Nuclear Development

ACTINIDE AND FISSION PRODUCT PARTITIONING AND TRANSMUTATION

**6th Information Exchange Meeting
Madrid, Spain,
11-13 December 2000**

In co-operation with the
European Commission

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Hosted by CIEMAT and ENRESA

*Centro de Investigaciones Energéticas,
Medioambientales y Tecnológicas*

*Empresa Nacional de
Residuos Radioactivos, SA*

NUCLEAR ENERGY AGENCY
ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

Pursuant to Article 1 of the Convention signed in Paris on 14th December 1960, and which came into force on 30th September 1961, the Organisation for Economic Co-operation and Development (OECD) shall promote policies designed:

- to achieve the highest sustainable economic growth and employment and a rising standard of living in Member countries, while maintaining financial stability, and thus to contribute to the development of the world economy;
- to contribute to sound economic expansion in Member as well as non-member countries in the process of economic development; and
- to contribute to the expansion of world trade on a multilateral, non-discriminatory basis in accordance with international obligations.

The original Member countries of the OECD are Austria, Belgium, Canada, Denmark, France, Germany, Greece, Iceland, Ireland, Italy, Luxembourg, the Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States. The following countries became Members subsequently through accession at the dates indicated hereafter: Japan (28th April 1964), Finland (28th January 1969), Australia (7th June 1971), New Zealand (29th May 1973), Mexico (18th May 1994), the Czech Republic (21st December 1995), Hungary (7th May 1996), Poland (22nd November 1996), Korea (12th December 1996) and the Slovak Republic (14 December 2000). The Commission of the European Communities takes part in the work of the OECD (Article 13 of the OECD Convention).

NUCLEAR ENERGY AGENCY

The OECD Nuclear Energy Agency (NEA) was established on 1st February 1958 under the name of the OEEC European Nuclear Energy Agency. It received its present designation on 20th April 1972, when Japan became its first non-European full Member. NEA membership today consists of 27 OECD Member countries: Australia, Austria, Belgium, Canada, Czech Republic, Denmark, Finland, France, Germany, Greece, Hungary, Iceland, Ireland, Italy, Japan, Luxembourg, Mexico, the Netherlands, Norway, Portugal, Republic of Korea, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States. The Commission of the European Communities also takes part in the work of the Agency.

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, as well as
- to provide authoritative assessments and to forge common understandings on key issues, as input to government decisions on nuclear energy policy and to broader OECD policy analyses in areas such as energy and sustainable development.

Specific areas of competence of the NEA include safety and regulation of nuclear activities, radioactive waste management, radiological protection, nuclear science, economic and technical analyses of the nuclear fuel cycle, nuclear law and liability, and public information. The NEA Data Bank provides nuclear data and computer program services for participating countries.

In these and related tasks, the NEA works in close collaboration with the International Atomic Energy Agency in Vienna, with which it has a Co-operation Agreement, as well as with other international organisations in the nuclear field.

FOREWORD

The objective of the OECD/NEA Information Exchange Programme on Actinide and Fission Product Partitioning and Transmutation, established in 1989, is to enhance the value of basic research in this area by facilitating the exchange of information and discussions of programmes, experimental procedures and results. This Programme was established under the auspices of the NEA Committee for Technical and Economic Studies on Nuclear Energy Development and the Fuel Cycle and is jointly co-ordinated by the NEA Nuclear Development Division and the NEA Nuclear Science Division.

The scope of the Programme includes information on all current and past research related to the following areas:

1. Physical and chemical properties of elements generated in the nuclear fuel cycle:

   a) Chemical properties and behaviour of the actinide species in aqueous and organic solution.
   b) Analytical techniques and methods.
   c) Physical and chemical properties of various actinide compounds.
   d) Collection and evaluation of nuclear and thermodynamic data of relevant elements.

2. Partitioning technology:

   a) Partitioning of high-level liquid waste with wet and dry processes.
   b) Platinum-group metals-recovery technology.
   c) Fabrication technology of the fuel and target materials.
   d) Partitioning in the reprocessing process.

3. Transmutation:

   a) Transmutation with fast reactors.
   b) Transmutation with TRU burner reactors.
   c) Transmutation with proton accelerators.
   d) Transmutation with electron accelerators.

4. Applications

Other activities related to nuclear data, benchmark exercises and more basic science studies in relation to this Programme are conducted by the NEA Nuclear Science Division and the NEA Data Bank.

The Programme is open to all interested NEA Member countries contributing to the information exchange activities and the Commission of the European Communities. All participants designated a liaison officer who is a member of the Liaison Group (see Annex 1, CD-ROM).

The Information Exchange Meetings form an integral part of the Programme and are intended to provide a biennial review of the state of the art of partitioning and transmutation. They are co-organised by the NEA Secretariat and major laboratories in Member countries.

An overview of NEA activities on partitioning and transmutation and relevant publications are available at http://www.nea.fr.html/pt/welcome.html

These proceedings include the papers presented at the 6th Information Exchange Meeting in Madrid (Spain) on 11-13 December 2000, held in co-operation with the European Commission. The opinions expressed are those of the authors only, and do not necessarily reflect the views of any OECD/NEA Member country or international organisation. These proceedings were co-edited by OECD/NEA and the European Commission. They are published on the responsibility of the Secretary-General of the OECD.

TABLE OF CONTENTS

EXECUTIVE SUMMARY

More than 160 participants from 15 countries and three international organisations gathered for the sixth time since 1989 to exchange information on the various aspects of partitioning and transmutation (P&T). This 6th OECD/NEA Information Exchange Meeting was generously hosted by CIEMAT and ENRESA and was held in co-operation with the European Commission.

Since 1989, the OECD/NEA has conducted an international Information Exchange Programme on Actinide and Fission Product Partitioning and Transmutation within which the most visible activity has been the biennial Information Exchange Meeting. Previous meetings were held in Mito City (1990), Argonne National Laboratory (1992), Cadarache (1994), Mito City (1996) and Mol (1998). This 6th meeting closed the first ten years of information exchange and opened the discussion on what the next years should bring. Indeed, while the objectives of these meetings have remained unchanged, the focus has changed according to the developments and expectations in the subject field.

This 6th meeting highlighted the developments in P&T according to the themes which were the subject of 5 sessions, i.e.:

- International collaboration and institutional aspects were addressed in the first session "Overview of National and International Programmes on P&T".

- The role of P&T in advanced nuclear fuel cycles and especially the link with waste management was considered in Session II "The Nuclear Fuel Cycle and P&T".

- Partitioning was addressed in Session III.

- Session IV addressed basic physics aspects (i.e. nuclear data and experiments), material and fuel developments, as well as an insight into specific reactor physics aspects of transmutation systems.

- Finally, Session V addressed several concepts of transmutation systems and highlighted especially the safety considerations.

An overview of the presentations and discussions during these sessions has been given by the session chairs. This executive summary brings an overview of the discussions held during the technical sessions and reports on the panel discussion during the closing session.

The discussions during the closing session highlighted the current issues and situation of P&T research and development in the individual countries and on an international level. A lively session addressed several aspects:

- The increasing need for interaction between the P&T and the radioactive waste management community.

- The organisation, planning and role of international collaboration in the development of P&T and especially of ADS.

- Multi-purpose or single-purpose development of ADS.

- The need for consensus on figures of merit for P&T, etc.

P&T has made various advances in the past ten years. It has been shown that separation of the minor actinides is a feasible process, exhibiting high separation factors at the laboratory level; figures of merit are needed in the future. Indications are that all separation processes, hydro-reprocessing as well as pyro-reprocessing, are becoming so complex that simplicity and thus cost-reductions should become prime criteria for future development. A large variety of transmutation concepts have been proposed in response to which partitioning has been continuously adapted (increasing recovery yields). As further development in partitioning and in transmutation becomes more expensive, choices on performance and specific objectives (i.e. criteria and indicators) for P&T will be needed. Future reprocessing processes are closely linked to fuel-fabrication aspects (fissile content of fuel, type of fuel). Pyro-processes will most probably be on a batch basis with low throughput because of the limiting transfer capacity between process steps. Continuous pyro-processes may be envisaged in a few decades time. The specific question of whether to include curium in a transmutation scheme influences the partitioning processes to be developed to a pre-industrial stage. The discussions indicated that no common view exists in that respect. Whereas fuel fabricators indicated a receptiveness to separate the curium and to store it in order to decay to plutonium for recycling after about 100 years; others indicated that this (concentrated) storage would involve difficult problems such as criticality, heat removal, doses.

The discussion also covered the question of why P&T might be needed and the objectives involved. P&T may be justified in order to increase the utilisation of the natural uranium resources while minimising the possible impact of the long-lived radionuclides on the biosphere. Different strategies could be envisaged. Multiple recycling has to be considered both in "double strata" strategies and in standard critical reactors to consistently reduce the final potential radiotoxicity of waste (by factors up to 200-300). "Once-through" strategies would only allow lesser reductions (factors 30-40). It was pointed out that, to obtain significant P&T effectiveness in reducing the potential radiotoxicity of waste in a deep geological repository, process losses in the fuel cycle as low as 0.1% for all transuranics are required.

Another option which might improve the disposal strategy would be to adopt partitioning and conditioning (P&C) in which specific conditioning of the minor actinides (MAs) and some of the long-lived fission products (LLFPs) is applied.

Even before deploying P&T in a fuel cycle, questions arise about the role of plutonium in a truly sustainable nuclear option. Sustainable development may depend on a plutonium economy in order to extend the time span over which the natural resources are available to generate electricity. In this context, one should mention that the thorium option also involves specific problems that will require substantial developments in the area of fuel cycle.

One may not forget that the time scales for implementation of P&T and of geological disposal are very different. While P&T could indeed reduce the mass and radioactivity of long-lived waste, for example by a factor of 100, achieving this reduction would take several decades to equilibrium and a few centuries to achieve the potential impact on the final waste disposal. This means an institutional decision to act on P&T early and for several decades in order to reduce potential very long-term impacts. Currently, the time period from decision to implementation of geological disposal is only about 40-50 years.

There has been growing interest and activity in basic science supporting P&T. This is particularly the case in the high power proton accelerator field where a synergy can be envisaged between transmutation and other applications of intense neutron sources using these accelerators. In the nuclear physics field, nuclear data measurements have experienced a revival as deficiencies in data are

identified. In the fuel area, laboratories for MA-handling are being built in which, over the next decade, options should be experimentally proven. A difficulty however has been identified, i.e. the reduced number of fuel irradiation facilities. Other crucial elements in the development of fuel and materials relate to corrosion and irradiation resistance; these are supported in well-structured R&D programmes. New technologies emerging from other fields of science and technology could also play an interesting role in the R&D for P&T (hollow fibres, nano-materials, other uses of Pb-Bi).

The panel discussion indicated that streamlining and prioritisation of R&D is needed in the future as P&T-related R&D starts to demand more resources and, as mentioned above, would benefit of a convergence of ideas and consensus on the selection of desirable fuel cycles schemes. Choices will need to be made in the coming years. Criteria and indicators will therefore need to be identified for future guidance of work. Basic science developments (materials, nuclear data and simulation, chemistry, etc.) will be very important and this will demand an international collaborative effort. The link with waste management was considered as a priority for future work by international programmes. Finally, simplicity in P&T should be sought.

The closing session ended with the statement that "the P&T community will have to make up its mind!" in the near future in order to keep the R&D well supported and well focused on the ultimate objective. This issue will be the focus of the 7th Information Exchange Meeting which is tentatively planned to be held at the end of the year 2002 and will be hosted by the Republic of Korea.

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SCIENTIFIC PROGRAMME[1]

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Monday 11 December 2000

8:30-9:15   Registration

9:15-9:45   **Welcome addresses**

       Mrs. Lucila Izquierdo, General Secretary of External and Institutional Relations, CIEMAT
       Mr. M. Hugon, Co-ordinator, P&T and Future Systems, EC-DGXII
       Mr. Ph. Savelli, Deputy Director, Science Computing and Development, (OECD/NEA)

9:45-11:00   **Session I: Overview of National and International Programmes**
          J.L. Diaz-Diaz (CIEMAT) – Ph. Savelli (OECD/NEA)

- *Research and Development of Technologies for Partitioning and Transmutation of Long-lived Nuclides in Japan –Status and Evaluation–*, S. Aoki (STA).
- *French Research Programme to Reduce the Mass and Toxicity of Long-lived Highly Radioactive Nuclear Waste*, P. Bernard *et al.* (CEA).
- *The Status of the US Accelerator Transmutation of Waste Programme*, J. Bresee *et al.* (DOE, ANL).

11:00-11:20 Coffee break

11:20-12:50
- *IAEA Activities in the Area of Emerging Nuclear Energy Systems*, A. Stanculescu (IAEA).
- *Accelerator Driven Sub-critical Systems for Waste Transmutation: Co-operation and Co-ordination in Europe and the Role of the Technical Working Group*, M. Salvatores *et al.* (TWG)
- *Partitioning and Transmutation in the EURATOM Fifth Framework Programme*, M. Hugon *et al.* (EC).
- *Activities of OECD/NEA in the Frame of P&T*, L. Van den Durpel *et al.* (OECD/NEA).

12:50-13:00   **Introduction of poster sessions**

13:00-14:30 Lunch

14:30-17:00   **Session II: The Nuclear Fuel Cycle and P&T**
           J J. Bresee (DOE) - P. Schapira (CNRS)

       Overview paper *Closing the Nuclear Fuel Cycle: Issues and Perspectives,*
            P. Wydler (PSI) and L. Baetslé (SCK•CEN)

- *Recent Topics in R&D for the OMEGA Project in JAERI*, T. Osugi *et al.* (JAERI).
- *Transuranics Transmutation on Fertile and Inert Matrix Lead-bismuth Cooled ADS*, E. González *et al.* (CIEMAT).
- *Actinide and Fission Product Burning in Fast Reactors with a Moderator*, I. Krivitski *et al.* (IPPE).
- *Assessment of Nuclear Power Scenarios Allowing for Matrix Behaviour in Radiological Impact Modelling of Disposal Scenarios*, H. Boussier *et al.* (CEA).
- *Disposal of Partitioning-transmutation Wastes with Separate Management of High-heat Radionuclides*, Ch.W. Forsberg (ORNL).
- *The AMSTER Concept*, D. Lecarpentier *et al.* (EdF, Ministère de l'Éducation, CEA, CNRS).

17:00-17:20 Coffee break

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[1] The four scientific Sessions (II-V) included an invited overview paper, which is reproduced in this booklet. The contributed papers are available on the CD-ROM.

17:20-19:05
Session III: Partitioning
J.P. Glatz (ITU) - J. Laidler (ANL)

Overview paper *Overview of the Hydro-metallurgical and Pyro-metallurgical Processes Studied Worldwide for the Partitioning of High Active Nuclear Wastes*, Ch. Madic (CEA)

Sub-session III-A: Aqueous Reprocessing

- *Partitioning-separation of Metal Ions Using Heterocyclic Ligands*, M.J. Hudson *et al.* (University of Reading, CEA).
- *Separation of Minor Actinides from a Genuine MA/LN Fraction*, J.P. Glatz *et al.* (EC/JRC/ITU).
- *Partitioning Anionic Agents Based on 7,8-Dicarba-Nido-Undecaborate for the Remediation of Nuclear Wastes*, F. Teixidor *et al.* (Institut de Ciència de Materials de Barcelona).

19:05-19:15   End of the session and summary of the first day

Tuesday 12 December 2000

9:00-10:35
Session III: Partitioning (Cont'd)

Sub-session III-B: Dry Reprocessing

- *Pyrochemical Processing of Irradiated Transmuter Fuel*, J. Laidler *et al.* (ANL, DOE).
- *R&D of Pyrochemical Partitioning in the Czech Republic*, J. Uhlir (Řez).
- *Demonstration of Pyrometallurgical Processing for Metal Fuel and HLW*, J.P. Glatz *et al.* (CRIEPI, JRC/ITU).
- *Development of Plutonium Recovery Process by Molten Salt Electrorefining with Liquid Cadmium Cathode*, M. Iizuka *et al.* (CRIEPI, JAERI).

10:35-10:55   Coffee break

10:55-13:05
Session IV: Basic Physics, Materials and Fuels
S. Pilate (BN) – H. Takano (JAERI)

Overview paper *Transmutation: A Decade of Revival Issues, Relevant Experiments and Perspectives*, M. Salvatores (CEA)

Sub-session IV-A: Basic Physics

- *Nuclear Data Measurements for P&T and Future Plans in JNC*, K. Furutaka *et al.* (JNC).
- *New Data and Monte Carlo Simulations on Spallation Reactions Relevant for the Design of ADS*, J. Benlliure (Universidad de Santiago de Compostela).
- *The Muse Experiments for Sub-critical Neutronics Validation and Proposal for a Computer Benchmark on Simulation of Masurca Critical and Sub-critical Experiments*, R. Soule *et al.* (MUSE collaboration).
- *OECD/NEA Benchmark Calculations for Accelerator Driven Systems*, M. Cometto *et al.* (CEA/PSI, OECD/NEA).

13:05-14:30   Lunch

14:30-15:40
Sub-session IV-B: Materials

- *Stainless Steel Corrosion in Lead-bismuth under Temperature Gradient*, D. Gómez Briceño *et al.* (CIEMAT).
- *Accumulation of Activation Products in Pb-Bi, Tantalum, and Tungsten Targets of ADS*, G.V. Kiselev *et al.* (RF SSC ITEP).
- *Thermal and Stress Analysis of HYPER Target System*, T.Y. Song *et al.* (KAERI, Seoul National University, Gyeongsang National University).

15:40-16:40   Poster session

16:40-18:20   **Sub-session IV-C: Fuels & Targets**

- *Fuel/Target Concepts for Transmutation of Actinides*, D. Haas *et al.* (EC/JRC/ITU).
- *Americium Targets in Fast Reactors*, S. Pilate *et al.* (BN, EdF).
- *Research on Nitride Fuel and Pyrochemical Process for MA Transmutation*,
  Y. Arai *et al.* (JAERI).
- *Transmutation Studies in France: R&D Programme on Fuels and Targets*,
  M. Boidron *et al.* (CEA, EdF).
- *Fission Product Target Design for HYPER System*, W.S. Park *et al.* (KAERI,
  Seoul National University).

18:20-18:30   End of the session and summary of the second day.

20:30   Conference Dinner kindly offered by CIEMAT and ENRESA.

Wednesday 13 December 2000

9:00-11:15   **Session V: Transmutation Systems and Safety**
                              Y. Arai (JAERI) – W. Gudowski (KTH)

              Overview paper *Safety Considerations in Design of Fast Spectrum ADS for Transuranic or
              Minor Actinide Burning: A Status Report on Activities of the OECD/NEA Expert Group*,
                                          D. Wade (ANL)

- *Safety Analysis of Nitride Fuels in Cores Dedicated to Waste Transmutation*,
  J. Wallenius *et al.* (KTH).
- *Aspects of Severe Accidents in Transmutation Systems*, H. Wider *et al.* (EC/JRC/Ispra).
- *A Simple Model to Evaluate the Natural Convection Impact on the Core Transients in
  Liquid Metal Cooled ADS*, A. D'Angelo *et al.* (ENEA, Politecnico di Torino).
- *Comparative Study for Minor Actinide Transmutation in Various Fast Reactor Core
  Concepts*, S. Ohki (JNC).
- *Study on a Lead-bismuth Cooled Accelerator Driven Transmutation System*,
  H. Takano *et al.* (JAERI).

11:15-11:40   Coffee break

11:40-13:30
- *Transuranics Elimination in an Optimised Pebble-bed Sub-critical Reactor*,
  P. León *et al.* (ETSI, Soreq NRC).
- *Transmutation of Nuclear Wastes with Gas-cooled Pebble-bed ADS*, A. Abánades *et al.*
  (LAESA, ORNL, Universidad Politécnica de Valencia and Madrid).
- *Myrrha, a Multi-purpose ADS for R&D as First Step Towards Waste Transmutation –
  Current Status of the Project*, H. Aït Abderrahim *et al.* (SCK•CEN, IBA).
- *ADS: Status of the Studies Performed by the European Industry*, B. Carluec *et al.*
  (Framatome, Ansaldo).
- *Helium-cooled Reactor Technologies for Accelerator-transmutation of Nuclear Waste*,
  A. Baxter *et al.* (General Atomics).

13:30-14:45   Lunch

14:45-17:00   **Closing session: P&T in the Future?**
                              V. Gonzalez (ENRESA) – M. Salvatores (CEA)

- Session chair summaries.
- Chairman IEM Summary introducing key-questions and discussion.
- Closing remarks by Mr. M. Hugon, EC.
- Closing remarks by Mr. L. Van den Durpel, OECD/NEA.
- Closing address by Dr. A. Colino, President ENRESA.

17:00   End of the 6th IEM.

Poster session: Partitioning
M.J. Hudson (University of Reading)

- *Studies on Behaviour of Selenium and Zirconium in Purex Process*, A.G. Espartero *et al.* (CIEMAT).
- *Solubilization Studies of Rare Earth Oxydes and Oxohalides. Application of Electrochemical Techniques in Pyrochemical Processes*, C. Caravaca *et al.* (CIEMAT, Universidad de Valladolid).
- *Calix[6]arenes Functionalised with Malondiamides in the Upper Rim as Possible Extractants for Lanthanide and Actinide Cations*, S. Esperanza *et al.* (Universidad Autónoma de Madrid).
- *Actinide(III)/Lanthanide(III) Partitioning Using N-PR-BTP as Extractant: Extraction Kinetics and Extraction Test in a Hollow Fiber Module*, A. Geist *et al.* (FZK).
- *The Potential of Nano- and Microparticles for the Selective Complexation and Separation of Metal Ions/Radionuclides*, G. Grüttner *et al.* (Micromod, Universität Potsdam).
- *New Extractants for Partitioning of Fission Products*, B. Grüner *et al.* (Institute of Inorganic Chemistry, Nuclear Research Institute, Katchem).
- *Influence of Intermediate Chemical Reprocessing on Fuel Lifetime and Burn-up*, A.S. Gerasimov *et al.* (RF SSC ITEP).
- *Recent Progresses on Partitioning Study in Tsinghua University,* C. Song *et al.,* (Tsinghua University).

Poster session: Basic Physics: Nuclear Data and Experiments
and
Materials, Fuels and Targets
P. D'Hondt (SCK•CEN)

- *Design and Characteristics of the n_TOF Experiment at CERN*, D. Cano-Ott (CIEMAT).
- *Recent Capture Cross-sections Validation on ^{232}Th from 0.1 eV to 40 keV and Self-shielding Effect Evaluation*, A. Billebaud *et al.* (CNRS/IN2P3/UJF).
- *Double Differential Cross-section for Protons Emitted in Reactions of 96.5 MeV Neutrons on Enriched ^{208}Pb Targets*, F.R. Lecolley *et al.* (CNRS/IN2P3, Subatech, Uppsala University, ULB, IreS).
- *Measurements of Particule Emission Spectra in Proton Induced Reactions of Interest for the Development of Accelerator Driven Systems*, N. Marie *et al.* (CNRS/IN2P3, Subatech, IPN, ULB, CEA, IreS).
- *Intermediate Energy Neutron-induced Fission Cross-sections for Prospective Neutron Production Target in ADS*, A.N. Smirnov *et al.* (Khlopin, Uppsala University).
- *Nucleon-induced Fission Cross-sections Calculations and Development of Transmutation-activation Data Library for Transitive Energy Region 20-200 MeV*, S. Yavshits *et al.* (Khlopin, Institute of Nuclear Power Engineering).
- *Neutron Radiative Capture Cross-section of ^{232}Th in the Energy Range from 0.06 to 2 MeV*, D. Karamanis *et al.* (CEN, ISN, CERN).
- *Determination of the Neutron Fission Cross-section for ^{233}Pa from 0.5 to 10 MeV Using the Transfer Reaction ^{232}Th(^{3}He, pf)^{234}Pa*, M. Petit *et al.* (CEN, ISN).
- *Measurement of Double Differential Cross-sections for Light Charged Particles Production in Neutron Induced Reactions at 62.7 MeV on Lead Target*, M. Kerveno *et al.* (Subatech, IPN, Institute of Atomic Physics Bucharest, LPCC Caen).
- *High and Intermediate Energy Nuclear Data for Accelerator Driven Systems – The HINDAS Project*, J.P. Meulders *et al.* (UCL, KVI, University of Santiago de Compostela, CEA, Université de Liège, Subatech, FZ Juelich, NRCG, LPCC/CNRS/ISMRa/Université de Caen, ZSR-Hannover, Uppsala University, Darmstadt, Braunschweig, ETHZ-Zurich, PSI).
- *A Study on Burnable Absorber for a Fast Sub-critical Reactor HYPER*, Y.H. Kim *et al.* (KAERI, Seoul National University).

Poster session: Transmutation Systems
W.S. Park (KAERI)

- *MA and LLFP Transmutation in MTRs and ADSs: The Typical SCK•CEN Case of Transmutations in BR2 and Myrrha. Position with Respect to the Global Needs*, Ch. De Raedt *et al.* (SCK•CEN).
- *Enhancement of Actinide Incineration and Transmutation Rates in ADS EAP-80 Reactor Core with MOX PuO$_2$&UO$_2$ Fuel,* S. Kaltcheva-Kouzminova *et al.* (Petersburg Nuclear Physics Institute, ENEA).
- *Remarks on Kinetics Parameters of a Sub-critical Reactor for Nuclear Waste Incineration*, J. Blázquez (CIEMAT).
- *Noise Method for Monitoring the Sub-criticality in Accelerator Driven Systems*, J.L. Muñoz-Cobo *et al.* (Universidad Politécnica de Valencia, ORNL, LAESA).
- *Molten Salts as Possible Fuel Fluids for TRU Fuelled Systems: ISTC #1606 Approach*, V. Ignatiev *et al.* (RRC-Kurchatov, VNIITF).
- *Comparative Assessment of the Transmutation Efficiency of Plutonium and Minor Actinides in Fusion/Fission Hybrids and ADS*, M. Dahlfors *et al.* (Uppsala University, CERN).
- *Deep Underground Transmutor (Passive Heat Removal of LWR with Hard Neutron Energy Spectrum),* H. Takahashi (BNL).
- *Radiation Characteristics of PWR MOX Spent Fuel After Long-term Storage Before Transmutation in Accelerator Driven Systems*, B.R. Bergelson *et al.* (RF SSC ITEP).
- *Radiation Characteristics of Uranium-Thorium Spent Fuel in Long-term Storage for Following Transmutation in Accelerator Driven Systems*, B.R. Bergelson *et al.* (RF SSC ITEP).
- *International Co-operation on Creation of a Demonstration Transmutation Accelerator Driven System*, A.S. Gerasimov *et al.* (RF SSC ITEP).
- *On Necessity of Creation of Accelerator Driven System with High Density of Thermal Neutron Flux for Effective Transmutation of Minor Actinides*, A.S. Gerasimov *et al.* (RF SSC ITEP).
- *New Original Ideas on Accelerator Driven Systems in Russia as Base for Effective Incineration of Fission Products and Minor Actinides*, G.V. Kiselev (RF SSC ITEP).
- *Conditions of Plutonium, Americium and Curium Transmutation in Nuclear Facilities*, A.S. Gerasimov *et al.* (RF SSC ITEP).
- *Demonstration Accelerator Driven Complex for Effective Incineration of ^{99}Tc and ^{129}I*, A.S. Gerasimov *et al.* (RF SSC ITEP).
- *Critical and Sub-critical GT-MHRs for Waste Disposal and Proliferation-Resistant Fuel Cycles*, A. Ridikas *et al.* (CEA).
- *The Use of Pb-Bi Eutectic as the Coolant of an Accelerator Driven System,* A. Peña *et al.* (ETSII, JRC/Ispra).
- *One Way to Create Proliferation-protection of MOX Fuel,* V.B. Glebov *et al.* (MEPhI, SEC NRC).
- *Transmutation of Long-lived Nuclides in the Fuel Cycle of BREST-Type Reactors,* A.V. Lopatkin *et al.* (RDIPE).

WELCOME ADDRESS

Lucila Izquierdo
General Secretary of External and Institutional Relations
CIEMAT
Avda. Complutense 22, 28040 Madrid, Spain

Ladies and Gentlemen,

Dear Michel Hugon, Phillipe Savelli, dear participants,

It is my pleasure, as Secretary of External and Institutional Relations of CIEMAT and in the name of its Director General, to welcome all of you to CIEMAT for the 6th OECD/NEA Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation.

We are glad that Madrid joins the list of cities that have hosted this series of meetings that started in Mito and have got consecutive success at Argonne, Cadarache and Mol.

We would like to thank the organisers both NEA/OCDE and the European Commission for their invitation for the joint hosting of the meeting by CIEMAT and ENRESA (the Spanish body for radioactive waste management).

CIEMAT is the Spanish Public Organism for Research and Technological Development supported by the Ministry of Science and Technology responsible of finding solutions to improve the use of resources and energy generation systems, to develop alternative energy sources and to solve the problems of the Spanish companies regarding energy and its effects on the environment.

CIEMAT is largely involved in the research on future and present nuclear energy sources through the programmes of Nuclear Fusion and Nuclear Fission, whose activities include many projects related to the back-end of the nuclear fuel cycle.

At CIEMAT we find in Partitioning and Transmutation (P&T) one very interesting element for the Nuclear Waste management. Ideally, it will allow to achieve large reduction on the inventories of long-lived radioactive wastes contained in the nuclear waste, in particular the actinides, reducing the concerns about our use of nuclear energy for future generations. Not to forget the positive effect on the public acceptance of nuclear waste management programmes and the potential capability to produce huge amounts of electricity.

After the participation of CIEMAT in the FEAT and TARC experiments at CERN related to the Energy Amplifier project, already in 1994, the Nuclear Fission Department has initiated a wide P&T research programme in 1997.

This programme includes six main lines: the advanced hydro-metallurgic reprocessing techniques to separate all the Transuranium elements and some long-lived fission fragments from the spent fuel of the present nuclear power plants; the pyro-metallurgic technologies for new ADS fuel recycling; the corrosion of materials in molten lead alloys; the behaviour of materials in extreme irradiation and temperature conditions as expected for the spallation target windows in ADS systems; the computer simulation of transmutation devices and strategies; and the participation on basic experimental research for transmutation and ADS.

All this work is strongly integrated in the international research on P&T, including the participation on 6 contracts of the present 5th Framework Programme of the European Union, and on the activities on OCDE/NEA, IAEA and the European ADS Technical Working Group. In addition, we have established bilateral collaboration agreements directly with CEA and with the ITU through ENRESA. Further contracts with CRIEPI and contacts with several USA laboratories open our activities in the field outside Europe.

Inside Spain, all this research is performed in close collaboration with ENRESA and several Universities distributed over the Spanish geography.

I wish that the efforts of all of you present here, the laboratories and institutions from were you are coming, and the international organisations represented in the room, will make soon the P&T dream promises a reality. In this sense the opportunities of information exchanges provided by OECD/NEA and other forums, and the continuation and increase of the support from the national and international funding agencies, as well as the progressive involvement of industry, are key elements for the success. I am sure that, with your collaboration, this meeting will represent an important step forward in this direction.

I hope that beside the intense work schedule of the meeting you can find some time to enjoy Madrid and the surrounding cities.

I want finally thank all the speakers and poster authors for sharing their work and results with all of us, and for their collaboration in the organisation of the meeting, and to all of you for coming, and I hope, for your active participation on the meeting.

Thank you and welcome to CIEMAT.

WELCOME ADDRESS

Michel Hugon

Co-ordinator, P&T and Future Systems
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Ladies and Gentlemen,

It is a great pleasure for me to welcome you today to this 6th Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation.

I would also address more especially my warm welcome to the many young and enthusiastic people, who are starting to work in this very exciting field.

It is the second time that the European Commission co-organises this Information Exchange Meeting with the Nuclear Energy Agency of the OECD. I would like to express here my deep satisfaction of the excellent relationships that we have established with OECD/NEA in the field of Partitioning and Transmutation (P&T), where we share a common understanding. This synergy enables both international organisations to maximise the co-operation in this area between their different member countries and also to invite representatives of China and Russia to participate to this meeting.

I would also like to thank both CIEMAT and ENRESA for hosting this meeting and for the hard preparatory work they have done to make this meeting on its way to a great success.

I am happy that this meeting is taking place in Spain, a country which is very active in the field of nuclear waste management and disposal. Earlier this year, in March, an International Conference on the Safety of Radioactive Waste Management was held in Cordoba and organised by IAEA in co-operation with the EC, the OECD/NEA and the World Health Organisation. From what I heard, it was a great success.

As you all know, P&T aims at reducing the inventories of long-lived radionuclides in radioactive waste by separating them from the waste and then transmuting them into radionuclides with a shorter lifetime. However, there will be always a need for appropriate geological disposal for the existing high level waste and the waste containing the long-lived radionuclides, which cannot be transmuted. Nevertheless, the techniques used to implement P&T could alleviate the problems linked to waste disposal. P&T is still at the research and development stage, which will require long lead-times.

There has been a renewal of interest in P&T worldwide at the end of the eighties (OMEGA programme in Japan, SPIN programme in France). Meanwhile, sufficient progress has been made in accelerator technology to consider as feasible the use of accelerator driven systems (ADS) for waste incineration. Proposals to develop ADS have been made during the nineties by the Los Alamos National

Laboratory in the USA with the ATW (Accelerator driven Transmutation of Waste) programme, by CERN in Europe with the Energy Amplifier (EA) and by JAERI in Japan. In addition, there is a number of research activities on ADS going on in several EU countries (Belgium, France, Germany, Italy, Spain, Sweden), Czech Republic, Switzerland, Korea and Russia.

The interest for P&T in the EU is reflected in the increase of funding in this area over the last three EURATOM Framework Programmes, 4.8, 5.8 and about 26 million € for the 3rd, 4th and 5th Framework Programmes respectively. Research work is also carried out at the Joint Research Centre of the EC, mainly at the Institute for Transuranium Elements in Karlsruhe.

Ladies and Gentlemen, I would also like to take this opportunity to inform you about some recent thoughts about the future energy supply in Europe that has been developed by the European Commission. At the Industry-Energy Council on 5 December, the Vice-President of the Commission in charge of Energy and Transport, Ms. Loyola de Palacio, presented a Green Paper entitled "Towards a European Strategy for the Security of Energy Supply" in order to launch a debate.

