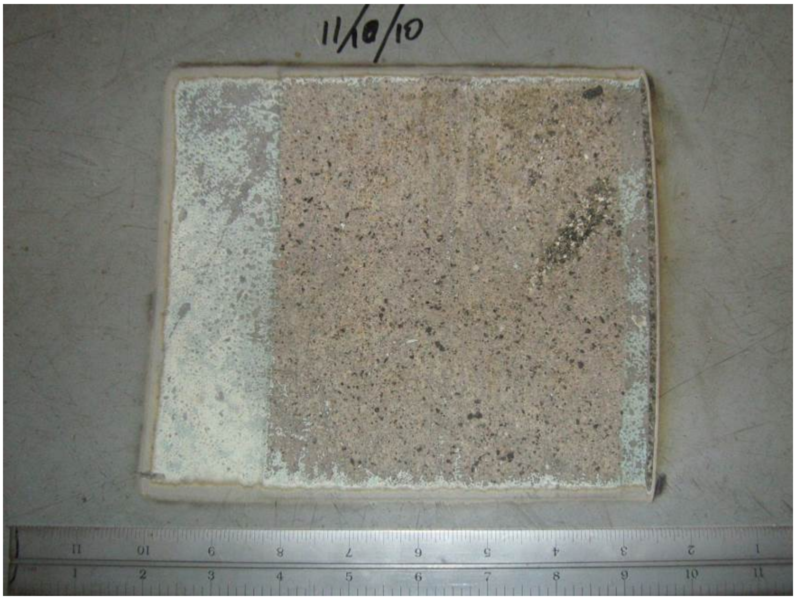
Figure 4.3: CleanLaser CL1000 Q-switched

Source: Oller (2011).

As another example of the state of the technology, EWI has been conducting internal research and development on laser-based coating removal. The majority of this work to date has been conducted on aircraft coatings on thin aluminium substrates. However, they have also done some work on low-observable ("stealth") coatings, marine coatings and carbon-fibre composite substrates. The scanner system removes the coating or substrate and collects the fumes in an exhaust hood. In 2010 EWI tested stripping a 6-inch path on a 12+ year old epoxy paint from the concrete floor in the laser lab. The epoxy paint was vaporised using a 15-kW fibre laser and sucked out through the exhaust system. The resulting floor surface can be seen in Figure 4.4.

The procedure took three passes at 10 kW. On the third pass, the laser was turned on later (2 inches from the start at the left). This shows the previous paint colour (light blue). On the right side of the stripe is the bare concrete surface after all paint was removed (Victor, 2010). EWI has also developed a robotic end effector, in addition to the one depicted in Figure 4.4, and conducted tests at the Southwest Research Institute (EWI, 2011).

Figure 4.4: Concrete floor covered with 12-year-old epoxy paint after EWI laser stripping



Source: Victor (2010).

PaR Systems has developed a ~1 000 W and 2 000 W high repetition rate, high power CO_2 laser system tested on Airbus and Boeing. The system is capable of achieving stripping at rates as high as 10.7 m^2 hr^{-1}. According to PaR, techno-economic studies have shown that the laser can out-perform the current conventional chemical stripping processes at this rate (PaR Systems, 2014). Sophisticated robotic systems that monitor paint thickness and adjust paint stripping rates have also been developed.

General Lasertronics Corporation also provides robotic laser ablation paint stripping applications for surface coating removal on all substrates (GLC, n.d.a). The Automatic Rotor Blade Stripping System (ARBSS) developed for aircraft combines the closed-loop, colour control laser work head with an industrial robotic system. It employs three colour-selective lasers that operate simultaneously and automatically to strip rotor blades. Once the rotor blade is secured in the fixture, the system requires no human intervention or observation. The ARBSS won the 2009 Maintenance Excellence Award from the United States Office of the Secretary of Defense and the 2010 Defense Manufacturing Excellence Award from the Aerospace Industries Association. The system reduced the time-to-strip by 90%, from about 24 hours (using manual labour and rotary sanders) to about 2 hours per blade at the H-53 rotor blade stripping facility at the Navy's Fleet Readiness Center-East. The Robotic Automated Coatings Removal System (RACRS, currently under development) extends this concept to automatically stripping an entire aircraft. The initial system is designed to strip the Navy's V-22 Osprey, but can be extended to other airframes.

CTC is an applied scientific research and development services organisation contracted by the United States Air Force to develop a robot-mounted laser ablation scanner capable of removing partial or entire paint layers from the surface of an aircraft. The Air Force requires a very accurate thickness measurement of the various layers. The Advanced Photonix, Inc. paint removal system, including an API T-Gauge® system coupled with a high-speed line scanner, will be an integral part of the laser paint removal process. The API THz system will scan the surface and develop a three-dimensional map that will provide feedback to the robotic motion control and accurately move the laser for proper paint removal (API, 2014).

Embedded contamination in the concrete can also be removed by laser scabbling of concrete. Cambridge-based TWI was contracted by the Nuclear Decommissioning Authority in the United Kingdom to develop single point tube cutting and concrete laser scabbling capabilities. In the laser scabbling process, the laser beam is applied to the surface of the concrete and its energy is absorbed, heating the concrete matrix and the concrete aggregate. Expansion of residual water vapour, probably in both the matrix and aggregate (and differential expansion between aggregate and matrix), causes the concrete to break up in a highly energetic fashion, leaving a rough scabbled surface consisting of matrix and aggregate. Scabbled concrete is collected in a HEPA-filtered system similar to a Hi-Vac. Some of the debris leaving the concrete surface was up to 20 mm in size. However, motion through the system reduced the size of the particles and the resulting debris had a high packing density. Work performed has indicated that a 5 kW power laser removed 1 m^2 of material to a depth greater than 10 mm in 110 minutes. A slower process speed will generally result in a deeper scabbled section. For concrete containing limestone aggregate, the deepest section has been measured at 22 mm, using a laser power of 5 kW and a travel speed of 100 mm/minute. For removal of large surface areas, a track overlap of 50% proved to be the most effective for producing a uniform depth in the scabbled profile. The process appeared to be independent of the attitude of the concrete. In multi-pass processing of the same track, the amount of concrete removed was seen to drop at each successive pass. For example, at 5 kW laser power and 300 mm/minute travel speed, the maximum depths of scabbling recorded for three successive passes of the beam were 10, 18 and 22 mm, respectively. Surface contaminants such as grease and paint had no effect on the scabbling process (Hilton and Khan, 2010).

AREVA has performed research on particle sizing and distribution in aerosols generated by laser ablation that should aid in the efficient design of ventilation capture systems to minimise secondary wastes associated with laser ablation coating stripping (Dewalle, et al., 2010).

Thus it appears that currently there are advanced systems available and eligible for further field testing and use at decommissioning facilities, and that they could greatly automate the decontamination process.

- Wiping and scabbling

Wiping or washing techniques are simple, easy, inexpensive and effective for removal of loose surface contamination. At this time, scabbling is the most effective method for removing embedded contamination in concrete and can be accomplished remotely with robotic end effectors. The rapid development of (NDA, 2010d, 2012b; John, 2012), laser scabbling will ensure its replacement for historically conventional methods (TWI, n.d.). If a laser rebar cutting capability was added to a laser scabbling head (Hilton and Khan, 2010) on a single end effector, there is the potential that concrete demolition and scabbling operations could be performed more efficiently. Japan is also developing laser scabbling capabilities in the wake of the Fukushima Dai-ichi meltdown. Experiments in Japan have shown that, in the case of a laser output of 5 kW and a run of 5 metres per minute, it is possible to scale a 4.8 m^2 area every hour when removing to a depth of 0.5 millimetres from the concrete surface. Similarly, when running at 2 metres per minute, it is possible to peel a 2 m^2 area every hour when removing to a depth of 1.7 millimetres. In other words, the time required to process a 100 m^2 area (10 m $\times$ 10 m area) is approximately 20 hours for a depth of 0.5 millimetres and approximately 48 hours for a depth of 1.7 millimetres (Arai, n.d.). As seen in Table 4.3, which provides wall scabbling rates based on a recent review of the scabbling technology (OECD/NEA, 2011), the current 5-10 kW lasers cannot yet achieve the scabbling rates of conventional machines and end effectors.

Table 4.3: NEA review comparison performance of wall shaving systems

Process	Cutter type	Project	Production rate (machine working time)	Avail. rate (%)	Remarks
Two-headed milling machine on forklift (figure)	Steel	CEA – ATUE	~10 m^2/h (max. 10 mm depth)	30%	Overall yield strongly impaired by an uneven surface (blocks)
Single-head milling machine on xy-frame	Diamond-tipped rotating disks	BP – Eurochemic	15-25 m^2/h (3 mm depth)	20%	Overall yield impaired by set-up time (~1 day)
Milling machine on Brokk carrier (figure and figure)	Diamond-tipped rotating disks	CEA – EL4	8 m^2/h (3 mm depth/pass)	50%	
PLB milling head (figure)	WC teeth	CEA – EL4	1.2 m^2/h (25 mm depth/pass)		Heavy tool, rough finishing
PLB milling head (figure)	WC teeth	CEA – AT1	1.5 m^2/h (25 mm depth/pass)		Heavy tool, rough finishing

Note: “Figure” indicates that the report contains an image of the tool.

Source: OECD/NEA (2011).

The depth will be increased when making several passes over the same area. Processing is possible for a wide area by gradually moving to the side. Although the removal efficiency is relatively high, a significant amount of time is required. However, the processing efficiency can be improved by increasing the laser output. Although the processing width is greatly affected by the laser output, it is possible to process a width of 50 to 60 millimetres in one pass when using an output of 5 or 10 kW (Arai, n.d.).

Continued development of this technology, increasing laser output and fibre length, could help further expedite extended scabbling operations, such as those at Trawsfynydd for the ponds’ scabbling that have a Magnox Optimised Decommissioning Programme (MODP) completion date of 2014, which was optimised from an LC35 completion date of 2022 (Parsons, 2007).

- Diamond wire cutting

Diamond wire cutting consists of a series of pulleys that draw a continuous loop of multi-strand wire equipped with a series of diamond beads. The wire is wrapped around the object to be cut and contact tension is applied on the wire by the pulleys. The spinning wire cuts a path through the concrete or even metal. Almost any thickness can be cut with this technique. Diamond wire cutting is versatile and has been used for cutting openings in containments and biological shields at operating and decommissioning facilities (Paratore, 2011; Ramanathan, 2012). Current uses require substantial set-up time that may include coring holes through which diamond wire is threaded. This can limit its use in high radiation or hazardous environment situations. Controls are required for highly contaminated items to reduce the possible spread of contamination due to swarf, which can be carried from the cutting area by the wire. This can result in significant secondary waste when water cooling of the wire is used. It is also possible to cut in dry conditions when the cutting wire is cooled by local injection of cold compressed air (-10 to -15°C). Dust emissions can be reduced using a sealed collection system located at the outlet of the wire. Dry cutting of reinforced concrete has been successfully demonstrated and applied at BR3 (Baryte concrete), Rheinsberg, KNK, the CIEMAT PIMIC project and WAK (OECD/NEA, 2011).

Dry diamond wire cutting was also performed in the United States to sever hot leg and cold leg nozzles close to the reactor vessel at Connecticut Yankee. HEPA ventilation on the enclosure at the outlet of the wire proved effective in controlling airborne radioactivity even though hot spots were present on the nozzles due to the thermal sleeves. Development of diamond wire end-effectors used for off-shore underwater cutting may have applications in decommissioning that warrant further R&D (Molfino and Zoppi, 2012).

- Nitrogen and carbon dioxide blasting

The initial form of CO_2 blasting applied in the nuclear industry was dry-ice blasting using CO_2 pellets. While this type of blasting can be very effective as a surface cleaning method, it has very limited abrasive force for the stripping of pure concrete and therefore is rather ineffective as a scabbling alternative. Enhancements for concrete abrasion additionally use a combination of laser and CO_2 ice blasting to increase local differences in temperature to cause spalling. The reachable abrasion depths in concrete are 5 mm. In contrast to all other blasting techniques CO_2 ice blasting does not produce any secondary waste as the dry ice immediately sublimates. At its current state of development this technology is not sufficiently abrasive to strip coatings or fixed contamination on a concrete substrate (OECD/NEA, 2011; Rigby, 2011). Liquid nitrojet jetting blasts liquid nitrogen at the surface under high pressure, and is capable of removing a thick layer of material (up to 30 mm in a single pass). Abrasives can be added to improve cutting. Liquid nitrojet jetting has been used successfully at West Valley to remove PBS coating on hot cells (Rigby, 2011). AREVA has used it successfully to decontaminate, cut or scabble different types of surfaces as metals, polymers and concrete (Moggia, 2011; AREVA, 2010). The following advantages have been clearly shown (OECD/NEA, 2011):

- The system is versatile, allowing the selective removal of coatings and paint from surfaces as well as the removal of a variable thickness of concrete (3-30 mm in a single pass).
- No generation of secondary waste.
- Insensitive to surface state (roughness, metallic inserts, etc.).
- An efficient process for coating stripping (10 m^2/h) and for concrete removal (2.5 m^2/h) for a 14 mm pass.
- It is operated remotely.

A drawback to these technologies is the health and safety factor due to the precautions required to work with cryogenic materials and the potential to create oxygen deficient atmospheres.

- Laser cutting

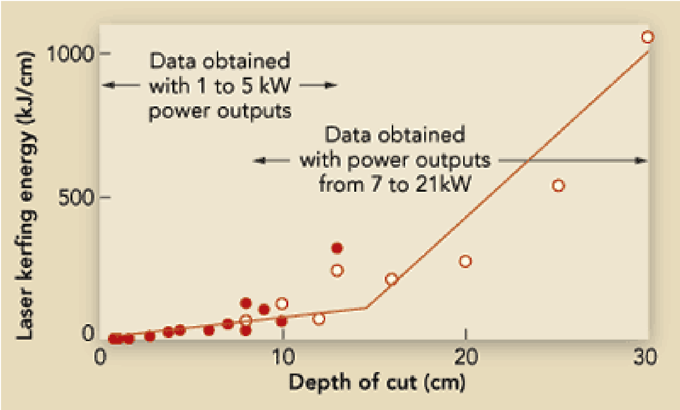
Because of their low generation of fumes and dust, and ease of remote control, the use of lasers for cutting concrete in the decommissioning of nuclear reactor facilities is an attractive proposition. Laser cutting of concrete has been evaluated for a number of years (Tirumala Rao, Kumar and Nath, 2005; Wignarajah and Nagai, 2005). The energy input required for cutting rises rapidly when the thickness of the concrete exceeds about 10 cm due to the fact that expulsion of the viscous, molten material produced in the kerf becomes difficult with increasing depth. The maximum thickness that can be cut with a single pass is about 30 cm (Wignarajah and Nagai, 2005).

Current technologies are capable of cutting concrete a few inches thick (Wignarajah and Nagai, 2005; Muto, Tei and Fujioka, 2007; Muto, et al., 2008). TWI Ltd demonstrated that a laser cutting head was able to cut thick-walled pipes, 25 mm thick 304 stainless steel plate, 50 mm thick C-Mn steel plate and concrete up to 87 mm thick. The LaserSnake project demonstrated single-side laser cutting for remote disassembly in confined spaces (OC Robotics, 2011).

Higher-powered lasers are being developed for the military by combining fibre-laser beams to get a high-quality single beam, either through coherent or spectral combination.

The result will be higher power in a single transverse mode beam. Developments such as these will improve cutting capabilities and efficiencies and warrant further research for applications to decommissioning (Hecht, 2012).

Figure 4.5: 2005 reported laboratory test results on laser cutting of concrete



Source: Wignarajah and Nagai (2005).

Future suggested R&D for concrete remediation, regeneration

- ***Description*** – Perform research into improved methods and technologies for removal of surface contamination, contaminated coatings and embedded contamination in concrete.
- ***Objectives*** – Continue to improve conventional decontamination by development of more effective chemical washes, treatments and strippable coatings. Optimising decontamination through the use of complimentary decontamination approaches, such as washes or chemical coatings coupled with electrokinesis. Develop more efficient and powerful tooling and automated, remote delivery systems. Develop decontamination approaches that compliment or expedite the de-planting effort rather than compete with it for space and resources. Continue improvement of conventional scabbling equipment and development of alternate approaches that: *i)* are easier to integrate into the de-planting process, such as laser scabbling; *ii)* will minimise secondary waste, such as nitrojetting.
- ***Desired deliverables*** – Better understanding of the mechanisms underlying concrete decontamination processes. More comparisons of decontamination effectiveness and efficiencies for radionuclides and waste forms. Development of automated applications and technologies that require minimal set-up/prep time and that minimise secondary waste issues. Technologies that complement the de-planting process.

Surface treatment and removal of contamination

Challenges

This suggested R&D deals with the decontamination of surfaces in general, as opposed to specifically for concrete. Many of the chemical washes and strippable coating applications discussed previously are also used for other materials, such as metal surfaces like cavity or fuel pool liners, or to decontaminate components such as glove boxes, electrical cabinets, pumps, tanks or valves, or other non-metal surfaces, such as tiles or cables in cable trays. Most chemical treatment applications require curing or drying times before any work in the vicinity is permitted. One of the challenges identified by the working group was to improve the drying process by using vacuuming or supplying dry air to the area. Better oxidation/surface removal laser technologies could eliminate this issue for areas such as reactor cavities because the contaminants are volatilised, captured and removed immediately by a ventilation system integrated into the tooling design.

Abrasive decontamination methods that incorporate secondary waste capture and grit recycling, or that use cryogenic materials (CO_2 or nitrogen) which minimise secondary waste when they sublimate, can also eliminate the overall decontamination time.

Another challenge that requires better surface decontamination and polishing methods is the decontamination of tritium in heavy water systems. Due to its high mobility and the ease with which it is adsorbed and desorbed in materials, tritium can contaminate concrete and metals at depth. A related challenge is the high cost of decontaminating large volumes of water.

Summary of current R&D for surface treatment and removal of contamination

- Using gels and foams to minimise secondary waste

As was noted in the section on transuranic contamination removal, chemical washes, strippable coatings and gels have been used effectively for contamination removal on a wide variety of materials and surfaces. The use of these materials can significantly decrease the amount, activity levels and waste classification of SSC (Kohli and Mittal, 2007; OECD/NEA, 1999). DeconGel® decontaminant is used for remediation of radiological and hazardous chemical substances at DOE sites. The application was developed and tested by the DOE and sent to Japan to assist with remediation efforts following the Fukushima Dai-ichi accident. Performance data and lessons learned on the use of this and other products at Fukushima Daiichi should be captured. The DOE reports that a pilot batch production of second generation prototype formulations [DeconGel 1108 (brushable) and DeconGel 1128 (sprayable)] contain an additive that provides improved efficacy for caesium, transuranic waste and hydrophobic contaminants such as PCB. The new formulations, which are non-flammable, have improved film toughness and peelability, and have reduced foaming compared to the original DeconGels. Tests of the new formulations were successfully completed in August 2011 at the Idaho National Laboratory under the direction of the EPA's National Homeland Security Research Center through the Technology Testing and Evaluation Program (TTEP). The tests showed improved decontamination efficacy for caesium on porous concrete (67% removal after two applications) as compared to DeconGel 1101 (45%) and to the primary peelable decontamination technology competitor (34%) removal after three applications; all were tested by the same method under TTEP (US DOE, 2011b).

Decontamination factors exceeding 900 were determined using electrochemical decontamination reagents with a surface electrode on stainless steel during recent evaluations in Japan (Mikheykin, 2012).

- Washing, wiping and pressure washing techniques for metal surfaces

The amount of metals that are expected to arise from the decommissioning of nuclear facilities and from the waste segregation initiative by LLWR should be sufficient to use the NDA metal treatment facilities to the design capacity. The capacity of NDA facilities is only a fraction, 4%, of what is available from the supply chain. The supply chain has an estimated capacity of 16 000 tonnes/year vs. 650 tonnes/year from the NDA facilities (NDA, 2009d). Therefore, it is important to develop decontamination technologies for scrap metal materials associated with decommissioning. The NDA metals treatment facilities have been evaluated and can handle uncomplicated geometries at low levels of contamination. However, the amount of secondary waste was in one case very high, well over what might be expected from similar commercial facilities.

The actual mechanisms underlying removal of loose surface contamination are being researched and theories to model and calculate the effectiveness of a removal technique are being refined. A recent thesis evaluated the factors identified by the Johnson, Kendall and Roberts (JKR) theory that affect the strength of the detachment force necessary to remove a particle of contaminant from a surface, and the roughness of the surface on which the contaminant is present, for predicting the efficiency of removal of loose contamination (Calderin Morales, 2010).

The JKR theory predicts the area of contact between a particle and a surface based on their adhesion force at the interfacial region, and the particle radii. Equation 2 of the theory is used to calculate the force necessary to detach a particle from a surface. Equation 4 relates the detachment load necessary to remove a particle from a surface with the particle radii and with the physical properties of the particle and the surface. Two methods were used to reach this objective: the first consisted of quantifying the contamination by weight and the second of quantifying the contamination by counting alpha and gamma particles. As a result, it was determined that for particles of 5 μm, the interaction between contaminant-wipe and contaminant-surface were significant. However, for particles between 37-149 μm, the contaminant-surface interaction was the only significant interaction affecting the amount of contamination removed (NDA, 2009d). Research for this thesis found that the interaction contaminant-wipe only played a significant role when used on particles of 5 μm average size, but not when the particles increased from 37-149 μm. A possible reason for this was the relation of contaminant particle size and the width of the wipes' microfibre. The interaction contaminant-wipe was significant when the particle size of the contaminant was smaller than the width of the microfibre (NDA, 2009d). This type of research is a good example of how examining the fundamental mechanisms of contamination, waste form and decontamination processes can provide a basis for the development of more effective decontamination methods and strategies such as development of wipes with a range of microfibre widths.

In the area of washing and wiping, some of the foam applications that are applied and vacuumed up could also be used with a wiping or scrubbing action before they are vacuumed up. A new piece of equipment for electro acid etch of weld areas may have applicability if scaled up to cover larger surface areas for decontaminating metal surfaces (IMD, 2011). Similarly, systems to wipe down and decontaminate waste boxes using an autonomous Cartesian robotic system for encapsulated waste boxes has been designed and implemented by Magnox (Sands, 2006).

While much of the discussion on gels, foams and abrasive blasting thus far is relevant to decontaminating external surfaces of pipes, pumps, tanks, etc., there are more aggressive means using high powered hydrolasers for component internal surfaces and structural surfaces. Today's equipment may be configured in a variety of ways for use on walls, ceilings or columns, and in confined spaces, underwater or in tanks. When combined with vacuum recovery of the waste stream, waste water recycling and remotely operated tools, hydrolasing is a highly effective remotely operated decontamination technique (Sands, 2006). Most hydrolasers used for aggressive decontamination operate in the 10 000 to 40 000 psi/g range and use between 6-20 gallons of water per minute, thus providing sufficient force to remove most paint and coatings. At the higher end of the range, concrete scabbling is performed. Pressures above 20 000 psi will also remove the lightly and tightly adhered oxide layers that form the radioactive film on component interiors (Sands, 2006).

Incorporation of hydrolasers onto remotely operated equipment with shrouded collection systems and water processing capabilities has also allowed them to be deployed effectively under water to conduct decontamination without losing water clarity (Farr, 2012; Jenkins, 2011; Fisher, 2011).

Abrasive blasting is a technology that is increasingly being studied for the realisation of three-dimensional micro decontamination using brittle materials such as glass and silicon. This etching process is based on the eroding properties of Al_2O_3 sharp particles projected at high speeds against a target substrate (Kim, et al., 2011). In an effort to improve the accuracy of the process, smaller particles with abrasion-resistant photosensitive polymers, such as polyurethane flexopolymers, have been proposed. Also, high-resolution electroplated masks have been used (Kim, et al., 2000); due to its intrinsic simplicity and economic feasibility, this technique has a large application potential, even for high resolution patterning. Studsvik Nuclear AB (Sweden) used a dry tube blasting method for the decontamination of heat exchangers for the first time in 1999 (Krause, 2007; Lindström, Wirendal and Lindberg, 2007). Abrasive blasting can also be used as an alternative to hydrolasing for *in situ* decontamination using systems similar to those used for processing steam generators. Incorporation of the spent grit into grouting to fill void spaces would minimise the secondary waste cost. In addition, abrasive honing techniques are being used to decontaminate component surfaces (IMD, 2010).

There are research efforts on decontamination methods to minimise secondary waste associated with surface washing, hydrolasing and abrasive blasting. Reactive CF_4/O_2 plasma is also being evaluated for *ex situ* decontamination of metals. Such methods would eliminate secondary waste (Kim, et al., 2000; Kim, 2012).

- Supercritical fluids

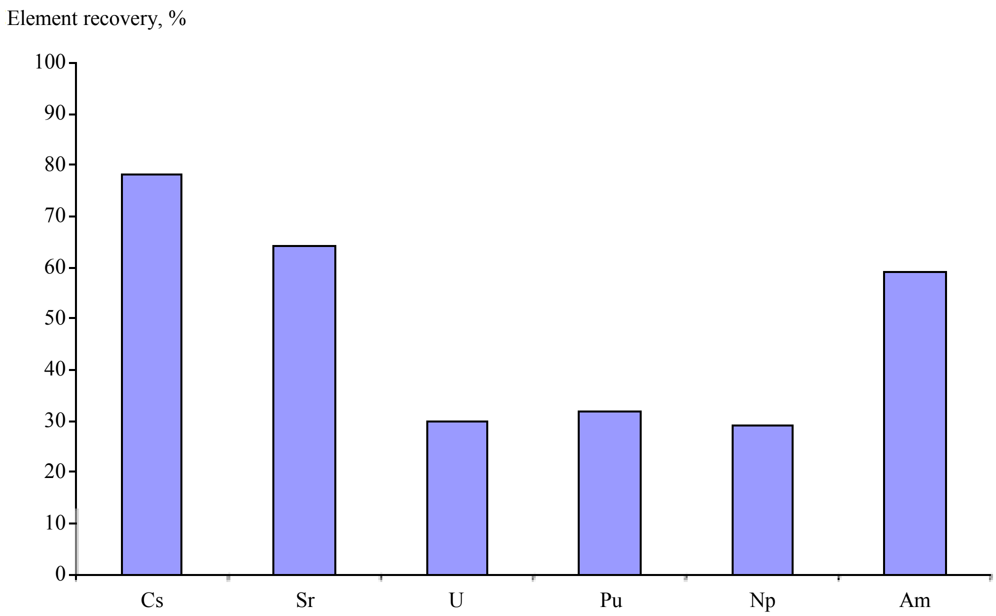
A supercritical fluid (SCF) is any substance at a temperature and pressure above its critical point, where distinct liquid and gaseous phases do not exist. It can effuse through solids like a gas and dissolve materials like a liquid. The supercritical phase results in a gaseous phase at one half the density of the liquid. When the mixture is close to the critical point, small changes in pressure or temperature result in large changes in density, allowing many properties of a supercritical fluid to be "fine-tuned". Carbon dioxide and water are the most commonly used supercritical fluids, although other mixtures are being tested for radioactive decontamination. In supercritical extraction the contaminated surface is exposed to supercritical CO_2 and radioactive materials are absorbed into the fluid. When the supercritical fluid becomes subcritical, contaminates precipitate out of the fluid and can be collected as a sludge or on filter media. The CO_2 can then be reused. The low volume of secondary radioactive liquid waste has aroused considerable interest in supercritical fluid use in decontamination technologies (Park, et al., 2012; Sawada, et al., 2005; Koh, et al., 2008; Shadrin, Murzin and Romanovskiy, 2005).

The advantages of supercritical fluids such as liquid carbon dioxide and freons have been demonstrated for the reprocessing of nuclear power plant spent fuel (NPP SNF), management of radioactive wastes (HLW), and for decontamination of equipment and work clothing. Fluids containing solutions of β-diketones (hexafluoroacetylacetone, HFA), tributylphosphate (TBP) and other neutral and acidic organophosphorus reagents like di-2-ethylhexylphosporic acid (D2EHPA) in supercritical liquid CO_2 permit extraction of the actinides (U, Th, Pu, Np, Am) and lanthanide cations in micro- and macro-quantities. Supercritical fluid extraction (SFE) using acids such as ώ-hydroperfluoropropionic acid (PFVA) may also be used for recovery of radionuclides such as ^{137}Cs, ^{90}Sr and ^{60}Co (Shadrin, Murzin and Romanovskiy, 2005).

Investigations in developing decontamination technologies and the creation of equipment for surface purification in CO2 medium are being conducted at the CEA (France), JAERI and Nagoya University (Japan), Kyung Hee University (Korea), Delft University (the Netherlands), LANL and INEL (United States) and at the Radium Institute and Mining Chemical Combine (Russia) (Shadrin, Murzin and Romanovskiy, 2005; Shimizu, et al., 2006).

Figure 4.6: Recovery of elements by water containing supercritical CO_2 into polyether – OP-7 and ώ-hydroperfluoropropionic acid

40 mg OP-7; 0.2 mmole H_2O; 30 μmole acid; 300 atm. 80°C; 20 min



Source: Shadrin, Murzin and Romanovskiy (2005).

- Laser process for surface decontamination

The use of lasers for cutting, scabbling and paint stripping have already been discussed in previous sections of this report. Fibre optic lasers capable of stripping oxide coatings have been and are continuing to be developed. As with laser scabbling and cutting, the great advantage of these systems is the ease of collecting the aerosols generated compared to chemical reagents, water from hydrolasing or grit from blasting. A laser beam hitting a surface can bring the surface temperature up to 2 000°F. Laser cleaning is based on contaminated surface ablation or heating, which results in contamination desorption. Laser ablation removes matter using high-energy laser pulses, resulting in solid material damage, similar to evaporation or sublimation. Laser ablation has been proposed as an *in situ* method for metal surface cleaning. For industrial surface decontamination and cleaning, the application of modern, reliable, powerful fibre lasers (100-1 000 W mean power) is becoming regarded as quite reasonable and efficient. Lasertronics and other companies have developed end effector-mounted and hand-held lasers capable of stripping metallic oxide layers. The Lasertronics system cleans surfaces to free-release levels (Cargill, 2011). India is developing lasers to decontaminate surfaces and MOX fuel tubes after pellet insertion (GLC, n.d.b; Gupta, 2011).

A review of previous laser decontamination studies was included in a recent doctoral thesis (Leontyev, 2011). A high-power Q-switched Nd:YAG laser was demonstrated as being very efficient in removing fixed metal surface contamination in United States experiments reported in 1995. A Q-switched Nd:YAG laser system of 100 ns pulse duration and a repetition rate that could be varied as much as 100 Hz-30 kHz was used. A 200 W maximum output power was reached when the repetition rate exceeded 9 kHz. For simulated stainless steel contaminations, this laser achieved decontamination rates of 93-100%, with most of the contamination removed in the first pass. Two types of real samples, Haynes 25 and lead brick, were also examined in the United States study (Leontyev, 2011). Haynes 25 is a cobalt-nickel-chromium-tungsten alloy with excellent high-temperature strength and good resistance to oxidising environments up to 980°C (1 795°F) for long exposures. It also has excellent resistance to sulphidation. The Haynes 25 decontamination stopped at DF = 3.46 after four passes (150 W, 5 kHz and 10 cm/s). The lead brick contamination dropped to almost background level after one pass. For a certain zone, DF was only 2 and it did not increase with more passes (Leontyev, 2011).

Results of two experiments conducted in France on metal samples from reactors were also reviewed in the thesis (Leontyev, 2011). The studies used four different wavelengths of available industrial lasers with the following parameters (Leontyev, 2011):

- Nd:YAG laser: 1 064 nm, 7 ns, 700 mJ, 30 Hz, 21 W;
- frequency doubled Nd:YAG laser: 512 nm, 7 ns, 250 mJ, 30 Hz, 7.5 W;
- XeCl excimer laser: 308 nm, 30 ns, 300 mJ, 5 Hz, 1.5 W;
- KrF excimer laser: 248 nm, 30 ns, 400 mJ, 1 Hz, 0.4 W.

Three types of samples were used: *i)* 304 stainless steel samples contaminated with ^{137}Cs but with no data on the initial activity; *ii)* samples of vapour generator tubes of 304 stainless steel that had ^{60}Co at 105 Bq/cm^2 and an oxide film thickness of 5-20 μm; *iii)* samples of vapour generator tubes of Inconel with initial ^{60}Co contamination levels of 20 500 Bq/cm^2.

Different types of contaminated surfaces and ablated matter collecting systems were used in the experiments. Cleaning was performed in air under flowing water film and under film containing nitric acid. Other experimental results indicate that the decontamination factors (the ratios of the concentrations of contaminant before and after cleaning) can be dramatically increased when laser ablation is performed under a layer of water, acid wash, gels and other coatings (Padma Nilayav, Biswas and Kumar, 2010; Kameo, Nakashima and Hirabayashi, 2004; Won, et al., 2011).

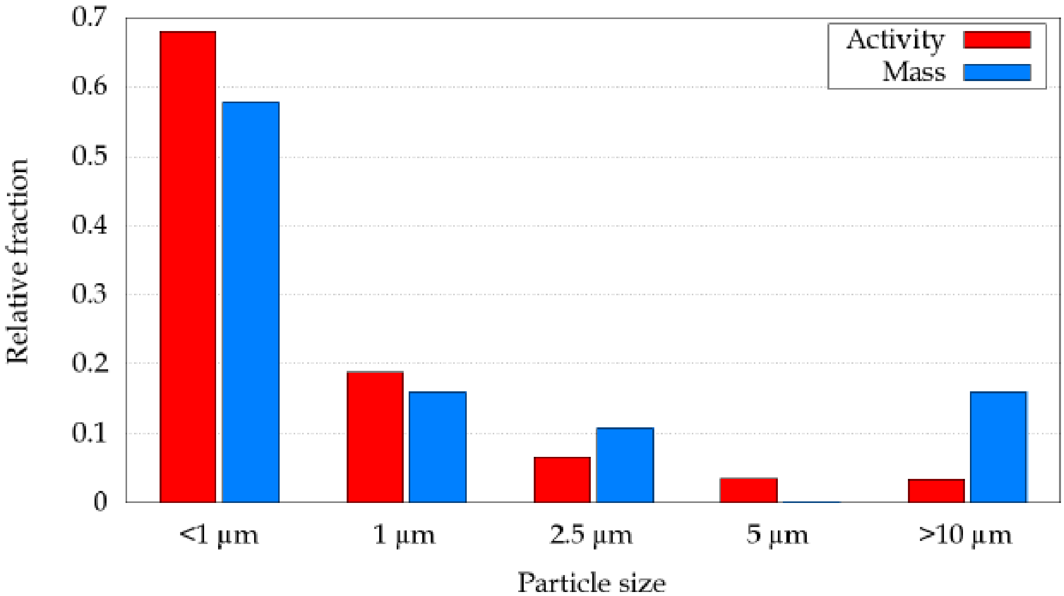
Liquid-assisted laser treatment has proven to be more effective than non-treated dry laser ablation of surface oxides. Under laser pulse action, water can turn into vapour and initiate a shock wave that can expel particles from the surface. It was demonstrated that the particle removal from the surfaces by laser light was more efficient with the presence of water on the surface than in ambient air. This water-assisted surface cleaning from particles is also referred to as "steam laser cleaning" (SLC). In the case of steam cleaning, a liquid film or capillary-condensed water on the surface is superheated by a laser pulse and subsequently results in its explosive evaporation. The expanding vapour ejects the particles from the surface (Leontyev, 2011).

For ablation in air with aspiration, the decontamination factor does not exceed 10, even for very high incident fluences. In contrast, the factor increases rapidly with the incident fluence for ablation under a water film. For ablation under diluted nitric acid, the factor can achieve 300. In almost all cases, the "best wavelengths" were 1 064 nm and 532 nm (Leontyev, 2011). The feasibility tests to select the most appropriate light source were performed with surrogate samples using a continuous CO_2 laser, a continuous Nd:YAG laser and a pulsed Nd:YAG laser. The pulsed Nd:YAG laser was found to be the most efficient among them (Leontyev, 2011).

For decontamination of a hard alloy (such as Inconel), the decontamination factor significantly increased when the sample was covered by HNO_3, as opposed to just air or water. It was also shown that with increased acid concentration, the decontamination factor increased for the same total incident fluence (Leontyev, 2011).

A French project focusing on the excimer XeCl laser for decontamination of metal components was launched at CEA Saclay in 2000. The system was designed and used for decontamination of real samples from vapour generator tubes made of stainless steel and Inconel. Aluminium samples from the fuel waste retreatment facility were also under evaluation in other studies. In these experiments, a XeCl laser (80 W mean power) and optical fibre transport of the laser beam to the sample was used. The results were "quite satisfactory" for stainless steel and aluminium; for Inconel, however, the decontamination factor did not exceed 2. This result may be associated with the presence of deep cracks on the surface of the Inconel sample under study. For the stainless steel, these cracks filled with the oxide were less important than for Inconel, where they were deeper than 150 μm (Leontyev, 2011). In Russia, studies using 100-150 W lasers at the Bochvar Research Institute attained a decontamination factor of about 20 on artificial samples. Experiments on real samples (the pieces of Raschig rings) revealed decontamination factors in the range of 20-82 after three passes. Simultaneous particle size studies indicated that the stainless steel sample ejected particle sizes of less than 1 micron on average (Leontyev, 2011).

Figure 4.7: Bochvar Research Institute size distribution of particles produced during cleaning of stainless steel



Source: Won, et al. (2011).

As a continuation of these studies, a prototype mobile laser decontamination system was scheduled for construction at the end of 2011. This system will be able to handle metallic tubing constructions of a complex surface of 0.5-1.5 m diameter and up to 2 m length (Leontyev, 2011).

Better understanding of the decontamination rates attainable with lasers is required. One of the aims of the study that provided the above background on laser decontamination was to conduct experiments on the effectiveness of laser ablation (decontamination factor, the rate of ablation, damage to the metal substrate) for cleaning metal surfaces with a contaminated layer. Radioactive contamination of the oxide layer was simulated by introducing europium (Eu) and sodium (Na) as mock radioactive contaminants. A decontamination factor of 300 for the Eu-contaminated sample was obtained after deposition of 372 J/cm^2. For the Na-contaminated sample, the decontamination factor was 1 500 after deposition of 186 J/cm^2. Such decontamination factors are considered sufficient to clean radioactive wastes. A productivity rate of 0.001 m^2/W × hour was demonstrated for oxidised metals. The study used a DetriLaser of the specifications shown in Table 4.4.

Safety interlocks, laser focal lengths and target shapes need to be considered in the development and deployment of lasers for surface decontamination.

Table 4.4: Specifications of the DetriLaser set-up

Wavelength	1.06 μm
Average laser power	2-20 W
Pulse repetition rate	20 kHz
Pulse energy	From 0.1 mJ up to 1 mJ
Pulse duration	120 ns (full width at half maximum)
Beam quality factor	$M^2 = 1.5$
Laser beam radius in the waist position	53 μm at the 1/*e* intensity
Laser fluence in the waist position	1-10.5 J/cm^2
Scanning speed	Up to 9 000 mm/s
Maximum scan field	290 × 290 mm^2

Future suggested R&D for surface treatment and removal of contamination

- ***Description*** – To develop more effective and efficient physical and chemical decontamination methods for removable and fixed contamination removal from metallic waste substrates. Investigate improvements to current methods such as wiping, washing, gels, foams, strippable coating, hydrolasing and abrasive blasting. Continue development on supercritical fluids and wet/dry laser decontamination to achieve greater understanding of effectiveness and productivity and to develop field-testable equipment. Consider and investigate overall decontamination effectiveness and efficiency improvements by using a combination of methods.
- ***Objectives*** – To develop new approaches to decontamination, improve existing under-developed equipment and methods and to reach a greater understanding of their proper applications, effectiveness and efficiencies.
- ***Desired deliverables*** – New surface decontamination equipment, methods and reagents that can be used for *in situ* and *ex situ* (post-removal) of metallic SSC. Continued testing and development of potential decontamination systems such as lasers or supercritical fluids for commercial applications and viability.

Heels and residues

Challenges

- Removing heels and residuals resulting from fuel reprocessing

Clean-up of tanks containing highly radioactive wastes associated with spent nuclear fuel reprocessing and high-level wastes has proven to be a difficult and drawn-out process. These wastes are typically stored in tanks or vaults with heels and residuals remaining after waste removal and stabilisation, such as vitrification of high-level waste (HLW) are complete. The methods and processes used to remove bulk nuclear waste from waste tanks have been implemented at many nuclear facilities around the world. However, after the bulk volume of the waste is removed from accessible tanks, a significant amount of residual highly radioactive waste can remain in the heel (Martin and Rood, 2011).

A tank heel is the amount of liquid remaining in each tank after removal of the bulk waste to the greatest extent possible by use of the existing transfer equipment, such as ejectors. The tank residual is the amount of radioactive waste remaining in each tank, the removal of which is not considered to be technically and economically practical. This could be the tank heel or the amount of radioactive waste remaining after additional removal using methods other than the existing transfer equipment (US DOE, 2002). This dilemma presents a significant waste removal problem for numerous sites for a large variety of reasons, among which two issues have become prominent: *i)* obstructions to tank access; *ii)* waste forms which are varied and complex in constituents (Martin and

Rood, 2011). Chemical treatment of heels produces large quantities of liquid waste. Since these are concentrated high-level wastes, high dose rates are encountered when handling and processing these materials. The methods available require aggressive chemical and mechanical processes which can damage the tank and result in unwanted releases of the tank contents or releases from the components (pumps, pipes, pressure vessels, etc.) used as part of systems for demobilising the tank. R&D on removing or stabilising tank heels has taken place at the DOE's national laboratories and at major universities (US DOE, 2002). There is also an issue with residuals in other components associated with these waste such as pumps and pipes where better decontamination and lockdown techniques are required.

As an example of the challenges this poses, operations are under way at Savannah River in the United States to decommission the underground tanks following removal/ disposition of their bulk HLW. Once the tanks are cleaned they will be filled with grout for permanent closure (Keefer, et al., 2012). The Savannah River site started removing plutonium-uranium extraction (PUREX) plant waste from Tanks 5 and 6 in 1973. Each tank contained 730 000 gallons of waste. In 2005/2006, there were ~34 000 gallons of sludge heel in Tank 5 and ~25 000 gallons of heel in Tank 6. The sludge heels were removed from 2005 through 2008 using mechanical (three mixer pumps) and chemical treatments after which there remained ~3 300 gallons of heel material in Tank 5 and ~3 500 gallons in Tanks 6. First, Tank 5 was cleaned using a feed and bleed method, with the filtrate (feed) being supplied by the well water system. The Tank 6 filtrate was supplied by a recirculation loop between the receipt tank and Tank 6. Results achieved in 2.5 days of feed and bleed in Tank 5 were approximately equal to the results achieved after 10 days of operation in Tank 6 (Vitali, 2010).

There are other applications at operating and decommissioning facilities where improved heel and residual decontamination techniques would be of benefit. Reactor water storage tanks, spent resin tanks, and sump collection and equipment drain tanks often have sludge heels with radiation levels in the mSv/hr to hundreds of mSv/hr range and are challenging to desludge and demolish, especially when high levels of transuranics are present.

- Magnox dissolution technology projects

During the operational lifetime of Magnox reactors, such as at the Bradwell Power Station, spent fuel assemblies were processed on site to remove the cladding and ship the fuel pins to Sellafield for reprocessing at its B205 plant. Magnox (magnesium non-oxidising) fuel assemblies used natural uranium (i.e. unenriched) as fuel and a non-oxidising magnox alloy fuel cladding to cover the uranium metal and contain fission products. Magnox alloy consists mainly of magnesium with small amounts of aluminium and other metals. The fuel assembly design included cooling fins to enhance heat transfer. This alloy must be maintained at a high pH of 11.5 in the ponds and vaults to prevent dissolution in water. The cladding is easily corroded in an aqueous environment and large quantities of corroded sludge have accumulated (primarily at the Sellafield site).

A stripping process was used for the magnox alloy fuel element casing to separate the fuel pins for reprocessing. This generated metallic fuel element debris (FED) which accumulated in the Magnox reactors ponds and vaults. After the fuel itself, FED is the largest radioactive waste stream at the Magnox reactors. The FED was placed in active waste vaults for storage, along with other forms of intermediate-level waste (ILW) such as activated cables and springs that were both loose and in packaging. The various materials in the vaults must be retrieved packaged and processed (Magnox, 2008). Part of the ILW retrieval packaging and storage process is aimed specifically at FED processing and volume reduction through dissolution in nitric acid.

England's Dungeness A was the first site to introduce FED dissolution more than 10 years ago. The waste was dissolved in an acid solvent and then treated to capture the majority of radioactivity. The effluent was then discharged within the site's authorised discharge limits. Assessments by the Nuclear Installations Inspectorate and Environment Agency have demonstrated the process had negligible environmental impact. Following the success of the Dungeness programme, other Magnox sites are adopting this approach.

Summary of current R&D for heels and residues

- Removal of heels and residuals

The DOE has targeted up to 19 billion dollars at Savannah River Site (SRS) and Hanford to develop "transformational tank waste technology". Heel removal is the intermediate phase of the waste retrieval and tank cleaning process at SRS, and is intended to decrease the volume of waste in the tank prior to treatment with oxalic acid. The goal is to reduce the residual amount of radioactive sludge wastes to less than 37 900 litres (10 000 gallons) of wet solids. Reducing the quantity of residual waste solids in the tank prior to acid cleaning reduces the amount of acid required as well as the amount of excess acid that could impact ongoing waste management processes (Keefer, et al., 2012).

A recent cost benefit analysis for removal or treatment of PUREX heel material from Tank 18 at the Savannah River Site evaluated 50 potential alternative technologies (SRR, 2012). These included various grouts with sorbent options, acid or ionic liquid dissolution with chelating agents, ammonium hydroxide leaching, use of surfactants, reverse surface charge sludge deflocing, ultrasonic cleaning with and without acids, vacuuming and sluicing using robotics, etc. For the 46 technologies evaluated and eliminated from further consideration, the most frequent reason given was "technology not developed", meaning that the technologies were unproved and there remained too many uncertainties with field application and performance to risk deployment. Four technologies chosen for further consideration were:

- robotics/modified Sand Mantis – with liquid mobilisation and vacuuming;
- articulating arm – with liquid mobilisation and vacuuming;
- recirculation line (feed and bleed) – mixing and recirculation with pumps;
- acid cleaning with robotic support – direct acid cleaning controlled by robotics.

The robotic modified Sand Mantis was chosen for SRS Tank 18 and 19 heel removal (Huff, 2009). The Sand Mantis is equipped with a low-volume, high-pressure spray nozzle and an eductor for heel removals (SRR, 2012). The DOE demonstrated successful use of a combination of bulk retrieval technologies for tank decontamination using mixer pumps and hydrolances, and mechanical heel removal using the robotic Sand Mantis in tanks with no cooling coils. A functional design criteria for a heel management system was recently issued by the DOE (Campbell, 2012).

A versatile remote manipulator alternative to the underwater remotely operated vehicles (ROV) such as the Sand Mantis was recently developed in England at the Trawsfynydd nuclear power station. S.A. Technology, working in partnership with the Energy*Solutions*-owned Magnox Ltd and ACTUS, designed and built two long-reach manipulators for retrieval of waste from three separate storage tanks. The Rotary Deployment Arm (RDA1 and RDA2) is a highly capable and versatile solution for the site's tank cleaning dilemma. The RDA is designed to complete a variety of clean-up tasks within four main areas on site, including the Resin Vaults (RV2 and RV3), Main Sludge Vault (MSV), and Pond North Void (PNV). These tanks are different sizes and have different technical constraints, making it difficult to design a single solution that could meet each tank's requirements. The RDA is capable of being deployed through penetrations with diameters of 10 inches and larger. The RDA has a vertical reach of 32 feet, and a horizontal reach of 15 feet when fully extended. The RDA consists of mast and forearm assemblies constructed from carbon fibre and stainless steel components, as well as electromechanical and hydraulic components to provide actuation. These mast and forearm assemblies reside inside the tanks during decontamination operations (Padma Nilayav, Biswas and Kumar, 2010).

Figure 4.8: Trawsfynydd RDA and end effectors for tank de-sludging and clean-out

Source: Image courtesy of S.A. Technology.

At the plant room floor level, the RDA is mounted inside a stationary support frame that allows the RDA to be deployed into and retrieved from the various tanks, and provides cable management for the variety of required services. The RDA support frame also provides containment for operation, and wash-down capability for contamination control, as well as wheeled transportation. The frame itself is mounted to each of the plant room floors during operations. In its retrieved state, the RDA fits completely within the support frame, which is about the size of a large refrigerator. The RDA system includes a hydraulic power unit, electrical enclosure and integrated control system to provide motive power and control. End effector tooling for the system is designed to meet a variety of operational requirements for each tank and void (Martin and Rood, 2011).

The DOE is developing "Enhanced Chemical Cleaning" for tanks with cooling coils, using oxalic acid with an oxalate decomposition step to minimise impact on salt waste processing and vitrification. Savannah River and Hanford are sharing lessons learned as both sites develop new tools for tank waste characterisation, retrieval and closure (Olinger, 2011).

The remaining waste in Tank 12 at SRS after heel removal had a high aluminium concentration. Aluminium dissolution by caustic leaching was identified as a treatment step to reduce the volume of remaining solids and prepare the tank for acid cleaning. Dissolution was performed in Tank 12 over a two-month period in July and August 2011. Sample results indicated that 16 440 kg of aluminium oxide (boehmite) had been dissolved, representing 60% of the starting inventory. The evolution resulted in reducing the sludge solids' volume by 22 300 litres (5 900 gallons), preparing the tank for chemical cleaning with oxalic acid (Keefer, et al., 2012).

Similarly highly active liquor (HAL) waste is being stored at Sellafield in the United Kingdom. The HAL is produced at Sellafield from the evaporation of raffinates in fuel reprocessing plants that contain the fission products and waste actinides. HAL is stored in a number of highly active storage tanks (HAST) located in the HAL Evaporation and Storage plant (HALES) at Sellafield. The HAST are high-integrity stainless steel tanks that feature cooling systems in order to manage decay heat. In 1990, the Waste Vitrification Plant (WVP) began converting the HAL into glass to retain the hazardous radioactivity in an immobile form. Vitrification enables long-term passive storage of the waste. Until recently, six new HAST were to be constructed to replace capacity from an ageing 21-tank complex that started storing the site's liquid reprocessing wastes in 1955. The NDA expects to complete reprocessing at Thorp by 2018 and Magnox reprocessing between

2017-2018. Decommissioning of HAL tanks will take place from 2023-2056 (Sellafield Ltd, 2008; 2012d).

- Fuel element debris (FED) dissolution

Dissolution technologies are being developed for the Magnox reactors and Sellafield (NDA, 2010a). Bradwell has led the development of an updated dissolution programme for use at the Magnox reactors. The dissolution programme makes use of the latest technology and incorporates the lessons learned from Dungeness. This has led to dissolution replacing encapsulation of FED as the preferred method in the Bradwell site's Lifetime Plan (Magnox, 2008). The improved Bradwell dissolution FED handling system development was co-ordinated with Dungeness, Sizewell A, the BNLS AWVR project and Sellafield B30 and B41 projects. In addition, lessons learned from the Hunterston experience as pertains to use of Brokk hydraulic arms were incorporated in the design (Magnox, 2008).

The debris to be handled and processed in the dissolution system consists mainly of Magnox splitters and minor quantities of fuel fragments and activated components (Nimonic springs, end fittings and thermocouple assemblies). In addition to these materials, the retrieval and processing scope will include the contaminated gravel layer at the bottom of the vaults and the fuel element debris (FED) corrosion products which are assumed to be present in the gravel (Magnox, 2008; Atkins, 2010).

Subsequent to FED dissolution the remaining un-dissolved packaged waste will be encapsulated/solidified in a combined wet and solid facility, to produce a passively safe waste form. The solidified waste will be placed in an on-site ILW interim storage facility until an off-site deep repository or alternative ILW disposal facility is available (Magnox, 2008). Dissolution of the Magnox FED retrieved from the Bradwell vaults is estimated to save 269 m^3 of packaged container space within the national Geological Disposal Facility (GDF). Lessons learned from the Bradwell development are now helping Sizewell A, Hinkley Point A and Oldbury undertake similar changes to their lifetime plans. The four sites combined have more than 2 000 m^3 of raw FED on site that needs to be managed, meaning that the safe and efficient use of dissolution will reduce waste volumes by 97% (Magnox, 2008).

The Bradwell Dissolution Plant will be sited right beside the reactors and directly over the vaults containing the debris. All the FED handling process are planned to use robotics. First the FED will be retrieved, sorted and then crushed. It will then be stored in box-like "vaults" on site. FED oxidation produces hydrogen that must be monitored constantly and the storage vaults use interlocking systems to ensure that any fire hazard is kept to a minimum. The FED will then be placed in vats and saturated with a mix of water and weak nitric acid for four hours. The resulting liquor, containing the dissolved FED, will then be decontaminated to remove radioactivity and heavy metals using an ion exchange media. Each stage is carefully monitored and, when the radionuclide levels are considered to be at sufficiently low, the decontaminated liquid containing magnesium salts and other metallic species will be discharged into the Blackwater Estuary (Magnox, 2008).

Along with the FED retrieval and dissolution systems, plans for a Mobile Aqueous Effluent Treatment Plant (MAETP) with the Aqueous Discharge Abatement Project (ADAP) related to the FED project are being developed. Factory acceptance testing (FAT) for early FED retrieval (EFR) was completed on 8 September 2011 (Sexton, 2011b). The factory acceptance testing results have been approved by an Independent Nuclear Safety Assessment for the installation of the equipment onsite. Modifications to the vault crane and the installation of a gantry crane, building and equipment have commenced. The setting-out of the EFR building was successfully achieved and installation of the equipment continued through September and October. The early operation of the EFR will provide a buffer stock for the planned FED dissolution plant and reduce the design risks of implementing the entire system through early learning. At this time the completion of factory acceptance testing of equipment for the dissolution plant is behind target. Testing was to be completed in 2012 (NDA, 2012a; Hinkley, 2010).

Future suggested R&D for heels and residues

- ***Description*** – Improve mechanical and chemical methods to deploy equipment, remove heels and decontaminate tank residues to increase safety and efficiency of heel removal from systems and components such as tanks.
- ***Objectives*** – To develop mechanical solutions that integrate with the waste transfer, heels removal, final decontamination and decommissioning of the high-level waste storage systems. To develop a wider array of chemical treatments for heel and residual waste forms that will assist removal and decontamination systems and components while minimising secondary waste issues and volumes.
- ***Desired deliverables*** – Improved robotic or remotely-operated mechanical and chemical decontamination methods to remove heels and residuals.

Optimising the use of robotics

Challenges

As evidenced by previous discussions on developing and applying robotic capabilities to dismantling and decontamination tasks, robots are being used at decommissioning projects in diverse ways. Although robotics have been used in nuclear power for over 30 years (Moore, 1985), their mainstreaming into the performance of D&D tasks lags far behind that of other robotics industrial and service sectors. Member countries identified the high cost of development of robotics technology as an obstacle to obtaining a suite of robotic and/or remote technologies (platforms and tools) for efficient operations in high radiation or contaminated areas.

Key challenges that need to be addressed in order to overcome high costs and more fully develop and integrate robotics into decommissioning projects include:

- Develop a fuller and more broadly held knowledge and appreciation of the robotics capabilities that currently exist and where they have been used successfully on decommissioning projects.
- Stop re-inventing technologies that already exist, then abandoning the equipment when the project is over. If every decommissioning project insisted on designing and fabricating their own excavators, cranes and other equipment from scratch they would also be prohibitively expensive.
- Manage decommissionings to become a consistent reliable patron of the robotics industry and enable the costs of new developments and advances to be spread over multiple decommissioning projects.
- Integrate the newer, more technically adept generation into the current older and vested generation of D&D managers who mistrust the technology and think it is simpler and more cost-effective to throw manpower at a task.
- Fund research further down the R&D pipe line to influence development and testing of robotic capabilities that are applicable to nuclear D&D instead of trying to back-fit them after they have been developed for other applications in other industries and the military.

