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Improving Evaluations of Source Terms for Severe Accidents at Nuclear Installations

Final Report of the Source Term Evaluation and Mitigation Project (STEM)

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NUCLEAR ENERGY AGENCY
COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS

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Acknowledgements

This report was written by C. Mun, J. Colombani, M.N. Ohnet and O. Leroy of the Institut de Radioprotection et de Sûreté Nucléaire (IRSN). The IRSN performed the experiments for the Nuclear Energy (NEA) Agency Source Term Evaluation and Mitigation (STEM) project. The IRSN acknowledges the support of the NEA in hosting the project. Furthermore, the IRSN is grateful for the support and guidance from all the STEM partners: Electricité de France (EDF), Canadian Nuclear Laboratories (CNL), the Teknologian tutkimuskeskus (VTT, Finland), the Nuclear Research Institute (NRI, Czech Republic), the Gesellschaft für Anlagen– und Reaktorsicherheit (GRS, Germany), the Korea Atomic Energy Research Institute (KAERI), the Korea Institute for Nuclear Safety (KINS) and the US Nuclear Regulatory Commission (US NRC).

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List of abbreviations and acronyms

CNL	Canadian Nuclear Laboratories
CSNI	Committee on the Safety of Nuclear Installations (NEA)
EDF	Electricité de France
IRSN	Institut de Radioprotection et de Sûreté Nucléaire (Institute of radiation protection and nuclear safety, France)
KAERI	Korea Atomic Energy Research Institute
KINS	Korea Institute for Nuclear Safety
MB	Management Board
NEA	Nuclear Energy Agency
NRI	Nuclear Research Institute (Czech Republic)
PRG	Programme Review Group
RCS	Reactor cooling system
SA	Severe accident
ST	Source term
STEM	Source Term Evaluation and Mitigation (NEA project)

Executive summary

The Source Term Evaluation and Mitigation (STEM) project, hosted by the Nuclear Energy Agency (NEA) and operated by the Institut de Radioprotection et de Sûreté Nucléaire (French Institute of radiation protection and nuclear safety, IRSN), was launched in mid-2011 in order to improve the evaluation of source term (ST) for a severe accident (SA) at a nuclear power plant (NPP) and to reduce uncertainties concerning specific phenomena related to the chemistry of two major fission products: iodine and ruthenium (Ru). Three main issues were considered for these two radionuclides:

1. medium-term iodine releases, with specific attention to the stability of iodine aerosol particles under radiation (decomposition induced by radiation producing gaseous iodine species);
2. short-term and medium-term iodine-paint interactions under radiation;
3. ruthenium transport chemistry in order to determine the speciation of Ru, in particular the partition between gaseous and condensed forms during its transport through the reactor cooling system (RCS).

The STEM/IODINE results significantly contributed to increasing knowledge in this area, and it has led to improvements of the modelling of key phenomena concerning iodine chemistry in the different calculation tools of all the involved partners. For example, in the ASTEC code, these improvements mainly concern: 1) the interaction of I_2 and CH_3I with paint under irradiation; 2) the formation and radiolytic decomposition of gaseous and deposited iodine oxide aerosols; 3) The radiolytic conversion of gaseous I_2 into CH_3I and; 4) the interaction of I_2 with steel and aerosols. Concerning STEM/RUTHENIUM results, in the same way, a preliminary Ru transport model through the RCS was implemented in the ASTEC code, for example, allowing a first attempt to integrate a possible Ru source term in probabilistic safety assessment level 2 (PSA-2) tools. From this, the source term evaluation performed with PSA-2 tools concluded that ruthenium radiological consequences could be quite significant.

These STEM results have helped to precisely identify some of the major remaining issues, which are proposed for investigation during the second phase of the project. Indeed, according to the discussions during last STEM Programme Review Group (PRG) and Management Board (MB) meetings in June 2015 and the conclusions from the NEA/NUGENIA-SARNET International Workshop on the Progress in Iodine Behaviour for NPP Accident Analysis and Management (March 2015), a second phase of the project, called STEM-2, has been launched within the project framework of the NEA. This four-year follow-up project is focused on medium- and long-term releases and with four main items, as prioritised by the STEM PRG:

1. The effect of the ageing of paints on iodine behaviour (both under normal operation conditions and under accidental conditions): i.e. irradiation tests (medium- and long-term irradiation periods) on paints aged in representative reactor conditions and loaded with iodine, to confirm iodine release processes observed with un-aged paints (including adsorption/desorption kinetics);
2. The study of the stability of iodine oxide aerosols:
    i. Iodine oxides radiolytic decomposition;

    ii. Iodine oxides decomposition by carbon monoxide and/or hydrogen;

3. Radiolytic oxidation of multi-component iodine aerosols;
4. Complementary tests based on ruthenium revaporisation processes with representative oxidative conditions (atm. radiolysis simulants) with the presence of gaseous pollutants representative of RCS conditions.