The starting points are:

- If no measures are taken, in the next 20 to 30 years, about 70% of the Union's energy requirements will have to be covered by imported products (today 50%). The energy dependence of the Union will be increasingly alarming. This will affect all sectors of the economy.

- The fight against the climate change is difficult: inversion of the trends is more difficult than it appeared to be three years ago. Thus, while the Union stabilised its emissions of greenhouse gases in 2000, the forecasts of the European Environment Agency consider that they will increase by 5.2% between now and 2010.

The Green Paper offers for discussion a plan for a long-term energy strategy, in 5 main fields:

- A genuine change in consumer behaviour and energy consumption.

- A truly alternative transport policy.

- Doubling the share of renewable energies from 6 to 12% in the energy balance between now and 2010 (financial measures).

- Solutions at the Community level (e.g. reinforced strategic oil and gas stocks, a fiscal policy for energy to steer towards more environmentally friendly sources).

- To analyse the medium-term contribution of nuclear power taking into account the phasing out decisions of the majority of the Member States and issues related to waste management, global warming, security of supply and sustainable development.

It is proposed that the European Union must retain its leading position in the field of *civil* nuclear technology, in order to retain the necessary expertise and develop more efficient fission reactors and enable fusion to become a reality.

Research on the technologies of waste management and their practical implementation under optimum safety conditions has actively to be continued. This applies to geological disposal as well as to partitioning and transmutation.

Ladies and Gentlemen, our work these coming days are thus of great interest. I wish you all a very fruitful and successful meeting as well as a nice stay in Madrid.

Thank you for your attention.

WELCOME ADDRESS

Philippe Savelli
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Ladies and Gentlemen,

It is a real pleasure for me to welcome you to this 6th Information Exchange Meeting organised by the OECD Nuclear Energy Agency (NEA).

Back in 1988, the Japanese government asked the NEA to launch an international information exchange programme on partitioning and transmutation (P&T). At that time, only a few countries were really active in this field. In the 1960s and 1970s, preliminary studies and experiments had been conducted in the USA, Japan and within several European countries, as well as the European Commission. The conclusions of some of the assessments published in the late 1970s and early 1980s clearly stated that the transmutation of minor actinides was considered theoretically possible, but that it was not obvious whether the potential long-term risk reduction for the waste disposal site was overall beneficial, because of the increase in short-term risks for the workers. Those studies also concluded that there were no obvious direct cost or safety incentives for P&T of actinides for waste management purposes. However, it was recognised that further investigation of advanced reprocessing techniques for conditioning of plutonium and minor actinides would be valuable.

A second phase of interest in P&T emerged at the end of the 1980s, partly based on the growing awareness of the difficulties in licensing large nuclear waste repositories and certain delays in the related R&D projects. There was a need to re-examine the validity of the P&T option in the light of the more recent results. This led Japan, France, USA and other countries to start new studies, complemented by an experimental R&D programme.

In the early 1990s, new assessment reports were published by France and the USA, as well as studies conducted under the auspices of IAEA or EC. The Nuclear Energy Agency undertook a systems study in early 1996 and published the *Status and Assessment Report on Minor Actinide and Fission Product Partitioning and Transmutation* in April 1999. It is worth emphasising four of the main conclusions of this report:

- P&T will not replace the need for appropriate geological disposal of high level waste.

- The recycling of plutonium and minor actinides could stabilise the transuranium nuclide inventory. However, multiple recycling of transuranium nuclides is a long-term venture for which it may take decades to reach equilibrium.

- Partitioning methods for long-lived radiotoxic elements have been developed on a laboratory scale and could be very useful to condition separated long-lived nuclides in appropriate matrices or in irradiation targets. These matrices could be selected to be less soluble than glass in geological media.

- Last but not least, fundamental R&D for the implementation of P&T needs long lead-times and would require large investments in dedicated fast neutron spectrum devices, extension of reprocessing plants and the construction of remotely manipulated fuel and target fabrication plants.

During the 1990s, we also noticed a renewed interest in accelerator driven systems (ADS). Today, as the participation in this meeting shows, several countries active in P&T emphasise the ADS-line. We have seen increasing international activity, especially in Europe; a growing number of bilateral and multilateral co-operations have been established. Examples of these are the collaboration of Japanese institutes with European Joint Research Centres, the 5th Framework R&D projects sponsored by the European Commission, the Technical Working Group in Europe under the chairmanship of Carlo Rubia, the ISTC activities with our Russian colleagues and the foreseen increased collaboration between USA and France.

It was in response to this emerging interest that the NEA launched new studies under the auspices of its Nuclear Development and Nuclear Science Committees. Both committees have, together with the NEA Data Bank, developed several well co-ordinated activities, covering a diverse set of issues related to P&T, such as nuclear data and benchmarks, partitioning techniques and also more strategic systems studies. Today, more exchange with the NEA Radioactive Waste Management Committee is sought and we view Session II (The Nuclear Fuel Cycle and P&T) of this meeting as a welcome step in this direction. A new Working Party on Scientific Issues in P&T has been launched and, in fact, held its first meeting yesterday here in Madrid. Other Working Parties and Expert Groups, as well as specific Workshops and Information Exchange Meetings will remain part of our work programme and they will be tailored in response to your demands. In addition, our P&T activities are now organised as a horizontal project and in that respect, a single NEA web page on P&T will announce all our projects and programmes in the future. A separate presentation in Session I this morning will cover our activities more in detail.

Ladies and Gentlemen, in the light of these past and ongoing developments, I consider it appropriate to raise two items that I regard as important for future activities on P&T.

The first is the increasing importance that nuclear power could play in response to the need for a more sustainable energy development. During the debate on climate change in den Hague two weeks ago, some delegates indicated that nuclear should be recognised as part of a future energy mix. It is for example encouraging to note that the European energy and transport commissioner, Ms. Loyola de Palacio, recognises this role of nuclear, despite some EC countries having embarked on a nuclear phase-out strategy.

If the concern for our future would be translated into a continued demand for nuclear energy, several of the developments discussed in this meeting could help reply to some of the questions regarding nuclear energy. The public would only accept an increased use of nuclear, if today's concerns about safety, waste and proliferation could be satisfied. P&T is one approach that could contribute to the sustainability of nuclear energy.

A second item relates to the assessment of P&T and especially the question of objectives and indicators to be applied. Society demands more clear objectives and indicators before embarking on

developments. This will surely become the case if society accepts increased reliance on nuclear energy. Society claims an economically viable energy resource, showing an excellent safety level, dealing in an efficient way with waste and other residuals and finally respecting the environment in the short and long term. Therefore, nuclear, and clearly also P&T, will have to face this kind of evaluation. We consider that an honest reflection on applicable objectives and criteria would be a worthwhile undertaking in the future.

In this context, we are all aware of the declining trend in nuclear education and availability of infrastructure. In today's context of deregulation and increasing competitive pressure on the utilities and on research institutes, P&T will have to face this additional challenge of limited resources and infrastructure. However, a positive factor is that the different P&T projects have presented new and challenging scientific problems that are attracting young scientists to enter the nuclear field.

This Information Exchange Meeting is again in co-operation with the European Commission and I wish to thank them for their valuable support. I believe that the co-operation we have established is a good example of how scarce resources can be shared, based on mutual understanding.

In ending my talk, I would first of all like to thank CIEMAT and its Director-General Dr. Felix Yndurain Muñoz, as well as ENRESA and its President Dr. Antonio Colino, who are jointly hosting this meeting and have ensured the success of, what I am convinced, will be a very enjoyable stay here in Madrid.

Ladies and Gentlemen, may I wish you a fruitful meeting. The numerous participation gives me confidence that the scientific programme has captured your interest and that these Information Exchange Meetings respond to your wishes. I am glad that we can also welcome participation from non-OECD countries. I invite all of you to help us shape our activities in the future and your advice or comments will certainly be taken into account and be reflected in our future programme of work.

Thank you for your attention.

SESSION I

Overview of National and International Programmes

Chairs: J.L. Diaz-Diaz (CIEMAT) – Ph. Savelli (OECD/NEA)

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SUMMARY

This session gave an overview of the major activities in the field of partitioning and transmutation (P&T) in some of the Member countries, i.e. Japan, France and the USA as well as by international groups (TWG) and organisations (EC, IAEA, NEA).

Significant progress has been made in many countries since the previous Information Exchange Meeting in Mol, November 1998. The OMEGA programme in Japan underwent a review by the AEC's Advisory Committee on Nuclear Fuel Cycle Back-End Policy that released a report entitled *Research and Development of Technologies for Partitioning and Transmutation of Long-lived Nuclide: Status and Evaluation Report* (March 2000). This report gave an overview of ongoing and planned R&D by the different governmental and private organisations in Japan and identified future activities. The report concluded that the R&D at the three research institutes (JAERI, JNC and CRIEPI) had resulted in the establishment of processes for P&T technology with the expected performance. The aims of the Phase I R&D have thus been achieved according the expectations included in the OMEGA-programme. R&D in Phase II has experienced some delays, the primary reason being that Japan is redefining its entire FBR programme, and facilities to handle MAs and other materials have yet to be constructed. The report also mentioned that in carrying out further R&D, it would be important to promote co-operation with domestic and foreign organisations in order that experimental facilities – including those for engineering experiments – can be used efficiently.

The three Japanese research organisations indicated in the report that a common issues is the implementation of experiments to demonstrate processes using actual HLLW. In addition, the preparation of a database on fuel irradiation behaviour for performance analysis and the development of fuel fabrication technology were considered an issue as well.

While several R&D activities are planned in the future, e.g. economic aspects, P&T technology as part of the fuel cycle, system design and others, it was considered appropriate to conduct R&D in these areas on a time schedule compatible with nuclear fuel cycle R&D. At present, feasibility studies on commercialised FBRs and related fuel cycles system is being carried out under the collaborative efforts of JNC, electric utilities, CRIEPI and JAERI. In this study, R&D scenarios toward commercialisation of fast reactor system will be reviewed by about the year 2005. Thus, around the year 2005 is deemed to be an appropriate time to reconsider all R&D scenarios of P&T including the

use of FBRs for transmutation together with power generation, and the double-strata fuel cycle. Thereafter, progress, results and R&D policy will be checked and reviewed every five years or so. Evaluations of P&T technology system concepts, and reviews of introduction scenarios, should also be conducted.

In France, research is conducted with the goal of establishing by 2006 the scientific feasibility of transmutation in various types of nuclear reactors (PWR, innovative reactors) and the technical feasibility of intensive separation downstream from reprocessing at La Hague, as well as of the specific conditioning of separated long-lived radionuclides. The research is conducted in co-operation with partners in the nuclear industry, EdF, COGEMA and FRAMATOME, as well as with CNRS and universities. It benefits form significant co-operation at the European and international level. It is constantly evaluated by the National Evaluation Commission, which draws up and publishes an evaluation report annually. The procedure involves identifying a set of complementary scientific and technical solutions, which serve to define open and flexible strategies for the back-end of the cycle and lay the groundwork for a decision in 2006.

Concerning partitioning research, a reference programme has been defined for an advanced separation process for the main long-lived radionuclides in waste. The families of extractors were defined, the principal reference molecule synthesised, and their performances verified experimentally on real radioactive solutions in the ATALANTE facility in order to reach the stage of scientific feasibility in 2001. The next stage will be that of technical feasibility, moving from the molecule to the overall chemical process, which will be defined and validated in 2005. Experimental studies on fuel for the transmutation in fast neutron reactors have been launched, in particular in the PHENIX reactor, whose irradiation programme has focused on this research since 1998 and which therefore has been the object of inspection, renovation and maintenance, in view of a power increase in 2001.

In addition, teams at CEA and CNRS, in co-operation with industrial partners, have provided the technical data for a request for an experimental demonstration model of a hybrid reactor for transmutation, in a European and international framework.

Since the last meeting on P&T in 1998 in Mol, the US accelerator transmutation of waste (ATW) programme has changed significantly. Two years ago, the only effort was the preparation of a research plan for developing ATW technology. Today, a significant research effort is underway, and the US is seeking opportunities to collaborate with other national programmes. Transmutation R&D in the US initially has been focused on ADS and has involved a series of trade-off studies. In all cases, it has been assumed that uranium remaining in civilian spent fuel elements would be recovered, probably by a modified PUREX process called UREX. Initial studies of the UREX process have shown that the uranium product will meet US Class C requirements and could be disposed of as low level waste or be stored for possible future use in a nuclear fuel cycle. Various combinations of proton accelerator designs, spallation neutron sources, and transmutation target have been evaluated for technology readiness, and assumed irradiated targets have been studied for the effectiveness of chemical processing to recycle untransmuted long-lived isotopes. These evaluations have resulted in a base-line design which includes a linear proton accelerator, a lead-bismuth spallation target, and sodium-cooled metallic or ceramic dispersion transmutation target/blanket non-fertile fuel elements. Another interesting transmutation system design currently being evaluated consists of a "dual strata" approach which would involve a thermal critical reactor within which plutonium and minor actinides would fission and $^{99}Tc/^{129}I$ would be subjected to a thermal neutron flux.

The ATW programme during the Fiscal Year 2001 involved approximately a doubling of the Fiscal Year 2000 funding. This will allow an expansion of experimental programmes, and DOE's Office of Nuclear Energy, Science and Technology (NE) is actively seeking opportunities for

collaborative research with foreign ADS programmes. Meanwhile, the programme is being reorganised to combine the objectives of the DOE Defence Programme's Accelerator Production of Tritium (APT) programme with those of NE's ATW efforts. The combined programme is known as Advanced Accelerator Application (AAA), and it will be administered by NE. Congress has requested a report by March 1, 2001 on how the new activity will be carried out.

In Europe, a Technical Working Group (TWG) was established with the task of identifying the critical technical issues in which R&D is needed to develop an European demonstration programme of ADS over a 10-year time scale. The TWG, currently extended to ETWG, started in 2000 an intensive work aimed at defining a European Roadmap towards an experimental ADS, called XADS. The roadmap document is expected to be issued in the first half of 2001. The first goal of the roadmap is to propose a technological route to reduce the risks associated with nuclear waste, based on the transmutation of this waste using an ADS. The second and main goal of the roadmap is to prepare a detailed technical programme, with cost estimates, which could lead to the demonstration of an experimental ADS in 10 years. The programme as described in the roadmap will lead to a rationalisation of human resources and experimental facilities, a training ground for young researchers, the development of innovative fuels and reprocessing technology, spin-offs in the fields of accelerators, spallation sources, liquid metal technology, radioisotope production and actinide physics and chemistry. Hence, a final goal of the roadmap is to identify possible synergies and rationalisation that this programme could have within the nuclear community, indicate potential spin-offs, show how competence can be maintained in a currently stagnating field.

The European Commission has included in its previous Framework Programmes and in the current ongoing Fifth one, FP5 (1998-2002), several activities related to P&T. These activities address the chemical separation of long-lived radionuclides and the acquisitions of technological and basic data, necessary for the development of an ADS. Collaboration is also being implemented in this field between scientists of the European Union (EU) and the Commonwealth of Independent States (CIS).

The interest for P&T in the EU is reflected in the increase of funding over the EURATOM Framework Programme, i.e. 4.8, 5.8 and about 26 million € for the Third, Fourth and Fifth Framework Programmes respectively.

The P&T projects in FP5 have been grouped in three clusters. The experimental investigation of efficient hydro-metallurgical and pyrochemical processes for the chemical separation of long-lived radionuclides from HLW is carried out in the cluster on partitioning. The work on transmutation is mainly related to the acquisition of data, both technological and basic, necessary for the development of an ADS. The cluster on transmutation-technological support deals with the investigation of radiation damage induced by spallation reactions in materials, of the corrosion of structural materials by lead alloy and of fuels and targets for actinide incineration. In the cluster transmutation-basic studies, basic nuclear data for transmutation and ADS engineering design are collected and sub-critical neutronics are investigated. Additional projects will be funded in 2001, such as preliminary engineering design studies for an ADS demonstrator, complementary projects for technological support and networking.

The Commissioner responsible for research in the EC launched the idea of an "European Research Area" in January 2000. The intention is to contribute to the creation of better overall working conditions for research in Europe. In view of the future research programme, the EURATOM Scientific and Technical Committee has prepared a report on the strategic issues to be considered in the development of the appropriate nuclear energy research strategies in a 20-50 year perspective. In the area of nuclear fission, continued support should be given to maintain and develop the competence needed to ensure the safety of existing and future reactors. In addition, support should be given to explore the potential for improving present fission technology form a sustainable development point of

view, i.e. better use of uranium and other nuclear fuels, whilst reducing the amount of long-lived radioactive waste produced.

The session ended with two presentations by the international nuclear agencies, IAEA and NEA, showing their programmes of work in the area of P&T. A more detailed overview of their activities is given in the respective papers. The need for further international co-operation was once again repeated and strengthening of existing co-operations between countries as well as in the framework of international organisations has been requested in order to secure the effective use of scarce resources. This need for international co-operation will also be needed as prioritisation of R&D will be needed in the nearby future while decisions for new infrastructure will be requested.

The discussion during this session already highlighted the need for convergence of R&D in the future. A selection of fuel cycle schemes and an associated precise work-programme or strategy was requested where new international studies should focus on fuel cycle impacts and R&D needs.

One may remark that quite significant resources are spent in R&D in the various areas of P&T. While some countries perform in essence theoretical studies on possible P&T scenarios, others are embarked in experimental programmes and commit resources for construction of specific facilities. In the meantime, these countries also recognise that P&T is a long-term endeavour and that no immediate decisions are needed before about 2005. In France and Japan, a review of the P&T programme in the light of a long-term back-end policy is foreseen by the middle of this decade, while real implementation of a P&T scheme would still need an additional 20 years.

SESSION II

The Nuclear Fuel Cycle and P&T

Chairs: J. Bresee (DOE) – J.P. Schapira (CNRS)

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SUMMARY

The introduction by L.H. Baetslé and P. Wydler to this session gave a general overview of new partitioning and transmutation (P&T) fuel cycles based on present technologies (LWR, FR, uranium-plutonium fuels) altogether with their main issues. In order to minimise back end risks, plutonium management has to be addressed first. On the other hand, any P&T fuel cycle assessment has to take into account all the wastes and nuclear materials involved as well as their global impact (radiological, heat, secondary solid wastes, liquid and gaseous effluents). P&T strategy based on multirecycle in FRs leads to a waste radiotoxicity reduction factor of about 3 to 10 (depending on the date) if only plutonium is recycled and of about 100 if all the TRU are recycled within different scenarios (double strata, double components). These figures do not take into account the fuel cycle inventory radiotoxicity. They are strong constraints related to safety coefficients degradation (this can be alleviated by using sub-criticality), to neutron economy and to performances (inventory, burn-up achievable, losses). This paper shows that a factor 100 of mass reduction for TRU can be achieved if losses are less than 0.18% and burn-up greater than 15%, which are real challenges. In this respect, the normal PUREX process might be inadequate. Finally, new options such as pyrochemistry and the use of sub-criticality with accelerator driven systems, which allow the use of fertile free fuels, will probably be needed to achieve such performances. A comprehensive view of the principal actinide transmutation strategies is given using evolutionary and innovative approaches and according to the principal driving force: resource, waste, and proliferation. Concerning long-lived fission products, such as [129]I, ^{99}Tc, [135]Cs, [93]Zr and [126]Sn, this paper gives a good review of the various difficulties to include them in a P&T strategy (if chemically separated, most of them might be embedded in a more stable matrix than glass). Finally, the impacts of various P&T options on geological disposal are described.

The other papers describe some work related to P&T carried out in various laboratories as well as a new proposal. In the T. Osugi *et al.* paper, some recent results related to the JAERI double strata strategy are given: nitrides fuels, pyrochemical processes, 800 MW$_{th}$ ADS Pb-Bi target and coolant. Technical issues related to ADS will be studied using the experimental facility at KEK, with the prospect for commercial ADS around 2035. A 800 MW$_{th}$ ADS based on the lead coolant, Rubbia's concept (but with forced convection) is studied at CIEMAT in Spain, with the emphasis on new types of fuel (thorium and nitride inert matrix) applied to TRU burning in LWR (normal MOX fuel) then in ADS.

W. Forsberg proposes to reduce the high-level waste heat loading by chemically separating five heat generating nuclei: Cs, Sr (both to be put in an interim storage for heat decay) Pu, Am and Cm (to be transmuted by P&T). The remnant wastes containing the low-heat radionuclides will be geologically disposed at low cost (no interim storage and reduced disposal surface needed).

H. Boussier *et al.* calculates the potential risk of geological disposal within various P&T scenarios described in the overview paper. The originality of this paper is to consider not the global inventory radiotoxicity but that of the quantities which escapes from the waste deduced from the matrix fraction subjected to alteration over time. However, the difficulty is to get accurate values for such fractions.

The session ended up with the presentation by J. Vergnes *et al.* from EdF of a new concept of a molten salt fuel critical and thermal reactor which is able to produce energy with a very low amount of long lived wastes. In such a reactor called AMSTER (Actinides Molten Salt Transmuter) actinides are recycled, fission products and eventually ^{236}U continuously extracted. At equilibrium and in a fissile isogenerating mode, only fertile material (natural uranium or thorium) is fed into the reactor. AMSTER can incinerate TRU produced in PWR or recycle its own actinides only. First theoretical studies show that a reduction by several decades in the TRU quantities is expected, leading to a "clean energy" reactor, especially if the thorium fuel cycle is used.

CLOSING THE NUCLEAR FUEL CYCLE: ISSUES AND PERSPECTIVES

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Abstract

Partitioning and transmutation (P&T) aims at making nuclear energy more sustainable from the viewpoint of the back-end of the fuel cycle by minimising the high-level waste with respect to its mass, radiotoxicity and (possibly) repository risk. P&T mainly deals with the management – i.e. transmutation and/or special conditioning and confinement – of minor actinides and fission products, but involves the closure of the fuel cycle for plutonium as a necessary first or parallel step. The conditions for a completely closed fuel cycle, the goals for transmutation, and the implications for the reactor and fuel cycle technology are overviewed and discussed, and the currently favoured transmutation strategies are compared with respect to achievable waste radiotoxicity reduction and impact on the releases of potentially troublesome actinides from a repository for vitrified high-level waste.

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[2] Under contract with Federal Office of Energy, CH-3003 Bern, Switzerland.

1. From the open fuel cycle to the semi-closed fuel cycles for plutonium management

The LWR once-through fuel cycle at a mean burn-up of 50 GWd/t_{HM} produces (a) spent fuel consisting of fission products (1.1 t/GWe-a), irradiated uranium, plutonium, and minor actinides (about 20 t/GWe-a, mostly uranium), and (b) depleted uranium from the enrichment process (about 170-190 t/GWe-a, depending on the ^{235}U concentration of the tails).

If these materials are not further utilised, they have to be considered as nuclear waste. The preferred option is currently to store the irradiated fuel elements after appropriate cooling in suitable geological formations. The LWR once-through fuel cycle has the advantage that it avoids the difficulties of reprocessing; however, it can only extract about half a percent of the energy content of the mined uranium.

Depleted uranium stored as UF_6 is a chemical hazard and becomes radiotoxic in the long-term. Therefore, it has to be transformed into a more stable form (e.g. UO_2 or U_3O_8) and appropriately stored for use in future breeder reactors, or adequately disposed. In the nuclear waste discussion, not much attention has yet been given to the management of the increasing stocks of depleted uranium.

1.1 Incentive for reprocessing

The incentive to reprocess irradiated LWR fuel arises primarily from the desire to improve the *uranium utilisation* and implies the recycling of the bred plutonium (about 250 kg/GWe-a) in MOX-LWRs or future high-conversion and fast-spectrum reactors. Multi-recycling of uranium and plutonium in LWRs in the so-called self-sufficient mode would allow the uranium utilisation to improve by about a factor of two, but involves a higher ^{235}U and plutonium enrichment due to the "degradation" of the uranium and plutonium isotopic composition. A much better (close to 100%) uranium utilisation could be achieved with fast reactors (FR).

Industrial reprocessing is currently based on the PUREX process which allows "clean" uranium and plutonium to be separated from the fuel and was initially developed for military applications. The remaining high-level waste (HLW), consisting mostly of fission products and minor actinides, is converted into a stable form for ultimate disposal, with the normal method being the storage of vitrified HLW in geologic repositories.

The separation of uranium and plutonium from the spent fuel has the advantages of reducing the actinide mass and the plutonium content of the HLW. In combination with vitrification, it minimises the risk of a clandestine recovery of fissile material from a waste repository; however, it does not significantly reduce the long-term toxicity of the HLW. Drawbacks are the extra investment in complex technology for reprocessing and α-active fuel fabrication, and the potential proliferation risk associated with the handling of pure fissile materials. Since the balance of advantages and drawbacks depends on regional boundary conditions and political factors, there are currently contradicting policies regarding the recycling of plutonium.

## 1.2 Plutonium stock management

In some countries, an early commitment to industrial reprocessing, combined with the delayed introduction of fast reactors, has led to large stocks of separated plutonium. The available fabrication capacity for LWR-MOX fuel of about 300 t_{HM}/a, however, will now allow some 25 tPu/a to be recycled, which balances the current output of the reprocessing facilities [1].

Relative to the self-sufficient recycling mode, which has a zero plutonium balance, the plutonium consumption of an LWR can be enhanced by increasing the number of MOX fuel elements in the core, up to a full MOX core. An even higher plutonium consumption – up to the theoretical limit indicated in Figure 1 for uranium-free concepts – could be achieved by reducing the uranium content of the fuel. However, it should be noticed that LWRs alone cannot completely burn plutonium because the buildup of the even isotopes in a thermal neutron spectrum constrains the number of recycles to two or three at most. The remaining degraded plutonium has to be disposed, or stored until it can be utilised in fast reactors, which offer similar net consumption rates as the LWRs (cf. Figure 1).

It is obvious that plutonium stocks can be managed effectively with LWRs and fast reactors; new types of burner reactors or reprocessing methods are not needed, but plutonium could, of course, also be managed with other types of reactors. The respective issues, including fuel developments, have been discussed in the framework of working parties and workshops of OECD/NEA [2,3] and in many international conferences. It should be emphasised that the closure of the fuel cycle for plutonium is a prerequisite for, but not a direct issue of P&T. Therefore, plutonium management as such is not in the focus of this paper.

Figure 1. **Net plutonium production of different reactor types (kg/GWe-a)**
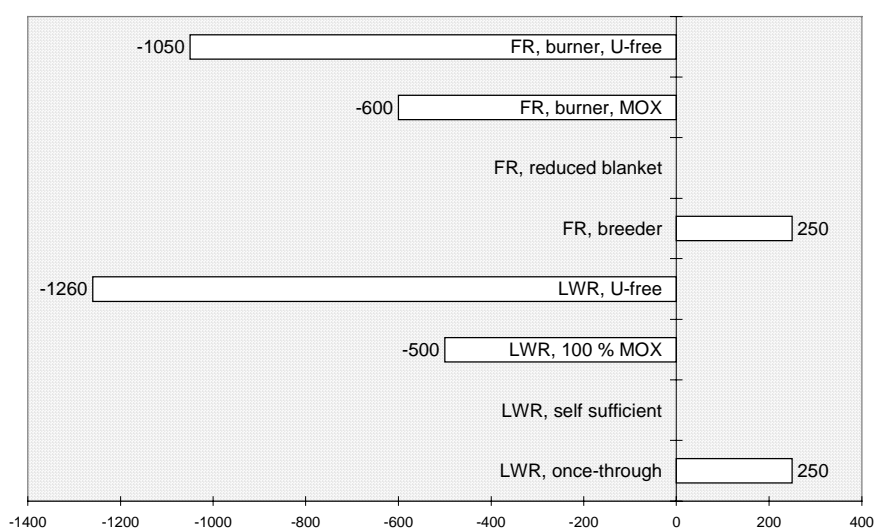
2. Fully closed fuel cycles with P&T

P&T aims at making nuclear energy more sustainable from the viewpoint of the *back-end of the fuel cycle* and implies the separation and further utilisation of valuable materials as well as the minimisation of the remaining HLW with respect to its mass, radiotoxicity and (perhaps) repository risk. It thus responds to current concerns of the public and politicians who are not satisfied with a radiological hazard which extends over millions of years, although the associated long-term risk in terms of annual individual dose is very small.

Figures 2 and 3 show the radiotoxicity of the HLW produced for a 120 GWe-a scenario after reprocessing and the resulting annual individual dose, estimated for vitrified waste emplaced in cristalline host rock [4]. Evaluations for other scenarios and repository concepts give comparable results [5,6] and confirm the following general observations:

- From the viewpoint of the *radiotoxicity*, which plays a role in accidental intrusion scenarios, P&T must first be concerned with the actinides, particularly the minor actinides americium and neptunium, the toxicity of the fission products (shaded in Figure 2) laying at least two orders of magnitude below that of the actinides after 10^3 years.

- From the viewpoint of the *long-term risk* of a geologic repository, the relatively mobile fission products are more important than the actinides, the fission products ^{135}Cs, ^{79}Se, ^{99}Tc and ^{126}Sn being dominant dose contributors in vitrified HLW scenarios[3].

- The fission product risk peaks in the time span 10^4 to 10^6 years after the closure of a repository, whereas an actinide risk arises "only" after one million years.

2.1 Goal for minor actinide mass reduction

Figure 2 shows that the radiotoxicity of the actinides requires more than ten-thousand years to decay to the toxicity level, $U_{nat}(LWR)$, given by the consumed natural uranium. With a hundred-fold reduction in the actinide content, this goal could be reached already after a few hundred years. The fact that the "natural toxicity" level for a pure fast reactor strategy, $U_{nat}(FR)$, is about hundred times smaller than that for an LWR strategy speaks also for the goal of a reduction in the actinide content of the HLW by a *factor of 100*.

It is obvious that a hundred-fold reduction of the actinide mass cannot be achieved in a single pass through a reactor. Hence, *multi-recycling* will be essential. In fact, the ideal P&T system has a fuel cycle which is fully closed for the actinides, meaning that only fission products are separated from the spent fuel and all actinides are returned to the reactor, together with a "top-up" (make-up) of new fuel replacing the fuel which was fissioned. It is also clear that such a system must be operated for many decades before the core – and hence the composition of the discharged fuel, which determines the specific waste radiotoxicity – reach an equilibrium.

2.2 Goal for actinide recovery

In practice, the actinides cannot be recovered completely from the spent fuel, and the remainder will go to waste. For a system with a fully closed fuel cycle, the mass of actinides going to waste is:

$$M^W = \delta \ L \ M^F$$

where M^F is the total mass of actinides fissioned, L is the actinide loss fraction during reprocessing and fuel fabrication, and the burn-up factor, δ, can be evaluated from the fraction of heavy metal fissioned, B, as $(1 - B) / B$. Under equilibrium conditions, M^F equals the top-up fuel mass, M^T, which, in general, can be divided into the mass, M^B, of transuranic or minor actinides to be burnt (i.e. transmuted and ultimately fissioned), and a diluent, usually consisting of fertile uranium (normal critical burner cores are not suited for fertile-free top-up fuel).

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[3] The long-lived fission product ^{129}I, which is known to dominate the repository risk in direct storage scenarios, is not present in vitrified HLW because it is discharged to the sea during reprocessing. Since sea disposal will not be a desirable feature of a "clean" nuclear energy, ^{129}I is also a candidate for P&T.

