Towards reducing the uncertainties on Source Term Evaluations: an IRSN/CEA/EDF R&D programme

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Abstract:

Many researches already carried out in the field of LWR severe accidents significantly improved the knowledge of physical phenomena involved in such accidents. They resulted in the development of increasingly better qualified assessment tools. However, major uncertainties still remain in some fields, concerning the assessment of risks for populations and the environment. IRSN, CEA, and EDF are therefore building up a co-operative research programme on this topic, based on separate-effect experiments. The results of these separate-effect experiments, would allow to improve models used for Source Term evaluation studies. The programme is fourfold and will address (i) iodine behaviour, (ii) boron carbide effects, (iii) air ingress, (iv) release of fission products. The programme is proposed for international co-operation.

1. BACKGROUND

The main technical issues for nuclear reactor safety in the event of a severe accident are the containment integrity and the characteristics of the source term to the environment. Since the TMI2 accident in 1979, progress in research has made possible the estimation of the potential consequences of a rather large spectrum of severe accident sequences. However, issues remain, in particular for source term, where knowledge is too scarce and a need for reducing uncertainties in source term evaluations was identified.

This is particularly the case for source term related to the release from the reactor core and transport through the reactor coolant system to the containment vessel of radioisotopes that are strong radio-contaminants (see e.g. [1]) and that can be volatile at relatively low temperature (down to 400 K) such as iodine ^{131}I and ruthenium isotopes ^{103}Ru and ^{106}Ru, under the tetroxide form RuO_4.

The challenge is the confidence we can place in the source term considered in the emergency plans:

- Are margins taken into account for the production of organic iodine sufficient or on the contrary too high, knowing that this compound is not retained by the filters used for containment venting?
- Is the ruthenium source term, an element potentially as radiotoxical as iodine and cesium combined, really greatly underestimated, as NRC inquiries might suppose it?

Among other programmes, but with a specific importance due to its integtral nature, the Phebus FP programme (see e.g. [2]) has played and is playing an important role in the qualification of calculation tools used for safety assessment. It also evidenced that certain phenomena were not well enough taken into account or quantified, concerning for instance iodine chemistry and fission product behaviour. These phenomena need to be further investigated in a complementary programme.

IRSN, CEA, and EDF are therefore building up a co-operative research programme on this topic, based on separate-effect experiments. The analysis of the results of the experiments will be the basis for the improvement of models used for Source Term evaluation studies. This programme spreads over a 6-year period (2005-2010).

2. IODINE

The most recent studies conducted at IRSN on the re-assessment of the S3 source term, which correponds to a delayed and filtered release to the environment, as well as within the context of level 2 PSA, confirmed that iodine was the main contributor to the short-term radiological risk in case of severe accident. Works tend to demonstrate that the S3 source term in organic iodine would be of the same order of magnitude than this previously assessed for 900MWe plant units, but to be multiplied by 2 or 3 for 1300 and 1400MWe plant units. In these studies, two prevailing factors are ill-quantified: the quantity of gaseous iodine released at the break (phenomenon highlighted in the Phébus-FP tests [3, 4 and quite misunderstood) and the production of organic iodine through reaction with paints in the gas phase of the containment building for which there is a great dispersal of experimental results. This point is especially important given the very low retention of organic iodine by the filters used for containment venting (U5 procedure for French reactors).

Besides, studies conducted within the context of the ICHEMM project [5] of the European 5th Framework Programme, which mainly aimed at studying the mechanisms of destruction of gaseous iodine in a containment building and the possible mitigation means, showed the potential importance of gaseous iodine reactions with the air radiolysis products in a containment building. Theoretical models were developed but have not yet been experimentally validated.

Following this result, IRSN has initiated several experimental programmes, that are part of the co-operative programme described in this paper:

- analytical tests in the EPICUR facility concerning iodine chemistry in the containment building under radiation (see figure 1),
- analytical tests in the CHIP facility dealing with iodine chemistry in the primary system, focused on the production of gaseous iodine at high temperature and the chemistry out of equilibrium (seefigure 2),
- PARIS analytical tests (see figure 3), carried out by FRAMATOME-ANP upon IRSN's request and dedicated to the study of reactions between air and iodine radiolysis products in a containment building (reactions tending to reduce the volatile iodine concentration), with surfaces of various types (steel, paints, silver).

Besides, the Phébus FPT-3 integral test, to be performed in 2004, should provide new results on iodine behaviour in conditions that had never been studied until now (absence of trapping by silver in liquid phase, increased mass transfer in "evaporating sump" conditions, reaction with methane in gaseous phase ...).