It should also be noted that an analytical working group will be set up to promote a final comprehensive analysis of the results for reactor case source term evaluation, including mitigation aspects. Finally, parts of the experimental investigations will be discussed and shared with the Canadian Nuclear Laboratories (CNL), the operating agent for the forthcoming NEA Behaviour of Iodine Project phase 3 [BIP-3].

1. Introduction

The Source Term Evaluation and Mitigation (STEM) project [1], hosted by the Nuclear Energy Agency (NEA) and operated by the Institut de Radioprotection et de Sûreté Nucléaire (French Institute of radiation protection and nuclear safety, IRSN), was launched in mid-2011 in order to improve the evaluation of source term (ST) for a severe accident (SA) on a nuclear power plant (NPP) and to reduce uncertainties on specific phenomena related to the chemistry of two major fission products: iodine and ruthenium (Ru). Three main issues were addressed concerning these two radionuclides:

1. medium-term iodine releases with specific attention to the stability of iodine aerosol particles under radiation (decomposition induced by radiation producing gaseous iodine species);
2. short-term and medium-term iodine-paint interactions under radiation;
3. ruthenium transport chemistry in order to determine the speciation of Ru, in particular the partition between gaseous and condensed forms, during its transport through the reactor cooling system (RCS).

The present report summarises the main outcomes of the STEM project. The first chapter deals with the main outcomes concerning the iodine part (EPICUR facility), while the second chapter focuses on the main outcomes related to the ruthenium part (START bench).

2. Main outcomes of STEM/IODINE part

The Source Term Evaluation and Mitigation (STEM)/IODINE experiments, carried out in the EPICUR facility, were focused on the releases of volatile molecular iodine (I_2) and organic iodides (RI) from representative painted coupons loaded with molecular iodine [2] or iodine aerosol species (CsI, CdI_2, IOx[1]) and placed in the gaseous phase of an irradiation vessel for ≥ 30 hours to evaluate medium-term releases. Six tests were performed to study the releases of iodine from epoxy painted coupons loaded with I_2 ("LD tests" series). Eight tests were performed to study the releases of iodine from CsI (& CdI_2) aerosols deposited on epoxy painted coupons, but also deposited on quartz and stainless steel coupons to make the data interpretation easier by preventing any interaction with paint [3]. Four tests were performed with IOx deposited on epoxy painted or quartz substrates. For all these tests series, the effect of the parameters of main interest (temperature, relative humidity, initial concentration of iodine deposited on the sample surface, etc.) on the releases of I_2 and RI (and on the global volatilisation, defined as the difference between the initial quantity deposited on the coupon before irradiation and the quantity of iodine remaining on the coupon after irradiation) are presented and discussed below. Further details are given in "Main findings of the IRSN experimental programs performed on iodine chemistry in severe accident conditions" [4].

2.1. Releases from epoxy painted coupons loaded with I_2

The experimental results of the LD1 to six tests obtained in the STEM/EPICUR programme identify trends on the effects of the investigated parameters on the RI, I_2 and global releases from epoxy paint loaded with iodine exposed to radiations for more than 30 hours:

- A typical profile of iodine species releases from the epoxy paint was observed for this set of tests: RI and I_2 releases are fast and usually significant during the first hours of the irradiation phase (< 10 hours) whereas the release kinetics for both species are significantly lower after that time (see **Figure 1**).