Figure 2. **Radiotoxicity of LWR spent fuel after uranium and plutonium separation**
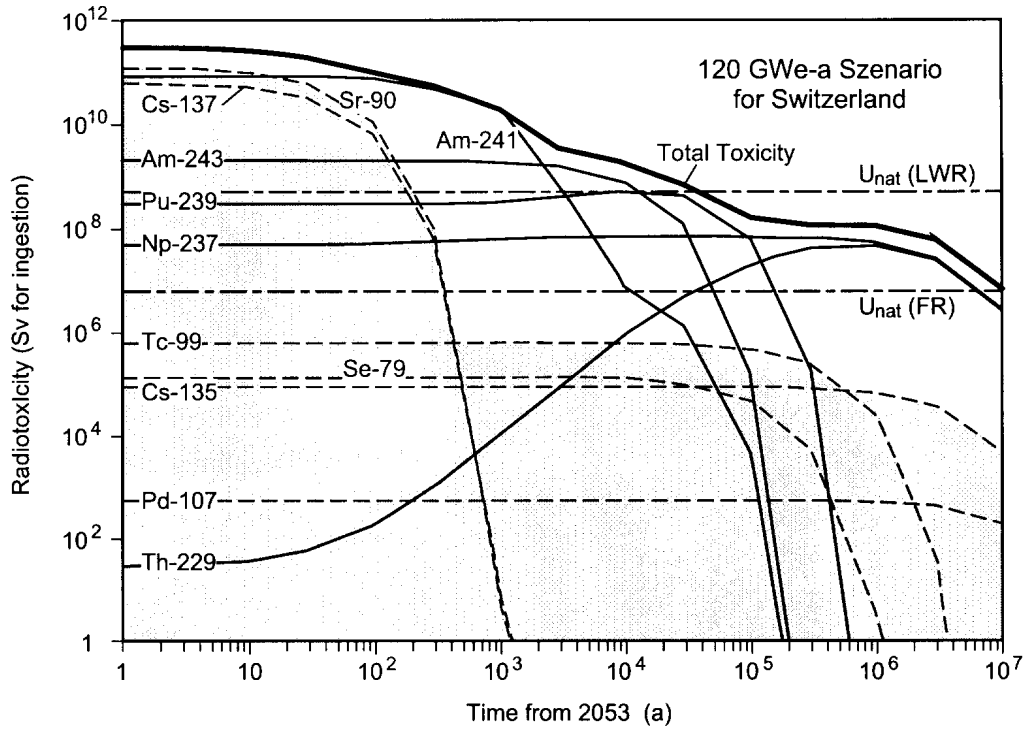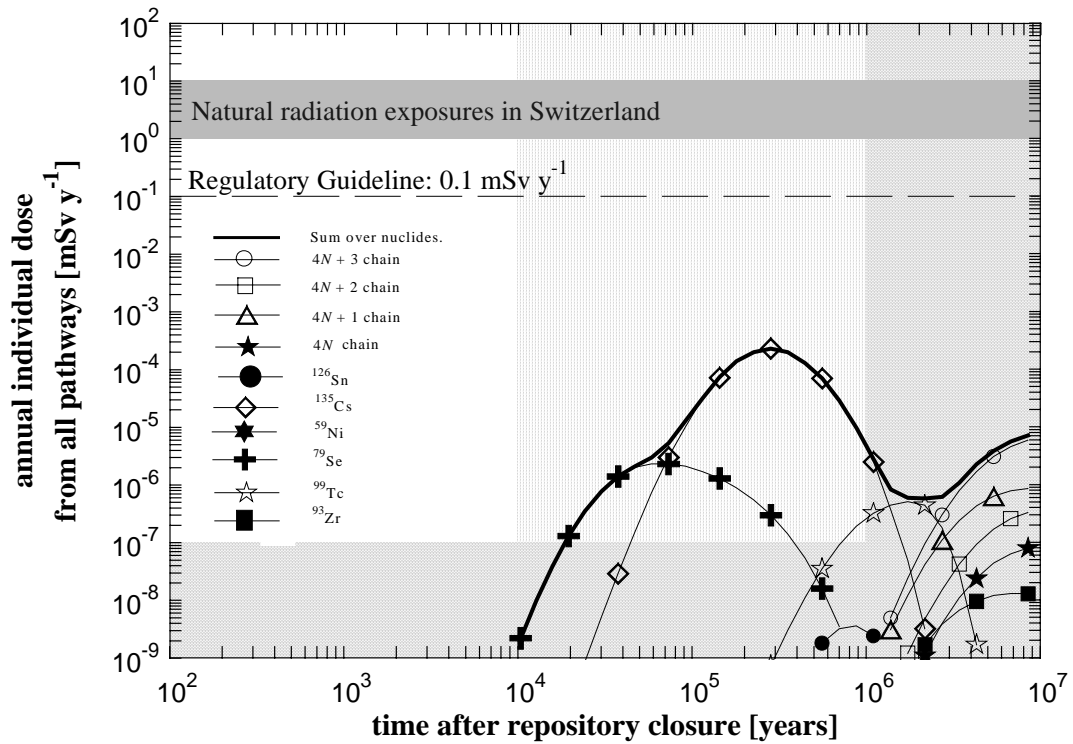

Figure 3. **Annual individual dose for vitrified HLW emplaced in cristalline host rock**

Denoting the transuranic (TRU) or minor actinide (MA) fraction of the top-up fuel, M^B/M^T, by τ and the "waste mass reduction factor", M^B/M^W, by R^M, one obtains the simple expression

$$L = \tau / (\delta \ R^M),$$

which gives the allowable losses as a function of the waste mass reduction factor. For the desired reduction factor of 100, an achievable average fuel burn-up of 15%, and a top-up fuel without a fertile component ($\tau = 1$), the expression yields $L = 0.18\%$. Since average burn-ups beyond 15% have not yet been proven with known fuel technologies, a *target value of 99.9%* for the actinide recovery yield must consequently be set for an effective P&T system.

3.   Neutronic requirements for fully closed fuel cycles and role of the ADS

For neutronic reasons, not all reactors can operate with a fully closed fuel cycle. To assess the suitability of a reactor in terms of neutron multiplication, the production-to-absorption ratio of the actinides in the equilibrium core, η_{ec}, is a useful parameter. Alternatively, the overall neutron balance for the complete fissioning of actinides can be measured in terms of the "fuel neutron production parameter" – D [7].

An η_{ec} value smaller than 1 means that the fuel of the equilibrium core cannot maintain a chain reaction; a negative -D value indicates that an actinide or an actinide mixture cannot be completely fissioned. It can be shown that the parameters are mainly influenced by the neutron spectrum and flux of the system and that the two approaches lead to the same conclusions.

The η_{ec} and -D values in Table 1 refer to realistic reactor concepts including an ATW-type sub-critical thermal transmuter, a CAPRA-type fast plutonium and MA burner, a critical (sodium-cooled) fast TRU burner, a sub-critical (LBE-cooled) fast TRU burner, and a dedicated sub-critical MA burner. For the actinide feed, plutonium and TRU mixtures separated from PWR spent fuel and the MA mixture from the first stratum of a typical "double strata" fuel cycle strategy [8] are assumed. The values demonstrate that minor actinides cannot be completely burnt in thermal systems and that fast plutonium and TRU burners offer more surplus neutrons than the respective thermal systems. The surplus neutrons could be used for burning fission products.

Table 1. **Neutronic performance of plutonium, minor actinide and transuranics burners**

Actinide feed [a]	Thermal (ADS)		Fast (critical)		Fast ADS	
	η_{ec}	-D	η_{ec}	-D	η_{ec}	-D
Plutonium	1.15[b]	0.40[b]	1.64	1.18	1.80	1.34
Minor actinides	0.89	-0.37	1.28	0.71	1.33	0.79
Transuranics	1.11	0.30	2.00	1.52	1.75	1.29

[a] Plutonium and TRU from PWR spent fuel with a burn-up of 50 GWd/t_{HM}, minor actinides from a park with PWR-UOX reactors (70%), PWR-MOX reactors (10%), and CAPRA reactors (20%).

[b] A MOX-PWR with self-sufficient plutonium recycling has a similar neutron economy.

3.1 Core design constraints

In practice, the design of a TRU or MA burner core, like that of any reactor core, is not only constrained by the above-mentioned basic neutronic criterion, but also by performance and safety parameters, such as the reactivity swing during burn-up, coolant void reactivity effect, Doppler coefficient, effective delayed-neutron fraction, etc. In particular, for a sodium-cooled fast reactor core, the substitution of normal MOX fuel by TRU- or MA-dominated fuel has an unfavourable influence on several of these parameters, and this is one of the reasons for the recent revival of various fast and thermal reactor concepts which were studied in the past, but have not been introduced as commercial systems. For example, the (positive) coolant void effect in sodium-cooled fast reactors, which deteriorates in a minor actinide burning regime, can be mitigated by substituting the sodium by *lead*, or even eliminated by substituting the liquid metal by a *gas coolant*.

To ensure that a critical burner core performs satisfactorily and has acceptable safety parameters, it is usually necessary to blend the TRU or minor actinides with the fertile materials uranium or thorium. However, blending reduces the transmutation effectiveness of the system. In this context, accelerator-driven systems offer interesting *additional parameters of freedom* by removing the criticality constraint and increasing the safety margin to prompt criticality. The latter feature is particulary important for MA burners, which are difficult to control as critical systems because the effective delayed-neutron fraction is only about half of that of a normal fast reactor.

In response to the new core design issues raised by P&T and the increased interest in advanced reactor technology in general, government and industry funded design teams in Europe, the Far East and the USA are currently spending a considerable effort on the optimisation of a broad range of advanced reactor designs featuring both critical and accelerator-driven cores.

3.2 Transmutation effectiveness

Various, sometimes conflicting definitions for transmutation effectiveness, usually based on the minor actinide balance of a particular core, can be found in the literature (see, for instance, [6]). For a system with fully closed fuel cycle, the difficulty of defining a core-specific transmutation effectiveness is circumvented by defining an "overall transmutation effectiveness" as the relative content of the top-up fuel in transuranic and minor actinides, M^B/M^T, i.e. the already discussed quantity τ.

It is interesting to notice that the overall transmutation effectiveness does not depend directly on the choice of the neutron spectrum, the fuel type and the coolant, but is governed by the above-mentioned performance and safety constraints. For a critical TRU burner based on liquid metal technology, τ is smaller than about 0.5, and in the case of homogeneous MA recycling in a EFR-type fast reactor τ is less than 0.1. The possibility to operate sub-critical MA and TRU burner cores with a fertile-free top-up fuel and hence 100% overall transmutation effectiveness, i.e. $\tau = 1$, is probably the *most important advantage of accelerator-driven systems*.

3.3 Neutronic transmutation effect

Using the same notation as before, the radiotoxicity reduction relative to the top-up fuel, $R^T(t)$, is:

$$R^T(t) = R^N(t) \ M^T / M^W$$

or, in terms of the fuel burn-up and the fuel loss,

$$R^T(t) = R^N(t) / (\delta \ L)$$

where $R^N(t)$ is a time-dependent "neutronic transmutation factor"[4]. It should be noticed that the toxicity reduction relative to the actinides to be burnt, which is of direct relevance for the assessment of transmutation schemes, equals R^T for all practical purposes because the toxicity of the fertile component of the top-up fuel is negligible compared with that of the actinides to be burnt.

Figure 4 compares the neutronic transmutation factors for three systems: (a) an ALMR-type critical TRU burner, (b) an ATW-type sub-critical TRU burner with a fast-neutron spectrum, and (c) a minor actinide burner operating in the P&T cycle of a double strata fuel cycle. It can be seen that the critical burner has a small "neutronic" advantage over the sub-critical burners, but the difference is not significant compared with the goal for the total toxicity reduction by a factor of 100.

The general conclusions to be drawn from this discussion are that:

- Regarding the neutronic transmutation factor, no single transmutation system has a significant advantage over other systems.

- Radiotoxicity reduction has to be achieved primarily by an actinide mass reduction which implies the maximisation of fuel burn-up and the minimisation of reprocessing and fuel fabrication losses.

The importance of advanced reprocessing and fuel technologies for P&T is thus confirmed.

Figure 4. **Neutronic transmutation factors for transuranics and minor actinide burners**

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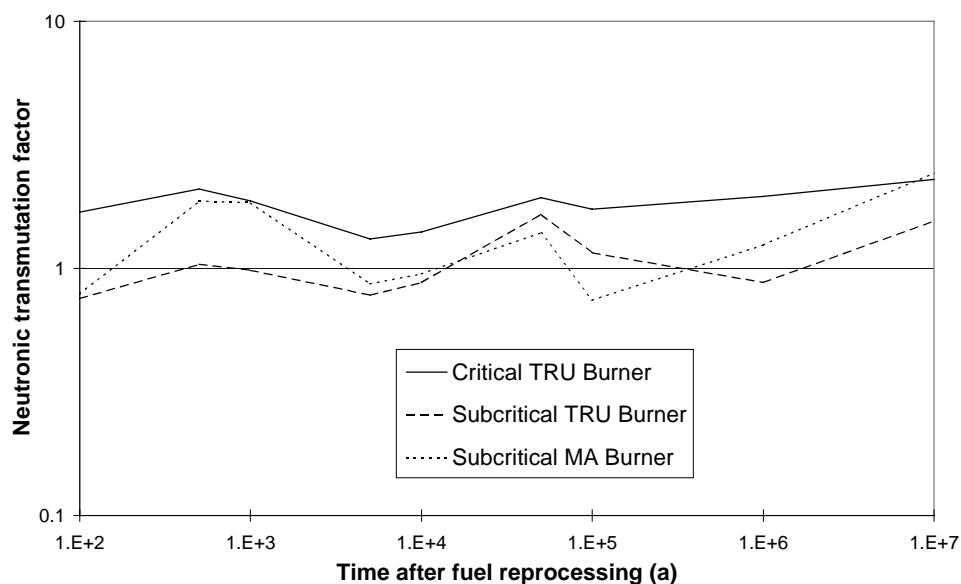
[4] $R^N(t)$ is a characteristic of the core and is sometimes called "neutronic toxicity reduction".

4. Fuel cycle problems and challenges

Current proposals for P&T technology rely on the aqueous (PUREX-type) reprocessing of spent fuel as a preliminary step preceding minor actinide partitioning. But, even in case of pyrometallurgical processing of TRUs, a mechanical head-end and an aqueous processing step (called UREX) for the prior removal of uranium as the main fertile element in any case precedes the sequence of pyrometallurgical separation steps.

The drastic increase of the plutonium content from 15% up to 43% and the fuel burn-up from 80 GWd/t_{HM} up to 210 GWd/t_{HM} for fast reactor concepts such as CAPRA make aqueous reprocessing more difficult because of the low solubility of plutonium and the radiation damage to the organic extractant (tributyl phosphate). Industrial "pilot" scale work at Dounreay and Marcoule has shown that, in specific technological conditions (pin-chopper, fast contactors), aqueous reprocessing can be considered as valid for fast reactor and future ADS fuel, if the decay heat can be mitigated by cooling or by dilution with LWR fuel. Special chopping or shearing systems have been developed for fast reactor (FR) fuel, e.g. for FFTF, in order to combine the single or multiple pin shearing with a more economical bundle-shear approach.

Spent fuel arising from a reactor park composed of 70% LWR-UOX reactors, 10% LWR-MOX reactors and 20% FR-MOX reactors has to be reprocessed in order to guarantee the plutonium recycle requirements in a stable scenario. The corresponding spent fuel discharges per 100 Gwe-a delivered by the reactor park are summarised in Table 2. Table 3 gives the residual decay heat for these fuel types, assuming the reprocessing operations to be carried out 5 years after the discharge of the fuel from the reactor.

Table 2. **Spent fuel arisings from a composite nuclear reactor park per 100 GWe-a**

Fuel type	Burn-up (GWd/t_{HM})	Delivered energy (GWe)	Spent fuel discharge (t_{HM})
LWR-UOX	50	69.7	1 520
LWR-MOX	50	10.3	224
FR-MOX	150-200	20.0	124-153

Table 3. **Decay heat of spent fuel 5 years after discharge from reactor**

Fuel type	Burn-up (GWd/t_{HM})	Total decay heat (kW/t_{HM})	Fission products (kW/t_{HM})	Actinides (kW/t_{HM})
LWR-UOX	50	2.76	2.28	0.477
LWR-MOX	50	6.56	1.97	4.60
FR-MOX	150	30.3	6.60	23.7
FR-MOX	210	33.7	8.14	25.6

4.1 Issues of aqueous reprocessing

The present state-of-the-art in aqueous reprocessing based on the PUREX process is almost satisfactory with regard to uranium and plutonium separation. Recovery yields of nearly 99.7% have been achieved and 99.9% is potentially achievable in a near future. However, reprocessing of industrial

quantities of LWR-MOX and FR-MOX within short cooling times will require gradual adaptations of the PUREX flow-sheet and involve the construction of additional extraction rigs.

In order to improve the plutonium dissolution yield and to avoid solvent radiation peaks during the extraction, a separate dissolver dedicated to FR-MOX treatment could be installed and connected to the main dissolver by a metering system. By connecting the dedicated FR-MOX dissolver to the main LWR dissolver, a constant radiation level can be kept throughout a given processing campaign. The second dissolver could also serve as "residue dissolver" by making use of highly oxidising compounds (e.g. electrochemically generated Ag(II)) to dissolve the insoluble fraction of the initial plutonium inventory and to reduce the transfer of insoluble plutonium residue to the waste stream.

The decay heat of the separated plutonium fraction enters the organic phase and remains in contact with the actinide fraction (U, Pu, Np) until separation and purification of the major actinides occurs. The ^{238}Pu concentration in the plutonium stream is the main source of radiation damage to the tributyl phosphate extractant, a strong source of neutrons in the separation plant, and an additional radiolysis agent during the oxalate precipitation and conversion to PuO_2. On the other hand, the high ^{238}Pu concentration in the separated PuO_2 for the same reasons (heat, spontaneous neutrons) improves the non-proliferation resistance of the product during storage. The MOX fuel fabrication will have to adapt its handling technology to reduce the radiation dose to the workers in the plant and during the transport of MOX fuel assemblies.

The separation of the minor actinides is currently under intense investigation throughout the OECD/NEA countries. The separation of the bulk of ^{237}Np from the aqueous product solution during the reprocessing operations is technically feasible but requires an adaptation of the first cycle extraction flow-sheet. The residual ^{237}Np which goes directly to the HLW solution could, in principle, be recovered by recycling the HLW solution through a secondary recovery cycle after a valence adjustment. With liquid extraction processes, the separation yield can be increased up to the desired value of 99.9% by increasing the number of extraction stages.

The separation of americium and curium is much more difficult, but considerable progress has been made during the past decade. Bulk separation of minor actinides together with lanthanides has been demonstrated under hot-laboratory conditions with several new processes such as TRUEX, DIDPA, DIAMEX and TRPO [6,9]. The separation of minor actinides from lanthanides, however, remains an obstacle for the industrial up-scaling of Am-Cm separation from HLW. Several promising laboratory methods – e.g. the ALINA and CYANEX301 processes using DTPA [10] and BTP [11] as specific extractants for the separation of minor actinides from lanthanides – have been tested at the ITU of JRC Karlsruhe. Yields of 99% and 97.6% have been achieved for americium and curium, respectively. Further improvements are necessary in order to include these methods in a cycle of multiple reprocessing.

By progressively incorporating an additional extraction rig for minor actinides from HLW coupled to a conventional vitrification process, a much less toxic HLW could be obtained, keeping the actinides in the fuel cycle for ultimate fissioning, and keeping the fission products in the glass.

4.2 Pyrochemical reprocessing

The proliferation risk potentially associated with the clean plutonium produced by the aqueous reprocessing has drawn the attention on the pyrochemical reprocessing which makes it difficult to separate individual TRU elements. Whereas the pioneering work was performed by ANL in the USA [12], most of the recent advances have been made in Russia [13] and Japan [14]. In the Russian

Institute of Atomic Research (RIAR), a pyrochemical process has been demonstrated with highly irradiated spent oxide fuel with burn-ups of 210-240 GWd/t_{HM}. Good results were obtained during the demonstration. The recovered PuO_2 will be mixed with UO_2 and processed by vibropacking into fresh FR-MOX fuel. Pyrochemical reprocessing of metallic fuel was developed in the USA in the framework of the integral fast reactor (IFR) programme with capability for actinide burning. Recovery yields of 95% for uranium and 99% for mixtures of uranium and minor actinides have been obtained on laboratory and pilot-plant scales at the Idaho Argonne West Laboratories.

The advantages and disadvantages of the pyrochemical reprocessing can be summarised as follows:

- Highly concentrated TRU product streams can be handled without major radiation degradation of the reagents, allowing the facilities to be more compact than aqueous reprocessing facilities for the same TRU throughput. Because of the absence of water, much smaller criticality risks during purification and metallic fuel re-fabrication arise when processing industrial quantities of TRUs.

- Since all TRU elements remain together throughout the process, the proliferation risk is much reduced. The separation of the TRUs from the lanthanides, however, is difficult, requiring the development of multistage countercurrent extraction using highly corrosive reagents at high temperature. The most challenging issue is the selection and industrial manufacture of corrosion resistant equipment which must be designed for remote operation and maintenance.

- For spent LWR-UOX fuel, a genuine pyrochemical process has to cope with the problem of the elimination of the uranium, which is the major constituent of the LWR fuel. Therefore, a mixed approach using a (P)UREX-type process for uranium-neptunium elimination has been selected as the first step of the "ATW road map" project. Fluoride volatility has also been suggested as an alternative, but the mixed volatility of U-Pu and zirconium leads to complex waste streams which can be difficult to control.

- Starting from metal or nitride TRU fuel, a complete pyrometallurgical process involving a series of electro-refining steps in a wide range of molten salt baths has recently attracted much interest throughout the nuclear research communities in the USA, Russia, Japan and France. Most of the respective flow-sheets remain in the pre-conceptual phase and will require several decades of R&D to become a mature technology comparable to the present aqueous reprocessing.

- Whereas the aqueous reprocessing consists of an independent industrial process which supports a large reactor park and can operate independently on continental or even world scales, the pyrochemical process will predominantly be applied in small facilities installed in the immediate vicinity of the reactors.

- However, in the long-term, pyrochemical reprocessing will become indispensable, if very hot fuel has to be multi-recycled in fast reactor or ADS facilities on an industrial scale with a limited out-of-pile inventory.

5. Principal actinide transmutation strategies

Depending on regional boundary conditions and political factors, countries with P&T programmes have developed different transmutation strategies. As to the transmutation of actinides, Table 4 provides an overview of the principal approaches and indicates respective driving forces. In

view of the historic development, the table distinguishes between evolutionary (right) and innovative (left) approaches.

The *evolutionary approach*, adopted mainly in Europe and Japan, aims at closing the fuel cycle in successive steps, starting with the recycling of plutonium in LWRs and later in fast reactors using conventional reprocessing and MOX fuel technology, and finally complementing the system with a dedicated P&T cycle which features MA burners with fast neutron spectra. This approach has the advantage that it can respond flexibly to changes in the priorities for plutonium and minor actinide management, and that new technologies have to be developed only for a comparatively small number of MA burners which support a large system of conventional LWRs and fast reactors.

Table 4. **Principal actinide transmutation strategies**

TRU burning		Pu recycling	
Separation of uranium and TRU from spent LWR fuel, TRU remain together. TRU recycled in thermal or fast critical or sub-critical reactors with fully closed fuel cycles. Dry reprocessing particularly suited for closed fuel cycles and highly active fuels. Principal driving force: non-proliferation		Separation of uranium and Pu from spent LWR fuel. Pu recycled in thermal and later in fast reactors (limited number of "thermal" recycles). PUREX-type wet reprocessing methods are appropriate. Flexible thermal: fast reactor ratio from about 4 to zero. Possibility to move to a pure fast reactor strategy. Principal driving force: resource management	
With thermal neutrons	**With fast neutrons**	**Without transmutation**	**With MA transmutation**
TRU burning in thermal reactor.	**TRU burning in FR** Flexible thermal: fast reactor ratio from about 2 to zero. Possibility to move to a pure FR strategy (IFR system).	**Pu burning** Fully closed fuel cycle for Pu. MA and FP conditioning by vitrification and/or dedicated insolubilisation with ceramics technology.	**Double strata fuel cycle** MAs (and FPs) burnt in a fully closed P&T cycle. Fast spectrum required for MA transmutation, ADS has safety advantages.
TRU burning in thermal ADS Pure burner strategy. (thermal ATW system)	**TRU burning in fast ADS** Thermal:fast reactor ratio of about 3. Possibility to move to a pure ADS strategy by adding fertile fuel (Energy Amplifier).	Principal driving force: waste management.	Dry reprocessing particularly suited for fully closed cycle. One MA burner supports some 15 "normal" reactors. Principal driving force: waste management.

The *innovative approach*, first suggested in the USA, aims at co-processing plutonium and minor actinides to avoid the use of technologies with a potentially high proliferation risk. After the initial

separation of the uranium from the LWR spent fuel, the unseparated transuranic actinides are recycled in a transuranic burner with a closed fuel cycle based on pyrochemical reprocessing. Compared with the double strata strategy, the number of burners in the equivalent system is four to six times larger, but the burners are not subjected to a (fast) neutron-spectrum condition. Nevertheless, most of the currently evaluated critical and sub-critical transuranic burners feature a fast neutron spectrum.

5.1 Strategies studied by OECD/NEA

An expert group of the OECD/NEA Nuclear Development Committee is currently comparing the principal actinide burning and transmutation strategies in more detail. The investigated strategies comprise:

a) Plutonium burning in LWRs and CAPRA-type fast reactors.

b) The double strata strategy with LWRs and CAPRA reactors in the first stratum and accelerator-driven MA burners in the second stratum.

c) TRU burning in critical fast reactors (IFR concept).

d) TRU burning in sub-critical fast reactors (ADS).

e) A heterogeneous strategy in which americium and curium are recycled in targets.

Table 5 summarises the most important assumptions for strategies a to d, and Figure 5 gives preliminary results for the achievable actinide waste radiotoxicity reduction relative to the open fuel cycle. The figure shows that transmutation strategies b, c and d all meet the goal of a hundred-fold reduction, and confirms that plutonium recycling alone is not effective in reducing the actinide waste radiotoxicity.

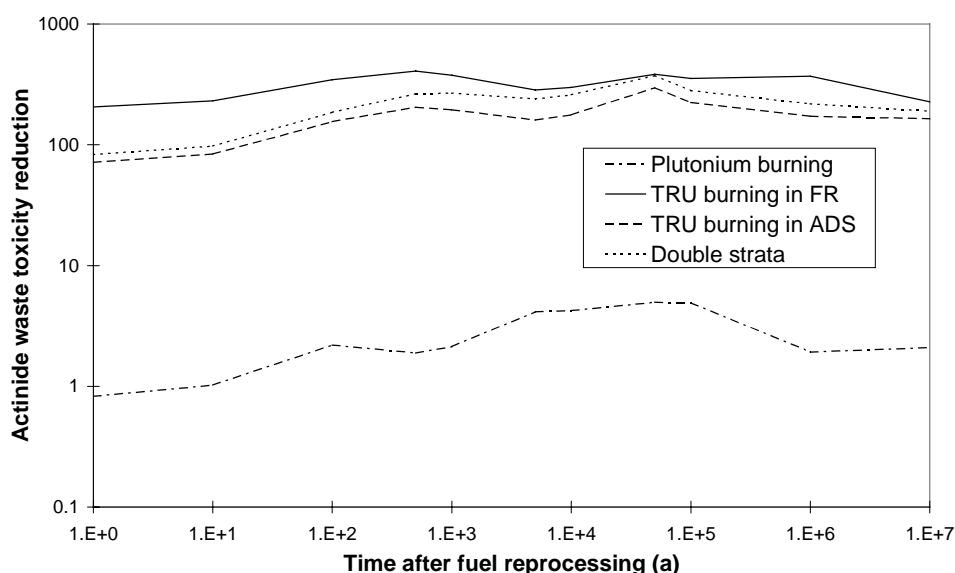Figure 5. **Actinide waste radiotoxicity reduction relative to open fuel cycle (Preliminary results of OECD/NEA study)**

43

Table 5. **Assumptions for transmutation strategies compared by OECD/NEA**

Strategies	Reactor/ADS	Fuel	Av. burn-up (GWd/t$_{HM}$)	Reprocessing method	Recovery yield (%)
All	LWR (N4)	UOX/MOX	50	wet	99.9
a, b	FR (CAPRA)	MOX	185	wet	99.9
b	MA Burner, ADS	AcN-ZrN	140	dry	99.9
c	TRU Burner, IFR	Ac-Zr	140	dry	99.9
d	TRU Burner, ADS	Ac-Zr	140	dry	99.9

6. Fission product transmutation

The neutron capture process is currently the only promising nuclear reaction for transmuting fission products. Other processes which have been proposed in the past rely on technologies which are still at a very early stage of development (e.g. fusion neutron sources) and generally suffer from a poor energy balance. The capture process consumes neutrons but, theoretically, fast reactors could deliver *enough surplus neutrons* to allow the potentially troublesome long-lived fission products to be completely transmuted to shorter-lived or stable species (cf. Table 1).

The transmutation of a fission product makes sense only if the reaction rate (microscopic cross-section times neutron flux) is higher than the natural decay rate of the nuclide. With the practically achievable neutron fluxes, this condition cannot be met for the most abundant fission products ^{137}Cs and ^{90}Sr with half-lives of only about 30 years, i.e. these fission products are "non-transmutable". However, since their radioactive life is limited to less than 300 years, they can be safely enclosed using engineered barriers only. On the other hand, the fission products determine the size of the vitrified waste repository which, consequently, *cannot be much reduced* by P&T operations.

The fission products which influence the long-term risk of a repository are, in order of radiologic importance, ^{129}I, ^{99}Tc, ^{135}Cs, ^{93}Zr, and ^{126}Sn. Activation products (^{14}C and ^{36}Cl) can also contribute to the dose. Obviously, the relative contribution of these nuclides to the integral risk varies according to the type of repository host rock. In the following, the problems associated with the transmutation of these five fission products are discussed separately. It should be noted that the determination of the separation yield and the decontamination factor (DF) for HLW depends to a great extent on policy decisions and that, depending on the nuclide, special *conditioning and confinement* is an alternative to transmutation.

6.1 Iodine-129 ($T_{1/2} = 1.6 \cdot 10^7$ a)

In most of the direct storage concepts for spent fuel, ^{129}I is the first nuclide to enter into the biosphere due to its very high mobility. During aqueous reprocessing, iodine is removed from the dissolver solution with a yield of 95-98%. To improve the separation yield, more complex chemical treatments are necessary. Better separation yields may also be achieved with high-temperature pyrochemical processes. Special methods for conditioning in the form of AgI, PbIO$_4$, etc. have been developed but, because of the extremely long half-life, the eventual migration to the environment cannot be excluded.

Since the radiotoxicity of ^{129}I is the highest of the fission products and equivalent to that of actinides, it would be advisable to increase the separation yield to achieve a DF of about 1 000. The necessity of an isotopic separation and the limited stability of the target material, however, make the transmutation of ^{129}I difficult, and conditioning and confinement seems to be the best method to reduce its radiological impact. Nevertheless, the present method of diluting iodine in the sea may still be defendable because of the enormous dilution of ^{129}I in the (natural) ^{127}I present in seawater. The storage in a salt dome, consisting of evaporated seawater, is an another alternative which still has its merits.

6.2 Technetium-99 ($T_{1/2} = 2.1 \cdot 10^5$ a)

The radiologic significance of ^{99}Tc is important if the repository surroundings are slightly oxidic. In reducing conditions ^{99}Tc is remarkably stable and insoluble as technetium metal or TcO_2 suboxide. Partitioning of ^{99}Tc is not an easy task because it occurs as insoluble metal and as soluble technetate ion in solution. Separation from aqueous effluents is possible in an advanced PUREX scheme, but recovery from insoluble residues is difficult, with the present recovery yield at best reaching 80%. Improving this yield significantly implies the development of new separation technologies such as the not yet proven conversion of the technetium into a single chemical species. Alternatively, a group separation together with the platinum metals may be carried out using pyrometallurgical processes. If separated in metallic form, transmutation appears to be feasible because of its stability and relatively large neutron capture cross-section.

6.3 Caesium-135 ($T_{1/2} = 2.3 \cdot 10^6$ a), Zirconium-93 ($T_{1/2} = 1.5 \cdot 10^6$ a) and Tin-126 ($T_{1/2} = 1.0 \cdot 10^5$ a)

Caesium occurs in the form of the isotopes 133 (stable), 135 and 137. In terms of radiologic significance, ^{137}Cs is the major constituent of HLW (see above). The activity of the long-lived ^{135}Cs in HLW is a million times lower. However, once released from a matrix as glass, caesium is very mobile. Transformation of ^{135}Cs to stable ^{136}Ba is possible from a neutronics point of view, but probably impracticable because a close to 100% isotopic separation efficiency would be required (traces of ^{133}Cs in the target would generate new ^{135}Cs during the irradiation).

Zirconium-93 is somewhat similar to ^{135}Cs, it has also a very long half-life and a small isotopic abundance (about 14% of the total Zr). An isotopic separation would be necessary, and its transformation to stable ^{94}Zr would be very slow because the thermal capture cross-section is about five times smaller than that of ^{135}Cs.

Tin-126 is partly soluble in HLW from aqueous reprocessing but occurs also as an insoluble residue, similar to technetium. Isolation involves a special treatment of the HLW, and the use of isotopic separation techniques. Transmutation of ^{126}Sn is questionable due to the very low neutron capture cross-section.

7. Consequences for geologic disposal

As mentioned before, the primary concern of geologic repositories are possible releases of the relatively mobile fission products. Since the fission product yields are not very sensitive to the fuel composition and the neutron spectrum of the reactor, the fission product risk depends primarily on the number of fissions, i.e. the energy, produced in the fuel. This means that the fission product risk cannot be much influenced by the actinide transmutation strategy and can only be mitigated by

separating troublesome fission products from the waste. The choice of the reactor can, however, influence the in-situ transmutation of a fission product. For example, capture of thermal neutrons in the precursor ^{135}Xe reduces the ^{135}Cs production in an LWR by as much as 70% [15].

As to the influence of different transmutation strategies on the release of actinides from a repository, the usual approach is to perform complete nuclide transport calculations. Such an integral approach was chosen, for instance, by the authors of a recent study of the European Commission who compared nuclide fluxes at the clay-aquifer interface [16]. However, a difficulty of this approach is the strong dependence of the results on the host rock characteristics, which can vary considerably depending on the structure of the host rock and the storage concept (storage in salt, clay or granite). For generic studies, it may therefore be preferable to perform site-independent comparisons on the basis of releases of potentially troublesome actinides from the well-defined near-field of the repository[5], allowing a subsequent folding with site-specific geosphere and biosphere responses to be performed independently and according to the needs of specific repository projects [15].

Adopting the latter approach, near-field release rates have been evaluated for the strategies with vitrification investigated in the framework of the afore-mentioned OECD/NEA study. The results in Figure 6 indicate a strongly non-linear relationship between release rates and actinide concentrations in the glass. Important conclusions are that the plutonium burning strategy generally increases the maximum release rates, and the addition of the P&T cycle does not reduce maximum release rates for all potentially troublesome actinides.

Figure 6. **Maximum release rates from near-field relative
to LWR once-through/vitrification case
(Strategies investigated in the framework of the OECD/NEA study)**

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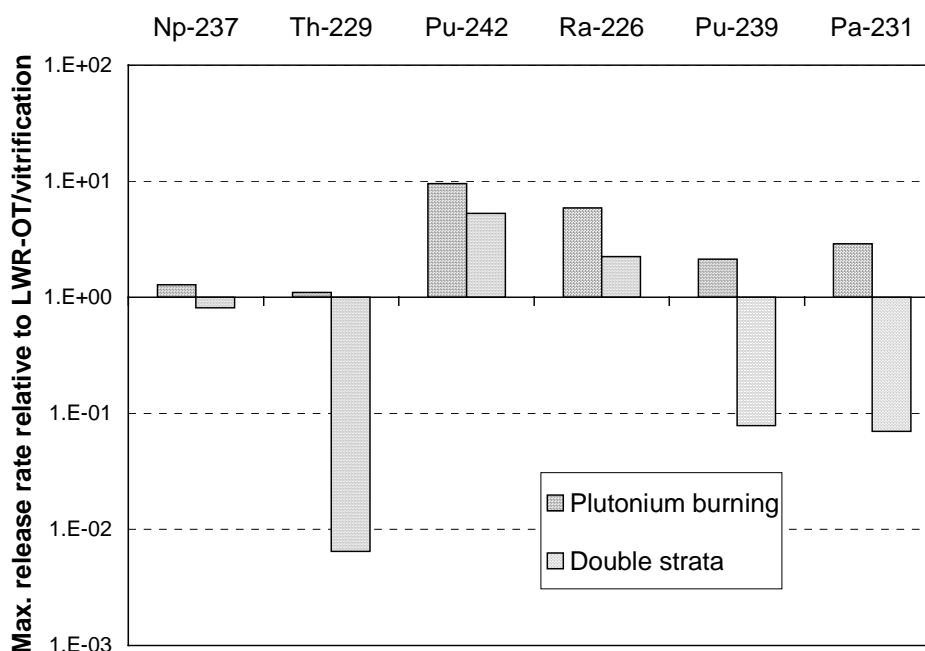
[5] Actinides to be considered in this context are ^{231}Pa, ^{237}Np and their respective daughter products ^{227}Ac and ^{229}Th, ^{226}Ra, a decay product of ^{234}U, and the long-lived plutonium isotopes 239 and 242.