An analysis of the reasons why Japanese robots did not play a prominent role in the Fukushima nuclear disaster response is instructive regarding the above challenges. The 1999 Tokai-mura criticality accident in Japan resulted in two deaths and prompted the Japanese government to consider whether robotic capabilities should be added to their nuclear emergency response capabilities. The Intervention on Accidents (INTRA) Robotics Group in France has maintained a fleet of robotic response capabilities in the event of a major nuclear accident since 1988 (Groupe INTRA, n.d.). France and Germany are among a handful of countries in the world with a robust, staged robotic emergency response capability on stand-by as part of their nuclear generation resources. The "pilots" or "robotocists" maintain their training, have developed procedures and participate in drills on a regular basis. There are a number of indoor, outdoor and civil engineering robotic devices (e.g. bulldozers, dump trucks, excavators) available for deployment at INTRA.

In January 2000, the then-Ministry of Economy, Trade and Industry allocated JPY 3 billion for the development of "nuclear disaster relief systems". As is characteristic of the nuclear industry, the Japanese attempted to replicate France's robotic emergency response model by developing their own nuclear response robots domestically (Yasuyuki, 2011). The Manufacturing Science and Technology Center received a budget and requested four proposals from Mitsubishi, Hitachi, Toshiba and the French firm Cybernetics. Work was started in June 2000 to develop the different types of robots, six units in total. The budget was limited to one fiscal year, so the development work was carried out hastily, with prototyping completed in about seven months. The demonstration runs took place barely within the set time of one fiscal year, occurring on 22-23 March 2001. TEPCO, the Electric Power Industry Central Research Institute, and the Japan Nuclear Cycle Development Institute did not find the robots suitable for immediate deployment. They raised a number of reasons for their conclusion, including the low reliability of remote control and the excessively large size of machines that were meant to move around in the small spaces of reactor buildings. It is likely that an underlying anti-robot opinion evident in the statement "it is quicker and easier to send personnel there while ensuring their safety" also influenced the decision to abandon further development of emergency response robotic capabilities (Yasuyuki, 2011). The project was abandoned and the robots became curiosity relics at museums and universities.

The first mistake the Japanese made was to try "develop" what France and Germany had already deployed instead of just buying it off the shelf and refining it. The second mistake was to believe that throwing manpower at a nuclear accident would be cheaper and easier than using robotics.

The last three challenges are illustrated by an evaluation of the reasons why the United States robotics industry was able to quickly deploy robots to assist with the Fukushima recovery. The United States has had a massive incentive since the 9/11 attack to develop robotics technology for defence and homeland security, and a massive laboratory in the form of two wars (Afghanistan and Iraq) in which to develop them. PackBots and a Small Unmanned Ground Vehicle (SUGV) were first put into operation to enter caves harbouring enemies or to dispose of roadside improvised explosive devices. Although a PackBot costs USD 120 000 per unit, and an SUGV costs USD 130 000-200 000, more than 3 500 units in total were sold to the military (Yasuyuki, 2011). The United States military has embraced the use of robotics and provided a steady revenue to the robotics industry as opposed to the nuclear D&D industry, which has had a fits-and-starts, feast-or-famine pattern since the 1980s.

The article we have cited (Yasuyuki, 2011) sums it up rather well with the following. Mr. Sanji points out that, "Japan has insufficient patrons to fund the development of disaster relief robots, and there are no operators to maintain and train the robots." Power companies in Europe and the military in the United States clearly play the role of patrons, and they are tightly coupled with the nuclear power operators by capital and trading relationships. In Japan, however, the government spends some development budget on occasions like the JCO accident, but these expenditures and developments never take root as a permanent disaster prevention system. The bias against robotics and the intermittent funding of their development for nuclear applications is clearly evident in many countries other than Japan.

The problem with continuing on that path illustrates the final point in the list of challenges. If there had been no 9/11 and no subsequent wars, would those robotic technologies have been available to deal with the multi-reactor emergency that unfolded in Japan? In the face of that event, the nuclear industry depended on robotics developed for homeland security and military applications to meet emergency response and D&D challenges. In 2006 a review of "Resources for Nuclear and Radiation Disaster Response" discussed the extensive array of government and military assets readying to respond to nuclear and radiation disasters and asked if we can ever be fully prepared. This assessment did not include any mention of robotics or remotely operated equipment (Maiello and Groves, 2006).

In contrast Dr. Robin Murphy of the Center for Robot-Assisted Search and Rescue (CRASAR) in Texas (CRASAR, 2014) describes how she and Dr. Satoshi Tadokoro of Tohoku University began collaborating in 1998 and how he started the International Rescue System Institute (IRS) in Japan in 2002 shortly after CRASAR was created in Texas. With international funding they were focused on developing search and rescue robotics for search and rescue after earthquakes, floods, tornados and other disasters. The Quince robot was under development for search and rescue missions at IRS. They had just finished a productive week of working at Disaster City with the IRS successfully testing Quince's improved mobility in different rubble piles and tunnels. This testing benefitted the deployment of Quince at Fukushima when the tsunami struck (Murphy, 2012). This instance in which robotics developed in Japan were successfully deployed was, once again, technology that was developed outside the nuclear power industry and was then back-fitted and adapted for use at Fukushima.

Clearly we should learn the lessons of Fukushima and a consolidated effort should be made to embrace and develop robotic technologies for decommissioning, emergency response and more broadly in the nuclear industry (Resende, 2012). It is the surest path to success in the global effort to decommission nuclear facilities in a cost effective and timely manner.

Figure 4.9: Quince being tested at disaster city rubble pile the week before the Fukushima Daiichi NPP accident

Summary of current R&D for optimising the use of robotics

- Three-dimensional integrated gamma-ray and vision systems

The Nuclear Decommissioning Authority and Magnox have sufficient decommissioning work ahead of them to ensure that robotic applications can have a payback period that spans many projects and they are clearly making an exceptional effort to develop such decommissioning technologies. This coincides with a national push to invest in cutting-edge robotics technology, such as autonomous and semi-autonomous robots whose capabilities align with the NDA's objectives for remote monitoring of stores and facilities in care and maintenance (EPSRC, 2012). With a little imagination, autonomous capabilities could be adapted for performance of mundane activities such as monitoring ILW stores and site environs, dust suppression during demolition, scanning and scabbling, handling and transport of debris, or remediating walls and floors surfaces. The list of decommissioning robotic developments spawned by this effort in the United Kingdom is impressive. As noted throughout the report, robotic applications are being evaluated by the NDA throughout the estate for laser cutting and concrete scabbling, for vault, tanks and ponds clean-up, and for component removal at Trawsfynydd, Bradwell, Dounray, Chapel Cross, Hinkley Point, Sellafield, etc.

Another example of innovative development of robotic capabilities is the use of the NVisage™ gamma camera and FARO LS 880 Laser Scanner modelling system at the United Kingdom Sellafield Separation Plant. The two systems were simultaneously deployed at different vantage points, using a robotic arm to create a precise 3-D model of the shear cell overlaid with radiation levels. The shear cell contains the majority of in-cell equipment from operations; the internals associated with the shear pack have been removed and placed in adjacent facilities. Bulky items (e.g. the dissolver basket, basket tipper, feed envelope, vessels and the shear support and feed column) all remain. The cell is lined with a stainless steel liner that was painted; however, this coating is peeling off the walls, creating debris in the bottom of the cell. Background dose rates above the shield door are 11.58 mSv/hr (γ) and 18.96 mSv/hr ($\beta\gamma$). To progress the characterisation of the shear cell, the project engaged REACT Engineering Ltd and Multipass 3-D Laser Scans Ltd to carry out laser scanning using a FARO LS 880 laser scanner that precisely mapped the contents of the shear cell. The data obtained would also be used to create 3-D model images of the shear cell integrated with the data from the gamma scanner (MacGregor, Slater and Mort, 2010). This is an example of R&D work that can be used to remotely survey and construct 3-D files for use in decommissioning planning and geostatistical evaluations of the data. Adaptation of this technology to include alpha-camera-type imaging of UV emissions from ionised air would be of great benefit for characterising and remediating hot cells and other areas with extremely high transuranic contamination hazards.

A series of 200 photographs were used to create 3-D models of a robotic manipulator and cell systems and structures in France. The 3-D model was used to develop a virtual reality mock-up that was used to practice and plan for the use of a robotic arm and end effectors to demolish the cell components (Chabal, et al., 2011). Similarly, a powered remote manipulator arm (PRM) was developed at Sellafield to isolate and remove redundant pipework in a high radiation area, and to clean and seal a contaminated pond wall. Significant preparation work had to be carried out before work could start on the job. This included the construction of a full-scale mock-up of the facility in Whitehaven to prove the equipment, method and safety of the procedure, along with providing a low-risk environment in which to train the operators. A project manager stated:

> *The full operation was practiced again and again in the test facility for 80 000 operating hours. Through the intense practice, we were able to satisfy ourselves and our regulators that the job could be flawlessly executed and that every eventuality has been considered and prepared for. At all times safety was the over-riding priority and although the job was high risk, it was a job that was long overdue.* (Nuclear Street, 2012a)

While these training and mock-up innovations undoubtedly led to a much more flawless execution of the work, they also certainly considerably drove up the costs for using robotics. This has echoes of the lessons learned from the Japanese experience discussed in the challenges section where emergency response robots were shelved based on a less than flawless performance. The nuclear industry needs to stop redesigning manipulator arms and come to grips with its inherent prejudices against using robots and adopt more realistic risk assessments in order to control costs. While this effort is to be applauded, we should ask ourselves, if workers had been used for these jobs instead of robots, would a similar level of effort with mock-up simulations, and forty man-years of mock-up rehearsals have been expended to "satisfy ourselves and our regulators that the job could be flawlessly executed"? After all, a robot cannot become ill, injure itself or be overexposed to radiation or hazardous substances.

A new approach for autonomous robot navigation uses a cheap, Microsoft Kinect sensor to map the entire 3-D space a robot would navigate and to store and use the information to identify changes in the memorised 3-D image. The navigation system is based on a technique called simultaneous localization and mapping (SLAM). It is being developed at the Massachusetts Institute of Technology (MIT) in the United States and will allow robots to constantly update a 3-D map as they learn new information over time. The MIT team previously tested the approach on robots equipped with expensive laser scanners, but a paper presented in 2012 at the International Conference on Robotics and Automation indicates they have now shown how a robot can locate itself within such a map with just a low-cost Kinect-like camera (R&D, 2011b).

The team tested the system on a robotic wheelchair, a PR2 robot developed by Willow Garage in California, and in a portable sensor suite worn by a human volunteer. They found it could locate itself within a 3-D map of its surroundings while traveling at up to 1.5 m/sec. This opens up exciting new possibilities in robot research and engineering for robots that fly or navigate in environments with stairs, ramps and all sorts of other indoor architectural elements (R&D, 2011b). Thus it appears that 3-D modelling of a room or space may be as simple as driving a robot around and then downloading the image map from its memory (Knight, 2012; R&D, 2011c).

- Develop flexible robots with the possibility to mount different tools

There are any number of robotics manufacturers with a wide variety of end effectors that can easily be changed out to perform a variety of decommissioning tasks; Brokk, K.T. Grant, Husqvarna Construction, Stanley LaBounty, etc., have all developed equipment capable of using a wide range of end effectors. S.A. Robotics/Technology, iRobot PackBots and the QinetiQ Talon were outfitted with chemical, biological, radiological, nuclear and explosive (CBRNE) detection kits and cameras to perform inspections and monitor conditions inside the Fukushima facility. Underwater crawlers such as the Scarab and Sand Mantis have been used for many years for underwater inspections, cleaning and retrieval applications. PAR, IWT and OCrobotics have developed and tested state-of-the-art end effectors for laser cutting, scabbling and paint stripping. The marine shipping industry has used magnetic wheeled crawlers with hydrolasers and capture hoods to remove hazardous paints for nearly a decade.

Remotely operated construction vehicles such as bobcats, dump trucks, bulldozers and full-size excavators are now available or easily and quickly outfitted to be operated robotically, as evidenced by the initial clean-up at Fukushima. Petrol fuel units can be used in environments where exhaust fumes and carbon monoxide are not a concern. The Japanese firm Yoshikawa Co. provided large remotely controlled excavators and trucks to Fukushima (Farr, 2011a). QinetiQ North America offers Robotic Appliqué Kits that convert Bobcat loaders into unmanned vehicles in 15 minutes. The kits permit remote operation of all 70 Bobcat vehicle attachments, such as shovels, buckets, grapples, tree cutters and tools to break through walls and doors (Farr, 2011a).

The French industrial consortium Groupe INTRA has a series of robots for emergency response that can operate from a considerable distance while providing real time response over many kilometres. Devices maintained by INTRA range from indoor and outdoor exploratory robots to giant earth removal trucks, bulldozers and excavators that can withstand harsh environments (Groupe INTRA, n.d.). Until recently, many systems in these robots have been proprietary, as standard equipment has not yet met system performance requirements. This has made it difficult to evolve the systems without changing the software. However, commercial boards and software are becoming viable for use in the most challenging designs. INTRA, with the help of the CEA, is replacing 15-year-old systems with Pentium 4 based boards, linked to specialist interface and communications boards while moving to a new software architecture. This combination is allowing robot systems to be updated quickly and efficiently without introducing software flaws and problems. This will also ensure the continued development and broader applications for these machines as autonomous and semi-autonomous software capabilities are developed (Wiegand, 2010).

There are almost always pre-existing machines that can be modified slightly or used as-is in lieu of developing expensive new site-specific technologies (Farr, 2012). A review of the current robotics capabilities and lessons learned for the nuclear industry has not been conducted for nearly a decade (EPRI, 2004a). Tokyo Electric Power Company is trying to come to grips with the inferential and anecdotal understanding of robotic capabilities that pervades the industry in order to expedite the application of robotics to the pressing issues facing the severely damaged decommissioning of the Fukushima units. At their International Symposium the company demonstrated (METI, 2012b) a systematic plan for characterising the contamination in the reactor buildings and testing decontamination methods and remote technologies (Sakai, 2012; Garrec, 2012). TEPCO is developing a "technical catalogue" for R&D technologies based on the plant manufacturers' database for Japanese nuclear facilities. They are also seeking to include other applicable remote decontamination technologies and D&D services in the catalogue through voluntary submission of applications (METI, 2012a). The catalogue should provide a comprehensive snapshot of the decontamination and robotics capabilities available in the industry.

While the nuclear industry is trying to determine what robotic capabilities are available and redesigning existing equipment, many new developments in robotic capabilities are being designed and tested for other industries and applications. Robot sales grew by 68% in 2011 (Moore, 2012) and this growth is accelerating robotics R&D, dramatically expanding the nature and capabilities of robots. The use of robotics is becoming more common on D&D projects as the global number of decommissioning projects accelerates. Mobile autonomous and semi-autonomous capabilities similar to those that allow military drones to fly missions with little or no human intervention are leading to the development of mobile robots that can perform routine tasks and assist humans (Brumson, 2012). This includes humanoid or near humanoid designs to assist in geriatric care, nursing and surgical procedures. The United States Navy is developing an autonomous humanoid robot to assist in firefighting aboard vessels. Its robot has voice recognition software, can take commands and follows the attention of the crew chief. The Navy plans to test the robot in a realistic firefighting environment on board the ex-USS Shadwell in late September 2013 (R&D, 2012c). Honda has an autonomous robot that can open and pour from containers, among other tasks (R&D, 2011a). Other developments include tiny spheres to monitor pipe internals, designed with the nuclear industry in mind (Chu, 2011). Tiny flexible robots are also being developed to perform surveillances in tight spaces (Chang, 2011), and silicone-based autonomous jellyfish robots that use water to generate energy, and synthetic muscles to propel themselves, have been developed by the United States Navy (R&D, 2012d). The robotic jellyfish prototypes are also being evaluated for environmental monitoring and spill remediation (R&D, 2012e).

The nuclear industry has used magnetic-wheeled robotic crawlers for reactor vessel weld inspections for some time. Shipyards have been using magnetic-wheeled autonomous robots to strip hazardous paint from vessels in dry-dock, with hydrolasers and vacuum capture hoods, for nearly a decade (Dasgupta, 2011). New wall-crawling robots have been developed for non-magnetic surfaces such as stainless steel or concrete (R&D, 2011d; Sellafield Ltd, 2012b). Other R&D areas are developing the capability to design and print out functioning robots from a laptop using 3-D laser printing (R&D, 2012a). This capability is rapidly evolving with expansion and evolution of "Fab-Labs" (Gershenfeld, 2012). Another development allowed handicapped personnel to control robots using brain waves (Jordans, 2012). General Motors and NASA collaborated to develop robotic gloves for auto workers (R&D, 2012b) and robotic systems with a human-like sense of touch are being developed (Heng, 2011). The capabilities and diversity of applications to which robotics can be applied is already substantial and growing rapidly as software, materials and design become more sophisticated.

Future suggested R&D for optimising the use of robotics

- ***Description*** – Develop, test and deploy off-the-shelf, re-usable, multi-functional and highly adaptable robotics for decommissioning of reactors and facilities of various sizes/designs.
- ***Objectives*** – To develop more cost effective and versatile robotics applications for decontamination and to integrate automated equipment and processes into the execution of decommissioning in ways that reduce replication of existing capabilities and use emerging robotics capabilities to facilitate D&D.
- ***Desired deliverables*** – Robotic platforms and tooling that expand current decontamination capabilities, minimise secondary waste generation and facilitate and integrate with the decommissioning process.

Bulk soil remediation and bio-remediation

Challenges

According to the DOE there are 79 million cubic metres of contaminated solid environmental media associated with the nuclear weapons complexes, of which 70% is contaminated with radionuclides. In addition, there are about 1 800 million cubic metres of contaminated soil, of which 57% is contaminated with radionuclides (US DOE, 1997; French, 2012).

Development of soil remediation technologies has been a priority since the Chernobyl disaster in 1986 and has gained even more urgency in the wake of the Fukushima accident in Japan. The working party identified the following challenges for bulk soil remediation:

- treating large volumes of soils;
- storage of large volumes of contaminated soils;
- reusing contaminated soils.

Following the nuclear disaster at Chernobyl, the Soviet government chose long-term evacuation over extensive decontamination; as a result, the plants and animals near Chernobyl inhabit an environment that is both largely devoid of humans and severely contaminated by radioactive fallout. The meltdown of the three reactors at Fukushima also contaminated large areas of farmland and forests. This has provided a new impetus for research on remediation of soils. The Japanese have established eight locations for study of soil decontamination (OECD/NEA, 2012b), with 25 proposals funded for improved/ innovative decontamination technologies. These include topsoil removal, turf stripping, and the fixation and excavation of soils. Three large construction firms have been awarded contracts from the Japan Atomic Energy Agency to test the effectiveness and efficiency of various decontamination technologies at 19 model sites throughout the Fukushima Prefecture. The results of these experiments will guide the large-scale decontamination effort. Japan's decontamination efforts are focused mostly on radionuclides ^{134}Cs and ^{137}Cs. These nuclides have been found in all of Japan's prefectures but are most highly concentrated within an oblong swath that extends about 50 kilometres northwest of the plant, and to a lesser extent throughout the eastern and central Fukushima Prefecture (Bird, 2012; Yoshida and Kanda, 2012). Currently test methods involve removal of the top 5-10 cm of top soil. The Japanese government has estimated the clean-up cost to be about USD 14 billion, and to generate at least 100 million cubic metres of soil, enough to fill 80 domed baseball stadiums – and the effort is estimated to take decades.

The renewed focus on soil decontamination methods is bound to lead to insights and developments applicable to decommissioning facilities with soils requiring remediation. Many of these evaluations and R&D initiatives focus on response to a nuclear accident, radioactive dispersal device (RDD) or dirty bomb, but are they are also applicable to containing, controlling and remediating contaminants in the environment.

Summary of current R&D for bulk soil remediation (including bio remediation)

- Phytoremediation

Phytoremediation is an emerging technology in which plants are used to absorb elements from a polluted environment and to bio-magnify (e.g. concentrate) them by metabolising them into various biomolecules in their tissues. Phytoremediation refers collectively to all plant-based technologies, but primarily uses green plants to remediate contaminated sites. It can be classified according to the main mechanism involved in the process (US EPA, 2001):

- rhizofiltration – technique involving plant roots in the uptake of contaminants;
- phytoextraction – technique involving the total body of the plant in the uptake of contaminants from soil;
- phytotransformation – applicable to both soil and water and involving the degradation of contaminants through plant metabolism;
- phytostimulation or plant-assisted bio-remediation – also used for both soil and water and involves the stimulation of microbial biodegradation through the activities of plants in the root zone (rhizosphere);
- phytostabilisation – technique that reduces the mobility and migration potential of contaminants in soil.

Phytoremediation is an attractive alternative to many of the currently practiced *in situ* and *ex situ* technologies for a variety of reasons, including low capital and maintenance costs, non-invasiveness, easy start-up, high public acceptance and the pleasant landscape that emerges as a final product (Pathak, et al., 2012; Whicker, et al., 2004). Phytoremediation has been evaluated and tested extensively as a method of remediating contaminated soils after the Chernobyl (Klubicová, et al., 2011) disaster and with renewed vigour following the Fukushima disaster (Entry, et al., 1997; Smith, n.d.; IAEA, 2011d; Yasutaka, 2012; Nakayama, 2012; Suzuki and Saito, 2012; Hirayama, 2012). The limited potential of this method and disappointing test results using sunflower seeds and various other potential plant species led the Japanese government to remove the top 5 cm of topsoil as the preferred remediation method. There are several issues that limit the potential for phytoremediation to become a viable decommissioning soil remediation methodology, the most obvious of which is root depth; a plant cannot absorb contaminants that are out of reach of its root system. Thus, phytoremediation is not viable for contamination deeper than a metre and at maximum two metres. The second issue is that most nuclides of concern are positively charged and are thus tightly bound in soils and especially in soils with high organic and clay components, as evidenced by the soil water partitioning coefficients (Kds) (Kozai, et al., 2012). A plant cannot absorb a radionuclide through its roots unless it is available in the interstitial water of the soil. Reacting soils to make them more acidic has been shown to make more contaminants available in the soil pore water and increase concentrations in plants, but it also lowers the productivity and yield (Vandenhove, Van Hees and Van Winckel, 2001). In addition, a given species of plant may exhibit selective uptake of one element or another (Co, Cs, U) but it is unlikely any that single species will effectively sequester all the nuclides in a contaminant waste stream or fingerprint (Vandenhove, Van Hees and Van Winckel, 2001; Malik, et al., 2000; Ramaswamia, Carr and Burkhardt, 2001). Finally, the very act of tilling the soil in order to plant a phytoremediation crop has the potential to greatly increase the volume of contaminated soil by distributing contamination isolated to a relatively thin top layer throughout the tilled depth (Fujiwaraa, et al., 2012). Given all these constraints, transgenic (genetically engineered) plants specially designed for phytoremediation likely hold the greatest promise for making this a viable soil remediation method (Eapen and D'Souza, 2005; Ruiz and Daniell, 2009).

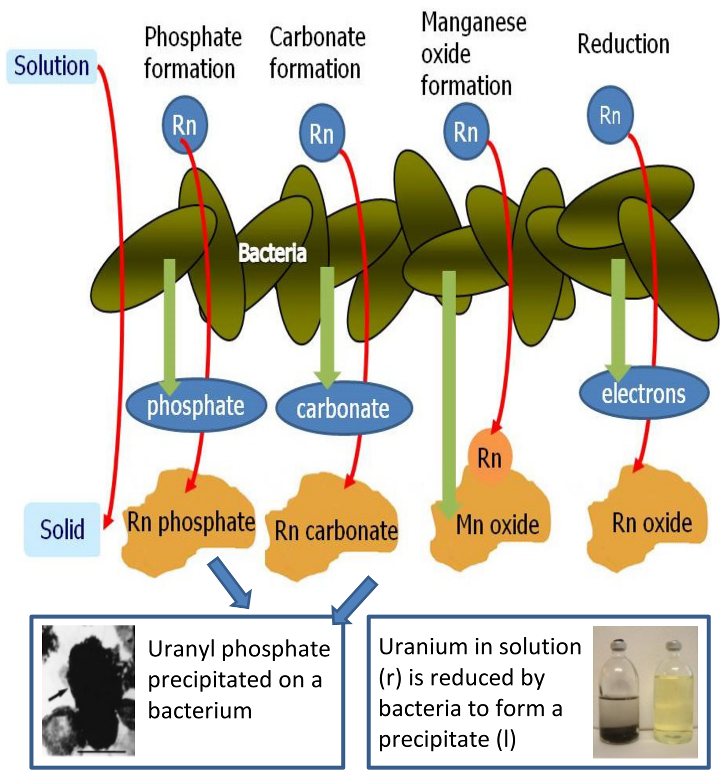
- Biogeochemical application in nuclear decommissioning and waste disposal

Ongoing research explores the use of microbial technologies to decrease the risk of contamination resulting from the decommissioning of nuclear sites and the construction of repositories for nuclear waste (Paterson-Beedle, et al., 2012). The objective is to reduce the potential for migration of radionuclides in soils and rocks using special properties of the bacteria that are present in the local environment and micro-environments. The project will investigate two different bacterial properties (Mackay, et al., 2009):

- how micro-organisms can be used to trap radionuclides within the soil/rock through the mineral depositions they form and consequently prevent their transport to the human environment (Figure 4.10);
- how some bacteria can be encouraged to produce minerals (e.g. calcite) in soils and rocks that will block or clog any pathways for fluid flow (Figure 4.11).

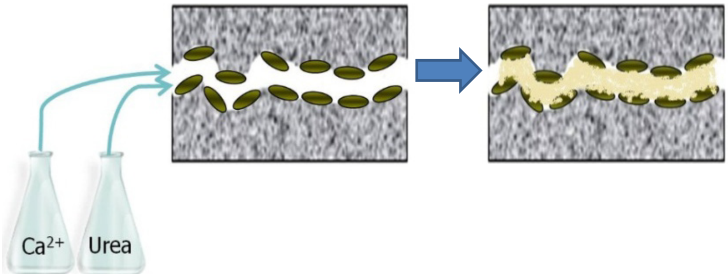
Biogeochemical studies include soils and rocks expected at decommissioning sites and repositories to gain a better understanding of variations in microbiological properties. These projects include extensive laboratory research (under controlled conditions) and investigations in the field. The processes of mineral deposition and radionuclide capture will be imaged over time and space in three dimensions using complex technologies such as magnetic resonance techniques. Mineral deposition includes using bacteria to clog fractures in rocks through the production and deposition of calcite. Radionuclide capture includes investigating the uptake and capture of radionuclides by bacterially-generated hydroxyapatite (bio-HA) mineral formations produced by the bacteria (Mackay, et al., 2009). Computer models will be developed to simulate the basic biological and chemical processes taking place. The main findings of the project will directly benefit the nuclear industry and the public, reducing risks from radionuclide migration and contributing to economical clean-up strategies (Mackay, et al., 2009).

Figure 4.10: Illustration of microbial capture from Birmingham University



Source: Mackay, et al. (2009).

Figure 4.11: Illustration of microbial calcite clogging from Birmingham University



Source: Mackay, et al. (2009).

- Soil washing and thermal/chemical/biological treatment of soils

This is a very broad area of soil treatment that includes both *in situ* (EUGRIS, n.d.b) and *ex situ* (EURGIS, n.d.a) treatment methods. *In situ* treatment technologies are chemical, physical, biological, thermal or electrical processes that remove, degrade, chemically modify, stabilise or encapsulate contaminants within soil or groundwater (matrices) without removing those matrices from the ground. *In situ* treatments have several advantages over *ex situ* treatments; for example, they generally involve less physical disruption of a site. The EURGIS portal for soil and water management in Europe provides a good overview of *in situ* treatment technologies (EUGRIS, n.d.b). Biological treatments were discussed previously in the two previous subsections on phytoremediation and biogeochemical treatments.

Physical/chemical treatments

Physical/chemical treatments use the physical and/or chemical properties of the contaminants or of the contaminated medium to destroy (i.e. chemically convert), separate or contain the contamination. In the physical processes the phase transfer of pollutants is induced. In the chemical processes the chemical structure (and then the behaviour) of the pollutants is changed by means of chemical reactions to produce less toxic or more easily separated compounds from the solid matrix (EUGRIS, n.d.b).

These treatments are typically cost effective and can be completed in short time periods (in comparison with biological treatment). Equipment is readily available and is generally not engineering or energy-intensive (EUGRIS, n.d.b). Certain *ex situ* physical/chemical treatment technologies are sensitive to certain soil parameters. For example, the presence of clay or organic materials in soil causes variations in horizontal and vertical hydraulic parameters that, in turn, cause variations in physical/chemical process performance (EUGRIS, n.d.a).

Soil flushing

In situ soil flushing is the extraction of contaminants from the soil with water or other aqueous solutions. It is accomplished by passing the extraction fluid through in-place soils using either infiltration or injection. Extraction fluids must generally be recovered from the underlying aquifer. The process is most applicable to inorganic contaminants, including radionuclides, but may be applied to organic contamination (Gombert, 1994). The additives for flushing could remain in low amounts in the soil and need to be monitored. Soil flushing requires the solution to be contained and recaptured. Low permeability or heterogeneous soils are difficult to treat. Above-ground separation and treatment costs for recovered fluids can drive the economics of the process (EUGRIS, n.d.b). Surfactants may be added to the extraction fluid to increase the solubility of organic compounds and of non-aqueous phase liquids. This technology is readily available through vendors (IAEA, 1999e). Japan's laboratory tests on soil flushing in two sandy soils suggest that about 70-90% of the radioactive caesium is removable (Yasutaka, 2012).

Thermal treatments

Thermal treatments offer quick clean-up times but are typically the most costly treatment group. This difference, however, is higher with *in situ* than with *ex situ* applications. Cost drivers are energy and equipment, and they are both capital- and O&M-intensive (Ohsugi, et al., 2012).

Thermal processes use heat to burn, melt, decompose or destroy the contaminants and increase their volatility. They are applicable for volatile radiological contaminants such as tritium and carbon-14 (EUGRIS, n.d.b). The Japanese are evaluating thermal treatment processes to volatilise ^{137}Cs from soil. Tests found that, by adding unspecified high-performance accelerants, increased *ex situ* volatilisation of caesium could be achieved with thermal treatment of the soil (Miura, et al., 2012). Another thermal treatment investigated in Japan was the use of pyrolysis to remove radiocaesium from organic materials (Ohsugi, et al., 2012).

A type of *ex situ* soil washing treatment for remediation of contaminated lands in Japan was presented in February 2012. Studies of the distribution of ^{137}Cs in soil samples showed that the majority of the ^{137}Cs (e.g. 60-80%) is compartmentalised in the fines or silt fraction. This is similar to the concentration of radionuclides in the paste as opposed to the aggregate in concrete. Removal of fines less than 0.05 mm by hydroseparation would provide a decontamination factor of 3. Dissolution and separation of fines is assisted by ultrasound in separation of the soil suspension in water. The chemical leaching step involves the application of strong stable element cations, such as Fe^{3+}, K^+, Cs^+ and Mn^+, to disassociate the radioactive Cs and Sr cations from the soil particles. This is similar to the practice of regenerating cation or anion resin beds by overwhelming the active sites with ionic species, causing the dissociation of the contaminant from the resin. Kinetics show an optimal desorption of 15-30% over the course of 16 days, with results varying widely based on molarity of the cationic solutions applied and the soil types treated. As was discussed with regard to the partitioning of contaminants in pore water versus on soil particles, the efficacy of this method is directly related to the negatively charged clay and organic constituent compositions of the soil. Thus, this method is the most effective on sands with less negatively charged material and fine clay-based particles. The degree of ^{137}Cs extraction varies from 2-40% by Fe(III) $> Cs^+ > NH_4^+ \approx K^+ \approx Na^+$. Higher temperatures increase chemical extraction by up to 50%. The combined hydroextraction and chemical leaching process can achieve decontamination factors of 4 to 5 (Toropova and Davydov, 2012). Evaluation of soil washing effectiveness using strong acids is also being performed in Japan (Yasutaka, 2012).

Another approach applicable to *ex situ* soil remediation uses colloid-stable selective sorbents to uptake ^{137}Cs. The new approach uses immobilisation of transition metals' ferrocyanides (cobalt, nickel and copper) in nano-sized carboxylic latex emulsions. The effects of ferrocyanide composition, pH and media salinity on the sorption properties of the colloid stable sorbents toward caesium ions were studied in solutions containing up to 200 g/L of sodium nitrate or potassium chloride. The sorption capacities of the colloid sorbents based on mixed potassium/transition metals' ferrocyanides were in the range of 1.3-1.5 mol Cs/mol ferrocyanide, with the highest value found for the copper ferrocyanide. It was shown that the obtained colloid-stable sorbents were capable of penetrating bulk materials without filtration, making them applicable for decontamination of solids (e.g. soils, zeolites, spent ion-exchange resins contaminated with caesium radionuclides). After decontamination of liquid or solid radioactive wastes the colloid-stable sorbents can be separated easily from solutions by precipitation with cationic flocculants, providing a concentration of radionuclides in the small volume of precipitates formed (Avramenko, et al., 2011). Similarly, electrokinetic separation is being tested in some systems (Kim, et al., 2010).

Another current extraction method involves the use of super critical fluid extraction (SCFE). Research has been conducted on the ability of SCFE as a method of remediating soil classified as mixed transuranic (TRU) waste. The range of contamination levels and contaminants that can be treated with SCFE varies widely. Thus, this methodology can be used for treating soils contaminated with transuranics and hazardous organic materials that could qualify as mixed TRU waste (Castelo-Grande and Barbosa, 2003; Shadrin, et al., 2008; Fox, et al., 2000).

Nanotechnology

Nanotechnology is still relatively in its infancy, but is rapidly evolving. It holds promise in remediating sites cost effectively and addressing challenging site conditions (US EPA, 2008b). Ongoing research at the bench and pilot scale is investigating particles such as self-assembled monolayers on mesoporous supports (SAMMS™), dendrimers, carbon nanotubes and metalloporphyrinogens to determine how to apply their unique chemical and physical properties for full-scale remediation (US EPA, 2008b, 2011c, 2011d, 2012).

Nanoscale materials can be grouped into three categories: natural, incidental and engineered. Examples of naturally occurring nanoscale materials include clays, organic matter and iron oxides within soil that play an important role in biogeochemical processes. Incidental nanoscale materials enter the environment through atmospheric emissions, solid or liquid waste streams from nanoscale material production facilities, agricultural operations, fuel combustion and weathering. Engineered or manufactured nanoscale materials are designed with specific properties in mind and may be released into the environment through industrial or environmental applications. Nanoscale materials may be produced via a "top-down" approach, such as by milling or grinding macroscale materials or, most commonly, via a "bottom-up" approach, such as borohydride reduction, which creates nanoscale materials from component atoms or molecules.

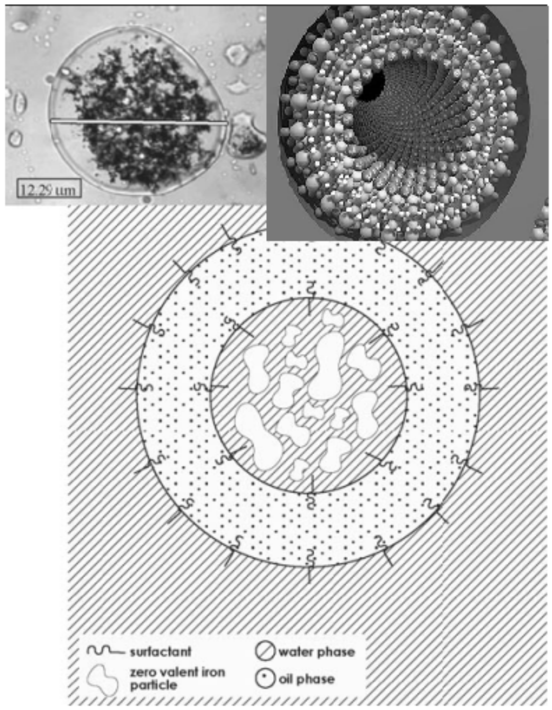
Nanoparticles can be highly reactive due to their large surface area to volume ratio and the presence of a greater number of reactive sites. This allows for increased contact with contaminants, thereby resulting in rapid reduction of contaminant concentrations. Due to their minute size, nanoparticles may pervade very small spaces in the subsurface and remain suspended in groundwater, which would allow the particles to travel farther than macro-sized particles and achieve wider distribution (US EPA, 2008b).

An increasing variety of nanomaterials with environmental applications have been developed over the past several years. Research indicates that nanoparticles such as nZVI, bi-metallic nanoscale particles (BNP) and emulsified zero-valent iron (EZVI) may chemically reduce the following contaminants effectively: perchloroethylene (PCE), TCE, cis-1, 2-dichloroethylene (c-DCE), vinyl chloride (VC), and 1-1-1-tetrachloroethane (TCA), along with polychlorinated biphenyls (PCB), halogenated aromatics, nitroaromatics and metals such as arsenic or chromium (US EPA, 2008b).

Some materials can be made with surface functional groups to serve as adsorbents to scavenge specific contaminants from waste streams. SAMMS™ particles consist of a nanoporous ceramic substrate coated with a monolayer of functional groups tailored to preferentially bind the target contaminant. The functional molecules covalently bond to the silica surface, leaving the other end group available to bind to a variety of contaminants. According to researchers, SAMMS™ particles maintain good chemical and thermal stability and can be readily reused or restored (US EPA, 2008b).

Figure 4.12 shows a schematic of a functionalised nano-sized pore within a SAMMS™ particle; particle has a large surface area to allow for quick sorption kinetics. Contaminants successfully sorbed to SAMMS™ particles include radionuclides, mercury, chromate, arsenate, pertechnetate and selenite. The EPA reports on the SAMMS™ Adsorbents website (SAMMS Absorbents, 2014) that the sorbent has shown positive results in pilot scale tests in the remediation of mercury in well water with a high concentration of dissolved solids, aqueous mercury in low concentrations, highly radioactive mercuric waste and gaseous elemental mercury (US EPA, 2008b).

Figure 4.12: Functional nano-sized pore within a SAMMS™



Source: US EPA (2008b).

Future suggested R&D for bulk soil remediation (including bio-remediation)

- ***Description*** – Develop *in situ* and *ex situ* methods for bulk soil remediation. This includes phytoremediation, bacteriological and other biological *in situ* and *ex situ* approaches, as well as physical/chemical treatments and extraction methodologies such as nanotechnology developments.
- ***Objectives*** – Develop new effective methodologies that allow extraction and concentration of contaminants from bulk soils to enable minimisation of waste generated and reclamation of soils for environmental restoration.
- ***Desired deliverables*** – New experimental and field tested *in situ* and *ex situ* bulk soil remediation methodologies.

Fixing contamination in soil and the use of engineered barriers

Challenges

Given the current state of bulk soil remediation, excavation and removal, use of engineered barriers is the most often used and frequently viable option for bulk soil and groundwater remediation. The large volumes of waste and the environmental impact of large-scale excavations and treatments makes other forms of interventions to contain and control contaminants less desirable until more viable remediation technologies are developed or until a facility's final license termination. This may entail controlling soil contaminants in place for prolonged periods at facilities where care and maintenance or SAFSTOR is an intermediate decommissioning phase. The following challenges have been associated with this issue:

- high cost of controlling the spread of soil contamination to groundwater;
- immobilisation as a temporary solution.

Preventing the spread of soil contamination to groundwater or through groundwater can require installation of physical or engineered barriers and removal and treatment of water from the isolated contaminated land. The on-site response to the Fukushima disaster entailed industrial scale applications of polymer fixatives to site buildings and soil to minimise the dispersal of contaminants. Intervention or containment measures are applied when extensive subsurface contamination precludes treatment or excavation of the material such as in fractured bedrock. In general, containment technologies are applicable to all forms and types of contaminants and consist primarily of surface caps, cut-off walls and bottom barriers. Clay/oil caps and impervious membranes are used to cap contaminants and prevent migration. Surface sealants/stabilisers are used to stabilise or cover waste deposits to control erosion, prevent surface water infiltration, provide dust and vapour control and contain contaminated wastes. Bituminous or sulphur membranes can be sprayed on to the site surface to form an impermeable barrier. Soil additives can be used to increase stability and strength, reduce permeability and/or reduce the shrinking/swelling behaviour of the soil. Additives include chemical stabilisers and dispersants (e.g. latex emulsions, plastic films), cement, lime and bentonite. These technologies are effective only as short-term measures; further R&D is needed to develop more economical, long-term measures to fix contaminants in soil.

Summary of current R&D for fixing contamination in soil and use of engineered barriers

- Immobilisation of radionuclides via *in situ* incorporation into stable mineral phase

An NDA study started in 2010 focused on the geochemical behaviour of high activity fission products such as ^{137}Cs and ^{90}Sr, and long-lived actinides such as uranium in sediments and sludges. The study examines mineral precipitation and incorporation reactions as a potential route to irreversibly fix elements such as Sr, Cs and U in sediments to render them unreactive over long time scales and prevent further migration. Experiments will be performed in the presence of glacial till, clay and soil materials typical of United Kingdom contaminated sites (Burke, Shaw and Trivedi, 2010).

In microcosms (e.g. small localised microenvironments), mineral precipitation will be induced via manipulation of the groundwater geochemistry and microbiology to form either overgrowths of new minerals on sediment particles or recrystallisation of native minerals. The study will investigate the uptake rates and mechanisms of Sr, Cs and U during precipitation of the three most common stable mineral types in oxic conditions: ferric oxides [addition and oxidation of Fe(II,0) containing phases – e.g. magnetite, nZVI or green rust], carbonates (Ca) and urea-containing solution, and silicates (pH increase to pH 13+ inducing recrystallisation). All of these interventions are designed to produce a temporary change in groundwater conditions, which will ultimately return to background conditions, but with a permanent change in mineral assemblage and radionuclide speciation (Burke, Shaw and Trivedi, 2010).

Advanced X-ray [including synchrotron methods, high resolution X-ray diffraction (μ-XAS) and electron microscope (SEM/TEM) techniques] will be used to characterise the induced changes in mineralogy and in radionuclide speciation. Such treatment will provide the protocols necessary for remediation and characterisation of core samples (with low radiological hazard) collected from contaminated sites. This will allow the assessment of the potential effectiveness of mineral incorporation reactions for *in situ* grouting of contaminated land or the *ex situ* stabilisation of contaminated soils or sludges (Burke, Shaw and Trivedi, 2010).

Initial results have been published related to this research for Sr sorption in sediments. The results indicate that the Sr removal is greatest in systems with the highest initial nitrate loading and consequently more alkaline conditions at the end of denitrification (Thorpe, et al., 2012b). After denitrification, a limited re-release of Sr^{2+} back into solution occurred coincident with the onset of metal [Mn(IV) and Fe(III)] reduction, which caused minor pH changes in all microcosms with the exception of the bicarbonate-buffered system with its initial nitrate of 100 mM and final pH >9. In this system~95% of Sr^{2+} remained associated with the sediment throughout the progression of bioreduction. Analysis of this pH 9 system using X-ray absorption spectroscopy (XAS) and electron microscopy coupled to thermodynamic modelling showed that Sr^{2+} became partially incorporated within carbonate phases that were formed at a higher pH. This is in contrast to all other systems, where the final pH was less than 9. The XAS analysis showed that outer sphere Sr^{2+} sorption predominated. These results provide insight into the likely environmental fate of a significant radioactive contaminant ^{90}Sr, during changes in sediment biogeochemistry induced by bioreduction in nitrate-impacted nuclear contaminated environments (Thorpe, et al., 2012b).

These results correspond with the existing knowledge of cementitious system sequestration and immobilisation of Sr and other cationic radionuclide species under high pH conditions with calcium carbonate (Thorpe, et al., 2012b). Similarly, results have been published for the influence of abiotic versus microbial transformation of uranium valences that greatly affect the solubility and mobility of uranium contaminants in soils and sediments (Law, et al., 2011).

Differences in the soil/sediment mineralogy and pore water geochemistry have a considerable effect on the potential for radionuclide remobilisation in groundwater. This impacts the efficacy of groundwater remediations both in the short term during active remediation and in the longer term due to passive infiltration of regional groundwaters. Soil types with high distribution coefficients (Kds) retain the majority of the contamination on soil particles with minimal concentrations available in the pore water for treatment and removal. The sorption and incorporation of radionuclides within sediments is controlled by a number of factors, including the composition of the contaminant source, groundwater geochemistry, mineralogy, reaction time and the microbial community present.

This means that site-specific conditions will control the speciation of radionuclides; however, there has been little characterisation of *in situ* radionuclide speciation or mineral association due to high radiological hazards involved. This hampers the adoption of effective remediation strategies (such as “pump and treat” or other *in situ* soil washing techniques) as there is currently little grasp of how to select an effective radionuclide removal strategy based on contaminant speciation in the subsurface. A University of Manchester thesis used laboratory-scale experiments to investigate the mechanisms and effectiveness of radionuclide remobilisation of key associated radionuclides (^{137}Cs, ^{90}Sr and U), and how this varies with remediation techniques (i.e. different leaching solutions) and changing speciation (e.g. ^{90}Sr sorbed to iron oxides versus ^{90}Sr partitioned to calcite) (Thorpe, 2012a).

To understand which phases are the most likely source terms in sediments, characterisation protocols will also be developed to determine the likely speciation of these radionuclides in soils and sediments relevant to United Kingdom contaminated land legacies (Burke and Shaw, 2012). The sorption edge for ^{90}Sr was observed between pH 4-6 with maximum sorption occurring (Kd ~ 103 L kg^{-1} at pH 6-8). At ionic strengths above 10 mmol L^{-1}, and at pH values between 6 and 8, cation exchange processes reduced ^{90}Sr uptake to the sediment. These results suggest that over long periods, ^{90}Sr in contaminated sediments will remain primarily in weakly bound surface complexes. Therefore, if the groundwater ionic strength increases (e.g. by saline intrusion related to sea level rise or by design during site remediation) then substantial remobilisation of ^{90}Sr is to be expected (Wallace, et al., 2012).

Another report on *ex situ* soil treatment shows that caesium can be immobilised in soils with 96.4% efficiency by ball milling with nano-metallic Ca/PO_4 (Reddy Mallampati, et al., 2012). Ball milling is a type of grinder that rotates around a horizontal axis, partially filled with the material to be ground plus the grinding medium. Different materials are used as media, including ceramic balls, flint pebbles and stainless steel balls. An internal cascading effect reduces the material to a fine powder (Montinaro, et al., 2007). Ball milling treatment is a promising treatment for the remediation of caesium-contaminated soil in dry conditions. Results show that immobilisation efficiency increases from 56.4% in the absence of treatment to 89.9%, 91.5% and 97.7% when the soil is ball-milled for 30, 60 and 120 minutes, respectively. The addition of nano-metallic Ca/PO_4 increased the immobilisation efficiency to about 96.4% and decreased the ball milling time. Use of nano-metallic Ca/PO_4 over a short milling time also decreases Cs leaching (Reddy Mallampati, et al., 2012). Therefore, ball milling with nano-metallic Ca/PO_4 treatment may be potentially applicable for the remediation of radioactive Cs-contaminated soil in dry conditions (Burke, Shaw and Trivedi, 2010).

- Use of groundwater monitoring wells

Nanotechnology injection

As noted above in the subsection *Soil washing and thermal/chemical/biological treatment of soils*, due to their extremely small size, nanotechnologies are currently being used to chemically react with and break down organic contaminants *in situ*. Contaminants successfully sorbed to SAMMS™ particles, including radionuclides (US EPA, 2008b, 2011c, 2012a) and Prussian Blue nanoparticles, are being tested for caesium fixation and sorption to reduce crop uptake in Japan (Kawamoto, et al., 2012). Delivery of such subsurface technologies may provide an effective means of sorbing soluble contaminants prior to their reaching groundwater.

In situ chromate reduction and heavy metal fixation

At many impacted sites chromate and heavy metal contaminants are also present. In addition, advances dealing with the contaminants are often translatable to radionuclides. *In situ* chromate reduction is an innovative technology. The approach consists of *in situ* reduction of chromates with a ferrous salt, and fixation of the metals using a destabilised aqueous sodium silicate solution. The silica treatment serves two purposes: it reacts with the metal and metal hydroxides to reduce metal solubility and it lowers soil permeability, thereby reducing the leaching rate of the treated soils. The primary objective of this technology is to remediate heavy metal contamination in soil (IAEA, 1999e). Available *in situ* technologies or treatment approaches for chromate contamination use chemical reduction and fixation for remediation [e.g. geochemical fixation, permeable reactive barriers (PRB) and reactive zones]. Other types of *in situ* approaches under development include enhanced extraction, electrokinetics and biological processes that can be used in phytoremediation and natural attenuation, and within PRB and reactive zones (US EPA, 2013a; Wuana and Okieimen, 2011).

Chemical oxidation/reduction

Chemical and/or microbial reducing agents can be injected into contaminated soil or unconfined aquifers to create a subsurface treatment barrier to immobilise or destroy target contaminants. Then water containing the reaction by-products and any remaining reagent is pumped back out and processed. The treatment barrier is a zone of favourable redox potential. The goal is to effectively transform dissolved metals and radionuclides to less soluble forms and to promote the destruction of organics, especially chlorinated hydrocarbons. This innovative technology allows *in situ* treatment of groundwater contaminants and avoids disposal costs, since metals and radionuclides are immobilised in place (IAEA, 1999e; US EPA, 2014).

Reactive gas injection

Feasibility studies for treating unsaturated soils by injection of reactive gases are being tested. Dilute mixtures of hydrogen sulphide in air or nitrogen are being used to treat soils contaminated with heavy metals, while chromate or uranium-contaminated soils are being treated with hydrogen sulphide and sulphur dioxide gas mixtures diluted by inert gases. Reactive gas injection is an innovative technology still under experimentation (IAEA, 1999e). This technology is being tested to immobilise uranium at the United States Hanford facility (Szecsody, et al., 2010).

Sparging

Air sparging is a technology in which air is bubbled through a contaminated aquifer creating an underground stripper that removes contaminants by volatilisation. The air bubbles carry the contaminants to a vapour extraction system. Air sparging operates at high flow rates to maintain increased contact between groundwater and soil. Target contaminants include volatile organic compounds (VOC) and fuels. A variation of the technology is to pull a vacuum on a groundwater well, lifting contaminated groundwater up into the well. Some of the VOC in the contaminated groundwater are transferred to air bubbles that rise and are collected at the top of the well by vapour extraction. The partially treated groundwater is never brought to the surface. It is forced into the saturated zone and the process is repeated. As groundwater is circulated through the treatment system *in situ*, contaminant concentrations are gradually reduced (IAEA, 1999e).

- Thermal fixation, passive/reactive barriers, grout walls and engineered wetlands

Thermal treatment processes range from low to extremely high temperatures to remove or completely destroy polluting constituents in wastes. There are basically two classes of *in situ* thermal treatment systems. The most extreme is *in situ* vitrification, which serves to destroy the waste and immobilise radioactive contaminants in a glass matrix that forms as a result of soil melting. At the other end of the spectrum are thermal enhancements designed to drive off organics and thus reduce radioactive transport by eliminating chelation effects. As noted previously, thermal treatment offers quick clean-up times but is typically the most costly. *In situ* thermal treatment is typically used for the treatment of soils, sludges, sediments and groundwater and does not apply to bulk wastes (IAEA, 1999e; Yoon, et al., 2012; US EPA, 2013c; Ahmad, et al., 2012; Dresel, et al., 2011).

Polymers such as lignosulphonate and latex have been applied in the aftermath of Chernobyl and Fukushima efforts to control contamination throughout the site involved spraying polymers to prevent dispersal of radioactive materials via wind and precipitation. Applications to soil tend to be polymeric films (latex, etc.) and polymeric structure formers (interpolyelectrolyte complexes, IPEC, etc.) (Zezin, et al., 2012; Mikheykan, 2011; Watanabe, 2012). This approach has been used in combination with sorbents to treat farmland contaminated with radiocaesium in Japan. Adsorbents based on dibenzo-20-crown 6-ether (DB_20C_6) with high caesium selectivity have been newly developed (Yamaguchi, et al., 2012). The effectiveness of these caesium sorbents has been demonstrated through field tests in Iitate-mura, Fukushima. A technique for decontaminating farm soil surfaces by combining poly-ion complex and bentonite clay, which enables a fixation of soil surface while suppressing dust discharge, succeeded in removing more than 90% of radioactive caesium from farm and grass soils. A radioactivity removal rate of 91-96% has been accomplished by spraying a poly-ion complex solution on the fields after mowing (Itoh, et al., 2012). A report on the application of $K_4\,[Fe(CN)_6]\cdot 3H_2O$ at a rate of 1.3 g/kg in soil finds that it reduces the fraction of exchangeable ^{137}Cs 100-fold (100 times). This method is effective for plots with contamination concentrated in the top 1-2 cm of soil (Epifanova and Tertyshnik, 2012).

A permeable reactive barrier (PRB) is defined as an *in situ* method for remediating contaminated groundwater that combines a passive chemical or biological treatment zone with subsurface fluid flow management. Treatment media may include zero-valent iron, chelators, sorbents and microbes to address a wide variety of groundwater contaminants, such as chlorinated solvents, other organics, metals, inorganics and radionuclides. The contaminants are concentrated and either degraded or retained in the barrier material that may need to be replaced periodically. There are approximately 100 PRB operating in the United States and at least 25 internationally. PRB can be installed as permanent or semi-permanent units. The most commonly used configuration is that of a continuous trench in which the treatment material is backfilled. The trench is perpendicular to and intersects the groundwater plume. Another frequently used configuration is the funnel and gate, in which low-permeability walls (the funnel) direct the groundwater plume toward a permeable treatment zone (the gate) (US EPA, 2013b).

- Adaptation with foam-reducing liquid waste

In addition to mineral and polymeric fixatives, foams and emulsions are being evaluated to prevent further migration of radionuclides after release to the environment. Emulsions and foams can be applied to minimise water intrusions and the generation of radioactive waste water associated with run-off (Fox and Medina, 2005). A comprehensive review of the use of fixatives to contain and control contamination as part of the response to radioactive dispersal devices (RDD) found that there is a lack of quantitative information on the performance of potential materials under field conditions (Al-Tabbaa and Perera, 2002; Parra, Medina and Conca, 2009). Development and testing of some comprehensive response capabilities that could be used to impede the migration of radionuclides in soil have been tested, such as part of the United States DOE field demonstration results of the Contamination Control Unit (CCU). The CCU was developed by the Buried Waste Integration Demonstration (BWID) for the US Department of Energy (DOE) Office of Technology Development. The CCU is a self-contained, field-deployable, trailer-mounted system designed to control contamination spread at the site of TRU handling operations. The CCU utilizes a vacuum system and is capable of dispensing soil fixatives, dust suppression agents and misted water. The soil fixative application system uses a soil fixative (3M foam) mixed with water to create a pale yellow foam material that is dispensed using volume expansion nozzles. This product provides a long-term, vapour suppressing foam for covering uneven contaminated soil (e.g. steep sloping dig faces) (Thompson, Freeman and Wixom, 1993).

Future suggested R&D for fixing contamination in soil and use of engineered barriers

- ***Description*** – Develop and test *in situ* and *ex situ* methodologies to fix, contain and control contaminants in soil to prevent airborne dispersal migration to groundwater.
- ***Objectives*** – Develop soil contamination fixation and intervention methods for long-term and short-term containment soil contaminants that are effective for various soil types and environmental conditions.
- ***Desired deliverables*** – Technologies for fixing and intervening in soil contamination to prescribed limits and prevent migration to groundwater.

Methods for decontaminating large volumes of water and chemicals to low levels

Challenges

Decommissioning often requires removal and treatment of large volumes of water due to groundwater intrusion into excavations during soil remediation and into below-grade structures during demolition. Pre-existing groundwater contamination plumes associated with facility operation can also require remediation prior to facility release from regulatory control. Unlike high-purity, high-activity water treated by conventional water processing systems, groundwater- and construction-related water often have high conductivity and turbidity, and can contain organic contaminants such as hydrocarbons and polymers. The high conductivity is caused by cations such as K^+ and Ca^+ in soil and rubblised concrete as well as some anions. These compete with contaminants for active sites on ion exchange resins and can overwhelm and remove contaminant ions from resin media. Similarly, active sites in zeolites and activated charcoals can also be rendered inefficient by non-contaminant constituents in groundwater and construction water. High turbidity from suspended solids can quickly clog filtration media and oils, and polymers from pre-existing leaks and spills or uncured or dissolved surface contamination fixatives can coat resins and clog filtration media. Demolition-related construction water is often in contact with rubblised concrete and concrete dust, creating high pH cementitious conditions that can challenge conventional water treatment systems.