- The ratio of RI released (RI released expressed in percentage of the initial activity loaded on the coupon, i.e. before irradiation) is stable for initial concentration of iodine on the paint lower than ~10-3 mol(I).m-2, and then this release ratio decreases for higher initial concentration of iodine. This phenomenon is probably due to the different (and preferential) chemical interaction sites for iodine-paint interactions depending on the initial concentration of iodine. The RI release is not significantly affected when the temperature increases from 80°C to 120°C or when the dose rate increases from 1.2 to 3.8 kGy.h-1. The RI release decreases as the relative humidity (RH) increases from 20 to 60%. In conclusion, regarding the conversion rate of iodine deposited on the paint into RI: the radiolytic conversion rate is mainly affected by the RH present in the irradiation vessel, an increase of the RH from 20 to 60% leads to a decrease of the conversion rate by a factor of about 3. This phenomenon may be explained by the fact that iodine diffuses more deeply in the epoxy matrix with a higher amount of steam, and then reacts chemically with the epoxy paint on its way to the deep layers. It might be more complex

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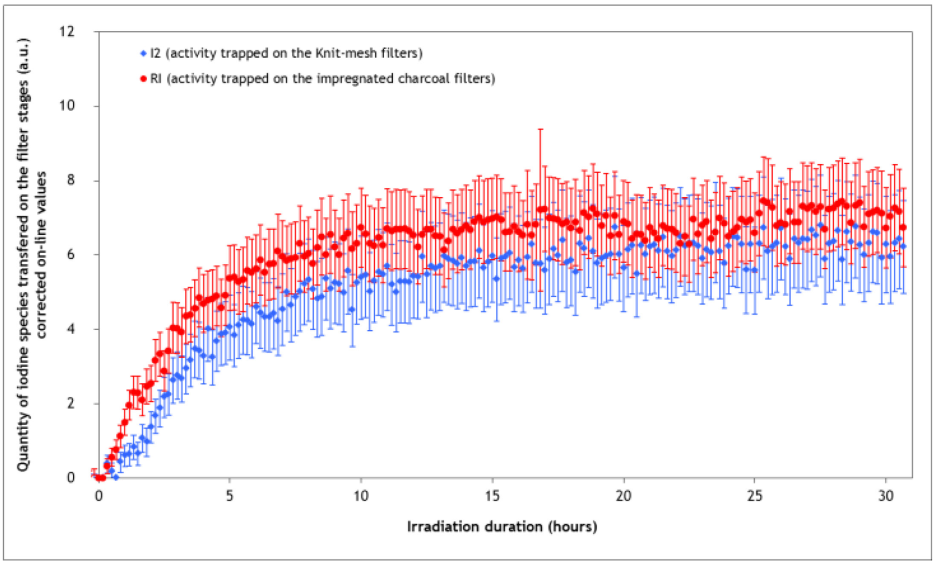
[1] IOx (or IxOy) stands for iodine oxides.

and difficult to get it back into the gaseous phase leading to a decrease of the RI release at a higher RH.

Figure 1. General trend of the release kinetics under radiation of I_2 (blue curve) and RI (red curve) from epoxy painted coupons pre-loaded with I_2



Source: IRSN.

- The ratio of I_2 released increases as the initial concentration of iodine on the paint increases; this phenomenon is probably due to the different (and preferential) chemical interaction sites for iodine-paint interactions depending on the initial concentration of iodine. The I_2 release decreases drastically as the temperature increases from 80°C to 120°C or as the dose rate increases from 1.2 to 3.8 kGy.h-1. The I_2 release decreases more moderately as the RH increases from 20 to 60%. In conclusion, the release of I_2 is mainly affected by the temperature and the dose rate: an increase of these parameters leads to a decrease of the release of I_2 by a factor higher than 5. A possible explanation of the temperature effect is that iodine interaction with paint could be enhanced at a higher temperature, leading to better iodine bounding to the paint and thus to a decrease in the I_2 release. A possible explanation of the dose rate effect is that the increase of this parameter leads to an increase of the concentration of radiation-induced radicals in the paint, which could promote the chemical reactions between iodine and paint-radicals and/or a better diffusion of steam and iodine between the polymer chains that leads to a more efficient iodine trapping in the deep layers of the paint. These two phenomena lead to a decrease of the I_2 release.
- The global release is linked to the RI and I_2 releases and is thus affected by the most influent parameters on the RI and I_2 releases.

2.2. Releases from coupons loaded with CsI (& CdI_2) under radiation

Previously, the importance of this process was suspected following analysis of the PHEBUS-FP tests, but it has not been quantified up until now, which is why tests investigating the effects of irradiation on deposited iodine aerosols (like CsI or CdI_2) were performed in STEM. The experimental results of the AER-CsI test series (AER-1 to 6 and AER-11) obtained in the STEM/EPICUR programme determined the effects of the investigated parameters on the aerosols I_2, RI and global releases from CsI aerosols deposited on several surfaces and exposed to radiation for 30 hours under temperature and humid air.