8. Summary

The principal points and conclusions arising from this overview discussion of the nuclear fuel cycle and P&T are summarised as follows:

The closure of the nuclear fuel cycle with P&T will be a long-term endeavour and becomes a central issue in the development of a future sustainable nuclear energy system; the P&T strategy will directly influence the choices of new reactor and reprocessing technologies.

Plutonium recycling is a first step in this direction. Plutonium can be managed effectively with existing LWRs which, later on, should be complemented with fast reactors to burn the plutonium completely. The necessary extension of the proven, PUREX-type reprocessing technology to cope with the highly active fuels arising from plutonium-burning strategies appears to be feasible. The motivation for the utilisation of plutonium is its energy content, but not a mitigation of the long-term radiological hazard associated with the back-end of the fuel cycle.

Partitioning and transmutation aims at making the fuel cycle more sustainable from the viewpoint of the back-end by reducing the HLW radiotoxicity and (possibly) the migration of radiotoxic nuclides from HLW repositories to the biosphere. To this end, it introduces the separation and transmutation (or, alternatively, improved immobilisation) of minor actinides and fission products, assuming that the fuel cycle is already (or simultaneously) closed for plutonium.

Transmutation implies the development of advanced and innovative reactor and fuel cycle technologies, including ADS reactor technology, fuels with very high burn-up capability, and pyro-chemical reprocessing methods. It can be shown that the goals of transmutation cannot be achieved without the implementation of fast-neutron-spectrum systems in some form, and that the overriding requirement is that for high fuel burn-ups and actinide recovery yields. Regarding neutronics, it appears that no single transmutation system has a significant advantage over other systems.

As to the separation of minor actinides from HLW using aqueous reprocessing, promising new processes have recently been developed. The results achieved at laboratory and pilot-plant scales give confidence that the required high recovery yields can ultimately be achieved on an industrial scale. Nevertheless, the long-term future appears to belong to the pyrochemical reprocessing method, which is not yet mature, but is intrinsically better suited for a fully closed fuel cycle with highly active fuels and may be more proliferation resistant because all actinides remain together, and the relatively compact plants can be collocated with the reactors.

A comparison of the double strata and the TRU burning strategy indicates that, under comparable assumptions, the two strategies are equivalent and that both have the potential of achieving a hundred-fold radiotoxicity reduction of the actinides in the HLW. These (or any other) actinide transmutation strategies, however, are not effective in reducing the total mass of the HLW which is dominated by the fission products and, hence, mainly depends on the total energy produced.

The primary risk of geologic repositories is related to the release of long-lived fission products. With the exception of ^{99}Tc, however, the transmutation of long-lived fission products appears to be difficult because of low neutron reaction cross-sections and the necessity of isotopic separations. This means that, for most fission products, special conditioning and confinement is the only practical method to reduce the radiological impact. Finally, it is shown that, due to the non-linear relationship between release rates from a repository and actinide concentrations in the glass, P&T does not necessarily result in reduced release rates for all potentially troublesome actinides as one could expect.

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SESSION III

Partitioning

Chairs: J.P. Glatz (ITU) – J. Laidler (ANL)

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SUMMARY

Professor Charles Madic of DEA-DCC (Saclay) presented an overview paper on chemical partitioning in which he described in considerable detail the French processes for the extraction of minor actinides and their separation from lanthanide elements. He noted that the number of aqueous processes is burgeoning and that the processes are becoming too complex. He registered a plea for process simplification and for reduction in the size of process equipment, achievable perhaps by pre-concentration of the solutions to be processed.

Professor Michael Hudson of University of Reading gave a richly detailed description of the structure and characteristics of heterocyclic ligands that can be exceedingly useful in the extraction of lanthanides and minor actinides. He presented a comprehensive model for designing ligands to function as specialized extractants for specific lanthanide elements.

Dr. Jean-Paul Glatz of the European Commission Joint Research Centre/Institute for Transuranium Elements (EC-JRC/ITU) reported on the successful demonstration of minor actinide/lanthanide separations using a nitric acid PUREX raffinate solution containing minor actinides and lanthanides. Bis-triazinepyridine (BTP) was used as the extractant in a 16-stage centrifugal contactor train operating in the counter-current mode. Reasonable decontamination factors and recovery efficiencies were achieved. The work is particularly noteworthy because it is the first demonstration of minor actinide/lanthanide separation using an actual waste stream.

Dr. James Laidler of Argonne National Laboratory presented a paper describing a pyrochemical process being developed for use with a non-fertile metallic transmuter blanket fuel. This chloride volatility process involves digestion of the inert zirconium matrix by formation of volatile $ZrCl_4$. Transuranic elements are subsequently recovered from the residual salt by electrowinning. The unit operations comprising this process have all been successfully demonstrated with simulated fuel.

Dr. Jan Uhlir of the Nuclear Research Institute, Rez, Czech Republic, proposed the use of a fluoride volatility method as a continuous or semi-continuous process for partitioning molten salt fuels in a molten salt transmutation reactor scheme. He maintained that a practical near-term application of fluoride volatility processing may be as a means for processing oxide fuels to remove uranium and

recover transuranics for fissioning in a transmuter system. He cited experience in processing BOR-60 oxide fuel at Dimitrovgrad in the 1980s.

The CRIEPI/Transuranium Institute collaborative effort was described by Dr. Jean-Paul Glatz. This study involves the processing of metal alloy fuels (U-Pu-Zr and U-Pu-MA-Ln-Zr) using molten salt electrorefining and reductive extraction. A capability for small-scale hot processing has been established at EC-JRC/ITU, and preliminary experiments have been carried out with the electrorefiner, including both solid and liquid cathode deposition. A molten salt/metal reductive extraction process has been used to demonstrate the cleanup of electrolyte salt as well as for the treatment of high-level liquid waste.

Dr. M. Iizuka of CRIEPI described their work in development of the liquid cadmium cathode. The work was performed with a small (9 mm dia.) cathode crucible, without stirring or agitation. The result was that complete recovery of Pu could be obtained at low current densities and low concentrations, but that Pu loss occurred at higher current densities by rapid growth of dendrites. The work led to a projection of a Pu collection rate of nearly 300 grams per hour in a crucible of practical dimensions, with Pu loadings in the cathode approaching 5 wt.%.

OVERVIEW OF THE HYDROMETALLURGICAL AND PYRO-METALLURGICAL PROCESSES STUDIED WORLDWIDE FOR THE PARTITIONING OF HIGH ACTIVE NUCLEAR WASTES

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Abstract

For more than 10 years, a revival of the interest for the partitioning of high active nuclear wastes (HAWs) exists worldwide in connection with possible improvements of the management of the HAWs actually produced and with futuristic nuclear fuel cycles. The main aim of the partitioning processes is to separate, from the complex mixtures of HAWs, the long-lived radionuclides (LLRNs), belonging either to the minor actinides (MAs) or to the fission products (FPs) families of elements, in order to prepare fuels and/or targets suitable for their subsequent transmutation (P&T strategy). An other possible strategy consists in the special conditioning of the separated LLRNs into stable dedicated matrices (P&C strategy).

The LLRNs considered for partitioning are the MAs, neptunium, americium and curium, but also the FPs, caesium, technetium and iodine.

Most of the partitioning processes studied so far belongs to the domain of hydrometallurgy, but, recently, a new impetus was observed in the field of pyrometallurgical processes.

The main aim of this talk is to present a brief status of the development of both "hydro" and "pyro" processes for the partitioning of LLRNs developed worldwide, with a special emphasis on the benefits/drawbacks of each process.

1. Introduction

Since the end of the 80s, a renewal of interest is observed worldwide for LLRNs partitioning techniques from nuclear wastes (HAWs). This interest is connected with two main fields:

- The conventional LWR closed fuel cycle using the PUREX process. New management methods for nuclear wastes are considered, the so-called partitioning-transmutation (P&T) and partitioning-conditioning (P&C) scenarios. For this domain, hydrometallurgy is the main route for LLRNs partitioning process development, while pyrometallurgy is also subject of some research.

- "New" fuel cycles associated with the development of fast reactors (FRs), accelerator driven systems (ADSs) and fused salt reactors (FSRs). For this field, pyrometallurgy is the main route considered for spent fuel reprocessing and wastes partitioning, while a small interest still remains for the hydrometallurgical route.

The aim of the present article is to give a brief overview of the progress realised worldwide in the recent years in the field of partitioning of LLRNs by hydrometallurgical and pyrometallurgical processes.

2. Partitioning processes: an overview

2.1 General considerations [1]

2.1.1 Target elements for the separations

Actinides: for P&T and P&C scenarios, the elements considered for partitioning are the so-called minor actinides (MAs): neptunium (Np), americium (Am), and curium (Cm), while for "new" fuel cycles scenarios, uranium (U), plutonium (Pu) and the MAs are all concerned with partitioning/reprocessing process development.

Fission products: for P&T and P&C scenarios, iodine (I), technetium (Tc) and caesium (Cs) are the three main elements considered for partitioning. Some P&C scenarios also consider the partitioning of caesium and strontium.

In Japan, the separation of the platinum group metals (PGMs) from nuclear wastes is also studied for industrial uses of the separated PGMs.

2.1.2 Goals for the separations

The most important goals for the separations are:

- Minimisation of the *long-term radiotoxic inventory* of the wastes conditioned in "conventional matrices", e.g. in glasses (removal of LLRNs of MAs and FPs families).

- Minimisation of the *heat load* of the conditioned wastes (removal of ^{137}Cs and ^{90}Sr).

- More often, *high purities* of the separated LLRNs are required, for target or fuel fabrication for subsequent transmutations of these LLRNs.

2.1.3 Consequences

Owing to the fact that efficient separation methods are needed with low losses of LLRNs and high purities of the separated LLRNs, *multi-stage processes are most often necessary.*

2.2 Hydro processes for actinides and FPs partitioning

2.2.1 Examples of separation strategies

Examples of separation strategies are given for some countries and some research organisations.

2.2.1.1 Japan

- JNC
  For FRs closed fuel cycle, JNC develops an integrated approach based on hydrometallurgical steps including the: (i) dissolution of MOX FR spent fuels with an aqueous nitric acid solution, (ii) iodine volatilisation, (iii) electrolytic extraction of technetium (Tc), palladium (Pd) and selenium (Se), (iv) crystallisation of most of the uranium contained within the spent fuel dissolution liquor in the form of hydrated uranyl nitrate, (v) single PUREX extraction step for recovery of remaining U+Pu+Np, (vi) SETFICS process for Am and Cm partitioning.

- JAERI
  JAERI proposed to separate Np and Tc during the implementation of the PUREX process. This organisation develops also, since many years, the so-called four-group partitioning process for the treatment of the wastes issuing the reprocessing by the PUREX process of UOX or MOX LWR spent fuels. This partitioning process includes the following steps: (i) MAs partitioning (Np, Am and Cm), (ii) Cs+Sr extraction, (iii) PGMs extraction. The remaining mixture of wastes constitutes the 4th category of elements of the initial mixture treated.

2.2.1.2 USA

The situation in the USA is peculiar because partitioning processes developed concern the treatment of defence wastes in particular those accumulated at DOE's Hanford site during the cold war. Several processes were developed for the partitioning of radionuclides from the wastes: (i) TRUEX process for transuranic extraction, (ii) SREX process for Sr removal and (iii) CSEX process for Cs extraction. It should be also noted that in 1999, a Report named *A Roadmap for Developing Accelerator Transmutation of Wastes (ATW) Technology* was published by the DOE [2], which considers the possible treatment of the LWR spent fuels accumulated in the USA in order to separate: (i) U for final disposition as low level waste and (ii) TRUs for burning in ATW systems. The processes considered for these separations are: (i) the UREX process, which consists in a modified PUREX process aiming to only extract U, (ii) pyrometallurgical partitioning process for TRU separation from the UREX wastes and for the ATW fuel cycle.

2.2.1.3 France (CEA)

The strategy developed by the CEA for partitioning the nuclear wastes of LWR closed cycle concerns 6 LLRNs to separate from the wastes: 3 MAs (Np, Am and Cm) and 3 FPs (I, Tc and Cs). This strategy is based on the development of successive liquid-liquid extraction processes: (i) the improved PUREX process for U, Pu, Np, I and Tc separations, (ii) the DIAMEX process for trivalent Am+Cm extraction (FP lanthanides (III), Ln, are co-extracted), (iii) the SANEX process for

Am+Cm/Ln(III) separation, (iv) the SESAME process for Am/Cm separation, (v) the CALIX-CROWN process for Cs separation.

In some organisations (e.g. in USA, Japan, Czech Republic or Russian Federation), instead of developing a succession of separation processes for peculiar LLRNs, the integration of processes for MAs and FPs extraction are studied. For example, the use of a solvent containing a mixture of cobalt dicarbollide+dioxide of diphosphine allows the combined extraction of Cs+Sr+(Am+Cm)+Ln.

2.2.2 Minor actinides partitioning

One cycle processes

- DIDPA process (JAERI, Japan)

    This process for MAs partitioning is based on the use of di-isodecylphosphoric acid (DIDPA). The extraction mechanism is the following:

$$M^{n+} + \overline{n\,(HA)_2} \Leftrightarrow \overline{M(HA_2)_n} + n\,H^+ \tag{1}$$

    The separation of the TRU elements is done by successive stripping from the loaded solvent, including the use of diethylenetriaminopentaacetic acid (DTPA) complexing agent for actinides(III)/Ln(III) separation (TALSPEAK like process, vide infra).

    The DIDPA process was recently tested successfully in the BECKY hot-cell at NUCEF (JAERI, Tokai-Mura).

    Among the possible drawbacks of this process one can mention the: (i) required feed acid adjustment, (ii) solvent degradation and its delicate clean-up, (iii) limited solvent loading with metal ions.

- SETFICS (JNC, Japan)

    The SETFICS process constitutes a modification of the TRUEX process (vide infra) based on the use of the extractant di-isobutyl-phenyl-octylcarbamoylmethylphosphine oxide

$$(\Phi C_8 H_{17} P(O) CH_2 C(O)(i\text{-}C_4 H_9)_2 = CMPO)$$

    The extraction mechanism is as follows:

$$M^{n+} + n\,NO_3^- + \overline{m\,CMPO} \Leftrightarrow \overline{M(NO_3)_n(CMPO)_m} \tag{2}$$
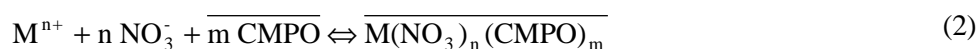
    The separation of TRUs is done by successive stripping from the loaded solvent, including also the use of DTPA for An(III)/Ln(III) separation. This process has not been tested yet with genuine HAWs. The possible drawbacks of this process are: (i) the limited stripping efficiency, (ii) the management of salts and DTPA containing effluents.

- PALADIN (CEA, France)

    This process is based on the use of a mixture of extractants: a malonamide (DIAMEX process extractant)+di-ethylhexylphosphoric acid (HDEHP), the extractant of the TALSPEAK process.

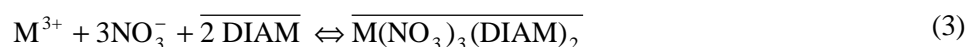The extraction and separation mechanisms are the following:

Extraction:

$$M^{3+} + 3NO_3^- + \overline{2\ DIAM} \Leftrightarrow \overline{M(NO_3)_3(DIAM)_2}$$ (3)

An(III)/Ln(III) separation: done after contacting the loaded solvent with a pH adjusted aqueous solution containing DTPA selective trivalent actinide complexing agent. For pH range HDEHP is the extractant, while at the metal nitrate extraction step, carried out with acidic feeds 3 to 5 mol/L in nitric acid, trivalent An and Ln are extracted with the malonamide.

This process was recently successfully tested in the ATALANTE facility (CEA/Marcoule, France).

The possible drawbacks of this process are the: (i) necessity to use 2 extractants, (ii) need of pH adjustment, (iii) co-extraction of numerous ions, (iv) solvent clean-up not yet defined.

Multi-cycle processes

- 1st step: An+Ln co-extraction

TRUEX (USA, Japan, Russian Federation, Italy and India)

This process is based on the use of the CMPO extractant. This process was developed by Horwitz (ANL) and Schulz (Hanford) in the USA in the 80s. The advantages of the TRUEX process are the following: (i) it can extract An (and Ln) salts from acidic feeds, (ii) its efficiency has been demonstrated with genuine HAWs, (iii) a large experience has been gained worldwide. The main drawbacks of the TRUEX process are the: (i) necessity to use large concentration of tri-n-butylphosphate (TBP) as solvent modifier added to the solvent to prevent third phase formation, (ii) stripping of metal ions which are not so efficient, (iii) delicate solvent clean-up.

DIAMEX (France, Italy, Germany, Europe, Japan, USA and India)

This process is based on the use of a malonamide extractant. The main interests of the process are: (i) An (and Ln) salts can be extracted from acidic feeds, (ii) its efficiency has been demonstrated on genuine HAWs, (France, Europe), (iii) no secondary solid wastes generated owing to the CHON character of the malonamide extractant. Its main drawback relies in the partial co-extraction of palladium (Pd) and ruthenium (Ru) with the MAs.

A process based on a new type of diamide, a diglycolamide (DGA), which is a terdendate ligand having better affinity for An(III) than the malonamide, is under development at JAERI (Tokai, Japan).

TRPO (INET, Tsinghua University, China)

The TRPO process is based on the use of a mixture of tri-alkyl phosphine oxides (R3P(O), with R = alkyl groups) as extractant. This process has been tested successfully in China with genuine HAW. Its main drawbacks concern the necessity: (i) to adjust the feed acidity, (ii) to use a concentrated nitric acid solution for An(III)+Ln(III) stripping, which complicates the subsequent An(III)/Ln(III) partitioning step.

- 2nd step: An(III)/Ln(III) separation

TALSPEAK and CTH processes

The TALSPEAK process, developed at ORNL (USA) in the sixties and then adapted (CTH process) at Chalmers University, Göteborg, Sweden, can be considered as the reference process for An(III)/Ln(III) group separation. It is based on the use of HDEHP as extractant and DTPA as the selective An(III) complexing agent. The An(III)/Ln(III) separation is performed by the selective stripping of An(III) from the HDEHP solvent loaded with the mixture of An(III)+Ln(III) under the action of an aqueous solution containing DTPA and an hydroxocarboxylic acid, like lactic, glycolic or citric acids. The advantages of this process are: (i) the large experience gained worldwide, (ii) its good efficiency. Among the main drawbacks one can cite: (i) the necessity to adjust the pH of the feed, (ii) the limited solvent loading of metal ions, (iii) the difficult solvent clean-up.

SANEX concept (acidic S-bearing extractants)

– CYANEX 301 process (China, USA, Germany)

The CYANEX 301 extractant consists in a dialkyldithiophosphinic acid (R2PSSH, with R = an alkyl group). Its use for An(III)/Ln(III) was first proposed by Zhu at Beijing (China) in 1995. The main interest of the process relies in: (i) the large efficiency for An(III)/Ln(III) separation, (ii) the fact that the process has been tested with genuine An(III)+Ln(III) mixtures. Nevertheless, for an efficient use of this process the feed solution should be adjusted to pH 3 to 5, which is not so easy to carried out industrially. Moreover, the solvent clean-up is also a weak point.

– ALINA process (Germany)

To cope with the main drawbacks of the CYANEX 301 process mentioned above, Odoj and Modolo at Jülich (Germany) proposed the use of a synergistic mixture made of bis(chlorophenyl)dithio-phosphinic acid ((ClΦ)2PSSH)+tri-n-octylphosphine oxide (TOPO) to perform the An(III)/Ln(III) group separation. If the separation factors between An(III) and Ln(III) are less than those observed with CYANEX 301, the concentration of nitric acid in the feed can be as high as 1.5 mol/L, which makes the ALINA process more attractive than the CYANEX 301 one. The ALINA process was successfully tested with genuine wastes. The possible drawbacks of this process are: (i) the solvent clean-up process not yet defined, (ii) the generation of P- and S-bearing wastes (from the degraded extractants) which should be managed.

SANEX concept (neutral N-bearing extractants)

– BTPs (Germany, France, Europe)

After the discovery by Kolarik at FZ Karlsruhe (Germany) of the astonishing properties of the bis-triazinyl-1,2,4-pyridines (BTPs) for An(III)/Ln(III) separation, a process was readily developed and tested in the frame of the European so-called NEWPART project [3]. Successful hot tests were achieved both at the CEA/Marcoule and at the ITU in Karlsruhe using the n-propyl-BTP. Large efficiency of the BTP process was obtained. One should mention also that the feed of the n-propyl-BTP process can be acidic ([HNO$_3$] = 1 mol/L). Nevertheless, even if this system seems very promising, an instability of the n-propyl-BTP

extractant was observed. As a consequence, efforts are underway at the CEA to modify the solvent formulation to suppress this major drawback.

– TMAHDPTZ+octanoïc acid (CEA, France)

A synergistic mixture made of the terdendate N-ligand, 2-(3,5,5-trimethylhexanoylamino)-4,6-di-(pyridin-2-yl)-1,3,5-triazine (TMAHDPTZ), and octanoïc acid was developed at CEA/Marcoule. A process flowsheet was defined and successfully tested with genuine effluent with good efficiency. The main drawbacks of this process are: (i) the required pH adjustment of the feed, (ii) the management of secondary wastes not yet defined.

- 3rd step: Am/Cm separation

For this step, processes based on the selective oxidation of Am at the +VI or +V oxidation states are developed, the curium remaining unchanged as Cm(III), allowing simple Am/Cm separation processes to be defined.

SESAME process (CEA, France, Hitachi, Japan)

In strong oxidising conditions, Am can be oxidised from Am(III) to Am(VI). This can be done, for example, by electrolysis in the presence of heteropolyanions (HPA) acting as catalyst. The so-generated Am(VI) can be separated from Cm(III) by extraction, for example by TBP. This is the principle of the so-called SESAME process developed at CEA/Marcoule. At Hitachi (Hitachi city, Japan), oxidation of Am to Am(VI) is obtained by the use of ammonium persulphate. Then, Am(VI) is extracted by TBP. The SESAME process exhibits a great efficiency for Am/Cm separation. A large experience was obtained at the CEA at pilot scale during the last twenty years with a SESAME like process (kg amounts of ^{241}Am were purified). Nevertheless, the industrialisation of the process is faced with difficulties such as: (i) the instability of Am(VI), (ii) the non-easiness to develop a multi-stage process, (iii) the generation of secondary solid waste (made of HPA constituents).

Am(V) precipitation (JNC, Japan)

The selective precipitation of double carbonate of Am(V) and potassium (K) is one of the oldest method for Am/Cm or Am/Ln separations, developed at the end of the 60's in the USA. This method requires the use a 2 mol/L K_2CO_3 solution in which the mixture of Am(III) and Cm(III) is dissolved. After chemical or electrochemical oxidation of Am(III) to Am(V), Am(V) precipitates from the solution as the solid crystalline $K_5AmO_2(CO_3)3$ nH_2O, while Cm(III) remains in solution. After filtration, Am is separated from Cm. This process: (i) is simple, (ii) is selective for Am, (iii) has been largely used worldwide. The process main drawbacks are: (i) the Am losses with Cm, which are not so low, (ii) the fact that it exists only one stage for the process, (iii) the large amounts of secondary wastes generated.

2.2.3 Fission products partitioning

Iodine (^{129}I)

The separation of iodine is done just after the spent fuel dissolution step within the PUREX process. Oxidation of iodide ion, I^-, into elemental iodine (I_2) induces its transfer to the dissolver off-gases (DOGs) where iodine is recovered through DOGs basic washing. To recover most of the iodine spent fuel inventory at that step, slight improvement of the efficiency of the transfer of iodine from the dissolution liquor to the DOGs seems required.

Technetium (^{99}Tc)

The soluble fraction of Tc contained in the spent fuels exists in the dissolution liquor as Tc(VII) (TcO_4^-). Its co-extraction with Zr(IV), then U(VI) and Pu(IV), by TBP is well known. So, for example, the separation of the Tc soluble fraction is achieved through a solvent special scrubbing step in the course of the implementation of the PUREX process at COGEMA La Hague reprocessing plants. If high Tc partitioning yield is required, the main problem concerns the recovery of the Tc fraction that is contained within the solid insoluble residues remaining after spent fuel dissolution. A special process is required for this Tc recovery, which actually does not exist.

Caesium and strontium or caesium alone

Many processes were developed worldwide in this field, including the use of:

– Inorganic sorbents, like for the JAERI's 4 group partitioning process.

– Crown-ether extractants, like for the SREX and CSEX processes developed in the USA (ANL).

– Cobalt dicarbollide extractants, as developed in Czech Republic, Russia and Western Europe.

– Calix-crown extractants, as developed in France, Western Europe and in the USA.

Most of these processes were successfully tested with radioactive effluents.

2.3. Pyro processes for actinide partitioning [4]

2.3.1 Selected media and possible separation techniques

Selected media

Most of the "pyro" processes developed so-far are based on the use of one or two of the following high temperature liquid phases:

- Fused salts. The most popular fused salts studied are:
  - Molten chloride eutectic, such as LiCl+KCl.
  - Molten fluoride eutectic, such as $LiF+CaF_2$.

- Fused metal, such as Cd, Bi, Al, etc.

Separation techniques

To partition the actinides contained within the fused salt baths, three main techniques are studied and developed:

- Actinide electrodeposition on solid (pyrographite or metal) or liquid metal cathodes.

- Liquid-liquid extraction of actinide between fused salt bath and a metal bath containing a reductive metal solute (Li for example).

- Actinide oxide precipitation from the fused salt under the proper control of the oxygen thermodynamic activity within the salt bath.

2.3.2 Examples of strategies and "pyro" processes

2.3.2.1 USA (ANL, Chicago)

A "pyro" process was developed at ANL in relation with the treatment of FR metallic fuels (EBR II's type) for stabilisation of these Na bonded fuels. The aims of the process is limited. It consists in the separation of the spent fuel into three major fluxes: (i) most of the uranium as a low level waste, (ii) cladding+noble metals+Zr as metallic waste, (iii) TRUs+FPs+Na+salt incorporation into a zeolithe matrix in order to obtain a ceramic waste after hot pressing. The key step consists, after the oxidative dissolution of the spent metallic fuel in LiCl+KCl eutectic bath at 500°C, into the separation of most of the uranium by electrorefining on a solid cathode. A demonstration campaign involving the treatment of 100 core assemblies (0.4 ton of spent HEU) and 25 blanket assemblies (1.2 tons of spent depleted U) was successfully carried out at Argonne West in the recent years. License for pyroprocessing the whole EBR II spent fuel inventory was obtained recently.

2.3.2.2 Russian Federation (RIAR, Dimitrovgrad)

The pyro-process developed at RIAR concerns the treatment of spent oxide fuels (UOX and MOX) in order to recover U and Pu for MOX fuel re-fabrication by the vibro-compaction process.

The spent oxide fuel is dissolved by chloration in a Li, Na, K, Cs chloride fused salt bath at 650-700°C. Separation of U, Pu or mixture of U+Pu from the salt bath can be obtained by electrodeposition or precipitation. For example:

- U can be separated as UO_2 (which is a good electric conductor) by electrodeposition on a cathode made of pyrographite, while chlorine gas is generated at the anode.

- As PuO_2 is a bad electric conductor, it cannot be electrodeposited on solid cathode. But PuO_2 can be selectively separated by precipitation after bubbling a mixture of Cl_2+O_2 gases into the fused salt bath.

- Under the addition of a mixture of Cl_2+O_2 gases into the fused salt bath, which stabilises Pu as oxychlorides, electrolysis generates a deposit of $(U,Pu)O_2$ onto the pyrographite cathode while chlorine gas evolves at the anode.

An important experience with spent fuel pyroprocessing has been obtained at RIAR with the treatment of:

- 3.3 kg of UO_2 spent fuel (1% burn-up) from the VK-50 reactor, done in 1968.

- 2.5 kg of UO_2 spent fuel (7.7% burn-up) from the BOR-50 reactor, done in 1972-73.

- 4.1 kg of $(U,Pu)O_2$ spent fuel (4.7% burn-up) from the BN-350 reactor, done in 1991.

- 3.5 kg of $(U,Pu)O_2$ spent fuel (21-24% burn-up) from the BOR-60 reactor, done in 1995.

*2.3.2.3 Japan*

- CRIEPI

   The "pyro" process developed at CRIEPI concerns both the treatment of spent fuels from LWRs (oxide fuels) or FRs (metallic fuels) and the partitioning of TRU elements from the wastes issuing the reprocessing of spent LWR fuels by the PUREX process. The fused salt selected is the LiCl+KCl eutectic in which the spent fuels or the oxides of high active wastes

are dissolved by a carbo-chloration technique. After dissolution, U can be electrodeposited as a metal on a solid cathode, then the TRUs can be recovered by electrolysis using a liquid cadmium or bismuth cathode. CRIEPI is also studying the partitioning of MAs by liquid-liquid extraction using a LiCl+KCl salt bath and Cd or Bi metallic solvents containing Li as a reducing agent. In this case, liquid-liquid extraction corresponds to the reductive transfer of a metal from the salt bath, where it exists as M^{n+}cation, to the metal solvent, where M exists as a M^0 solute.

The equation of the extraction reaction can be written as follows:

$$M^{n+}_{(salt)} + n\, Li^0_{(Cd\ or\ Bi\ metal\ phase)} \Leftrightarrow M^0_{(Cd\ or\ Bi\ metal\ phase)} + n\, Li^+_{(salt)} \tag{4}$$
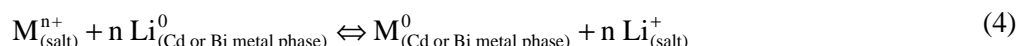
Large expertise has been gained by CRIEPI in this field but only with surrogates of actinides. A joint CRIEPI-ITU programme is under way to test the process with actinides.

- JAERI

  JAERI is studying pyroprocessing for the possible treatment of nitride, oxide or metallic spent fuels in order to prepare nitride fuels enriched with ^{15}N for FRs. After dissolution of the spent fuels into a LiCl+KCl eutectic salt bath, the actinides will be electrodeposited on solid or liquid (Cd) cathodes. The recovered actinide metals will then be converted into actinides nitrides after their dissolution in liquid cadmium. The nitruration agent will be N_2 or Li_3N.

- JNC

  JNC is also engaged in the development of pyro processes aiming to reprocess FR spent fuels. The method selected are similar to those studied by CRIEPI: (i) choice of LiCl+KCl eutectic bath, (ii) electrodeposition method, (iii) liquid-liquid extraction between salt bath and liquid Cd metal.

2.3.2.4 France

Two years ago, the CEA has launched a programme dedicated to the partitioning of MAs by "pyro" processing. A research team was created at CEA/Marcoule and special hot facilities have been created. The programme selected is rather wide. It will consider both chloride and fluoride melts and the most important separation techniques known to be effective in "pyroprocessing", i.e. (i) electrodeposition, (ii) oxide precipitation, (iii) liquid-liquid extraction between fused salt bath and a metallic solvent. The results obtained to-date concern mainly the basic understanding of the chemistry of actinides (U, Pu and Am) in solution in the fused melts. Process developments are also underway and active tests on irradiated objects are foreseen to be done before 2005.

2.3.2.5 Czech Republic

At Rez Institute, Czech scientists are developing a process based on the dual use of actinide hexafluoride volatilisation and pyroprocessing of the wastes from a fluoride melt. This research programme is connected with the interest of Czech Republic for the development of the molten salt reactor (MSR) technology. Facilities are under construction at Rez Institute for testing the pyro-processes.

3. Conclusions and perspectives

3.1 Conclusions

Numerous concepts have been consolidated or newly developed during the last few years, both in "Hydro" and "Pyro" processing of HAWs or spent fuels and targets for "new" nuclear systems.

Tests on "real objects" were carried out successfully in several countries, including the EBR II demonstration test at Argonne-West (UA) on pyroprocessing of FRs spent fuels.

In the domain of "Hydro", blooming of concepts is observed. Multi-step processes look promising but most of the systems developed so far appear complex. Efforts to simplify the processes seem required.

In the domain of "Pyro", strong consolidations of "old concepts" were obtained, including fluoride volatilisation.

3.2 Perspectives

- Hydro

    It seems important to work in order to increase the "simplicity" and "compacity" of the MAs and LLFPs separation processes. Some routes for improvement can be proposed:
    – One cycle process.
    – Consideration of High Active Concentrates instead of High Active Raffinates for process development (large volume reduction factor).
    – Integration of MAs and LLFPs separation processes.
    – Consideration of possibly new LLFPs for partitioning.
    – Maintaining alive the "CHON principle" (minimisation of secondary solid wastes).

- Pyro

    Directions for improving the processes appear to be:
    – Minimisation of TRU losses in wastes and increase of the purities of the separated.
    – Actinides which can be obtained through the combined use of several separation techniques and multi-stage techniques.
    – The waste problem, which is mostly corrosion related owing to the aggressive.
    – Character of the media and the high process temperatures, needs to be precisely estimated.
    – Consideration of the possible separation of LLFPs.

- Collaborations

    It seems a pressing necessity to maintain, or best to increase, the collaborations in this complex field at:
    – National levels: maintain or create network(s) between academic and applied research bodies. As example in France it exist two networks working under the auspices of the

December 1991 Nuclear Waste Act: the so-called PRACTIS and NOMADE Groupes de Recherches.