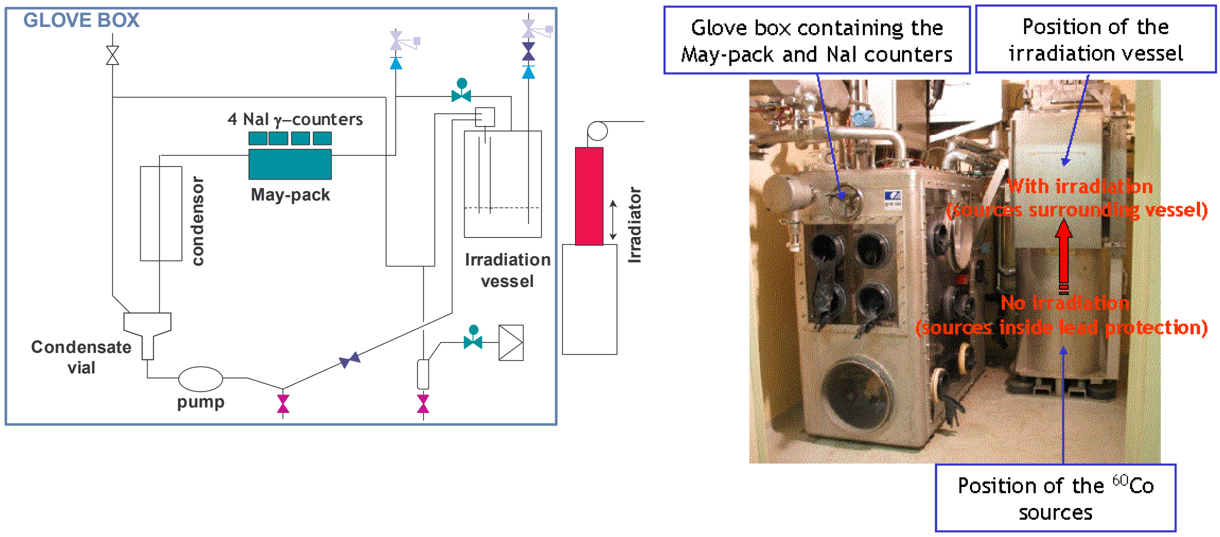

Figure 1: EPICUR: schematic view and picture of the facility

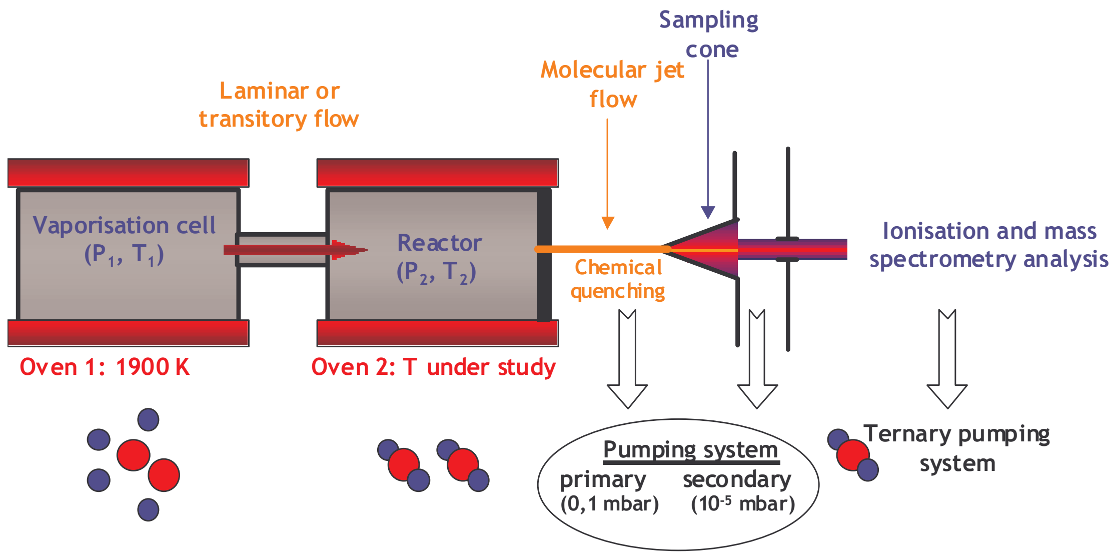

Figure 2: principles of the CHIP test device for the analytical tests

3. BORON CARBIDE

A number of reactors, including BWRs, VVERs and the most recent French reactors use boron carbide (B_4C) as an absorber material. B_4C oxidation by steam, following its initial degradation, produces borated and carbonated species, especially methane, which may have an influence on the chemistry of fission products (formation of organic iodine through homogeneous reaction), thus on their volatility and the source term. Until now, this effect has not been explicitly taken into account, especially in the molecular/organic iodine conversion rates:

- is this rate only concerned by the uncertainties related to paints or not?
- are the reactions of destruction under radiation prevailing?