The global release is almost total from CsI aerosols deposited on quartz and stainless steel surfaces and irradiated under air at 50% RH, whereas this global release is incomplete from CsI aerosols deposited on epoxy paint surfaces (from 15 to 77% of global release depending on the dose rate and the initial concentration of CsI on the epoxy paint surface). This phenomenon can be explained by the CsI interactions enhanced in the case of epoxy paint and/or to the I_2 affinity for the epoxy paint leading to a “re-adsorption” process. The global release is linked to the aerosols, as well as I_2 and RI releases, and is thus affected by the most influent parameters on these releases.

- The aerosol release is important under certain irradiation conditions. These released aerosols are assumed to be iodine oxide species; they may be formed by reaction of I_2 (produced from CsI aerosols irradiation) and oxidative products formed by air radiolysis during the irradiation phase. The aerosol release decreases with epoxy paint surfaces, compared to quartz and stainless steel surfaces, because of the affinity of I_2 for epoxy paint surfaces (the reactions between I_2 and air radiolysis products are disadvantaged in favour of a phenomena of “re-adsorption” of I_2 on the epoxy paint). The aerosol release decreases at higher RH and high temperature, due to the thermal instability at high temperature of the iodine oxide species and their reactions of decomposition under humid conditions, which leads to I_2 production, also explaining the effect of several parameters on the I_2 release. The aerosol release increases at high dose rate, probably due to an increase of the concentration of air radiolysis products promoting the formation of iodine oxide species. This release decreases when the CsI concentration (and the particle size) decreases, probably due to the decrease of the I_2 release at low concentrations.

- Two typical profiles of I_2 release from CsI aerosols are observed, depending on the nature of the surface. The release of I_2 from CsI aerosols deposited on quartz or stainless steel surfaces starts in the early hours of the irradiation phase and presents two kinetics: the release is fast and important during the first hours of the irradiation phase, whereas the release kinetics are significantly lower after that time. In contrast, the release from CsI aerosols deposited on epoxy painted surfaces starts later and displays only one kinetic rate during the irradiation phase. Moreover, the final total release of I_2 from CsI aerosol deposited on epoxy paint surfaces is less than that from CsI deposited on quartz and stainless steel surfaces. This difference of behaviour may be due to the affinity of I_2 with the epoxy paint. Indeed, the radiolysis of CsI aerosols may produce I_2 that is released from quartz or stainless steel surfaces, whereas it may also be re-adsorbed in case of epoxy painted surfaces. Otherwise, the I_2 release increases at high RH (50% at 80°C), probably because the deliquescence point of CsI is almost reached (~57% at 80°C) and/or the decomposition of iodine oxide species into I_2 is enhanced at high RH. This release decreases at a high dose rate as a result of an increase of the concentration of radiation-induced radicals in the epoxy paint promoting the chemical reactions between I_2 and radio-induced paint-radicals and/or an increase of the concentration of air radiolysis products promoting the formation of iodine oxide species. The I_2 release decreases moderately as the CsI concentration (and the particle size) decreases, probably due to CsI aerosol interactions with the epoxy surface enhanced at low concentrations.

- The RI release was low in most of the tests. The RI release observed in the test performed with quartz or stainless steel coupons can only be attributed to the presence of organic contaminants in the loop, in the irradiation vessel or on the coupon or to contamination sources of the carrier gas.

The experimental results of the specific AER-12 test – aimed at studying the formation and release of volatile iodine species from CdI_2 aerosols under irradiation in "standard" conditions (irradiation during 30 hours in air with steam at 50% of relative humidity at 80°C) – have shown that the radiolysis of CdI_2 aerosols deposited on a quartz coupon leads to an almost total global release (~98%). The main species volatilised are molecular iodine (63.0% at the end of the test) and species suspected to be aerosols (13.4% at the end of the test). The molecular iodine volatilisation is attributed to the decomposition under radiation of CdI_2 aerosols deposited on the quartz coupon. The species trapped on the quartz fibre filter might be aerosol species; these aerosols produced from CdI_2 radiolysis are assumed to be iodine oxide aerosols. **The results of this AER-12 test are thus consistent with those obtained with CsI aerosols deposited on a quartz coupon and irradiated in similar conditions.**