These factors often render conventional water treatment applications inefficient and unable to keep up with the water removal and processing demands necessary to support decommissioning activities. This creates water storage and transport challenges since treatment and release of the collected water often lags far behind the excavation or demolition activity. In addition, large volumes of secondary waste from filtration media such as charcoal beds and conventional filters, as well as spent ion exchange media, are often generated and require disposal as radioactive waste. The following challenges associated with treating large volumes of water associated with decommissioning were identified by the working group:

- the associated cost of treatment and the volumes of secondary waste produced;
- post-operative clean-outs of effluents and groundwater;
- the limitation to organic liquids.

Early treatment remedies for groundwater contamination were primarily "pump and treat" operations. Because of the relatively high cost and often lengthy operating periods for these remedies, the use of *in situ* treatment technologies is increasing (US EPA, 2008b). Typically these large volumes of decommissioning-related water have relatively low levels of contaminants in comparison to the lower volume, more radioactive water from draining systems collected in sumps and from decontamination activities associated with the dismantlement phase of decommissioning. The water treatment challenges that unfolded at Fukushima where in-plant piping systems, including lube oil systems, were damaged by the earthquake/tsunami, inundated with sea water and silt and subsequently rendered highly radioactive by direct sea water injections into the cores of four reactors was an exception to this general rule and created high activity water that posed all the challenges of processing and treatment created by D&D activities. Water processing systems had to be developed and deployed to treat large volumes of extremely high activity water (up to 1-2 Sv per hour on the surface). The water was of very low quality, containing fouling suspended solids, biological polymers and oils, as well as ion-exchange-depleting, high-conductivity sea water rich in competing anions and cations. In addition, the release and dispersal of radioactivity across Japan has required development and deployment of water treatment systems to decontaminate swimming pools, ponds and groundwater. Thus, there is a great interest and a push for systematic research and development of water treatment technologies to address these challenges (Abdel Rahman, Ibrahium and Hung, 2011).

Summary of current R&D for decontaminating large volumes of water and chemicals to low levels

- Physical and chemical treatments for stabilisation and containment

Perhaps the most ambitious project for the treatment of contaminated groundwater to date is ongoing at the US DOE's Hanford Facility. Clean-up of the site has been under way since 1989 and will likely continue for another 40 years or more. Many different chemicals and radioactive materials are present in Hanford's groundwater. Contaminants include chemicals such as carbon tetrachloride, chromium and nitrate and radioactive materials such as uranium, strontium-90, technetium-99, tritium and iodine-129. More than 70 square miles of groundwater is contaminated above regulatory standards. Some of those contaminants, chromium, nitrate, uranium, technetium, tritium and strontium have reached the Columbia River (Oregon DOE, 2012).

Eliminating the flow of contaminants in groundwater to the Columbia River and eventually cleaning up the groundwater is one of the most difficult challenges for the

Hanford clean-up. Initially, there were several interim groundwater remediation systems in operation, including five pump and treat systems, two passive groundwater treatment systems along the Columbia River, and a pump and treat system and soil vapour extraction unit on the Central Plateau. Over 40 million gallons of groundwater were treated every month with these interim systems. Development of new water treatment facilities using newly developed technologies and processes has greatly increased capacity in new installations and lowered secondary waste generation costs. The new systems draw groundwater from networks of over 50 and up to hundreds of wells to treat groundwater at rates of 2-5 cubic metres per minute (Oregon DOE, 2012; Reyes-Mills, 2011; Cary, 2012).

Development of resins and other media that have higher specificity and sorption capacities for contaminants in water is ongoing. Ion exchange water processing provides a conventional method of processing large volumes of water. The DOE has sponsored development of water treatment systems at Hanford capable of processing large volumes of waste water associated with the waste vitrification process and other systems for remediation of groundwater using a pump and treat method. The development of higher capacity and selectivity resins for hexavalent chromium has led to a dramatic lowering in spent resin generation rates, allowing the system to be run for 18 months without requiring demineraliser media change-out (US DOE, 2012). The new plant is expected to pump up to 2 000-2 500 gallons of water a minute and operate 24 hours a day. When the water is cleaned to drinking water standards, it will be re-injected into the ground in key places to contain contamination and push it toward the wells that pump out the water. Over the lifetime of the plant, it is expected to treat 25 billion gallons of groundwater and remove 77 000-110 000 pounds of carbon tetrachloride. It will also remove radioactive technetium-99 and iodine-129, plus chromium, trichloroethene and nitrates (Cary, 2012). Pump and treat efficiencies in different soil types using samples from contaminated sites in the United Kingdom are also being evaluated (Burke and Shaw, 2012).

Hanford has also experimented with a number of other technologies, such as an underground chemical barrier that was created near Hanford's D Reactor to convert hexavalent chromium in groundwater into a less mobile and less toxic form as water flowed through the barrier. Parts of the barrier failed, making pump and treat necessary to augment that portion of the barrier that is working. A different type of chemical barrier using calcium phosphate was formed near Hanford's N Reactor. As radioactive strontium in groundwater flows through the barrier, it binds to the calcium phosphate (also called apatite). This barrier shows promise and is being expanded (Oregon DOE, 2012).

Hanford has also tested a process called biostimulation, using molasses and vegetable oil to feed tiny micro-organisms (bacteria) in the soil, which simultaneously consume oxygen in the groundwater. This soil and groundwater chemistry is altered, forming redox conditions and changing chromium to a less mobile and less toxic form (Oregon DOE, 2012).

The difficult conditions at the Fukushima reactors have required development of innovative water treatment systems capable of treating the large volumes of water that collected in the buildings and that accumulated in the forebay and intake structure. The disaster has fostered a remarkable effort by the Japanese to understand and develop improved decontamination and remediation technologies (Farr, 2011a, 2011b, 2011c). The Japanese National Institute for Materials Science (NIMS) is collecting basic data on natural minerals produced in various regions and inorganic materials with different chemical compositions as a tool for selecting suitable materials for sorbing radionuclides under different environmental conditions. They will make this information available in an NIMS Materials database (MatNavi) (NIMS, 2012). The use of natural minerals such as zeolite as adsorbents is under study as the most promising approach. But even natural minerals having the same group name have varying adsorption capacities, depending on the chemical composition and regions from where the material was derived. Performance also varies greatly depending on use conditions, such as the radioactivity concentrations and the pH of the water being treated. It is necessary to select the optimum adsorbent for the conditions at each site.

NIMS is collecting basic data on natural minerals produced in various regions and inorganic materials with different chemical compositions as a tool for selecting suitable materials. The focus is on adsorbents for caesium, strontium and iodine. NIMS has collected nearly 800 basic data items for 60 species of materials from various localities and with various chemical compositions. The water treatment applications being studied range from sea water, which was used to cool the reactor core and is accumulating at the power plant site, to contaminated soil in the immediate vicinity of the plant and in the larger region around the plant (such as rice paddies, fields, orchards, woods and forests, surface waters, buildings and roadways). The distribution of contaminated water is also extremely diverse, encompassing sea water, river water, ponds, lakes, pools, agricultural water, etc. To remove radioactive substances from this diverse range of sites, NIMS is performing experiments and collecting data on many types of adsorbents under a wide variety of conditions. Database construction is being carried out by a total of seven universities, four independent administrative institutions (IAI), and one foundation under Dr. Hirohisa Yamada, Group Leader of the Functional Geomaterials Group at NIMS (2012). This research is bound to yield information on material selection and application as well as treatment system methods and processes that will be beneficial for facility decommissionings (Blet, 2012).

The use of Chitosan, a polysaccharide biopolymer, and its derivatives in combination with other biological and organic materials is also being evaluated for sorption of radionuclides (Homeland, 2011; Muzzarelli, 2011). Chitosan and its derivatives have been used in many diverse applications, including coatings in food packaging, numerous biomedical applications and as a flocculent in waste water treatment. Chitosan has been found to have affinity for a number of metals as well as various dyes, due in part to its availability of amine and hydroxyl functional groups capable of complexation. The use of chitosan as a potential media for decontamination of aqueous materials has been investigated for various radionuclides, including uraniums, ^{60}Co, ^{137}Cs, ^{85}Sr, ^{60}Co, ^{152}Eu and ^{238}U (Holfeltz and Paulenova, 2012; Abdul Nishad, et al., 2012; Galamboš, Suchánek, Rosskopfová, 2012). Chitosan availability, biodegradability and biocompatibility make it an appealing media for decontamination of large-volume waste water streams or other environmental materials that are not candidates for traditional chemical decontamination agents (Holfeltz and Paulenova, 2012).

The properties of inorganic materials such as zeolite are also being studied intensely to better understand the sorption mechanism. Nonatitanate and zeolite are effective sorbents in systems with low calcium ions and low salinity, thus limiting their application for large-scale treatment of decommissioning waste waters (Merceille, et al., 2012). Titanosilicates have also shown promise for treating waste waters (Popa and Pavel, 2012). Crystalline silico-titanates (CST) developed at Sandia Laboratories are being used to treat more than 43 million gallons of contaminated waste water at Fukushima. The inorganic, molecularly engineered, CST ion exchangers can be sized specifically for caesium and other elements. They are effective for lowering highly concentrated radioactivity levels from contaminated water. The remaining lower concentration of radioactive waste can be treated in a more economical and less hazardous way (SNL, 2012).

Mesoporous materials and some (hybrid) mesoporous solids have been investigated as solid ligands to remove actinides and fission product contaminants from liquid effluents, but also as model materials to investigate radiation defects as possible nuclear waste disposal forms and as functional materials to be placed in or close to new forms of nuclear waste matrices. A mesoporous material is a material containing pores with diameters between 2-50 nm. Some modified mesoporous materials can be synthesised to have larger pores and have potential for application in water treatment systems. Synthesised mesoporous materials possess high surface areas, large pore sizes, narrow pore size distributions and high thermal, hydrothermal and mechanical stabilities. Modified mesoporous materials are found to have high selectivity and capacity in the adsorption and separation of the transition and heavy metal ions in the aqueous solution.

In addition, they showed great selectivity and capacity for the adsorption and separation of the radioactive materials in aqueous medium (Makowski, et al., 2012; Al-Othman, 2006).

Multi-wall carbon nanotubes (MWCNT) are also being developed as a promising technology for contaminant clean-up and water processing (Yavari, Huang and Ahmadi, 2011; Gupta, Agarwal and Saleh, 2011; Tan, et al., 2008; Burke and Shaw, 2012).

Future suggested R&D for decontaminating large volumes of water or chemicals contaminated to low levels

- ***Description*** – The working group had no recommended R&D activities to be investigated for treating large volumes of water; however, it is evident that remediation efforts and research and development in the United Kingdom, Japan and the United States for current groundwater remediation challenges are delivering new insights into treatment processes and effective approaches. Further research and development of emerging technologies such as mesoporous materials and carbon nanotubes warrants further research and development by the industry.
- ***Objectives*** – To develop and test emerging materials and nanotechnologies for contaminant isolation and removal in systems capable of treating large volumes of water while minimising the generation of secondary wastes.
- ***Desired deliverables*** – Further research on the design of new materials and their modification to increase contaminant collection and sequestration efficiencies. Laboratory and field tests of new treatment technologies and processes capable of being scaled for treatment of large waste water volumes associated with decommissioning facility demolition and groundwater remediation.

Suggested areas of future collaboration

Suggested collaboration to resolve common national programme issues include: *i)* the application of new physical and chemical processes to remediate concrete, such as laser cleaning, scabbling, nitrogen blasting, gel coating, as well as crushing/disposal using volumetric criteria; *ii)* minimising the limiting factors that currently prevent the use of robotic technology in high radiation or contaminated areas, including the high cost associated with both robotic technology and development of specialised tools.

5. Materials and waste management

Theme overview

This section focuses on a wide spectrum of practices and processes for the treatment and disposal decommissioning wastes. This includes such areas as conditioning, handling, transport, interim storage, treatment for recycling, disposal, material clearance and entombment. The working group analysed a wide array of issues associated with nuclear decommissioning facility waste management, as illustrated in Table 5.1.

Table 5.1: Waste management issues

Item	Description
1	Management of problematic wastes – chemical (PCB, asbestos, etc.) and mixed waste
2	Treatment/removal (including mineralisation) of organic materials (bituminised waste, resins, oils, nitrates), activated sodium
3	Treatment of reactive metals (high-temperature processes, melting) and managing gas generation
4	Development of dynamic chemo-toxic inventories from chemical reactions during storage
5	Management of depleted uranium
6	Improved segregation of waste (separation of long-lived components from LLW); segregation of VLLW
7	Clearance and recycling of low contaminated materials
8	Treatment (including characterisation, reporting) of materials with hard-to-measure nuclides, e.g. using melting for characterisation
9	Monitoring of waste packages during interim storage and long-term management (traceability) of waste package data
10	Handling and treatment of degraded waste packages
11	Conditioning of waste (different grouts, foam concrete, etc.; improving waste incorporation)
12	Avoiding radiolysis inside the casks (because of beta/gamma emitters and water)
13	Long-term monitoring of entombed facilities including avoidance of voids
14	Long-term performance of waste forms (e.g. concrete, impact of super-plasticisers on radionuclide migration)

The management of problematic and mixed waste and of organic materials, even if not involving large quantities, is a common high cost issue within almost all countries. Some initiatives have already been launched, mainly in the field of volume reduction, but a great amount of work remains in relation to the final disposal of organic residues. The clearance and recycling of low-level contaminated materials, even if raising some challenges, is considered by most countries to be mainly policy related and based on sound technical aspects.

Regarding conditioning of waste, although several countries do not envisage the modification of their own methodologies, others suggest the possibility of optimising the known processes (e.g. cementation) by investigating the interactions of waste and encapsulation matrices to improve long-term behaviour and/or incorporation factors.

Summary of current practices and guidance

Current practices for the most vexing waste forms such as high-level waste, mixed hazardous waste and radioactively contaminated organic wastes rely heavily on stabilisation technologies that increase waste volumes and require costly development, testing and approval processes. In addition, not all countries have suitable climates and geologies for disposal of such wastes. Many countries have "orphan wastes" containing transuranics, asbestos, PCB, chromium, etc. Stabilisation and disposal in landfills is currently the only viable and most widely used option for disposal. High-level wastes (HLW) left over from spent fuel reprocessing contain high levels of transuranics with half-lives of hundreds of thousands of years. The current approach is to vitrify the HLW to stabilise it for disposal in geological repositories. It is very difficult to predict with certainty the repository conditions and stability of the waste form over such long time horizons. Similarly, graphite reactors' long-lived constituents such as ^{14}C and ^{36}Cl are highly mobile in the environment. Technologies that recover and concentrate the long-lived radioactive constituents from such waste forms would reduce the complexity of viable disposable solutions and greatly reduce the high-level waste volumes. They would also allow recovered transuranics to be used in MOX fuel for fast neutron reactors thereby eliminating the long-lived radionuclides and simplifying disposal options. It may also be viable to use recycled graphite in a new generation of reactors.

Stabilisation is also a common practice for treating organic waste prior to disposal. Pyrolysis and WOX incineration technologies are being used to a limited extent to volume reduce such waste, leaving contaminated salts that are more amenable to cementitious and vitrification stabilisation methods.

On the other end of the spectrum, the vast majority of decommissioning waste volumes are free of or have very low levels of contamination, and practices are being developed in Europe to minimise waste volumes requiring disposal at low-level radioactive waste facilities. The IAEA (2004) has formulated concentrations below which materials are suitable for disposal at municipal landfills. The very low-level waste (VLLW) concentrations result in less than 10 μSv per year of exposure to members of the public transporting and handling the waste, and to future residents at the site.

The United Kingdom has adopted these levels (CEWG, 2006) and is successfully using them at facilities such as Bradwell to clear materials (Sexton, 2011b). Similar provisions are available in the United States under 10 CFR 20.2002 (US NRC, 2013a) alternate disposal provisions (< 50 μSv/yr) and through the state of Tennessee's Bulk Survey for Release (BSFR) programme (< 10 μSv/yr) (Tennessee DOEC, n.d.). The alternate disposal provisions allow licensees to model disposal locations using fate and transport software such as RESRAD or RESRAD-OFFSITE to estimate doses to future residents, and to perform time motion estimates for doses to workers transporting and handling the waste. As this is authorised on a case-by-case basis in the United States and requires publication in the Federal Register and public comment on each petition to the NRC, alternate disposal authorisations require advance notifications of 6 months to a year.

Big Rock Point disposed of waste at a local landfill using this provision, and Humboldt Bay has obtained permission for disposals at a licensed hazardous waste facility which, due to the design, climate and geology of the site, can receive somewhat higher concentrations than municipal landfills. Under the BSFR programme in Tennessee, several municipal landfills have been modelled by waste processors, such as Studsvik, and they have been licensed to dispose of VLLW with concentrations shown to result in exposure of less than 10 μSv/yr to a hypothetical resident farmer occupying the landfill after the post-closure monitoring period. The BSFR programme was used extensively by Connecticut Yankee for marginally contaminated soil and concrete disposals.

Table 5.2: Guidance documents on materials and waste management

Facility type	Phase	Region	Document
All types	All phases	International	*Monitoring for Compliance with Exclusion, Exemption and Clearance Values*, IAEA (2005)
All types	All phases	International	*Application of the Concepts of Exclusion, Exemption and Clearance*, IAEA (2004)
All types	All phases	International	*Radioactivity Measurements at Regulatory Release Levels*, OECD/NEA (2006)
All types	All phases	International	*SADRWMS Project – International Project on Safety Assessment Driving Radioactive Waste Management Solutions*, IAEA (2013g)
All types	All phases	International	*The Safety Case and Safety Assessment for Predisposal Management of Radioactive Waste*, IAEA (2013h)
All types	All phases	International	*The Management System for the Processing, Handling and Storage of Radioactive Waste*, IAEA (2008c)
All types	All phases	International	*The Management System for the Disposal of Radioactive Waste*, IAEA (2008b)
All types	All phases	International	*Classification of Radioactive Waste*, IAEA (2009b)
All types	All phases	International	*Disposal of Radioactive Waste*, IAEA (2011b)
All types	All phases	International	*Predisposal Management of Radioactive Waste*, IAEA (2009e)
All types	All phases	International	*Storage of Radioactive Waste*, IAEA (2006f)
All types	All phases	Europe	*Opinion of the Group of Experts Established Under Article 31 of the Euratom Treaty on the Revised Basic Safety Standards for the Protection of the Health of Workers and the General Public Against the Dangers Arising from Ionising Radiation*, EC (2010)
All types	All phases	Europe	*Recommended Radiological Protection Criteria for the Recycling of Metals from the Dismantling of Nuclear Installations*, EC (1998)
All types	All phases	Europe	*Definition of Clearance Levels for the Release of Radioactively Contaminated Buildings and Building Rubble*, EC (1999)
All types	All phases	Europe	*Practical Use of the Concepts of Clearance and Exemption, Part I – Guidance on General Clearance Levels for Practices*, EC (2000a)
All types	Decommissioning	International	*Managing Low Radioactivity Material from the Decommissioning of Nuclear Facilities*, IAEA (2008d)
All types	Decommissioning, care and maintenance, waste storage facilities	International	*Predisposal Management of Low and Intermediate Level Radioactive Waste*, IAEA (2003c)
Fuel cycle	Decommissioning, care and maintenance, waste storage facilities	International	*Predisposal Management of High Level Radioactive Waste*, IAEA (2003b)
Fuel cycle and waste processing facilities	Operational and decommissioning	International	*Application of Thermal Technologies for Processing of Radioactive Waste*, IAEA (2006a)
Waste processing, storage and disposal	All phases	International	*CRAFT Project – International Project on Complementary Safety Reports: Development and Application to Waste Management Facilities*, IAEA (2013a)
Waste disposal facilities	Pre-operational	International	*Siting of Near Surface Disposal Facilities*, IAEA (1994)
Waste disposal facilities	All phases	International	*PRISM: Practical Illustration and Use of the Safety Case Concept in the Management of Near-Surface Disposal*, IAEA (2013e)
Waste disposal facilities	All phases	International	*Borehole Disposal Facilities for Radioactive Waste*, IAEA (2009a)

Table 5.2: Guidance documents on materials and waste management (*cont'd*)

Facility type	Phase	Region	Document
Waste disposal facilities	All phases	International	*GEOSAF – International Project on Demonstrating the Safety of Geological Disposal*, IAEA (2014a)
Waste disposal facilities	All phases	International	*Geological Disposal Facilities for Radioactive Waste*, IAEA (2011c)
Waste disposal facilities	All phases	International	The Safety Case and Safety Assessment for the Disposal of Radioactive Waste, IAEA (2012b)

Summary of challenges and R&D needs

The following provides a summary of the materials and waste management challenges cited by the respondents within the "Future R&D and Innovation Needs for Decommissioning" Task Group.

- *Management of problematic and mixed waste*. Many member countries indicated there were problems with "orphan waste" for which there is no clear disposal path. Wastes containing PCB and asbestos were commonly cited as a decommissioning challenge that should be addressed. Similarly, management of water containing hexavalent chromium, which is a highly carcinogenic form of chromium in the +6 oxidation state, was commonly cited as a challenge. Chromium was commonly used in secondary systems, such as component cooling, as an oxidation inhibitor in many of the initial reactors designed. This has led to issues with hexavalent chromium water generated when draining systems in preparation for D&D and also with groundwater contamination from leaking systems containing chromium or on-site disposal practices in some of the older facilities such as Hanford in the United States. Mixed waste streams for which the waste form is either not determined or not optimised could lead to low waste loading factors increasing the disposal volume of the stabilised waste form, hence increasing the number of final waste packages (FWP) required. Management of Pu-contaminated waste forms and tritiated waste were also identified as problem waste forms with limited disposal options.

- *Treatment/removal of organic materials*. Organic materials decompose in storage and disposal and do not lend themselves to conventional cementitious stabilisation methods. Decomposition changes the chemical make-up of the waste, potentially degrading the stabilisation material and enhancing the mobility of radionuclides and hazardous materials. Even management of low-level organic waste forms (bituminised waste, resins from PWR operations, trichloroethylene liquid waste and contaminated oils) is problematic. In this sense development of new waste conditioning techniques poses a challenge across many waste forms.

- *Treatment of reactive metals*. Potential long-term gas production in a repository by corrosion of metals due to possible water infiltration is also a concern. Reactor internals are activated to levels unsuitable for conventional land disposal. The high-level waste or Greater-Than-Class C (GTCC) wastes are currently being stored in inerted canisters at spent fuel storage facilities until suitable long-term disposal facilities are constructed. Wastes from sodium-cooled reactors and sodium-bonded fuels are also highly reactive with water and require passivation prior to disposal.

- *Chemical reactions and monitoring during storage*. There is also uncertainty regarding waste forms and chemical reactions and changes in the final repository. The potential for radiolysis and generation of highly reactive peroxides and free radicals in high-activity wastes further complicates understanding of the long-term chemical changes in waste forms. More studies of the material inventory of wastes and investigation of possible interactions and reactions are needed. The issue may increase in importance as part of licensing the final repositories, as requirements from the authorities are still somewhat uncertain. In addition, the capability to monitor waste integrity and physical/chemical parameters during storage will be required to verify that the repository environment and wastes are behaving as predicted and to identify potentially threatening situations. The planning, equipment and facilities to handle degraded waste packages is also a challenge that has been identified by some countries.

- *Management of depleted uranium*. Several countries have large stocks of depleted uranium in both oxide and hexafluoride form. Due to the long time horizons required for safe disposal and the corrosive and volatile nature of hexafluorides, these waste present disposal problems. A further complicating factor is that this material is potentially usable as an energy source in MOX fuel for fast neutron reactors or breeder reactors.
- *Improved segregation of waste*. Several countries have limited land areas with few locations suitable for land disposal facilities. Waste volume reduction through improved segregation of waste is a priority to reduce disposal costs and the waste volumes requiring disposition to low-level waste facilities.
- *Clearance of low contaminated materials*. Many countries are interested in developing clearance methodologies similar to those being implemented in the United Kingdom for very low-level wastes. Under such strategies, very low-level waste, where potential exposures are less than 10 μSv/year, are allowed to be disposed of in municipal landfills, thus avoiding disposal of low risk materials at low-level waste facilities. This helps to greatly conserve the capacity of low-level waste disposal facilities for higher-risk waste.

Suggested additional research and development

In light of both the material and waste management challenges identified by the respondents, and of current R&D, the following additional R&D topics are suggested.

Management of problematic and mixed waste

Challenges

There are many challenges associated with the management of problematic wastes such as magnesium-rich sludges associated with Magnox fuel storage, encapsulation, and storage of high-activity-level and intermediate-level wastes. Challenges also arise with: *i)* treating and managing hazardous and radioactive wastes such as contaminated asbestos and radioactive waste containing hexavalent chromium; *ii)* the development and deployment of modular processing and treatment systems that process the wastes directly or are required for their collection and clean-up. The working group identified the following challenges under this topic:

- management of problematic wastes – chemical (PCB, asbestos, etc.) and mixed waste;
- research on waste forms for plutonium-containing species;
- development of a methodology to calculate CLEMC (CL for small areas of elevated activity);
- safety and security of recycled orphan sources and disposition path identification;
- tritiated waste desiccation.

Summary of current R&D for management of problematic and mixed waste

- Reduction treatment of hexavalent chromium into trivalent chromium

Hexavalent chromium is a heavy metal that is a human carcinogen by the inhalation route of exposure and a possible human carcinogen by the oral route of exposure. Due to the use of chromated solutions as a corrosion inhibitor in nuclear facility systems, some sites have solid wastes and waste water that require treatment as part of decommissioning. In addition, hexavalent chromium wastes were not regulated until the 1970s, and prior on-site disposal practices have led to contaminated soil and groundwater issues with hexavalent chromium at some nuclear facilities (French, 2012). Reduction to trivalent chromium converts hexavalent chromium, Cr(VI), to a less mobile, less toxic form, Cr(III). Zero-valent iron (ZVI) is often used as a Cr(VI)-reducing agent (Ratnadeepa, et al. (2010). A considerable volume of research has been carried out to investigate the mechanism and kinetics of Cr(VI) reduction with ZVI, as well as the influence of various parameters controlling the reduction efficiency (Gheju, 2011; Haruo, et al., 2004; Djouider, 2012).

Removal of hexavalent chromium from waste water typically involves chemical reduction of the chromium. A recent study evaluated the effectiveness of Fe(II)-treated faujasite [zeolite Fe(II)-Y] for reduction of Cr(VI) and immobilisation (adsorption/ co-precipitation) of the Cr(III) reaction product. The Fe(II)-faujasite material effectively removed high concentrations of dissolved Cr(VI) from aqueous solution resulting in Cr solid loadings as high as 0.30 mmol Cr per gram Fe(II)-faujasite or ~1.5% Cr (w:w). Results of Cr K-edge X-ray absorption near edge spectroscopy (XANES) confirmed that the oxidation state of Cr in Cr(VI)-treated Fe(II)-faujasite was Cr(III) (Kiser and Manning, 2010). The higher concentrations achieved could be effective in minimising waste volumes for treatment of Cr(VI) contaminated groundwaters. Chromium-specific ion exchange resins are also available for water treatment (Milkey, 2010; Oregon DOE, 2012).

Bioremediation using bacteria is also being investigated for *in situ* and *ex situ* reduction and sequestration of Cr(VI) to Cr(III) (Schaffner, et al., 2008). A recent investigation involved microbial remediation of Cr(VI) without producing any by-product. Bacterial cultures tolerating high concentrations of Cr were isolated from a soil sample collected from the chromite-contaminated sites of Sukinda, and their bioaccumulation properties were investigated. Strains capable of growing at 250 mg/L Cr(VI) were considered as Cr resistant. The experimental investigation showed the maximum specific Cr uptake at pH 7 and temperature 30°C. At about 50 mg/L initial Cr(VI) concentrations, uptake of the selected potential strain exceeded 98% within 12 h of incubation. The bacterial isolate was identified by 16S rRNA sequencing as Brevebacterium casei (Das and Mishra, 2010).

As noted in US EPA (2011c) and Wuana and Okieimen (2011), there are a variety of groundwater remediation techniques being evaluated, including nanoparticles such as nZVI, bi-metallic nanoscale particles (BNP), and emulsified zero-valent iron (EZVI) for *in situ* hexavalent chromium remediation and carbon nanotubes (Xie, Wang and Xu, 2012; Gholipour, Rafsanjani and Goharrizi, 2011; US NRC, 2008b) that may be applicable for waste treatment.

- Management of asbestos

Asbestos materials are being thermally and chemically treated to alter the fibres such that they are non-hazardous and can be recycled. Thermally treated asbestos is being recycled for use in cement, ceramics and other products (EEN, 2012; Bernardo, et al., 2011; Ding, Peng and Chen, 2012; Faik, 2012). Asbestos waste (pure chrysotile asbestos and asbestos cement) was treated under hydrothermal conditions using different acids in various temperatures in order to produce a material that is non-toxic and can be used as an adsorbent for petroleum pollutants (Kousaiti, et al., 2010).

- Development of alternative encapsulants for problematic species

Encapsulation systems based on Portland cement (PC) promote corrosion of reactive metals such as uranium, aluminium and magnesium used in the United Kingdom Magnox fuel design. Alternative encapsulation matrices based on non-ordinary Portland cement (OPC) are being pursued to treat historic legacy wastes within the United Kingdom's intermediate-level waste (ILW) inventory. Currently these wastes are encapsulated in composite OPC cement systems that incorporate high replacement with blast furnace slag of pulverised fuel ash. However, the high alkalinity of these cements can lead to high corrosion rates, with reactive metals found in some wastes releasing hydrogen and forming expansive corrosion products. Two alternative commercial products, calcium sulphoaluminate (CSA) and magnesium phosphate (MP) cement, which react with a different hydration chemistry and which may allow wastes containing these metals to be encapsulated with lower reactivity, are being studied. The results indicate that grouts can be formulated from both cements over a range of water contents and reactant ratios that have significantly improved fluidity in comparison to typical OPC cements. All designed mixes had set in 24 hours with zero bleed. The pH values in the plastic state were in the range 10-11 for CSA and 5-7 for MP cements. In addition, a marked reduction in aluminium corrosion rate has been observed in both types of cements compared to a composite OPC system. These results are encouraging in that both cement types can provide a possible alternative to OPC (NDA, 2010a; Hayes and Godfrey, 2007). The results have also confirmed compliance of this material against NDA RWMD guidelines for strength and expansion (Covill, et al., 2011; Montague, Vandeperre and Hayes, 2012). Recycled concrete aggregate is also being evaluated as an LLW and ILW encapsulant (NDA, 2011b; Butcher, 2011; INL, 2007).

- Silicone polymers as encapsulants

Silicone polymers are being studied as alternatives to grout for encapsulation of reactive metal wastes and other problematic materials, including ion exchange resins and some polymers. Cement will not stick to any surface that repels water, so the materials can be poorly encapsulated (Nathan, 2011).

Babcock FY2009/10 research on silicone polymers investigated their capacity to encapsulate nuclear waste. This project involved irradiation trials, drum trials, physical testing (e.g. tear resistance) and thermal testing (NDA, 2011b). Siloxanes, or silicone polymers with an inorganic backbone and organic side chains, offer a number of features to make them attractive as a waste encapsulant These include two added advantages over epoxy, in that they cure near room temperature (simplifying plant design), and long-term radiation degradation results in the gradual loss of the organic side chains as low molecular weight gases, ultimately leaving a waste form based on a silicate matrix, or quartz-like structure. This essentially creates a radiation-induced, low-temperature vitrification process (Black, n.d.). Other studies have found that adding iron as a reducing agent can reduce the mobility of chromium in silicone foams used as encapsulates. Thus additives to control ionic species in the waste material also warrant investigation (Miller and Duirk, 2011). The addition of iron rust formed under anoxic conditions has also been shown to immobilise neptunium (Nathan, 2011).

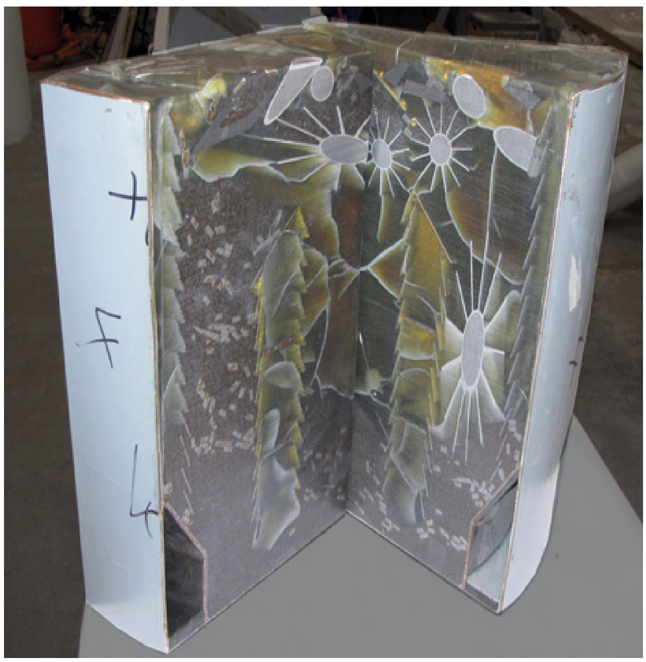
Physical tests have been carried out on siloxanes because there was a concern that the rubbery form might be too soft. However, this fear proved to be unfounded. A drum filled with sharp-edged bars and rods, encapsulated in siloxane rubber, was dropped from 15 metres and the bars stayed in the same position. The flexibility seems to be of benefit since cement encapsulants would be expected to crack when dropped from a similar height (Black, n.d.).

Much of the research has focused on assessing how the silicone and epoxy encapsulation materials will respond to radiation doses, which is complicated by the length of time the wastes must be kept safely encapsulated. Steven Black (n.d.) of Babcock Engineering consultancy stated, "Over the 10 000 years we need to keep the waste safe, we can expect a radiation dosage of 10 million grays…We've delivered that dosage over a short period of time and we've applied a smaller dose of radiation very slowly and compared that with an accelerated dose, and we get identical characteristics."

- Ability of epoxy resins to encapsulate wet wastes

A vinyl ester styrene is in use at a waste-treatment plant at Trawsfynydd in Wales to encapsulate ion exchange resins. However, styrene polymers use a volatile precursor material that is a fire risk with some applications. They are also difficult to formulate and if the waste contains any water, they may not cure effectively. To get around this, Babcock is looking at different polymers such as epoxy resins. This could be useful for one of the biggest single waste problems in the United Kingdom – the Windscale Piles (Nathan, 2011).

Figure 5.1: Wastes from the Windscale Pile 1 fire, often sharp-edged and metallic, could be stored safely by polymer encapsulation



Source: Nathan (2011).

Steven Black of Babcock Engineering consultancy stated:

The polymer is mixed with a powdered material that is itself chemically inert, such as silica sand, blast-furnace slag, and glass microballoons. They bulk out the polymer, which means there is less epoxy to react, and they are designed to conduct heat, so they knock the overall temperature right back and also prevent shrinkage on curing. We've also had good results with powdered graphite as a filler, which is a double win, as we have a large amount of graphite waste to dispose of. (Nathan, 2011)

The other consequence is that it cuts the cost of encapsulation. "When we buy epoxy at the half-tonne scale, it is about £20 per kilo, whereas Portland cement is £80 per tonne," Black added. "There is a massive cost difference with polymers of any type, but we have to use them where there is no alternative." (Nathan, 2011)

- Uranium co-ordination chemistry in magnesium-rich systems

Co-ordination chemistry is the study of compounds formed between metal ions and other neutral or negatively charged molecules. The study of uranium co-ordination chemistry in magnesium-rich systems is a United Kingdom PhD project that will complete a matrix of experiments to determine the quantities and the co-ordination chemistry of uranium adsorbed onto or co-precipitated with magnesium hydroxide and magnesium carbonate phases from aqueous solutions. This is an effort to better understand Magnox reactor spent fuel sludge chemistry. The research is intended to help answer questions concerning the co-ordination chemistry of uranium in fuel pond sludges and how the uranium will behave when sludge is removed (Veelen, 2011).

- Modular/mobile effluent retrieval and transfer plants

This is an item from the 2008/2009 NDA research portfolio (NDA, 2009c) which states that the application of a modular effluent treatment plant has the potential to accelerate decommissioning activities by decoupling abatement requirements from limitations imposed by existing infrastructure. This project investigates the functional requirements of a modular plant, the characteristics of key effluent types, and examples of both existing modular plants and current technologies that may be employed in a modular format. The development of these types of systems is important, since existing systems may be outdated or dysfunctional at the time of decommissioning if the facility has been in SAFSTOR or care and maintenance. The operation of the Enhanced Actinide Removal Plant (EARP) at Sellafield, used to remove actinides via flocculation, is an example of a modular waste treatment system deployed in the 1990s (Irons, n.d.). Another paper from 2011 describes various modular waste treatment systems developed for use in encapsulating and treating waste (Phillips, Houghton and Crawford, 2008). Modular treatment systems developed off-site and deployed to the Sellafield Ponds also played a key role in controlling radioactivity levels in the ponds to support clean-up activities (Calvin, 2011).

- Alternative waste package design

One of the most costly aspects of decommissioning a nuclear licensed site is the management and interim storage of radioactive waste until ultimate disposal. The construction of new waste treatment facilities or shielded stores is costly at a time of restricted budgets and expenditure constraints. Self-shielded packages are now becoming a popular concept, given the ongoing drive to find the most cost-efficient and effective means of interim storage. This generally assuming a storage period of up to 100 years for intermediate-level waste (ILW), in line with current regulatory guidance (Drake, n.d.).

The objectives of a robust self-shielded waste container/container system include:

- provision of a transportable and disposable waste container/container system that ensures sufficient radiation shielding and containment of contents, such that the resulting packages can be managed in the long-term without the need for additional shielding or physical protection;
- compatibility with the existing design for the Magnox interim storage facility (ISF);
- evidence that the waste container/container system can be integrated with systems and/or equipment for retrieval and conditioning of waste would be desirable;
- suitability for an interim storage period of up to 150 years in an ISF and subsequent disposal within the geological disposal facility (target lifetime of a waste package is 500 years);
- provision of an IP-2 and/or Type B transportable container/container system for Magnox ILW under the International Atomic Energy Agency (IAEA) transport regulations and licensing for multilateral transport within the United Kingdom;
- capability of the waste container/container system to be licensed for transport of fissile material will be required (GO, 2012).

Self-shielded packages, also known as “mini-stores”, offer a number of benefits: being suitable for interim storage in an unshielded building, allowing personnel entry, providing weather-proof cover, and requiring considerably less capital expenditure than building and storing waste in conventional remotely handled stores. There may still be a need for some amount of environmental control, but the self-shielded approach simplifies storage requirements and, by avoiding the need to build a shielded store, is said to provide greater flexibility in the decommissioning programme. However, the wide and expanding range of self-shielded options, with widely differing capacities, construction materials and costs, and each with their own advantages and disadvantages, makes selection of the optimum design challenging.

Available shielded package designs include 2 m or 4 m boxes, ductile cast iron containers (DCIC), TRU-Shield drums and WAGR boxes, as well as overpacking options such as the ModuCube. As a further option, an innovative approach by Babcock is driving a development of the TRU-Shield container model to meet United Kingdom requirements as a self-shielded waste package for interim storage and disposal. The design is based on the concept of a lead-lined stainless steel drum, with a lead thickness of 50-75 mm as required. With a 1.75 tonne container mass, and payload volume up to 305 litres, these containers are relatively small, handled easily and potentially suitable for transport on public roads. Waste can be encapsulated or un-encapsulated, and the containers would be particularly suitable for specific purposes such as smaller quantities of waste form; for example, a reprocessing, plutonium or fuel manufacture plant where fissile content is a concern. Outfitted with integral mixing paddles, the containers could also be used for liquid wastes. Waste could either be directly loaded into the TRU-shield or into a drum liner if later removal is required.

A different approach is the ModuCube mini-store, an “overpack” providing a shielded enclosure able to receive four unshielded 500 litre drums or one 3 m^3 box. This is not designed for use as a transport or final disposal package, its main benefit being to provide reusable shielding and interim storage of encapsulated waste in pre-existing approved disposal packages. Each option has its benefits and relative “best use”. While a single approach may be tempting to enable use of common infrastructure, in reality, there will always be a need for a range of designs to suit different needs (Drake, n.d.).

While ILW waste is being prepared and placed in the various packages described above there is still a great deal of research to be done to improve container designs, understand their long-term performance properties and to optimise placement of the various waste forms in the most suitable containers. This includes ensuring that transportation worthiness is factored into the designs for final waste packages (Abkowitz, Metlay and Mote, 2011). The long-term performance and potential failure mechanisms of the packages in the ILW store and geologic repository environments also need to be considered (King and Padovani, 2011; Serco, 2011b; Rebak, 2011; Janin, et al., 2011; Quintessa Ltd., 2011, Ghahari, et al., 2011).

R&D in this area should be closely aligned with relevant guidelines and objectives of the NDA and other national and international organisations developing interim storage and repository technologies (NDA, 2011a; Majhu, 2011; Bergström, Pers and Almén, 2011, Zuloaga, et al., 2011).

Future suggested R&D for management of problematic and mixed waste

- ***Description*** – Research and development on problematic and mixed wastes to advance the knowledge of waste attributes, and to develop and test improved technologies for waste collection, processing, sequestration, treatment and storage.
- ***Objectives –*** Research to better understand waste chemistry and environmental interactions with encapsulants and containers for problematic and mixed wastes. Develop improved treatment, neutralisation and sequestration technologies for problematic and mixed wastes, in conjunction with modular/mobile collection and

treatment systems and supporting systems. Continue long-term performance evaluation of waste package materials and integrate state-of-the-art knowledge of waste attributes, container material performance, ILW stores and geological repository design and waste handling into better waste packaging designs.

- ***Desired deliverables*** – Improved understanding of waste chemistry and chemical interactions; improved understanding and technologies for waste neutralisation and encapsulation; improved modular/mobile systems for collection, processing and treatment of problematic and mixed waste forms; improved, more versatile and reliable package designs that integrate with ILW stores and geologic repository designs and operations.

Treatment/removal of organic materials

Challenges

The working group identified the challenge of managing resins and other organic waste. As mentioned above, cement does not adhere to any surface that is hydrophobic and repels water. Organic waste materials, e.g. bead resins, can be poorly encapsulated using conventional Portland cement-based encapsulants. Polymer-based encapsulation technologies such as silicone and epoxy are being tested and developed as solutions to ensure the long-term stability of organic wastes (Nathan, 2011). In addition to encapsulation technologies, other technologies such as wet oxidation, pyrolysis and mineralisation are being developed to digest and remove the organic material or to alter its properties to make it non-hydrophobic. Wet oxidation, pyrolysis and vitrification processes have been in use since the 1990s in the nuclear industry primarily, to volume-reduce and destroy organic wastes such as resins, oils and sludges that contain organic constituents or chelating agents (IAEA, 2006a). These technologies are being widely tested and deployed at facility decommissionings in the United States and Europe. However, there is still extensive research to be performed on the various end product waste forms and their interactions with encapsulation media or the characteristics of the vitrified material. The extent to which these technologies can be adapted to process other forms of wastes with high levels of organic material, such as filter cartridges and dry active waste (DAW), has yet to be determined. Other technologies, such as the sorption and electrochemical destruction of oils and oily waste waters, hold promise for processing contaminated organic wastes.

Summary of current R&D for treatment/removal of organic materials

- Wet oxidation process to treat contaminated resins from PWR/BWR reactors

There are three main types of wet oxidation processes, based on temperature: cold wet oxidation, high-temperature oxidation and supercritical thermal oxidation (Katagiri, et al., 2011; Green, 2009). The cold wet oxidation process is a mild process that is conducted at ambient temperatures and used extensively in the pulp and paper industry. It uses soluble oxidants such as hydrogen peroxide combined with a catalyst to remove low molecular weight material, such as sugar acids and phenols. There is an emphasis on using oxygen-based oxidants to avoid problems with the formation of haloform compounds. This process only converts soluble organic compounds rather than totally oxidising all materials. Ion exchange resins have resistant polystyrene resin at ambient temperatures, so this process will only destroy chelating agents (Green, 2009).

The high-temperature wet oxidation process (WOX), a unique wet oxidation process operating at 100°C under atmospheric pressure, can be used for decomposing organic substances in wastes (Kim, et al., 2007). More resilient materials can be treated in this way, including aromatic compounds and some polymers. Carbonaceous material is converted to CO_2 or carbonates, but there may be further treatment required to treat the residual ash. An example of this process is used in the ModulOx process, developed by Lawrence Livermore Laboratories in the United States, which uses a different oxide-sodium persulphate (Green, 2009). This process is not totally effective at destroying all ion exchange resins and probably also needs an autoclave process (Green, 2009). Supercritical thermal oxidation involves the method above but uses temperatures above the critical temperature of water (374°C). There are problems with depressurising and waste disposal, so this process is operated as a batch process. This method has a high probability of destroying all organic material.

Treated wet oxidisation waste such as resins and filter sludges have a reduced volume and can then be encapsulated using cement (Sasaki, et al., 2011; Gunale, et al., 2009). The process also destroys chelating agents that pose a problem for long-term encapsulation and enhance the transport of radioactive species in the environment (Green, 2009).

A proprietary wet oxidation process has been developed to consume the organic material in liquid sludges produced by municipal waste water treatment plants. The process will be used to treat 100 m^3 of spent ion exchange waste from the operation of the Enrico Fermi nuclear power plant in Trino, near Turin, Italy. The process will reduce the volume of waste by 85%, help reduce its flammability and make it more chemically stable. Sogin, the Italian state decommissioning agency, developed and carried out the treatment with Ansaldo Nucleare and the Swiss firm Granit Technologies. The consortium began a two-year, EUR 13 million project in 2009 to apply a wet oxidation process to the spent nuclear resins. The spent resins represent almost a quarter of the total, current on-site waste that needs to be treated and conditioned. The resins are currently stored in 106 stainless steel vessels (about 1 m^3 capacity each) in a temporary storage area on site. The estimated total activity was about 26 TBq at the end of 2005 (NEI, 2009).

The WOX process was also evaluated for treatment of spent resins for the Tsuruga nuclear power station Unit 1, owned by the Japan Atomic Power Company (JAPC), which will terminate its commercial operation in 2016 (Sasaki, et al., 2011). The United Kingdom wet oxidation process used at Winfrith is the ModulOx process. The findings of ^{14}C in some resin streams lead to Winfrith's investigation into the likely impact of the ModulOx wet oxidation process on the levels of ^{14}C in spent resin. Results indicated that this wet oxidation process can be used to remove >98% of the ^{14}C present in the original waste, as well as destroying the chelating agents. The performance of WOX can be altered through manipulation of the catalyst, the contents of the catalyst and the volume of H_2O_2, with or without an electrochemical potential (Green, 2009).

- Detoxification process

Nanotechnology can also be used to remove organic chelating agents from nuclear waste. A recent study in Finland evaluated the degradation of simulated mixed organics commonly found in nuclear waste streams under a combined influence of sonication and magnetic field. Nanoscale bi-metallic iron-nickel was used as a source of Fenton reaction. The data were fitted to obey second-order kinetics. The extent of degradation was TBP-EDTA-citric acid, greater than TBP-EDTA, greater than TBP alone. The influence of the three variables that govern degradation behaviour, namely sonication energy, magnetic field and time, were evaluated with a response surface methodology. The model could predict the ratio of total organic carbon content to a maximum error of only 6% (Ambashta and Sillanpää, 2011).

- Mineralisation of organic radioactive materials

Fuji Electric developed a low-pressure oxygen process (LPOP) using plasma technology for mild decomposition and mineralisation of organic material such as ion exchange resin. This method is suitable for radioactive spent resin volume/weight reduction and stabilisation for final disposal. The ion exchange resins are vaporised and decomposed into gas phase with pyrolysis, and then are decomposed and oxidised with low-pressure plasma activity based on oxygen. The process is performed under moderate conditions with incineration temperatures ranging from 400-700°C, and using a low-pressure (low-temperature) inductively coupled plasma 10-50 Pa (Katagiri, et al., 2010). Similarly difficult organic constituents in radioactive wastes have also been digested using a photolytic advanced oxidation process (AOP), followed by biodegradation in the second stage (Makgato and Nkhalambayausi-Chirwa, 2010).

- Pyrolysis waste treatment

Pyrolysis using fluidised bed steam reforming (FBSR) provides a low-temperature (700-750°C), continuous method by which to process wastes that are high in organics, nitrates, sulphates/sulphides or other aqueous components (Jantzen and Crawford, 2010). The THOR® FBSR process has been shown in previous test programmes to effectively convert several types of liquid radioactive waste simulants into solid products. FBSR is being considered as a potential technology for immobilising a wide variety of radioactive wastes at the Hanford Site, the Idaho National Laboratory (INL) and the Savannah River Site (SRS). Waste liquids may be high in organics, nitrates/nitrites, halides and/or sulphates. They include LAW at DOE sites in the United States and other waste streams that may be generated by the advanced nuclear fuel cycle flow sheets that are being considered by the Global Nuclear Energy Partnership (GNEP) initiative (Qafoku, et al., 2010).

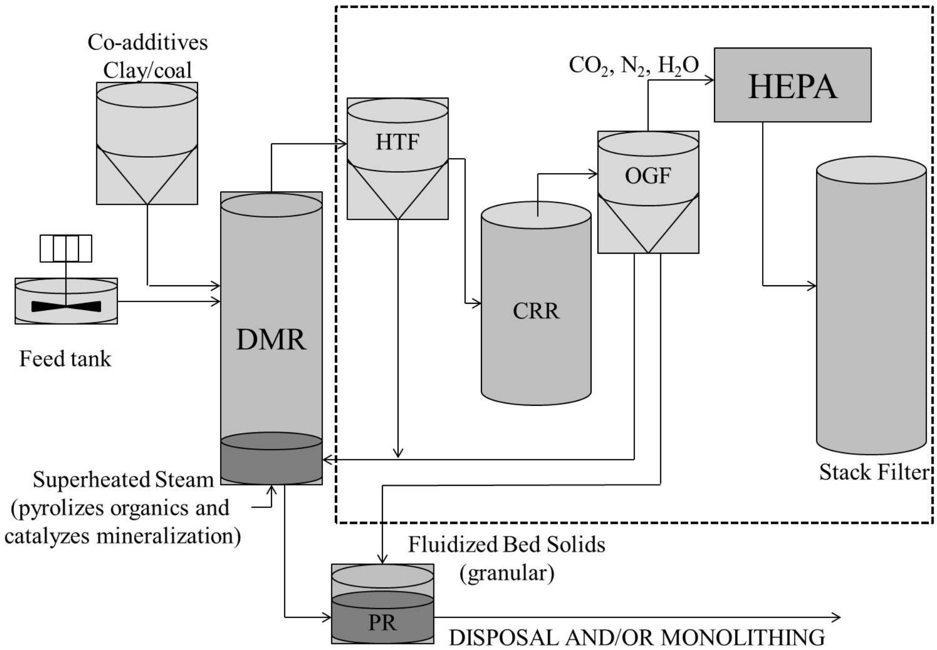
The objective of FBSR-related work has been to create geophases (minerals) that would provide leach-resistant (durable) waste forms for immobilising the contaminants that are present in different waste liquids. The FBSR mineral waste form is a granular product but can subsequently be encapsulated into a monolith for disposal if necessary. The mineral components of the waste form are primarily Na-Al-Si (sodium aluminosilicate, NAS), feldspathoid minerals with cage-like and ring structures, and iron-bearing spinel minerals. The cage- and ring-structured minerals atomically bond radionuclides like ^{99}Tc and ^{137}Cs and anions such as SO_4, I, F and Cl. The spinel minerals appear to stabilise hazardous waste species such as Cr and Ni (Qafoku, et al., 2010).

The primary end-product of pyrolysis/steam reforming is a granular flowable material of a highly inorganic nature, where the inorganic materials are mostly metal oxides and salts. THOR Treatment Technologies, LLC (TTT) and the Savannah River National Laboratory (SRNL) demonstrated the feasibility of converting the FBSR granular product encapsulated into a geopolymer matrix using an early low-activity waste (LAW) secondary waste stream composed of LAW off-gas treatment condensates that would normally be recycled within the WTP plant (Studsvik AB, 2011). Extensive characterisation work has been conducted on the fluidised bed steam reforming (FBSR) granular product for use at the United States DOE Hanford facility, including mineralogy, the product consistency test (PCT), the pressurised unsaturated flow (PUF) test and the single-pass flow-through test (SPFT). Work has been initiated to characterise the retention and release of radionuclides and constituents of concern, but much more work is needed (Qafoku, et al., 2010). Testing at the Savannah River Site indicated that for mineral waste forms, as in glass, the molecular structure controls contaminant release by establishing the distribution of ion exchange sites, hydrolysis sites and the access of water to those sites. The durability testing (product consistency test; ASTM C1285) of the FBSR mineral waste form has shown that the FBSR product is more durable than glass and that an Al-buffering mechanism controls the release of alkali (Na, K and Cs) elements and the solution pH controls the release of the other constituents like Re (simulant for ^{99}Tc), S and Si. This mechanism is known to occur in nature during weathering of aluminosilicate mineral analogues. Additional testing using the single-pass flow-through test indicates that the FBSR mineral product is more durable than a glass made from the same waste by ~2 orders of magnitude (Jantzen and Crawford, 2010).

Still, a recent report on Hanford's FBSR wastes concluded that only limited work has been conducted to characterise the FBSR waste form as suitable for its secondary wastes. A number of different binders, including cements and high-aluminium cements, geopolymers, hydroceramic cements and Ceramicrete have been evaluated at the laboratory scale for encapsulating the FBSR granular product to form a monolithic waste form. A geopolymer was selected for the most recent FBSR waste form characterisation but a final decision on the binder material has not been made. The FBSR waste form has been shown to pass the toxicity characteristic leaching procedure (TCLP) required to meet the dangerous waste limitations at Hanford's Integrated Disposal Facility (IDF). As with any waste form, if the concentrations of the constituents of concern are too high, the waste form will not pass TCLP. At the expected concentrations in the secondary wastes, the FBSR product will easily pass TCLP. The FBSR waste form monoliths pass the 500 psi compressive strength requirement. Candidate binders include cements, geopolymers and Ceramicrete. Table 1.2 of the PNNL-20704 report provides a summary of testing recommendations for containerised grout technology and waste form performance. Table 2.1 of the report provides a summary of the FBSR pilot-scale sodium aluminosilicate waste form preparation tests (Qafoku, N.P., et al., 2010).

Figure 5.2: FBSR sodium aluminosilicate (NAS) waste form dual processing flow sheet

DMR = denitration and mineralising reformer; PR = product receiver;
HTF = high temperature filter (material recycled to DMR);
CRR = carbon reduction reformer (treats gases only);
OGF = off-gas filter; HEPA = high-efficiency particulate air filter



Source: Neeway, et al. (2012).

In 1999, the Studsvik facility in Erwin, Tennessee demonstrated the ability to commercialise the FBSR process. The facility uses steam reforming based on a process known as THermal Organic Reduction (THOR®). The THOR® FBSR process is being used commercially to process liquid radioactive waste streams, including ion exchange resins, charcoal, graphite, sludge, oil and solvents that contain up to ~4.5×10^5 Sv/hr. Steam reforming thermally treats wastes at temperatures ranging from 625-750°C using a fluidised bed reformer. During mineralisation with superheated steam, organic matter is converted to carbon dioxide and steam, while nitrates and nitrites are reduced to nitrogen. The non-volatile solids in the residue are converted to water-insoluble, stable crystalline minerals that incorporate contaminants (Neeway, et al., 2012).

