Progress In Modelling Fission-Product Release : An Update With Respect To IRSN's Release Codes

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ABSTRACT

IRSN presented in detail at this conference in 2003 the modelling of its release codes, ASTEC/ELSA and MFPR. Progress relating to key objectives is described here. ELSA aims to calculate reliably and rapidly release of fission products (FPs) and structural materials from a degrading core for all in-vessel configurations. Two areas were targeted for improvement: release of so-called semi-volatile FPs and release of structural materials. For the mechanistic code MFPR developed by IBRAE (Russian Academy of Sciences) and IRSN, with the new objective of analysis of design-basis loss-of-coolant accidents, modelling of fission-gas release has been improved mainly addressing processes occurring during normal operation. Further improvement depends in part on results of experiments and analyses that IRSN has launched with partners.

1. INTRODUCTION

IRSN develops two computer codes for calculating fission-product (FP) release from light-water reactor fuel, MFPR (developed by IRSN and IBRAE, the Nuclear Safety Institute of the Russian Academy of Science) and ASTEC/ELSA. The two codes have different objectives:

- ASTEC/ELSA, part of the integral code ASTEC (Van Dorsselaere et al. (2004)), is a semi-empirical tool aiming to calculate reliably and rapidly the release of all fission products (FPs), actinides and structural materials from a degrading core covering all in-vessel configurations during a severe accident;
- MFPR employs a highly mechanistic approach modelling phenomena at the fuel-grain and sub-grain scale with the aim of gaining insight into the state of the fuel and its FPs during any type of transient, severe accident or otherwise.

Two years ago at this conference IRSN presented in some detail the modelling of these two release codes, Dubourg et al. (2003), Nicaise and Ozrin (2003), Plumecocq et al. (2003). Since then progress has been made in some key areas that

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were deemed unsatisfactory. With respect to ELSA, two areas were identified for improvement:

- improvement of release of so-called semi-volatile FPs (Ba, Mo, Ru, etc.) since they can contribute substantially to the source term and play a major role in core degradation via decay heating;
- addition release of structural materials other than from Ag-In-Cd alloy and boron carbide (already modelled) since these influence FP speciation during transport and constitute significant sources of aerosol to the containment.

Regarding the MFPR code, modelling of fission-gas release has been improved with the main objective of improving the capacity to analyse design-basis loss-of-coolant accidents (LOCAs). The improvements mostly address processes occurring during normal reactor operation.

2. IMPROVEMENT OF THE ASTEC/ELSA CODE

It was concluded in a recent benchmark exercise for severe-accident codes comparing calculations with data from the Phébus FPT1 experiment that, in general, code modelling of structural-material release was in need of significant improvement, Clément et al. (2005). In relation to ELSA, it was decided to improve modelling of silver, indium and cadmium release from molten control-rod alloy and begin adding release of structural elements beyond silver, indium, cadmium and boron (from boron carbide used in some French PWR control rods as well as in BWR control blades). In effect, significant releases from structural alloys in the core are known to occur where the following elements are concerned: chromium, iron, niobium, nickel, tin and zirconium. Of these, the most urgent addition required was tin since it represents the most significant mass contribution to emissions from the core and is known to react with the FP tellurium affecting both its release from fuel rods (trapping on the unoxidized Zircaloy cladding) and transport in the RCS (formation of tin telluride).

It has been observed that tin release can be correlated with the extent of oxidation of the cladding where this is very clearly seen in the Phébus FPT-1 experiment (see Figure 1). On this basis a simple linear-proportionality model for tin release from Zircaloy cladding has been added; best results are obtained if the fraction of tin released is proportional to 0.6 of the oxidized fraction of Zircaloy, Haste and Plumecocq (2003). It can be expected that this simple model will be reliable in steam-rich conditions such as those of the FPT1 experiment; however, checking its performance against an experiment in which conditions are much more reducing, such as FPT-2, is required.

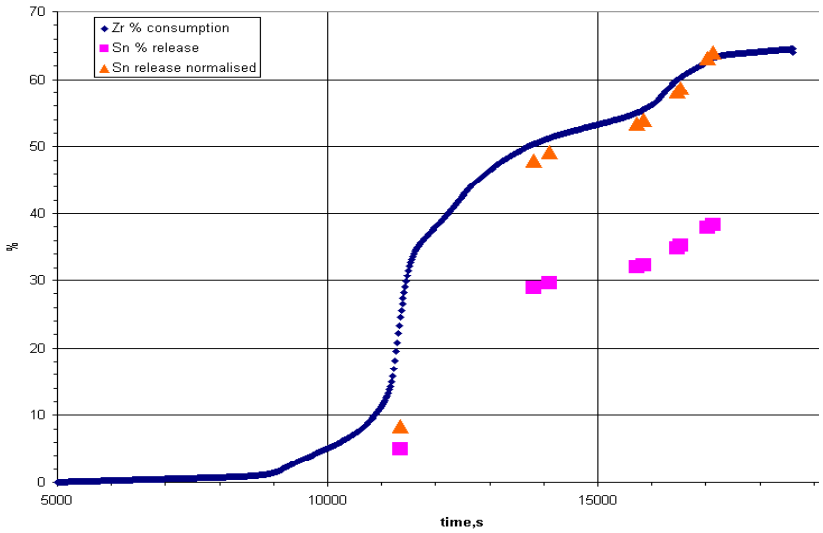

Figure 1: correlation between tin release and extent of Zircaloy oxidation (from H_2 production)