- Bi-national levels: numerous collaborations exist, e.g. CRIEPI-ANL, CEA-JNC, CEA-JAERI, etc.

- Regional level. As example at the European level it exists common works partly financed by the EU, e.g. the PARTNEW, CALIXPART and PYROREP programmes within the 5th FWP of EU (2000-2003). The role of ITU at Karlsruhe is also very important for European and wider collaborations,

- At the International level, the roles of OECD/NEA for Workshops and Working Parties managements and also of IAEA appear essential.

So, within a few years, one predicts that a large array of robust "hydro" and "pyro" processes will be available for the definition of new scenarios for the management of nuclear wastes generated through LWRs and FRs closed fuel cycles, but also for the fuel cycles of futuristic nuclear systems, such as the MSRs or the ADSs.

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SESSION IV

Basic Physics, Materials and Fuels

Chairs: S. Pilate (BN) – H. Takano (JAERI)

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SUMMARY

Session IV was subdivided into three parts devoted respectively to basic physics, materials, fuels and targets.

1. Basic physics

Nuclear data measurements performed or supported by JNC in Japan concern fission and capture cross-sections for Am, Np and long-lived fission product nuclides. In the case of ^{99}Tc, activation measurements revealed significantly different from previous results. It was noted that a co-operation with ORNL is planned.

Residue production in spallation reactions is being studied by researchers from Spain, Germany and France. New experiments are made at Saturne and NESSI (GSI) to identify the numerous isotopes created; an accuracy of 10% on their production is aimed at.

The experimental programme MUSE in MASURCA at CEA-Cadarache, launched in 1995, now continues with 16 different partners and with EC funding. In the present MUSE-4 configuration, a strong pulsed neutron source is fed from the coupled accelerator GENEPI. The k-value will progressively be decreased to 0.95. After the Na-cooled core, a gas-cooled core will be mocked-up.

In parallel, a complementary programme SAD will be run at Dubna (RF), using real spallation sources produced by a synchrotron.

A simplified version of MUSE-4 in RZ geometry is also proposed as a benchmark; the calculations already available indicate differences of e.g. 0.8% on the k-level.

Results of ADS benchmark calculations have been gathered by OECD/NEA and PSI/CEA. NEA had already organised a first, preliminary ADS exercise in 1994. This follow-up exercise started in 1999. Pb-Bi is retained as ADS target and reactor coolant. Two cores are considered: at start-up and at equilibrium. The external source is pre-defined. Significant discrepancies can be observed among the

7 solutions, obtained with the 3 basic data files ENDF/B-VI, JEF 2.2 and JENDL 3.2. The k-values differ by as much as 3% dk, and this is not only an effect of basic data libraries.

A next exercise is planned on a transient ADS benchmark (beam trip). It was pointed out that the partners should give more details on their data processing.

2.   Materials

The corrosion of stainless steels in a Pb-Bi circuit has been studied by CIEMAT for temperatures ranging from 400°C (cold leg) to 550°C (hot leg). The loop was made of austenitic steel while test samples were made of 2 martensitic steels. The oxide layer formation was recorded after different operation times up to 3 000 hours. A gas with 10 ppm O_2 was bubbling in the hot leg.

The oxide protection layer grew with time. The coolant dissolved some elements of the steel, mainly nickel.

Such experiments, crucial for the use of Pb-Bi coolants, should be made again, provided the exact O_2 activity be well monitored.

Two other papers were devoted:

- A Russian one, to the production of residues from spallation (as above).

- A Korean one, to thermal and stress analysis for the HYPER target; HYPER is the accelerator driven system developed by KAERI, based on the use of Pb-Bi coolant (another KAERI paper also considered the problem of transmuting ^{99}Tc and ^{129}I in HYPER, what is very difficult).

3.   Fuels and targets

While a paper by industrial companies stressed the interest to develop practical concepts of Am targets, to be placed in special, moderated positions in fast reactor cores, two very interesting papers by ITU and CEA described the experimental programmes devoted to new fuels and targets in Europe. Both are complementary, and the EC sponsorship stimulates a vast European collaboration.

Different promising concepts will be examined and tested, as for example IMMOX (Inert Matrix MOX), THOMOX (where ThO_2 is a "quasi-inert matrix"), MATINA (with macromasses instead of micro-dispersion), ECRIX/CAMIX/COCHIX to be loaded in Phénix.

Two laboratories for minor actinides have recently been built in Europe, the one at Marcoule (ATALANTE) and the other one at ITU Karlsruhe, allowing handling Am in the kg range.

In Japan, JAERI also builds a new facility TRU-HITEC, for high-temperature chemistry of Am and Cm. Available at Tokai in 2002, it will allow to handle dozens of grams of Am and Np, in addition to Pu. The research is centered there on nitride fuel with inert matrix, and on pyro-processing (as described in a JAERI paper on ADS transmutation in Session V).

The set-up of these three laboratories for minor actinides handling is a concrete result of the "decade of revival" for transmutation research, which had been illustrated in the bright overview paper introducing Session IV, given by Mr. M. Salvatores.

The coming decade should now help identify the most efficient transmutation options thanks to irradiation experiments.

TRANSMUTATION: A DECADE OF REVIVAL ISSUES, RELEVANT EXPERIMENTS AND PERSPECTIVES

OVERVIEW PAPER

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Abstract

For more than a decade, transmutation studies have been again a topic of wide interest and have triggered numerous international activities, like bilateral/multilateral collaborations, information exchanges, state-of-the-art reports, conferences, but also some co-ordinated programmes and experiments.

It is legitimate to ask at this point, whether transmutation studies are still "fashionable" and why; what is known, what has been done and what should be done.

Since the motivations of national programmes are often different, due to a different context, we will take for granted that transmutation is generally seen as an option for the back-end of the fuel cycle in order to reduce the burden of potential geological storages of radioactive wastes (whatever their nature).

Finally, we also acknowledge the fact that some highly respected scientists have at several occasions during this decade expressed their doubts about the value of the transmutation option. A typical example is the position expressed by Pigford and Rasmussen, reporting the results of a study for the US National Research Council (IAEA-TECDOC-990, 2/12/1997).

1. Introduction

To give a state-of-the-art of the transmutation studies, one could make use of international publications or proceedings of the specialised conferences that have been mushrooming in this field. Of course a significant example is the OECD/NEA state-of-the-art report published in 1999: "Actinide and Fission Product Partitioning and Transmutation. Status and Assessment Report".

We will limit our analysis to a few points that we consider of special relevance. Successively, we will review some ongoing research and experimental validation studies, in order to provide a list of relevant expected results, which should have impact to shape (or re-shape) future programmes.

In this perspective, we will also indicate which are in our opinion the "missing" experiments or studies, the absence of which could jeopardise the process of decision making.

The nuclear energy "environment" is a changing one, and it is of interest to review some activities/studies/concept proposals, which are not strictly speaking in the "transmutation" domain, but which could have an impact on the conclusions which could be drawn on the potential role of transmutation.

Finally we will attempt to summarise a list of open questions and an analysis of possible (re)orientation of priorities. Of course, this paper does not deal with chemistry issues, but rather with reactor and fuel cycle technology, since the physics of transmutation is today well understood (see for example [1]).

2. Where are we?

Transmutation is of course an R&D endeavour. The potential "customers" of such R&D can be found more in the society at large and its political representative bodies than in industry. By the way, it is not evident that even a fundamental feature of transmutation (i.e. the need to reprocess the fuel) is clearly understood in that context.

In so far as customers, one should not forget that utilities look probably with some apprehension and scepticism to studies which could offer options for the back-end of the fuel cycle but which could potentially have a non negligible impact on the cost of the electricity generation, without a clear definition of criteria to evaluate costs versus benefits, and in a frame of a highly competitive environment. In fact, utilities are ready to contribute to the R&D studies but to establish sound figures for induced or direct costs on the kWh!

The nuclear reactor and fuel cycle industry has an obvious interest to be well informed of the transmutation R&D issues and results, but does not finance these activities other than marginally. This oversimplified analysis is only meant to make clear that there is an inherent difficulty to evaluate the real status of the research in the transmutation field with respect to the potential utilisation of the technology in a reasonable time horizon within stated performance and cost criteria.

In view of this difficulty, we have preferred to single out some specific scientific results, which could characterise our present understanding of transmutation and its use as an option for the fuel cycle.

2.1 The IFR concept and the homogeneous recycling

The IFR concept [2] is still the most outstanding example of an "inherently" transmutation concept in the so-called "homogeneous" recycling mode (see Figure 1 and [3]). The IFR concept can be seen as an energy producing system capable to recycle Pu and minor actinides (MA), to reach equilibrium, both stabilising the Pu and MA mass flows, and sending to the wastes only a very small fraction of the radiotoxic isotopes. This fraction is of the order of 0.1% or less, according to the announced performances of the pyrochemical process involved, which has still to be demonstrated at large scale in the frame of the transmutation application.

Figure 1. **Pu and MA management in P/T strategies[6]**
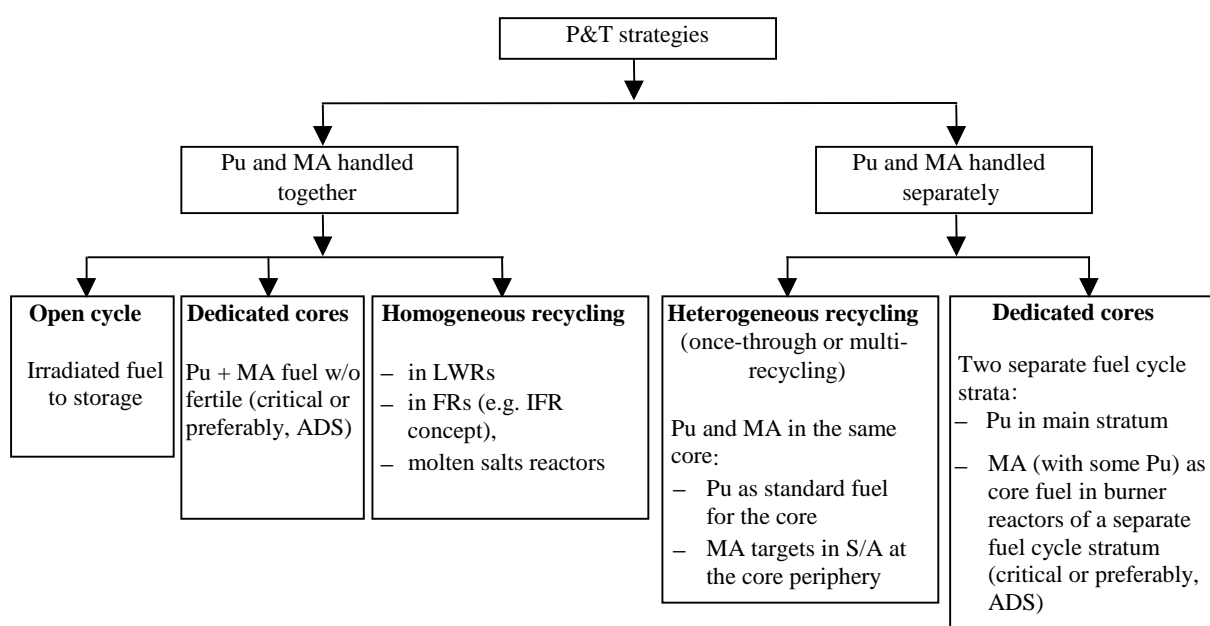
The appealing aspects of the IFR concept in the frame of transmutation are:

- The concept is mainly designed to produce energy, making an optimised use of resources and using a robust reactor and fuel cycle layout.

- The fuel cycle does not imply the separation of Pu and MA.

- The concept can accommodate in principle several options in terms of reactor size, reactor coolant, waste-forms, etc.

In general, the homogeneous recycling has equivalent performances for whatever the type of fuel in the fast reactor. In fact, if the losses at reprocessing are assumed to be of the order of 0.1%, the homogeneous recycling allows to reach a reduction of the potential radiotoxicity with respect to the open cycle scenario of a factor of 200 and more, and this over all the time scale ($10^2 \rightarrow 10^6$ years) [4]. However, the consequences on the fuel cycle have to be taken into account (see Table 1) and their impact evaluated.

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[6] If LLFP management is required, they can in principle be handled in the different scenarios as targets to be irradiated at the periphery of the different core types.

Table 1. **Consequences on the fuel cycle of MA recycling in FR[a)].**
Variation expressed as ratio with respect to the corresponding values
for the reference case: PWR-MOX Pu content: 12%, taken as 1.

		PWR – MOX 12%	FR: Multi-recycling of Pu, Am, Cm	FR: Multi-recycling of Pu, once-through irradiation of Am + Cm targets[d)]
Fabrication	Activity heat due to: α β γ Neutron source	1 1 1 1 1	0.1 0.5 0.2 1.5 30	0.1 1.7 0.4 8.7 104
Reprocessing[b)]	Activity heat due to: α β γ Neutron source	1 1 1 1 1	0.1 0.2 0.5 0.2 0.4	0.10 0.11 0.09 0.06 245[c)]

a) Oxide fuel, EFR type.
b) 5 years cooling time.
c) Effect due mainly to ^{252}Cf.
d) Heterogeneous recycling total fission rate: 90% (see text).

2.2 *Heterogeneous recycling and its potential limitations*

An option has been explored, mainly in Europe and in particular at CEA in France [5] and at JNC in Japan [6], to perform the transmutation of MA in the form of targets to be loaded in critical cores of a "standard" type. The mode of recycling has been called "heterogeneous" (see Figure 1), the potential advantage being to concentrate in a specific fuel cycle the handling of a reduced inventory of MA (separated from Plutonium). The major obstacles to that approach are:

- The very high irradiation doses needed to fission a significant amount of MA (which implies very high damage rates).

- The need to separate Am and Cm from Pu and to keep them (Am and Cm) together, in order to reach high values (~30) for the radio-toxicity reduction [3].

- The need to load the MA targets in a very large fraction (~30 ÷ 50%) of the reactor park, possibly made of fast reactors, due to their favourable characteristics for this mode of recycling (high fluxes, which can be easily tailored in energy to increase fission rates).

- Consequences on the power distributions and their evolution with time.

In any case, the consequences on the fuel cycle are relevant, if one wants to reach a factor of radiotoxicity reduction of ~30 ÷ 40 (see Table 1 and [7]).

2.3 Dedicated systems

Making again reference to the scheme of Figure 1, a possible approach to keep the MA fuel cycle and the transmutation technology separated from the electricity production, is the one which calls for the use of "dedicated" cores, where the fuel is heavily (>30%) loaded with MA, the rest being, e.g. plutonium (the ratio Pu/(Pu + MA) being <0.3). Work performed at JAERI in Japan [8] and in Europe [9], has shown that critical "dedicated" cores can have difficulties, related to the safety parameters degradation, due in particular to a very low delayed neutron fraction (<0.2% $\Delta k/k$) according to the fraction of Pu (and its isotopic vector) in the core, and to a reduced Doppler effect. This characteristic has indirectly helped to promote the accelerator driven sub-critical systems (ADS) and the so-called "double strata" fuel cycle concept [8].

It should be said, however, that a convincing, scientifically based comparison of a critical and a corresponding (i.e. same fuel, same power, same coolant) sub-critical core is still lacking, in particular in terms of safety performances for a well defined range of MA/Pu ratios and for a variety of Pu isotopic vectors. As far as transmutation performances, obviously both critical and sub-critical systems (loaded with the same fuel and for the same power) are equivalent. The "dedicated" cores approach implies that ~3-5% of a power park (in other term a "support" ratio of ≈20) is needed to stabilise MA and Pu mass flows (see for example §3.3.3).

The "dedicated" core approach has been a powerful incentive to support ADS research studies.

It is also worth to notice, that the concept of a "dedicated" core applies both to a strategy of reduction mainly of MA or of reduction of Pu and MA (see Figure 1). In fact, Pu and MA can be kept together and their inventory reduced drastically, using a fuel without "fertile" (i.e. uranium) support. In this case (where the Pu/(Pu + MA) ratio is of the order of 0.7 ÷ 0.8), ADS can still offer a valuable core concept.

2.4 Long lived fission products (LLFP)

The transmutation of LLFP has always been a controversial issue. A better understanding of the list of potential candidates for transmutation has been achieved [10]. However, there is no doubt that the task of transmutation, even if limited to ^{99}Tc and ^{129}I (^{135}Cs being out of question) is a formidable one, in terms of fuel cycle requirements. In fact, the relatively slow pace of transmutation even in the most optimised neutron environment (i.e. high flux and well-tailored thermalized spectrum), implies the presence in the fuel cycle at equilibrium of a very large amount of these fission products [11]. The use of ADS can slightly help, in terms of neutron availability, but the decrease of the support ratio (unless ^{129}I isotopic separation is envisaged) makes all the scenarios of LLFP transmutation very unlikely.

3. What is being done?

For the four areas indicated above, that we have chosen as relevant to assess the potential of the transmutation technology, it is important to review ongoing research and experimental validation studies, in order to evaluate the time horizon for their possible implementation.

3.1 The homogeneous recycling and the IFR concept

Unfortunately, after a period of intensive development, the IFR programme has been stopped. No significant activities have been left alive on the metal fuel development and the pyroprocess, with the

exception of activities started at CRIEPI (Japan) and now extended to TUI-Karlsruhe. These new activities concern also the fuel reprocessing (see [12] at this conference). In particular, an experiment (METAPHIX) is planned, in order to irradiate metal fuel pins, loaded with MA and rare earths (RE) ([13] and Figure 2).

Figure 2. **Arrangement of fuel pins in a rig**
for the METAPHIX experiment (CRIEPI-TUI) (from [13])
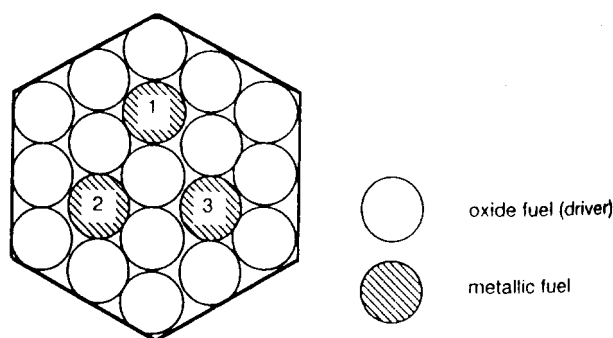
Nine metallic fuel pins are prepared for the METAPHIX irradiation study: three pins of UPuZr, three pins of UPuZr-MA2%-RE2%, three pins of UPuZr-MA5%, and UPuZr-MA5%-RE5%. They are planned to be inserted in the positions 1, 2 and 3, respectively, in the rig. Three rigs consisting of three sample metallic fuel pins and sixteen driver oxide pins will be prepared. Three rigs correspond to three different values of the burn-up: 1.5, 5 and >10%, respectively.

As far as homogeneous recycling in standard oxide fuels, some experimental knowledge has been obtained with the SUPERFACT experiment [14]. More will come from experimental programmes conceived at JNC and which should take place in JOYO beyond 2003. On the contrary, no experience exists on MA-loaded oxide fuels in standard light water reactors.

Finally, it has to be noted that the present revival of interest for pyroprocessing techniques, has been largely motivated by the relevance of these techniques to handle "hot" fuels, like those which are foreseen for transmutation. It has to be noticed that a modest but significant programme, PYROREP, has been launched as an EU contract for the 5th FWP.

3.2 Heterogeneous recycling

Apart from conceptual studies at JNC and CEA, experimental activities have been launched in Europe (e.g. the EFTTRA collaboration) and some useful indications have been gathered [15]. The EFTTRA-T4 and T4-bis experiments concern ^{241}Am, at a 12% volume fraction, inside a matrix of $MgAl_2O_4$, for a maximum fission rate of 28%. The swelling due to the decay of the ^{242}Cm produced by neutron capture, has been relevant, and triggered further research on the form of inert matrix/actinide fabrication (e.g. micro-dispersion versus macro-dispersion). It is worth to notice that experiments performed up to now, did not cover the presence of ^{243}Am and the presence of Cm.

Further experiments are planned in France, and in particular the ECRIX experiment, which should take place in PHENIX and the CAMIX and COCHIX experiments (J.C. Garnier, CEA, Private communication), also planned in PHENIX. The CAMIX experiment will provide information on "micro-dispersion" of a $(Am, Zr, Y)O_{2-x}$ compound in MgO and COCHIX information on the same

compound "macro-dispersed" in MgO or $(Zr_{0.6} Y_{0.4})O_{1.8}$. All 3 experiments are planned to reach a fission rate equivalent to 30 at%.

A significant global experiment is presently worked-out in the frame of the collaboration between MINATOM (Russia) and CEA (France) with the participation of FZK and TUI-Karlsruhe, as partners of CEA. In this experiment (AMBOINE), Am targets AmO_2+UO_2 and AmO_2+MgO should be fabricated at RIAR according to the VIPAC process. These targets should be irradiated in BOR-60 and reprocessed by pyroprocess after irradiation again at RIAR, providing in that way a full validation of the whole fabrication – irradiation – reprocessing cycle for CERCER targets (S. Pillon, CEA – Private communication).

3.3 Dedicated systems

3.3.1 Fuels for dedicated systems

For both critical and sub-critical dedicated cores, the major issue in the path towards feasibility demonstration, is the fuel development. Many candidates have been considered (see for example Table 2), but limited experimental work has been done, in order to characterise the basic properties of these potential fuels, their fabrication processes and their behaviour under irradiation.

Table 2. **Dedicated Pu + MA fuels (adapted from [16])**

Metal fuels	– Need to improve thermal properties $\Rightarrow$ add non-fissile metal with high melting point (e.g. Zr) $\Rightarrow$ Pu-MA-Zr alloy. – However: mutual solubility of Np and Zr?
Oxide fuels	– Mixed transmutation oxides as a logical extension of MOX. – However: smaller margin to melting (low thermal conductivity).
Nitride fuels	– Good thermal behaviour. – However: need to enrich in ^{15}N. – Lower stability against decomposition at high temperatures.
Composite fuels: the role of Zr	Ad-hoc "tailoring": – MgO + (Zr, An)O_{2-X} (CERAMIC-CERAMIC). – Zr + (Zr, An)O_{2-X} (CERAMIC-METALLIC). – Zr + (An, Zr) alloy (METAL-METAL). However, fabrication can be difficult (also: size and distribution of the disperse actinide phase).
Coated particle fuels	Special form of composite fuels. However in the case of fast spectra, little is known on potential candidates (TiN?). $\Rightarrow$ A generic problem: the high He production under irradiation.

A common feature for these fuels is to be fertile-free, or, at least, "U-free", since Th is sometimes considered as an acceptable support, in particular for strategies that promote the replacement of the U-cycle with the Th-cycle.

Well-structured programmes for fuel development are missing in practically all the major transmutation programmes. A significant exception is the JAERI programme, focused on nitride fuels.

The CONFIRM project, sponsored by the EU 5th FWP is also devoted to nitride fuels ((Pu, Zr) N and (Am, Zr) N). The project aims to the fabrication, characterisation and irradiation of these fuels, and addresses also the issue of ^{15}N enrichment.

Finally, we recall the initiative of the European Technical Working Group (TWG) on ADS, chaired by Professor Rubbia, which has set up an ad-hoc task force an Fuel Fabrication and Processing, in order to produce a state of the art report and an agreed work plan in the frame of a road-mapping towards ADS deployment [16].

As far as reprocessing, the situation is, obviously, not fully satisfactory either. The use of dedicated fuels imposes their reprocessing in all considering schemes. Again, the programme proposed by JAERI, includes laboratory scale experiments of reprocessing, together with a flow diagram of a process to enrich in ^{15}N the fuel [17].

The present European efforts are reviewed in [12,16]. A programme for pyrochemistry development is also being set up in France at CEA. The rationale for it can be found in the report: "Assessment of Pyrochemical Processes for Separation/Transmutation Strategies: Proposed Areas of Research – CEA/PG – DRRV/Dir/00-92, March 2000".

3.3.2 ADS systems

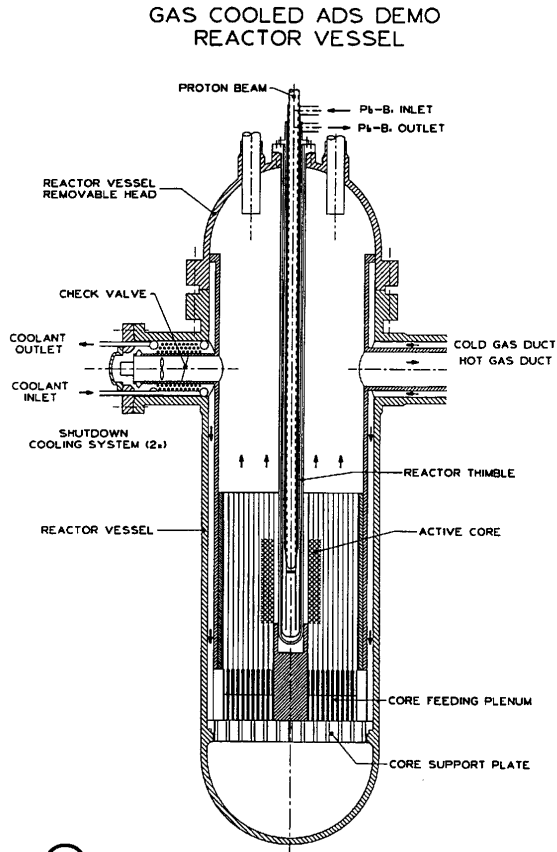A special case is the research activity in the ADS domain.

In the last two years, relevant initiatives have taken place. The ATW Roadmapping in the US [18], should give rise to a focused programme in very near future. The joint KEK-JAERI project, has given a place to ADS development in Japan, in the frame of a multipurpose facility [19].

In France, the GEDEON programme [20] has gathered a large community of physicists around the basic physics items of research for ADS (nuclear data, spallation physics, sub-critical core neutronics, materials, but also pyrochemistry, molten salts, thorium and system studies).

In Europe the Technical Working group mentioned above has been established. In that frame two concepts for ADS are being studied (see Figure 3) and a rationale is emerging for a "step-by-step" validation and demonstration of the ADS concept (see Figure 4) and its waste transmutation potential, in the frame of a specific road-mapping, which is being finalised at present. Few comments will be made in what follows, on some ongoing experimental steps, like the MEGAPIE project and the MUSE programme. Finally, the European Union is supporting a number of projects, in the frame of the 5th R&D Framework Programme.

The issues and programmes related to high power proton accelerators (HPPA), although essential, will not be dealt with here. We only remind the R&D work that has been initiated on the topic of "accelerator reliability" and which has been the subject of two NEA Nuclear Science Committee workshops (Mito, 1998, Aix-en-Provence, 1999).

Figure 3. **Sketch of ADS, liquid metal cooled (right) and gas cooled (left) (not to scale),
representative of the European EADS proposals (ANSALDO and FRAMATOME).
Potentially the same fuel assembly (e.g. SNR-300 S/A with MOX fuel).**

GAS COOLED ADS DEMO
REACTOR VESSEL

PROTON BEAM
Pb-B. INLET
Pb-B. OUTLET
REACTOR VESSEL
REMOVABLE HEAD
CHECK VALVE
COOLANT OUTLET
COOLANT INLET
COLD GAS DUCT
HOT GAS DUCT
SHUTDOWN COOLING SYSTEM (2.)
REACTOR THIMBLE
REACTOR VESSEL
ACTIVE CORE
CORE FEEDING PLENUM
CORE SUPPORT PLATE
FRAMATOME
REALISATIONS NUCLEAIRES

ANSALDO
Ansaldo Nucleare
Ramo d'Azienda di Finmeccanica S.p.A.

1. CORE
2. REACTOR VESSELS
3. ROTATING PLUGS
4. ACS
5. TARGET UNIT
6. SA TRANSFER MACHINE
7. INTERMEDIATE HEAT EXCHANGER
8. SA HANDLING CHANNEL
9. SA BASKET
10. COVER GAS COOLER

Figure 4. **A step-by-step approach to the validation and demonstration of the ADS concept**
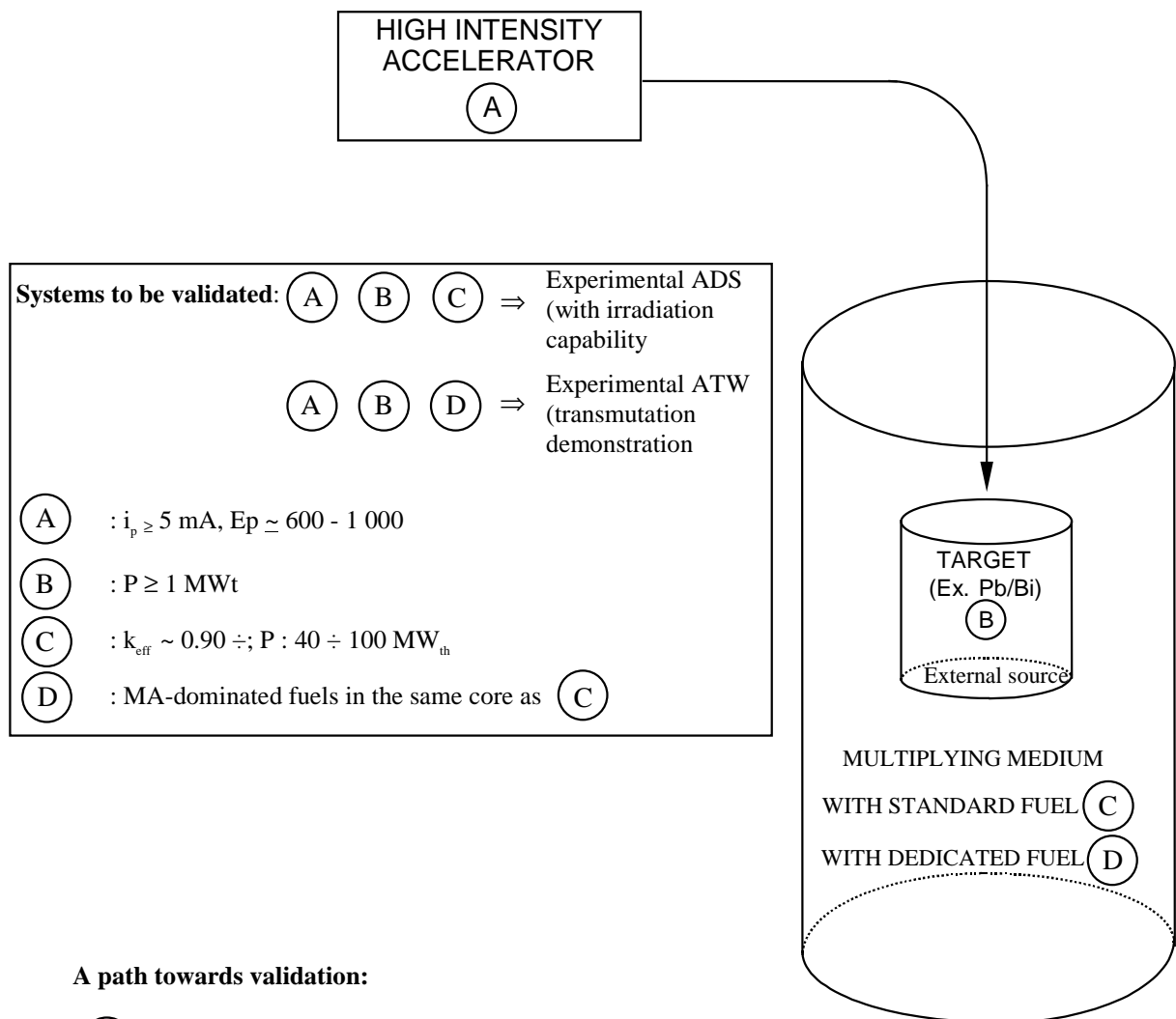
A path towards validation:

(A) : the IPH project (High Intensity Proton Injector) or TRASCO
program and follow-up programs (e.g. superconducting cavities)

(A) + (B) : the MEGAPIE project (with "known" (A))

(B) + (C) : the MUSE programme (with "known" (B))

(D)

Next steps:

(A) + (B) : spallation source $(1 \div 5 \text{ MW}_{th})$

(A) + (B) + (C) : experimental ADS $(40 \div 100 \text{ MW}_{th}; k_{eff} \simeq 0.90 \div 0.98)$ with standard fuel
(e.g. SNR-300 MOX fuel) and high flux $(\approx 10^{15} \text{ n/cm}^2 .\text{s})$ (time horizon ≈ 2015)

(A) + (B) + (D) : experimental ATW (time horizon ≈ 2025)

3.3.2.1 The MEGAPIE project [40]

MEGAPIE is an international (CEA, PSI, FZK, CNRS France, ENEA, SCK•CEN. JAERI will join soon) experiment to be carried out in the SINQ target location at the Paul Scherrer Institute in Switzerland and aims at demonstrating the safe operation of a liquid metal target at a beam power in the region of 1 MW. The minimum design service life will be 1 year (6 000 mAh).

The target material will be the PbBi eutectic mixture. Existing facilities and equipment at PSI will be used to the largest possible extent. In fact, the MEGAPIE target will be used in the existing target block of SINQ.

A vertical cut through this target block and parts of the proton beam line is shown in Figure 5.