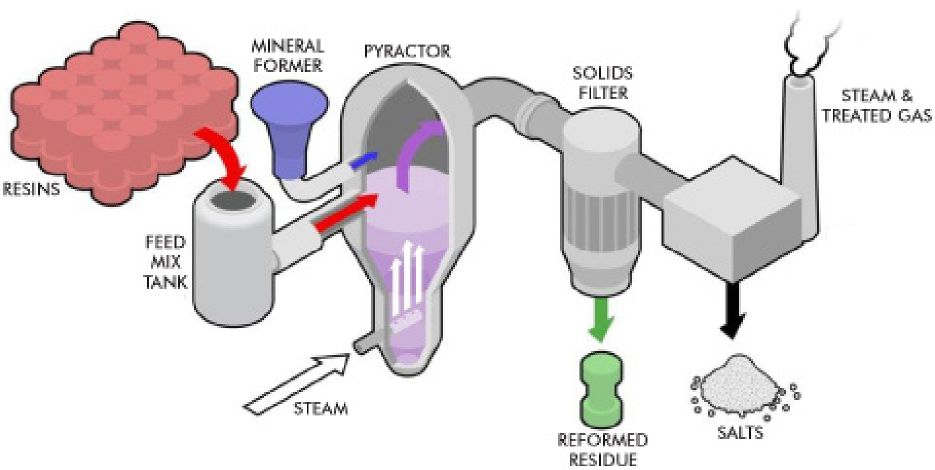
The Studsvik Erwin THOR® FBSR facility, in the joint venture SEMPRASAFE with EnergySolutions, is being used to volume-reduce resins of relatively low activity and high-activity ion exchange resins that until recently had no viable disposal path in the United States. The resultant blended granular product is acceptable for disposal as Class A waste at an NRC licensed landfill (Rogers and Stevens, 2012). THOR® reduces resin volumes by approximately 5:1 (Robinson, 2008; Studsvik AB, n.d.; Mason and Myers, 2010). Thus, depending on the encapsulation ratios, there is potential to conserve disposal volumes by reducing the overall volumes of the blended resins. The process also reduces the risks posed by undisposed high-activity resins by removing them from interim storage facilities across the country and placing them in licensed, engineered landfills in a stabilised condition. Cartridge filters have also been processed using the Erwin THOR® facility. Other organic waste forms, such as plastics associated with bulk dry active waste, may also be processed and sized to provide a flowable low-level activity feedstock that can be used to down-blend and dispose of higher activity waste in a safe and economical manner.

Studsvik, in collaboration with Westinghouse in Västerås, has built a pyrolysis facility mainly for burning operational waste (Studsvik AB, 2011). Studsvik's patented THOR® fluidised bed technology steam reforming ensures that organic material can be destroyed at relatively low temperatures without releasing harmful substances.

Studsvik/Kobelco is also considering development of a THOR® facility for Tokyo Electric Power Companies (TEPCO) to treat organic wastes from waste water at the troubled Fukushima Dai-ichi facility. Studsvik has a goal of building a THOR® process in Japan (Malm, 2012).

Figure 5.3: THOR® technology for processing spent ion exchange resins

SEMPRASAFE utilises Studsvik's THOR® process, using heat to reform spent resins into a reformed residue



Source: Rogers and Stevens (2012).

Pyrolysis is also being considered for treatment of Hanford liquid secondary wastes from sludge treatment processes. The insoluble sodium aluminosilicate mineral form is the preferred FBSR product for the Hanford tank wastes because the liquid secondary wastes will be converted to stable solid waste forms that will be disposed of in the Integrated Disposal Facility (IDF). Process additives such as magnetite are added to iron-bearing spinel minerals that sequester Cr and Ni in the waste (Qafoku, et al., 2010). The granular product is then encapsulated in a binder material to form a monolithic form to limit dispersability and to provide some structural integrity for subsidence prevention in the disposal facility. The FBSR process has been demonstrated at a pilot scale with non-radioactive simulants of Hanford Envelope C (AN-107) and Envelope A (salt cake) tank wastes, and with a simulant of the LAW melter off-gas submerged-bed scrubber liquid effluent (Qafoku, et al., 2010).

A bench-scale steam reformer (BSR) was designed and constructed at the Savannah River National Laboratory (SRNL) to treat actual radioactive wastes, confirm the findings of the non-radioactive FBSR pilot scale tests, and to qualify the waste form for applications at Hanford (Jantzen, et al., 2011). The process has also been demonstrated with the Idaho National Laboratory (INL) sodium-bearing waste (SBW) (Qafoku, et al., 2010). An FBSR facility is being designed and constructed at Idaho National Engineering and Environmental Laboratory (INEEL) for the treatment of SBW, which will be sent for disposal at the Waste Isolation Pilot Plant (WIPP) in Carlsbad, New Mexico. Another such facility is being considered for converting Savannah River Site (SRS) salt supernate waste (Tank 48), containing nitrates and caesium tetraphenyl borate (CsTPB) to carbonate or silicate minerals which are compatible with subsequent vitrification (Neeway, et al., 2012).

- Vitrification of ion exchange resins

A Studentship at the University of Sheffield received NDA funding for research on the vitrification of ion exchange resins in support of the HPA Wet ILW Retrieval & Processing Project, with industrial supervision by Magnox South Ltd (NDA, 2009c; Magnox, 2008). This is one of the first projects to investigate the vitrification of intermediate-level waste (ILW) in the United Kingdom. This process is being evaluated at the Hinkley Point Power Station by Sheffield's Imobilisation Science Laboratory (ISL). Legacy wet wastes, consisting of resins, sludges and filtration sands are targeted for a range of glass formulations that are suitable for vitrification (Matthews, 2010). Three different technology demonstrations have been completed to date. Bench-scale testing and ranking of the SLC/RWMD-specified criteria has four glass compositions optimised and the off-gas chemistry determined. Analysis of the four bounding waste permutations are being conducted on six glasses (Nuclear Research Centre, 2012).

Four waste mixtures or permutations are under consideration for volume reduction and immobilisation by vitrification. The inorganic fractions of several of the wastes are suitable for vitrification since they largely consist of SiO_2, MgO, Fe_2O_3, Na_2O, Al_2O_3 and CaO. However, difficulties may arise from the high organic and sulphurous contents of certain waste streams, particularly spent ion exchange (IEX) resins. IEX resin wastes may be the key factor in limiting waste loading, and possible thermal pre-treatments of IEX resin to decrease C and S contents prior to vitrification have been investigated. Results suggest that low-temperature (90°C) pre-treatment is more favourable than high-temperature (250, 450, 1 000°C) pre-treatment. A thorough desktop study has provided initial candidate glass compositions that have been down-selected on the basis of glass-forming ability, melting temperature, viscosity, liquid temperature, chemical durability and potential sulphate capacity. Early results for two of the candidate glass formulations indicate that formation of an amorphous product with at least 35 wt.% (dry waste) loading is achievable for HPA IEX resin wastes (Bingham, et al., 2008).

- Coating of ion exchange resin beads

As discussed previously, organic ion exchange materials may be encapsulated in cement; however, there are issues related to the reaction of hydrophobic organic resins to Portland cement and proper curing and long-term stability of the encapsulated waste. The major issues with the encapsulation of organic ion exchange materials are (Utton and Godfrey, 2010):

- The organic materials' reaction with the cement and the ion exchange of cement ions with waste ions, e.g. borate. This can cause retardation of the cement set.
- Resin expansion causing fracturing of the waste form and potential release of activity.

- Radiation instability of the resin, causing cement matrix expansion and break-up of the waste form.

Conventional physical pre-treatment methods used to minimise the effect of the resins on the hydration of cements include de-watering, drying, crushing and grinding. However, the resins re-expand on contact with water. Chemical treatments prior to cement encapsulation involve neutralising the resins to prevent unwanted ion exchange. For example, anionic resins may be treated with $Ca(NO_3)_2$ and cationic resins, with $Ca(OH)_2$ solutions to saturate sorption sites (Utton and Godfrey, 2010). Another technique employed is to pre-swell the resin with a sodium hydroxide solution. The long-term stability trial for this is ongoing. Alternative waste packaging solutions for ion exchange resin provides improved waste loading and improved waste form stability (Magnox, 2008). Research has also been conducted to develop novel cement systems to minimise the pre-treatment process and to make the cement more compatible with the ion exchange resins (Utton and Godfrey, 2010).

- Electrochemical destruction of contaminated oil and oil water mixes

Arvia® Technology Ltd has developed a waste treatment method, the Arvia Titan, for treating contaminated oils and oil-water liquids. The technology removes and destroys organic contaminants and oil, using a method that is free of process chemicals, is energy efficient and produces little solid or liquid waste for disposal (Arvia/Magnox, 2011).

Figure 5.4: The Arvia® Titan on site at Trawsfynydd

The Arvia Titan is a single unit, using a unique adsorption material called Nyex®. The key elements of the process with regard to oil destruction are as follows (Arvia/Magnox, 2011):

- *Adsorption* – A flow of compressed air is used to mix the Nyex® adsorbent with the emulsified oil waste in the reactor. This mixing is vigorous and leads to rapid adsorption of the emulsified oil onto the surface of the Nyex®.

- *Sedimentation* – The fluidising air is switched off and the dense Nyex® particles settle rapidly between the plates of an electrode bed, comprised of a series of graphite bipolar plate electrodes at the base of the reactor.
- *Electrochemical destruction* – A direct electric current is passed through the electrode bed across the conductive surface of the Nyex®, which destroys the oil through direct and indirect oxidation, creating water, carbon dioxide and hydrogen gas. This serves to regenerate the adsorbent surface of the Nyex® for reuse, and the cycle repeats. Since Nyex® is highly conductive, the power needed to destroy the pollutants is very low.

Future suggested R&D for treatment/removal of organic materials

- ***Description*** – Research and development on organic materials in decommissioning wastes, to advance the knowledge of waste attributes and to develop and test improved technologies for treatment and storage technologies.
- ***Objectives*** – Better understanding of organic waste and organic waste treatment residue chemistry and interactions with encapsulants. Improved organic waste oxidation and reduction technologies and as well as pre-treatment and methods.
- ***Desired deliverables*** – Improved understanding of organic waste, post-treatment residue chemistry and chemical interactions. Improved understanding and technologies for processing and stabilising organic wastes and residues.

Management of depleted uranium

Challenges

The world has large stockpiles of depleted uranium that require recycling, reuse or disposal. While conversion to depleted uranium oxides renders the material equivalent to natural uranium, conventional LLW disposal in shallow landfills is only suitable for arid regions where uranium and the daughter nuclides such as radium will not impact groundwater aquifers. Many countries do not possess such environmental conditions and so must develop means to encapsulate and sequester depleted uranium to minimise transport from the disposal facility. The depletion levels of the ^{235}U in depleted uranium vary, but over 85% of the material is comprised of ^{238}U. Long-term disposal is very challenging since a progeny of the principle radionuclide (^{238}U) is radon-222 (^{222}Rn), which is a highly mobile, unreactive noble gas.

Depleted uranium can also be viewed as a resource from which further extraction of fissile material can be achieved and that can be used in breeder reactors to generate more fissile material. Both approaches have been and are being used successfully and may play a future role in determining the overall fate of this material. The challenge is to evaluate the economics and likelihood of each eventuality and to improve the understandings/technologies to maintain all options as viable alternatives while global nuclear development shifts in response to technological developments, energy needs and socio-political dynamics.

Summary of current R&D for management of depleted uranium

- Encapsulating and conversions for uranium

Part of the NDA's 2009/10 research portfolio (NDA, 2010a) targets the development of encapsulants for the management of its managed uranics inventory (Bennett, Higgo and Wickham (2001). The inventory comprises a wide range of material types, including magnox-depleted uranium (MDU), thorp product uranium (TPU), uranium hexafluoride and uranium metal. This project involved a review of the issues associated with the encapsulation of uranics, identification of potential encapsulants and initial experimental work (Cook, Addington and Utley, 2011). Depleted uranium is produced during the manufacture of nuclear fuel and fuel reprocessing. In depleted uranium, the proportion of the fissile component (^{235}U) in the fuel is reduced compared to the non-fissile radionuclide, ^{238}U. By definition, depleted uranium contains ^{235}U with a concentration lower than the 0.711% occurring in nature. Like plutonium, depleted uranium is currently regarded as a resource with potential nuclear and industrial uses. However, should United Kingdom stocks be declared surplus to requirements, then approximately 100 000 tonnes of depleted uranium could require disposal. At present, depleted uranium arising from nuclear fuel manufacture is stored mainly in the form of uranium hexafluoride. The depleted uranium inventory includes that used in breeder reactors for plutonium production. Forms of depleted uranium that are potentially acceptable for long-term storage and disposal include uranium tetrafluoride (UF_4), uranium oxides (UO_2 and U_3O_8) and uranium metal.

A new plant in Ohio (United States) has been built to convert the depleted uranium hexafluoride (DUF_6) left over from uranium enrichment into uranium oxide. Meanwhile, the enrichment company Urenco has agreed to assist with the licensing and permitting process for a new deconversion plant in New Mexico (WNN, 2010; US NRC, 2009a). Up to 3 170 tonnes per year of LLW could be sent for disposal annually to this proposed facility (US NRC, 2012). The NRC states in its environmental impact statement for the facility (2011) that the resultant depleted uranium oxide is acceptable for disposal as LLW and there is enough existing national disposal capacity to accept the LLW generated by the proposed facility. The NRC has revised regulations to allow shallow land disposal of depleted uranium at facilities such as those in Texas and Utah where there is very little rainfall and depleted uranium in its oxide state resembles natural uranium in terms of solubility and specific activity. However, none of the forms are suitable for disposal in wet environments such as the United Kingdom; therefore, research on alternative disposal options or technologies is required (US NRC, 2011f, 2012).

- Further reprocessing for ^{235}U enrichment

The amount of uranium mined annually accounts for only two-thirds of the world's nuclear demand, with the shortfall made up by re-enriching depleted tails, repurposed military uranium and reprocessed nuclear fuel (Marvel and May, 2010). Enrichment raises the percentage of the fissile uranium isotope ^{235}U from its natural 0.71% to a reactor grade of up to 5%. The depleted uranium waste stream is typically about 0.3%. Since 1996, depleted uranium tails from Western Europe enrichers Urenco and Eurodif are being sent to Russia for re-enrichment (Diehl, 2004; IAEA, 2009g). In Russia, the imported tails are, instead of natural uranium, fed into surplus enrichment cascades. The product obtained from re-enrichment is mostly natural-equivalent uranium plus some reactor-grade low-enriched uranium (Diehl, 2004).

Similar reprocessing of tails is being carried out in the United States where USEC Inc. announced that it has entered into a multi-party arrangement with Energy Northwest, the Bonneville Power Administration (BPA), the Tennessee Valley Authority (TVA) and the DOE to extend uranium enrichment operations at the Paducah Gaseous Diffusion Plant in Kentucky. Under the agreements, the DOE will provide high-assay depleted uranium hexafluoride, also known as tails, to Energy Northwest. Energy Northwest has contracted with USEC to re-enrich the tails into low-enriched uranium. Energy Northwest will use a portion of the low-enriched uranium for its Columbia Nuclear Generating Station and will sell the remainder to TVA for use in its reactors. TVA will supply the power for the re-enrichment under an agreement to extend the existing USEC-TVA power contract (GAO, 2011; Nuclear Street, 2012b). Enrichment technologies such as separation of isotopes by laser excitation (SILEX) could render extraction of fissile ^{235}U from depleted uranium tails even more cost-effective and efficient, making the existing tails a valuable resource for underpinning future growth in nuclear capacity.

Nearly all current power reactors are of the "thermal neutron" design, and their capability to extract the potential energy in the uranium fuel is limited to less than 1% of that available. The remainder of the potential energy is left unused in the spent fuel and in the depleted uranium. The reuse of depleted uranium to produce fissile material in fast neutron breeder reactors (ANS, 2005) is another potential alternative to disposal in regions that lack sufficiently arid conditions to render shallow landfill disposal feasible (Tendall and Binder, 2011). This extends the energy recovered from the mined uranium 100-fold.

Fast reactors can convert ^{238}U into fissile material at rates faster than it is consumed, making it economically feasible to utilise ores with very low uranium concentrations. A suitable technology has already been proven on a small scale. Used fuel from thermal reactors and the depleted uranium from the enrichment process can be utilised in fast reactors, and that energy alone would be sufficient to supply the nation's needs for several hundred years (ANS, 2005). Fast neutron breeder reactors in conjunction with fuel reprocessing would not only eliminate the depleted uranium disposal issue but would also eliminate high-level waste disposal issues. Virtually all long-lived heavy elements, including the Am and Cu transuranic actinides, are eliminated during fast reactor operation, leaving a small amount of fission product waste that requires assured isolation from the environment for less than 500 years. This is a much simpler proposition than the design of geologic repositories that must maintain integrity of HLW for many hundreds of thousands of years.

Figure 5.5: Portsmouth (Ohio) depleted uranium cylinder storage yard

Source: US DOE.

GE-Hitachi Nuclear Energy (GEH) has proposed the construction of a nuclear power plant comprising two Prism fast reactors at Sellafield to assist the United Kingdom in disposing of its reactor-grade plutonium stockpile. This fuel would consist of a mix of plutonium metal and depleted uranium (WNN, 2011). After 45-90 days of irradiation in the reactor fuel, GEH said that the fuel would be brought up to a "spent fuel standard" of radioactivity, after which it could be stored in air-cooled silos. It would then be suitable for disposal alongside the United Kingdom's other high-level forms of radioactive waste. The entire United Kingdom plutonium stockpile could be irradiated in such a plant within five years, during which time the plant could also generate electricity from fission of the additional ^{239}Pu created from the ^{238}U in the depleted uranium. Once all of the stockpile has been irradiated, the Prism plant could then start reusing the fuel solely for electricity generation. With fuel staying in the reactor for about six years, and one-third removed every two years, the plant could operate for up to 60 years. Although used Prism fuel could be recycled after removal of fission products, GEH's proposal does not include a reprocessing plant at Sellafield (although one could be added later).

Future suggested R&D for management of depleted uranium

- ***Description*** – R&D is required to develop a greater understanding of potential impacts, uses and alternatives for depleted uranium disposal, reuse and recycling. This entails environmental impacts and considerations for disposal in LLW landfills, the means to sequester and contain depleted uranium and daughter radionuclides in non-arid regions, and proposals/practices for reuse and recycling of the material.
- ***Objectives*** – To obtain better environmental assessments and models of depleted uranium disposal impacts under various climatic conditions, and to develop encapsulation/containment strategies and technologies for depleted uranium disposals. The R&D effort should also evaluate continued reuse and recycling options, and integration with front-end supply and back-end reuse in strategies for nuclear infrastructures given advancements in separation and enrichment technologies, designs and potential fuel reprocessing without HLW.
- ***Desired deliverables*** – Models for evaluation of the long-term environmental impact of disposal of depleted uranium that accounts for the fate and transport of the material and daughter nuclides in the environment for shallow land disposal facilities in various climates and regions. Technologies for stabilisation and sequestration of depleted uranium and daughter radionuclides to facilitate disposal in non-arid regions. Better understanding, models and technologies for the reuse and recycling of depleted uranium in the existing and planned global nuclear infrastructure.

Improved segregation of waste

Challenges

The OECD/NEA working party identified the study of field deployable systems to sort and segregate decommissioning debris based on the waste acceptance criteria (WAC) of the disposal facility as a challenge for future decommissioning R&D. This includes technologies that implement the material sentencing processes to minimise waste volumes and maximise the unrestricted release, recycling and sentencing of materials such as VLLW. Effective segregation minimises the more costly and long-term disposal challenges for low-, intermediate- and high-level wastes. This is a major challenge for improving the efficiency and cost effectiveness of decommissioning in many countries with limited disposal options.

In addition, the segregation of long- and short-lived radionuclides from waste streams is a prime consideration to ensure sufficient capacity in high- and Intermediate-level repositories, and to minimise the challenges presented to the engineering of waste treatment and sequestration processes that must isolate wastes for time frames of hundreds to thousands of years. As noted above in the subsection entitled *Reprocessing for ^{235}U enrichment*, if fast breeder technology is developed, separation of long-lived constituents from waste and burning them in MOX fuel could greatly simplify disposal facility design.

Summary of current R&D for improved segregation of waste

- Monitoring on an industrial scale

The decommissioning of a facility can be viewed as an exercise in materials handling and sentencing. The decommissioning process involves the same principles regardless of the end state. Material must be removed, sentenced and disposed of or recycled and reused if a facility is to be brought to: *i)* Greenfield, where all SSC will be characterised, assayed and sentenced for material release or shipment as waste; *ii)* Brownfield, where some SSC will be characterised, assayed and released from regulatory control.

The proper implementation of segregation and sentencing requires multi-disciplinary, integrated decommissioning planning and execution. Every decommissioning activity has constraints. Schedule, budget, health and safety, radiological and environmental requirements, engineering requirements and personnel and equipment availability are examples of constraints that must be identified and factored into most decommissioning activities. Data gaps, such as the amounts and types of materials, contaminants present, re-recycling options, waste disposal requirements and equipment effectiveness must be identified in the segregation and sentencing planning process. Closure of data gaps by obtaining the required information must be completed to support efficient planning and execution of a project or activity.

Development of segregation and sentencing technologies must be integrated into the overall decommissioning process. Optimal design and use of these technologies will depend upon the quality of the generic planning for segregation and sentencing of the materials. Planning must include physical and contaminant characterisation of materials; including a physical inventory of material types (metal, concrete, soil, large components, miscellaneous components, etc.), quantities and forms. Segregation often begins in the deplanting process. Some components are part of an asset recovery programme that has targeted them for recycling or reuse. Characterisation must also identify the contaminants present (radiological and hazardous), their levels and distributions on each material type.

The deplanting plans for SSC to be removed must include segregation and sentencing requirements as constraints that have been addressed. The sentencing monitoring/assay methods, e.g. smear sampling, direct scans (e.g. frisking), *in situ* gamma spectroscopy, truck monitors, tool monitors and confirmatory sampling must be planned. The action levels and data quality objectives associated with each material sentencing option must be planned and understood (US NRC, 2009b). Segregation and sentencing may require preparation of the SSC for assay, sampling and monitoring, such as segregation by material type (e.g. cables, concrete, steel, soil), sizing to allow use of conveyors or box monitors and decontamination or surface treatments such as paint removal. The applicable disposal facility waste acceptance criteria (WAC) and transportation and packaging requirements must be understood. Development of broadly applicable, field deployable, automated segregation and sentencing equipment will require the above issues to be thought through and addressed for the decommissioning process in general.

Segregation and sentencing are currently labour-intensive processes that typically require extensive hands-on material handling and packaging as well as hands-on assay and sampling. With proper characterisation, sentencing and material handling planning, current technologies exist that are capable of automating the material sentencing and waste segregation process in an autonomous and semi-autonomous manner. As is the case with manufacturing and production, the benefits of automated robotic technologies are best realised for processes that have been standardised and properly sequenced. Meeting the sentencing and segregation challenges will require some assembly-line-like standardisation of the decommissioning material handling and removal processes.

Research is currently being conducted in Lithuania for the streamlining of waste management practices. This has included an emphasis on the progress in such practices and the completion of construction projects for a new solid radioactive waste management and storage facility, disposal facilities for very low, low and intermediate short-lived radioactive wastes, a new free-release measurement facility, and a new spent nuclear fuel interim storage facility (Poskas, et al., 2012). These characterisation, sentencing planning, and material sentencing and assay practices are illustrative of an integrated understanding and planning approach that lends itself to more automated handling and segregation of waste materials.

Another example of material characterisation, sentencing and handling strategies that lends itself to integration with automated systems is the decommissioning of the nuclear facilities at the Institute of Nuclear Energy Research (INER), which had been decommissioned and decontaminated successively over recent years. Since dismantling was a complex task, the main goal of waste minimisation required the set-up of procedures, criteria of free release, strict follow-up and traceability at all steps. This study gave an overview of the efforts on the non-destructive assay (NDA) of relatively large volumes of waste and the sampling of contaminated waste with radiochemical analysis to determine the radionuclide vectors (e.g. waste streams/fingerprints and scaling factors). Technical achievements in this research and experiences in free-release planning and measurement of very low-level radioactivity with high throughput for scrapped metal at INER could offer a reference for segregation and sentencing decision making (Wei, et al., 2009).

The robotic capabilities, mote characterisation and assay capabilities, and the geostatistical integration of data to understand contaminant levels and distributions mentioned previously in this report demonstrate that the tool kit exists to develop more automated screening, material sentencing and handling systems. Autonomous and semi-autonomous robotic systems now have the capability to evaluate the items being sentenced and factor that into the assay and material disposition and segregation process. In addition, the array of cutting, sizing and material processing technologies that can be integrated into automated systems lends itself to the development of deplanting and processing of materials into forms that standardise geometries and facilitate automated assay and handling systems.

Autonomous and semi-autonomous robotic systems capable of object recognition for dismantling and demolition tasks outside the nuclear industry are already being considered (Cruz-Ramírez, et al., 2011). New robotic systems are going to play an essential role in future dismantling services for renewing office interiors in buildings. In dismantling tasks, robots are expected to be able to find and remove very small parts such as screws and bolts. Such recognition of small parts is difficult for robots. The article describes a vision-based hierarchical recognition applied to dismantling tasks where large structures are detected first, allowing the small parts attached to these structures to be detected more easily (Cruz-Ramírez, et al., 2011).

For example, after dismantling ceiling panels intended for reuse, it is necessary to remove the screws that once held these panels to the light gauge steel (LGS). Following the detection of the LGS, a robot arm with a stereo camera on its tip can track and compute a trajectory near that structure to detect the screws. The large structure is detected by using a process of line detection in 2-D; its 3-D pose is measured with the stereo camera. During the motion along the structure, the screws are detected through the application of a multi-template matching process to every captured image. This detection is followed by a support vector machine (SVM), which recognises a screw candidate with a high true positive rate and a low false positive one. These rates are improved with a temporal multi-image integration for tracking the screw candidates. In the experiment described by Cruz-Ramírez, et al. (2011), 10 actual screws distributed in 1.1 m increments along a linear segment on the LGS were successfully recognised with a few false positives and with a final computed 3-D position of 2 mm in average. The feasibility of the methodology was evaluated by experimentation under various lighting conditions in a realistic environment. The experimental results show that the method works well for application at an actual dismantling site (Cruz-Ramírez, et al., 2011). Thus robotic material recognition systems capable of identifying objects in waste streams for proper segregation and assay are already being developed for non-nuclear purposes.

A recent paper argued for an innovative project at a major nuclear facility advocating the design and application of robotic technology to address site-specific needs for safely removing major structures and cleaning and decontaminating areas of its processing plant. The paper found that robots are frequently the answer, but commercially available units may not be the suitable solution, so designing custom robotics may be the best answer. Firms considering approaches to handling hazardous clean-up conditions can look to the experience at Dounreay in Scotland for designing robotic answers for special-needs handling applications. This may be the first major hazardous site to design and employ custom robotic techniques to handle most clean-up tasks (Bloss, 2010). As noted above and throughout this report, robotic capabilities are rapidly evolving and a comprehensive review of robots available within and outside the nuclear industry should be conducted before embarking on a project designing and manufacturing another specialty robot.

Another paper published over a decade ago described an automated robotic work cell that was designed, tested and demonstrated to classify hazardous waste stream items with previously unknown characteristics. The object attributes being quantified were radiation signature, metal content and object orientation and volume. Multi-sensor information was used to make segregation decisions and to perform automatic grasping of objects. The work cell control programme used an off-line programming system as a server to perform both simulation control and actual hardware control of the work cell. The overall work cell layout, sensor specifications, work cell supervisory control, 2-D vision-based automate grasp planning and object classification algorithms are discussed (Holliday, et al., 1993).

Another recent paper described a glove box dismantling facility developed as a centralised decommissioning workshop to dismantle glove boxes and recover nuclear fuel material residuals from the glove boxes. The facility possessed one power manipulator arm and six master slave manipulator arms to remotely size-reduce contaminated glove boxes. This article describes the facility and introduces the size-reduction procedures. Data obtained from one of the glove box size-reduction activities, performed by both remote and manual methods, are analysed and a comparison of these two methods is discussed (Kitamura, Watahiki and Kashiro, 2011).

More universal guidelines and requirements for waste sentencing and acceptance criteria are also key elements required to enable capital expenditures and payback over multiple projects for development of more globally-applicable automated waste segregation and handling technologies. The varying waste classification schemes and release from regulatory control criteria among member countries inhibit the development of more universal segregation and sentencing systems with proper assay and detection threshold capabilities (Muchová and Eder, 2010a; Muchová, Eder and Villanueva, 2011; Wei, et al., 2009; Gunter, 2011).

More automated systems for separation and recovery of demolition commodities are being developed outside the industry, such as those for processing and recycling scrap cables. The dominant means of recovering the metal from cable scrap in developed countries is automated cable chopping. This process usually includes pre-sorting, cable chopping, granulation, screening and density separation. If a dry electrostatic system or wet separation (e.g. cyclones, tables) is used, the metal content may be reduced to less than 0.1%, which will consequently increase the value of the recovered plastic. In general, the overall metal recovery is around 94-99%. A less costly and just as environmentally sound process for material separation is cable stripping, but it is a process with much lower throughput. Cable stripping machines are also used in most developed countries by utilities, cable manufacturers, cable chopping companies and metal scrap dealers. The advantage of stripping, in contrast to chopping, is the purity of the recovered jacketing and insulation materials. They are completely free of conducting metal and, if the user is careful in segregating the cable scrap before it is processed, the tailings can consist of one type of polymer. This way, the tailings, both metal and polymer, become more easily recyclable (Muchová, Eder and Villanueva, 2011). Merger of these sizing and separation technologies with robotic recognition assay and segregation capabilities is an example of advances that have yet to be developed and deployed for facility decommissioning.

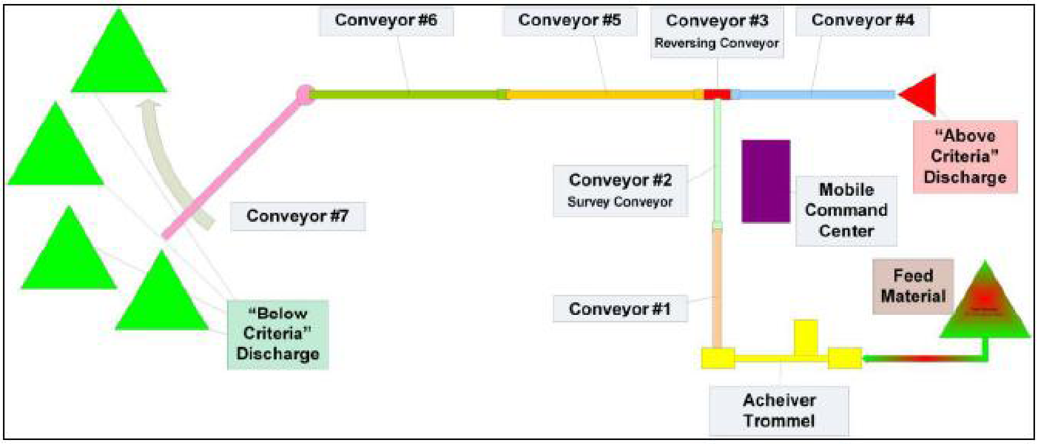
Perhaps one of the best glimpses into the future and example of an integration and deployment of automated assay, screening and segregation for a decommissioning project is the soil screening and segregation methodology and technology brought to bear in decommissioning the Plum Brook Reactor Facility in Ohio. Because large volumes of soil were expected from site remediation, NASA requested that the decommissioning contractor provide an automated soil survey sorter system for the final status survey (FSS) of excavated soil. Mactec Development Corp. was contracted to provide this service (ANS, 2010b; Mann, 2012).

The material was delivered from the feed stockpiles by heavy equipment to a screener for removal of large debris (> 4 in.). Material was passed through the screener and fed into a rotating drum-trammel that further screened the feed material (with a 2.5 in. screen) and delivered a steady stream to the first conveyor section. Material rejected by screening equipment was staged in stockpiles designated for disposal as radwaste (ANS, 2010b; Mann, 2012).

Screened feed material proceeded to a second conveyor where it passed under the detector system. This assembly consisted of two large NaI gamma scintillation detectors ($4 \times 4 \times 16$ in.) coupled to a gamma spectroscopy system. The soil passing beneath was scanned by the detectors that were calibrated to measure the ^{137}Cs activity concentration in the soil. This in effect implemented the FSS plan's scan survey requirement for the soil (ANS, 2010b; Mann, 2012).

Conveyor No. 3, a reversing conveyor, was located immediately downstream of the detection conveyor. It directed the flow of material passing beneath the detectors to the appropriate discharge conveyor. The reversing conveyor is shown in Exhibit 12 of Appendix A of the report. The diversion control set point (DCS), was set at a count rate equivalent to 5.2 pCi/g, one-half of the DCGL (10.3 pCi/g, surrogate DCGL for soils where ^{137}Cs predominates). Soil determined to be below the DCS was directed by Conveyor 3 to the clean discharge path via Conveyors 5 and 6 to stacking Conveyor No. 7. The clean discharge stacking conveyor is shown in Exhibit 13 of Appendix A the report, showing the system delivering soil to the clean discharge stockpile area (ANS, 2010b; Mann, 2012).

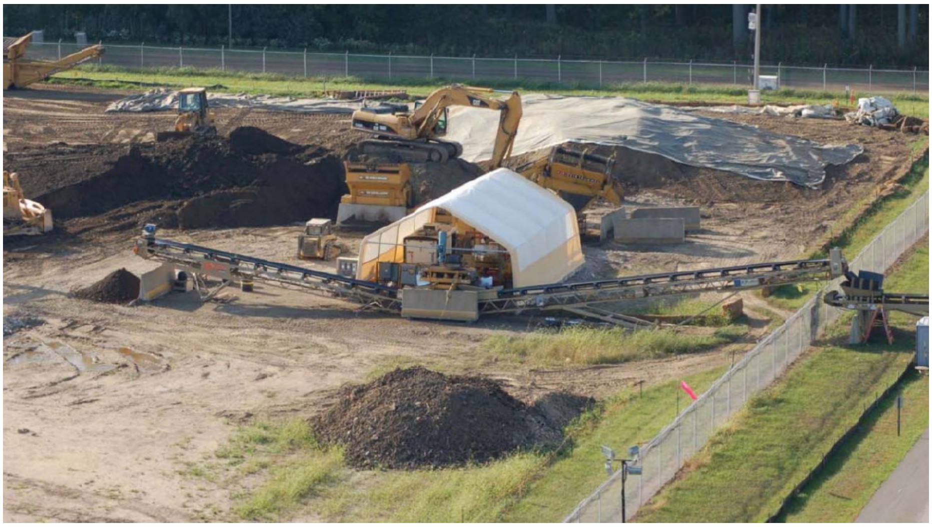
Figure 5.6: Schematic of Mactec Orion ScanSort Conveyor



When soil that exceeded the DCS was detected, the conveyor control system reversed the direction of Conveyor No. 3, the sorting conveyor, and the material was directed to Conveyor No. 4 for discharge to the "above criteria" soil pile. There the material was staged for radwaste shipment. The "above criteria" discharge extends from the left in the Exhibit 12 photo in the report (ANS, 2010b; Mann, 2012).

To meet the FSS plan requirement for collection and analysis of systematic samples from each soil survey unit, samples were collected from the ScanSort "clean discharge" conveyor. One litre grab samples were manually collected by a qualified FSS technician from the clean discharge conveyor at regular intervals to ensure that a minimum of 15 samples were collected per 500 tonnes of clean discharge soil. The clean discharge soil was delivered to individual 500-tonne piles by the stacking conveyor. The piles were identified as numbered batches, each equivalent to a Class 1 soil survey unit; 211 batches in all. The one-litre grab samples were analysed by gamma spectroscopy at the on-site counting laboratory (ANS, 2010b; Mann, 2012).

Figure 5.7: OrionScan sort layout at Plum Brook



Clean discharge piles were maintained with FSS isolation controls until all the FSS systematic sample concentrations were confirmed to be below applicable DCGL and verification surveys were performed on selected clean discharge piles. The verification survey of selected piles included *in situ* gamma scans and samples collected for gamma spectroscopy analysis by the on-site counting laboratory. After successful completion of the verification surveys, individual piles were moved to a large, clean material stockpile for use as backfill in excavated areas on the site (ANS, 2010b; Mann, 2012).

An additional detector system, called the auxiliary detector system, was added to the ScanSort conveyor system to meet the FSS plan requirement for replicate quality control scans of a minimum of 5% of scanned soil. This system, configured and calibrated to the same specifications as the primary detector set-up, was installed above Conveyor No. 6. It assayed all "below criteria" soil from batch 139 through 211, about 34 300 tonnes. The auxiliary system did not identify any soil above the diversion control set point of 5.2 pCi/g (ANS, 2010b; Mann, 2012).

The ScanSort system was operated from August 2009 to August 2010, with two pauses to allow for excavation to replenish feed material stockpiles. Altogether slightly over 97 000 tonnes of soil were processed by the ScanSort system, over 99% of the material assayed was below the 5.2 pCi/g DCS (ANS, 2010b; Mann, 2012).

Systems incorporating these types of principles must be developed for concrete, rebar, hangers, cables, pipes and system components to characterise, deplant, process, assay and segregate the limited variety of materials that are common to all decommissioning facilities. In order to make these systems cost effective and viable they need to be reused and improved at multiple decommissionings rather than fabricated and thrown away only to be redesigned and replicated for future projects.

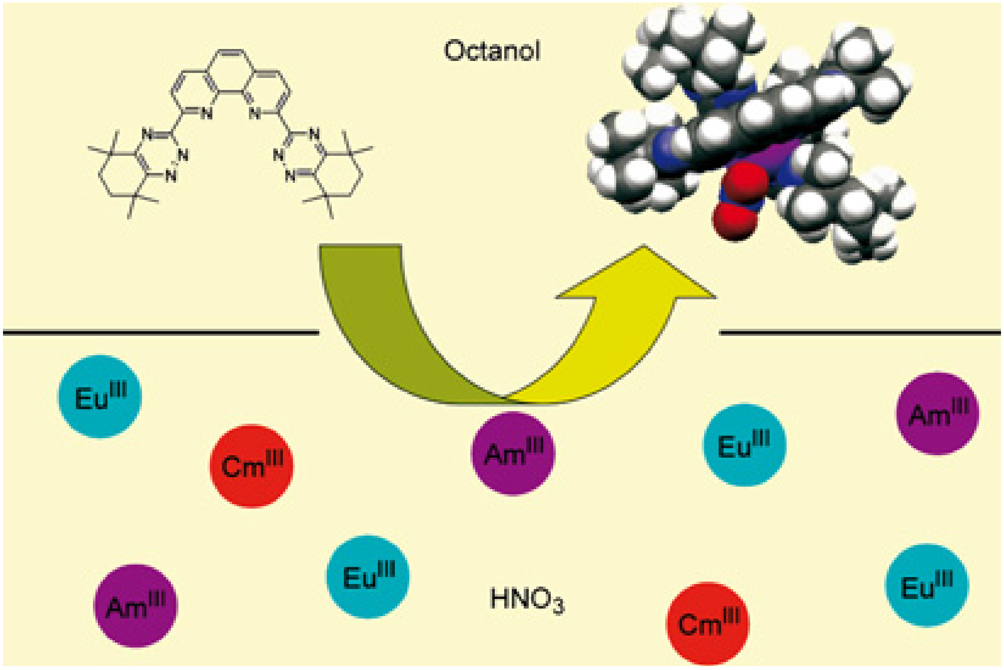
- Separate and transmutate transuranics from fuel reprocessing HLW and HAL

Transuranic actinide recycling by separation and transmutation is being considered worldwide and particularly in several European countries as one of the most promising strategies to reduce the inventory of radioactive waste and to optimise the use of natural resources. With its multidisciplinary consortium of 34 partners from 12 European countries plus Australia and Japan, the European Research Project ACSEPT (Actinide Recycling by Separation and Transmutation) is contributing to the development of this strategy by studying both hydrometallurgical and pyrochemical partitioning routes (Bourga, et al., 2009).

The PUREX process is used to chemically separate the dissolved spent fuel into uranium, plutonium and higher actinides with fission products. The final products are uranylnitrate, plutonium nitrate and high-level waste. The total capacity of commercial reprocessing facilities is currently about 4 500 tHM/year in France, the United Kingdom, Russia, Japan and India (Kessler, 2012). The uranium and plutonium products can be converted into oxides and fabricated into uranium/plutonium mixed-oxide fuel elements. The latter can be loaded into light water reactor or fast breeder reactor cores. Thorium/uranium fuel can be reprocessed using the THOREX process. The thorium/uranium-233 fuel also can be fabricated into mixed-oxide fuel elements and loaded into light water or fast breeder reactors. The remaining wastes are classified into high-, medium- and low-level waste (Kessler, 2012). Minor actinides americium and curium cannot currently be separated from contaminating lanthanides, which prevents them from being reused along with the plutonium and uranium. The actinides are responsible for the long-lived fractions in the high-level wastes requiring eventual disposal in geologic repositories.

The recent development of ligands capable of selectively binding actinides offers the possibility of integrating this separation technology into spent fuel reprocessing systems and eliminating the high-level waste resulting from current reprocessing systems (Lewis, et al., 2011; Lewis, Hudson and Harwood, 2011).

Figure 5.8: Harwood team's ligand for binding actinides



Source: Hadlington (2011).

The ligand binds exclusively to actinides, allowing them to be separated simply. The team that developed the ligand, which includes researchers from Belgium, France and Germany, modified an earlier ligand and achieved vastly improved selection. The original ligand was based on a backbone of four nitrogen-containing aromatic rings. This showed good selectivity, but was liable to attack by radioactively-generated radicals (Hadlington, 2011).

Harwood's team modified the molecule by applying atomic scaffolding to stiffen the structure into a rigid horseshoe shape, and replaced vulnerable benzylic hydrogens with more stable tetramethylcyclohexyl groups (Hadlington, 2011).

The effect of these modifications was startling. A separation ratio of around 20 to 1, which is 20-fold more actinides removed by the ligand than lanthanides, is considered good. The new ligand, 2,9-bis(1,2,4-triazin-3-yl)-1,10-phenanthroline, achieved a separation ratio ranging between 68 and 400 to 1 (Hadlington, 2011).

The team reported that the ligand meets all the major criteria required of it for use in a separations process. Its synthesis can be scaled up to the level of tonnes, it is relatively inexpensive to manufacture, it is resistant to acid hydrolysis and radiolysis, and it has the appropriate kinetics for selective extraction and stripping of actinides (Hadlington, 2011). It could be considered when rethinking more difficult extractions and separation approaches (Nichols, et al., 2011; Lewis, et al., 2012a, 2012b; Jung, et al., 2012). The separated actinides could be used to produce fissionable materials in breeder reactors and the lanthanides would have applications in medical technologies (Lewis, et al., 2012c; 2012d; Forsström, 2012).

- Opportunities to segregate short/long-lived intermediate-level wastes

Intermediate-level waste (ILW) contains higher amounts of radioactivity than low-level waste that is suitable for disposal at shallow land disposal facilities in many countries. Some ILW requires shielding due to these higher activity concentrations. ILW typically comprises resins, chemical sludges and metal fuel cladding, as well as contaminated materials from reactor decommissioning. Smaller items and any non-solids may be solidified in concrete or bitumen for disposal. It makes up some 7% of the volume and has 4% of the radioactivity of all radwaste. Intermediate-level waste can be subdivided into material that has a short half-lives (ILW-SL) and material that has a long half-lives (ILW-LL), with the latter category requiring more long-term storage and control capabilities.

It is widely accepted in member countries that radioactive waste of low and intermediate activity with limited concentration of long-lived radionuclides (ILW-SL) can be safely disposed of in near-surface facilities, either above or below ground levels. The safety of this concept is based on the performance of engineered and natural barriers combined with a period of institutional control of the repository. Waste that poses a potential risk in the long term due to its high activity or high content of long-lived nuclides should be disposed of in geological formations at depth, allowing for unrestricted use of the land after the closure of the disposal facility. Some of the subsurface repositories may accept a certain amount of such a waste type. With the exception of the Waste Isolation Pilot Plant (WIPP) facility in the United States, no other repository has thus far been designed exclusively for this purpose. When considering the wide spectrum of activities of long-lived radionuclides present in waste, however, this simplified division between near surface and deep geological destinations does not seem to be completely practical. Therefore, a number of member countries are considering alternative approaches to the disposal of non-heat-generating waste containing long-lived radionuclides, ranging from adapting facilities to designing specific ones.

Separation of radioactive waste into two categories, that which is acceptable for near surface disposal and that which should be directed to a geological repository, seems simple but complications can arise when defining the boundary between the two categories and, in particular, when deciding the most appropriate disposal route for wastes that contain relatively low concentrations of long-lived radionuclides. This waste category is generated within the nuclear fuel cycle, but can also arise in countries that do not operate nuclear power plants (NPP). Wastes of this type may not be large in volume or high in radioactivity, but their radiological, physical and chemical properties can make their management difficult (Carlsson, 2006). Some examples of ILW-LL are given below:

- waste generated during reactor operation;
- decommissioning waste;

- wastes from reprocessing plants;
- wastes from radiopharmaceutical production;
- wastes arising from accidents;
- used sealed sources;
- concentrates and waste from research laboratories.

Radiological properties are governed by the radionuclides present in the waste, and the risks induced by them decrease with elapsed time. Other properties depend on how the waste is processed and may also change with time. However, the potential impact of waste disposal on the environment depends on the combination of both groups of characteristics, together with the engineered and natural components of the disposal system. A number of radiological and non-radiological parameters can be used to characterise radioactive waste for disposal, but the following, which are significant in considering different disposal approaches and options, were selected and discussed in a recent study on the classification of waste (IAEA, 2009d):

- half-life and activity;
- radiotoxicity and chemotoxicity;
- waste amount;
- waste form properties and waste conditioning.

There is an international consensus that, for long-lived wastes, isolation is best achieved through geological disposal, the goal of which is to ensure passive protection of humans and the environment from the radiotoxic species that the waste may contain. Only geological disposal allows the possibility of isolating radioactive waste for a period of time that is sufficiently long to allow the radioactivity to decay to safe levels. Institutional control may include active measures such as monitoring, surveillance and facility maintenance. Institutional control may also include passive measures such as the placement of markers and restricted use of the affected land. In broad terms, the combination of engineered and natural barriers together with institutional control aims to provide adequate containment of the radionuclides and isolation of the waste. ILW-SL management has been solved in a number of countries through the construction and operation of near surface repositories, some of which are now closed and under institutional control. Questions remain, however, on the safe disposal of ILW-LL and high-level waste (HLW). It is generally recognised that spent nuclear fuel (SNF), HLW and high inventories of other ILW-LL should be disposed of at sufficient depths in stable geological formations. Some countries require that almost all radioactive waste be disposed of in geological formations, without regard to their radiochemical characteristics.

The existing Swedish repository for low- and intermediate-level waste (SFR) is licensed for disposal of short-lived waste originating from the operation and maintenance of Swedish nuclear power plants. The repository is foreseen to be extended to accommodate short-lived waste from the future decommissioning of nuclear power plants. Long-lived waste from operations, maintenance and eventual decommissioning will be stored before its disposal in a geological repository. This repository can be built either as a further extension of the SFR facility or as a separate repository (OECD/NEA, 2004).

Studies have been carried out on the removal of radioactive cobalt (^{60}Co) from synthetic intermediate-level waste (ILW) and decontamination waste using neat polyurethane (PU) foam as well as n-tributyl phosphate-polyurethane (TBP-PU) foam. The radioactive cobalt has been extracted on the PU foam as cobalt thiocyanate from the ILW. The maximum amount of cobalt removal has been observed when the concentration of thiocyanate in the solution is about 0.4 M. Cobalt can be separated from decontamination waste containing ethylenediaminetetraacetic acid (EDTA) and iron(II). The extent of cobalt extraction is slow and the separation of iron and cobalt is better with the neat PU foam compared to the TBP-PU foam. Column studies have been carried out in order to extend these studies to the plant scale. The capacities of the PU foams for cobalt have been determined. The effect of density and the surface area of PU foam have been investigated. Fourier transform infrared (FTIR) spectral studies have been conducted to discern the interaction between PU foam and cobalt thiocyanate species (Rao, et al., 1997).

Future suggested R&D for improved segregation of waste

- ***Description*** – R&D is required to develop modular, field deployable technologies and processes to size sort, assay and sentence decommissioning wastes. These include more automated processes for sorting and sentencing concrete rubble, metals and soils associated with decommissioning. In addition, a development of technologies to extract, sort and separate long-lived radionuclides from sludges, liquors and ion exchange resins could greatly reduce the engineering challenges and required capacities to safely dispose of very long-lived wastes. Separation of shorter-lived highly-activated wastes from transuranic-laden high-activity wastes can also change the strategy and requirements for disposals of intermediate-level wastes.
- ***Objectives*** – To explore assembly and use of existing technologies for separation and sorting waste streams in automated, field-deployable systems and to improve the assay and automated sentencing of wastes to alleviate manual sorting and assay constraints and gain higher efficiencies and outcomes to optimise the waste management hierarchy. The physical extraction of long-lived radionuclides from some waste streams is an objective that warrants further R&D. In addition, better strategies and technologies for identifying and separating intermediate-level wastes, with relatively short half-lives on the order of tens to hundreds of years, from those with half-lives in the thousands and tens of thousands of years should be a priority for future R&D in this area.
- ***Desired deliverables*** – Automated waste processing, segregation and assay systems that are modular and field-deployable to decommissionings, and the development of the subassemblies, components and instrumentation that can be integrated into such systems are desired outcomes for future R&D. Processes and equipment to extract long-lived actinides from transuranic waste streams and strategies and technologies to identify and separate ILW-LL and ILW-SL are also desired outcomes.

Clearance of low-level contaminated materials

Challenges

The OECD working group sited the treatment of contaminated metals by melting and new paths for recycling materials of low contamination levels as a common area of interest among member countries. Melting of metals for recycling decontaminates the product ingots for fission products, uranium and transuranics that are partitioned to the off-gas, slag and fly ash, with minor residual levels left in the product ingot. Continued pre-treatment, decontamination and separation processes are desirable, however, since metallic activation products such as cobalt and nickel remain in the recycled metal and the slag and fly ash will frequently contain heavy metal concentrations, such as lead and cadmium, which result in a mixed waste (e.g. hazardous waste with by-product radionuclides). This limits the potential to meet recycling concentration limits and creates issues repatriating the secondary waste to the countries of origin since such facilities are currently limited to the United States, Germany and Sweden. Also, there is currently limited potential for reuse of recycled metals that do not meet the European Commission concentration limits within the nuclear industry. Uses have thus far been limited to reuse for waste shipping containers and shielding.

Similarly, recycling and reuse of concrete aggregate is limited, with very few attempts at deploying efficient, industrial scale processes to separate concrete from rebar and then size, assay and sentence it. Technologies that separate Portland cement from the fill aggregate have been developed and are in use outside the nuclear industry. The recycling and regeneration of concrete is much more energy efficient than manufacturing new concrete. Still, the recycled concrete tends to be used as backfill, to create pads or as fill for container void spaces rather than as a resource for construction of the next generation of reactors.

Summary of current R&D for clearance of low-level contaminated materials

- Employing wastes as part of grout

The United Kingdom national inventory includes 4.4 million m^3 of raw LLW. Packaging increases the volume by a factor of 1.5 to 6.4 million m^3 (if disposed as LLWR). As seen in Figures 5.9 and 5.10, the overwhelming majority of the decommissioning waste is scrap metal, soils and rubble. The maximum vault capacity of the low-level waste repository (LLWR) located near the village of Drigg is only 1.7 million m^3. This United Kingdom repository can last until 2130, but only if the space is managed wisely (Rossiter, 2012; Bloodworth, 2012).

Figure 5.9: United Kingdom forecast of decommissioning LLW

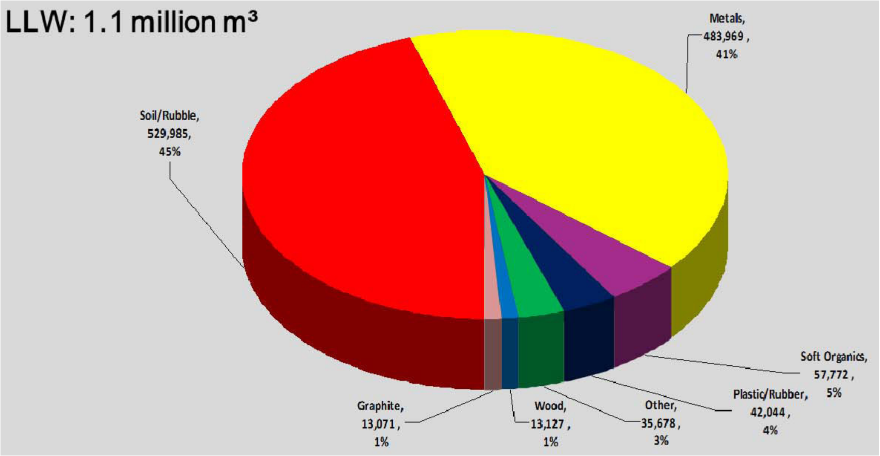
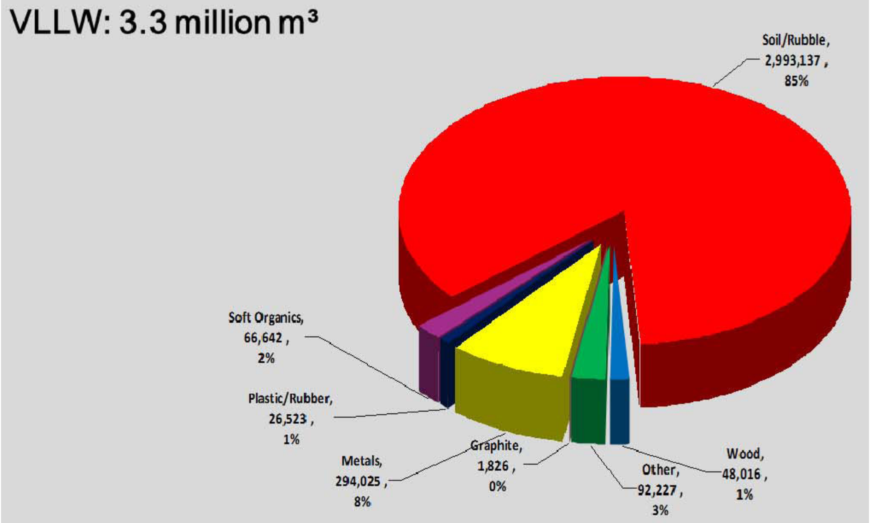


Figure 5.10: United Kingdom forecast of decommissioning VLLW



For many categories of ILW the most common method of conditioning has been the encapsulation of the waste with a cementitious (or occasionally polymer-based) grout within a stainless steel waste container. Historically, such an approach has resulted in waste packages that are compatible with the needs for interim storage, transport and disposal in a geological disposal facility (GDF) (NDA, 2009a). It has also been the general practice for the primary matrix of waste and encapsulating material (i.e. the waste form) to be supplemented by an additional layer of inactive cementitious material to provide a "grout cap" (NDA, 2009a). To this end, other waste materials (liquid or solid) can be blended into grout material to fill void spaces and minimise the overall waste volume. Container void spaces for fuel skips repackaged at Bradwell are being filled with contaminated desiccant grout and volume optimised with other waste streams (i.e. super compacted pucks) before being consigned to the LLWR for final disposal at Drigg (Sexton, 2011b). At Sellafield Industrial, recovery and beneficiation of fuel ash is being investigated as a means to guarantee the high specification supply the industry needs. Grout made from the beneficiated ash has to be tested for a number of product quality parameters, such as fluidity of grout, dimensional stability and strength (Butcher, 2011). The capping material must be stable and not undergo chemical or physical changes (such as carbonation or oxidation) that would unacceptably enhance the risk of releasing radioactive particulates, especially during interim storage and transport, or otherwise affect performance of the grout cap. Properties that make use of cementitious materials for capping desirable are that the materials are readily available, relatively inexpensive and not onerous to handle. Materials such as Portland cement (OPC) combined with either ground granulated blast furnace slag (BFS) or pulverised fly ash (PFA) are routinely used to provide suitable fluid capping grouts (NDA, 2009a). Other applications include the use of recovered radioactive concrete as fill for ILW (Butcher, 2011).

- Soil and concrete aggregate screening and clearance

Over the decommissioning lifetime of the Sellafield site alone, an estimated volume of 2 000 000 m^3 of waste concrete will arise. The bulk of this waste is likely to be clean/ exempt waste. There is a major effort in the United Kingdom to recycle and reuse both clean and contaminated concrete (Butcher, 2011). Concrete reuse has also included void space backfill and to create matt for ILW stores (Joyce, 2012).

An example of soil screening was discussed earlier, wherein soil was excavated, monitored and sorted on an industrial scale (Lombardo, Lopez and Lively, 2011). Similar methodologies and equipment can be used to monitor other materials such as aggregate concrete. Another example of screening technology is the methods used to survey for spent nuclear fuel fragments in reactor component burial trenches at Hanford in the United States (Bland, 2011). The aim was to identify and remediate the trench fragments more efficiently. Gamma spectroscopy instrumentation coupled with unique spectral analytical techniques was mounted on excavation equipment for real-time monitoring.