Concerning silver, indium and cadmium release, this was improved by making it consistent with the molten-pool modelling already included in ELSA, Plumecocq et al. (2003). The analogy that can be made is to depict the control-rod situation as evaporation of the molten-alloy pool in the control rod stub with additional release from candling (before this freezes in a cooler region), Haste and Plumecocq (2003). The pool surface can be taken to be at the temperature at which the stainless-steel control-rod canister melts, i.e., the melting point of Fe_3O_4 given the partly oxidized state of the canister during a severe accident. The improved performance of ELSA is shown in comparison with experimental results from the Phébus FPT1 test, see Figure 1. Acknowledging somewhat sparse data points for these elements, the results can be considered fairly satisfactory except for two aspects: the release of tin starts too early and indium release is significantly overestimated. The first problem arises from use of the Urbanic correlation for Zircaloy oxidation which is known to overestimate. The second problem is more subtle where, unlike silver and cadmium, indium evaporation is affected by unaccounted-for chemistry at the surface of the molten alloy (In, In_2O, InOH and In_2O_3). Indeed, the requirement for further improvement of releases from control rods is recognized in the frame of the SARNET programme, Maliverney et al. (2005).

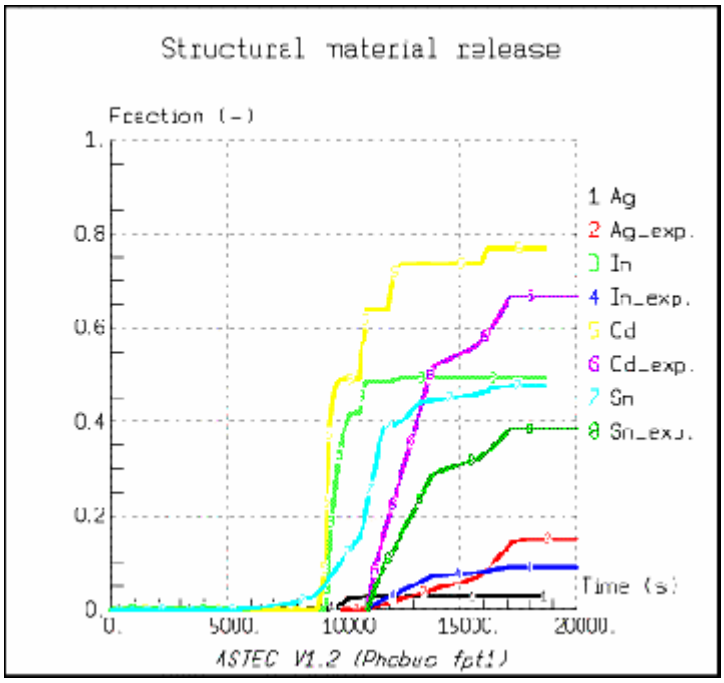

Figure 2: structural element release for the Phébus FPT1 experiment

On semi-volatile FPs, for ELSA these are FPs requiring special treatment due to their non-negligible release and sometimes high release sensitivity to the oxygen potential prevailing in the fuel. The approach in ELSA is based on the premise that evaporation from a solid phase (the $UO_{2\pm x}$ solid solution or a precipitate) into open fuel porosities is the rate-limiting step. On the basis of observation, the previous version of ELSA designated eight FPs as semi-volatile, viz. Mo, Ru, Sr, Ba, La, Ce, Eu and Y. From this starting point, the correlations for the total vapour pressures of these FPs have been overhauled re-assessing the main evaporating species with in some cases identification of new species that are more credible.

Release of strontium, lanthanum, cerium, and europium can be modelled on the basis simply of the metal vapour or its monoxide as the key species determining the total vapour pressure over the UO_2 solid solution. The same is done for barium except that it is treated as a separate phase where this is actually a pragmatic compromise given large uncertainty in the actual forms of the barium in the fuel: it appears to behave as though in the form of a pure BaO(c) phase whereas it can be expected that other species such as $BaUO_3$(c) and $BaZrO_3$(c) exist in the fuel. Indeed, in order to fit data from the VERCORS tests, Ducros et al. (2001), the vapour pressure produced by this simplification is further enhanced by an empirical factor

equal to 3.6. Lastly with respect to this FP, we note that additional work is required regarding its interaction with cladding where its trapping as barium zirconate is not modelled at present.