Figure 5. **Vertical cut through the target block**
and part of the proton beam transport line of SINQ
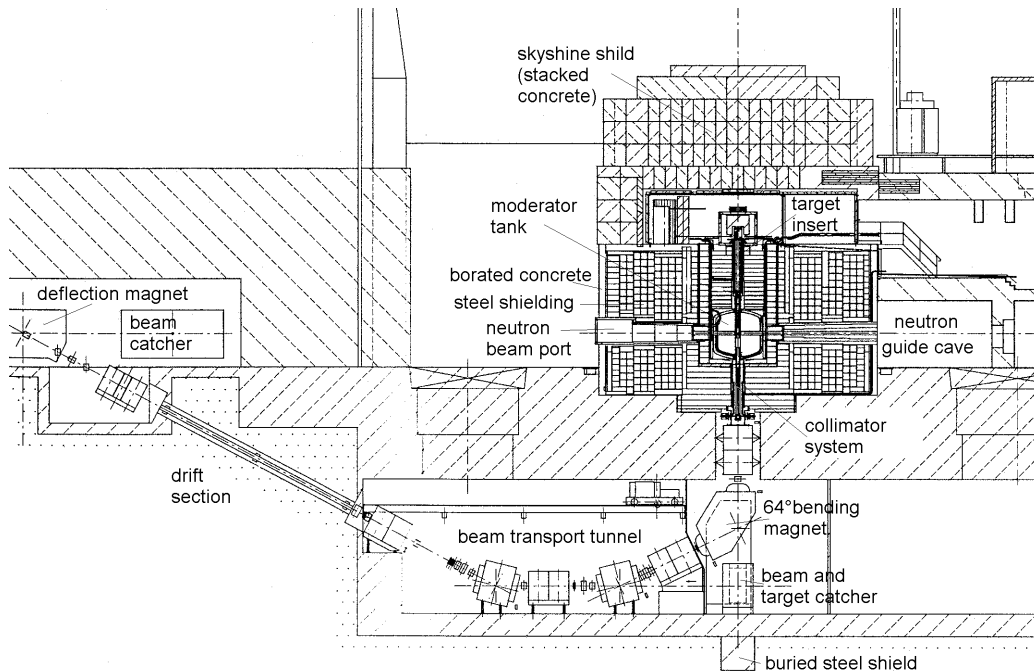
The target's outer dimensions must be such that it fits into the target position of the SINQ facility, the existing target exchange flask including its contamination protection devices and the existing target storage positions.

The target will be designed for 1 MW of beam power at a proton energy of 575 MeV, i.e. a total beam current of $i_P =$ 1.74 mA.

It is also important to realise that the stability of beam delivery cannot be guaranteed at all times. The MEGAPIE heat removal system must be able to cope with frequent short beam trips and occasional unstable operation i.e. up to days long shutdown periods.

A sketch of the MEGAPIE spallation target is given in Figure 6.

The major objectives of the MEGAPIE initiative are:

- Full feasibility demonstration of a spallation target system.

- Evaluation of radiation and damage effects of structures and beam window in a realistic spallation spectrum.

- Effectiveness of the window cooling under realistic conditions.

- Liquid metal/metal interactions under radiation and stress.

- Post irradiation examinations (PIE).

- Demonstration of decommissioning.

Figure 6. **Sketch of the 1 MW exploratory liquid lead-bismuth spallation target MEGAPIE**
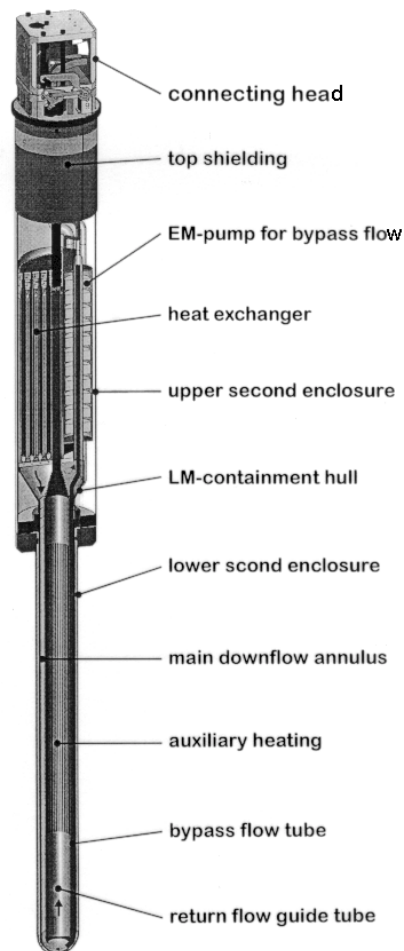
It has to be reminded that two EU contracts, established in the frame of the 5th FWP [21], SPIRE (material irradiation) and TECLA (physico-chemical properties of lead alloys: corrosion...), provide a relevant R&D back-up to the MEGAPIE project. Moreover, experimental laboratories have been launched in support of these activities (like the KALLA laboratory in FZK-Karlsruhe) or are re-oriented (like the ENEA laboratory in Brasimone: the CIRCE loop).

3.3.2.2 The MUSE experiments

The MUSE experiments, launched in 1995 [22], provide a simulation of the neutronics of a source-driven sub-critical system, using the physics characteristics of the separation of the effects due to the presence of an external neutron source from the effects of the neutron multiplication. In fact for a wide range of sub-criticality values (e.g. k_{eff}: 0.9 ÷ 0.99) the space dependence of the energy distribution of the source neutrons is quickly (in approximately one mean free path) replaced by the fission-dominated neutron energy distribution.

In practice, external known neutron sources have been introduced at the centre of a sub-critical configuration in the MASURCA reactor. The more recent of these experiments is made of a deuton accelerator and a target (deuterium or tritium) at the centre of a configuration, where actual target materials (like lead) are loaded, to provide the neutron diffusion representative of an actual spallation target (see Figure 7 and [23]). The neutrons issued from (d,d) and (d,t) reactions provide a reasonable simulation of the spallation neutrons, in terms of energy distribution (see Figure 8).

Static (e.g. flux distributions, spectrum indexes, importance of source neutrons) and kinetic parameters (e.g. time dependence of neutron population, effective delayed neutron fraction, with appropriate weighting, etc.) have been or will be measured (see [23]). Sub-criticality itself, is measured by static and dynamic techniques.

Finally, the proposed experiment MUSE-4 start-up procedure i.e. 1) critical configuration with accelerator hole but no beam, 2) sub-critical configuration with accelerator hole but no beam, 3) same, but with beam on, allows to establish a precise reactivity scale in step 1, which can be used both to calibrate eventual control rods and to measure in a standard way (e.g. with the modified source multiplication, MSM, method) the level of sub-criticality of steps 2 and 3.

The MUSE-4 experiments, described in a separate paper at this conference [23], are also partly supported by an EU contract for the 5th FWP.

3.3.2.3 Streamlining basic physics experiments

The ADS research development has also motivated a significant number of experimental activities in the field of spallation physics and nuclear data measurement and evaluation (mainly actinides and LLFP). Examples will be found in papers at this conference.

A major experiment takes place at GSI, defined in order to gather much needed information on spallation product yields and distributions in (A, Z) [24].

Also in this area, the EU supports projects in the frame of contracts for the 5th FWP [21].

If present uncertainties in nuclear data allow making reasonable pre-conceptual design assessments, future detailed studies will require more accurate data, with drastically reduced uncertainties. The relevant sensitivity studies have started (see [25]), but they have not yet tackled in satisfactory way the problem of the accuracy needs in the intermediate (i.e. 20 MeV ≤ E ≤ 200 MeV) energy range.

Figure 7. **The MASURCA installation for the MUSE programme**

The GENEPI
deuton accelerator

Vertical loading of
MASURCA tube

MUSE configuration

Deuton beam (from GENEPI)

☐	MASURCA tube (10×10 cm)
▦	Steel shielding
▨	NaSS reflector
❚❚	Core (Na-UO$_2$ - PUO$_2$)
▓	Lead (simulating spallation target)
▒	Tritium or deuterium target

Beam pulse width $\simeq$ 1 µsec

Source intensity $\approx 2 \times 10^{10}$ n/s

Sub-critical core: $k_{eff} = 0.95 \div 0.98$
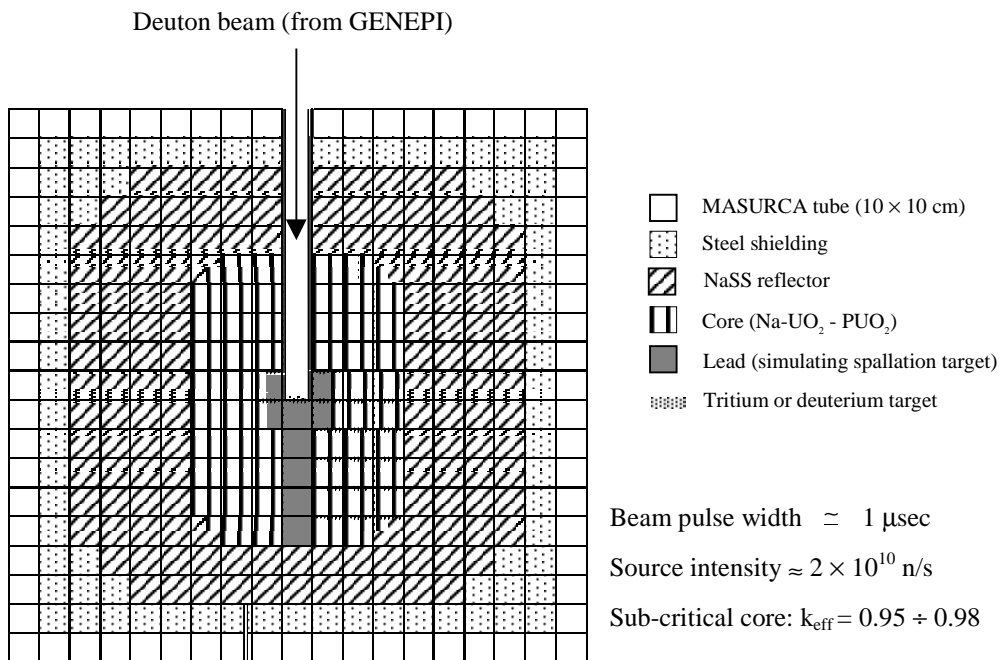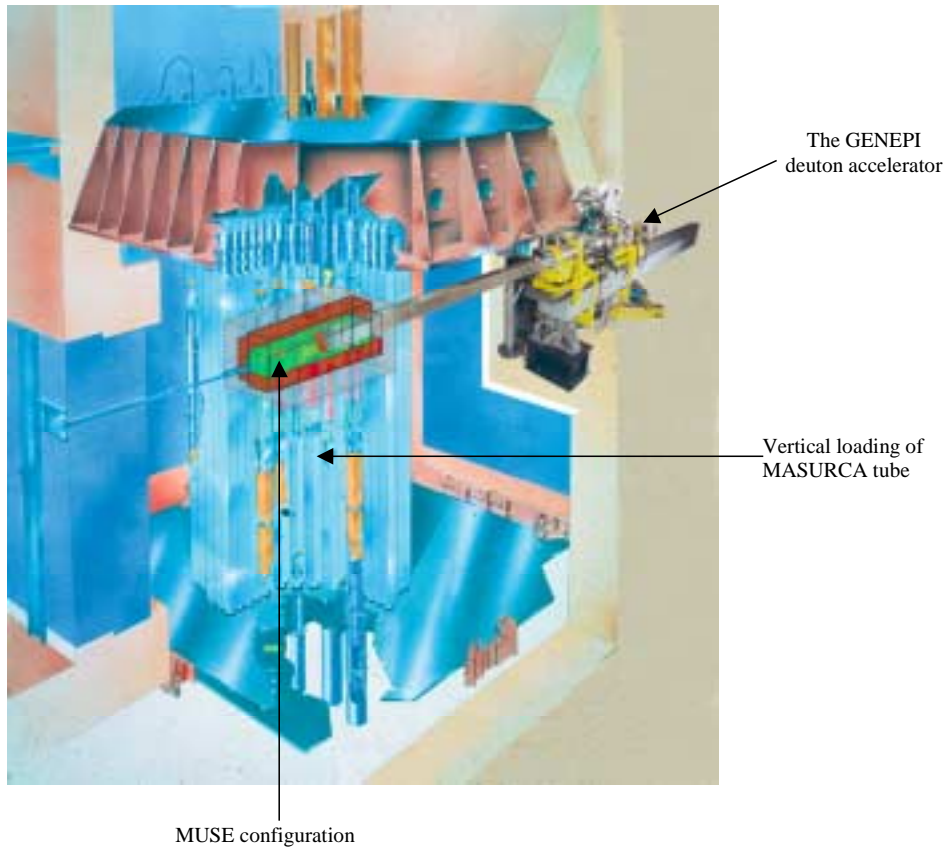
A sub-critical MUSE-4 configuration

Figure 8. **Comparison of the neutron spectra obtained with (D,D) and (D,T) neutron sources (as in MUSE experiments), with the reference spallation source, in the same configuration**
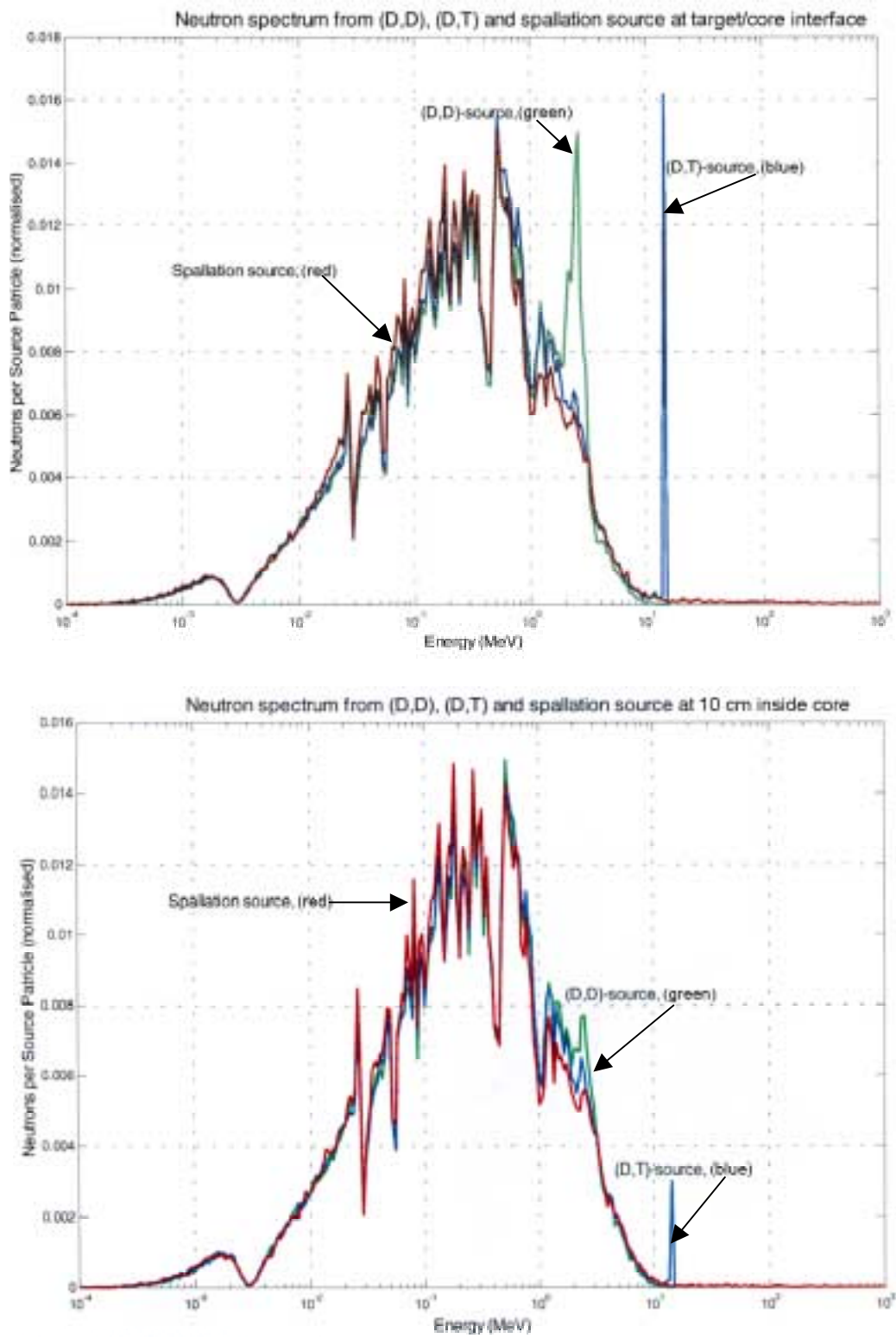
3.3.3 Scenarios studies

Scenario studies have allowed during this decade to get a global picture of the transmutation potential, mass flows at equilibrium, and consequences on the power park structure. In the illustration of Figure 9 [26], the same type of ADS is used in order to transmute MA (double strata approach), or Pu + MA (double component type of power park [27]).

The fractions of ADS in the park at equilibrium are shown (respectively 3.4% and 16%), and also the MA and Pu yearly mass flows, including total losses towards a deep geological storage.

3.4 LLFP

In this area, after the performance of irradiation experiments on ^{99}Tc and Iodine [28], not much is being done apart from conceptual studies, that underline the need to use high fluxes and thermalised spectra, like in the so-called "Leakage-with-Slowing Down" (LSD) approach [11].

Projects related to the transmutation of ^{90}Sr and ^{137}Cs have finally been abandoned everywhere.

Cs transmutation is more and more considered as non-realistic (even with isotopic separation). Other activation products have been mentioned as candidates for transmutation, but the inherent difficulty of high neutron-consuming processes has discouraged further experimental programmes.

Figure 9. **Scenarios at equilibrium for a 60 GWe Park – Mass flows/year (t) (only TRU)**

Double strata: to transmute MA (separated from Pu).
Double component: to transmute Pu + MA (non-separated).
Same type of sub-critical ADS (gas-cooled, particle-fuel).
Power: 1 500 MW$_{th}$.
Initial sub-criticality: k$_{eff}$ = 0.98 (i$_p$ ≈ 17 mA, E$_p$ = 1 GeV).
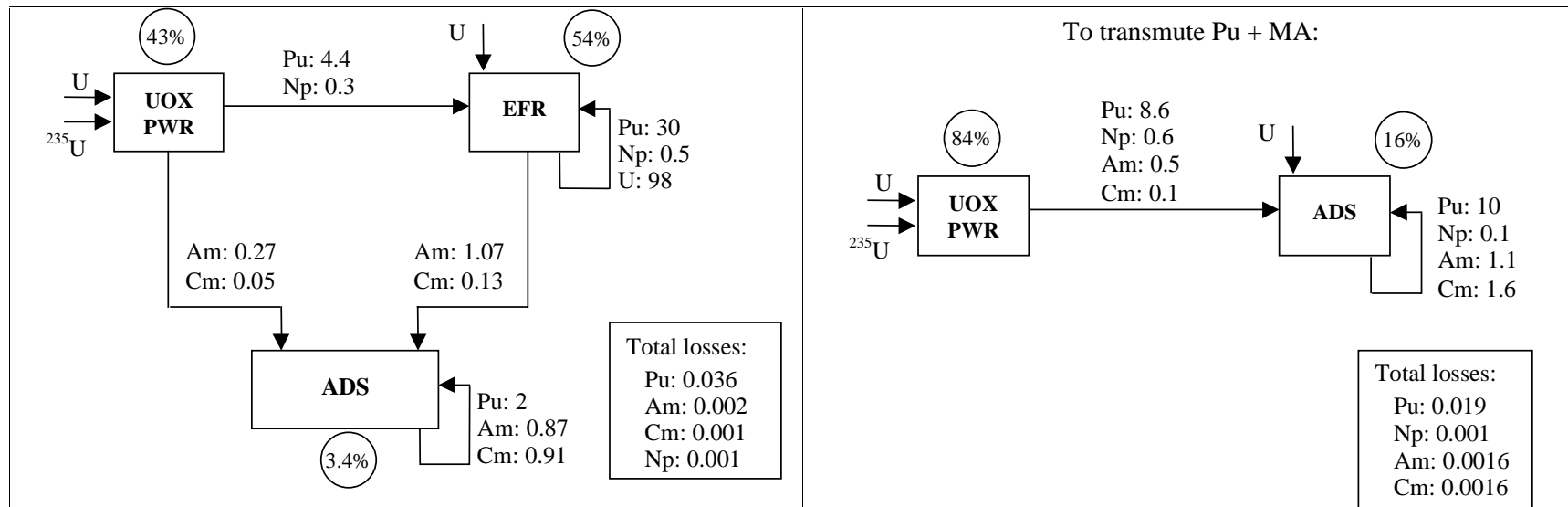
(%) : fraction in the power park.
UOX-PWR: 4.9% ^{235}U enrichment. BU: 60 GWd/t.
Cooling time: 5 years.
Ageing before irradiation: 2 years.
Losses to the wastes: U, Pu, MA= 0.1%.

4. Major missing points

The analysis of the ongoing activities and of their relevance allows attempting an indication of areas where further efforts are needed, in order to consolidate the present knowledge and to provide elements to judge feasibility.

- It seems that what is probably still needed with a high priority is a (re-)assessment of the criteria to judge the performance of transmutation systems. Transmutation as a waste management option, is indissolubly related to a constant or expanded use of nuclear energy and its impact should be evaluated on the full fuel cycle (cost and licensing of new installations, doses to the workers, secondary wastes, acceptability, ...).

- Experimentation about fuels is a priority. No concept can be considered seriously, if the appropriate fuels are not defined, which means characterised, fabricated, irradiated and reprocessed. Now, very limited facilities are available to deal with MA fuel fabrication and reprocessing (wet or dry routes) and their workload is already very demanding. Moreover, the problem of Cm has been up to now somewhat "forgotten" and, on the contrary, it can be crucial to define an optimum transmutation strategy or even to identify potential "show stoppers".

  To start with compounds and fuel basic properties assessment should be a priority. An international co-ordination and share of work should be envisaged.

  Also, irradiation tools with fast neutrons will be dramatically reduced in the coming years with the remarkable exception of the JOYO reactor (at least up to ~2015). Again, an international initiative could be envisaged to harmonise programmes and to allow the best use of existing resources.

  Reprocessing of irradiated fuels should be foreseen as an essential step of any programme on fuels, for homogeneous recycling of fuels, heterogeneous recycling of targets, or dedicated fuels. The priority, in the opinion of the author is with fuels for homogeneous recycling and with dedicated fuels, if the double strata, (or the "double component") approach is accepted.

- In the case of heterogeneous recycling, the feasibility of a fission rate >90% should be experimentally verified, and that demonstration should be made in the case of a target containing both Am and Cm. In fact, multi-recycling of targets should be avoided.

- It is more and more evident that transmutation studies could not necessarily require the separation of individual MA and MA from Pu. Quite the opposite option seems to be more attractive [3]. In that respect, partitioning/conditioning strategy [29] could represent an option to be investigated, and which could justify partitioning by itself.

- In the case of ADS, an Experimental ADS (EADS) realisation at the 2015 horizon, is a need, in order to prove the technology at a significant scale. The priority is the engineering concept (i.e. component coupling, control, reliability and licensing) validation. The double role of an EADS as a facility to validate the concept but also able to provide the appropriate fast neutron field for advanced fuel irradiation at high damage rates, is a strong point to be made.

  The present status and the necessary research in the accelerator field are the subject of another paper at this meeting. It is however necessary to remind here that this is an essential issue, since the expected performances of the "dedicated" high power accelerators are very demanding in terms of reliability and availability.

- LLFP transmutation research, if at all needed, should concentrate on a realistic approach for ^{129}I handling, if any. Reliable target materials and high transmutation rates should be the priority

goals in this field. Since once-through transmutation is hard to be envisaged, the recovery of iodine in the irradiated target and its reprocessing, should also be the object of research.

5. A changing environment

The research in transmutation experienced a revival in the mid-eighties, essentially in the context of waste management within programmes which gave a definite value to Plutonium and which implied the reprocessing of irradiated fuel.

The transmutation approach was successively identified in some countries with the approach to Pu elimination (both weapon and civil Pu). Reactor physics problems were indeed very similar. In that way, the interests of the two previously separated communities (i.e. Pu = resource versus Pu = liability), were somewhat federated, in particular in terms of fuels development and their reprocessing by pyroprocesses.

The third step in the evolution of the transmutation approach is underway at present, since the objectives of a "Generation IV" or, in general, the objectives of a future nuclear power development (beyond the horizon 2030-2050) are being globally re-discussed.

Transmutation and waste minimisation are then part of the potential criteria to define future energy systems (reactor plus fuel cycle).

In this changing environment, it can be useful to single out some concepts or research areas, which can have impact on the future of transmutation studies.

5.1 Evolutionary reactor concepts

A few well worked-out reactor concepts have emerged in the last few years, which, besides attractive safety and economics characteristics, have a potential to be "inherent" MA transmuters in the homogeneous recycling mode.

Besides the IFR concept, often quoted as a paradigm in the present paper and the Energy Amplifier proposed by C. Rubbia, we can remind the BREST lead-cooled fast reactor concept developed in Russia [30], the CAPRA reactor in France [31], the SCR (Super-critical Water Cooled) concept, developed in Japan [32], but also the APA concept [33], despite the fact that it concerns mostly an innovative assembly design for PWRs.

In particular, the nitride fuel foreseen for the BREST reactor, favours the MA transmutation by neutron spectrum hardening. Using a pyrochemical process, it is possible to envisage for this fuel, by multi-recycling, the transmutation of the actinide produced during irradiation. This mode, close to the one indicated for the IFR concept, has the same advantages and of course similar drawbacks, in particular due to the build-up of spontaneous fission neutron emitters (Cm isotopes, cf. isotopes, see §2.1 and Table 1).

Finally, interest in gas-cooled fast reactors has been renewed, in particular to keep open the fast reactor (FR) option, due to FR flexibility with respect to resources utilisation and their potential for waste minimisation. In view of the "political" opposition to Na as coolant in some countries, the gas cooling is being revisited.

The potential of any fast reactor, whatever the fuel type (oxide, nitride, metal) and whatever the coolant [34], indicates that the priority for GCFRs is to design a viable reactor (in terms of safety) with a realistic fuel form (e.g. particle, avoiding as far as possible graphite) for which no firm candidate has been proposed up to now.

5.2 Molten salts

Molten salt reactors, besides their specific interest as energy producing systems, have also a number of perceived advantages for transmutation, in particular:

- High burn-up potential (up to 600 MWd/t) limited only by absorption due to fission products, minimising the quantity of fuel to be reprocessed (a few litres of salt per day).

- Actinide losses minimised in the ultimate waste form (0.01-1% of the actinide inventory).

Molten salts characteristics result also in increased flexibility:

- Continuous input of purified salt fuel and output of irradiated salt: by adjusting the TRU concentrations, reactivity can be controlled without the use of poison or fertile material.

- Long-lived fission products (Zr, I, Tc, Cs, ...) can be added directly to the salt with no detrimental effect on its physicochemical properties.

- In the case of thorium cycle, fertile thorium can be added to the salt to fabricate ^{233}U rather than TRUs; the ^{233}U can be "quickly" extracted without ^{232}U.

Recent studies on molten salts concepts [35,36] point out the application to MA and Pu elimination, but also indicate the way towards improved fuel cycle and waste management scenarios (e.g. the TASSE system [37]).

5.3 The thorium cycle

A recent study of the European Union (in the frame of a contract for the completed 4th FWP, to be continued in the 5th FWP) has addressed the issue of "Thorium as a Waste Management Option" [38].

The objective of the work was a re-assessment of Thorium cycles in the context of limitation of nuclear waste production and prospects for waste burning. The aim was to obtain a review of the major steps of the fuel cycle, focusing to the waste aspect. A restriction was made regarding reactor types: PWR, FR and ADS.

The final report of that study shows that there are important advantages of thorium cycles with respect to the waste issue that we will quote in detail from [38]:

- Long-lived radio-toxicity of mining waste is expected to be relatively small, which leads to more manageable waste as compared to the uranium case.

- Fabrication of Th/Pu-MOX fuels is comparable with U/Pu-MOX fabrication methods as long as fresh Th, fresh U and recycled Pu are used. Recycling of U bred from Th, however, needs remote handling and reprocessing techniques specific to Thorium.

- The use of Thorium in PWRs always requires make-up fuel and therefore a self-sustaining mode is impossible in such a reactor.

- To reduce the radio-toxicity of PWR waste in an once-through mode, one has to avoid ^{238}U and therefore use thorium together with make-up fuel like ^{233}U or highly-enriched ^{235}U. Advantages in terms of waste radio-toxicity are seen during the first 10 000 years of disposal. Recycling gives a further reduction of radio-toxicity up to 10 000 to 50 000 years of disposal.

- The long-term residual risk of directly disposed fuel in a thorium matrix is still not known very well, but there are indications on improved performance. Further experimental work is needed to clarify this point.

- Th-assisted Pu burning, using a Th/Pu-MOX type of fuel in a PWR, is an attractive option with respect to mass reduction of Pu.

- Fast neutron reactors and accelerator-driven systems offer both (with similar characteristics) the possibility of a closed Th cycle without make-up fuel, except to start the cycle, reducing mining needs and radiological risks. Full recycling of actinides gives impressively low radio-toxicity results for the wastes over a long period of disposal, starting after the bulk of fission products has decayed.

Non-proliferation concerns are also treated in the report.

The interest of the Th cycle should justify a number of experimental developments, in particular:

- Reprocessing and fabrication techniques of Th fuels could be extended from laboratory scale to industrial scale and further optimised.

- Co-extraction of actinides by pyrochemistry in molten salts, aiming at losses of the order of 0.1% should be demonstrated.

- Nuclear data and physico-chemical data should be established or improved.

- Some simple irradiation experiments should be foreseen, (as it is the case in the new thorium project for the EU 5th FWP).

Finally, the considerations of [37], further enhance the potential of thorium, if powerful accelerators are used.

5.4 *Multipurpose neutron source installations*

Recently, the "transmutation" community has become involved in the discussions around "multipurpose" facilities, based on a high power proton accelerator, which provides neutrons, by spallation on one (or several) target(s) for different applications.

The most known example is of course the joint KEK-JAERI project [19]. A new initiative is under study in Europe. The ADS experimental installation could be one of the "potential" customers of the neutrons (as it is in Japan), and, consequently, the transmutation community could be interested both to the possibility to demonstrate the concept, and to irradiate the dedicated fuels and targets needed to assess feasibility (see §4).

However, the need to single out a "leading" customer, can somewhat jeopardise the performance allowable for the "lesser" customers. A typical example, is the debate on the pulsed or continuous mode of operation of the high intensity proton accelerator.

6. Healthy criticism

In the reference quoted at the beginning of this report, Rasmussen and Pigford express their doubts about the value of P&T with arguments that should be carefully taken into account still today. Three of their arguments seem of particular relevance, besides the economical and institutional issues, which, although of fundamental importance, have to be adapted to each specific situation:

1. The total inventory of untransmuted radioactivity in the reactor and fuel cycle must also (besides what is sent to the repository as losses at reprocessing) be considered as a potential waste and it takes centuries to reduce it.

2. In the search for an adequate measure of performance, the repository "intrusion" scenario, is claimed to be the most affected by P&T. However, if one considers "intrusion" in a repository, why not to consider "intrusion" in the installations of the fuel cycle, where most of the inventory is kept!

3. P&T will increase to significant amounts new secondary wastes.

As far as arguments 1 and 2, it is clear (and should be always made clear in front of any type of audience), that P&T strategies are definitely associated to an (expanded) use of nuclear energy, with fuel (re)processing and relevant investments in new facilities. However, this (expanded) use of nuclear energy can be made acceptable to the public by the very fact that the burden to repositories is reduced by P&T, and that potentially physical means to eliminate all nuclear materials are provided by the same P&T technologies, even if they should be operated for long periods of time.

Finally, the problem of secondary wastes, and the more general problem of the impact of P&T on the fuel cycle installations, often mentioned in the present report has to be carefully quantified, in particular in terms of social acceptability.

In conclusions, arguments against P&T can be seen simply as arguments in favour of a simplified fuel cycle, and not necessarily in favour of the once-through cycle based on Uranium utilisation.

7. Conclusions and perspectives

Transmutation of wastes has been revisited in the last decade and, although no spectacular breakthrough has been made, a number of significant results have been obtained.

Besides the relevant results in the aqueous chemical separation process domain (which have not been reviewed here), one can quote:

- Understanding of the physics of transmutation and of the "neutron availability" concept.

- Understanding of the role of innovative fuels (including molten salts and particle fuels) to improve the characteristics of the fuel cycle and to minimise wastes.

- Understanding, in that context, of the potential of pyrochemical processes both for fuel fabrication and for irradiated fuel reprocessing.

- Understanding of the role of ADS to handle Pu and MA, but also to provide an option for an extended use of the thorium cycle.

- Understanding of the role of fast neutron spectra and their flexibility. In this frame, the discussion around the coolants for FR would benefit from a better international agreement on pro and cons of the different options.

Since fuels play a central role in all scenarios of waste minimisation and nuclear power development, an international share of efforts around nitrides, oxides and metals should be organised in order to insure an optimum use of resources in the few existing laboratories to handle very active fuels. In that frame, the availability of irradiation facilities, in particular able to provide fast spectra (and high damage rates) is a key point and a major concern.

No convincing case can be made in favour of transmutation, without the full experimental demonstration of its feasibility. Experiments are then needed and the relevant installations should be kept available, with enough experienced teams. Besides the case of the installations for fuel characterisation, fabrication and irradiation, often mentioned in this report, installations related to basic physics (nuclear data and neutronics) will remain vital for all scenarios of development.

In the field of ADS, the development of high power proton accelerators and the construction of a $60 \div 100\,\mathrm{MW_{th}}$ Experimental facility, at a realistic but not too far away, time horizon, seem to be necessary in order not to loose credibility.

International initiatives should be upgraded and, besides the very valuable information exchange goal should address the practical share of work in key fields and should help to focus on some most promising concepts, promoting joint experiments and avoiding dispersion of efforts. In this respect, a co-ordinated activity on pyrochemical processing is strongly suggested.

Finally, the results of the new study of OECD/NEA presently underway will certainly help to better understand and to agree on the relative merits of two of the major options (i.e. critical fast reactors and ADS) for waste transmutation [39].

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SESSION V

Transmutation Systems and Safety

Chairs: Y. Arai (JAERI) – W. Gudowski (KTH)

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SUMMARY

Session V started with a very interesting and important overview paper by Dr. Dave Wade (ANL) summarising safety and operational concerns for ADS. Some of these concerns were also addressed in some later presentations but this paper gave an insight in a logic deduction of the safety and operational aspects that is defining an ADS.

Different presentations were given on concepts or precise projects for ADS facilities. One should remark that some of these proposals involve multi-purpose machines based on Pb-Bi technology (i.e. KEK/JAERI, Myrrha) where some proposals involve gas-cooled systems (i.e. GA, LAESA pebble-bed). One may remark that our community could benefit of checking non-nuclear experiences with Pb-Bi coolants in order to shorten our learning curve.