The Japanese have also been investigating the development of high-quality aggregate recovery technology with the objective of recovering coarse and fine aggregates from decommissioning concrete waste and meeting the Japan Architecture Society's standard (JASS5N) for a nuclear facility. The aggregate recovery rate can be increased to 70% or more to sentence more material to recycling, making it possible to recycle up to 500 000 tonnes of concrete wastes. The mechanical grinding method for the recycling of aggregate (a process of removing hydrated concrete adhering to raw aggregate by grinding it with a crusher) allows recycled coarse aggregate to meet the JASS5N standard. In the case of the selective heating method (a process for renewing aggregate by selectively heating cement paste with microwaves for crusher-based separation and removal), both coarse and fine aggregates meet JASS5N. The whole-heating method (a process of grinding with a crusher after whole-heating treatment to recover the aggregate) has also enabled both coarse and fine aggregates to satisfy JASS5N (Paajanen and Lehto, 1992).

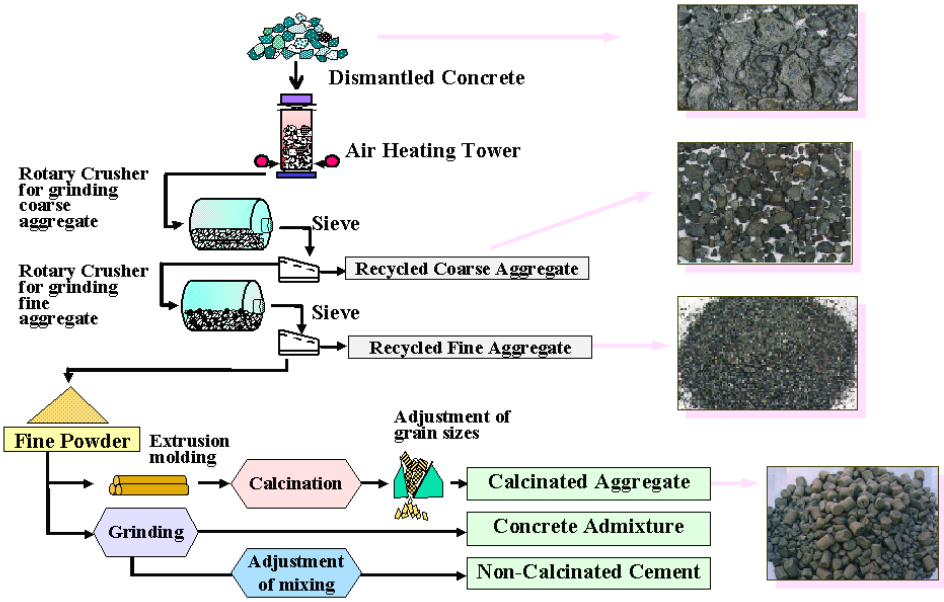
Another process for recovering the powder derived from the high-quality aggregate and which meets the JIS and JASS5N cement standards, is being developed to recycle concrete wastes. It was possible to manufacture Portland cement to JIS standards using a burnt cement manufacturing process. Japan's Recycling Technique for Radioactive Concrete Recycling technology is used to recycle radioactive concrete into solid material, mortar or waste form is carried out to establish the production technique for solidifying materials and for sludge filler waste form (Bloodworth, 2012; Kelly, Butcher and Adamson, 2011; Saishu, et al., 2000).

Figure 5.11: NaI PDA and electronics mounted on excavator boom

Figure 5.12: Steel plate welded in bucket creates uniform soil thickness for sodium iodide monitoring

Figure 5.13: Gamma detectors perform gross gamma screening of trench material

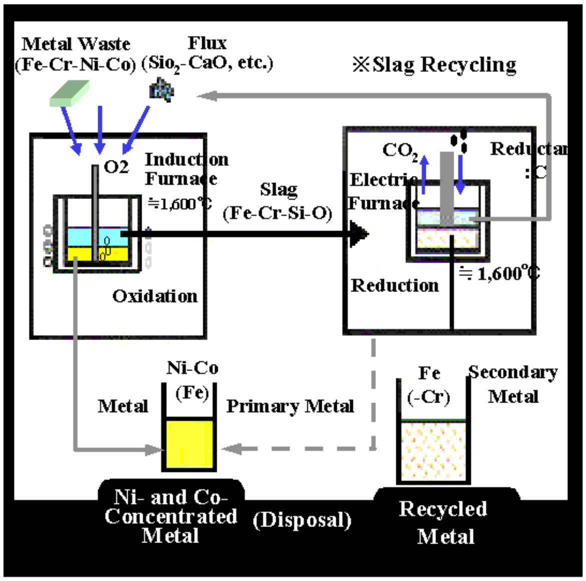
Figure 5.14: NUPEC concrete aggregate recycling

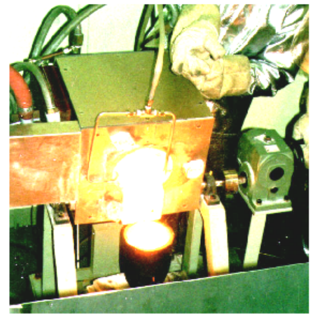


Japan is also developing a technology for scrap metal recycling called pyrometallurgical separation technology to expand the recycling range of metal decommissioning wastes. This process separates Ni and Co from metals, and can achieve a decontamination factor (DF) of 10 or more and a recyclable metal recovery rate of 60% or more. This is being developed for a target reduction of processing loads by some 3 000 tonnes of material.

An oxygen sparging method of separation, using selective oxidation based on oxidative potentials of Co and Ni, is reported to have achieved a DF of 100 or more for Ni in relation to stainless steel, and 10 or more for Co in relation to both carbon steel and stainless steel. This results in a recyclable metal recovery rate of 60% or more. Another molten metal casting technology uses low-level metal wastes by melting and filling packages with the molten metal in place of mortar, etc., as a volume reduction method. The metal filling rate has reached 95% or more in a simulated waste (Saishu, et al., 2000; Robinson, 2011).

Figure 5.15: NUPEC cobalt and nickel oxidative decontamination process





Removal of Recycled Metal (after secondary reduction)

Radiometric clearance verification equipment used in Europe may also be automated and scaled up to automate the clearance of materials. Verification of the clearance levels uses a statistical approach based on the measurements of a representative number of "material units" packed in suitable containers. The container measurements are performed by a commercial total gamma counting chain (TGMC) knowing the final nuclide vector (Klein, et al., 1999; Requejo, et al., 2012; Wörlen, 2011).

Figure 5.16: Total gamma counting chain (TGMC)

TGMC measurement station

Additional checking station

For every representative container the parameter Xn is calculated. When it is less than the unit it confirms the respect of the clearance level for the whole homogeneous group (Requejo, et al., 2012):

$$X_n = \sum_i \frac{a_{i,n}}{a_i^L}$$

where: a_i = the specific activity (for both mass and surface concentrations) of the nuclide *i;*

a_i^L = the corresponding clearance level;

n = the dimension of the representative container group.

For the released metal intended to be processed in a foundry, the foundry licensee will assure the 1/10 proportion mixture with metals that originated elsewhere (Requejo, et al., 2012).

Future suggested R&D for clearance of low-level contaminated materials

- ***Description*** – Research and development is required to develop technologies and processes to decontaminate and segregate radionuclides into manageable secondary waste streams and to process low-level waste into products that meet the quality standards that allow for recycling and reuse options. Technologies are also required to more efficiently sort, size, assay and sentence materials.
- ***Objectives*** – To explore the assembly and use of existing technologies for separation and sorting of low-level waste streams in automated, field-deployable systems and to improve the assay and automated sentencing of wastes to alleviate manual sorting and assay constraints. Decontamination and segregation of radionuclides into manageable secondary waste streams is also desirable.
- ***Desired deliverables*** – Automated low-level waste processing, segregation, sizing and assay systems that are modular and field-deployable to decommissioning and the development of the subassemblies, components and instrumentation that can be integrated into such systems are desired outcomes for future R&D. Processes and equipment to segregate radionuclides into manageable secondary waste streams and to remove non-radiological contaminants are also desirable; this will yield higher-grade end-products with more options for recycling and reuse within the nuclear industry.

New waste conditioning techniques

Challenges

The OECD working group cited the following issues as challenges for conditioning waste to ensure the long-term stability and retention of contaminants:

- evaluation of matrix characteristics (compression resistance, lixiviation sciviation factor, thermal cycle resistance, etc.) as relates to inclusion of chemical components (e.g. sulphates) and waste incorporation factor;
- studies for new conditioning matrixes (fill materials);
- management of graphite.

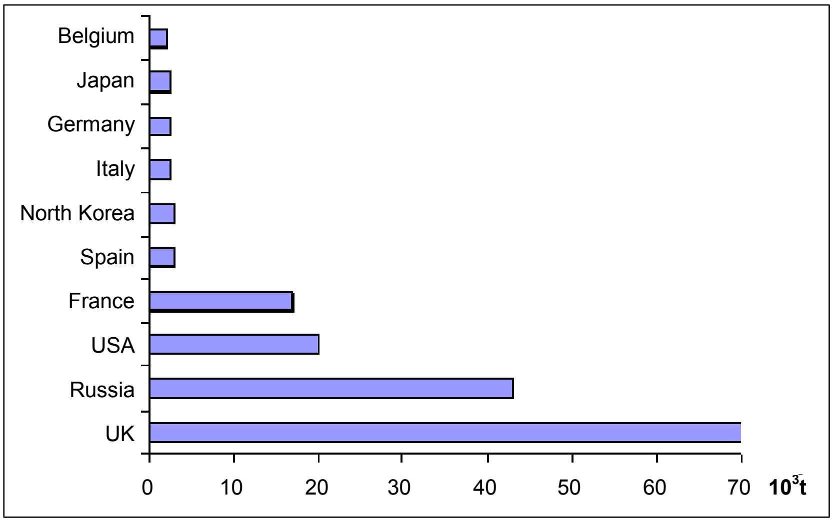
The extremely long time horizons over which intermediate-level and high-level wastes will remain hazardous and need to be sequestered from the environment make the design and testing of conditioning techniques such as encapsulation and container designs a challenge. In addition to the time horizons, reactions within the waste, with the containers and in repository conditions over protracted time frames are also important and require more R&D in order to enhance the certainty of the overall performance of the conditioned waste in final form.

Summary of current R&D for new waste conditioning techniques

- Wetting agents and dry/porous grouts for encapsulating graphite

Radioactive graphite waste from decommissioning activities on NDA sites will account for a large proportion of the total mass of unconditioned wastes destined for disposal. The United Kingdom has about 80 000 tonnes of irradiated moderator or reflector graphite materials from 47 reactors, principally from the Magnox or advanced gas-cooled reactors (ACR). This is a large portion of the world's graphite (Lee, 2006).

Figure 5.17: Metric tonnes of graphite waste



Source: Lee (2006).

Disposal is also problematic and several options exist, ranging from shallow land burial to deep geologic disposal. The current baseline strategy for reactor graphite wastes in the United Kingdom is to encapsulate them upon retrieval, using a cementitious grout, in stainless steel “Nirex” containers in accordance with NDA Radioactive Waste Management Directorate (RWMD) Letter of Compliance (LoC) specifications.

Following interim storage, the containers will be transferred to the United Kingdom’s planned geological disposal facility (GDF), when it becomes available (currently scheduled for around 2040) (Meehan, et al., 2011). The current baseline strategy is to use 3 m^3 Nirex boxes for operational wastes and 4 m^3 Nirex boxes for final decommissioning and site clearance (FD&SC) wastes (Meehan, et al., 2011).

The dismantling of the reactor graphite will generate graphite dust. The main issue with encapsulating graphite powder in cement is that it is hydrophobic and difficult to wet, and can therefore float on top of the cement grout (NDA, 2009c). Work on the immobilisation of graphite dust is still at the early stages of development (Wise, 1999; Rudisill, 1999). Several alternative methods are currently under consideration at the Windscale Pile 1 project. These include:

- encapsulation of small quantities of dust in mixing bowls in the dismantling cell, with the mixing bowls being disposed of in the final package;
- supercompaction of the dust in sacrificial cans, using a binder such as clay to provide a more stable product;
- encapsulation of the dust in some form of polymer acceptable to the Nirex repository (Wise, 1999).

Nirex requires waste forms to be fully immobilised in a cementitious monolith. However, the density of irradiated reactor graphite at 1.6 te/m^3 is less than the typical grout density of 1.8 te/m^3; hence, flotation of the graphite can occur during grouting and in certain cases, depending on the loading of the graphite in a package, could cause its steel box furniture to float. This would result in a waste form where the graphite was not fully immobilised by the grout. The flotation of the graphite can be overcome by the use of an anti-flotation device as part of the furniture inside the final package. This has to be designed to withstand the uplift forces caused by the graphite. Processing will have to ensure that the formation of dust from the graphite blocks is minimised and that grout capping effectively immobilises small amounts on the surface (Wise, 1999).

UKAEA has undertaken extensive studies to consider the best practicable options for disposing of these graphite liabilities in a safe manner while minimising the associated costs and technical risks. These options include, but are not limited to, disposal as low-level waste, incineration or encapsulation and disposal as intermediate-level waste. Nirex waste package specifications require that all measures shall be taken to ensure that the activity in waste is effectively immobilised and loose particulate material is minimised. Nirex requires graphite dust to be intimately immobilised to ensure that the package will perform in a predictable manner in an impact or fire accident. Intimate immobilisation would be achieved by the encapsulation of graphite dust in some form of matrix. This would normally be a cementitious matrix. However, the fact that the dust does not wet easily would require some form of wetting agent to get large quantities of dust to combine with the encapsulation grout. This wetting agent and its degradation products would have to be acceptable to the disposal environment. When only small quantities of dust are present, preliminary grouting trials at WAGR have demonstrated that the dust can be incorporated in the grout matrix. Another layer of inactive grout applied above the encapsulation grout as a cap would seal in any remaining graphite dust on the surface (Wise, 1999).

Research on graphite waste encapsulation at the University of Manchester consists of investigating the interaction of graphite waste and matrix. This includes studying the graphite's solid and powder microstructure using optical microscopy and scanning electron microscopy (SEM) to investigate the interaction of graphite waste and encapsulant. The mechanical properties of the encapsulated graphite, such as strength/compression, density and porosity, are being investigated to study the change in performance of irradiated graphite after encapsulation and the determination of water in encapsulation material after curing. Derived data is used to support calculations of change in total volume of waste after encapsulation (Hagos, 2010).

Long-term leaching tests under repository conditions are also being studied at the University of Manchester. These consist of two simulated groundwater environments, using demineralised water as the control, to quantify the leach rate of ^{3}H, ^{14}C and ^{36}Cl. Studies of encapsulation materials use a 3:1 mix of blast furnace slag (BFS) to ordinary Portland cement (OPC) or a 3:1 mix of pulverised fuel ash (PFA) to the ordinary Portland cement (OPC) (Hagos, 2010).

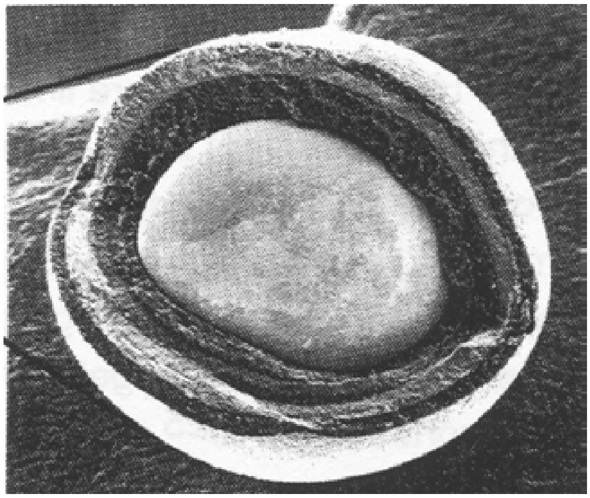
There is also work rendering irradiated graphite core material into silicon carbide (SiC) that can be used as fill-in grout material. This would provide both a disposal path and a waste form that does not require wetting agents. SiC is also a constituent in the PBMR refractory fuel, core and structure (Chavda, Ojovan and Zhang, 2011).

- Cementitious barriers and *in situ* technologies

The hyper-alkaline plume created by cementitious water associated with a geologic repository is diluted by groundwater and buffered by geologic materials so that down-plume conditions will approximate and in time equilibrate with ambient groundwater conditions. In addition to down-plume or down-field changes, it should also be recognised that the pH and ionic composition of cementitious water changes with time as the ageing of the concrete progresses (Berner, 1999). Waste disposal facilities provide a good framework for evaluating the formation and effects of cement derived hyper-alkaline plumes (Crossland and Vines, 2001; GTS, n.d.). Krupka and Serne (1998) state:

> *Cementitious materials have several important uses in low level waste (LLW) disposal facilities. These include the use of cement as a waste form (i.e. solidification) for LLW as well as its use as a backfill and construction material for the LLW storage vault. The formulations of these cements are expected to contain substantial amounts of Portland cement. Therefore, the long-term behavior of hydrated cements and their constituent phases in natural ground waters is important to the performance assessment (PA) of LLW disposal systems and the potential release of radionuclides.* (Krupka and Serne, 1998; Soler, et al., 2006; OECD/NEA, 2012a; Serco, 2012; SREL, 2011)

Figure 5.18: SiC/UO_2/graphite



Source: Lee (2006).

The Cementitious Barriers Partnership (CBP) works to improve understanding and prediction of the long-term structural, hydraulic and chemical behaviour of cementitious materials and waste forms used primarily in nuclear waste disposal. The individual CBP tasks all focus on development of computational tools, laboratory and field experimental data, test methods and guidance documents relative to cement barrier performance. The CBP is developing software modules that will be used as an integrated framework for better understanding and prediction of cementitious barrier performance (CBP, n.d.). Research is ongoing on 13 tasks aimed at developing and refining performance assessment software codes. This includes installation of engineered test beds. Reports are available on-line at the referenced URL (CBP, n.d.; Kosson, et al., 2008; FZJ, 2011; Serco, 2011c).

Encapsulation in cement is the favoured method in the United Kingdom for disposal of intermediate- and low-level radioactive wastes. It is typical to use composite cement systems incorporating blast furnace slag (BFS) or pulverised fuel ash (PFA) as these offer several advantages over Portland cement, notably a lower heat of hydration. The use of these mineral additions from a waste product that would itself need a disposal route is advantageous because the decreased amount of Portland cement used provides a reduction in cost and energy consumption (Sharp, et al., 2003; NRC, 2011).

Vitrification of some intermediate-level United Kingdom wastes is currently being investigated as a potential option for waste immobilisation and conditioning. It is being considered since it may offer the potential benefit of volume reduction as compared to immobilisation with cement grouts. It may also reduce the uncertainty associated with the long-term degradation of some wastes, such as organic materials. One option for the disposal of the resulting vitrified ILW product would be to place it in a geological disposal facility (GDF) in a high pH environment with cemented ILW and a cementitious backfill. However, at present there is a lack of information on vitrified waste-cement interactions, in particular the consequences of the high pH of cement pore solution. Hence, this report reviews the current understanding of the aqueous durability of vitrified and vitreous waste forms with a particular emphasis on the effects of highly alkaline conditions as an input to future optimisation studies for a GDF. Durability test methods are reviewed and, as most durability studies on glasses have been conducted on vitrified high-level waste (HLW), the durability of vitrified HLW along with other vitrified and vitreous wastes is examined in detail. The performance of glasses in cement and concrete is also reviewed. Overall, the literature suggests that highly alkaline conditions, such as those provided by a cement-based near field, could have an adverse effect on the performance of vitrified ILW (Serco, 2011a).

The *in situ* decommissioning (ISD) approach was used as the end state for the safe and cost-effective disposition of a structurally sound, robust nuclear facility. This approach involves filling all below-grade areas with flowable, self-levelling cementitious materials (grout) to entomb a massive nuclear building. The Savannah River Site (SRS) is implementing ISD at two reactor facilities by filling all subsurface areas with zero-bleed, flowable-fill, gravel grout No. 8 diutan gum (ZB-FF-8-D) grout and placing a water-resistant concrete slab over the filled area. This grout was designed by the Savannah River National Laboratory (SRNL) for uncongested dry areas in the reactor building. The 105-R reactor disassembly basin was filled with the aforementioned material. Once the grout is poured, heat is anticipated due to the hydration of the prozzolanic cements in the mix (Lima, et al., 2011; Lagos and Roelant, 2012; Monetti, 2012).

An *in situ* demolition technique of hot waxing pipes was used to allow open air demolition of drain lines highly contaminated with europium at the TRA-632 hot cells at the Advanced Test Reactor (ATR) Complex at the Idaho Nuclear Technology and Engineering Center (INTEC). The D&D labour force also successfully tackled removing three old nuclear reactors, digging up massive tanks and demolishing numerous large structures within a tightly confined area, all with no recordable injuries. Throughout the demanding process, the workers often were compelled to pioneer innovative solutions to the vexing, surprising problems that arose, taxing their resourcefulness. Among their ideas was injecting molten wax into the radioactive drain lines under the hot cells to encapsulate highly dispersible europium isotopes before the lines were extracted. This saved significant time and money by allowing open-air demo/removal work to be done (Mendiola, 2012).

- Securing the value from long-term monitoring of simulant ILW packages

Intermediate-level wastes (ILW) will require monitoring for hundreds of years to ensure that package integrity and waste forms are performing as planned and that radionuclides remain sequestered (Wilding, et al., 1991). In addition, those facilities in SAFSTOR or care and maintenance will require environmental monitoring and routine surveillances to ensure hazardous materials are contained and controlled. A total of 26 first-generation gas-cooled Magnox reactors were built in the United Kingdom in the 1950s and 1960s. In 1988 Berkeley was the first to shut down. In December 2010 its two reactors became the first to start a 60-year wait for core radioactivity to degrade before final decommissioning. This is the first such decommissioning milestone in the United Kingdom nuclear industry. Starting in 2074, the reactor vessels, heat exchangers and primary circuits of the reactors will be dismantled and the reactor buildings demolished, along with infrastructure built to facilitate these activities. Within nine years the final site clearance would be completed, the nuclear site license withdrawn and the land released for alternative use (NEI, 2011). SAFSTOR preparation at Berkeley first began in 1993 with the decommissioning of both reactors' eight primary gas circuits. This required the removal of the top and bottom gas ducts, sealing of reactor vessel inlets and outlet ducts, and removal of the boilers, gas circulators and motors. It also entailed the installation of new temperature and humidity monitoring instrumentation as one aspect of a planned long-term reactor condition and monitoring programme. Access will normally only be required every five years for basic monitoring and maintenance checks until final site clearance (NEI, 2011).

In 2015 Bradwell will become the first of the United Kingdom's Magnox nuclear stations to enter a care and maintenance (C&M) state. In doing so it will play a major part in demonstrating that the nuclear industry can manage its legacy safely and effectively. The acceleration preferred at Bradwell involves bringing the C&M state forward from the previously planned 2027 to 2015. In the United Kingdom context, when a plant/facility/installation is kept in a state of C&M, it is made safe for a planned period of quiescence in which the buildings are sealed and the waste is stored in a passively safe condition. Decommissioning activities will recommence after this period. C&M at Magnox sites begins when the only significant buildings on a site are the reactor buildings and an intermediate-level waste storage facility that will be removed at the dismantling stage. By the time Bradwell reaches C&M, all buildings on site except for the reactor buildings, interim storage facility for DCIC, ponds and capped building, and the security office will have been deplanted and demolished. The graphite cores of the reactors will remain intact, safely housed inside the vessels' solid concrete bioshield walls. The building will be completely clad to protect it from the elements and restrict any unauthorised entry. In short, by 2015 Bradwell will be in a passively safe state for the duration of care and maintenance (Sexton, 2011a).

In reaching this state, the site teams will have demonstrated the integrated application of a number of alternative technologies and the overall benefits that could be rolled out to other sites, and played a major part in demonstrating that Magnox Ltd, EnergySolutions, the NDA and the industry in general can manage its legacy safely and effectively (Sexton, 2011a). C&M will be followed, in Bradwell's case, by final site clearance between 2083 and 2092 (Sexton, 2011a).

A range of ILW materials such as sludge, gravel, resins, desiccants and other solid materials created from various processes during generation are also contained within Bradwell underground vaults. Other ILW may arise during decommissioning activities; they will be retrieved and treated, categorised and stored safely in ductile cast iron containers (DCIC, commonly known as yellow boxes), which provide the necessary radiation shielding. The DCIC each hold about 2 m^3 of waste and can be purchased as individual boxes, as needed. This innovative approach means that as waste minimisation techniques such as dissolution advance, the amount of storage space required can be adapted easily. This has the potential to avoid the significant capital costs associated with traditional ILW stores. Bradwell is modifying one of the redundant gas circulator halls to house the DCIC until a purpose-built weather protection facility is available (Sexton, 2011a).

ILW is stored for prolonged periods in the C&M phase but will eventually be transferred to the geologic repository. One paper proposes a system for monitoring nuclear waste stored in sealed underground repositories after a period of several decades. The focus of the work is on the selection and prototyping of a critical part of the system, the energy source. In order to select the energy source from a number of possible options, a trade-off study is implemented using a weighted decision matrix. From this trade-off study a system based on a magnetic spring to store energy is chosen, primarily due to its low number of parts and no self-degrading effects. When required, some of this stored energy is converted to the electrical domain and supplied to a wireless sensor node in a conditioned form. A prototype of the concept is presented and the supply of energy to a wireless sensor node is demonstrated (Constantinou, et al., 2011).

NDA research on waste package behaviour includes the continuation of targeted research programmes started in 2009/10, such as the following (NDA, 2010a; 2011b):

- "Smart" coupons for proactive waste package monitoring (NNL) – development of technical specification for a "Smart" coupon.
- Remote salt deposition measurement – laser-induced breakdown spectroscopy (LIBS) technology feasibility study; comprehensive assessment of the ability of LIBS to quantify the presence of salt on stainless steel for remote, in-store, waste package monitoring.
- Phased inter modulation (PIM) for the measurement of corrosion on waste container lifting features (Babcock); experimental investigation of the technique. This project will continue in FY2010/11.
- Inductively coupled technology for imbedded sensors (NNL); demonstration of technique using waste container filled with simulant waste.

- Telemetry systems; development of a generic design of a system for remote store monitoring and control.

Instrumentel has successfully completed a collaborative project with the National Nuclear Laboratory (NNL) to develop and demonstrate a battery-less wireless telemetry system capable of monitoring the contents of ILW waste containers. The system consists of a no-battery, wireless transponder sealed within a container that is connected to seven sensors monitoring strain, light and temperature from different regions. The sealed system is both powered and interrogated by an external reader through an industry standard, sintered metal filter, thereby maintaining the integrity of the container without the need for wire feed-throughs. Measurements can be taken from within the sealed container and transmitted to a safer environment, allowing continuous monitoring of the internal contents of the containers to be safely undertaken. A similar project monitoring concrete block performance is being conducted at the Savannah River's P reactor (Instrumentel, n.d., 2012; NDA, 2010a).

The *In Situ* Decommissioning (ISD) Sensor Network Test Bed at Savannah River National Laboratory (SRNL) was established to gain experience with installing sensors on massive, hardened concrete monoliths and using remote monitoring and communication systems to verify and validate sensor data collected on the concrete blocks' structural integrity. During SRS P Reactor decommissioning, large concrete blocks were removed from an exterior wall of the structure and a preliminary sensor system was designed and deployed for the structural monitoring (Lagos and Roelant, 2012; Zeigler, et al., 2011).

- Characterisation of hardened cements incorporating simulant ILW

For over 20 years, Magnox North and its predecessors have implemented a programme of work to establish the long-term stability of their ILW packages. This has involved preparing simulant ILW packages and monitoring their performance over time. The work has generic applicability across the nuclear industry, and is a significant component of the overall case to demonstrate that robust interim storage and disposal of ILW is sustainable. This project developed a database for the current samples across the full NDA estate, reviewed the processes associated with such a programme (e.g. protocol for adding new samples), and developed the business case for a possible national programme (NDA, 2010a). The NDA is also funding PhD research on ILW and the University of Leeds is funding PhD studentship on the characterisation of hardened cements incorporating simulant ILW (NDA, 2010a).

A range of magnesium hydroxide waste sludges arising from the reprocessing of nuclear fuel exists in the United Kingdom and requires safe long-term disposal. Similar wastes undergo a cementation process in order to immobilise radioactive material prior to disposal. Simulant magnesium hydroxide sludges were prepared, and their subsequent interactions with composite cement systems based on the partial replacement of ordinary Portland cement (OPC) with blast furnace slag and pulverised fuel ash were studied. This work concluded that there was little reaction between the sludge and any of the composite cements during hydration. Apart from a small quantity of a hydrotalcite-type phase containing magnesium from the sludge, the main phases detected were C-S-H and unreacted brucite. This indicates that the magnesium in the sludges is encapsulated by the cement rather than being immobilised or chemically bound within the hardened matrix (Collier and Milestone, 2010).

Radioactive iron (Fe^{3+}) hydroxide flocs are produced during the reprocessing of nuclear fuel at the United Kingdom's Sellafield facility. The flocs must be pre-treated with slaked lime before encapsulation in a pulverised fuel ash/ordinary Portland cement composite to produce a crack-free waste form. Results obtained after investigating the fate of the iron in the floc during cementation indicate that the iron in the floc reacts by substituting into and adsorbing onto the C-S-H phase formed during hydration. A small quantity is also substituted into a crystalline katoite phase, $Ca_3AlFe(SiO_4)(OH)_8$. These substitutions are significant because the iron in the floc is rendered chemically immobile within the cement rather than simply being physically encapsulated. No new phases are formed after 12 years of cement hydration and examination of the 12-year-old sample indicates that the durability of the waste form appears to be high (Collier, et al., 2009).

A sample of high-level radioactive tank waste was characterised to provide a basis for developing a waste simulant. The simulant is required for pilot-scale testing of pre-treatment processes in a non-radiological facility. The waste material examined was derived from the bismuth phosphate process, which was the first industrial process implemented to separate plutonium from irradiated nuclear fuel. The bismuth phosphate process sludge is a complex mixture rich in bismuth, iron, sodium, phosphorus, silicon and uranium. The form of phosphorus in this particular tank waste material is of specific importance because it is the primary component (other than water-soluble sodium salts) that must be removed from the HLW solids by pre-treatment. This work shows unequivocally that the phosphorus in this waste material is not present as bismuth phosphate. Rather, the phosphorus appears to be incorporated mostly into an amorphous iron(III) phosphate phase. The bismuth in the sludge solids is best described as $BiFeO_3$. The behaviour of phosphorus during caustic leaching of the bismuth phosphate process sludge solids is also discussed (Lumetta, et al., 2009).

- Superplasticisers

Organic superplasticisers are a special category of high-range water-reducing (HRWR) admixtures that have been used in the concrete industry over the last four decades. ASTM Int. (formerly the American Society for Testing and Materials) has specified a range of concrete admixture types. Within this range, superplasticisers are described as a specific, highly efficient class of water-reducing admixture that are either water-reducing (Type F) or water-reducing and water-retarding (Type G). As a result, they are designed to achieve the following functions in a concrete/grout formulation (Hayes, Angus and Garland, 2012):

- Permit large water reductions while maintaining fluidity.
- Impart extreme workability without increasing water content or promoting bleed liquor (Khatib and Mangat, 1999).
- Impart high early strength development, largely as a result of allowing lower water content mixes to be used. This can result in a reduction in both the permeability of concrete (thereby reducing chloride-ion penetration to limit corrosion of metallic wastes) and the cost of concrete construction (reduced cement contents).

An NDA report on the current status of superplastisisers released in 2012 (Hayes, Angus and Garland) is part of an ongoing programme of research conducted by the NDA and its contractors. It is a component of the research into the implementation of geological disposal for radioactive wastes in the United Kingdom. As a result of the potential issues raised by the long-term supply of current nuclear industry specification cement powders, a number of waste producers have considered the use of organic superplasticisers in encapsulation grout formulations such that British and European standard cement powders could potentially be used to meet the required grout performance criteria for the encapsulation of ILW. The report's aim is to collate the relevant information regarding the current state of development of superplasticisers for nuclear waste packaging applications and their use in general construction applications in order to provide a view as to their potential use in NDA-RWMD's illustrative concept designs for a geological disposal facility (GDF). The report also identifies the knowledge gaps required to satisfy its acceptance for incorporation in both this application and in the construction of the GDF (Hayes, Angus and Garland, 2012).

There are five generic classes of chemicals that are used as organic superplasticisers:

- sulphonated melamine-formaldehyde (SMF) condensate salts;
- sulphonated naphthalene-formaldehyde (SNF) condensate salts;
- modified lignosulphonates;

- vinyl co-polymers;
- aliphatic polycarboxylate polyether graft co-polymers (known as "comb polymers"; new generation superplasticisers) (BASF, 2010, n.d.).

The mode of action of the class of materials listed for the first four classes is essentially to act as surfactants in which fluidity enhancement is achieved by both: *i)* electrostatic repulsion effects between cement particles, thus preventing their agglomeration; *ii)* the increase in the solid-liquid affinity of cement particles, which tends to aid their dispersion. In addition, they act to disperse particles via steric hindrance effects. The combination of these factors brings about deglomeration of cement particles in solution, leading to a rapid dispersion of the individual cement grains and improvements in both fluidity and subsequent hydration. The polycarboxylate-based superplasticisers that began to be developed in the United States, Germany and Japan in the early 1980s are currently the only class of superplasticiser in general use (Hayes, Angus and Garland, 2012).

Since the current class of polycarboxylate ether (PCE)-based superplasticisers are likely to remain available, and cement powders are likely to remain similar to current standards during the time scales of proposed use in waste encapsulation grouts, an experimental assessment of the current class of PCE superplasticisers regarding radionuclide mobility could be considered to provide an understanding of:

- the impact of solubility enhancement to a GDF safety case;
- the nature of complexes formed with actinides in terms of potential competing species in a GDF environment, and the effect of various components within the PCE;
- the partitioning of PCE superplasticiser and minor components into the cement pore solution over time and its effect on GDF post-closure conditions of radiation and alkaline hydrolysis, and the extent to which they partition and their impact on radionuclide solubility;
- sorption mechanisms of the superplasticisers on cement/rock strata that may mitigate radionuclide release into the near field (Hayes, Angus and Garland, 2012).

The relevance of international research to the United Kingdom scenario is limited since this has focused on low-pH cements and on sulphate-resistant Portland cement (SRPC), although the use of SRPC would likely be appropriate in United Kingdom disposal scenarios and might also be desirable in encapsulation grouts. A number of programmes were carried out within the nuclear industry from the late 1980s to examine a variety of commercial melamine, naphthalene and vinyl co-polymer superplasticisers in a range of BFS/OPC grout formulations. These studies measured a wide range of fresh and hardened grout properties, including permeability and pore water chemistry assessment, in addition to a limited series of gamma irradiation trials. In general, the trials showed that all types of superplasticiser greatly enhanced fluidity and marginally reduced water/solids (w/s) ratios without significant modification of other fresh and hardened grout properties. Furthermore, the grouts tested were shown to be dimensionally stable with low gas generation "G" values recorded after exposure up to a 9 MGy total dose (Hayes, Angus and Garland, 2012).

The use of such admixtures may allow lower water content grout formulations to be used, with the potential advantage of reducing "bleed" liquor. The amount of water added to ensure sufficient fluidity is still in excess of that required for cement hydration and can therefore result in residual "bleed" liquor being expelled from the cured product and an increase in the porosity/permeability of the waste form. The resultant bleed liquor may require removal prior to waste package completion, leading to secondary waste generation, additional processing equipment and delays to the packaging process. In addition, the excess water within matrix pores may promote reactive metal corrosion over prolonged time scales and hence serve to reduce the long-term integrity of ILW products. In the context of waste encapsulation, there may also be disadvantages, such as a reduced rate of gas release, which could result in pressurisation and cracking of waste forms.

Early studies conducted in the United Kingdom and Japan found that commercially available superplasticisers enhanced the solubility of uranium, technetium, plutonium and americium by several orders of magnitude in free solution tests. However, further studies on cured BFS/OPC, PFA/OPC and Nirex Reference Vault Backfill (NRVB) grouts that had been cured for 28 days and incorporated 1 v/w% of an SNF superplasticiser showed plutonium concentrations close to the expected value for cement equilibrated solutions. The added superplasticiser appeared to have little effect on solubility. Superplasticisers were assessed in terms of their impact on the release of radionuclides out of the package and, in this respect, solubility and sorption mechanisms were the two key factors in terms of mitigating radionuclide release from the near-field.

The latest generation polycarboxylate ether graft co-polymer (PCE) superplasticisers became readily available by the year 2000. The PCE superplasticiser studied at this time was ADVA Cast 550, a commercial product supplied by Grace Construction Products, which had been shown to produce highly fluid BFS/OPC grouts. Initial complexation studies incorporating ADVA Cast 550 and model degradation products (i.e. succinic, glutaric and tricarballylic acids) in saturated calcium hydroxide solution to simulate cement pore water and backfill indicated that the solubility of nickel (II), europium (III) (as a surrogate for americium) and cerium (IV) (as a surrogate for Pu (IV) were not significantly enhanced by the presence of ADVA Cast 550 and model degradation products. However, it should also be noted that long-term hydrolysis of this backbone under cementitious near-field conditions would be expected to yield a high molecular weight polycarboxylic acid, which may be more strongly complexing than the original PMA. Thus, the fate of the superplasticiser in the cement microstructure and how it may partition and behave in the cement pore fluid, including the formation of degradation products, will be a major factor in assessing the long-term performance with respect to solubilising radionuclides under geological disposal facility near-field conditions (Hayes, Angus and Garland, 2012).

After initial studies, ADVA Cast 550 was replaced on the commercial market by ADVA Cast 551, a product described by Grace as being very similar. Therefore, recent studies carried out for NDA have investigated the benefits and feasibility of adopting superplasticised grout formulations based on ADVA Cast 551 in existing encapsulation plants. In 2009 NDA-RWMD commissioned work to assess the effect of ADVA Cast 551 on the solubility of plutonium (IV) and uranium (VI) in 0.01 mol dm^{-3} sodium hydroxide solution at pH 12 and near saturated calcium hydroxide solution at pH 12. The study showed that the solubility of plutonium and uranium were increased by up to four orders of magnitude due to the presence of ADVA Cast 551 at varying concentrations, with some evidence of colloid-like behaviour. Enhancement of uranium (VI) solubility was negligible at the lowest concentration studied (0.01% ADVA Cast 551) in near saturated calcium hydroxide solution. Further studies have indicated that the solubility of americium was also enhanced by four orders of magnitude and thorium by three orders of magnitude in saturated calcium hydroxide solutions containing 0.001% ADVA Cast 551 concentrations.

Gamma irradiation also reduced the solubility enhancement, presumably due to radiolytic degradation of the superplasticiser within the grout. There is uncertainty in applying the results of these trials, all undertaken in aqueous solutions, to the behaviour of superplasticised grout pore water in terms of enhancing the mobility of radionuclides from waste forms (or concrete construction facilities) (Hayes, Angus and Garland, 2012).

Previous studies have indicated that the leaching of several superplasticisers (including polycarboxlated ether co-polymer) from concrete is minimal, with the suggestion that the superplasticiser may reduce the total leaching behaviour by reducing the permeability of concrete. Further studies have also suggest that ADVA Cast 551 may sorb onto OPC; there is consequently the potential that if radionuclides bind to the superplasticiser in solution, the resultant complex may then also sorb onto cement surfaces and therefore act to inhibit the release of radionuclides into the near-field (Hayes, Angus and Garland, 2012).

Continued research provides an opportunity for a broader assessment of the impact of superplasticisers (and other organic additives) in a national geological disposal facility (GDF), including a range of construction applications as well as the proposed backfill grout. It is therefore anticipated that this type of "realistic" experiment, aimed at understanding the effect of actual cement pore solutions on actinide solubility over time, will be important for assessing superplasticiser behaviour in the GDF.

- Long-term suitability of epoxy resins

A number of wastes within the nuclear industry are not compatible with traditional cementation waste encapsulation processes. Polymeric encapsulation matrices offer solutions that can remove the undesirable effects associated with cement. A project in the United Kingdom began to investigate the long-term suitability of epoxy-based resins for encapsulating problematic wastes by reviewing epoxy resin chemistry and identifying future research requirements (NDA, 2009c; Nirex, 2005).

To date, organic polymers have not been widely employed in the immobilisation of radioactive wastes in the United Kingdom. Increasingly, however, waste packagers are considering the use of organic polymers and there are a number of ongoing research programmes aimed at investigating the properties/performance of specific materials and establishing their suitability for immobilising certain wastes, particularly for niche applications. A Guidance Note was issued to provide generic advice on the potential use of organic polymers as waste immobilisation materials in order to assist waste packagers in the development of alternative waste encapsulation strategies. This Guidance Note was intended to be the first of two such documents to consider the use of organic polymers for waste encapsulation. The principal aim of Part 1 of this document is to identify the types of organic polymers that have been employed and are proposed for the immobilisation of radioactive wastes throughout the world, and to provide information on them and discuss their relative merits and limitations with respect to their practicality of use, performance and properties. Part 2, a follow-up document, will examine the consequences of the use of such materials, in particular their evolution and degradation, in the latter stages of the Phased Geological Repository Concept (PGRC) (Nirex, 2005).

Nirex has investigated the degradation of alternative encapsulants such as polymers. Work has mainly focused on radiolytic degradation, which is clearly an important ageing mechanism for polymer systems (and arguably less so for cements). A comprehensive review of ageing mechanisms and identification of analogues for alternative systems in general is needed to bring the knowledge base up to a comparable level to that of cement. One of the potential attractions of alternatives such as polymers is their relatively simple composition, and the possibility of water-free compositions that would greatly reduce the interaction of the encapsulant with the waste (for example, negating water-driven corrosion of metallic waste components) (Environment Agency, 2008).

- Concrete as a barrier function in repositories

The study of cementitious materials is an area of intense and ongoing research and development to secure materials from release over extremely long time horizons in geologic repositories. Cementitious materials will be used in almost every programme for geological disposal of radioactive waste (OECD/NEA, 2009). They have been proposed for use as roadways and floors, tunnel linings, waste conditioning matrices, waste packages, overpacks, buffers, backfills, tunnel plugs and seals, and fracture grouts. After construction and use of the repositories, large quantities (e.g. several million kilograms) may remain in HLW and spent fuel repositories after closure. Even larger quantities of cementitious materials will remain in repositories for United Kingdom LLW and ILW disposal facilities due to the practice of grouting the containers and trenches. The disposal of long-lived radioactive wastes requires an understanding and assessment of the interactions of cementitious materials with other repository materials, host rocks and groundwaters over thousands of years (OECD/NEA, 2009).

The first permanent disposal facility for HLW to be built in the world is likely to be located in northern Europe. Sites at Forsmark in Sweden and Eurajoki in Finland have been selected. In Finland, site works commenced in preparation to apply for a construction license in 2012 and start operation in 2031. In Sweden, site works on the underground facility could commence in 2013, with full construction beginning in 2015, and operation by 2023. Finnish and Swedish HLW will be disposed of in crystalline bedrock at a depth of almost 500 metres (ANSTO, 2011).

Selecting a suitable design for all waste repositories will involve various modelling studies aimed at considering effects over longer time scales than can be experimentally accessed. Studies of natural systems, such as the cement-like rocks at Maquarin in Jordan, can provide useful information on the processes that occur in such systems. This type of information may help to build confidence that performance assessment models include the relevant effects and processes, and may also provide other qualitative safety arguments (OECD/NEA, 2012a). Potentially negative effects that have to be assessed and managed by using the safety case to inform disposal system design include (OECD/NEA, 2012a):

- the uses of different cementitious materials in various repository designs;
- the evolution of cementitious materials over long time scales (1 000s to 100 000s of years);
- the interaction of cementitious materials with surrounding components of the repository (e.g. waste, container, buffer, backfill, host rock).

An important concern regarding the use of cementitious materials in geologic repositories for HLW and spent fuel is their interaction with the bentonite buffer, backfill material and the host rock close to the repository near field. For this reason, the EC Project on Engineering Studies and Demonstrations of Repository Designs (ESDRED) has developed a low-pH < 11 concrete formulation as an alternative to standard ordinary Portland cement (OPC) concrete formulations, with the aim of reducing the interaction of the cementitious materials with the near-field components. Maintaining pore fluid pH < 11 is considered acceptable for preventing or reducing the alteration of the bentonite EBS. The development of the low-pH concrete involved laboratory work as well, as field testing at the Äspö and Grimsel underground research laboratories and at the Hagerbach tunnel site in Switzerland (OECD/NEA, 2012a). Similar to this interest in the interactions between bedrock and cementitious materials is the study of interactions with clay materials used for repositories and for lining and capping LLW landfills (Gaucher and Blanc, 2006).

Figure 5.19: Application of low-pH cement to seal rock fractures

Structural fatigue of low-pH cementitious materials is another area of research and concern. Microcracks were shown by a researcher at Lille University in France to initiate at the interface of the aggregate and the hardened cement paste. Experiments used aggregates in the 1-6 mm size range. Smaller aggregates resulted in higher desiccation strength and smaller damage densities, as well as a higher mechanical strength of the hardened cement-based material. Numerical modelling of fracture propagation was performed using the XFEM code, which uses a damage force criterion and is a code applied largely in mechanical engineering. The approach developed in the study can be used to evaluate the effects of desiccation on the degradation of cement-based materials in a ventilated repository for radioactive waste (OECD/NEA, 2012a). Experiments on water diffusivity and chloride transport for reinforced structural concretes are also being carried out (Zhao, 2012).

Based on these developments, thermodynamic modelling, coupled with kinetic equations that describe the dissolution of clinker (the paste constituent used in concrete) as a function of time, can be used to:

- quantify the liquid and solid phase compositions of ordinary Portland cement and blended cements during the hydration process;
- evaluate compositional changes that occur in cementitious materials due to the use of various aggregates and other mineral additives (e.g. silica fume and blast furnace slag);
- predict degradation of cement in contact with the repository environment.

Cementitious materials are used as chemical barriers for post-closure containment and retention of radionuclides within a disposal facility by imposing conditions that minimise radionuclide solubility and provide sites for radionuclide sorption. The mineralogy and other properties of cementitious materials that contribute to their physical and chemical barrier performance within the engineered barrier system, however, will evolve in geologic repositories post-closure due to several processes, including (OECD/NEA, 2012a):

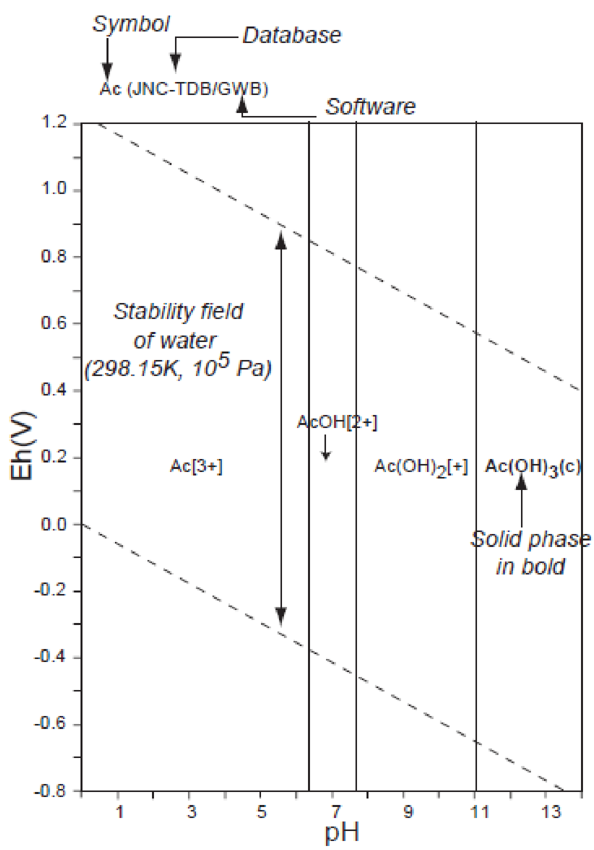
- leaching;
- reaction with groundwater solutes;
- hydration and crystallisation;
- reaction with wastes, their degradation products, and with non-cementitious waste forms;
- cracking.

Reaction of cementitious materials with groundwater will lead to changes in the mineralogical composition of the cements, accompanied by changes in porosity and permeability. Cracking can lead to localised water flow along the cracks and preferential leaching or deposition of reaction products. These processes can also alter the sorption properties of the cementitious materials. The heterogeneous distribution of cementitious backfill and waste in the repository result in additional complexities (OECD/NEA, 2012a). Predictive calculation models based on a discussion of the incongruent dissolution/ precipitation of the C-S-H system are being developed and refined to better understand the ageing process and the long-term performance implications. A coupled chemical equilibria-mass transport code for porous media (CCT-P), was developed to predict the alteration behaviour of cement materials (Sugiyama, 2012). Models such as these are normally coupled with various thermodynamic databases, such as FACT, HATCHES and the OECD/NEA *Chemical Thermodynamics* series 1-5 to predict elemental complexing and speciation under oxidative/reduction conditions (Eh) at various pH (Takeno, 2005). There is variation among the thermodynamic databases with regard to complexing and speciation behaviour. Taneko provides a comparison of Eh-pH diagrams (see Figure 5.20) from seven thermodynamic databases that is very useful for understanding complexing and speciation of contaminant elements (Takeno, 2005). Since thermodynamic modelling is constantly evolving, it may be time to update this atlas. Cement monolith tank leaching experiments have also been conducted to better understand the mechanisms of the ageing process and for refining the CCT-P code (Sugiyama, 2012).

At a research level, models of such interactions have improved considerably over recent years but, in contrast, safety assessments tend to represent interactions between cementitious materials and other repository components using rather simple and conservative approaches. These simple performance assessment representations may be easier to defend but still tend to give a pessimistic view of disposal system performance. In particular, some potentially positive effects of such interactions (e.g. clogging of fractures in the host rock as a result of precipitation of cement-related minerals) have been identified but are not included in safety assessment models (OECD/NEA, 2012a).

Rheology is the study of the flow of matter, primarily in the liquid state, but also as "soft solids" or solids under conditions in which they respond with plastic flow rather than deforming elastically in response to an applied force. It applies to substances with a complex molecular structure, such as muds, sludges, suspensions, polymers and other glass formers (e.g. silicates), as well as many foods and additives, bodily fluids and other biological materials. Rheology of cement-based materials is controlled by the interactions at the particle level. Research is being conducted to investigate particle interactions and rheological properties of cement-based materials at the micro- and macro-scales (Lomboy, 2012).

Figure 5.20: Legend of Taneko Eh-pH diagrams



The fracture properties of cement paste, mortar and concrete are highly related in nature. A lattice fracture model that captures detailed crack information with high computational efficiency and stability has been developed for a PhD thesis (Qian, 2012). It also enables investigation into the relationship between the material structure and the fracture properties of concrete. This can be achieved by projecting the lattice network on top of the original material structure of concrete. Research is being conducted on a parallel computing code using a parameter-passing, multi-scale modelling scheme which, when coupled to the lattice fracture model, enables study of the relationship of the fracture processes in cement paste, mortar and concrete for the model. This reduces computational time and enables the analysis on even larger lattice systems. A multi-scale fracture modelling procedure is proposed and demonstrated. Three levels are defined, including micrometre scale for cement paste, millimetre scale for mortar and centimetre scale for concrete. The lattice fracture model is applied at each scale respectively. The inputs required at a certain scale are obtained by the simulation at a lower scale. At the lowest scale in question, the micrometre scale for cement paste, the inputs are determined by laboratory experiments and/or nanoscale modelling from literature (Qian, 2012).

Future suggested R&D for new waste conditioning techniques

- ***Description*** – Research and development is required to understand and model the long-term performance of waste conditioning options. These include physical performance, chemical interaction, the sequestration of radionuclides in the waste for encapsulation of hydrophobic organic wastes that are not tightly bound to cementitious encapsulants, and the performance of cementitious systems.
- ***Objectives*** – To increase the fundamental understanding of physical properties, chemical reactions and retention of radionuclides in current conditioning options. The performance of simulated conditioned wastes under repository conditions also requires more research. The overall objective should be to develop waste conditioning materials and processes that have a high level of confidence in their long-term performance.
- ***Desired deliverables*** – Fundamental data on physical/chemical properties and performance of conditioned waste, improved understanding and modelling of waste performance under disposal conditions, and improved waste conditioning substrates and processes are the desired deliverables of this R&D effort.

Suggested areas of future collaboration

Despite some variability among the responses on R&D requirements and priorities from the countries' representatives, the working group considers that the following issues are of very high priority as they have high impact on costs or dose, and some could be issues for future collaboration:

- managing problematic wastes – chemical (PCB, asbestos, etc.) and mixed waste;
- treatment/removal (including mineralisation) of organic materials (bituminised waste, resins, oils, nitrates) and activated sodium;
- conditioning of waste (different grouts, foam concrete, etc.; improving waste incorporation);
- long-term performance of waste forms (e.g. concrete, impact of superplasticisers on radionuclide migration).

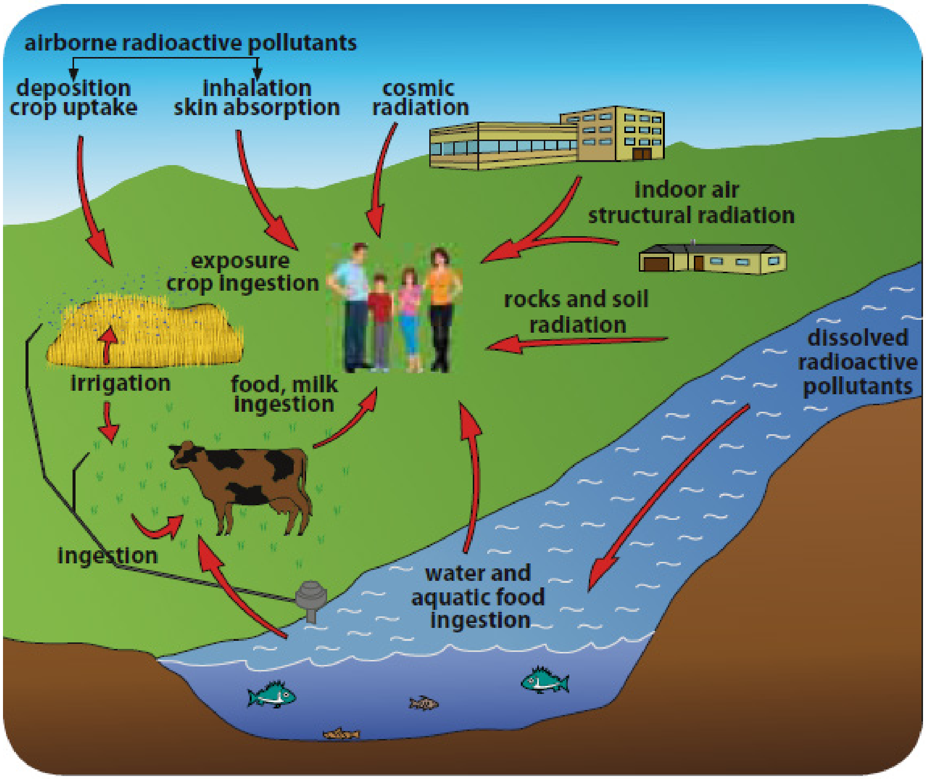
Two more issues ranked as high priority and were selected for possible collaborative R&D activities: *i)* treatment of reactive metals (high-temperature processes, melting) and managing gas generation; *ii)* clearance and recycling of low-contaminated materials. The remaining issues ranked at lower priorities or were not considered as issues warranting future R&D activities.

6. Site characterisation and environmental monitoring

Theme overview

This theme focuses on characterisation, modelling and clearance assays to support and verify conceptual site models and to demonstrate compliance with license termination criteria. This entails characterisation and assay to determine the radionuclide profiles present prior to final decommissioning activities, and using radionuclide fate and transport models to provide a risk-based evaluation of the significance of radionuclides remaining after delicensing. It is the final phase of the iterative characterisation process (discussed in Chapter 2) and is in many ways related to the R&D discussed regarding facility characterisation prior to demolition since the targeted remediations and interventions necessary to achieve an acceptable end state are integral to meeting license termination criteria. It also involves the final status surveys and sampling required to ensure acceptable levels have been achieved for release of the facility and post-closure monitoring to verify that end state conditions are met.