The vapour pressure of ruthenium is based on the sum of the pressures arising from the metal and its four oxides over the metal, i.e., Ru(c) as a separate phase due to the insignificant solubility of ruthenium in urania, Kleykamp (1985). However, in highly oxidizing, low-temperature conditions (such as may occur with air ingress following vessel melt-through in a PWR severe accident) ruthenium dioxide is the stable phase, not the metal. Hence, the sum of the same five vapour pressures is calculated over RuO_2(c)and the final pressure used for ruthenium is the minimum of that over the metal and that over the dioxide. We note that it may incorrect to treat the dioxide as entirely insoluble with respect to urania, Busker et al. (2003).

Concerning molybdenum, the chemistry may be quite complicated implicating the noble-metal precipitate phase, a number of molybdates, the dioxide and the trioxide. The evolution of the speciation in the fuel as a function of oxygen partial pressure and temperature is not yet well understood except in oxidizing conditions where these favour formation of caesium molybdate. Hence, in ELSA a simplification is made where the model is to calculate the vapour pressure based on evaporation of this species (i.e., congruent, or purely thermal evaporation) in oxidizing conditions; in reducing conditions, an empirical reduction of the caesium molybdate vapour pressure is applied using a reduction factor of 0.6.

Finally, for yttrium, reassessment for this FP has led to it being re-classed as a non-volatile, i.e., having a volatility comparable to or lower than that of urania itself and no longer requiring special treatment as a semi-volatile.

3. IMPROVEMENT OF THE MFPR CODE

Turning to the mechanistic MFPR code, modelling of fission-gas release has been improved with, in particular, the objective of analysis of design-basis loss-of-coolant accidents. The improvements mainly address processes occurring during normal reactor operation: modelling for evolution of point-defect concentration; modelling for evolution of dislocation density; fuel densification; and an improved model for grain growth. What follows can only summarize this work where more detail on most aspects can be found in Veshchunov et al. (2004) and Veshchunov et al. (2005).

Microscopic defects in the UO_2 crystal structure can strongly influence fission-gas release. These defects include point defects, such as vacancies, interstitials and fission atoms and extended defects, such as bubbles, pores and dislocations. Initially during steady-state irradiation, while the density of the generated dislocations remains relatively low, the effect of extended defects on point defects is weak and intra-granular bubbles formed from the solid solution of gas atoms in urania can be considered to be at equilibrium. In this case, the situation is simple since the defect structure of the crystal is practically excluded from consideration. However, at higher burn-ups, the dislocation density significantly increases and influences the evolution

of intra-granular bubbles with, notably, substantial suppression of generation of the intra-granular-bubbles. This leads to stabilization of intra-granular-bubble concentration in the late stage of irradiation accompanied by a noticeable increase in their mean size.

In power-transient and/or annealing conditions, the approximation of equilibrium bubbles is no longer valid and interactions of bubbles with point defects and dislocations occur. Indeed, during bubble growth and coalescence, extended defects (such as dislocation loops uniformly distributed in the grain bulk) may act as the main sources of vacancies (giving bubble equilibrium) and afford the equilibrium concentration of the point defects in the crystal bulk. This could explain enhanced bubble growth by dislocation motion observed in annealing conditions, Une and Kashibe (1990), Zacharie et al. (1998).

In MFPR the effect of point and extended defects on gas behaviour is now included explicitly. In non-equilibrium situations, a new mechanism for gas release due to dislocation motion has been implemented. The mechanism considers sweeping of bubbles and their delivery to the grain boundaries by dislocation segments climbing in the course of vacancy generation during annealing. Extensive validation of the MFPR dislocation-creep model showed that experimental results for gas release during post-irradiation annealing could be adequately simulated over the whole temperature range of the tests with an appropriate choice of initial dislocation density. Therefore, a new model for the dislocation evolution during irradiation has been implemented to provide appropriate initial conditions for the post-irradiation annealing and transient regimes.

Preliminary comparison with experimental data for the irradiation regime is very encouraging where comparisons have been made with data for dislocation density, Nogita and Une (1994), as well as with intra-granular-bubble concentration and mean size, Kashibe et al. (1993). This latter comparison is shown in Figure 3 where it is seen that satisfactory prediction of is obtained in the late stage of irradiation. Results for the dislocation density evolution obtained in this validation (not shown) are also in adequate agreement with measurements. The performance of MFPR is also shown with respect to data from the ADAGIO programme where, here, test 1065 provides the distribution of fission gases between intra- and inter-granular bubbles as a function of radial location in a fuel sample of 60GWd/t burn-up, Noirot et al. (2004). The experimental technique used is the same as that developed by AECL for the GBI tests, Elder et al. (1994), Dickson et al. (2000). Table 1 shows good agreement with the data where, we note, this post-irradiation distribution is critical in determining subsequent behaviour during a transient.