It should also be remarked that pyroprocessing technology can be applied to pebble-bed's TRISO-type of fuel which indicates the potential for closed fuel cycles by recycling this fuel.

The session also showed good arguments in favour of nitride fuels.

SAFETY CONSIDERATIONS IN DESIGN OF FAST SPECTRUM ADS FOR TRANSURANIC OR MINOR ACTINIDE BURNING: A STATUS REPORT ON ACTIVITIES OF THE OECD/NEA EXPERT GROUP

OVERVIEW PAPER

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Abstract

The Nuclear Development Committee of the OECD/NEA convened an expert group for a "Comparative Study of Accelerator Driven Systems (ADS) and Fast Reactors (FR) in Advanced Nuclear Fuel Cycles". The expert group has studied complexes (i.e. energy parks) of fission-based energy production and associated waste management facilities comprised of thermal and fast reactors, and ADS. With a goal to minimise transuranic (TRU) flows to the repository per unit of useful energy provided by the complex, the expert group has studied homogenous and heterogeneous recycle of TRU and minor actinides (MA) in the facilities of the complex using aqueous or dry recycle in single and double strata architectures. In the complexes considered by the expert group the ADS is always assigned a TRU or MA (and sometimes a LLFP) incineration mission – with useful energy production only as a secondary ADS goal to partially offset the cost of its construction and operation.

Ancillary issues have also been considered – including ADS safety challenges and strategies for resolving them. This paper reports on the status of the expert group's considerations of ADS safety strategy.

1. Introduction

The term ADS comprehensively includes all non-self sustaining fissioning neutron multiplying assemblies which are driven by an external neutron source provided by a charged particle accelerator and a neutron producing target. ADS systems under current study worldwide include both thermal and fast neutron multiplying media comprised of either liquid or solid (lattice) fuel and driven by either cyclotron or linear proton accelerators and spallation targets (liquid and solid) of various heavy metals. The underlying missions targeted for ADS systems span the range from nuclear waste incineration with ancillary power production through power production with integral waste self-incineration to finally, excess neutron production for the purpose of isotope production via neutron capture reactions on targets.

The OECD/NEA expert group on "Comparative Study of ADS and FR's in Advanced Fuel Cycles" [1] confined its scope of inquiry to a subset of ADS configurations – those targeted for nuclear waste incineration with ancillary power production, and specifically those which operate on a fast neutron spectrum with a solid fuel pin lattice. Moreover, the expert group set a requirement of maximum "support ratio" (i.e. maximum energy from the reactors in the complex compared to energy from the ADS in the complex) which leads to inert matrix fuel (i.e. ^{238}U and ^{232}Th – free fuel) for the ADS. The scope of this discussion of safety strategy is similarly confined in scope. Even within the limited scope, a range of possibilities exists. The ADS might be a minor actinide (MA) burner or a TRU burner; the physics and safety characteristics of these cases differ because of differences in their values of β_{eff} (which helps to set the degree of sub-criticality of the ADS) and in their reactivity burn-up swing (which helps to set the control strategy). The choice of coolant (liquid metal or gas) and fuel type (oxide, nitride, metal) also distinguishes members of the ADS class considered by the expert group. The choice of recycle (partitioning) technology (aqueous, dry) directly affects the architecture of the energy complex and indirectly affects the ADS itself. Figure 1 illustrates the several energy producing complexes which were considered by the expert group and identifies the waste management function of the ADS studied in the single strata architecture (3B) and (the double strata architecture (4)).

The fast spectrum, solid fuel, waste incinerating class of ADS considered by the expert group shares with all ADS a distinction from critical reactors in relying on an external neutron source rather than a self generated delayed neutron source for maintaining the neutron population in balance – with attendant changes in dynamic response and in control strategy. However, the class of ADS considered here offers design and safety challenges which are unique vis-à-vis other ADS classes in the areas of burn-up control compensation and reactivity feedback characteristics; these unique challenges are traceable to a small number of salient design features which derive directly from the requirements of the TRU or MA incineration mission – with ancillary power production. The salient design features of the ADS whose safety features were considered by the OECD/NEA expert group include the following:

- Fertile-free transuranic or minor actinide fuel.

- Multiple recycle of fuel to (near) complete fission incineration of transuranics.

- Fast neutron spectrum.

- Choice of coolant (Na, Pb-Bi or He).

- Sub-delayed critical operating state.

- External neutron source created via spallation reactions of high-energy protons on a heavy atom spallation target.

The approach taken by the expert group to identify ADS safety issues and discuss strategies for addressing them was as follows. First, the top level safety functions to be satisfied for any fission chain reacting system (reactor or ADS) were enumerated. Then, the distinguishing features of the class of ADS considered here were traced back to the mission assigned to it in the complex; as a way to indicate which features (and safety issues) would be changed by a change in mission requirements. Then, an impact matrix was constructed (with "safety function" columns and "distinguishing feature" rows) to identify where the ADS distinguishing features have raised safety-relevant challenges which are different from the more familiar situation for a fast reactor. Finally, for each identified challenge, a set of alternative safety strategies for addressing it were discussed with the views that:

Figure 1. **Nuclear fuel cycle schemes**
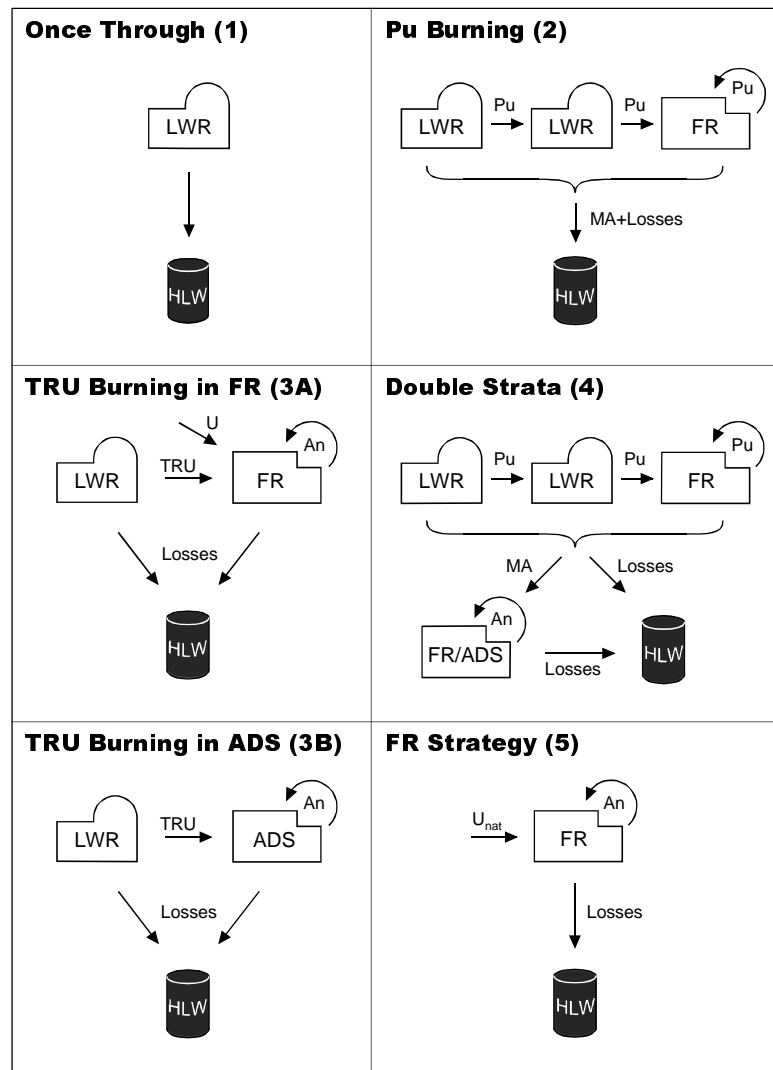
- Safety should be "designed in" from the outset.

- The vast experience base from fast reactor development should be exploited where possible, e.g.:
    - Defence in depth principles.
    - Single failure criterion.
    - Exploit passive safety principles.

And that in devising ADS strategies, one must:

- Bear in mind that safety implications on recycle and re-fabrication operations accrue to choices made for the ADS *per se* and must be factored in.

The work of the expert group is ongoing with a target for completion by late spring 2001. This paper provides a status report.

2. Safety functions and strategies for fissioning systems

2.1 *Basic safety functions for fissioning systems*

At a basic level, there are six safety functions to be fulfilled when operating fissioning systems.

1) The nuclear fuel must remain contained within a controlled space because of its radiotoxicity; this is traditionally accomplished by use of multiple containment barriers.

2) Shielding must be kept in place between fissioning and fissioned fuel and humans to avoid suffering radiation damage.

3) A heat-transport path must be in place to carry energy away from the fissioning medium to a heat sink; usually an energy conversion plant.

4) The rate of release of fission energy in the chain reacting medium must be regulated to remain in balance with the rate of energy delivery to the heat sink, so as not to overheat the containment barriers around the fuel and challenge their integrity; a capacity to store heat in the reacting medium and heat transport channel will buffer mismatches of short duration or small amplitude.

5) Since some 5% of the energy liberated from each fission event is initially retained in nuclear bonds of unstable fission products, and since these fission products subsequently decay according to their natural radioactive-decay time constants, a means must be provided for transporting heat from the fission products and transuranics in the fuel for all times subsequent to the fission event. Failure to satisfy the latter two safety functions could lead to overheating of the fuel with the potential to defeat the integrity of the containment and shielding.

6) Operation of the fissioning device in a quasi steady state mode requires a balance of neutron production and destruction rates from one fission chain generation to the next – even as the composition of the chain reacting medium changes due to transmutation and as the absorption, leakage, and neutron production properties of the fissioning medium change with changes in composition and temperatures.

2.2 Safety strategies

Strategies to fulfil the six basic safety functions have been developed and refined over many years for conventional (critical) reactors. The strategy employs defence in depth such that any single failure will not defeat the strategies for meeting safety functions and thereby result in unacceptable release of radiotoxicity; multiple barriers (fuel cladding, primary coolant boundary, and reactor containment building) are used to prevent release of radiation even under accident conditions. Highly reliable (diverse and redundant) systems for controlling and terminating the chain reaction are used to match heat production to removal rate. Highly reliable, redundant/diverse systems for decay heat removal are provided. High quality construction and verification norms minimise manufacturing flaws, and rigorous maintenance, formal procedures, and exhaustive training and certification of operators and maintenance workers are used to minimise the occurrence of human error which could subvert the achievement of the safety functions. Once safety is "designed into" the system, its efficacy is judged by an independent safety regulative authority on a plant-by-plant basis prior to deployment and during its operation.

In recent years, the fast reactor safety design strategy has gone beyond those traditional measures, and the system architecture consisting of the reactor heat source coupled to the balance-of-plant heat engine is configured to achieve the safety functions by exploiting the natural laws of physics to the maximum degree achievable. This *passive safety approach* partially supplants the traditional engineered devices by exploiting passive systems or inherent characteristics that play the role of "functional redundancies" (i.e. they can, in case of failure of the upstream line of defence achieve the same safety related mission); the approach is so implemented to assure safe response[7] – even if the engineered systems which require assured sources of power and highly reliable "active" sensing and switching equipment were to fail, or if multiple, compounding failures and human errors were to occur simultaneously. The passive safety approach can be applied for all the defence in depth levels, i.e. accident prevention, accident management and consequence mitigation. The passive concepts can employ inherent reactivity feedbacks to keep heat production and removal in balance. Designs having minimal reactivity loss upon burn-up and minimal reactivity vested in control rods can preclude reactivity addition accidents. Designs having large margins to damage temperatures and large thermal mass provide reactivity feedbacks with room to operate without reaching damage temperatures or conditions. Designs using buoyancy-driven flows and always-operating heat transport paths to ambient remove decay heat without systematic reliance on switching of valve alignments or active monitoring. These passive safety approaches for fast reactors have been demonstrated [2] in full-scale tests at EBR-II, RAPSODIE, FFTF, BOR-60, etc.

Given that the safety approaches for FRs are well known, the plan for this paper is to first describe the chain of logic which gives rise to the salient differences between FRs and that class of ADS studied by the OECD/NEA expert group. Then a broad survey is made of each salient difference of the ADS design as compared to a FR to identify which of the six basic safety functions might be affected by this particular salient difference. Following that, for each case having an identified difference; potential strategies for fulfilling the function for an ADS are discussed.

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[7] A passive system should be theoretically more reliable than an active one. The reasons are that it does not need any external input or energy to operate and it relies only upon natural physical laws (e.g. gravity, natural convection, conduction, etc.) and/or on inherent characteristics (properties of materials, internally stored energy, etc.) and/or "intelligent" use of the energy that is inherently available in the system (e.g. decay heat, chemical reactions, etc.). Nevertheless passive devices can be subject to specific kinds of failure like, e.g. structural failure, physical degradation, blocking, etc. Generally speaking, the reliability of passive systems depends upon:

- The environment that can interfere with the expected performance.
- The physical phenomena that can deviate from the expectation.
- The single components reliability.

3. Minor actinide and/or transuranic burning ADS design rationale and distinguishing design features

Overall purpose; support ratio, and fertile-free fuel – First, the overall purpose of the class of ADS considered here is to function as one element of an integrated nuclear power enterprise comprised of conventional and advanced power reactors for energy production and ADS for reducing the radiotoxicity of the nuclear waste produced by these power reactors – prior to its entombment in a geologic repository. The radiotoxic materials targeted for incineration may be minor actinides (MA) or may be transuranics (TRU), depending on the configuration of the overall enterprise. The ADS also may incinerate selected fission products (see Figure 1, which illustrates the several power production complexes considered by the expert group). The ATW [3] (US design) is an example of a TRU incinerator such as case 3B; the JAERI double strata ADS [4] is an example of a MA incinerator such as case 4.

The transuranics are fissioned in the ADS to transmute them to fission products with the concomitant release of heat amounting to about 1 gm TRU incinerated per MW_{th} day energy release. *For a fissioning device, the incineration rate of TRU depends on the power rating of the heat removal equipment – be it an ADS or a reactor* and while the ADS plant will likely use the liberated heat for power production to offset the cost of its operation, its primary function is to reduce the transuranic and long-lived fission product inventories emanating from the power reactors deployed in the nuclear energy complex. The "support ratio" of the integrated power producing complex is the ratio of power of the reactors in the enterprise to the power of the ADSs in the enterprise. A large support ratio is targeted for the class of ADS designed for waste incineration with ancillary power production considered here so as to relax the demands on ADS cost and energy conversion ratio – inasmuch as the ADS will then comprise a smaller segment of the overall integrated energy supply complex. The primary purpose of the ADS is to *maximise incineration rate per unit of heat that has to be removed. Fertile-free fuel is the first salient design feature shared by proposed ADS systems of the class considered here* – to avoid *in situ* production and incineration of new transuranics. A 3 000 MW_{th} ADS plant operating for 300 days per year transmutes about 900 kg of TRU into nearly 900 kg of fission products and releases 900 Gigawatt days of thermal energy.

Multiple recycle – The ADS will operate on a closed fuel cycle with a feedstream of transuranics or minor actinides arriving from the power producing reactors and with fission-product-containing (largely actinide-free) waste forms leaving destined for a geologic repository. Internal multiple recycle of the ADS fuel will be required to reconstitute the fuel into fresh cladding because the fluence required for total fission consumption exceeds the neutron damage endurance of any known cladding. Recycle is required also to inject new feedstock into the ADS lattice to sustain the neutron multiplication within its design range as well as to extract the fission products destined for geologic disposal. Although not unique to ADS, a *need for multiple recycle constitutes a second salient feature of the ADS considered here.*

Except for the "once-through cycle" (Case 1 of Figure 1), the recycle step in the overall complex is where the waste stream to the geologic repository is generated. It is comprised of fission products and of TRU or MA trace losses which escape the recycle/refab. processes back to the ADS or FR. These trace losses of TRU or MA to waste are to be minimised if the ADS is to achieve its assigned mission; it is clear that both the trace loss per recycle pass and the number of recycle passes fully control the ADS contribution to the complex's total loss – and that therefore a high average discharge burn-up from the ADS is desirable. Moreover, since the radiotoxicity per gram and also the half life of the various TRU or MA isotopes vary, it is desirable that the transuranic isotopic spectrum achieved upon multiple recycle should be favourable in terms of long-term toxicity (including accounting all post emplacement decay daughters[8]) – the ADS neutron spectrum is controlling in that regard.

Fast neutron spectrum – Upon multiple recycle to achieve total fission incineration, the TRU or the MA isotopic composition of the LWR or FR spent fuel feedstock evolves to an asymptotic ADS recycle feed

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[8] For example for the US (oxidising environment) geologic repository, ^{237}Np (a post emplacement daughter in the $^{241}Pu \rightarrow {}^{241}Am$ decay chain) dominates the long term toxicity.

composition; this composition depends on the neutron spectrum to which it is subjected. (see Table 1) The ADS considered here is designed to operate on fission chains in the fast neutron range so that all transuranic elements stand a good chance to fission upon a single neutron absorption and thereby to minimize the development of an isotopic spectrum which is skewed toward heavier mass transuranic isotopes. Table 2 shows [5] that a fast neutron spectrum (having high fission probability for all TRU) is entirely essential to achieve total consumption in an MA burning ADS and it is preferable to a thermal spectrum for TRU burning. Table 3 indicates [6] that the asymptopic isotopic spectrum from multi-recycle in a fast spectrum will lead to a more favourable long term radiotoxicity burden that does that arising from thermal spectrum burning. *A fast neutron spectrum is the third salient design feature of the class of ADS systems considered here.*

Table 1. **Equilibrium distribution of transuranic isotopic masses for high fluence exposure to thermal and fast neutron spectra**

Isotope	Thermal neutron spectrum	Fast neutron spectrum
^{237}Np	5.51	0.75
^{238}Pu	4.17	0.89
^{239}Pu	23.03	66.75
^{240}Pu	10.49	24.48
^{241}Pu	9.48	2.98
^{242}Pu	3.89	1.86
^{241}Am	0.54	0.97
^{242m}Am	0.02	0.07
^{243}Am	8.11	0.44
^{242}Cm	0.18	0.40
^{243}Cm	0.02	0.03
^{244}Cm	17.85	0.28
^{245}Cm	1.27	0.07
^{246}Cm	11.71	0.03
^{247}Cm	0.75	2.E-3
^{248}Cm	2.77	6.E-4
^{249}Bk	0.05	1.E-5
^{249}Cf	0.03	4.E-5
^{250}Cf	0.03	7.E-6
^{251}Cf	0.02	9.E-7
^{252}Cf	0.08	4.E-8
Total	100.00	100.0

Note: all values are atom % of transuranic inventory built up as a result of extended exposure to a neutron flux. (Calculated as the steady-state solution of the depletion-chain equations independent of criticality considerations.)

Table 2. Values of D_j (neutron consumption per fission) for isotopes j or for a fuel type ($D_j<0$: implies excess neutron self production; $D_j \geq 0$ implies a source of extra neutrons is required)

Isotope (or fuel type)	Neutron spectra and flux level φ (n/cm^2.s)			
	Fast spectrum	Standard PWR		Highly thermalized
	10^{15}	10^{14}	10^{16}	10^{16}
Th (with extraction of ^{238}Pa)	-0.39	-0.24	-0.24	-0.27
Th (without extraction of ^{238}Pa)	-0.38	-0.20	1.22	1.14
^{238}U	-0.62	0.07	0.05	0.1
^{238}Pu	-1.36	0.17	0.042	-0.13
^{239}Pu	-1.46	-0.67	-0.79	-1.07
^{240}Pu	-0.96	0.44	0.085	0.14
^{241}Pu	-1.24	-0.56	-0.91	-0.86
^{242}Pu	-0.44	1.76	1.10	1.12
^{237}Np	-0.59	1.12	0.53	-0.463
^{241}Am	-0.62	1.12	0.076	-0.54
^{243}Am	-0.60	0.82	0.16	0.21
^{244}Cm	-1.39	-0.15	-0.53	-0.48
^{245}Cm	-2.51	-1.48	-1.46	-1.37
D_{TRU} (discharge from a PWR)	-1.17	-0.05	-0.35	-0.54
$D_{TRUPu + Np}$ (discharge from a PWR)	-0.70	1.1	0.3	0.4
D_{TRU} (discharge from a PWR)	-1.1	-0.20	-0.40	-0.53

Choice of coolant – Although not a design feature which distinguishes ADS from fast reactor concepts, the choice of coolant plays a strong role in core design strategy and safety strategy for fast reactor and ADS alike. It is useful for clarifying the discussions to explicitly include coolant choice among the ADS distinguishing features. Since the neutron spectrum is to be fast, the candidate coolants are sodium, heavy liquid metals (e.g. Pb or Pb-Bi) and gas.

Features shared with fast reactors – As indicated in Figure 1, fast reactors are themselves employed for TRU or MA consumption in several of the fuel cycle schemes studied by the expert group and whereas the features discussed above – i.e. fast neutron spectrum, multiple recycle, and alternate coolant choices are shared by those FRs with the ADS, the fast reactors do not employ fertile-free fuel. The neutronic properties of fertile-free fuel *dictated by the a-priori requirement to maximise support ratio* motivate the features of the ADS which most clearly distinguish it from a fast reactor.

Features which are unique to the ADS – Fertile-free fuel is prescribed for the ADS – motivated by the goal to maximise ADS support ratio in the power producing energy complex. The neutronics properties of fertile-free TRU or MA fuel – its η value and its delayed neutron fraction – give rise to the remaining distinguishing ADS features: specifically sub-critical operating state driven by a spallation neutron source.

Table 3. **Radiotoxicity data (CD = Cancer Dose Hazard)**

Isotope	Toxicity factor CD/Ci	Half-life Years	Toxicity factor CD/g
Actinides and their daughters			
^{210}Pb	455.0	22.3	3.48E4
^{223}Ra	15.6	0.03	7.99E5
^{226}Ra	36.3	1.60E3	3.59E1
^{227}Ac	1185.0	21.8	8.58E4
^{229}Th	127.3	7.3E3	2.72E1
^{230}Th	19.1	7.54E4	3.94E-1
^{231}Pa	372.0	3.28E4	1.76E-1
^{234}U	7.59	2.46E5	4.71E-2
^{235}U	7.23	7.04E8	1.56E-5
^{236}U	7.50	2.34E7	4.85E-4
^{238}U	6.97	4.47E9	2.34E-6
^{237}Np	197.2	2.14E6	1.39E-1
^{238}Pu	246.1	87.7	4.22E3
^{239}Pu	267.5	2.41E4	1.66E1
^{240}Pu	267.5	6.56E3	6.08E1
^{242}Pu	267.5	3.75E5	1.65E0
^{241}Am	272.9	433	9.36E2
^{242m}Am	267.5	141	2.80E4
^{243}Am	272.9	7.37E3	5.45E1
^{242}Cm	6.90	0.45	2.29E4
^{243}Cm	196.9	29.1	9.96E3
^{244}Cm	163.0	18.1	1.32E4
^{245}Cm	284.0	8.5E3	4.88E1
^{246}Cm	284.0	4.8E3	8.67E1
Short-lived fission products			
^{90}Sr	16.7	29.1	2.28E3
^{0}Y	0.60	7.3E-3	3.26E5
^{137}Cs	5.77	30.2	4.99E2
Long-lived fission products			
^{99}Tc	0.17	2.13E5	2.28E-3
^{129}I	64.8	1.57E7	1.15E-2
^{93}Zr	0.095	1.5E6	2.44E-4
^{135}Cs	0.84	2.3E6	9.68E-4
^{14}C	0.20	5.73E3	8.92E-1
^{59}Ni	0.08	7.6E4	6.38E-3
^{63}Ni	0.03	100	1.70E0
^{126}Sn	1.70	1.0E5	4.83E-2

Table 4. **Delayed neutron fraction**

Isotope	γ_d/γ_{total}
^{238}U	0.0151
^{232}Th	0.0209
^{235}U	0.00673
^{239}Pu	0.00187
^{241}Pu	0.00462
^{242}Pu	0.00573
^{237}Np	0.00334
^{241}Am	0.00114
^{243}Am	0.00198
^{242}Cm	0.00033

$\Rightarrow$ 10% Fertile fission raises β in fertile containing fast reactor fuel

$$\begin{array}{cc} \beta(^{238}U) & \beta(^{239}Pu) \\ 0.10 \times 0.0151 \quad + & 0.90 \times 0.00187 \\ = 0.00151 \quad + & 0.00168 \end{array}$$

$$= 0.00319$$

(Nearly doubles β vis-à-vis fertile-free fuel)

Sub-delayed critical operating state – Transuranic fuel containing no fertile (^{238}U and ^{232}Th) atoms exhibits a delayed neutron fraction for fast fission which is in the range of 0.0015 to 0.0020 i.e., about half the value for a conventional fast reactor and about a sixth the value for a conventional LWR. Table 4 displays β for fast fission of various actinide isotopes and shows that even at only 10% contribution to fissions, as is typical for a fast reactor, fertile ^{238}U or ^{232}Th contribute very significantly to delayed neutron fraction – doubling its value from what applies for fertile-free fuel. For fertile-free TRU or MA fuel compositions the delayed neutron fraction is remarkably small and therefore the margin to prompt critical is correspondingly small. This feature, when combined with reactivity feedback considerations discussed next, leads to further salient design features of ADS *specifically as a safety strategy approach.*

The neutron leakage in a fast neutron lattice is sensitive to the assembly geometry because of the long neutron mean free path. Subtle thermo-structural-induced geometry changes which are dependent on power to flow ratio (P/F) – such as fuel bowing, grid plate expansion, etc. – change the neutron leakage fraction in response to power and flow changes. For example Figure 2 illustrates for the FFTF sodium cooled fast reactor the normalised power to flow ratio dependence of the radial expansion plus bowing component of thermostructural reactivity feedback. Several features are notable [10]; first the amplitude is nontrivial with respect to β over the range $0 < P/F < 1$; clamping and duct wall tolerances and stiffness are designed [7] so as to assure negative bowing reactivity feedback at P/F in the vicinity of the operating point, P/F $\simeq$ 1; and reactivity increases with decreasing P/F may become indeterminant [7] at low values of P/F owing to the "unlocking" of above core load pad structural contact. Numerous other leakage dependant thermostructural reactivity feedbacks (grid plate expansion, fuel axial expansion, etc.) are also individually nontrivial in amplitude compared to β, as illustrated in Figure 3 for a power change transient in the modular PRISM reactor [8].

ADS designs using fertile-free fuel have high values of k_∞ and correspondingly high neutron leakage fractions [9]. With a reduced delayed neutron fraction of 0.002 or less and even assuming an unrealistically small neutron leakage fraction of only 5%, a change in leakage fraction of only a few per cent of its value – induced by thermostructural feedbacks – would exceed the .002 $\Delta k/k$ offset from prompt critical. Not only is it impossible to design for and to control thermostructural response to that degree of precision [10], but variability as well as controllability is the issue here. In an ADS functioning as a waste burner, the fuel composition itself and its η value and β value can be expected

to vary from loading to loading as LWR spent fuel and/or FR spent fuel of differing burn-up, differing cooling times and differing origins supply the ADS fuel feedstock. These feedstock variabilities change not only k_∞ and the concomitant leakage fraction and resulting amplitudes of thermostructural feedbacks, but they also change the delayed neutron fraction and offset from prompt criticality itself. Consider the effect on core k_∞ of even small variability in $^{239}Pu/^{241}Pu/^{241}Am$ ratios in ADS feedstock[9] as indicated by their vastly differing η values illustrated in Figure 4. Or, referring to Table 4, consider the effect on β of the transformation of ^{241}Pu (14.35 year half life) to ^{241}Am over different cooling periods prior to introduction into the ADS – a factor of four change in β contribution.

Figure 2. Reactivity from radial core expansion as a function of normalised power-to-flow ratio, comparing the FFTF correlation and the SASSYS/SAS4A calculation
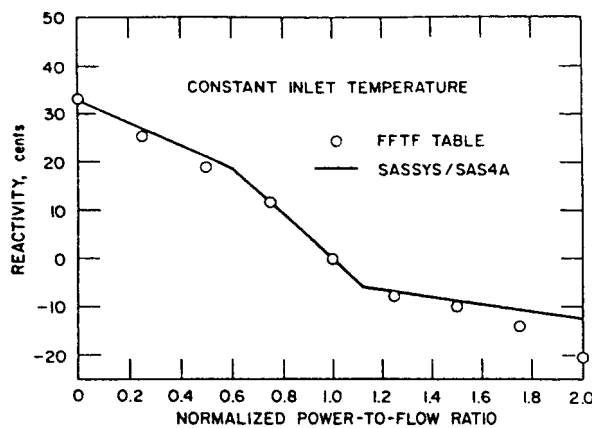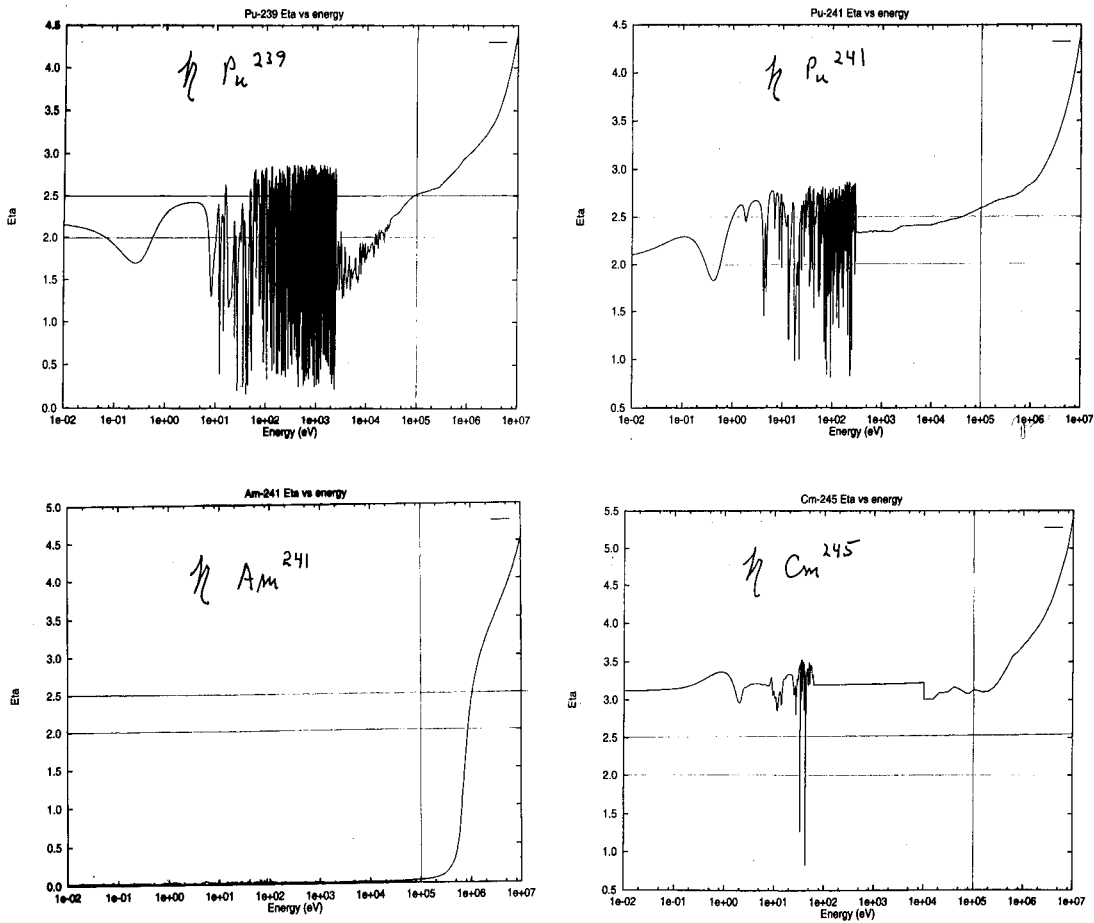

$(\beta = 0.003)$

Figure 3. Unprotected transient overpower for ALMR ($\beta = 0.003$)

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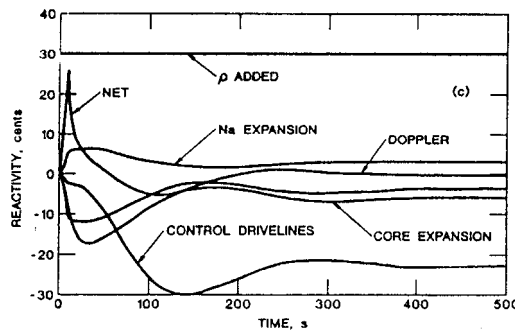
[9] Local power peaking in reload assemblies also are affected by these variabilities.

Figure 4. η (Neutrons released per neutron absorbed) for several isotopes

Taken all together, the variability and non controllability of the reactivity state of a fertile-free MA or TRU lattice relative to the reduced offset from delayed critical to prompt critical requires a safety strategy to assure that no potential can exist for power/flow induced reactivity feedback to carry the lattice into the super prompt critical range – even accounting for the variability in β, k_∞, and leakage fraction which result from feedstock of varying composition. The ADS strategy is one approach to this need – an increased offset of operating state from prompt critical is achieved by operating sub delayed critical. So as to avoid any potential for power/flow induced reactivity feedbacks to inadvertently carry the system into the super prompt critical regime, the geometry and composition of the ADS lattice is configured so that the operating margin to prompt critical will always substantially exceed the maximum power/flow reactivity feedback – while accounting for expected variability in the values of η and β owing to differing feedstock compositions. But the resulting offset then exceeds the value of the delayed neutron fraction itself – so it makes the operating point of the ADS lattice sub-delayed critical. An external source is required, therefore, to drive a continuing fission reaction. The fissioning system multiplies the externally supplied neutron source. *A sub-delayed critical operating state driven by an external neutron source is the fourth and defining design feature of all ADS.*

Spallation neutron source – At 1 gm of TRU or MA incinerated per $MW_{thermal}$ day, ADS facility heat ratings must lie in the range of 1 000 MW_{th} or more to support any reasonably – sized energy complex. The size of the neutron source required to drive a sub-delayed-critical ADS depends on both

106

the desired heat rating and on the degree of neutron self multiplication of the lattice, which depends on the degree of sub-criticality (see Equation 1, below). With the required offset from prompt critical no less than 2 or 3% $\Delta k/k$ (i.e. a source neutron fission chain multiplication no greater than 30 to 50), it is clear that no passive neutron-emitting source is strong enough to meet the requirement for $\sim 1\,000$ MW$_{th}$ power rating. However, plausible extensions in proton beam current capability which are now achieved in linear accelerators (i.e. beams of multi megawatt levels), could achieve required neutron source strength by driving a heavy metal spallation target. *This leads to the fifth salient design feature of an ADS; namely the external source must derive from a spallation neutron target driven by a high power proton accelerator.*

3.1 Summary of distinguishing features for TRU or MA burning ADS

The distinguishing features of the type of ADS considered by the expert group derive directly from the mission assigned to it in the energy complex: namely – *TRU or MA (and LLFP) incineration for waste management in the integrated energy complex with power generation only secondary to partially offset cost,* – combined with the *a-priori assumptions* on scope of cases considered by the expert group: namely *fast spectrum*, s*olid fuel*, *and maximised support ratio.*

The resulting distinguishing features are:

Those shared with FR:

- Fast neutron spectrum.