Figure 6.1: Radiation pathways during decommissioning closing and decommissioning



Source: Samseth, et al. (2012).

Site characterisation establishes the contaminants of concern, their concentrations/ levels concentrations (e.g. source-term) and their spatial distributions. This information is used for derivation of dose/risk values using exposure scenarios and dose modelling analysis to assess compliance with site release dose criteria. It also includes environmental sampling and monitoring data to assess accidental releases and supplement on-site characterisation. In some instances a combined risk estimate that includes radiological and hazardous materials may also be required to delicense a facility. Adequate characterisation is also necessary to assess environmental impacts.

Another concern is the development of detection techniques and methods, particularly remote detection of contamination, transport of contaminants in subsurface media and 3-D modelling. This theme also addresses potential groundwater contamination and 3-D contaminant plume modelling as well as potential transport to reach receptor points in the context of environmental monitoring. Issues addressed by this working group include:

- adequacy of characterisation for release (the extent required), performance period to demonstrate compliance (the duration required) and where and how the grid density is established;
- acceptance of residual activity: to what vertical and horizontal extents is the contamination to be characterised and how to convert dose into concentration (and *vice versa*), using exposure pathway analysis and scenarios;
- use of remote detection equipment (including automation and robotic techniques) for long-term monitoring of contaminated land or hard-to-reach areas, especially areas with high dose areas;
- environmental impact of soil decontamination and use of statistical tools and models to assess contaminant transport via environmental media;
- detection limits and equipment for mobile nuclides in soil and groundwater (e.g. tritium, strontium and caesium);
- 3-D modelling of subsurface soil and groundwater to simulate radionuclide movements and benchmarking of such models.

The group considered the first three issues as being of high priority for future R&D, with the remaining of medium priority.

Summary of current practices and guidance

License termination criteria has generally transitioned from quantitative limits on surface activities and concentrations to risk-based criteria with dose equivalent limits ranging from 10 μSv/year (HSE, 2008) to 250 μSv/year (IAEA, 2006d) to a future site occupant. Compliance with the clearance criteria is demonstrated through final characterisation and dose estimation to post-closure occupants of the facility using fate and transport models. Variations of two scenarios are generally used for the post-closure occupant: *i)* industrial use where portions of the site such as buildings or grounds continued to be used for commercial (e.g. non-residential) purposes; *ii)* a resident farmer scenario where agricultural pathways are modelled. Table 6.1 provides a list of current practice guidance documents used to model sites and demonstrate compliance with delicensing dose criteria.

Current characterisation, modelling and clearance practices have generally adopted similar approaches and standards in various member states. A typical approach is to establish a conceptual site model that represents the end state of the facility with regard to future land use and the physical attributes of potential remaining contamination. This includes the radionuclides of concern associated with the facility as well as the contaminated media and spatial distribution based upon characterisation data. The conceptual site model also includes the land use, hydrogeological, climatology and key parameters such as distribution coefficients for radionuclides as well as identification of the potential pathways for exposure to future occupants of the site. The conceptual site model is then used as input to computer models such as RESRAD for resident farmer agricultural pathways or RESRAD-BUILD for industrial scenarios involving use of site buildings (Yu, n.d.).

Table 6.1: Guidance documents for license termination surveys and modelling

Facility type	Phase	Region	Document
All types	Decommissioning surveys and sampling	United States	*A Nonparametric Statistical Methodology for the Design and Analysis of Final Status Decommissioning Surveys*, US NRC (1998)
All types	Decommissioning surveys and sampling	United States	*A Subsurface Decision Model for Supporting Environmental Compliance*, US NRC (2011c)
All types	Decommissioning surveys and sampling	United States	*Minimum Detectable Concentrations with Typical Survey Instruments for Various Contaminants and Field Conditions*, US NRC (1995)
All types	Decommissioning surveys and sampling	United States	*Performance and Documentation of Radiological Surveys*, ANSI (2001)
All types	Decommissioning surveys and sampling	European Union	*European Radiation Survey and Site Execution Manual (EURSSEM)* (2010)
All types	Decommissioning surveys and sampling	European Union	*Definition of Clearance Levels for the Release of Radioactively Contaminated Buildings and Building Rubble*, EC (1999)
All types	Decommissioning surveys and sampling	European Union	*Recommended Radiological Protection Criteria for the Clearance of Buildings and Building Rubble from the Dismantling of Nuclear Installations*, EC (2000b)
All types	Decommissioning surveys and sampling	United States	*Consolidated Decommissioning Guidance: Characterization, Survey, and Determination of Radiological Criteria* (Vol. 2), US NRC (2006)
All types	Decommissioning surveys and sampling	United States	*Multi-Agency Radiation Survey and Site Investigation Manual MARSSIM* (Revision 1), US NRC (2002a)
All types	Decommissioning surveys and sampling	United States	*Multi-Agency Radiation Survey and Assessment of Materials and Equipment Manual (MARSAME)*, US NRC (2009b)
All types	Decommissioning surveys and sampling	United States	*Guidance on Choosing a Sampling Design for Environmental Data Collection for Use in Developing a Quality Assurance Project Plan*, US EPA (2002)
All types	Decommissioning surveys and sampling	United States	*Characterization in Support of decommissioning Using the Data Quality Objectives Process*, ANSI (2008)
All types	Decommissioning surveys and sampling	United States	*Residual Radioactive Contamination from Decommissioning: Technical Basis for Translating Contamination Levels to Annual Total Effective Dose Equivalent* (Vol. 1), US NRC (1992)
All types	Clearance level modelling	United States	*Residual Radioactive Contamination From Decommissioning: User's Manual DandD Version 2.1* (Vol. 2), US NRC (2001)
All types	Clearance level modelling	United States	*Residual Radioactive Contamination From Decommissioning: Parameter Analysis* (Vol. 3), US NRC (1999c)
All types	Clearance level modelling	United States	*Comparison of the Models and Assumptions Used in the DandD 1.0, RESRAD 5.61, and RESRAD-Build 1.50 Computer Codes with Respect to the Residential Farmer and Industrial Occupant Scenarios Provided in NUREG/CR-5512*, US NRC (1999a)
All types	Clearance level modelling	United States	*Probabilistic Dose Analysis Using Parameter Distributions Developed for RESRAD and RESRAD-Build Codes*, US NRC (2000b)
All types	Clearance level modelling	United States	*Development of Probabilistic RESRAD 6.0 and RESRAD-BUILD 3.0 Computer Codes*, US NRC (2000a)
All types	Clearance level modelling	United States	*Probabilistic Modules for the RESRAD and RESRAD-Build Computer Codes*, US NRC (2000c)
All types	Clearance level modelling	United States	*Application of Model Abstraction Techniques to Simulate Transport in Soils*, US NRC (2011b)
All types	Clearance level modelling	United States	*Lessons Learned in Detecting, Monitoring, Modeling and Remediating Radioactive Ground-Water Contamination*, US NRC (2011d)

Table 6.1: Guidance documents for license termination surveys and modelling (*cont'd*)

Facility type	Phase	Region	Document
All types	Clearance level modelling	United States	*Understanding Variation in Partition Coefficient, K_d, Values: The K_d Model, Methods of Measurement, and Application of Chemical Reaction Codes* (Vol. 1), US EPA (1999a)
All types	Clearance level modelling	United States	*Understanding Variation in Partition Coefficient, K_d, Values: Review of Geochemistry and Available K_d Values for Cadmium, Cesium, Chromium, Lead, Plutonium, Radon, Strontium, Thorium, Tritium (^3H), and Uranium* (Vol. 2), US EPA (1999b)
All types	Clearance level modelling	United States	*Understanding Variation in Partition Coefficient, K_d, Values: Review of Geochemistry and Available K_d Values for Americium, Arsenic, Curium, Iodine, Neptunium, Radium, and Technetium* (Vol. 3), US EPA (2004)
All types	Clearance level modelling	United States	*Information on Hydrologic Conceptual Models, Parameters, Uncertainty Analysis, and Data Sources for Dose Assessments at Decommissioning Sites*, US NRC (1999b)
All types	Clearance level modelling	United States	*Evaluation of Subsurface Radionuclide Transport at Commercial Nuclear Power Plants*, ANS (2010a)
All types	Clearance level modelling	International	*Modelling the Transfer of Radionuclides from Naturally Occurring Radioactive Material (NORM)*, Report of the NORM Working Group of EMRAS Theme 2, IAEA (2007b)
All types	Clearance level modelling	United States	*User's Manual for RESRAD·OFFSITE Version 2*, US NRC (2007)
All types	Clearance level modelling	United States	*User's Manual for RESRAD Version 6*, ANL (2001)
All types	Clearance level modelling	United States	User's Manual for RESRAD-BUILD Version 3, ANL (2003)
All types	Clearance level modelling	United States	*External Exposure Model Used in the RESRAD Code for Various Geometries of Contaminated Soil*, ANL (1998)
All types	Clearance level modelling	United States	*A Compilation of Radionuclide Transfer Factors for the Plant, Meat, Milk, and Aquatic Food Pathways and the Suggested Default Values for the RESRAD Code*, ANL (1993)
All types	Clearance level modelling	International	*Remediation Process for Areas Affected by Past Activities and Accidents Safety Guide*, IAEA (2007c)
All types	Clearance level modelling	International	*Release of Sites from Regulatory Control on Termination of Practices*, IAEA (2006e)
All types	Clearance level modelling	International	*Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments*, IAEA (2010a)
All types	Clearance level modelling	International	*Validation of Models Using Chernobyl Fallout Data from the Central Bohemia Region of the Czech Republic – Scenario CB. First Report of the VAMP Multiple Pathways Assessment Working Group*, IAEA (1995)
All types	Clearance level modelling	International	*Modelling of Radionuclide Interception and Loss Processes in Vegetation and of Transfer in Semi-Natural Ecosystems*, IAEA (1996)
All types	License termination process	United Kingdom	*The Delicensing Process for Existing Licensed Nuclear Sites*, ONR (2013)
All types	Decommissioning surveys and sampling	United Kingdom	*Delicensing Guidance, Guidance to Inspectors on the Interpretation and Implementation of the HSE Policy Criterion of No Danger for the Delicensing of Nuclear Sites*, HSE (2008)
All types	Decommissioning surveys and sampling	United Kingdom	*Remediation of Radioactively Contaminated Sites*, SEPA (2011)
All types	Decommissioning surveys and sampling	European Union	*Inventory of Best Practices in the Decommissioning of Nuclear Installations: Final Report*, EC (2006)
All types	Decommissioning surveys and sampling	International	*Release of Radioactive Materials and Buildings from Regulatory Control*, OECD/NEA (2008)

Table 6.1: Guidance documents for license termination surveys and modelling (*cont'd*)

Facility type	Phase	Region	Document
All types	Decommissioning surveys and sampling	International	*Determination of the Characteristic Limits (Decision Threshold, Detection Limit and Limits of the Confidence Interval) for Measurements of Ionizing Radiation – Fundamentals and Applications*, ISO (2010)
All types	Decommissioning surveys and sampling	International	*Decommissioning of Facilities Using Radioactive Material*, IAEA (2006c)
Reactors	Decommissioning surveys and sampling	International	*Decommissioning of Nuclear Power Plants and Research Reactors*, IAEA (1999b)
Fuel cycle	Decommissioning surveys and sampling	International	*Decommissioning of Nuclear Fuel Cycle Facilities*, IAEA (2001)
All types	Decommissioning surveys and sampling	International	*Safety Assessment for the Decommissioning of Facilities Using Radioactive Material*, IAEA (2009f)
All types	Decommissioning surveys and sampling	International	*Characterization and Dose Modeling of Soil, Sediment and Bedrock During Nuclear Power Plant Decommissioning*, EPRI (2009a)
All types	Decommissioning surveys and sampling	International	*Groundwater Monitoring Guidance for Nuclear Power Plants*, EPRI (2005c)
All types	Decommissioning surveys and sampling	International	*A Practical Guide for the Performance of Combined Risk Assessment at Nuclear Power Plant Decommissioning Sites*, EPRI (2005a)
All types	Decommissioning surveys and sampling	International	*Summary of Utility License Termination Documents and Lessons Learned*, EPRI (2002b)
All types	Decommissioning surveys and sampling	International	*Guide to Assessing Radiological Elements for License Termination of Nuclear Power Plants* EPRI (2002a)
All types	Decommissioning surveys and sampling	International	*Embedded Pipe Dose Calculation Method*, EPRI (2000a)
All types	Decommissioning surveys and sampling	International	*Use of Probabilistic Methods in Nuclear Power Plant Decommissioning Dose Analysis*, EPRI (2002d)
All types	Decommissioning surveys and sampling	International	*Trojan License Termination Plan Development Project*, EPRI (2002c)
All types	Decommissioning surveys and sampling	International	*Determining Background Radiation Levels in Support of Decommissioning Nuclear Facilities*, EPRI (2001b)

Most countries have adopted a sampling and survey regimen that is statistically based for final status surveys and sampling to demonstrate compliance with license termination criteria. EURSSEM (2010) and MARSSIM (US NRC, 2002a) assume uniform distributions of surface-contaminated land and structures and utilise statistical models predicated on this assumption to establish confidence levels associated with the source term determinations. These assumptions are limited by the nature of contaminating events which tend to be from localised spills and leaks and are rarely from large-scale airborne contamination events that would distribute source terms evenly in an impacted area. Consequently areas that are likely to exceed the clearance level (DCGL) require 100% scans which are very time consuming and costly. Data quality objectives are established and survey plans are designed to ensure adequate coverage and sensitivity to ensure any remaining source term meets the dose-based license termination criteria.

Current environmental monitoring practices rely on fixed location monitoring (such as environment air sample, thermoluminescent dosimeter locations and periodic sample collection and analysis regimens) associated with radiological environmental monitoring programmes (REMP) at operating and decommissioning facilities. This can also consist of confirmatory groundwater monitoring programmes that require scheduled sample collection and analysis from a set of monitoring wells which may persist after land and structure clearance and license termination is achieved. In the case of facilities placed in SAFSTOR or care and maintenance some form of environmental monitoring regimen will also be required. Such monitoring regimens are not dynamic or continuous and require a high degree of understanding of environmental conditions (e.g. wind speed and direction, site hydrology, site geology, background radionuclides distribution and variability) in order to be meaningfully interpreted.

Summary of challenges and R&D needs

The definition of end states for D&D facilities and of standards for release of materials for recycling will have a major impact on cost, schedule and risks to the public, workers and the environment. However, there is insufficient scientific basis for comparing the safety of various end states. Research should be directed toward understanding the fate and behaviour of treated and untreated contaminated material by determining the fundamental chemical species of the contaminants in the host material and how the species behave. The effect of time and changing ambient conditions should be considered in these investigations. Further research should be directed at incorporating these results into risk assessments to evaluate and compare the long-term safety provided by different end-state options. A better scientific underpinning for the selection of facility end states requires further development in the research of the actual health risk of residual levels of radioactive and hazardous materials, and the transport dynamics (e.g. fate and behaviour) of contaminants in D&D facilities (NRC, 2001).

Research is needed to understand the physical and chemical forms (speciation) of contaminants in building construction materials. Buildings may be in storage for decades before D&D and potentially in use for decades thereafter. Understanding the speciation and behaviour of the contaminants, how the speciation evolves with time and the impact of decontamination activities on the chemical speciation, is critical for developing a scientific basis for determining end states. Decontamination often uses chemical or biological processes that can impact the behaviour and performance of construction materials as well as the contaminants themselves. The use of chemicals or bacteria for decontamination can dramatically affect the local environment by changing pH or inducing chemical reduction or oxidation reactions through respiratory activity. This activity, coupled with physical changes due to material cutting, melting or polishing from decontamination efforts, can impact the behaviour of the contamination. For example, acids dissolve concrete; less dramatic reductions in pH can also have profound effects, but details of these changes over time and how they may affect the eventual release of contaminants are not well understood. Even if there are no decontamination activities the host material will change with time.

The behaviour of hazardous airborne species presents another opportunity for research. While water is the primary mover of contaminants in the ground (subsurface contamination), airborne pathways may be especially important in establishing a scientific basis for facility end states. Performance assessment modelling using fundamental fate and transport data could be developed as an important decision-making tool for establishing facility end states (NRC, 2001).

In addition, the available fate and transport models generally allow a narrow range on end-state contaminant scenarios to be addressed within a single model. Sites with several contamination sources such as contaminated land, subsurface contamination from leaks or spills, or with end states that leave contaminated structural materials such as basement floors and walls in the subsurface, often require the use of multiple models and different fate and transport softwares to predict the overall risk to a future occupant.

The challenges identified by the working group under this theme include:

- identification of reliable, adequate characterisation methods to identify subsurface radionuclide contamination and assess long-term transport via environmental media with minimal intrusive characterisation;

- selection, evaluation and benchmarking of contaminant transport codes and models in consideration of potential long-term environmental impacts (e.g. 1 000 years or more);
- proper assessment of the source terms, considering lack of data and selection of appropriate statistical methods and models using a probabilistic approach;
- establishing appropriate realistic scenarios for receptors (e.g. a representative person), considering land use for a specific performance period.

Suggested additional research and development

Adequate non-intrusive characterisation techniques for subsurface/volumetric media coupled with 3-D modelling

Challenges

Sampling at the end of the project to verify that clearance or license termination levels have been met, such as those developed using MARSSIM (US NRC, 2002a) and EURSSEM (2010), use random sampling designs based on the premise that contamination is homogenous. Biased samples based on process knowledge are added to the randomly generated sample and scan points to account for the inhomogeneous nature of most contaminants within a survey area. MARSSIM also applies only to release criteria for building surfaces and soils up to 15 cm deep. Thus, these frameworks are poorly suited to subsurface or volumetric media. The software such as RESRAD, RESRAD-OFFSITE and RESRAD-BUILD (Yu, n.d.) used to determine acceptable release levels are limited in their ability to model embedded and subsurface contaminants' fate and transport.

First appearing in the 1980s, the data quality objectives (DQO) process has motivated a number of follow-up guidance documents implementing the process for the NRC, DOE and EPA and has shaped the landscape of environmental investigations for the last 30 years. During this time, the environmental community has seen the emergence of advanced sampling and remote sensing technologies, statistical and mathematical models and decision support systems that deal with various aspects of site investigation. Members of the regulatory community, particularly at the Environmental Protection Agency (EPA), have called for a substantial update of the DQO process to integrate these new and powerful approaches into a second-generation DQO process. Unfortunately, the response to such calls for revision have been slow, primarily because the implications of such changes are difficult to ascertain (Stewart, 2011).

While no sweeping update has occurred, the EPA has articulated the Triad model. Triad represents a concerted effort by experts from the public and private sector to create a modern approach that lays the groundwork for a second-generation DQO process. Triad methodology spans the project life cycle, providing continuity between management practices, scientific methods and technological advances that emphasise the quality of the decision. At the centre of the Triad model is the conceptual site model (CSM), which is a representation of site knowledge that evolves over the course of investigation. CSM communicate knowledge about a variety of issues, including geology, exposure pathways, spatial distribution of contamination and transport mechanisms. Under Triad, the CSM drives data collection by identifying knowledge gaps. The CSM is reciprocally informed and evolved by the outcome of those data. Triad recognises the value of accurate laboratory analysis but also calls for the inclusion of screening and field detection methods that are typically faster and less expensive to collect. The combination of speed and reduced costs can result in a greater sampling density and better support for CSM evolution. Given these recent advances, it may be time to identify opportunities within regulatory guidance where Triad principles and geostatistical advances can be drawn together into the regulatory process (Stewart, 2011).

As discussed in Chapter 2, applications are currently being developed, used and tested for decommissioning that show promise for providing the same statistical confidence with fewer samples and more accurate determination of acceptable levels or derived concentration guidelines (DCGL) for embedded or subsurface contaminants. The US NRC's NUREG/CR-7021 (2011c) shown in license termination criteria has generally transitioned from quantitative limits on surface activities and concentrations to risk-based criteria with dose equivalent limits ranging from 10 μSv/year (ANS, 2010a) to 250 μSv/year (Ward, Gee and White, 1997) to a future site occupant.

Table 5.2 incorporates geostatistical methodologies into the license termination criteria for sites with subsurface contamination. However, this CSM method for increasing the statistical certainty that subsurface contamination meets license termination criteria has not yet been implemented. Surface scans prior to backfilling remediated soils and factoring in groundwater potential radiation exposures are still being used to verify license termination criteria. Final status surveys have not yet incorporated the geostatistical methods. As noted in Chapter 2 these tools are primarily being used only for planning and execution of remediations in Europe and in the United States.

The geostatistical methods infer the contaminant distributions through kriging and assign statistical confidence levels to the three-dimensional array, negating the need to rely on methods based on surface measurements. These tools can optimise the final status survey strategy by minimising the surface scanning and subsurface sampling required to have high statistical confidence that the release criteria have been met. As mentioned in Chapter 2, geostatistical cartographies have been successfully performed using ISATIS software (Desnoyers and Dubot, 2012b; Matzke, et al., 2007). Some studies conclude that conventional statistical [e.g. EURSSEM (2010), MARSSIM (US NRC, 2002a)] data processing and geostatistical data processing are complementary rather than in opposition to one another when applied to the proper radiological characterisation stage of a decommissioning and dismantling project (US DOE, 2009). Development of guidance documents, such as NUREG/CR-7021 (US NRC, 2011c), incorporating geostatistical methods into final status survey execution, as well as integration of geostatistics with survey and sample planning tools is required to take full advantage of the new geostatistical modelling capabilities.

In addition, complex and multi-hazard site investigations, remediations and clearance require more sophisticated integrated multi-disciplinary (e.g. hydrogeological, health physics, environmental) analytical tools to correctly model complex environmental interactions. Radiological and hazardous contaminants have been introduced into complex subsurface environments by way of: *i)* intentional disposals through injection wells, at disposal facilities, or in evaporation or seepage ponds (e.g. at some US DOE sites); *ii)* accidental spills and leaks from waste storage tanks, basins and transfer lines or accident locations like Chernobyl and Fukushima. Many sites have multiple sources of surface and subsurface contamination that require fate and transport modelling. Understanding contaminant fate and transport is difficult because of the complex subsurface environments that are characterised by multiple hydrological, geochemical and microbiological processes occurring at different scales, with significant heterogeneity and daunting measurement and observational constraints.

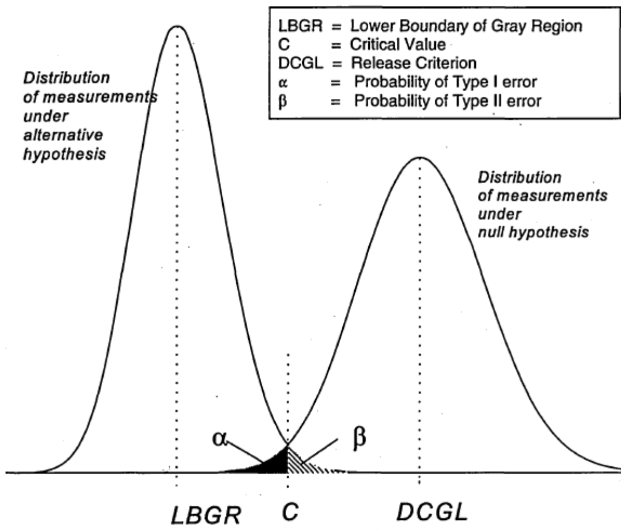
Summary of current R&D for characterisation using statistical sampling and 3-D modelling

- Development of 3-D models and statistical tools to simulate subsurface contamination

At the final status survey stage of site characterisation, statistical approaches enable the determination of the minimum number of samples required to be collected to meet desired statistical confidence levels. Classical statistical tests then allow validation that clearance levels are met. The decision to consider compliance with clearance levels is based on a statistical test and requires contaminant levels to be collected using random sampling designs. When some initial parameters are missing such as the true variance of the background levels, more advanced strategies, such as two-phase sampling designs, can be employed to implement a simple random sampling characterisation plan.

A large number of statistical tests are available: comparing average to a fixed threshold (as shown in Figure 6.2), comparing proportions, estimating the mean, constructing a confidence interval of the mean. However there should be consideration of the underlying hypotheses of these statistical tests such as the actual spatial randomness of values and the type of statistical distribution. (Desnoyers and Dubot, 2012b).

Figure 6.2: Critical value and null hypothesis underpinning MARSSIM and EURSSEM



For the final phase of characterisation to demonstrate compliance with clearance levels destructive samples are located according to the radiation survey results using maps of the survey unit. This is the judgemental part of the methodology which uses biased sampling based upon survey data in addition to random sampling based upon an assumed homogenous distribution of the contaminant. Additional sampling points might then be located using the same approach as that for the surface radiation mapping (reduction of uncertainty, intermediate probability validation).

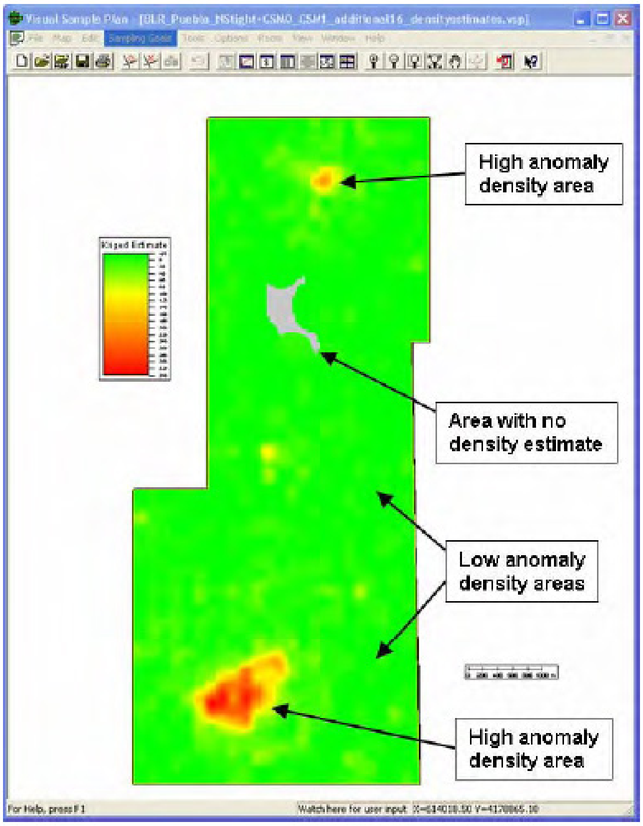
The vertical variability of the contamination with depth is normally significantly higher than the variability in the horizontal plane for any given contaminant location in media such as soil or concrete. Sampling resolution in the vertical direction has to be denser as a consequence (typically a few centimetres or less for building structures and a few dozens of centimetres for soils) in order to achieve high confidence levels for estimation of the total source term. Determining the correct number of samples for the final radiological characterisation relies on a statistical test to be performed at the end to demonstrate that clearance levels have been met with sufficient statistical certainty. Statistical tests and equations, such as the Sign Test or Wilcoxon Rank Sum Test, are widely used and quite easy to implement to get the required confidence level for decision making (Desnoyers and Dubot, 2012b; EURSSEM, 2010; US NRC, 1998, 2002a).

Visual Sample Plan (VSP) is a software tool used to design surveys and analyse data for MARSSIM (US NRC, 2002a) Final Status Surveys (US DOE, 2009; Matzke, et al., 2007). Most of the sampling designs validated are probability-based, meaning samples are located randomly (or on a randomly placed grid) and the number of samples is calculated such that, if the amount and spatial extent of contamination exceeds levels of concern, at least one of the samples would be taken from a contaminated area at least X% of the time. Hence, "validation" of the statistical sampling algorithms is defined to mean ensuring that the X% (confidence) is actually met. A Visual Sample Plan validation effort focused on four VSP sampling designs based on the following sampling objectives that were deemed pertinent for sampling within a building after a chemical or biological attack (Nuffer, et al., 2009):

- *Upper-tolerance-limit-based sampling* – Statement that X% confident that at least Y% of surface area is below some quantitative contaminant limit where only random samples are obtained.
- *Compliance sampling* – Statement that X% confident that at least Y% of surface area contains no detectable contamination where only random samples are obtained.
- *Combined judgment and random sampling* – Statement that X% confident that at least Y% of surface area contains no detectable contamination where both random and judgmental samples are obtained.
- *Hotspot sampling* – Statement that at least X% confident that any contaminated area greater than a given size and shape is sampled.

The VSP geostatistic module identifies locations within the transects that are identified as being high density (i.e. high number of anomalies within a specified amount of the surveyed transect area). The geostatistical anomaly density mapping is composed of two primary tasks. The first task is to model the spatial variability of the measured anomaly densities as determined from the geophysical transect data; it involves the development of a variogram based on the window-averaged transect density values. A variogram depicts how the variability of a set of values changes as the distance between the spatial locations of these values increases. The second task involves the estimation of anomaly density at unsampled locations within the study area through the geostatistical methodology known as kriging.

Figure 6.3: Results of kriging estimation displayed in VSP

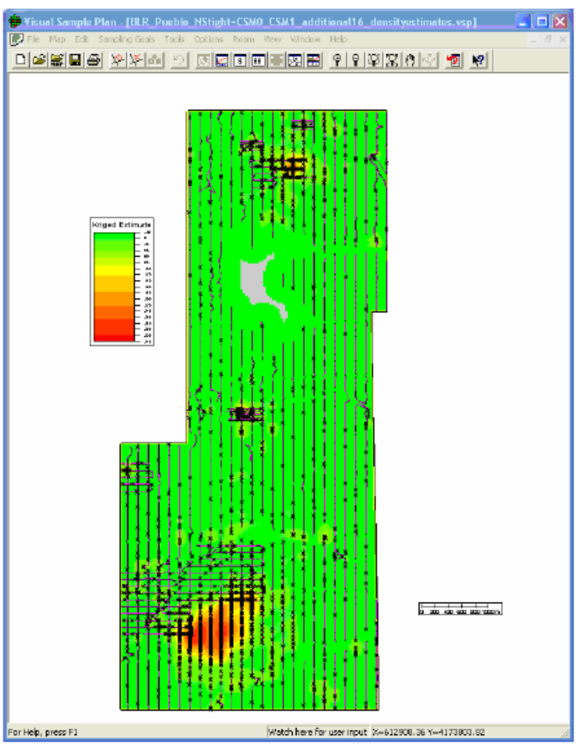


Kriging uses the model of spatial variation as captured by the variogram to provide an unbiased, minimum-variance estimate of the anomaly density. Kriging is the procedure that creates the final anomaly density map. To use these methods, GAM/GAMV and KT3D also must be installed (Matzke, et al., 2007).

In addition to the estimate of anomaly density, the kriging procedure generates a map of the estimation variance. Figure 6.4 shows the kriging estimation variance computed for the kriging results from the Pueblo Precision Bombing Range study area. The estimation variance shows the uncertainty of the kriging estimate and is a function of the data configuration and the variogram model. Estimates of anomaly density for locations on or near sample transects should be very accurate and, hence, have a low uncertainty. Conversely, estimates at distance from the sample transects are likely to be less accurate with a relatively high uncertainty. The estimation variance value reaches a maximum at distances greater than or equal to the variogram range away from the nearest data point. The map of estimation variance shows how the variance of the anomaly density changes across the study site.

Insights from ongoing US NRC reviews of field investigations involving radionuclide transport in the subsurface illustrate the need to test and confirm conceptual site models (CSM). The assumptions and parameterisation inherent to these CSM, which affect radionuclide release and transport, should be tested. In particular, the unsaturated zone where many leaks and spills originate needs detailed characterisation and confirmatory monitoring.

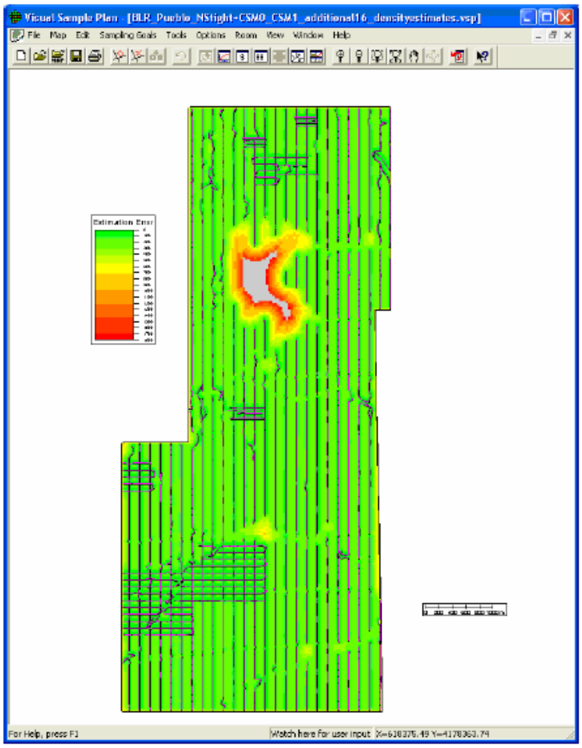
Figure 6.4: Kriging estimation and course-over-ground traces displayed in VSP



A dose assessment to determine risk-informed compliance with regulatory criteria is used to evaluate the need for and selection of remediation methods. If remediation is warranted, the choice of the employed remediation method(s) is based on site and source characterisation, modelling and monitoring data. These data should be used to the test the CSM. Remediation options range from highly-aggressive methods, such as pump, treat, monitor and recycle or release, to more passive methods, such as monitored natural attenuation. All successful remediation strategies involve monitoring programmes to determine their efficacy. This monitoring is coupled with performance assessment models using performance indicators (PI), which provide a measurable indication of remediation performance and are derived from analysis of the CSM and monitoring data (Nicholson, et al., 2012).

Figure 6.5: Kriging variance and course-over-ground traces displayed in VSP

Highest variance values in red; lowest values in green

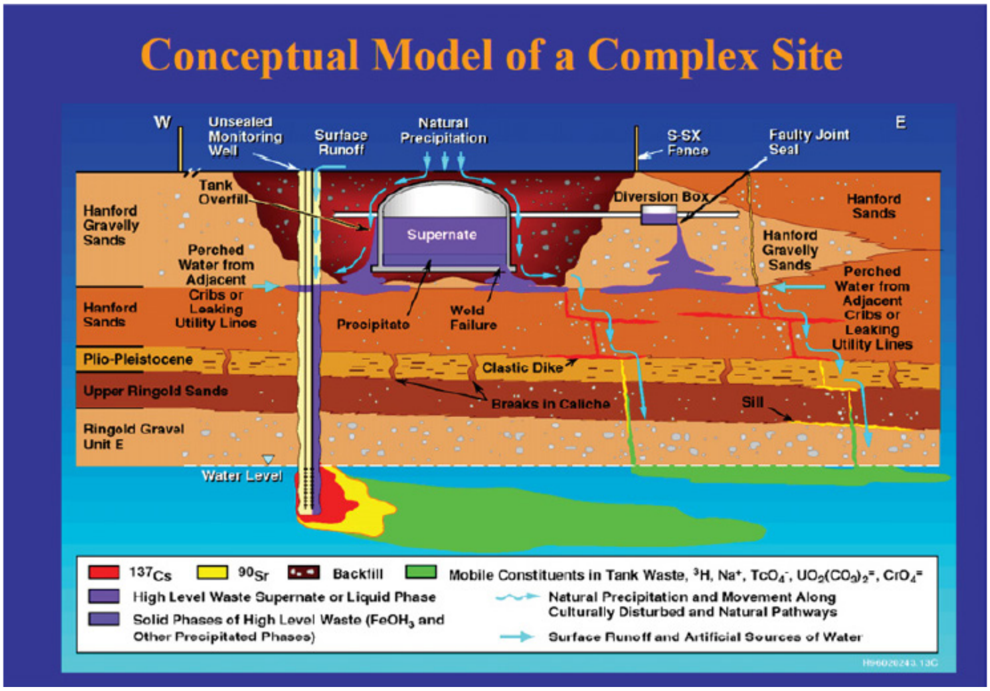


Recently, the American Nuclear Society and American National Standards Institute issued an industry-consensus standard ANSI/ANS-2.17-2010, *Evaluation of Radionuclide Migration in the Subsurface at Commercial Nuclear Power Plants* (2010a). This standard incorporates valuable guidance on a range of topics, e.g. hydrogeologic characterisation, CSM, performance assessments, mathematical modelling, performance-confirmation monitoring and information management. Its appendices provide listings of potentially relevant resources for conducting subsurface radionuclide transport characterisation, monitoring and modelling programmes.

Recent work focused on model abstraction techniques to judge whether simplifications used in modelling introduce significant uncertainties and/or errors. This research analyses multi-year watershed scale tracers and ecological/hydrometeorological studies. The approaches developed can be used to assess leak-generated infiltrations, percolations and migrations to groundwater that affect pathways (Nicholson, et al., 2012).

A dose assessment to determine risk-informed compliance with regulatory criteria is a primary consideration for the NRC. In particular, the potential for a public drinking water pathway can increase the public dose and escalate the need for and selection of remediation methods. To determine whether groundwater remediation is needed and what remediation strategy should be pursued, one must analyse the groundwater monitoring data to define the contaminant plume and its behaviour over time (Stewart, 2011).

Figure 6.6: Conceptual model of hydrogeologic/ engineered features affecting groundwater



Source: Ward, Gee and White (1997), Meyer and Gee (1999).

The Multi-Agency Radiological Site Survey Investigation Manual (MARSSIM) (US NRC, 2002a) is a regulatory guidance document regarding compliance evaluation of radiologically contaminated soils and buildings. Compliance is determined by comparing radiological measurements to established limits using a combination of hypothesis testing and scanning measurements. Scanning allows investigators to identify localised pockets of contamination missed during sampling and allows investigators to assess radiological exposure at different spatial scales. Scale is important in radiological dose assessment since regulatory limits can vary with the size of the contaminated area and sites are often evaluated at more than one scale. Unfortunately, scanning is not possible in the subsurface and direct application of MARSSIM breaks down. MARSSIM is a comprehensive decision framework for surface contamination but stops short of formalising a process for the subsurface (Stewart, 2011).

A subsurface decision framework called the Geospatial Extension to MARSSIM (GEM) is used to provide multi-scale subsurface decision support in the absence of scanning technologies. Based on geostatistical simulations of radiological activity, GEM recasts the decision rule as a multi-scale, geospatial decision rule called the regulatory limit rule (RLR). The RLR requires simultaneous compliance with all scales and depths of interest at every location throughout the site. The RLR is accompanied by a compliance test called the stochastic conceptual site model (SCSM). For those sites that fail compliance, a remedial design strategy is developed called the multi-scale remedial design model (MrDM) that spatially indicates volumes requiring remedial action. The MrDM is accompanied by a sample design strategy known as the multi-scale remedial sample design model (MrsDM) that refines this remedial action volume via careful placement of new sample locations. Finally, a new sample design called "check and cover" is presented that can support early sampling efforts by directly using prior knowledge about where contamination may exist (Stewart, 2011).

The development of the Spatial Analysis and Decision Assistance (SADA) software at the University of Tennessee Knoxville includes three-dimensional geostatistical capabilities for subsurface modelling to aid MARSSIM planning and surveys for final site clearance. SADA provides a number of critical MARSSIM tools for sample design and checks for compliance. These include a formal MARSSIM approach for individuals building a MARSSIM assessment from scratch. In addition, users can access various stages (available through the MARSSIM Quick tools) of the process to introduce a SADA mid-evaluation. Regulators can also use the Quick tools to check a licensee's work (UT, 2007). SADA is free software that incorporates tools from environmental assessment fields into an effective problem-solving environment. These tools include integrated modules for visualisation, geospatial analysis, statistical analysis, human health risk assessment, ecological risk assessment, cost/benefit analysis, sampling design and decision analysis. The capabilities of SADA can be used independently or collectively to address site-specific concerns when characterising a contaminated site, assessing risk, determining the location of future samples or when designing remedial action (MARSSIM, 2006).

Figure 6.7 demonstrates how SADA uses a geospatial estimator on only the points defined within the polygon. The polygon can also be used in this manner to generate screening or risk results of subsets of the input data.

SADA is an evolving freeware product targeted toward individuals performing environmental assessments in support of decision making. The primary objective is to create a user-friendly software package for environmental characterisation and decision making. This problem-solving environment applies and integrates a number of algorithms that can either be used in a stand-alone fashion or in the direct support of performing a site assessment. SADA processes and produces its information in a clear, transparent manner, directly supporting decision processes, and can serve as a communication tool between technical and non-technical audiences. The end result is that SADA can be used to facilitate decisions about a given site in a quick and cost-effective manner. SADA has a strong emphasis on the spatial distribution of contaminant data and is therefore best suited for anyone who needs to look at data within a spatial context (UT, 2007). Similar tools are being used in Europe for guide sampling, remediation and risk assessment (Berton, 2011).

Figure 6.7: Spatial analysis and decision assistance for subsurface modelling: 2-D

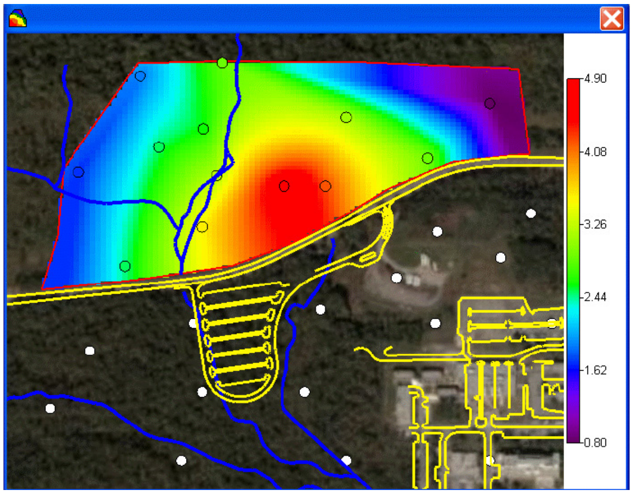
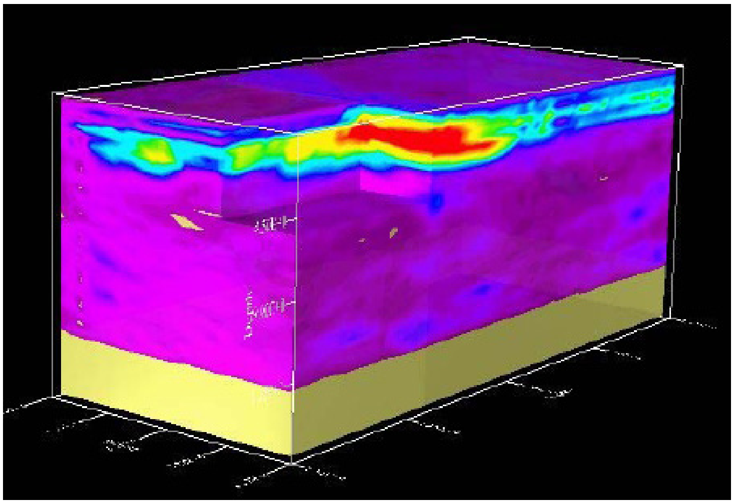


Figure 6.8: Data can also be visualised in true 3-D

SADA comes with a three-dimensional visualisation feature with a great deal of control over image rendering



- Development of integrated environmental models

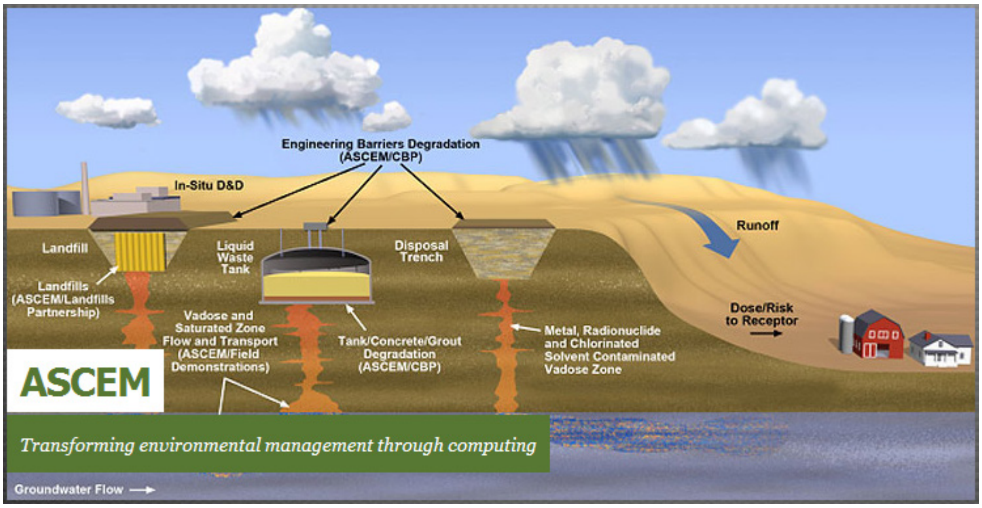
The National Academy of Sciences prioritised four research and development gaps for the DOE's Office of Environmental Management (DOE EM) groundwater and soil remediation roadmap as listed in Table 6.2.

Table 6.2: NAS prioritised groundwater and soil remediation research and development gaps

GS #	Gap	Priority
GS-1	Contaminant behaviour in the subsurface is poorly understood	High
GS-2	Site and contaminant source characteristics may limit the usefulness of baseline subsurface remediation technologies	Medium
GS-3	Long-term performance of trench caps, liners and reactive barriers cannot be assessed with current knowledge	Medium
GS-4	Long-term ability of cementitious materials to isolate wastes is not demonstrated	High

To close these gaps, the DOE EM, in collaboration with the DOD, EPA and NRC, is developing an advanced simulation capability for environmental management (ASCEM) that consists of a state-of-the-art scientific tool and approach for understanding and predicting contaminant fate and transport of both radiological and non-radiological contaminants in natural and engineered systems (Meza, et al., 2011). The mission of the DOE EM is to complete the safe clean-up of the environmental legacy from the nation's five decades of nuclear weapons development and government-sponsored nuclear energy research (NRC, 2009). This clean-up effort is one of the most complex, and technically challenging, in the world. It is projected to be ongoing for decades and to cost from USD 265-305 billion to complete (Meza, et al., 2011).

Figure 6.9: Graphic depiction of ASCEM prediction of contaminant fate and transport



Source: Meza, et al. (2011).

The ASCEM (US DOE, n.d.) uses modular and open source, high-performance software tool sets to facilitate integrated approaches to modelling and site characterisation that enable robust and standardised assessments of performance and risk for clean-up, intervention and closure activities (Meza, et al., 2011). The development of next-generation, science-based reactive flow and transport simulation capabilities and supporting modelling tool sets within a high-performance computing framework aims to address the DOE EM's waste storage and environmental clean-up challenges (US DOE, n.d.). Use of ASCEM will help to better estimate clean-up time and costs and reduce uncertainties and risks (Meza, et al., 2011). The integrated simulation framework is being developed along three major objectives or "thrust areas" (US DOE, n.d.; Hiergesell and Taylor, 2011):

- *Platform and integrated tool sets:* Provides the user with tool sets for model development and analysis, visualisation and management of data and simulation results. The framework will allow the use of disparate, multi-scale and often sparse information for subsurface property and process parameterisation. The platform and its interoperable structure will be user friendly; it will not require users to have extensive expertise with high-performance computing tools and interfaces. An important goal is to develop a set of modelling and simulation tools that can be used on a wide range of computer platforms ranging from desktop computers to the largest computing facilities available within the US DOE (n.d.).

- *Multi-process high performance computing simulator:* Provides the user with state-of-the-art flexible and extensive simulation capabilities for a range of modern computer architectures. The thrust area will develop a fate and transport simulator in support of environmental management applications and performance assessments that is designed to use the high-performance computing (HPC) power of modern architectures, from laptops to supercomputers. A graded, iterative approach to assessments generates a suite of conceptual models that span a range of complexity. Such models may require coupling of hydrological, biogeochemical, geo-mechanical and thermal processes in order to provide a comprehensive and accurate simulation of contaminant fate and transport. The platform and integrated tool sets, named "Akuna", provides the tools for users to generate this wide range of conceptual models, and "Amanzi" provides the flexible and extensible computational engine to simulate them. Developing Amanzi in a manner that is modular and extendable is achieved with a hierarchical and modular design that captures all the steps involved in translating a conceptual model to output for analysis. An object-oriented programming model is used to support this hierarchical view, with higher-level objects and much of the code being developed in the C++ language. Specifically, the design has high-level objects that abstract process models and their coupling, supporting tool sets that provide the building blocks for these high-level objects, and low-level objects and services that are used by the supporting tool sets (US DOE, n.d.).

- *Site application:* Provides the ASCEM developers and users with the expertise needed to address the DOE EM's environmental clean-up challenges. Tasks this area include (US DOE, n.d.):
  - A user requirements interface enhanced by engaging end users to inventory and document simulation needs across the DOE EM complex and to identify considerations related to efficient implementation of performance assessments. The end user interaction results provide input to requirement specifications for the platform and high performance computing development teams.
  - Establish demonstration site attributes by identifying and prioritising criteria for selecting sites at which to demonstrate the platform and the high-performance computing core. A range of key site conditions were examined and documented (e.g. humid vs. arid; porous granular vs. fractured rock materials; saturated vs. unsaturated; pH background and contaminant geochemistry, as well as other site conditions; and subsurface contamination vs. waste behaviour in tanks).
  - Establish site application working groups by using the criteria established in the second task to choose representative demonstration sites and establish working groups for demonstrations. Current working groups include: *i)* Attenuation-Based Remedies for the Subsurface (Savannah River Site F Area); *ii)* Deep Vadose Zone (Hanford site's BC Cribs and Trenches); *iii)* a representative Waste Tank Performance Assessment and preliminary efforts on mercury contamination (Oak Ridge National Laboratory).

The Attenuation-Based Remedies for the Subsurface Working Group is focused on the Savannah River Site (SRS) F Area and evaluation of remediation and natural attenuation approaches for uranium-contaminated groundwater. This group was the focus of the Phase I demonstration and efforts continue in Phase II.

The data-rich F Area provides an opportunity to test ASCEM capabilities on a complex remediation problem (US DOE, n.d.; Meza, et al., 2011). The SRS F Area seepage basins consist of three unlined, earthen surface impoundments that received ~7.1 billion litres (1.8 billion gallons) of acidic, low-level waste solutions. The acidic liquid waste (average influent pH of 2.9 originated from the processing of irradiated uranium in the F Area separations facility from 1950 through 1989. The plume currently extends from the basins to approximately 600 metres down-gradient at a stream and contains a large number of contaminants. Based on risk to potential receptors, the most hazardous contaminants are uranium isotopes, ^{90}Sr, ^{129}I, ^{99}Tc, tritium and nitrate. Groundwater is currently acidic, with pH values as low as 3.2 near the basins. As a result, the sediments that underlie the F Area have been exposed to acidic solutions for many decades. The following processes affecting the attenuation of uranium at the F Area were considered in this study (US DOE, n.d.):

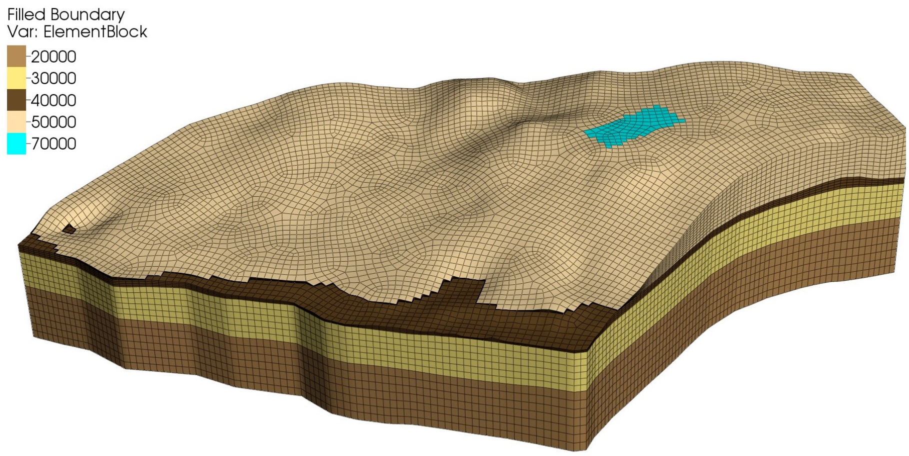
- *Adsorption/desorption:* Considered to be dominant natural attenuation mechanisms.
- *Dilution/mixing:* Dilution is also considered an important attenuation mechanism; dilution and mixing occur at the interfaces of the plume and uncontaminated water.
- *Mineral dissolution/precipitation:* These processes occur throughout the plume and are particularly important in slowing the advance of a leading pH gradient.
- *Aqueous reactions:* Occur between dissolved species; both can add and remove free protons from groundwater and occur throughout the plume.

The development of conceptual and numerical models (including the computational domain, boundary conditions and model parameters), is guided by 2-D and 3-D scoping studies conducted using the existing PFLOTRAN simulator (Figure 6.10) (US DOE, n.d.).

Figure 6.10 depicts the four major hydro-stratigraphic units that are included in this model of the F site. The horizontal resolution of this mesh is approximately 16 metres (US DOE, n.d.). The top layer (ID: 50 000) is the upper aquifer zone (UAZ), followed by the tan-clay confining zone (ID: 40 000), the lower aquifer zone (ID: 30 000), and the Gordon confining unit (ID: 20 000). The largest F basin (ID: 70 000) is shown in a greenish-blue cyan colour, and is where the contaminant source was positioned in this model. The simulation was run on 256 cores of the Franklin Cray XT4 system at NERSC. The 17-component chemistry model was used, and the retardation of the uranium plume relative to the non-reactive tracer is evident, as the tracer has already reached the site's 4-mile branch (US DOE, n.d.).

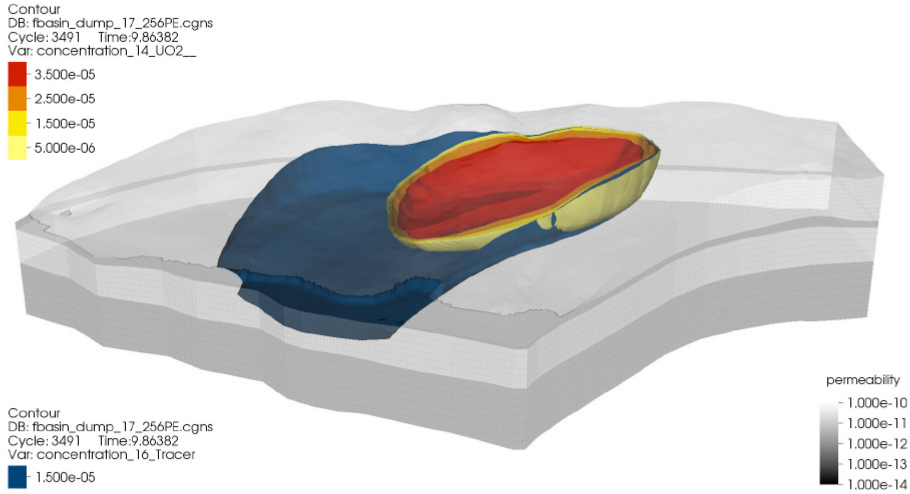
In order to demonstrate the capabilities of the Amanzi prototype, a numerical model of the F Area seepage basins was constructed. Figure 6.11 depicts the isosurfaces of the uranium plume (yellow and red) and a non-reactive tracer (blue) at 9.86 years for the unstructured mesh F Area seepage basin model described above (US DOE, n.d.).

Figure 6.10: PFLOTRAN simulator depicting an unstructured hexahedral mesh



Source: US DOE (n.d.).

Figure 6.11: Isosurfaces of the uranium plume at the SRS F area seepage basin



Source: US DOE (n.d.).

At the Hanford site, a Working Group is focused on the evaluation of innovative treatment technologies for recalcitrant contaminants in the deep vadose zone. The current effort is focused on an end-to-end demonstration of ASCEM using the relatively sparse data set from the site's BC cribs. The no-action case and conceptual model uncertainty are being evaluated for the Phase II demonstration.

The Waste Tank Performance Assessment working group at Oak Ridge National Laboratory is focused on demonstrating the ASCEM capabilities needed for DOE EM performance assessments. These assessments are associated with waste tank closures and LLW disposal in engineered containment systems. While a specific site is not being evaluated, representative problems are included as part of this working group's effort. The Oak Ridge Mercury Working Group is focused on initial scoping efforts to determine the nature of the demonstration. ASCEM capabilities need to be developed to support a demonstration at the site (US DOE, n.d.).