2.3. Releases from coupons loaded with iodine oxide Aerosols (IxOy)

Air radiolysis leads to the formation of air radiolytic products like NO_2, O_3 or HNO_3 [5] as well as shorter-lived radical intermediates that can oxidise iodine and lead to the formation of iodine oxides (IOx) particles that settle down onto surfaces [6]. IOx are small aerosol particles, whose composition and chemical behaviour, particularly under irradiation, are not well known. A specific device to generate this kind of aerosols has been designed, and then experiments were performed in the EPICUR facility. Thus, four dedicated tests: AER-7, AER-8, AER-9 and AER-10 tests, have been carried out in order to evaluate the influence of the temperature and the influence of the nature of the coupon surface on the volatilisations of iodine species. The AER-10 test was performed in the same conditions as the reference test AER-7 in order to evaluate the reproducibility of the data obtained. The main outcome is that the instability (at least partially) under radiation of such iodine oxides species is now clearly experimentally demonstrated. More precisely, the results of these tests have shown that:

- Regarding the tests performed with iodine oxide aerosols deposited on a quartz coupon and irradiated at 80°C, the values of global volatilisation and molecular iodine release are reproducible. The aerosol fraction is very low, and the low organic iodide species release is attributed to the presence of organic pollutions.

- The temperature increase from 80 to 120° leads to an increase of molecular iodine release from oxide aerosols deposited on the irradiated quartz coupon. The molecular iodine release is significant during the first sweeping phase at 120°C (without radiation). These two results, coupled with literature data concerning the decomposition temperatures of IxOy species, might indicate that the iodine oxide species produced during the loading phase and deposited onto the quartz coupon would be I_2O_4 rather than I_4O_9 or HIO_3.

- The irradiation at 80°C of iodine oxide aerosols deposited on an epoxy painted coupon leads to a higher global volatilisation by a factor of four than with iodine oxides deposited on a quartz coupon. This result is probably due to a more significant production of aerosols (more than 10%) and to an increase of the release of I_2 (by a factor of four). In spite of the presence of potential organic radical sources in the case of the epoxy coupon, the release of organic iodides species remains of the order of magnitude of the measured values corresponding to pollutions. The increase of I_2 release with an epoxy painted coupon might be linked to an increase of the residual quantity of I_2 fixed on the epoxy painted coupon during the loading phase or to species deposited on the epoxy painted coupon with different speciation being less stable under radiation.

These experimental results of the STEM/IODINE part allow a better understanding of the iodine-paint interaction with containment surfaces and will be used to improve the models currently implemented in calculation tools of all involved partners, such as the ASTEC code [7], or other calculation tools like COCOSYS-AIM, KICHE, and RAIM.

3. Iodine-paint interaction

A specific deliverable of the Source Term Evaluation and Mitigation (STEM) project concerned a literature survey report dealing with the status of current knowledge and understanding on iodine-paint interaction issue. This report provides an **overall status on the paints composition, insights of their resistance towards different stresses (temperature, irradiation, humidity) and their ageing chemical processes.**

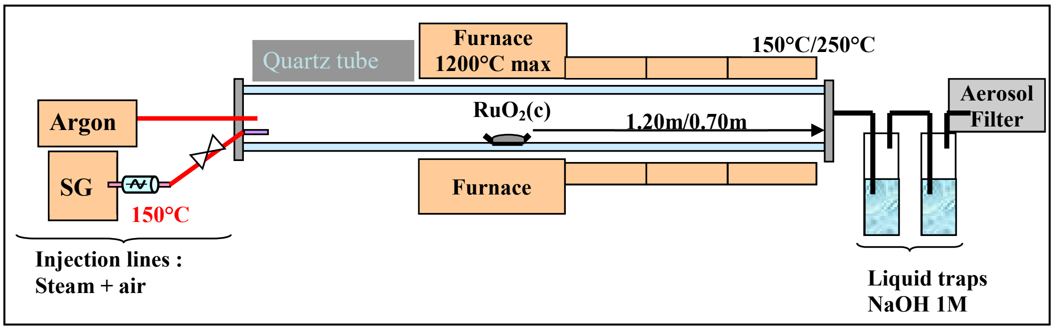
It also shows the likely chemical reactions between iodine and (aged) epoxy paint under irradiation and that, according to one article published in the literature [8] and some NEA Behaviour of Iodine Project (BIP) and BIP-2 test results, a pre-irradiation dose on a painted epoxy coupon (before the iodine loading) can reduce significantly the I_2 adsorption kinetics on the paint which could in turn lead to an increase of the gaseous iodine concentration, especially in the short term (<20 h). So, irradiation could have a significant influence on gaseous iodine concentration for doses received by the paint above 100 kGy as it could reduce I_2 adsorption rate and thus increase gaseous I_2 and probably gaseous CH_3I by I_2 radiolytic conversion reaction with organic compounds in the gaseous phase. As paints and more generally polymers can be exposed to several MGy in case of SA, it appears relevant to investigate the effect of paint ageing on: i) iodine adsorption kinetics and ii) iodine release kinetics from paints, both in normal operating conditions and during a SA by a high irradiation field composed of beta and/or gamma rays.