Studies were conducted in the past on the B_4C degradation [6 and oxidation within the context of European projects (PECO of FP3 and COLOSS [7] of FP5). These studies resulted in the development of models describing the degradation (generation of eutectic mixtures relocating in the core lower part to form partial blockages) and the release of oxidation products (boron oxide, boric acids, carbon oxides, methane, hydrogen) influencing the chemistry of fission products in the primary system and the containment building, and therefore the source term.

Models have already been integrated in the computation codes, but not yet fully validated. For example, the consideration of low partial oxygen pressures expected during oxidation phases and zircaloy cladding may be mentioned. This is also the case for some aspects relating to the relocation of eutectic mixtures in the core lower part, such as their oxidation.

The impact of B_4C oxidation products (borated and carbonated species) on the FP chemistry has been studied theoretically only. It should be noted that there is a close coupling between phenomena related to core degradation and source term, which resulted in including a test dedicated to the B_4C behaviour in the Phébus-FP test matrix (FPT-3 test), and launching a programme of analytic tests on VERDI (within the framework of the COLOSS European project) and INTERMEZZO facilities.

The experiments that are part of the co-operative programme are the following:

- analytical tests on the degradation of long B_4C rods (about 30 cm), to be carried out in the INTERMEZZO furnace within the BECARRE programme (see figure 4)
- analytical tests on the characterisation of emitted B_4C oxidation products, to be carried out also in the INTERMEZZO furnace within the BECARRE programme,

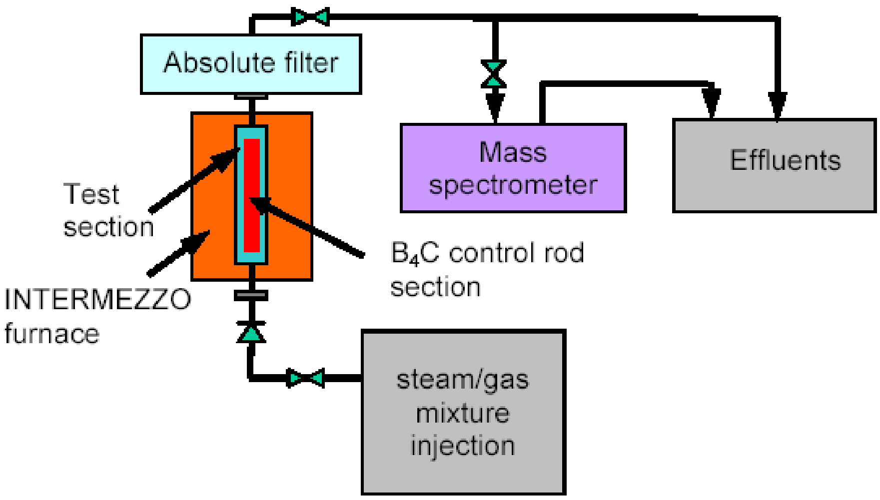

Figure 3: B4C control rod degradation test device

4 AIR INGRESS

An air ingress and its contact with fuel result in significant releases of some fission products. This is especially the case for ruthenium which has the same radiotoxicity as iodine in short term through ^{103}Ru isotope and as cesium in medium term through ^{106}Ru isotope. It is released in this case as a volatile FP (highlighted in AECL tests [8) and the tetra-oxide form may exist in gaseous form in the containment building (recently highlighted in AEKI RUSET tests [9]). Globally, the ruthenium release from the core may be 10 to 50 times higher than with steam only and the ruthenium tetra-oxide might represent a problem comparable with that of iodine. This behaviour is not currently recognised in source term studies. NRC pays particular attention to this specific point, especially for dry cask accidents [10]. Iodine might also be affected as its oxidation by air results in volatile forms. Thermodynamic calculations result in a complete revaporization of iodine that might be deposited on the structures before the air intake.

An air ingress may occur in various situations:

- during a reactor shutdown accident (core uncovering with the pressure vessel open),
- during a severe accident on a reactor after vessel melt-through,
- further to the emptying of a spent fuel storage pool, either in a nuclear power plant or in a reprocessing or storage plant,
- during a handling accident,
- during a transport accident.