In addition to these working groups, Site integration teams are being formed to evaluate specific problems at DOE EM sites. The difference from demonstration working groups is that these teams involve partnership and co-funding with DOE EM sites for applying specific ASCEM capabilities to target problems. The emphasis over time will shift from the demonstration working groups to site integration teams as ASCEM development and quality assurance matures (US DOE, n.d.).

A similar effort is under way at the International Atomic Energy Agency (IAEA) with the Modelling and Data for Radiological Impact Assessments (MODARIA) project (IAEA, 2014b). MODARIA continues some of the work of previous international exercises in the field of radioecological modelling and focuses on areas where uncertainties remain in the predictive capability of environmental models. These previous international exercises include BIOMOVS (Biospheric Model Validation Study) and BIOMOVS II, initiated by the Swedish Radiation Authority in 1985, and the programmes sponsored by the IAEA's VAMP (Validation of Model Predictions, 1988-1996), BIOMASS (Biosphere Modelling and Assessment, 1996-2001), EMRAS (Environmental Modelling for Radiation Safety, 2003-2007) and EMRAS II (2009-2011) (IAEA, 2013c, 2014b, 2014d).

The overall objective of the EMRAS programme was to enhance the capabilities of member countries to model radionuclide transfer in the environment and, thereby, to assess exposure levels of the public and biota in order to ensure an appropriate level of protection from the effects of ionising radiation associated with radionuclide releases, and from existing radionuclides in the environment. Specific objectives in the areas of radioactive release assessment, restoration of sites with radioactive residues, and environmental protection are (IAEA, 2013b):

- to test the accuracy of the predictions of models for assessing the transfer of radionuclides in the environment;
- to develop and improve models for particular environments and, where appropriate, to agree on generally applicable transfer parameter values;
- to provide a forum for the exchange of ideas, experience and research information.

Activities within the framework of the MODARIA programme emphasise improvement of environmental transfer models for reducing associated uncertainties or developing new approaches to strengthen the evaluation of the radiological impact to man, as well as to flora and fauna, arising from radionuclides in the environment (IAEA, 2014b; Sheppard, et al., 2011).

An example of an international effort where the ASCEM (US DOE, 2010a) and/or MODARIA capabilities can be tested and developed is the International Test Case Proposal for the Chernobyl Cooling Pond Decommissioning and Remediation, sponsored by the Interagency Steering Committee on Multimedia Environmental Modeling (ISCMEM) (2009, 2011; Faybishenko, 2011). The heavily contaminated water in the reactor basement and soil at the site reaches a total radioactivity greater than 200 TBq and includes ^{137}Cs-80%, ^{90}Sr-10%, ^{239,240,241}Pu-10% from the routine releases of contaminated water into the cooling pond (ISCMEM, 2011).

The ISCMEM is a non-OSTP affiliated organisation of nine federal agencies (NRC, EPA, USACE, DOE, USGS, NOAA, NRCS, Bureau of Rec, NSF) established by a memorandum of understanding (MOU) for the purpose of collaborating on the development of multimedia environmental models. This international project provides an opportunity for federal agencies to review and select information from long-term observations at the Chernobyl cooling pond and the surrounding 30 km watershed, which can be used as an analogue for improving the scientific basis and developing linkages in the areas of multimedia environmental modelling (ISCMEM, 2011). The focus of this effort is on:

- parameter estimation and modelling uncertainty (Nicholson and Hill, 2011);
- site characterisation and monitoring;
- human health risk and safety;
- loss of natural resources;
- clean-up and evaluation of remediation technologies on a large scale.

Figure 6.12: ^{137}Cs concentrations in the watershed of the Chernobyl cooling pond

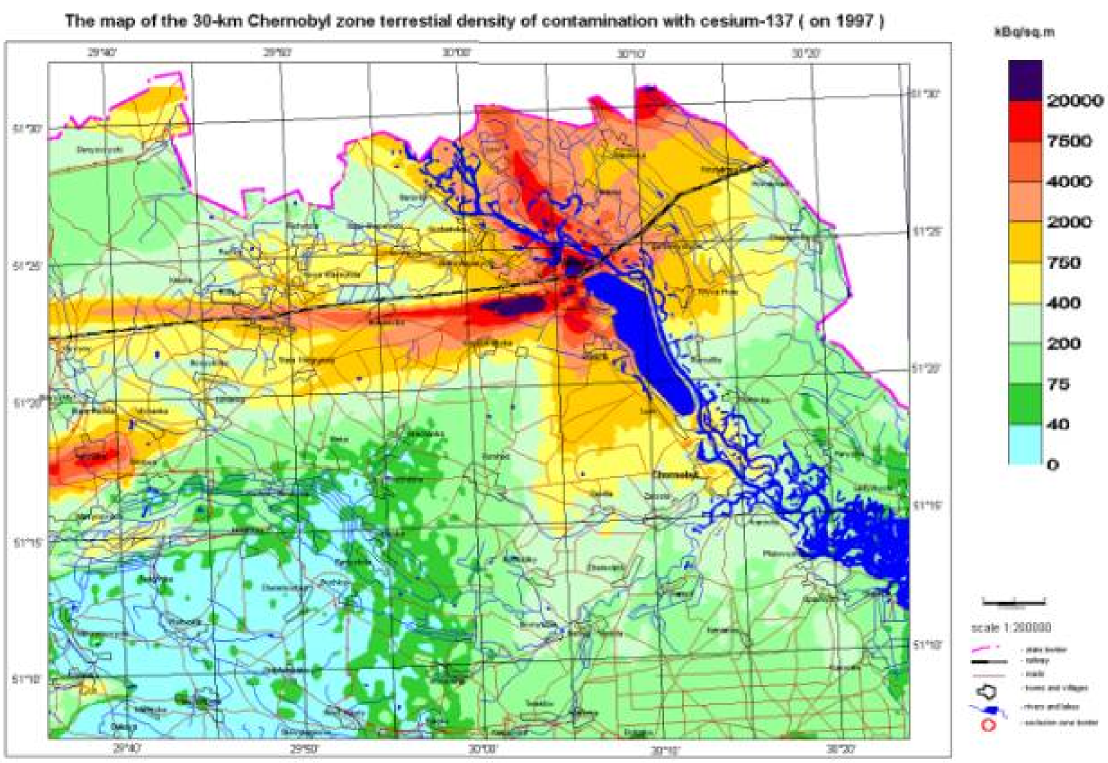
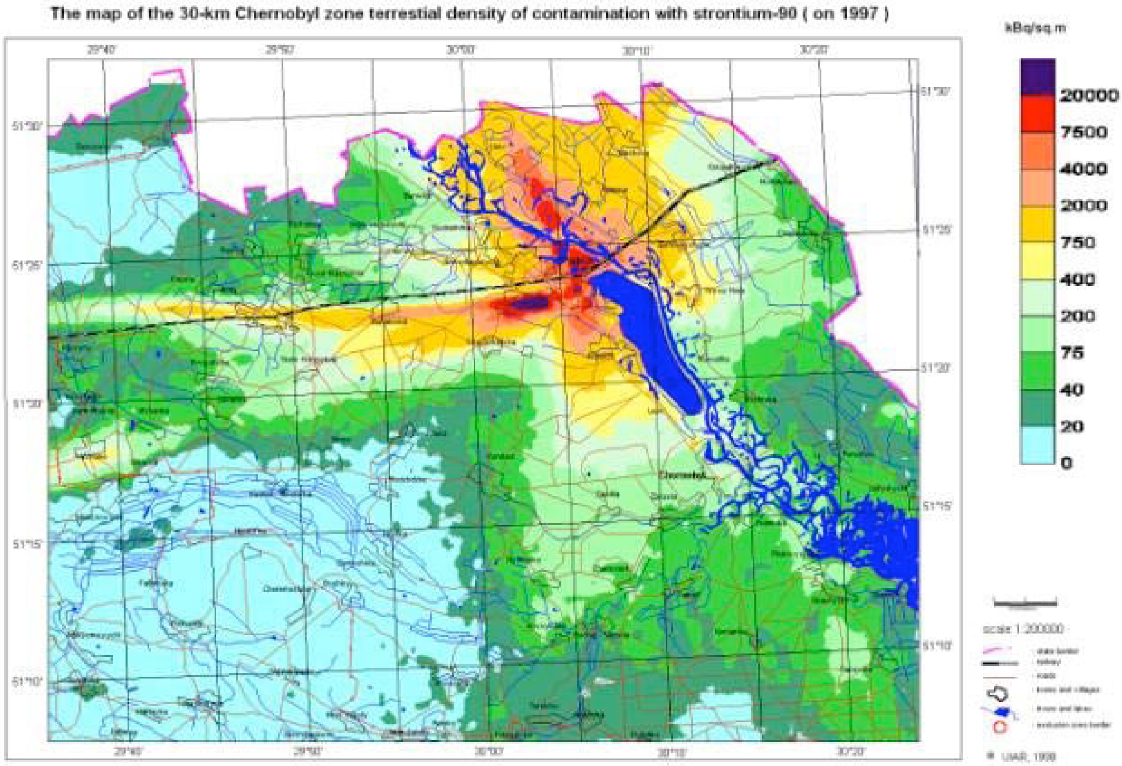


Figure 6.13: ^{90}Sr concentrations in the watershed of the Chernobyl cooling pond



If endorsed by ISCMEM, the Chernobyl cooling pond international case study will provide the opportunity for collaboration between different federal agencies and international organisations in accomplishing their missions associated with predicting the post-accident, long-term behaviour of radionuclides and remediation of radioactively contaminated sites (ISCMEM, 2011). The development of more powerful and accurate fate and transport models will also be required to estimate risks and prepare strategies for remediation of the Fukushima site (Ebihara, Yoshida and Takahashi, 2012).

Future suggested R&D for characterisation using statistical sampling and 3-D modelling

- ***Description*** – Research and development is required to develop methods, software applications and interfaces for instrumentation that allow geostatistical conceptual site models to be more efficiently developed and to minimise scanning and sampling required to achieve the statistical confidence necessary for final site clearance. This includes development of software that is scalable, depending on the complexity of the contaminants' distribution and the hydrogeological and environmental interactions at decommissioning sites.
- ***Objectives*** – The objectives of this R&D should be to expand the site closure criteria and statistical verification beyond the current, limited applications to building and soil surfaces and to integrate subsurface modelling and statistical confidence into risk estimates for final site clearance. Advances are also needed to more efficiently collect and upload data-supported conceptual site model development, and to use geostatistical modelling to meet acceptable confidence levels while minimising the scans and sampling required. Applications that are scalable from relatively simple contaminant distributions and environmental systems to large, complex systems where an accurate understanding of the fate and transport of the contaminants is required to ensure clearance criteria meet the site release risk objectives.

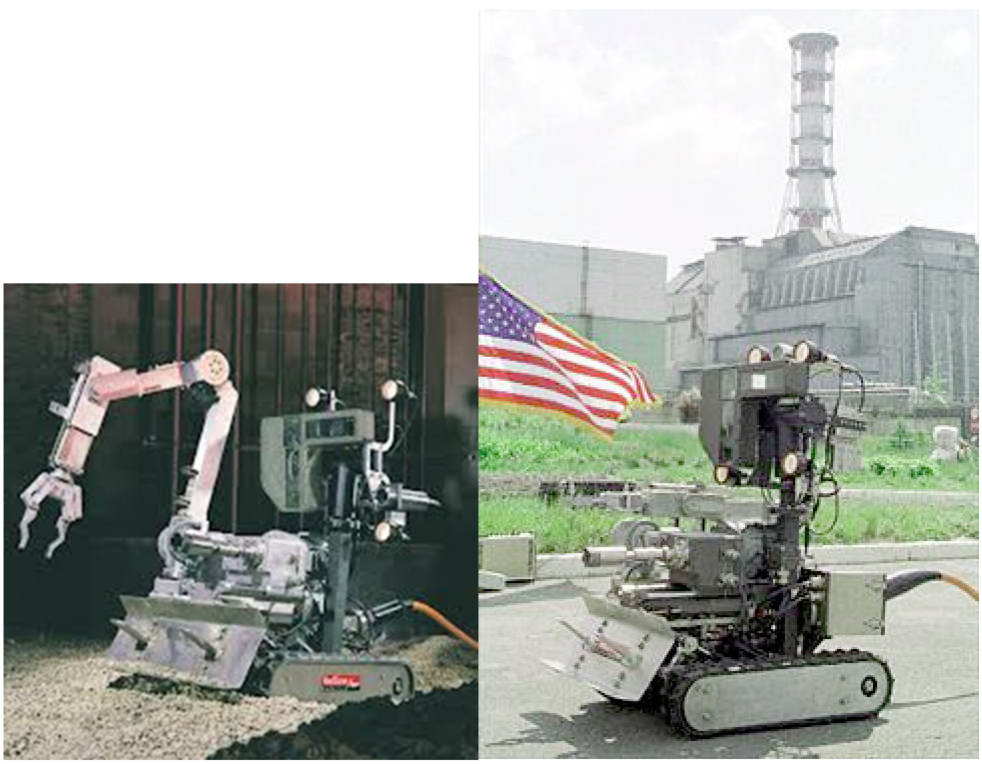
- ***Desired deliverables*** – Software, instrumentation and methods to incorporate subsurface and non-surficial contamination into site clearance criteria and risk assessments and to optimise the surveys and sampling required to meet the desired confidence levels for clearance. Improved scalable modelling of complex contaminant distributions and the fate and transport of radiological and non-radiological contaminants in complex natural systems.

Use of robotic techniques for sampling

Challenges

The current ongoing example of the challenges associated with sampling and surveying in high dose rate, hard-to-access areas is the recovery effort at Fukushima, which has relied heavily on robotics due to the hazardous environment created by the hydrogen explosions in the plants and the high radiation levels still present. However, robots were used at Chernobyl to access hazardous areas and to sample and monitor highly radioactive hot cells and storage tanks. The Pioneer, a robot developed by the DOE, NASA and RedZone Robotics and used for reconnaissance and sampling at Chernobyl's reactor 4 building in 1999, is a radiation-hardened remote reconnaissance system for structural analysis (Pioneer Project, n.d.).

Figure 6.14: The Pioneer robot developed for Chernobyl



Source: CPEO (n.d.).

The major functional capabilities Pioneer consisted of:

- a teleoperated mobile robot for deploying sensor and sampling payloads;
- a mapper for creating photorealistic 3-D models of the building interior;
- a core borer for cutting and retrieving samples of structural materials;
- a suite of radiation and other environmental sensors.

The Three Mile Island nuclear energy facility in Pennsylvania used six robots over a ten-year period for its recovery efforts. Since Fukushima, other companies such as Progress Energy are pursuing using robots as part of their day-to-day operations to better protect plant workers (Nuclear Energy Institute, 2012).

In an article about the Fukushima recovery Yoshino noted, "While only wired communication was used at Three Mile Island, today's robots have wired-wireless hybrid communications… Advancements in communication technologies have enabled the robots to send large amounts of data, allowing us to get more information than in the past." (Nuclear Energy Institute, 2012)

The R&D Engineering section at Savannah River National Laboratory (SRNL) engineers, integrates, tests and supports deployment of custom robotics, systems and tools for use in radioactive, hazardous or inaccessible environments. Mechanical and electrical engineers, computer control professionals, specialists, machinists, welders, electricians and mechanics all adapt and integrate commercially available technology with in-house designs to meet the needs of the Savannah River Site (SRS), the Department of Energy (DOE) and other governmental agency customers (Tibrea, et al., 2011).

The challenge is to build upon the experience being gained at Fukushima, and develop robotic capabilities that can be used for sampling in hazardous situations that take advantage of autonomous and semi-autonomous advances, as well as future and existing 3-D mapping, direct sampling and contaminant imaging capabilities. It is likely that a suite of robotic capabilities for constrained access to tanks, rooms, hot cells and for underwater survey and sampling will be necessary (Nuclear Energy Institute, 2012). An equal challenge will be to get robots deployed on actual decommissioning projects such that experience can be gained, operators can be trained, procedures and protocols developed, and a continuous improvement cycle can be implemented (Winfield, 2013).

Summary of current R&D for use of robotic techniques for sampling

- The 3-D GammaModeler™

The Three-Dimensional (3-D) GammaModeler™ visual and gamma ray imaging system remotely surveys large areas for gamma-ray emissions and displays the results in 3-D representations of the radiation sources. The 3-D capability of the GammaModeler™ allows the radiation environment inside an object to be determined. The system consists of four modules: a sensor head, a portable personal computer, a pan-and-tilt controller and a 3-D workstation. The sensor head is controlled remotely by the computer. Remote operation and control of the sensor head minimises operator exposure (CPEO, n.d.).

- Robotic sampling and survey at Fukushima Daiichi

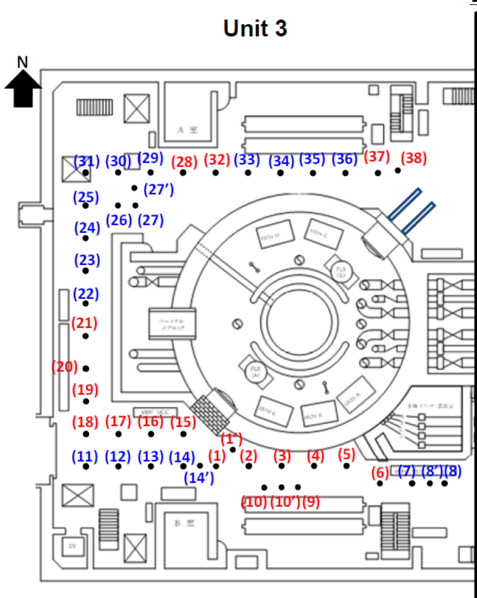
Robots were first deployed at Fukushima just weeks after the accident. Four robots were deployed by Massachusetts-based company iRobot, two PackBot 510s and two Warrior 710s designed for battlefield operations but modified for use at Fukushima. The PackBot came equipped with a full HazMat kit, which enabled it to measure temperature and detect gamma radiation, explosive gases and toxic chemicals, and feed that data to TEPCO controllers in real time (Nuclear Energy Institute, 2012; Kawatsuma, Fukushima and Okada, 2012; ASME, 2012).

Approximately three months after the accident, TEPCO brought another robot, the Quince, to the site to assist with the recovery effort. Experts at Japan's Chiba Institute of Technology, Tohoku University and the International Rescue System Institute jointly developed this robot. It is highly mobile and has been used for dust sampling as well as radiation dose and temperature measurements inside the facility's reactor buildings. Experts at the Chiba Institute of Technology performed several tests on the robot before sending it to Fukushima, including making it navigate a stairway built to mimic the conditions of those in the facility's reactor buildings, fall off a table more than 400 times to see which part of the robot broke first and crash into walls to see how resistant it was to shock (Nuclear Energy Institute, 2012).

During a redesign project to equip the robot for disaster response missions, TEPCO gave the institute two specific missions, one of which was to explore the inside and outside of the reactor buildings to perform dose rate measurements (Nuclear Energy Institute, 2012). The second mission TEPCO had for Quince was to sample contaminated water and install a water gauge in the basement of the reactor buildings. To succeed in the above two missions, the Chiba Institute redesigned Quince, and performed repeated operational tests to improve it. Finally, one of the robots was delivered to the Fukushima Dai-ichi nuclear power station in June 2011 (Nagatani, et al., 2011).

The Quince is the only unmanned vehicle that has been able to climb the narrow 90-cm-wide, 40°-steep stairways inside Fukushima's reactor buildings to measure radiation levels where the spent fuel pools are located. TEPCO used the data and video taken by Quince on the top floor of Unit 2, where radiation levels were still too high for workers to enter, to prepare for the removal of rods from the spent fuel pool. TEPCO used the first Quince at the site for five months before it became stranded inside Unit 2 due to a loss of communication caused by a tangled cable. Engineers modified the next two robots, fitting them with a fully automated wire reel system to rewind their 500-metre-long cables. Quince 2 also has a remote dust sampling system and Quince 3 has a 3-D laser scanner. The robots are designed to withstand more than 200 Sv of radiation, and can move easily over uneven debris. The Quinces are still being used at Fukushima, collecting critical data from difficult-to-reach areas, and TEPCO continues to look for innovative ways to use robots in its recovery efforts.

Figure 6.15: Fukushima Daiichi Unit 3 first floor reactor building results of robotic surveys



Dose rate measurement result *50mSv/h or more: indicated in red [Unit: mSv/h]

Point	5cm above the floor	150cm above the floor	Point	5cm above the floor	150cm above the floor
[1]	82.1	44.7	[19]	94.0	98.7
[1']	74.3	51.6	[20]	73.2	65.3
[2]	97.4	78.0	[21]	68.8	49.2
[3]	89.1	73.5	[21] Wall	65.4	42.4
[3] Wall	72.3	77.9	[22]	40.0	35.1
[4]	81.7	91.4	[23]	35.2	24.1
[5]	65.5	64.9	[24]	21.7	21.4
[6]	60.0	25.7	[25]	33.5	24.5
[7]	15.9	16.2	[26]	21.7	24.9
[8]	15.7	15.8	[27]	21.7	27.5
[8']	18.5	17.8	[27']	17.7	24.4
[9]	87.8	85.2	[28]	73.0	35.5
[10]	91.5	74.5	[29]	24.1	25.7
[10']	92.5	95.8	[30]	18.2	27.8
[11]	66.8	69.9	[31]	31.7	34.1
[12]	29.2	53.8	[32]	63.8	36.7
[13]	46.2	57.6	[33]	36.7	40.1
[14]	30.3	44.1	[34]	29.4	37.5
[14']	27.7	41.3	[34] Wall	27.1	37.4
[15]	69.8	41.3	[35]	27.7	41.7
[15] Wall	53.7	40.4	[36]	47.7	72.5
[16]	50.6	40.6	[37]	203.1	124.7
[17]	180.9	57.4	[38]	59.7	85.2
[18]	102.0	79.8	–	–	–

東京電力

Another robotic device used extensively for sampling and surveying at Fukushima was the Honeywell T-Hawk. The T-Hawk is a small helicopter-type device that flew over 40 missions in 3 months to assist TEPCO and authorities with operations in and around the facility. The T-Hawk allowed the nuclear engineers to see into the reactor buildings (in real time) from angles that an overhead satellite or plane could not see. The videos were very useful in determining the condition and structural integrity of the reactor buildings, but probably the most valuable data was from the radiological sensor mounted on the T-Hawk. It gathered data as the robot flew at low altitudes and next to the reactors, and the data was then assembled into highly accurate maps. The T-Hawk actually hovered in a plume rising from one of the reactors, providing direct sampling (Murphy, 2012).

The DOE's Idaho National Laboratory modified one of its TALON robots specifically for the Fukushima response. The TALON was outfitted with radiation-hardened cameras, GPS, night vision, and chemical, biological, radiological, nuclear and explosive detection kits. With these kits, it can identify more than 7 500 environmental hazards and has sensing capabilities from up to 3 280 feet. The TALON has also been used in military operations in Iraq and Afghanistan, and for decontamination at New York City's Ground Zero.

Robots were used to investigate the radiological conditions of the reactor buildings and acquire data useful for the "development of remote decontamination technology". Robotic investigations of radiation sources and dose rates have used a gamma camera and dosimeters, and have collected samples of dust, peeling paint and core drillings (TEPCO, 2012).

Figure 6.16: Gamma camera images of Fukushima Unit 3 reactor building first floor

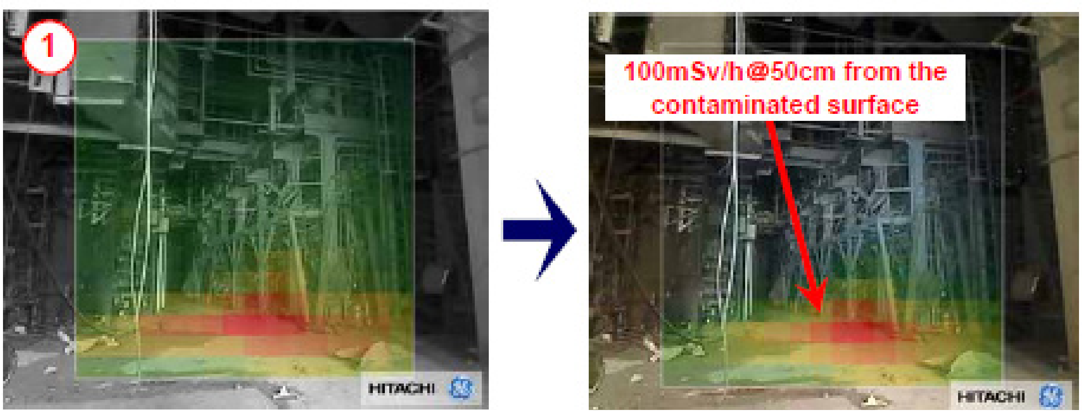


Image acquired at the large carry-in entrance before BG correction (Angle of elevation: 10 degrees)

Image acquired at the large carry-in entrance after BG correction (Angle of elevation: 10 degrees)

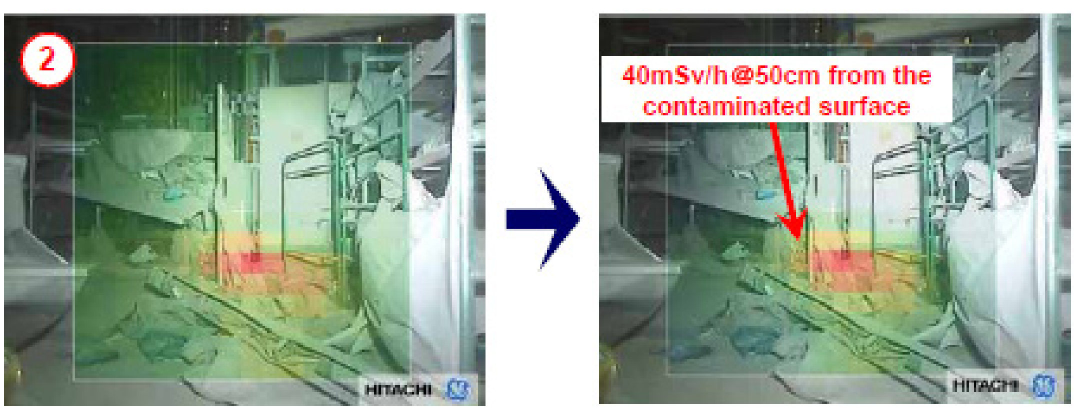


Image acquired at the northwest corner before BG correction (Angle of elevation: 0 degrees)

Image acquired at the northwest corner after BG correction (Angle of elevation: 0 degrees)

This sampling was performed to:

- confirm the validity of the sample (stable caesium: ^{133}Cs) used for decontamination testing in terms of the amount attached on the surface and the permeability;
- understand the contamination conditions (elemental composition) of each unit in order to select the optimum decontamination method.

Gamma-emitting nuclide analyses, beta measurements and alpha measurements of the contamination samples (dust, paint and concrete core) were obtained using an imaging plate (sensitive film) to confirm the level of contaminant permeability into the concrete core as well as the range of contaminant spreading in the planar direction. The sample surface was also studied with an electron microscope and the chemical composition was studied with an elemental analyser in order to confirm caesium and its distribution on the border between the paint and concrete and in cracks (TEPCO, 2012).

The DOE has listed the following gamma detection techniques as having remote characterisation value and deployment capability (CPEO, n.d.).

- RadScan 600 gamma-ray imaging system

The RadScan 600 gamma-ray imaging system was developed by British Nuclear Fuels Ltd. (BNFL). The RadScan system characterises contaminated sites containing high levels of surface radiation at a 12-inch distance. This system provides real-time data on the location and concentration levels of gamma radioactive material. The RadScan 600, with a single detector, employs spectroscopy to identify contamination and exposure level information along with isotopic information for all surfaces surveyed. Since the inspection head is operated remotely, worker exposure and access constraints typically associated with traditional hand-held survey instrumentation are minimised (CPEO, n.d.).

- *In situ* gamma spectroscopy with ISOCS

ISOCS is a complete *in situ* object counting system developed for use in a wide variety of measurement applications. Most radiological contamination situations do not result in uniform deposition of the contaminant material. Consequently, the selection of a small sample set to send to the laboratory is a difficult and imprecise task. One solution is to take very large samples and average them over the entire object or area. The gamma radiation detector uses a high-purity germanium crystal for high resolution and high efficiency as it identifies radioactive isotopes and provides real-time assays of the radioactive contents of containers, surfaces and samples. The system provides traditional spectra of counts as a function of gamma energy, which are then converted to radionuclide concentration using a software system. The entire system is mounted on a portable cart that allows for the rotation of the detector on a horizontal axis. The ISOCS does not produce an image (CPEO, n.d.). ISOCS have been used to support nearly all decommissioning efforts since the early 90s in the United States and most recently at Zion to assay 28 m^2 areas of un-impacted lands to verify there were no plant-related radionuclides in the un-impacted survey units.

- Remote mapping of gaseous contaminant plumes

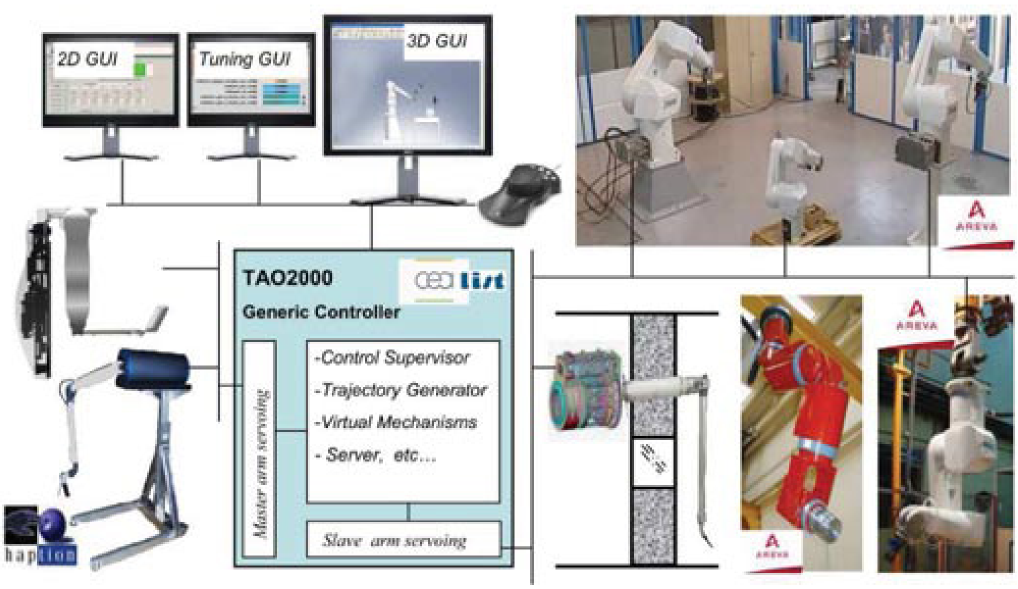
Recent work on the remote mapping of hazardous gas plumes based on infrared spectroscopy using imaging Fourier transform spectrometers may allow for better detection and resolution of airborne contaminants (Harig, et al., 2009). The method employed is based on the analysis of infrared radiation absorbed and emitted by the molecules of the clouds. While this is applicable to non-radiological contaminants identified by infrared spectroscopy, a similar method based on gamma emissions or ionised nitrogen ultraviolet emissions may be viable for remote real-time assay and monitoring of plumes containing radioactive materials. The result of the infrared spectrum analysis of the hyperspectral data, the so-called "hazardous cloud" image, is displayed by an overlay of the plume's image on the image of a video or infrared camera.

- CEA LIST computer-aided teleoperation

Fifteen years ago, the French Atomic Energy Agency Interactive Robotics Laboratory (CEA LIST) and AREVA formed a joint venture for an ambitious R&D programme in robotics and remote handling technologies as applied to France's spent fuel management facilities in order to cover the requirements of its different plant life cycles. CEA LIST is in charge of the conceptual studies and development of prototypes, and AREVA is in charge of the specifications and industrialisation of developments. Robotic developments at AREVA are applied to operational plants as well as those in decommissioning, e.g. at production plants for carrier systems, for welding and contamination checking, and for repairing, inspecting or clean-up (Geffard, et al., 2012).

Robotics systems can use either off-the-shelf industrial robots or devices specifically designed for teleoperations. As such, robotic intervention systems may be mounted either through the wall or on vehicles to fit different types of hot cells. Development carried out by CEA LIST and AREVA has focused on technological components that can be used in different types of systems. Some of them are now commonly used for maintenance operations at the AREVA NC (nuclear cycle) La Hague reprocessing plant. Since the first maintenance operation in 2005, several other successful interventions have been realised using the industrial MA23/RX170 telemanipulation system. Another robotic system under development since 2010 is the through-the-wall telerobot named MT200 TAO, TAO being a French acronym for "computer-aided teleoperation". It was based on the slave arm of the MSM MT200 and has been evaluated in an active production cell at the La Hague plant. Although these evaluations are ongoing, the positive results obtained have led to an update and industrialisation programme. All these developments are based on the same generic control software platform, called TAO2000 V2. It is the second release of the CEA LIST core software platform dedicated to computer-aided force-feedback teleoperation (Geffard, et al., 2012).

Figure 6.17: TAO2000 controller software architecture



Source: Geffard, et al. (2012).

TAO allows a distant operating person to feel the strength exerted by the slave arm or robot. This technology is implemented though TAO2000 and the telerobotics systems using motorised robots as master and slave arms, designed and qualified to respond to the LaHague hot cells' constraints and needs (Geffard, et al., 2012).

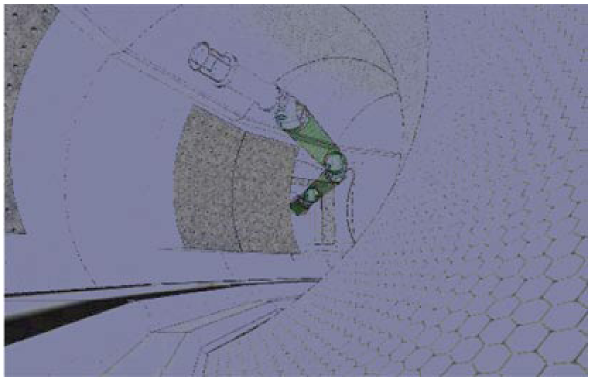
- CEA LIST long-reach manipulators

The Interactive Robotics Laboratory of CEA LIST is in charge of the development of remote handling technologies to meet energy industry requirements. It has developed advanced robotics systems for inspection or light intervention in hazardous environments with limited access, such as blind hot cells in the nuclear industry or the thermonuclear experimental Tokamak fusion reactor.

Long-reach manipulators, also called snake arm robots, are needed in various fields such as space, energy or medical domains to perform tasks that are usually unreachable for humans. For instance, manipulators can be used in inspection, maintenance and safety tasks under hazardous operation conditions at nuclear sites (Perrot, et al., 2012). The CEA LIST robotics team has more than 30 years of experience in force-feedback remote handling system design and control for application of robotics in hazardous environments.

Inspection robotics started in the late 1990s when AREVA NC identified the need for blind hot cell inspection tasks and light interventions with a manipulator. The requested system must be manoeuverable through horizontal, small-diameter wall engineering penetrations in a wide range of cells. To meet these requirements, CEA LIST developed a very challenging long-reach robotic carrier called PAC. This 6-m-long carrier with 11 joints weighs less than 30 kg and has a 1 kg payload. It is actuated by electrical motors and includes on-board hardened control electronics qualified up to 10 kGy. It can be remotely operated by means of a control system that includes a graphical user interface providing a virtual three-dimensional display as well as online collision avoidance capabilities and real-time dynamic simulation. This allows intuitive driving of the arm around obstacles (pipes, tanks, vessels, etc.). Another industrial robot (called LORA for long-reach arm) is currently under development. It will be a 9-m-long and made of 7 modules with 15 actuated joints.

Figure 6.18: PAC robotic long-reach manipulator, 3-D control and monitoring



In parallel, based on this experience, the CEA LIST and CEA IRFM (*Institut de recherche sur la fusion par confinement magnétique*) laboratories have developed a manipulator called the articulated inspection arm (AIA) for mini-invasive operations in nuclear fusion facility vessels. This project is being developed for remote handling activities for the next step of fusion reactors. Performed under a European Fusion Development Agreement work programme, the aim is to demonstrate the feasibility of close inspection (e.g. viewing and leak testing) of the diverter cassettes and the vacuum vessel first wall of the International

Thermonuclear Experimental Reactor (ITER). To carry out an intervention in a short time after plasma shutdown, the operation of the robot should be performed under Tokamak conditioning, i.e. high vacuum (10-6 Pa) and temperature (120°C) conditions. Both the PAC and the AIA must meet severe specifications: small penetration diameter, minimum reach of 6 m, high dexterity to move in constrained environments and many degrees of freedom for obstacle avoidance. These robotic systems are designed as multi-link carriers that must support significant weight, so they have similar architectures and designs and can carry the same process tools.

Figure 6.19: Long-reach snake arm PAC robot assembly view

Figure 6.20: Introduction in the cell (left) and pipe avoidance during inspection (right)

In view of future applications, long-reach robots could be assigned two types of missions: either in inspection (passive applications) or as carriers for intervention tools or diagnostics (active applications). Anticipating the prospect of in-vessel intervention needs for ITER, CEA has already identified interchangeable devices (diagnostics and tools) to be plugged in at the front head of a robotic arm. This covers applications such as vision camera, water leak localisation devices, diagnostics calibration and inner-component characterisation by laser systems (Perrot, et al., 2012). A laser ablation end-effector is also available. Plasma-wall interaction phenomena induce dust production and hydrogen trapping during pulses at fusion reactors; a laser treatment and diagnostic system are under development for the inventory and removal of these elements. An ytterbium fibre laser will be used for film ablation and for recovery of trapped tritium. At the same time a chemical analysis of the co-deposited layers can be done using the laser-induced breakdown spectroscopy analysis technique.

In summary, universal, long-reach carrier technology for inspection in nuclear environments has been designed, manufactured and successfully tested. Industrialisation of an AREVA inspection robot is beginning and the first LORA prototype is currently under procurement. These robots are based on an innovative, parallelogram architecture and an intuitive control mode using real-time simulation in a 3-D graphical environment. Future advanced tools are in development and further progress is being made with regard to robot modelling, motion simulation and geometric calibration, taking structure flexibilities into account. Operator assistance for complete robot monitoring will be increased due to the addition of high-level functions in 3-D supervision software and anti-collision management.

Figure 6.21: Deposited carbon on bottom limiter of Tore Supra; laser ablation and vacuum design

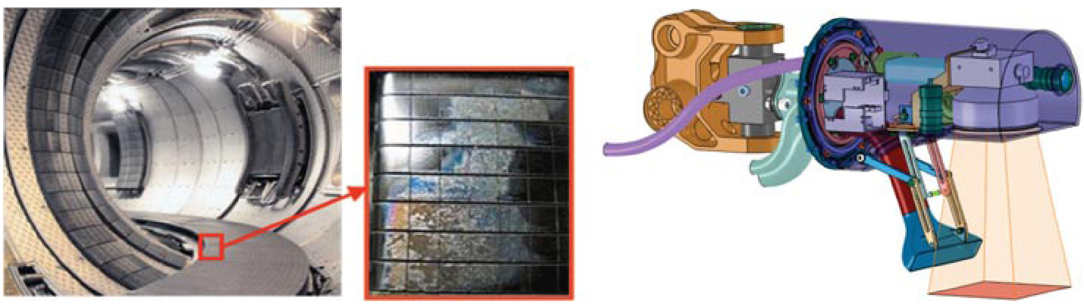


Figure 6.22: AIA robot inspection in Tore Supra Tokamak during air operation (left) and window view during operation under vacuum and at temperature (right)

- Spent fuel pyroprocessing cell trolley mounted servo manipulator system

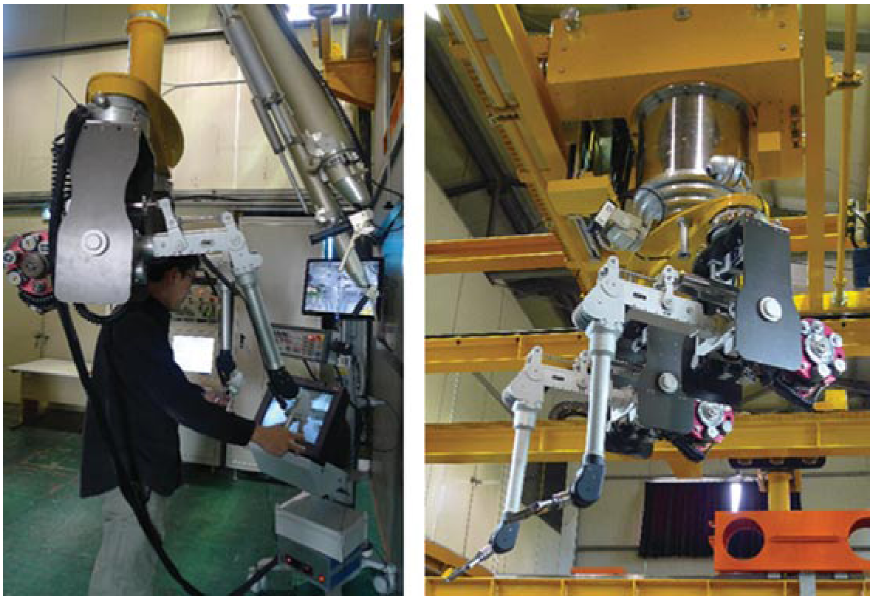
Korea Atomic Energy Research Institute (KAERI) has been developing technology for pyroprocessing, which is considered one of the most promising options for future nuclear cycles in the Republic of Korea. Pyroprocessing is a process for separating transuranics

(TRU) such as Pu, Np and Am from spent fuel in a high-temperature molten salt bath. Once separated, they can be transmuted in a fast reactor to produce even more fissile fuel for power generation. Thus, pyroprocessing reduces the amount of spent nuclear fuel and also dramatically decreases the disposal load through recycling and the elimination of toxic waste, particularly long-lived fission products in spent nuclear fuel (Buehler, Pouliot and Montambault, 2012).

Pyroprocessing is composed of several unit processes: voloxidation, electrolytic reduction, electrorefining, electrowinning and salt waste treatment. From 1997 to 2006, KAERI completed the Advanced Spent Fuel Conditioning Process (ACP) project, which focused on the development of an electrolytic reduction process to convert spent oxide fuel into a metallic form while separating the high-heat sources such as Cs and Sr from the U metal. This reduces the total heat, volume and radioactivity of the metal ingot to about a quarter of that of the spent fuel. To move one step forward toward successful demonstration of pyroprocessing technology, an integrated demonstration facility that includes all major unit processes is needed. To this end, KAERI completed the design of such a pyroprocessing demonstration facility, named the Pyroprocess Integrated Inactive Demonstration (PRIDE) facility.

PRIDE has an argon-atmosphere cell where all the operation and maintenance of process equipment must be performed remotely through master-slave manipulation. Conventional mechanical master-slave manipulators used in hot-cell facilities are limited in terms of workspace and payload due to their mechanical design and power transmission mechanism. Such conventional manipulators alone would not be sufficient in PRIDE. In the conceptual design stage of the PRIDE facility, process development researchers required a servo manipulator system with a handling capacity of 25 kgf, which is a design constraint on the maximum weight of the unit module of process equipment for maintenance, as well as with a dual-arm configuration for dexterous manipulation. With this motivation, KAERI developed a bridge-transported, master–slave, dual arm servo manipulator system as a part of the project (Lee, et al., 2012).

Figure 6.23: Bridge-transported bilateral servo manipulator system installed in RHEM

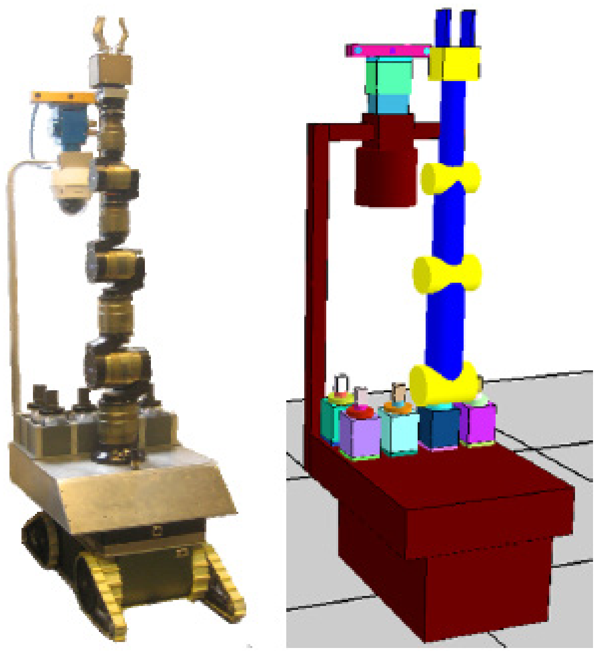


In addition to the commercial MSM telemanipulation systems, a specially-designed system called the BDSM, which is mounted on tracks inside the argon cell, is also provided for complete access to all areas in the cell. The BDSM consists of dual arm master manipulators, dual arm slave manipulators, a bridge transport system for the slave manipulators and a master transporter. The bridge transport system is composed of a trolley, a gantry and telescopic boom, and it is configured for the positioning and orientation of the slave manipulators. It also has redundant drives to allow movement to a maintenance area when drive failure occurs (Lee, et al., 2012).

- Autonomous robot for hazardous site sampling and investigation

An autonomous software system called CORBYS (2011), consisting of cognitive system modules, situation awareness architecture, self-organising information anticipatory architecture, etc., has been used with a reconnaissance robot for a demonstration project of a robotic system for examining hazardous environments. The autonomous robotic system that can be used for inspection of contaminated/hazardous environments was used to evaluate the genericity of the CORBYS cognitive techniques. The robot, RecoRob, was developed within a German national project "A Mobile Reconnaissance Robot for Investigation of Hazardous Environments – RecoRob)". The RecoRob is a mobile, outdoor robotic system designed for handling samples in unstructured hazardous/contaminated environments. It consists of a robot arm with 7 DOF (degrees of freedom) that is mounted on a mobile platform. The main focus of this project was the development of robot skills for autonomous sample drawing using dexterous manipulation while the robot is remotely navigated by a human operator. The robot is equipped with sensors for environment perception as well as with sensors for platform navigation and robot arm control. In the CORBYS project, two experimental set-ups will be designed in which the robot works in a team with a human to investigate contaminated/hazardous environments. Both experimental scenarios of the second demonstrator will be designed to test different functionalities of the CORBYS cognitive control architecture with an emphasis on alternating the human-robot lead-taking in exploratory scenarios (CORBYS, 2011).

Figure 6.24: RecoRob robotic system as hardware and mapped virtual reality



RecoRob uses a mobile ASENDRO platform from Robowatch (Robowatch Industries Ltd.), which consists of a variable drive system equipped with chains supported by swing arms capable of continuous rotation, enabling the platform to climb stairs and overcome obstacles. The system components to be actuated are: a SCHUNK 7 DOF lightweight robot arm for the object manipulation and a 2 DOF pan-tilt-head for steering the vision system.

The sensor system consists of an ATI Technologies force-torque sensor in the manipulators wrist, a PointGrey Bumblebee XB3 stereovision camera for 3-D environment reconstruction, a Samsung SNC dome camera for workspace observation and an NEC ThermoTracer IR camera for thermal inspection of the working area. For the computational power, there are two SPECTRA NISE 3140P2E industrial computers, and for communication a D-Link DAP-2590 access point. Additionally a NAVILOCK NL-302U GPS receiver is used for the aspired navigation and localisation purposes. The operator interaction is realised with a Getac M230N-5 rugged notebook, including wireless LAN and a touchscreen for intuitive operator input. There are two scenarios being tested (CORBYS, 2011):

- *Augmented teleoperation:* The robot is teleoperated by the human so that it is sent into the contaminated area to collect samples used to determine contamination levels. The robot's primary task is to follow the operator's instructions for navigation and manipulation. However, if communication fails, the robot has to be able to take the initiative in completing the task, despite the loss of human command. Two capabilities are considered critical:
  - *Sporadic navigation extrapolation*, when it is assumed that the communication is sporadically interrupted due to interferences at three-second intervals. The robot should use previous commands from the human operator and recorded sensory inputs to estimate the trajectory for the next few seconds, until the communication returns.
  - *Balance loss prevention*, when the CORBYS cognitive modules should detect dangerous tilting of the robot and enable a reflexive response by stopping the robot or moving it back toward the operator. In this way the module will endow the robot with the cognitive capability of reasoning to "veto" dangerous human operator commands in order to avoid tipping over (CORBYS, 2011).
- *Robot as a co-worker:* The robot works as a transportation robot helping the human to carry containers with collected samples. At the beginning of the investigation mission, the robot follows the human partner, keeping a constant distance between them. Based on sensory information (e.g. robot vision sensors and human, body-wearable sensors such as an inertial measurement unit), the robot analyses the human's behaviour and deduces the human's goal. If there is an unexpected change in human behaviour, such as a change in direction of movement indicating human intention to approach the robot, the robot has to change its behaviour so as to stop and allow the human to place the containers with the collected samples onto the robot (CORBYS, 2011).

One of the core functions of the RecoRob software architecture is sample handling. Therefore, a sample manager has been developed to control the inventory of sample containers and their current state. At the beginning of a new sample drawing task, the available sample containers are checked and an appropriate type is chosen. Each sample type has its own unique manipulation sequence defined and saved in the system. The only missing information for successful task execution is the target location which is defined by the operator. This procedure relates to one of the major project goals, that being the simplicity of the procedure: A maximum of autonomy in sample handling will be achieved using a minimum of operator inputs. The only essential input is related to the cognitive capabilities of the user, deciding where and what kind of sample needs to be drawn. All further information is either known *a priori* or can be obtained through sensory data input (CORBYS, 2011).

Future suggested R&D for use of robotic techniques for sampling

- ***Description*** – Research and development is required to continue to develop robotic sampling and survey capabilities for high radiation or hazardous environments. This includes both permanently installed systems for processing of high activity materials, such as spent fuel and irradiated graphite and hardware, and temporary, mobile and modular systems such as those used at Fukushima. Development of autonomous or semi-autonomous capabilities for situational awareness, navigation and sample collection should be explored to provide more robust capabilities.

- ***Objectives*** – Develop installed and modular robotic capabilities that can function semi-autonomously to sample and survey hard-to-access and/or hazardous environments, such as hot cells, system and component internals or activated material waste treatment enclosures. Capabilities should include *in situ* analyses such as gamma cameras, and sample collection for *ex situ* analysis.
- ***Desired deliverables*** – Radiation hardened, robust robotic platforms that are highly adaptable to survey and sampling situations encountered during decommissioning. These can include track, trolley, rail or permanently installed systems with robotic arms or extended reach systems. These devices should be capable of performing various survey and sampling tasks in a semi-autonomous manner.

Use of remote sensing and satellite technologies for characterisation and environmental monitoring

Challenges

The major challenges associated with this topic are the identification, modification and use of the rapidly developing, diverse and multi-faceted technological advancements in this area. Their integration into adaptable sustainable technologies and platforms for decommissioning that will not rapidly become obsolete and irrelevant is a major challenge. *Challenges in Green Environmental Chemistry*, a 2011 book published by the Royal Society of Chemistry (RSC), provides an excellent explanation of the technologies and challenges associated with remote sensing, direct detection and teledetection systems for environmental hazards that have analogies for nuclear decommissioning. Such capabilities revolve around direct detection of *in situ* contaminants that are reported through telemetry or remote sensing by way of satellite or autonomous/semi-autonomous vehicles. While these technological adaptations are promising and interesting, they are extensions of an antiquated framework that will be altered and absorbed by another new, pervasive and powerful development – the Internet of Things (IoT) (Sundmaeker, et al., 2010).

The premise of the Internet of Things (in line with the Cisco concept of the Internet of Everything) is that almost every object imaginable will become connected to one vast digital network wherein its infrastructure provides a platform for the combination of smart objects (i.e. wireless sensors, mobile robots), sensory networks and human beings, all using different but interoperable communication protocols, to develop a dynamic multimodal/heterogeneous network. Such a network can be deployed in inaccessible or remote spaces (oil platforms, mines, forests, tunnels, pipes, etc.) or in emergencies or hazardous situations (earthquakes, fire, floods, radiation areas, etc.). In this infrastructure, these different entities discover and explore each other and learn to take advantage of each other's data by pooling resources and dramatically enhancing the scope and reliability of the resulting services (Sundmaeker, et al., 2010).

The Cluster of European Research on the Internet of Things (CERP-IoT) has around 30 major research initiatives, platforms and networks working on projects identifying such technologies as radio frequency identification. CERP-IoT states we are just at the beginning and follows the prognostics that predict 50-100 billion devices to be connected by 2020. Environmental monitoring is just one of the wide array of applications that will be revolutionised by this inevitable interconnection and real-time monitoring. Wireless identifiable devices and the use of IoT technologies in green-related applications and environmental conservation are one of the most promising market segments in the future, and there will be increased deployment of wireless identifiable devices worldwide in environmentally-friendly programmes (Sundmaeker, et al., 2010). The day is fast approaching when data acquired in the field will be uploaded and available in real time for use in CSM construction, characterisation and license termination survey packages and in the geostatistical fate and transport models.

Standardisation efforts for radio frequency identification (RFID) and wireless sensor networks (WSN) are considering data rates of up to 1 Mb/s, heterogeneous sensor integration and different frequencies. This will open up new applications with positive impacts on society, such as remote data monitoring in disaster scenarios, ubiquitous connectivity for health monitors in body area networks and wireless broadband for rural areas. Secure communications are also a concern of end users. In the meantime, operators are looking beyond the capital expenditure costs of running RFID networks to minimising operational costs such as power consumption and site costs (installation, integration, maintenance).

IoT and wireless technologies can soon be used to advance the efficiency and effectiveness of numerous city and national environmental programmes, including the monitoring of vehicle emissions to help supervise air quality, the collection of recyclable materials, the reuse of packaging resources and electronic parts and the disposal of electronic waste. RFID can be used to identify electronic subcomponents of PC, mobile phones, and other consumer electronics products to increase the reuse of these parts and reduce waste. RFID continues to provide greater visibility into the supply chain by helping companies more efficiently track and manage inventories, thereby reducing unnecessary transportation requirements and fuel usage. As the CERP-IoT says, the true research work starts now (Sundmaeker, et al., 2010).

Summary of current R&D for use of remote sensing and satellite technologies

- Remote sensing of soil contamination

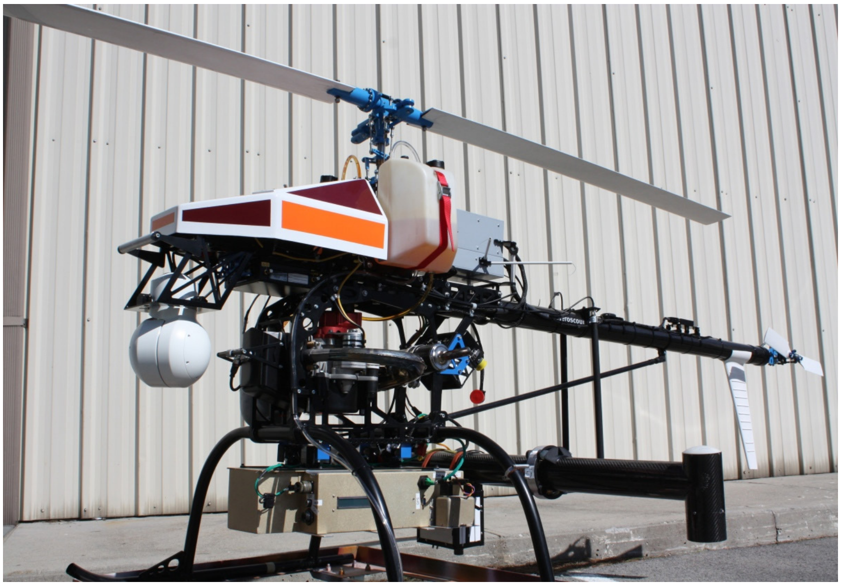
In case of a nuclear accident, decision makers rely on high-resolution and accurate information about the spatial distribution of radioactive contamination surrounding the accident site. However, the static nuclear monitoring networks of many European countries are generally too coarse to provide the desired level of spatial accuracy. In the Netherlands, authorities are considering a strategy in which measurement density is increased during an emergency using complementary mobile measuring devices. This raises the question, where should these mobile devices be placed? This article proposes a geostatistical methodology to optimise the allocation of mobile measurement devices, such that the expected weighted sum of false-positive and false-negative areas (i.e. false classification into safe and unsafe zones) is minimised. Radioactivity concentration is modelled as the sum of a deterministic trend and a zero-mean spatially correlated stochastic residual. The trend is defined as the outcome of a physical atmospheric dispersion model, NPK-PUFF. The residual is characterised by a semivariogram of differences between the outputs of various NPK-PUFF model runs, designed to reflect the effect of uncertainty in NPK-PUFF meteorological inputs (e.g. wind speed/direction). Spatial simulated annealing is used to obtain the optimal monitoring design, in which accessibility of sampling sites (e.g. distance to roads) is also considered. Although the methodology is computationally demanding, results are promising and the computational load may be considerably reduced to compute optimal mobile monitoring designs in nearly real time (Heuvelink, et al., 2010).