- Solid fuel lattice.

- Multiple recycle.

- Choice of coolant: Na, Pb-Bi, gas.

And those unique to ADS:

- Fertile free fuel.

- Sub-critical operating state.

- Spallation neutron source driven by a high power proton beam.

4. Safety-related challenges arising specifically from ADS design features

The salient design features of ADS give rise, in some cases, to different safety-related challenges and different approaches to fulfilling the six cardinal safety functions for fissioning systems as compared with the more familiar issues and safety strategy which apply for a fast reactor. Table 5, which tabulates salient design feature (matrix rows) versus required safety function (matrix columns), identifies where these differences exist. In Table 5 the effect of salient design features on strategy for meeting safety functions is indicated for both *normal operational safety* and for *off-normal safety* situations. The Table 5 impact matrix is overviewed here and the safety strategy options to accommodate the new issues are briefly discussed.

Table 5. **ADS Distinguishing feature vs. basic safety function impact matrix**

Distinguishing features	Neutron balance		Heat removal		Regulation of power/flow		Containment		Shielding		Decay heat removal	
	Normal	Off-normal	Normal	Off-normal	Normal	Off-normal	Normal	Off-normal	Normal	Off-normal	Normal	Off-normal
Fast spectrum	▨											
Choice of coolant			▨	▨		▨			▨			▨
Fertile-free fuel	▨				▨	▨						
Sub-critical state	▨				▨	▨						
Spallation source	▨		▨			▨		▨	▨	▨		▨
Multiple recycle		▨							▨		▨	

▨ Legend: new issues arise vis-à-vis a fast reactor.

Proton beam transport tube and spallation neutron source effects – The most readily obvious geometrical difference occurs via the introduction of the proton beam tube. First is its topological effect on the multiple containment barrier defence in depth containment safety strategy. In fast reactors, the fuel is contained first by its cladding (or by multiple layer ceramic barriers in particle fuel), then by the primary cooling circuit boundary and lastly by the containment building. For the ADS based on linacs, the proton beam tube penetrates the last of these and employs a metallic proton beam window as a topological continuation of the primary coolant boundary. The safety issue pertains to the preservation of defence in depth for the containment and shielding functions. In a fast reactor similar topologies result from steam lines which penetrate the containment, and from IHX tubes which comprise a topological extension of the primary coolant boundary. In BWRs, the steam lines penetrate both the containment and the reactor vessel. Fast acting values at the containment boundary of steam pipes and robust heat exchanger tube walls are the safety strategies used for these reactors. Fast acting valve safety strategies employed for reactors will be considered for the ADS. For the ADS, the beam window operates in an especially hostile environment in light of its temperature and the proton and neutron bombardment that it experiences; the hazard deriving from the multi-megawatt proton beam potentially impinging on any of these barrier boundaries or valves is unique in an ADS.

The beam tube introduces new issues as well in the area of shielding safety function – comprising a streaming path from the fissioning lattice to the exterior of the vessel as well as an unshieldable pathway for radiation activation of the bending magnets or accelerating structures of the accelerator. Finally, the several tens of centimetre diameter evacuated beam tube presents a new issue in the area of a potential positive reactivity effect should the beam tube flood and decrease the neutron leakage; the degree of reactivity offset from prompt critical must be sufficient to safely accommodate such potential flooding.

The presence of an strong spallation neutron source has an effect on power density peaking factor [11] and on the change in power peaking as k_∞ of the lattice changes with burn-up and as the ratio of source to fission multiplied neutrons is altered by changes in source strength or reactivity. Also, depending on beam tube entry geometry, the fuel-loading pattern may be non-azimuthally symmetric – again affecting power density profile. Tailored spatial k_∞ zoning can be applied and, a design strategy, which relies on, increased margins so as to accommodate local shifts in power/flow ratio – while undesirable for a dedicated power producer, is quite consistent with the ADS mission where power production is an ancillary function only.

The ADS power output is proportional to spallation source strength and sub-critical reactivity offset via the relationship:

$$P \, \alpha \, \frac{S}{\frac{1}{k}-1} = \frac{S}{-\rho} \tag{1}$$

$$where \; \rho = \frac{k-1}{k}$$

Asymptotic adjustment of power (or power density) scale to first order with source changes or reactivity changes as:

$$\frac{\delta P}{P_0} = \frac{\delta S}{S_0} - \frac{\delta P}{\rho_0} \tag{2}$$

While a favourable ADS safety feature derives from its asymptotic rather than rising period response to a positive reactivity insertion [12], a safety challenge still remains in assuring that positive source strength or source importance changes cannot take the ADS to damaging overpower conditions. Equation 2 indicates that e.g. at an beginning of cycle offset of $-\rho_o$ equals 3% $\Delta k/k$ and a burn-up reactivity loss of 6% $\Delta k/k$, the source for maintaining end of cycle power level would have to exceed beginning of cycle requirement by 100%, leading to a factor of two overpower potential should the full source strength be introduced prematurely. Options to minimise burn-up reactivity loss include multi-batch fuel loading [9] and optimal mixes of plutonium and minor actinides [13] to flatten the reactivity change with burn-up. However, given fertile-free fuel, it has proven impossible for ADS designers to achieve small burn-up reactivity loss; so that compensation by either external reactivity changes (control rods) or by source strength or importance changes is unavoidable. In every case then, an overpower potential exists.

At the other extreme, if ADS heat removal equipment were to fail (loss of flow or loss of heat sink), then the beam would be required to trip off within seconds to avoid overheating and melting of the fuel [14].

Such considerations of controlling ADS on the basis that the beam current will assume the functions assigned to control rod in fast reactors might lead to a "nuclear safety grade" designation for the accelerator equipment and its maintenance, or at least for its controller – having significant unfavourable cost implications. Alternately, the proton beam might be operated at 100% strength at all times with a safety grade scram circuit, while burn-up reactively loss could be compensated by (safety grade) control rod actuators. Using the same example as above, a control rod bank worth of 6% $\Delta k/k$ would accomplish the same burn-up reactivity compensation as a factor of two larger proton accelerator – with a significant favourable cost advantage likely. Or, mechanical adjustments of neutron source importance via changes in source location or spectral importance may be options. Even adjustable volume fraction mixes of various spallation target materials having differing neutron yield per proton might be considered. In all cases a safety hazard exists in potential for premature actuation of the excess source or reactivity prior to burnout of the lattice; it simply cannot be avoided, short of letting the power rating decrease with burn-up.

Coolant choice effects – The distinguishing characteristic of the coolant choices relate to system pressure, lattice power density, effect on neutron spectrum, and chemical activities – as tabulated in Table 6.

Table 6. **Coolant characteristic features**

	Na	**Pb-Bi**	**He**
System pressure	Low	Low	High
Lattice power density	High	Low	Lower
Neutron spectrum	Hard	Harder	Harder
Chemical activity	High	Low	Null

These distinguishing features permeate the entire design approach for ADS and fast reactor alike and influence the resulting safety strategies. High pressure gas cooling introduces a loss of coolant vulnerability but eliminates chemical compatibility issues. Gas cooling shares with Pb-Bi cooling the need for a low power density, open fuel pin lattice – which leads to potential for significant reactivity additions upon hypothetical pin disruption or compaction but reduces potential for blockage from foreign objects.

Freezing temperatures and coolant/structural/fuel chemical interactions and potential for "local fault propagation" into flow blockages are important safety relevant issues for liquid metal coolant choice, and given that every fertile-free fuel under consideration for ADS use lacks a data base of inservice experience, this issue will require a substantial R&D effort in every case.

The coolant voiding reactivity coefficient is of reduced safety relevance because the ADS provides an added degree of freedom in the sub-critical offset from prompt critical sufficient to cover voiding worths [15].

The high density of Pb alloy coolant introduces several new issues for ADS and fast reactor alike; first is the structural support and the seismic structural response of large reactor vessels when filled with dense lead alloy. Second is the design of refuelling equipment and fuel assembly hold-down devices for the case where the fuel and the structures are less dense than the coolant and tend to float in it. Its high boiling point, on the other hand, provides more than sufficient margin to boiling.

A significant safety-relevant issue for fast reactors and ADS also is the consequence of failing to maintain leak tightness of the primary coolant system. Rank ordering of coolant favours liquids over gas for this issue because only gas operates at above-ambient pressure. However, each coolant displays a vulnerability which is unique to itself. Since gas-cooled systems operate at high pressure, a loss of integrity *anywhere* in the gas heat transport circuit could lead to a loss of coolant accident. Loss of coolant accidents are of extremely low probability for liquid metal cooled systems using a pool layout but each liquid metal displays a safety vulnerability upon leakage of primary coolant. Sodium burns in air, creating an aerosol containing (24-hr γ-emitting) radioactive ^{24}Na. Pb-Bi alloy does not burn but none-the-less releases 138-day (α-emitting) ^{210}Po. Safety approaches have been developed in the fast reactor communities to mitigate and recover from Na and Pb-Bi leakage events and, as compared with a gas leakage loss of coolant vulnerability in a gas cooled fast reactor, the liquid coolant mitigation technologies are at a more mature state of development. However, in-service inspection and repair are a serious vulnerability for opaque liquid metal cooling as compared with gas cooling.

For fast spectrum ADS applications, safety-related issues upon loss of primary boundary integrity should be evaluated first at the particular point of vulnerability innate to ADS: the single thin-wall boundary between the transmuter coolant and the vacuum extension of the proton beam tube leading into the spallation target located at the centre of the core. The window operates in a hostile environment of proton and neutron damage and it alone lies between the centre of the fissioning lattice and the proton accelerating structures external to the containment building. Beam tube melt-through upon a beam misalignment similarly presents a loss of containment boundary vulnerability.

Fertile-free fuel effects – Fertile-free fuel has a high value of η (see Figure 4) and requires a design strategy for safely disposing of excess neutrons. The options are leakage or internal parasitic capture – either discrete absorbers or absorbers homogeneously mixed with the fuel. Thermostructural reactivity feedback variations can be reduced the smaller is the leakage fraction and this is desirable for reasons discussed above. Recycle/re-fabrication batch sizes may benefit from the homogeneous absorber option. On the other hand, radial k_∞ zoning using only a single fuel pin fabrication specification may be achievable using discrete absorber pins.

As already discussed above, the absence of internal conversion of fertile to fissile species with burn-up will place demands for burn-up reactivity compensation on other design approaches – such as source strength or source importance changes, batch refuelling, or absorber control rod changes. For minor actinide burners, in situ isotopic transmutations mitigate but do not eliminate this issue.

Optimised mixes of MA and Pu can be tailored [13] to limit burn-up swing; but in every case source or reactivity compensation strategies are needed.

An off-normal safety related challenge derives from fertile-free fuel – which excludes the traditional Doppler contributor to prompt negative reactivity power feedback in a FR. Small (but not zero) Doppler feedback has been accommodated (and beneficially exploited for a passive safety mechanism) in metal-fuelled fast reactors. However, an HCDA termination mechanism will have to be devised for an ADS having fertile free fuel [16], high melting point oxide-fuelled FRs traditionally rely heavily on prompt Doppler feedback to limit the pre-disassembly energy generation which controls severity of HCDAs. Inertial resistance to disassembly in an HCDA sequence is an additional issue with Pb-Bi cooling.

Pure TRU or MA fuel presents issues in recycle batch sizes and processing geometries because of a small critical mass. Experience exists with metal-fuel/pyro recycle where discrete rather than continuous processing is employed and batch size is limited by relatively larger fast criticality constraints; this issue would require special care in the case of continuous aqueous reprocessing having very small critical masses.

Sub-critical operating state and dynamics effects – A fundamental distinction between ADS and critical reactor safety-relevant control arises because of the dramatic differences in dynamic response of critical versus sub-critical source-driven lattices. In a source-driven system, a change in source strength or in source importance or a change in reactivity will cause the neutron population and power level to promptly[10] adjust to a new asymptotic level in accordance with Equation 2; whereas in a critical reactor a change in reactivity leads (absent reactivity feedbacks) to an asymptotic period (or exponential time change) of neutron population, the promptness of which is controllable by the reactivity insertion magnitude. In a critical reactor, the period of power adjustment is chosen to match the thermal and structural time constants – which are in the range of 0.1 to 100 seconds (see Figure 3).

The dynamics and control challenges can be illustrated under the excellent assumption that the neutron population, n(t) is in prompt quasi-static equilibrium with the source. For a reactor it is the delayed neutron source; for the ADS it is the external spallation source plus the delayed neutron source:

$$\frac{dn}{dt} = 0 = \frac{\rho - \beta}{\wedge} n + \lambda C + S$$

$$n(t) = \frac{\wedge}{\beta - \rho(t)} \left[\lambda C(t) + S(t) \right]; \quad [units] = \left[\frac{neutrons}{cm^3} * Vol \ of \ Core \right]$$

(3)

where:

$\wedge$	=	prompt neutron generation time ~ 10^{-7} [sec]
$1/\lambda$	=	delayed neutron precursor lifetime ~ 10 [sec]
β	=	delayed neutron fraction ~ .002
$\beta - \rho(t)$	=	$\beta - \rho o - \Delta\rho(t)$
$\beta - \rho_o$	=	reactivity offset from <u>prompt</u> critical $\left[\frac{\Delta k}{k} \right]$
$\Delta\rho(t)$	=	feedback + external control reactivity

---

[10] The adjustment will occur within 30 to 50 prompt neutron generation times for sub-criticality of 2 to 3% $\Delta k/k$. Given a generation time of ~10^{-7} sec, *prompt* means several microseconds adjustment times for an ADS.

The prompt neutron population establishes equilibrium immediately ($<10^{-6}$ sec) to any:

- external source change, $S(t)$
- delayed neutron precursor source change, $\lambda\,C\,(t)$
- reactivity change, $\rho(t) = -\rho_o + \Delta\rho(t)$

The relative nimbleness of the two sources which drive the neutron population is very different: $S(t)$ is fast and can change by 100% in 10^{-7} sec while the delayed neutron source, $\lambda\,C\,(t)$ is sluggish with a time constant for one Θ – folding factor of $1/\lambda \sim 10$ sec. Moreover, the delayed source has a memory of previous history of $n(t)$:

$$
\frac{d}{dt}C(t) = \frac{\beta\,n(t)}{\wedge} - \lambda\,C(t)
$$
$$
C(t) = \int_{-\infty}^{t} e^{-\lambda(t-\tau)}\,\frac{\beta}{\wedge}\,n(\tau)d\tau
$$
(4)

Finally, whereas for a critical reactor the delayed neutron source is the only source present, for the ADS the delayed source is but a very small fraction of the total source and it depends on sub-criticality level:

From Equations 3 and 4:

$$
\frac{S_o}{n_o} = \frac{-\rho_0}{\wedge}; \quad \frac{\lambda C_o}{n_o} = \frac{\beta}{\wedge}
$$
(5)

so, for the ADS:

$$
\frac{delayed\ source}{total\ source} = \frac{\lambda C_0}{\lambda C_0 + S_0} = \frac{\beta}{\beta - \rho_0} = \frac{\left(\begin{array}{c}delayed\ critical\ offset\\ from\ prompt\ critical\end{array}\right)}{\left(\begin{array}{c}total\ offset\ from\\ prompt\ critical\end{array}\right)}
$$
(6)

for example, given a 3%$\Delta k/k$ sub - critical state and $\beta = .0015, then = \dfrac{1}{21}$

Because it is only a small faction of the total source, and the neutron population adjusts promptly to changes in total source, the delayed source cannot be counted on in the ADS to slow down dynamic response of the neutron population (and concomitant power density) even though such a showdown is highly desirable because the time constants of heat removal, relaxation of thermal stresses, and relaxation of reactivity feedbacks all lie in the range of 0.1 to 100 seconds (see Figure 3). Since delayed neutrons won't buffer the differences between the prompt neutron power adjustment time and the slow thermostructural relaxation times, new control challenges arise for the ADS; specifically *the controller and actuator must themselves perform this moderation function* so that the control actuator (whether on source strength, source importance, or reactivity) must achieve:

- Very slow adjustments.
- Very precise changes.
- Be very reliable.

Moreover, the fuel is where neutron and heat removal time constants clash continually; giving rise to new requirements on the fuel also, specifically *it must be structurally tough to thermal shocks, and must have heat storage capacity to slow down heat release transients.*

Controller options include traditional control rod actuators as well as actuators controlling source strength or source importance (either spatial or spectral dependencies). As in the case of burn-up reactivity swing compensation, the control actuator will likely be required to assume a "nuclear safety grade" level of reliability.

In summary, the dependence on spallation source neutrons rather than on delayed neutrons to maintain the fission chain reaction in balance from one fission chain generation to the next leads to extremely abrupt response to control actions, reduced influence of power/flow dependent reactivity feedbacks, and puts added importance on the fuel and on the control actuator itself to moderate the vastly different time constants of nuclear and heat removal processes.

Beam reliability effects – Current multi megawatt proton beam accelerators have not been designed for second-to-second reliability; they trip off many times a week due to accelerator cavity sparking; they restart after a spectrum of time delays ranging from a fraction of a second to tens of minutes [17]. Since ADS power scales linearly with the source strength (Equation 2) such source trips lead to ADS power trips which in turn induce abrupt fuel and coolant temperature transients. Such temperature transients induce thermal stresses in the core support and heat transport heavy-walled structures; repeated trips give rise to life-shortening low cycle fatigue damage of these structures [18]. Moreover, if the restart delay exceeds several minutes, the balance of plant must undergo a major restart procedure to connect to the grid [4].

Accelerator designers are devising means to reduce the frequency of trips – but do not foresee means to reduce frequency to only a few per year (similar to frequency of unplanned reactor trips). Thus, design options to mitigate their effects on the transmuter core and the heat transport circuits have been studied. Options include multiple accelerators to avoid total loss of power given any single accelerator trip. Other options include power density de-rating to lessen the amplitude of temperatures swings. Thermal storage – in the fuel [18], in the coolant, and in the steam generator [4] – are also considered so as to mitigate the abruptness of downstream temperature swings.

Application of passive safety principles – For liquid metal cooled ADS, the passive decay heat removal strategies used for fast reactors apply without modification.

Passive power self-regulation based on thermostructural reactivity feedbacks which has been exploited for fast reactor passive safety [19] is precluded by the fundamental feature of ADS sub-critical source-driven systems. For an ADS, the operating point is offset from prompt criticality by $(\beta - \rho_o)$ where $-\rho_o$ is the sub-criticality operating point. This is compared to an offset of only β for a critical reactor. As is evident from observation of the denominator in Equation 3, the effect is to decrease sensitivity of ADS power level to reactivity feedbacks as compared to a reactor. Moreover, as is also evident from the inhomogeneous source term in Equations 3 and 4, the *power can never be driven to zero by reactivity changes as long as the spallation source is nonzero*. These differences give rise to a need in the ADS for different strategies for employing passive concepts to keep heat production and removal in balance. Specifically, some means for source strength or source importance to be adjusted passively in response to power changes is needed. Options include accelerator powered by ADS-generated electricity[11] [20]; or source – transmuter coupling which is dependent on coolant temperature or density. Absorber curtains or moderator curtains in the buffer surrounding the source or target spatial relocations (all activated by coolant temperature or density changes) affect coupling and might offer opportunities to apply passive *source* feedbacks analogous to the passive *reactivity*

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[11] The exceedingly long time constant for feedback presents a major challenge with this option.

feedbacks successfully exploited and demonstrated in fast reactors as the means to passively self regulate the heat production rate to match the heat removal rate.

Activation product effects – Given the ADS function to reduce waste losses to the repository from the fission energy complex per unit of useful energy from the complex, care must be taken in ADS design to minimise production of incremental waste. This is a safety-related issue not only for the long term, but for operational safety as well. Issues arise in choice of coolant [21], choice of spallation target material [22] and level of sub-criticality operating state and in beam misalignment and halo effects on activation of accelerator structures [23].

Recycle facility safety – The recycle and re-fabrication processes for TRU and MA fuel introduce safety issues of criticality, pyrophoricity and atmosphere control; these do not differ in character from the recycle safety issues for reactors intended for TRU or MA use. In either case, however, small critical mass of fertile-free fuel and the demands on shielding and atmosphere control when working with high concentrations of minor actinides (displaying characteristics of spontaneous fission, neutron emission, and low temperature volatility) raise new challenges compared to current practice with UOX or MOX fuel.

Accelerator safety – The accelerator brings with it the usual accelerator safety issues (high-voltage safety, control of worker dose owing to components activated by beam divergence, etc.). These issues are not peculiar to ADS applications except for shielding issues at the ADS/accelerator beam tube interface, already discussed.

5. Summary

The work of the expert group in studying the safety issues of a specific class of ADS – that employing fast neutron spectrum, solid fertile-free fuel, and multiple recycle – with a primary mission of TRU or MA incineration has comprised an ancillary element of the OECD/NEA "Comparative Study of Fast Reactors and ADS in Advanced Fuel Cycles". Safety-related challenges which derive from the distinguishing design features of the ADS for this specific mission have been identified. The expert group has discussed safety strategy options available for addressing each safety-relevant issue based on a presumption that safety should be designed in from the start; that relevant fast reactor safety principles and practices should be applied where applicable and that safety of the entire cycle (including recycle, refab, and waste disposal operations) should be kept in mind during each ADS safety-related design decision.

While the work is still ongoing, multiple options for addressing nearly all issues have been developed, drawing on experience from reactor safety approaches. An impact matrix (of ADS distinguishing design feature vs. required safety function) has been developed, and a tracking of distinguishing design feature back to specified ADS mission element has been produced. These materials will be useful to designers and safety analysts in optimising the design of their ADS concepts within their specific set of constraints and mission requirements.

Several issues merit special note. First, the issue of ADS dynamic response to reactivity or source changes and the achievement of buffering between nuclear and thermo/structural time constants without benefit of delayed neutron buffering is the area of greatest difference between fast reactor and ADS safety-related characteristics and an area where no precedents exist in the fast reactor experience base. Second, a prompt negative feedback mechanism for quenching HCDA sequences will have to be developed for the ADS which will rely on phenomenology other than the traditional Doppler coefficient of reactivity of fertile atoms in the fuel – or else the maximum support ratio requirement which dictated fertile-free fuel could be relaxed. Finally, opportunities for application of passive safety principles can be foreseen and should be exploited; straightforward adoptions are available for passive decay heat removal; innovation will be required to achieve passive power self-regulation.

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POSTER SESSION

Partitioning

Chair: M.J. Hudson (University of Reading)

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SUMMARY

Amongst the developments within the poster presentations there was a high degree of novelty and innovation. In all, there were twelve interesting poster presentations – the list for which is given elsewhere. All of the presentations were of a high standard and the authors are to be congratulated for the hard work that they put into the presentations. The paper by Suarez *et al.,* for example, indicated that selenium and zirconium isotopes remained in the raffinate within the PUREX process. Caravaca *et al.* showed that within electrochemical processes in pyrochemical systems, when LiCl/KCl is used as the electrolyte, the nucleation and crystal growth of the rare earth metal seems to be the controlling step for deposition. Following the success of recent European Projects, which studied amides, Almaraz *et al.* have managed to bind malondiamides onto calix[6]arenes which may have potential as solvent extraction reagents. Using hollow fibre techniques, Geist *et al.* showed that over ninety per cent of americium might be extracted from the feed phase when nPr-BTP is used as an extractant. The kinetics of the extraction seemed to be rather slow. Dicarbolide studies are also continuing and Plesek *et al.* showed that COSAN might be used for the separation of isotopes of strontium and actinides. The influence of intermediate chemical processing of nuclear fuel has been studied by Gerasimov *et al.* The extent of burn up increased with the amount of enrichment. Song *et al.* discussed the developments that have been taking place at Tsinghua University. They produced a flow sheet for the total partition process for commercial HLLW, which was the focus of much attention.

The paper by Gruettner *et al.* indicated that nanotechnology should be considered more seriously for the selective complexation of radionuclides. Especial importance must be directed in the future the great potential that nanoparticles and nanostructured materials may have in the future. Thus the particles themselves, or the functional groups on the surface may be used to interact with radionuclides.

POSTER SESSION

Basic Physics: Nuclear Data and Experiments
and
Materials, Fuels and Targets

Chair: P. D'Hondt (SCK•CEN)

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SUMMARY

As mentioned during the poster session introduction, research on ADS has incited a revival of interest in nuclear cross-sections of many nuclides in a large energy range with a special interest for the higher energies.

In this poster session, 12 papers were foreseen from which 11 were presented, a success in itself. I will go through each of them following the order of the programme to give you a flavour of what was presented.

The first paper was on the n-TOF experiment at CERN and was concerned with the actual design of the installation, with special emphasis on those aspects particular to the n-TOF: namely the excellent energy resolution and the high-energy spectrum of the neutrons. Worthwhile to mention is that the neutron energy spectrum induced by 20 GeV protons on a lead target, after a 200 m flight path only contain 7% of the neutrons with energies higher than 20 MeV.

The second poster reported on recent capture cross-sections validation on ^{232}Th from 0.1 eV to 40 keV. It was shown that it is possible to determine cross-sections with a precision of 5% by making use of a slowing-down spectrometer associated to a pulsed neutron source. These measurements performed at ISN (Grenoble) with GENEPI show good agreement with ENDF/B-VI and JEF-2.2 in the energy range 10 eV to 40 keV. Discrepancies were observed with JENDL 3.2 in the energy range from 300 eV to 3 keV.

The third poster reported on double differential cross-sections for protons and light charged particles emitted in reactions of 100 MeV neutrons on enriched ^{208}Pb targets. The measurements were performed at Upsalla, Sweden. Preliminary results were presented for different angles relative to the beam.

The fourth paper reported on measured double differential cross-sections of neutrons produced in reactions induced by a proton beam of 62.5 MeV on a lead target. This experiment was performed on the S-line of the CYCLONE facility of Louvain-la-Neuve in Belgium. Results were shown for 5 different angles.

The fifth paper reported on a joint work between Russian and Swedish colleagues. This work focused on neutron-induced fission cross-sections of tantalum, tungsten, lead, mercury, gold and bismuth. Results were presented for 10 different neutron energies ranging from the fission threshold up to 175 MeV.

Paper 6 was entitled: "Neutron Radiative Capture Cross-sections of ^{232}Th in the Energy Range from 60 keV to 2 MeV". This paper reported on the work performed at the 4 MW Van der Graaf of the CEN-Bordeaux. The activation technique was used and the cross-section was measured relative to the ^{197}Au(n,γ) standard cross-section up to 1 MeV. The results indicate that the cross-sections are close to the JENDL database for values up to 800 keV and over 1.4 MeV. For energies in the intermediate range, values are slightly lower to the ones from the libraries.

The seventh paper was related to the determination of the neutron fission cross-section for ^{233}Pa from 0.5 to 10 MeV using the transfer reaction technique. This common work of CEN-Bordeaux, CEN-Saclay and ISN-Grenoble is a first attempt to determine the neutron induced fission cross-section of ^{233}Pa in the fast neutron energy range as a product of the fission probability of ^{234}Pa and the same compound nucleus formation cross-section. Although the results are preliminary, they tend to agree with the JENDL evaluation at least for energies greater than 4 MeV.

The experiment described in the eighth paper has been performed at the CYCLONE installation of Louvain-la-Neuve. Double differential cross-sections for light charged particles production in neutron induced reactions at 62.7 MeV on lead target were presented. Special attention was devoted to the correction procedures coming from the use of a thick target and collimators. Measurements were done with good statistics and are in good agreement with other experimental data. The comparison with some well-known theoretical total production cross-sections data still shows large discrepancies. Need for improvements of the theoretical models are still necessary.

The HINDAS project, which was presented as paper 9, will provide similar data for Fe and U. The general objective of HINDAS, accepted within the 5th framework programme, is to obtain a complete understanding and modelling of nuclear reactions in the 20-200 MeV region, in order to build reliable and validated computational tools for the detailed design of the spallation module of an ADS.

The tenth paper was also concerned with an accepted project within the 5th framework programme, namely: the CONFIRM programme. This project aims at investigating the feasibility of a high burn-up, high linear rating uranium free fuel, by means of modelling, fabricating and irradiating transuranium nitride fuels. Some preliminary results from the safety analysis, pellet/pin design and data requirements for irradiation modelling were presented.

Finally, the eleventh paper was concerned with one of the major problems in ADS namely the large burn-up reactivity swing and the consequent unfavourable slanting of the radial power distribution over the depletion period. In this work two concepts of the burnable absorber application were considered, homogeneous and heterogeneous loading of B$_4$C. The homogeneous application of the B$_4$C burnable absorber can be effectively used in reducing the burn-up reactivity swing but is not favourable in terms of source neutron multiplication. Loading of burnable absorbers in the outer zones can minimise the parasitic spallation neutron absorption as well as mitigate the slanting phenomenon of the radial power distribution. Outer zone loading leads to longer cycle lengths compared to un-poisoned reference cores.

To conclude, this poster session included new information on cross-section measurement. There is still need for improvement of the theoretical models. New initiatives were presented in terms of programmes and installations and I am looking forward to seeing new results at the next Information Exchange Meeting in Korea.

POSTER SESSION

Transmutation Systems

Chair: Dr. T. Y. Song (KAERI)

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SUMMARY

In the poster session of transmutation system, all 18 papers were submitted. Many technical aspects related to transmutation system were described.

ITEP submitted 7 papers which are mainly related to accelerator driven system. They cover radiotoxicity and decay heat calculations of PWR spent fuels using Th, U and Pu. They also showed the calculation results of Pu, Am, Cu, Tc and I transmutation in ADS, and suggested some ideas for ADS.

B.R. Bergelson from ITEP presented changes of radiotoxicity and decay heat power of actinides from spent uranium and uranium-plutonium nuclear fuel of PWR-type reactors at long-term storage. In another paper, he presented the same changes from spent thorium-uranium nuclear fuel. A.S. Gerasimov from ITEP suggested the principal opportunity for development of a project for a Demonstration Transmutation ADS (DTADS) as an international collaboration in Russia. He also discussed the opportunity to use high thermal neutron flux for effective incineration of fission products and minor actinides. He analysed weapon-grade plutonium burning and transmutation of the americium and curium isotopes from spent fuel in reactor or accelerator-driven installations with various neutron fluxes and spectra. Finally, he discussed the design of ADS complex for transmutation of ^{99}Tc and ^{129}I. G.V. Kiselev from ITEP introduced ideas and suggestions related to ADS technology. They include broad range of topics such as accelerator, target, sub-critical blanket, sectioned blanket, necessity of high neutron flux, two blankets installation with fluid fuel etc.

Two other papers dealt with gas-cooled reactors and one paper dealt with the topic of molten salt. A. Baxter from General Atomics reported work on the development of two concepts using helium-cooled reactor technologies for transmutation. Both concepts make use of thermal and fast energy spectra. D. Ridikas from CEA discussed gas-cooled target and assemblies, and considered both fast and thermal sub-critical assemblies. It was suggested that the best features of both critical and sub-critical systems can be merged by combining the GT-MHR with an accelerator driven sub-critical assembly. V. Ignatiev from Kurchatov Institute reported molten salt reactor developed in the framework of the ISTC#1606. ISTC#1606 includes experimental study of the key properties of the selected molten salt fuel composition, experimental verification of structural materials and physics & fuel cycle considerations on molten salt reactor.

Five other papers related to ADS covered kinetics, sub-criticality measurement, fuels for Energy Amplifier, Pb-Bi coolant and comparison of neutron sources. Only one paper covered transmutation in a fast reactor. J. Blazquez from CIEMAT remarked the subtleties behind the questions related to ADS such as sub-criticality, spallation source, kinetic parameters etc. Y. Rugama from Universidad Politecnica de Valencia presented an absolute measurement technique for the sub-criticality determination based on the Stochastic Neutron and Photon Transport Theory. S. Kaltcheva-Kouzminova from Petersburg Nuclear Physics Institute presented neutronics calculations of the accelerator driven reactor core EAP-80 with UO_2 & PuO_2 MOX fuel elements and Pb-Bi coolant. A. Pena from ETSII e IT showed calculation results obtained by using two different CFD codes for Pb-Bi coolant in ADS. It shows that Pb-Bi coolant circulation by buoyancy forces is an important result. M. Dahlfors from Uppsala University showed a preliminary comparative assessment relevant to the transmutation efficiency of plutonium and minor actinides. It has been performed in the case of ANSALDO's Energy Amplifier Demonstration Facility with two different neutron sources. A.V. Lopatkin from Research and Development Institute of Power Engineering introduced RDIPE's work on a concept of a fast lead-cooled reactor with UN-PuN fuel (BREST series).

The other two papers are related to comparison of BR2 and MYRRHA in transmutation characteristics, and usage of ADS for proliferation creation of MOX fuel. Ch. De Raedt from SCK•CEN presented the performances of the high flux materials testing reactor BR2 and compared them with those of the ADS prototype MYRRHA in its present development stage. Finally, V.B. Glebov from MEPhI evaluated the potential of the accelerator driven systems for enhancing the proliferation resistance of LWR MOX fuel. ^{232}U is added to the MOX fuel and irradiated in the ADS blanket to create the inherent reaction barrier.

CD-ROM CONTENTS AND INSTRUCTIONS FOR USE

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