As for "reactor" scenarios, the analysis performed within the framework of the Phébus STLOC-1 test preparation [11] led to prioritise scenario 2. It is considered that the current reviews of the level 1 PSA at IRSN will result in significantly reducing the probability of occurrence of scenario 1. As for scenario 2, the most penalising case for safety seems to be the case where there is not much non oxidised cladding left in the reactor vessel. In this case, the "solid" fuel will probably remain in place for a while and the oxidation by air of this fuel should result in significant ruthenium releases, a part of which might be injected in gaseous form into the containment building. Besides, fission products deposited in the primary system during the previous accident phase will be oxidised by air. This may result in more volatile forms, in particular for iodine.

As for emptying of spent fuel storage pools or handling accidents, the main question is to know if these accidents may develop into "severe accident". This would be the case if the power of oxidation of the cladding by air cannot be compensated by convection and radiation, resulting in a sudden increase of temperatures and significant releases of fission products.

Air ingress calculations performed by IRSN have already been discussed at the Phébus Air Ingress Working Group. Their results confirm those of previous studies performed at Sandia National Laboratory.

The following elements are part of the co-operative programme:

On the source term aspect.

- experimental investigations on the behaviour of volatile ruthenium (tetra-oxide) under radiation in a containment building,
- in medium term, experimental studies of re-volatilisation of various fission products under air, using facilities of the CHIP programme.
- to complement the existing database, a "semi-integral" experiment is foreseen in the VERDON facility; this test will be devoted to release and transport of fission products from irradiated fuel under air, and is to be performed in 2008/2009; it will include measurements of FP release, deposition in a thermal-gradient tube and possibly re-vaporisation,

These studies could be completed in a longer term (after termination of the 2005-2010 programme) by an integral test in the Phébus facility. The past experience shows that such a test with a representative source of fission products and structure materials provides unique information in such a complex area (chemistry under radiation and out of equilibrium).

On the oxidation aspect of zircaloy and other alloys under air

- performance of new tests (MOZART programme) whose requirement are defined from a the state of the art, in order to: 1/ reduce uncertainties on kinetics of cladding oxidation in the 800-1200°C range, 2/ process the case of pre-oxidised, pre-hydrided cladding (to simulate the effect of irradiation), 3/ study the nitriding resulting from an oxygen starvation and the combustion of nitrides (production of ZrN, desquamation, dispersal and severe oxidation of particles), 4/ study the oxidation through air/water vapour/carbon oxides mixtures (see figure 5).

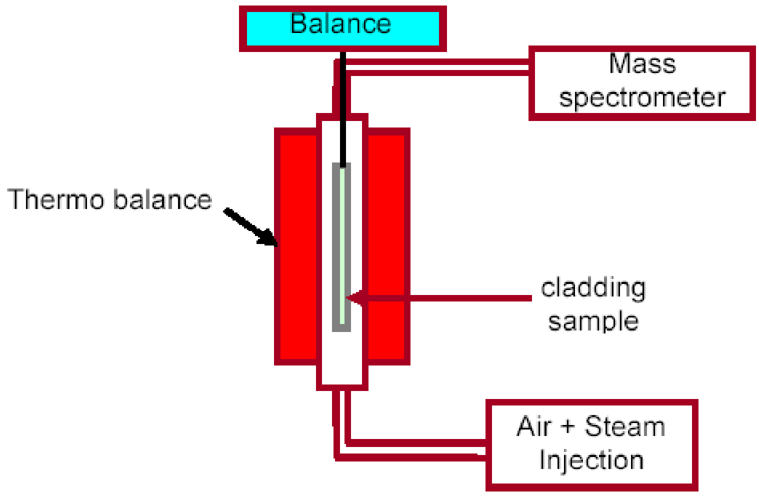

Figure 4: MOZART test device

5 FISSION PRODUCT RELEASE

The release of fission product from fuel has been mainly studied through small scale annealing tests, like those performed in the VERCORS facility [12]. Such experiments allow to build a data base on FP releases and more precisely, they provide relevant information about:

- the impact of the gas atmosphere on FP volatility (Vercors tests showed for example the strong impact of oxidizing / reducing conditions on the releases of semi-volatile FP such as Mo or Ba),
- the impact of burn up and fuel type (UO_2, MOX, …) on FP releases: for instance, significant releases at high temperature of Nd - 20 to 40%- at high burn up under oxidizing conditions were observed,
- and in some cases, the impact of material interaction on FP behaviour (trapping of Te and Sb in the cladding when not fully oxidized) and information about fuel relocation temperature (strong impact of the irradiation on the relocation temperature, but without significant evolution in the range 45 to 70 GWd/t).