These identified issues are integrated in the STEM-2 description of work proposal (see Chapter 5 “Conclusions”).

4. Main outcomes of STEM/Ru part

As already demonstrated by previous international or domestic experimental programmes [9][10][11][12], ruthenium species released from the core in case of a severe accident (SA) with oxidising conditions can reach the containment and form volatile ruthenium tetroxide. The transport of Ru species in the reactor cooling system (RCS) is much less understood. That is why, in the frame of the Source Term Evaluation and Mitigation (STEM)/RUTHENIUM programme aimed at ruthenium transport studies [13], a series of 22 vaporisation and revaporisation experiments have been performed in the START test facility (see **Figure 2**). Ru is vaporised from a crucible containing RuO_2 powder in a furnace at 1 200°C, and then transported through a thermal gradient tube, with an outlet temperature of 150°C to 250°C depending on the tests. The main studied parameters were the carrier gas which was a steam-air mixture at different ratios, the type of thermal gradient ("abrupt" or "smooth") in the tube, and the tube material (quartz or stainless steel).

Figure 2. Scheme of the START test facility



Source: IRSN.

The Ru vaporisation kinetics in the furnace with oxidising conditions was characterised, with a good reproducibility, for different carrier gas mixtures (H_2O/air) and can be considered constant and steam content dependent. The dry air condition is the most favourable condition for the vaporisation of RuO_2 from the crucible at 1 200°C.

The long duration tests (7 hours) show the existence of transient phenomena during the 1st hour (vaporisation phase), confirming thus the necessity of studying the ruthenium chemistry for several hours to observe the precise phenomenology. Whatever the gaseous mixture, most of the vaporised Ru is deposited in the quartz tube [i.e. >95% of the released initial inventory (i.i.)]. The total Ru transported downstream of this tube (gas and aerosols) represents a few percent of the Ru mass vaporised for long duration tests, and is mainly in gaseous form, which confirms the potential important impact of Ru on the source term in oxidising conditions. The main conclusions of the long duration vaporisation tests can be summarised as follows:

- In long duration vaporisation tests, an "abrupt" thermal profile promoted the transport of Ru at the tube outlet (x4 compared to a "smooth" one), but it favours also RuO_2 particle formation (aerosols) in the gaseous phase. On the contrary, there is only Ru gaseous species with a "smooth" gradient profile.

- No clear effect of steam on the total Ru transported (gas + aerosol) at the tube outlet was observed in the vaporisation tests. Nevertheless, whatever the tube material and the gradient thermal profile, a carrier gas of 60 wt% steam and 40 wt% air promotes the quantity of gaseous Ru transported.
- The tube material (quartz or stainless steel) has no significant effect on the total Ru transported at the tube outlet during the vaporisation phase, whatever the gaseous mixture. However, it has an impact on the gas/aerosol partition.

Two-thirds of the tests have been performed with a revaporisation phase, consisting of sweeping the steam-air mixture into the tube containing Ru deposits (without generating ruthenium from the crucible). Contrary to the tests studying vaporisation, no transient phenomenon was observed during this revaporisation phase. The main conclusions of the revaporisation tests can be summarised as follows:

- Whatever the parameters studied (material of the tube: quartz or stainless steel, thermal profile: “abrupt” or “smooth” and gaseous mixture: with or without steam), the ending of revaporisation phenomenon could not be reached after 29h of experiment in START test conditions. So, the given values are not a maximum. During all these revaporisation tests, the Ru is transported mainly in gaseous form to the tube outlet. However, there is no Ru transport (lower than the detection limit) in absence of air (H_2O/Ar mixture).
- Whatever the thermal profile, it was noticed that in the transport tube the Ru deposit profile changed between the vaporisation and revaporisation tests. The Ru deposit shifted from the higher to lower temperature zones during the revaporisation test. With regard to the total Ru transported at the tube outlet (mainly in gaseous form), it remains in the same order of magnitude ($\leq$ 1% i.i.), so it can be said that the thermal profile (abrupt or smooth) has no significant effect.
- There is a gaseous mixture composition effect on the Ru transported at the tube outlet. With a quartz tube and an abrupt or a smooth thermal profile, the gaseous Ru transport is enhanced with an excess of air. On the contrary, for the tests with stainless steel tube and an abrupt thermal profile, the gaseous Ru transport is promoted with an excess of steam. To conclude: with an abrupt profile, there is an effect of the tube material on the Ru transport during the revaporisation phase (in condition with a steam/air ratio of 60/40 wt%).
- The gaseous Ru amount transported at the tube outlet seems to be promoted with the stainless steel tube.

The transport of ruthenium in conditions similar to those prevailing in the RCS, during a PWR SA (stainless steel, abrupt thermal profile, steam in excess and an outlet temperature at about 170°C), during both vaporisation and revaporisation phases lead to comparable gaseous Ru releases at the transport tube outlet (~2% i.i.), in START experimental test conditions. ***However, this is not the maximum reachable value because the revaporisation phase was not finished at the end of the experiment (only 12 hours of revaporisation).***

The STEM/Ruthenium results allowed the participants to gain knowledge on the key parameters impacting the transport kinetics, the partition between gaseous RuO_4 and particles of RuO_2 and the extent of the revaporisation process of deposited species in the thermal gradient tube. A preliminary Ru transport model through the RCS was

implemented in the ASTEC code [7], for instance, and a similar approach can be taken for other fission products release and transport codes.

5. Conclusions

The Source Term Evaluation and Mitigation Project (STEM)/IODINE results significantly contributed to increasing knowledge and led to improvements of the modelling of key phenomena concerning iodine chemistry in the different calculation tools of all involved partners. For example, in the ASTEC code [7], these improvements mainly concern: i) the interaction of I_2 and CH_3I with paint under irradiation; ii) the formation and radiolytic decomposition of gaseous and deposited iodine oxide aerosols; iii) The radiolytic conversion of gaseous I_2 into CH_3I and; iv) the interaction of I_2 with steel and aerosols. Concerning STEM/RUTHENIUM results, in the same way, a preliminary Ru transport model through the RCS was implemented in the ASTEC code [7], for example allowing a first attempt to integrate possible Ru source term in PSA-2 tools. From this, the source term evaluation performed with PSA-2 tools concluded that ruthenium radiological consequences could be quite significant.

These STEM results have helped to precisely identify the major remaining issues that are proposed to be investigated during the second phase of the project. Indeed, according to the discussions during the last STEM Programme Review Group & Management Board meetings (June 2015) and the conclusions from the International NEA/NUGENIA-SARNET Workshop on the Progress in Iodine Behaviour for NPP Accident Analysis and Management (March 2015), a second phase of the project, named STEM-2, has been launched in the /NEA framework. This four-year follow-up project is focused on medium- and long-term releases and with four main items, as prioritised by the STEM Programme Review Group:

- The effect of the ageing of paints on iodine behaviour (both during normal operation conditions and during accidental conditions): i.e. irradiation tests (medium- and long-term irradiation periods) on paints aged in representative reactor conditions and loaded with iodine to confirm iodine release processes observed with un-aged paints (incl. adsorption/desorption kinetics);
- The study of iodine oxide aerosols stability:
    - *Iodine oxides radiolytic decomposition;*
    - *Iodine oxides decomposition by carbon monoxide and/or hydrogen;*
- Radiolytic oxidation of multi-components iodine aerosols;
- Complementary tests based on Ruthenium revaporisation processes with representative oxidative conditions (atm. Radiolysis simulants) + presence of gaseous pollutants representative of RCS conditions.

It should also be noted that an analytical working group has been set up to promote a final, comprehensive analysis of the results for reactor case source term evaluation, including mitigation aspects. Finally, parts of the experimental investigation conditions have been discussed and shared with the Canadian Nuclear Laboratories (CNL) (Operating Agent of the forthcoming third phase of the NEA Behaviour of Iodine Project [BIP-3][14]).

6. References

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