Recent events have highlighted the need for unmanned remote sensing in dangerous areas, particularly where structures have collapsed or explosions have occurred, to limit hazards to first responders and increase their efficiency in planning response operations. In the case of the Fukushima accident, an unmanned helicopter capable of obtaining overhead images, gathering radiation measurements, and mapping both the structural and radiation content of the environment would have given the response team invaluable data early in the disaster, thereby allowing them to understand the extent of the damage and areas where dangers to personnel existed. Obtaining situational awareness of the post-event environment requires mapping the radiation distribution of the area and localisation sources of high radioactive intensity. Conventional systems for these tasks require large, heavy, expensive equipment that necessitate the use of a full-size helicopter instead of an inexpensive, easily transportable unmanned aerial vehicle (UAV).

Therefore, the Unmanned Systems Lab (USL) at Virginia Tech has developed a sensing system to fit within the constraints of a small helicopter UAV platform and still complete the required mapping and localisation tasks. The new system is based on the Aeroscout B1-100 helicopter platform, which has a one-hour flight endurance and uses a COFDM radio system that gives the UAV an effective range of 7 km. The total weight of the remote sensing system is 90 kg, including the autonomous helicopter and sensing payloads for the radiation detection and imaging operations. The radiation detector payload is a sodium iodide crystal with associated software and novel search algorithms to rapidly and effectively map and locate sources of high radiation intensity. In addition to this detector, the sensing system also features a stereovision system to generate terrain maps of a region of interest: a DST OTUS-L170 Gimbal camera with laser range finding functionality to geo-locate points of interest, and a Cobham NETNode COFDM radio to provide this functionality at a maximum range of 7 km in scenarios with no sightline. An on-board generator powers the sensing system components while lithium-polymer batteries power the flight controller and communications radio. By incorporating this sensing technology into an unmanned aerial vehicle system, crucial situational awareness can be gathered about a post-disaster environment and response efforts can be expedited. The USL report details the radiation mapping and localisation capabilities of this system as well as the testing of the various search algorithms using simulated radiation data. The components of the system have been flight tested over several years and a new production flight platform has been built to enhance reliability and maintainability (Towler, Krawiec and Kochersberger, 2012).

Similarly, radiation dose rates were mapped by customising an autonomous cleaning robot, "Roomba", equipped with an H8 microcomputer and a scintillation counter to remotely control the vehicle and send measured dose rate data. The data obtained were arranged with position data, and then the distribution map of the radiation dose rate was produced. Manual, programed and autonomous driving tests were conducted and all performances were verified. That is, for each operational mode, the measurements with both moving and discrete moving were tried in and outside of a room. Consequently, it has been confirmed that remote sensing of radiation dose rate is possible by customising a currently marketed robot (Kobayashi, et al., 2012).

Figure 6.25: Sensor system components installed on an Aeroscout B1-100 helicopter



Another paper considered two scenarios for nuclear radiation detection using multiple UAV, from which contour mapping of the nuclear radiation is simulated. Then, for real applications, this paper presents a low-cost UAV platform with built-in formation flight control architecture together with a formulated standard flight test routine. Three experimental formation flight scenarios that imitate the nuclear detection missions are prepared for contour mapping of a nuclear radiation field in 3-D space (Han, et al., 2012).

There are also broader implications for the use of remote sensing technologies for long-term monitoring of contaminated lands and disposal sites. The use of imaging spectrometry offers the potential to monitor the Belarusian landscape at opportune spatial and temporal resolutions. Vegetation has been shown to be an important agent in the cycling of radioactive isotopes in the environment and therefore a useful indicator of radionuclide contamination. This pilot research has focused on assessing the spectral response from Pinus sylvestris (dominant on the Belarusian landscape) at differing ages and with varying levels of ^{137}Cs contamination. Continuum removal was applied to the spectra showing that, for older forests (circa 35 years), significant spectral differences between low- and high-contaminated sites exist at wavelengths that are causally related to foliar biochemicals. This was not the case for young forests (circa 15 years), where no significant differences were found. The results signify the potential to infer contamination levels from a spectra of forests, partitioned by age, thus indicating the possibility of using imaging spectrometry to monitor radionuclide contamination, a possibility warranting further investigation (Boyd, et al., 2006).

Another study investigated the use of hyperspectral remote sensing for characterising vegetation at capped hazardous waste sites. The management of water infiltration into the waste area is a key issue to prevent the migration of the hazardous constituents into the environment. Historically, the vegetation component of a hazardous waste capping system has been viewed as a means to stabilise the surface soils and prevent erosion. However, for some capping systems in arid and semi-arid climates, the vegetative cover has taken an increasingly functional role through the construction of evapotranspiration or water balance cover systems. The vegetative cover and soil systems are constructed to maintain a hydrologic balance, with the vegetation withdrawing water from the underlying soils on an annual basis, thereby minimising deep infiltration. Proper functioning of these types of systems depends on the development and maintenance of a robust plant community that can maintain water withdrawal capacity over the life of the capping system. Some *in situ* remediation strategies are also being implemented whereby the migration of subsurface contaminants is dependent on the water withdrawal capability of vegetation. Considered to be a type of phytoremediation, these strategies may be applicable where subsurface contaminants are potentially mobile, and management of vegetation can result in reduced infiltration and subsequent hydraulic control of the migration of contaminants in soils and shallow groundwater. In all such cases, the maintenance of a high evapotranspiration capacity through well-adapted and healthy plant communities is key to the proper and long-term stabilisation of the wastes.

Monitoring of these systems is commonly conducted by ground level observations by trained professionals and is becoming a significant cost element in the management of such systems. Consequently, there is a growing demand for an efficient and reliable approach to vegetation monitoring at waste remediation and stabilisation sites. Remote sensing technology can provide a cost-effective tool for this type of monitoring in harmony with information obtained from *in situ* investigation (Serrato, et al., 2012).

Using discrete wavelet transforms, [wavelet transforms are based on small wavelets with limited duration (Chun-Lin, 2010)] the spectral parameters of uranium mineralisation factors can be acquired, and the spectral identification pedigrees of typical quadrivalence and hexavalence uranium minerals can established. Furthermore, using hyperspectral remote sensing observation technology, the referenced study developed hyperspectral logging of drill cores and trench. This technology is capable of quickly processing lots of geological and spectral information, and the relationship between radioactive intensity and abnormal spectral characteristics of Fe^{3+} was established. These provided a remote sensing technical basis to uranium geology, and better results have been achieved in Taoshan uranium deposits in south China (Zhang, 2008).

- Remote groundwater monitoring

PNNL staff collaborated with Burge Environmental, Inc. to develop remote sensing systems to measure radionuclides such as ^{90}Sr, ^{99}Tc, ^{129}I, tritium and uranium in groundwater. These systems are capable of automated sample collection from shallow wells or aquifer tubes, sample pre-treatment and delivery of prepared samples to various radiochemical sensor modules. As shown in Figure 6.26, prototype systems have been deployed to various locations at the DOE's Hanford site: the 100-N Area [^{90}Sr (A)], the 200-W Area ZP-1 pump and treat plant [^{99}Tc (B)], and the 300 Area North [uranium (not shown)]. These systems are in varying phases of deployment and operation. PNNL staff and Burge Environmental, Inc. are also collaborating to develop a laboratory prototype of a tritium monitor for proof-of-concept performance testing. Future funding will enable the deployment of this technology into the field (PNNL, 2013).

Figure 6.26: Hanford's prototype ^{90}Sr monitor and ^{99}Tc pump and treat process monitor

The remote sensing systems may be used for remote monitoring of contamination plumes, in a treatment plant or in locations of active *in situ* remediation (e.g. to monitor contaminant concentrations going into and emerging from a remediation transect). Scientific efforts are expected to lead to the automatic uploading of analytical data into a remote server that can continuously perform two- or three-dimensional modelling of the subsurface plumes, or can actively monitor the near-real-time performance of pump and treat plants (PNNL, 2013). The Federal Remediation Technologies Roundtable in the United States also reports developments and information for remediation technologies, including innovative *in situ* monitoring technologies (FRTR, 2013). Another study provides an overview of remote sensing and monitoring techniques to detect leaks of radioactively contaminated water from structures (Sheen, 2012).

- Remote monitoring of airborne contaminants

The Fukushima nuclear accident showed the importance of timely monitoring and detection of radioactive emissions released from nuclear fuel cycle facilities. Nuclear power plants in continuous operation are a stationary source of gas-aerosol emissions that are presented in a ground surface layer persistently. Following radioactive emission, atypical effects can be observed, for example: areas with increased ionisation exhibit an increased concentration of some gases caused by photochemical reactions. The gases themselves and their characteristic radiation emitted in an excited state due to ionisation can be markers of radioactivity and can be monitored by a passive method. Hydrogen atoms and hydroxyl radicals are formed in a radioactive plume by radiolysis of water molecules and other hydrogen-containing air components due to the high-energy electrons from beta-decay of radionuclides. The hydrogen atom and hydroxyl radical can spontaneously radiate at 1 420 MHz and 1 665-1 667 MHz, respectively. A passive method of remote monitoring of radiation levels coupled with dispersion models uses the radiofrequencies of hydrogen and hydroxyl to model radioactive releases from of nuclear power plants. Monitoring the characteristic emission frequencies could allow radioactive concentrations to be monitored remotely in real time rather than relying on fixed-position radiation monitors and air sampling (Kolotkov and Penin, 2012).

Observations of microwave scattering from ambient room air ionised with a negative ion generator measured the frequency dependence of the radar cross-section of ionised air from 26.5-40 GHz (Ka-band) in a bistatic mode with an Agilent PNA-X series (model N5245A) vector network analyser. A detailed calibration scheme was published to minimise the effect of the stray background field and system frequency response on the target reflection. The feasibility of detecting the microwave reflection from ionised air portends many potential applications, such as remote sensing of atmospheric ionisation and remote detection of the radioactive ionisation of air (Liao, et al., 2011; Liao, et al., 2012).

- Remote sensing techniques and enhancement of detection limits

In addition to the remote sensing techniques using satellites and unmanned aerial vehicles discussed above, work is being done to install remote monitoring instrumentation at sites to update information automatically (ISCMEM, 2011). The REASoN project at the University of South Carolina is developing remote monitoring tools for monitoring hazardous waste sites (RSHDSS, 2010). Another project, conducted by the Savannah River Ecological Research Laboratory (SREL), is developing an automated stream monitoring system that responds to transient flow conditions in a manner designed to evaluate the impact of episodic precipitation events on the export of contaminants within the Tims Branch/Steed Pond System (TBSP). Concentrations of uranium (U) and nickel (Ni) are the primary contaminants of concern in the TBSP system. In 2010, an updated version of the online data reporting system was installed on the SREL network (Seaman, et al., 2011; SREL, 2012). The monitoring system consists of a YSI water quality probe, including turbidimeter, data logger, flow metre, depth gauge, ISCO sampler, batteries, solar panels, antenna and transmitter/receiver. The proposed system makes use of SREL's Federal Communications Commission dedicated transmission frequency to remotely monitor and control system performance, providing real-time data acquisition capability. This network experienced delays and issues that prevented it from becoming operational until late 2011 (SREL, 2012). Alternative vegetation studies for waste cap covers are also examining the development of an automated system for evaluating landfill closure cap performance behaviour using buried sensors, remote sensing, real-time systems (Seaman, et al., 2011).

Another Savannah River project, Par Pond, is a 2 640 acre man-made reservoir located in the eastern portion of the site. Par Pond was created in 1958 by constructing an earthen dam across Lower Three Runs Creek. Releases from the R Reactor in the form of process leaks, purges and make-up cooling water have contaminated Par Pond with ^{137}Cs and other radioactive and non-radioactive contaminants such as mercury. The current estimated inventory of ^{137}Cs associated with all sediments within the Par Pond reservoir is approximately 43 Ci, of which 9 Ci are present in the 1 340 acres of exposed sediments. The remaining 68 Ci of ^{137}Cs inventory in the Par Pond system is located in the sediments of the pre-cooler canal/pond system and Lower Three Runs Creek (US EPA, 1995). From June through September 1991, the level of Par Pond was lowered from 200 feet to 181 feet. The 181-ft-level was chosen to reduce the risk and consequences, in the unlikely event of a dam failure, of potential flooding in downstream communities (US EPA, 1995). This resulted in a reduction of the reservoir's surface area and volume by approximately 50% and 65%, respectively, and exposed 1 340 acres (5.3 km^2) of contaminated lakebed sediment (RSHDSS, 2010). The mobility of the radionuclides was much greater than expected. As a result, remote environmental monitoring processes were installed:

- *A real-time soil moisture monitoring system (D-Area phytoremediation) developed by Adcon Telemetry* (RSHDSS, 2010). Adcon develops and manufactures low-power radio networks for agriculture, water management, hydrographics, meteorology and numerous other applications. Examples of applications where their systems have been used include:

- water level: groundwater, channels, rivers/streams;
- water quality: DO, pH, EC;
- flow: closed pipe, open channel, stream gauging;
- rainfall: precipitation and precipitation intensity.

- *FDTAS-tritium analysis system in surface and groundwater in near real time* (RSHDSS, 2010). A field deployable tritium sampling system developed by the University of Georgia Center for Applied Isotope Studies (CAIS). The CAIS has developed and tested the Field-Deployable Tritium Analysis System (FDTAS) to perform near-real-time, *in situ* analysis of tritium in surface and groundwater samples. The FDTAS can be deployed in the field and controlled remotely by a technician in a laboratory. The unit can be programed to collect and analyse water samples using a portable LS counter optimised with a background reduction system. Tritium concentrations as low as 10 Bq/L (~270 pCi/L) can be detected (CAIS, n.d.).
- *Sol-gel indicators for process and environmental measurements* (RSHDSS, 2010). Sol-gel uses a luminescence intensity-based sensor system for dissolved oxygen (DO) measurement. The sensor film consists of tris(2,2-bipyridyl)ruthenium(II(Ru (bpy)3Cl_2 as a photosensitive indicator immobilised within the mixture of tetraethoxylsiane (TEOS) and methyl triethoxysilane (MTEOS) membrane. The laboratory experimental results show that the ratio of the fluorescence intensity of the sensor correlates well with DO concentration from 0-32.96 mg/l. Depending on the charge-coupled device (CCD) camera used, the proposed system can monitor the 0.9×0.9 cm areas with 9×9 μm resolutions and it can be used for oxygen distribution measurement in spatial and temporal scale for undersea monitoring applications. This is an example of how a critical groundwater and surface water parameter can be monitored in real time (Liu, Yu and Zhai, 2009). Similar sensing and telemetry strategies can be developed for monitoring hazardous and radioactive contaminants.

A recently published study describes deployment of a distributed point source monitoring system based on wireless sensor networks in an industrial site where dangerous substances are produced, used and stored. Although the system was for monitoring volatile organic compounds (VOC) the principles of detection and remote monitoring and transmission are an example of remote monitoring capabilities being developed. The system consists of a wireless sensor network (WSN) using photoionisation detectors (PID). It continuously monitors the VOC concentration on an unprecedented time/space scale. Internet connectivity is provided via TCP/IP in real time at a one-minute sampling rate, thus providing plant management, and environmental authorities if necessary, with an unprecedented tool for immediate warning in case critical events happen. The platform is organised into sub networks, each including a gateway unit wirelessly connected to the WSN nodes. Environmental and process data are forwarded to a remote server and made available to authorised users through a rich user interface that provides data rendering in various formats, in addition to world-wide access to the data. Furthermore, this system consists of an easily deployable stand-alone infrastructure with a high degree of scalability and reconfigurability, as well as minimal intrusiveness or obtrusiveness (Manes, et al., 2012; Directions Magazine, 2011).

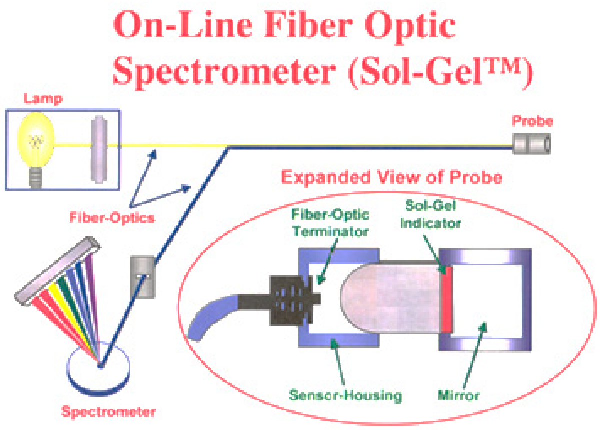
- Remote transmission of data using advanced satellite technology and GPS systems

The Internet of Things (IoT) is what happens when everyday ordinary objects have inter-connected microchips inside them. These microchips help not only keep track of other objects, but many of these devices sense their surrounding and report their findings to other machines as well as to the humans. Applications of IoT can be placed into four broad categories: *i)* environmental quality and protection, oceans and coasts; *ii)* climate change; *iii)* biodiversity, conservation; *iv)* environmental awareness. Environmental quality and protection covers issues of pollution, hazardous waste management, hazardous chemical management, waste disposal management and waste policy and information management (Campbell, 2011; Duplan, 2011). In Japan a nuclear radiation monitoring system uses crowd-sourcing radiation data from individual Geiger counters to give a national real-time map of radiation data, accessible to everyone via Pachube. Pachube is a data brokerage platform for the IoT, managing millions of data points per day from thousands of individuals, organisations and companies across the world. The platform is designed to allow things to "plug-in" to other things in real time so that, for example, buildings, weather stations, interactive environments, air quality monitors, networked energy monitors, virtual worlds and mobile sensor devices can all talk and respond to each other in real time. An open-source, customisable, mobile application allows users to match up their current location with radiation data and receive real-time estimates on radiation levels in their immediate vicinity (Dlodlo, 2012; Unz, Rogers and Waggoner, 2011).

Figure 6.27: Adcon Telemetry real-time soil moisture monitoring system

Figure 6.28: FDTAS-tritium analysis system in surface and groundwater in near real time

Figure 6.29: Sol-gel indicators for process and environmental measurements



Lessons can be drawn from the RESCATAME European Union (EU) project. RESCATAME is a pervasive air-quality sensors network for environmentally friendly urban traffic management. It is an EU-funded project to monitor air quality and urban traffic through a Waspmote sensor board. With data collected from sensors across the city, providing full-time geographic coverage at low cost, municipalities can efficiently achieve a way of better managing urban traffic in major cities. The Waspmotes measure parameters such as temperature, relative humidity, carbon monoxide, nitrogen dioxide, noise and particles. If any of these parameters go above an action level threshold, the system analyses the information and reacts by sending an alarm to a central node. Each Waspmote integrates a GPS to know where the sensor is located. It is also possible to transmit data via GPRS as a secondary radio module for better availability and redundancy in situations when it is critical to ensure the reception of the message, like possible fire alarms. The GPRS module operates in four different bands, meaning it supports any cellular provider and functions globally (Dlodlo, 2012).

A more time-honoured method, "Pigeonblog", provides an alternative means of participating in environmental air pollution data gathering. This project equips urban homing pigeons with GPS-enabled electronic pollution sensing devices capable of sending real-time, location-based air pollution and image data to an online mapping/blogging environment (Dlodlo, 2012).

The Internet of Things includes nanotechnology capable of being embedded on persons or things to monitor and report the data of interest (Akyildiz and Jornet, 2010). A current example of such a function was published in a nuclear decommissioning report that described a wireless application to allow remote data logging via a compact Bluetooth wireless adapter that plugs into handheld digital multimeters (DMM) (Sandrik, 2012). The solution includes two free mobile applications based on Google's Android operating system, one for basic monitoring and another for data logging. The "mobile metre" application enables real-time interaction with connected DMM on the screen of an Android-based smartphone or tablet. The "mobile logger" application simplifies data logging and remote monitoring. Agilent also offers a free data-logging application that runs on Windows-based PC (Yang, Yangb and Plotnick, 2012).

- Complex autonomous wireless networks for hazardous environment monitoring and response

The *International Journal of Distributed Sensor Networks* focuses on applied research and applications of sensor networks. A large number of important applications depend on sensor networks interfacing with the real world. These applications include medical, military, manufacturing, transportation, safety and environmental planning systems.

Many have been difficult to realise because of problems involved with inputting data from sensors directly into automated systems. Sensor fusion in the context of distributed sensor networks has emerged as the method of choice for resolving these problems (Manes, et al., 2012).

A special issue of the *International Journal of Distributed Sensor Networks* presented a series of papers on its website that report on remote-monitoring, autonomous networks that is very instructive relative to the development of such capabilities. Distributed networks of robots or sensors may be deployed in hazardous environments to effectively perform missions such as reconnaissance, surveillance, search and recovery or resource harvesting. When mobile robots are used to host the sensor and communication nodes for a distributed sensing network, the network may achieve improved capabilities for detection and classification of potential targets and other objects of interest (Manes, et al., 2012).

One of the papers describes a nodal decision model where each sensor in the network is required to collect local observations that are probably corrupted by noise, make a local decision regarding the presence or absence of an event and then send its local decision to a fusion centre. After that, the fusion centre makes the final decision depending on the results of all local decisions received and a decision fusion rule. The decision-making capability of each node is different owing to the dissimilar signal noise ratios and some other factors, so a specific sensor's contribution to the global decision should be constrained by its decision-making capability. Based on this idea, a novel linear decision fusion model for WSN is proposed to employ the constrained particle swarm optimisation algorithm and a typical penalty function to solve its problem. The emulation results indicate that the design is capable of achieving a very high accuracy in pinpointing areas of elevated concentrations (Manes, et al., 2012).

The paper also discusses the modelling and performance analysis of a network of chemical sensors with dynamic collaboration. Another paper evaluates the problem of environmental monitoring using a wireless network of chemical sensors with a limited energy supply. Since the conventional chemical sensors in active mode consume vast amounts of energy, an optimisation problem arises in the context of a balance between the energy consumption and the detection capabilities of such a network. A protocol based on "dynamic sensor collaboration" is employed. In the absence of any pollutant, the majority of sensors are in the sleep (passive) mode; a sensor is invoked (activated) by wake-up messages from its neighbours only when more information is required. The authors propose a mathematical model of a network of chemical sensors using this protocol (Manes, et al., 2012).

Another paper addresses the effects of motion on distributed detection in mobile *ad hoc* sensor networks. A large set of mobile wireless sensors observe their environment as they move about. The authors consider the subset of these sensors that each made observations about a brief, localised event at the time when near that location. As the sensors continue to move, one of them eventually finishes processing its observations, decides that an event of interest occurred, and wants to determine if other sensors confirm its results. This sensor thus assumes the role of a cluster-head (CH) and requests that all other sensors that collected observations at that time/location reply to it with their decisions. The motion of the sensors since the observation time determines how many wireless hops their decision must cross to reach the CH. The authors analyse the effect of this motion in this 1-D case by modelling each sensor's motion as a correlated random walk (CRW), which can account for realistic transient behaviour, geographical restrictions, and non-zero drift. The results allow a rapid characterisation of the time-dependence of the distributed detection algorithms being executed in realistic mobile sensor networks.

Another paper discusses density control, which is of great relevance for wireless sensor networks monitoring hazardous applications where sensors are deployed with high density. Owing to the multi-hop relay communication and many-to-one traffic characters in wireless sensor networks, the nodes closer to the sink tend to die faster, causing a bottleneck for improving the network lifetime (Ehlers, et al., 2012). The authors systematically investigate the theoretical aspects of the network load and the node density. Furthermore, the authors prove the accessibility condition to satisfy that all the working sensors exhaust their energy with the same ratio. By introducing the concept of the equivalent sensing radius, a novel algorithm for density control to achieve balanced energy consumption per node is thus proposed. Another paper discusses various topology management schemes for improving network parameters, such as capacity, lifetime, coverage and latency and by integrating a scheme to increase battery life and energy efficiency among the networked sensors. This is important since a wireless sensor network is composed of a large number of sensor nodes that are densely deployed in the field. These nodes monitor the environment, collect the data and route it to a sink. The main constraint is that the nodes in such a network have a battery of limited stored energy, and if the nodes start to die, the network lifetime gets reduced. Application for wireless communication networks for gas turbine engine testing and construction of a distributed AUV network for underwater 3-D plume-tracking operations are discussed (Manes, et al., 2012). It is quite evident that this is an area worthy of further decommissioning R&D for facilities that require long-term monitoring and response capabilities.

Future suggested R&D for use of remote sensing and satellite technologies

- ***Description*** – R&D are required to continue to develop remote sensing detectors, systems and networks for characterisation and monitoring of decommissioning facilities as well as hazardous material storage sites.
- ***Objectives*** – To develop remote or *in situ* deployable sensors for radioactive contaminants and radiation to monitor and characterise airborne, waterborne and soil contaminants as well as storage environs and waste package integrity. Develop transmission, communication, data analysis and response algorithms and deployment systems in fixed or mobile configurations that range from human to machine, machine to machine or machine to network, including autonomous deployment and monitoring capabilities.
- ***Desired deliverables*** – *In situ* or remote sensors for monitoring radiation levels and radioactive material concentrations in the environment. Wireless positioning and communications systems to communicate and analyse data that can function in complex networks and situations. Field-deployable platforms to house and transport the required instrumentation, equipment and power supplies.

Comparison of dose modelling and code/model results

Challenges

Dose modelling is performed to translate the dose- or risk-based regulatory delicensing or release criteria to measurable concentrations of radioactivity in soil and on building surfaces. Dose modelling considers how future receptors might be exposed to residual radioactivity that remains following the decommissioning of a site or building.

Dose models and codes are used to calculate the risk or dose associated with the facility end-state contaminant concentrations. The measurable concentrations that correlate to the acceptable risk or dose for release from regulatory control or license termination are called derived concentration guideline levels (DCGL).

The starting point for dose modelling is determining the post-decommissioning usage scenario likely to occur. These range from limited delicensing, where part of the facility remains under regulatory control, to industrial or residential occupancy scenarios after the facility is released, to the more conservative “resident farmer” scenarios where the property is occupied and used as a farm and the majority of food and water consumed is grown on the property.

Once the exposure scenario is chosen, the contaminants of concern must be identified, and the model parameters decided upon and input. These parameters include: location, area and depth of contamination remaining in soils or on structures; the hydrogeological parameters of the site; fate and transport parameters, such as distribution coefficients; the exposure pathways; and exposure durations, rates and dose conversion factors. Typically these codes allow probabilistic analysis of the model to be run with each input parameter assigned a statistical distribution around a mean and standard deviation. The code picks random values from within the distribution and runs the model using them to determine which parameters significantly alter the outcome of the dose or risk assigned. Often this process is underpinned by processes such as Latin hypercube sampling to ensure that values chosen randomly are representative of the entire distribution of possible values and have not been grouped by chance at one particular part of the distribution.

Input parameters that significantly alter the outcome are called "sensitive parameters" and either require further site-specific justifications for the values chosen or are chosen from the upper or lower quartile of the distribution to ensure that the modelled doses or risks are conservative. The probabilistic analysis must be performed for each contaminant of concern and the models typically calculate the fate and transport and resultant dose from the daughter radionuclides as well. Thus, even relatively simple contaminated zone and hydrogeological models require long computer run times on conventional personal computers to perform probabilistic analysis on radionuclides, such as ^{239}Pu or ^{241}Am and their many daughters. At sites contaminated with non-radiological contaminants such as heavy metals, asbestos or PCB, the fate transport and risk from residual levels of these contaminants must also be considered when determining acceptable end-state criteria based on the "combined risk" from radiological and non-radiological contaminants.

There is currently very little consensus on contaminants' fate and transport modelling and for dose calculation/risk assessment that underpins license termination or delicensing criteria. Despite recommendations from international organisations such as the International Atomic Energy Agency (IAEA) and the International Commission on Radiation Protection (ICRP), each nation tends to go it alone, developing their own software codes for models and using different values for critical parameters. International variation in values occur in parameters such as distribution coefficients in soils, dose conversion factors from inhalation and ingestion of radionuclides, occupancy factors, radionuclide uptake by food types and consumption rates, and even in the determination of the end-state dose/risk criteria acceptable for delicensing.

The most widely used modelling codes in the decommissioning industry in the United States are likely RESRAD for soil areas and RESRAD-BUILD or DandD for building surfaces. Both RESRAD codes were written and are maintained by Argonne National Laboratory. Another Argonne code, RESRAD-OFFSITE, can calculate doses to receptors adjacent to the site as well as those located within it. A geostatistical code ISATIS is used in Europe for fate and transport modelling and risk assessments.

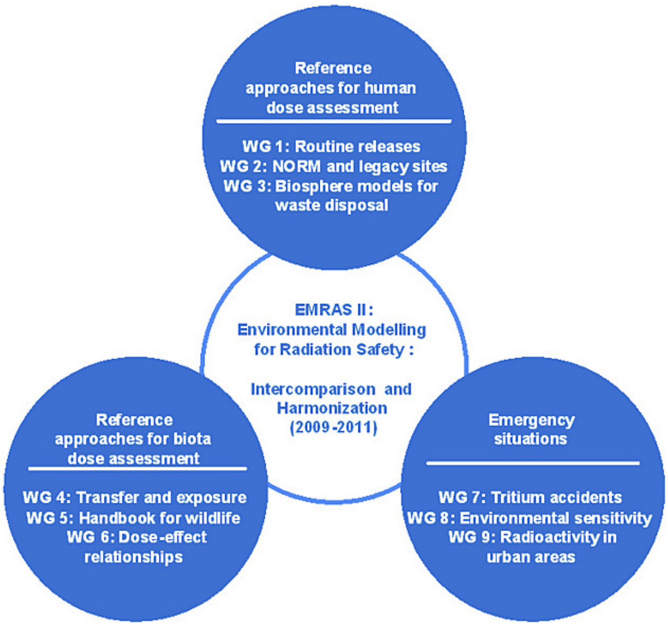
Efforts in the development of integrated environmental models, such as those described earlier in the subsection of that title, could go a long way toward solidifying international consensus and resolving these issues. One example is the Chernobyl cooling pond. The DOE's Advanced Simulation Capability for Environmental Management (ASCEM) or the IAEA's Environmental Modelling for Radiation Safety (EMRAS) could be brought to bear on such an issue requiring international collaboration.

Summary of current R&D for comparison of dose modelling codes using actual case studies

- Development of groundwater models and benchmarking

The bulk of United States and IAEA efforts to develop better, more fully integrated hydrogeological modelling of complex hazardous material sites were described earlier in the subsection entitled *Development of integrated environmental models* (Environment Agency, 2011). The EMRAS II project, which ran from 2009-2011, included environmental model enhancements and benchmarking. The general aim of this IAEA programme was to improve capabilities in the field of environmental radiation dose assessment by means of acquiring improved data for model testing, comparison, reaching consensus on modelling philosophies, approaches and parameter values, development of improved methods and the exchange of information.

Figure 6.30: EMRAS II working groups



As part of the EMRAS project the IAEA compared various modelling software and approaches to three hypothetical scenarios for testing models, and four real scenarios for sites contaminated with naturally occurring radioactive materials (NORM) (IAEA, 2007b; Trotti, et al., 2009). The IAEA report discusses the testing and further development of the hypothetical scenarios (Chapter 4), the testing of the real scenarios (Chapter 5) and the results of the development and testing the models (Chapter 6). A number of models were reviewed during the BIOMOVS, BIOMOVS II, VAMP and BIOMASS projects. However, most of these models were not considered suitable for the NORM project, because they only simulate the behaviour of single, specific radionuclide, and do not have the databases that would facilitate their use for simulating the behaviour of NORM, which includes many daughter nuclides in the environment. Models that could be used for NORM assessment fall into three categories (IAEA, 2007b): screening, compliance and detailed impact assessment. The models that were used are described in Appendix II of the IAEA report. The appendix also lists other models that were noted as potentially useful but not actually tested by project participants (IAEA, 2007b).

Table. 6.3: IAEA NORM study models used for hypothetical scenarios

Scenario	Models	Users
Point source	COMPLY [23] CAP-88 [24, 25] PC-CREAM [26] CROM [27]	J. Horyna, R. O'Brien J. Horyna P. McDonald, R. O'Brien, V. Amado D. Perez Sanchez
Area source	DOSDIM = HYDRUS RESRAD (onsite) [28, 29] RESRAD-OFFSITE [30,31] PRESTO v4.2	T. Zeevaert, G. Olyslaegers J. Horyna, R. O'Brien C. Yu, P. McDonald, R. O'Brien, V. Amado, J. Horyna J. Horyna
Area source + river	RESRAD-OFFSITE	C. Yu, P. McDonald, R. O'Brien

Results from the COMPLY/CAP-88 and PC-CREAM testing on the hypothetical point source showed the doses calculated by COMPLY were higher than those calculated by PC-CREAM. These are all software packages designed for performing radiological impact assessments of routine and continuous discharges of radionuclides to the environment. This is consistent with the different nature of the two models; COMPLY/CAP-88 is designed to check compliance with regulatory requirements and is deliberately conservative, whereas PC-CREAM is a very detailed impact assessment model. In view of this, the higher doses predicted by COMPLY/CAP-88 are acceptable. The hypothetical area source results showed that the general agreement between the DOSDIM and RESRAD results were good, considering the differences between the models. However, where DOSDIM + HYDRUS predicted that there would be no leaching of radionuclides from the waste if the waste was assumed to be clay, RESRAD did predict slow leaching. The real scenarios evaluated included (IAEA, 2007b):

- lignite power plant (LPP II) scenario;
- former gas mantle production plant (Camden) scenario;
- phosphogypsum disposal site (PGDS1);
- phosphogypsum disposal site (PGDS2).

Only the first scenario for the lignite power plant (LPP II) scenario was tested in detail by the working group. It was tested using PC-CREAM, COMPLY and CROM software packages. The doses estimated using PC-CREAM and COMPLY were similar in magnitude; this was encouraging, because of the uncertainties in the source terms and the differences between the models. The predicted ground surface concentrations from CROM are higher than the measured values, but of approximately the same order of magnitude (IAEA, 2007b).

The RESRAD programmes appear to meet most of these requirements. DOSDIM and HYDRUS have been used on several occasions, but it is not clear that the detailed documentation described above is readily available to a wide range of potential users. The methodologies used in PC-CREAM are described in detail but the databases in PC-CREAM do not include all the naturally occurring radionuclides, and must be added by the user (IAEA, 2007b).

There is a marked lack of models capable of handling area source situations where the geometry of the waste is complex (for example, varying waste thickness or multiple waste stacks). Many legacy sites have these characteristics. This may not be important in most cases, because measurement involves averaging in both time and space and inevitably tends to smooth out small-scale variations. Nevertheless, further work is needed on situations of this type to determine whether the available models are suitable for assessing the impact of such sites (IAEA, 2007b).

In a more recent presentation at the 2012 EU-NORM Symposium it was reported that a general assessment methodology was developed and tested on a few case studies. Modelling exercises were carried out for the PG stack of Gela (Italy) and the uranium tailings repository of Bellezane (France). The PG stack calculations were performed with RESRAD-OFFSITE as well as the Belgian software DOSDIM, with and without a clay barrier wall. The models showed that the clay barrier retards the breakthrough and reduces the ^{226}Ra dose by a factor of four, but comparison of the results between the models was not mentioned (EU-NORM, 2012).

The tailings repository in France was modelled using two scenarios, a current situation scenario and an intrusion scenario. Two models were used to perform the calculations, SATURN and RESRAD-OFFSITE. Both modelling results for the current situation scenario have shown that the human dose impact is trivial during the calculation time frame of 1 000 years; this is due to the very long travel time (more than 40 000 years) of the radionuclides to surface water and groundwater. For the intrusion scenario, both models give significant dose estimates but a meaningful comparison between the RESRAD-OFFSITE and the SATURN results is difficult to establish due to different modelling assumptions and approaches; for instance, in the modelling of the source term and the exposure pathways. In the RESRAD-OFFSITE calculations, pits 68 and 105 containing the tailings of the Bellezane repository had been replaced by an average single source term, while SATURN calculated contributions of both pits separately. SATURN focused on a few specific exposure pathways (neglecting, for example, the drinking water pathway) while RESRAD-OFFSITE included all of them. In spite of the different modelling assumptions, the quantitative results in the intrusion scenario for the dose from ingestion of plants turned out to be very similar in both models. At t = 100 yr, this dose amounts to 1.18 mSv/yr in the RESRAD-OFFSITE calculations while the SATURN calculations give 1.33 mSv/yr for pit 68 and 0.65 mSv/yr for pit 105 (EU-NORM, 2012).

The modelling exercises, and in particular the comparison between different models, could not be completed due to lack of time. The IAEA launched a follow-up programme to EMRAS II called MODARIA (Modelling and Data for Radiological Assessment) (2014b). The programme will continue the IAEA's activities in the field of testing, comparing and developing guidance on the application of models to assess exposures to humans and radiological impacts on the environment. This benchmarking of models should help develop some international consensus. The results of radiological assessments are used, e.g. in the evaluation of the radiological relevance of routine and accidental release of radionuclides, to support decision making in remediation work and for the performance assessment of radioactive waste disposals. MODARIA will provide an international focal point in this field and will help member countries to develop and maintain knowledge and competence in the areas of radioecology and environmental assessment. The overarching objective of the IAEA's activities in environmental modelling is to enhance the capabilities of member countries to simulate radionuclide transfer in the environment and, thereby, to assess exposure levels of the public and in the environment in order to ensure an appropriate level of protection from the effects of the ionising radiation associated with radionuclide releases and from existing radionuclides in the environment.

The specific objectives of the MODARIA project in the areas of radioactive release assessment, restoration of sites with radioactive residues and environmental protection are as follows (IAEA, 2014b):

- to test the performance of models developed for assessing the transfer of radionuclides in the environment and radiological impact to man and environment;
- to develop and improve models for particular environments and, where appropriate, to agree on data sets that are generally applicable in environmental transfer models;
- to provide an international forum for the exchange of experience, ideas and research information.

The MODARIA programme will run for four years and was launched at its first technical meeting held at IAEA headquarters in Vienna in November 2012 (IAEA, 2013c). The implementation of the MODARIA programme is led by a steering committee that will generally meet during the annual technical meeting if necessary; additional meetings may be organised. Potential topics were proposed to the participants in a document. The ideas for the programme had been developed during 2011-2012, taking into account the work of the now-completed EMRAS II programme, plus suggestions and views from a variety of sources.

Further work on the modelling of NORM and legacy sites could be carried out within MODARIA. Points of attention could be, among other things (EU-NORM, 2012):

- development of the capability to carry out probabilistic calculations and of rigorous methods of estimating uncertainties in model predictions and estimates;
- modelling of the effectiveness of different remediation actions for real sites;
- cross-over between working groups, e.g. submit real site data to other working groups for possible integration in their own modelling objectives.

- Localised elevated contamination or "hot spot" modelling

The primary goal of research described in a 2008 PhD thesis (Abelquist) was to develop a technically defensible approach for modelling the receptor dose due to smaller "hot spots" of residual radioactivity. Nearly 700 combinations of environmental pathways, radionuclides and hot spot sizes were evaluated in this work. The hot spot sizes that were studied ranged from 0.01 m^2 to 10 m^2 and included both building and land area exposure pathways. Dose modelling codes RESRAD, RESRAD-BUILD, and MicroShield were used to assess hot spot doses and develop pathway-specific area factors for 11 radionuclides.

The research identified particularly hot spot sensitive pathways, i.e. particularly sensitive to changes in the areal size of the contaminated area. The external radiation pathway was the most hot spot sensitive for 8 of the 11 radionuclides. Pathway-specific area factors derived for 11 radionuclides were generally much less restrictive than previous hot spot criteria. The research proposed a Bayesian statistical approach for assessing the acceptability of hot spots. A posterior distribution is generated based on the final status survey data that provides an estimate of the 99th percentile of the contaminant distribution. Hot spot compliance is demonstrated by comparing the upper tolerance limit, defined as the 95% upper confidence level on the 99th percentile of the contaminant distribution in the survey unit, with the 99th percentile derived concentration guideline level ($DCGL_{99th}$) value. The $DCGL_{99th}$ is the hot spot dose limit developed using the dose modelling research to establish the area factors mentioned above. The proposed approach provides a hot spot assessment approach that considers hot spots that may be present, but not found. Examples are provided to illustrate this approach (Abelquist, 2008).

Table 6.4 provides hot spot area factors based on the external radiation pathway for radionuclides of interest from the Fukushima accident (Abelquist, 2011).

Table 6.4: DCGL multiplication factors for Fukushima-related nuclides hot spots of varying sizes

	Hot spot size (m^2)						
Radionuclide	1 000	10	3	1	0.5	0.1	0.01
^{60}Co	1	2.30	4.73	11.4	21.3	100	990
^{129}I	1	1.76	3.14	6.93	12.6	57.6	575
^{137}Cs	1	2.18	4.42	10.6	19.8	93.1	918
^{239}Pu	1	1.92	3.63	8.41	15.5	72.4	713

These area factors were evaluated both when the receptor was located directly on the 1 m^2 soil hot spot (ranged from 6.6 to 11.4) and when the receptor was located 6 m from the hot spot (ranged from 650 to 785 – not shown in Table 6.4). It is worth emphasising that the area factors for external radiation pathway are generally the same regardless of the radionuclide. For example, the area factor ranged from roughly 7 to 11 for a 1 m^2 hot spot, from 12 to 21 for a 0.5 m^2 hot spot and 60 to 100 for a 0.1 m^2 hot spot (Abelquist, 2011).

- Dose modelling for non-human species

Since the United Nations Conference on Environment and Development in 1992 there has been an increasing focus on protecting the environment from all forms of stressors, including radioactivity. As a consequence, a series of international programmes has been aimed at establishing a framework for assessing potential impacts on, and protection of, non-human biota from releases of radioactive materials. However, despite extensive development work, there are presently no internationally recognised limiting criteria for determining the significance of impacts on non-human biota. A range of numerical benchmarks has been developed but it generally represents "screening levels" or similar values that are intended to trigger further consideration or assessment rather than regulatory responses. In the absence of an agreed basis for assessment, or detailed practical guidance on the appropriate actions to be taken in the event that existing benchmarks are exceeded, the existing levels may be inappropriately applied and resources misallocated (SKM Enviros, 2010; Higley, Alexakhin and McDonald, 2004).

The International Commission on Radiological Protection (ICRP) has widened the scope of its radiological protection system to include explicit consideration of environmental impacts, and has established a committee to develop an assessment methodology (ICRP, 2007). The IAEA has established working groups on the issue as part of its EMRAS programme, and the European Commission has funded a series of programmes to develop an assessment framework (FASSET and ERICA) and to investigate options for establishing numerical benchmarks and their application (PROTECT) (Andersson, et al., 2008). The EC FASSET and ERICA programmes ran consecutively from 2000 through 2007 and provided a European biota dose assessment methodology, which included a re-analysis of biota dose effects information and proposals for screening levels. The PROTECT programme focused specifically on the derivation of protection goals and further statistical analysis of effects data (SKM Enviros, 2010).

United Kingdom programmes include the development of the Environment Agency's approach for assessing the impact of ionising radiation on wildlife (R&D Publication 128), which has subsequently been applied in assessments of authorised facilities, within the context of United Kingdom conservation (Habitats) regulations. There have also been developments in other countries. In the United States, for example, the Department of Energy has established a working group and technical standard methodology for evaluating radiation doses to biota, which has been implemented as a software tool in RESRAD-BIOTA (SKM Enviros, 2010).

The most recently published benchmarks are those derived by the ICRP, as a consequence of a series of programmes funded by the EC. These benchmarks are referred to as "derived consideration reference levels" (DCRL) and "screening levels", respectively. These benchmarks were derived in different ways and with different purposes, but in both cases the levels were not intended to be used as regulatory limits, but as indicators of where further consideration is required (e.g. more detailed assessment). However, it is still necessary to determine an appropriate response in the event that such benchmarks are exceeded (SKM Enviros, 2010).

Table 6.5 lists numerical benchmarks proposed for protection of populations expressed in terms of dose rate from the NDA SKM Enviros report (2010). Regulatory interpretation continues to evolve, and there would be value in encouraging debate in scientific and regulatory communities to avoid potential future misallocation of resources and regulatory activities by applying dose criteria that are inappropriate (e.g. using screening values to limit operations). This is a rapidly evolving field of work; consideration of appropriate guidance and the possibility of other, higher benchmarks, is a matter of some urgency (SKM Enviros, 2010).

Since 2005, ICRP Committee 5 has been working to develop an assessment method by which environmental impacts may be directly assessed and managed, on the basis of proposals outlined in its Publication No. 91 (ICRP, 2003). The resultant approach was published in October 2009 (ICRP, 2009). This approach is based on the definition of a group of 12 "reference animals and plants". These entities perform an analogous function to "reference man" within the existing ICRP system of radiological protection for humans. The ultimate objective is to develop a common approach to human and environmental protection. The ICRP has established DCRL, defined as follows (SKM Enviros, 2010):

> *A band of dose rate within which there is likely to be some chance of deleterious effects of ionizing radiation occurring to individuals of that type of reference animal or plant (derived from knowledge of defined expected biological effects for that type of organism) that, when considered together with other relevant information, can be used as a point of reference to optimize the level of effort expended on environmental protection, dependent on the overall management objectives and the relevant exposure situation.* (ICRP, 2009)

Table 6.5: Numerical benchmarks proposed for protection of populations expressed in terms of dose rate

	IAEA (1992)	UNSCEAR (1996)	Environment Canada (2003)	ERICA (2008)	DCRLSs (ICRP, 2009)
	μGy/h				
Terrestrial					
Plants	400	400	100	10	
Reference pine tree					40-40
Reference wild grass					40-400
Animals	40	40-100		10	
Invertebrates			200		
Reference bee					400-4 000
Reference earthworm					400-4 000
Birds					
Reference duck					4-40
Mammals			100		
Reference deer					4-40
Reference rat					4-40
Aquatic					
Fresh water organisms	400	400		10	
Algae			100		
Macrophytes			100		
Benthic invertebrates			200		
Reference frog					40-400
Fish			20		
Reference trout					40-400
Marine organisms	400	400		10	
Reference crab					400-4 000
Reference flatfish					40-400
Reference brown seaweed					400-4 000
Deep sea organisms	1 000			10	

Source: SKM Enviros (2010).

The bands of DCRL are presented as an order of magnitude range for each of the reference animals and plants (RAP). The implication is that dose rates falling below the lower end of the band are unlikely to be of concern, while for doses between these levels, and above the higher level, some consideration of appropriate action may be required, depending on the situation. These values were derived largely on the basis of expert judgment (SKM Enviros, 2010).

These values have the advantage of not only being specific to different trophic levels, but also including an order of magnitude range around each value, thus allowing for a two-stage consideration of dose rate results. However, the limited number of organisms for which these values are provided pose a significant challenge to their application; interpolation between organism types is generally required. Furthermore, regulatory interpretation has yet to be clarified.

The reference organism approach is outlined in more detail in Larsson's report in the *Journal of Environmental Radioactivity* (2009). The ERICA project included the development of a software tool (Brown, et al., 2004) incorporating dose effects information from a broader range of sources than FASSET and a statistical analysis of such data that provided the basis for specification of a single dose rate screening level. This analysis was based on the European technical guidance document commonly used to derive screening levels for non-radioactive pollutants (EC/JRC, 2003)

The ERICA screening level (of 10 μGy/h, 1 mrem/h) is intended to be protective of the structure and function of generic ecosystems and organism groups. It was derived from a statistical analysis of radiation effects data, and represents the dose rate at which a 10% change in an observed effect may be expected to occur, relative to a control group, in 5% of species. This analysis was based on reproduction, morbidity and mortality from acute and chronic exposures.

The PROTECT project followed ERICA. Its mission included reflection on the suitability of different approaches available to meet the needs of the international community, and the development of dose rate thresholds for wildlife to help to determine the risk of exposure to ionising radiation (SKM Enviros, 2010).

Additional analyses were undertaken on the basis of reproductive effects that were found to be the most sensitive endpoints for the majority of species. A similar screening value was defined on the basis of an analysis for both aquatic and terrestrial ecosystems and the lowest dose rates at which a 10% change in the reproductive endpoint is observed. A safety factor of two was applied in view of the number and origin of data, the endpoints evaluated, the availability of supporting evidence and the data spread (SKM Enviros, 2010).

The form of future screening levels and other benchmarks will continue to be based on multi-organisation and international debate, e.g. within the ICRP and through existing IAEA work programmes. At an international level, there is a need to develop a more structured approach to dealing with situations in which current screening criteria or other benchmarks are exceeded, or where an existing assessment needs to be enhanced for other reasons.

Future suggested R&D for comparison of dose modelling codes using actual case studies

- ***Description*** – Research and development is required to develop more robust and versatile environmental fate, transport and risk assessment software tools. Existing tools should be benchmarked further against common modelling scenarios. The scenarios should integrate with and support the license termination processes delineated in EURSSEM and MARSSIM, including evaluations of inhomogenous source term distributions. Other research on the acceptable risk and dose basis for human and non-human species for delicensing needs to be conducted such that international standards can be harmonised.
- ***Objectives*** – Harmonise delicensing risk criteria for humans and non-human species. Develop more robust and versatile fate and transport models, and provide an international consensus on input parameters to underpin the models. Develop software tools and update EURSSEM and MARSSIM to integrate more powerful and accurate modelling capabilities into the license termination process.
- ***Desired deliverables*** – Studies that provide common risk-based criteria for release of facilities from regulatory control that address both human and non-human species. Studies that further the harmonisation of fate, transport and risk input parameters into modelling software and the update of existing software to include internationally agreed parameters. Bench testing of existing modelling software on common scenarios and improvement of existing software. Update to EURSSEM and MARSSIM processes to integrate improved modelling and geostatistical capabilities into the process for demonstrating risk/dose targets have been met at the desired statistical confidence levels.

Suggested areas of future collaboration

Areas of future collaboration may include:

- information exchange and joint testing of 3-D modelling for subsurface contaminant transport and groundwater modelling, as well as atmospheric and ocean plumes with emphasis on methods for treatment of uncertainties and sample optimisation;
- information exchange on advanced technologies for radiological characterisation, detection and monitoring using remote sensing, robotic techniques and satellite technologies;
- information exchange on approaches, methodologies, models and scenarios used to demonstrate compliance with clean-up and decommissioning criteria.

7. Conclusions

Clearly the need for research and development of more improved and efficient decommissioning technologies is vast and pressing. The challenge for the international community is to further these efforts and to get in front of technological developments and bring them to bear on decommissioning in a manner that optimises the limited funding for R&D available internationally. A collaborative, integrated, international effort is required to optimise funding and strategically target future R&D in order to deal with the legacies of the arms race and nuclear accidents such as Chernobyl and Fukushima Daiichi. A powerful economic incentive to fund development of more efficient decommissioning technologies currently exists due to the backlog of facilities awaiting or approaching decommissioning. This effort is necessary to fully exploit existing technologies for characterisation and risk assessment, dismantlement, remediation, material processing and sentencing in order to gain much needed efficiencies in the decommissioning process.

The nuclear industry has not fully exploited or implemented current technological capabilities, and too often relies on outdated technology to perform decommissioning tasks. Decommissioning across the globe is being executed largely through the force of manual labour, thereby requiring extensive personnel protection measures, engineering controls and costly, inefficient detailed work planning and monitoring to achieve the high levels of safety required. Although this approach is ubiquitous, it is clearly not as efficient as using remotely operated robotic technologies coupled with current technologies for end-effector and material handling tooling, assay, material segregation and sentencing. Modular, automated, remotely operated, broadly applicable technologies need to be assembled and field tested for dismantlement and material handling at actively decommissioning facilities.

If the D&D industry continues to reinvent current technologies for limited specialised tasks at isolated decommissionings and insists on demonstrating a higher level of performance than that which can be achieved using conventional manual methods, it will fall further behind in exploiting the ongoing developments in automated and robotic technologies that are occurring outside the industry. If lessons learnt to refine existing technologies and persistent R&D for decommissioning applications are not pursued, the industry will continue to have to adapt, tweak and assemble legacy technologies developed for semi-related applications.

Funding schemes must address the risk aversion of D&D managers towards deploying new technologies with the aim of deploying existing technology in the field for testing and development at decommissioning projects. A concerted effort is also required to end the "feast or famine" cycle of decommissioning, which does not foster continuous improvement of decommissioning expertise and technology, but instead instils a false sense of uniqueness that encourages wasteful reinvention of previously used technologies. International efforts to view facilities awaiting decommissioning as a fleet and to sequence decommissioning in order to develop a mature, stable supply chain that can spread the R&D costs of developed technologies over many decommissioning projects will be required to foster further R&D and ensure efficient improvements are brought to bear on the decommissioning challenges highlighted in this report. The isolated fits and starts of these projects in individual companies and countries leads to an unreliable and destructive economic environment that undermines sustained development of decommissioning expertise.

The difficulty in designing technologies and repositories to stabilise, package and sequester waste with exceedingly long half-lives also involves a major effort requiring large expenditures of R&D resources due to the uncertainties of the environment, material characteristics and interactions of materials in repositories over the exceedingly long durations required for the waste to decay to safe levels. An aggressive R&D effort aimed at technologies to extract and destroy or reuse these long-lived materials, such as actinides in high-level waste or activated graphite, should be a parallel path of R&D for dealing with the daunting challenges related to the safe, long-term disposal of these materials. This path necessitates the development of strategies to utilise and integrate legacy materials in new-build, next-generation reprocessing and fuel fabrication facilities, as well as nuclear power plants such as mixed-oxide fuels in fast neutron reactors.

Conservation of current and planned disposal facility infrastructure is also a pressing need in many countries, from geologic repositories for high- and intermediate-level waste to shallow land disposal facilities for low- and very-low-level waste. Many countries do not have the environmental conditions to easily accommodate construction and operation of such facilities. Given the expansive volumes of legacy waste around the world, prioritised research in handling, stabilisation, encapsulation and containment, and co-ordinated development of stable, efficient disposal options, are and will continue to be critical.

Decontamination, recycling and reuse of decommissioning materials is also an important factor for reducing the volume of materials requiring disposal. The recycling and reuse of contaminated metal, concrete and graphite within the nuclear industry would greatly ease the burden on waste disposal facilities. Most contaminants in concrete and steel are surficial with the overall bulk of the material having very low concentrations of contamination at depth. A large percentage of the materials that are not acceptable for very-low-level waste disposal are at concentrations that will be reached in new facilities, constructed with new materials after only a few years of operation. Integration of decommissioning material reuse in new build projects and the R&D necessary to process the materials such that they meet the new build material standards is a worthwhile consideration in order to continuously reduce the decommissioning waste burden on waste disposal facilities.

Similarly, there is a need to integrate modern geostatistical capabilities into the characterisation and final status survey methodologies used to demonstrate compliance with licence termination criterion. Update, scalable and modular fate and transport software needs to be developed and integrated with geostatistical capabilities to ensure that relatively simple end states and contaminant distributions, as well as complex sites with multiple surface and subsurface contaminated zones, can be modelled. There is also a need to take advantage of advances in telemetry and satellite-based Internet connectivity to integrate mapping and sample and survey data acquisition with the fate and transport models and routine monitoring tasks.

This report has identified many near-term and long-term technologies and capabilities that hold promise for making current and future decommissionings better, cheaper and faster. Development of these capabilities requires integrated planning and co-operation and a long-term commitment to foster decommissioning R&D and to have member countries implement new technologies in the field.

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R&D and Innovation Needs for Decommissioning Nuclear Facilities

Nuclear decommissioning activities can greatly benefit from research and development (R&D) projects. This report examines applicable emergent technologies, current research efforts and innovation needs to build a base of knowledge regarding the status of decommissioning technology and R&D. This base knowledge can be used to obtain consensus on future R&D that is worth funding. It can also assist in deciding how to collaborate and optimise the limited pool of financial resources available among NEA member countries for nuclear decommissioning R&D.

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