These data are also used for the interpretation of integral experiments (such as Phébus-FP) in which the complex phenomenology (in particular the coupling between fuel degradation, Hypotheses which have been made about the impact of burn-up and oxygen potential on fission product behaviour need to be assessed for being used in predictive calculation tools. This will be done by performing detailed examinations of irradiated and annealed fuel pellets, using micro-analytical techniques in order to determine the location of fission products in the various phases as well as the corresponding compounds.

In addition, several items need to be better addressed such as, for example, the fission product releases from MOX fuel (previous annealing tests were mainly devoted to UO_2), the impact of the high burn-up microstructure on the fission gases release, the impact of gas atmosphere on the fuel relocation temperature…

In France, following the shutdown of the VERCORS facility at the end of 2002, it was decided to build a new facility, VERDON, which will be located at CEA Cadarache and allow the performance of FP release experiments (see appendix 4). Taking into account the experience of VERCORS, this new facility will provide well controlled conditions for fuel temperature, gas flow, which could be steam, hydrogen, air, or an inert gas, with accurate measurements of FP releases (on-line gamma spectrometers), and a detailed characterization of the fuel. Depending on the objectives, tests can be run with an instrumented outlet line (TGT, impactors, iodine trapping device…) and the fuel can be re-irradiated for a few days in an experimental reactor in order to create short-lived FPs.

The cooperative programme will focus :

- on the detailed analyses of the “Vercors” fuel (micro-characterization),
- on the performance of four tests in 2008 / 2009 in the Verdon facility to investigate FP release from irradiated fuel: two tests with MOX fuel, one with high burn-up UO_2 fuel, and one test under air conditions (see figure 5).

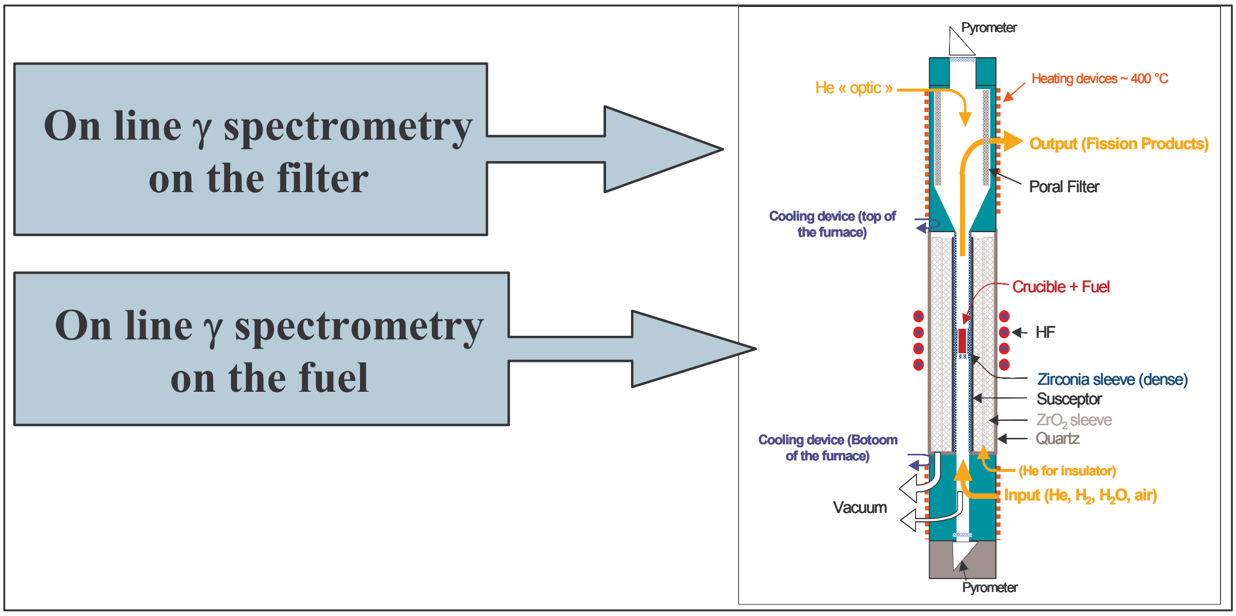

Figure 5: The VERDON furnace

6 CONCLUDING REMARKS

The source term separate effect tests programme presented in this paper is a joint IRSN/CEA/EDF project. It includes separate-effect experiments on iodine behaviour in the circuit and the containment, on boron carbide degradation and oxidation, on air ingress and on fission products release. The programme is using facilities located at Cadarache and operated by IRSN and CEA, some of them (CHIP and VERDON) being under construction. The project duration is six years, from 2005 to 2010.

It is expected that the outcomes of the programme will allow reducing the remaining uncertainties on source term evaluation studies, through improvement and better validation of the models used in calculation tools.

The programme is open for international participation. Discussions are ongoing with potential partners.

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