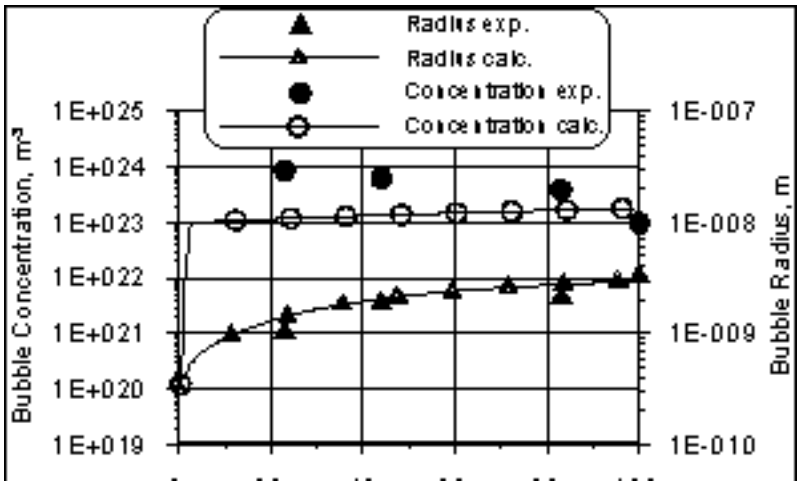

Figure 3: intra-granular-bubble concentration and bubble radius as a function of burn-up in steady-state irradiation (data from Kashibe et al. (1993)).

r (mm)	Gaz intra		Gaz inter		Rétention	
	exp	MFPR	exp	MFPR	exp	MFPR
0.9	73	83.8	14	6.8	87	90.6
1.3	73	84.2 *(79.9)*	14	7.0 *(6.72)*	87	91.2 *(86.6)*
2.0	73	84.6	14	7.6	87	92.2
2.4	87	84.7	8	8.2	95	92.9
3.0	87	84.6 *(79.3)*	8	9.0 *(8.96)*	95	93.6 *(88.4)*
3.7	88	83.8	8	9.5	96	93.3

Table 1: comparison of gas distribution between intra-and inter-granular bubbles with data from ADAGIO test 1065 (numbers in parentheses show some results obtained with the code before the latest improvements

Progressive extension of the modelled phenomena in the MFPR have turned it into a very valuable tool for exploring and understanding FP and fuel behaviour in a wide range of contexts such as normal operation, recovered LOCA and severe accident.

4. CONCLUSIONS

Improvements in IRSN's two FP-release codes have been described where specific areas were targeted for improvement.

With respect to the severe-accident code ASTEC/ELSA, two areas have received attention: improvement of release of so-called semi-volatile FPs (Ba, Mo, etc.) since these can contribute considerably to the source term and play a major role in core degradation via decay heating; and improvement of release of structural materials including that arising other than from control rods (already modelled) since these influence FP speciation and comprise significant sources of aerosol. Further effort will be required with respect to indium release and addition of other structural-material elements not yet considered (Fe, Cr, Ni, Zr and Nb).

With respect to the mechanistic release code MFPR, modelling of fission-gas release has been improved with, in particular, the new objective of analysis of design-basis loss-of-coolant accidents. Improvements have mainly address processes occurring during normal reactor operation since these condition behaviour during the subsequent transient. A model for dislocation generation and evolution during irradiation has been implemented along with a fuel-densification model and improved grain-growth model. Combined with the modelling describing the evolution of point defects and their interactions with gas bubbles, a self-consistent consideration of the whole system of point and extended defects in irradiated fuel has been obtained. This work has, as desired, significantly improved the predictive capability of the code in applications analysing design-basis and severe-accident LOCAs. Indeed, progressive extension of the phenomena modelled by MFPR have turned it into a valuable exploratory tool for understanding FP and fuel behaviour in a wide range of contexts.

Further improvement of modelling will depend on results of experiments and analyses that IRSN has already launched with partners. Two of these are micro-characterization of fuel for improving or confirming understanding of Cs and semi-volatile FP release and release of FPs - principally ruthenium - in air-ingress scenarios.

5. ACKNOWLEDGEMENTS

The authors thank their colleagues at IBRAE (Moscow), in particular Drs. Berdyshev, Ozrin, Shestak, Tarasov and Veshchunov, for their theoretical insights and numerical exploits embodied in the modelling of the MFPR code. The authors also thank Dr. Nicaise (IRSN) for useful ideas with respect to modelling molybdenum and barium release.

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