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PHEBUS FP RELEASE ANALYSIS USING A MICROSTRUCTURE-BASED CODE

ANALYSE DES ESSAIS PHEBUS FP DE LIBÉRATION DE PRODUITS DE FISSION À L'AIDE D'UN PROGRAMME DE CALCUL À MICROSTRUCTURE

L.N. CARLUCCI

Presented at the IAEA Technical Committee Meeting on Behaviour of Core Materials and Fission Product Release in Accident Conditions in Light Water Reactors CEN Cadarache, France 1992 March 16-20

Chalk River Laboratories

Laboratoires de Chalk River

Chalk River, Ontario K0J 1J0

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RÉSUMÉ

Le résultats des analyses de libération de produits de fission de deux pré-essais PHEBUS FP ont indiqué que le programme de calcul à microstructure, FREEDOM, prédit une différence importante d'ordre dans le temps et de pourcentage de libération des produits de fission gazeux, pour les deux essais. À titre d'indication de sa capacité de prédiction, on s'est également servi de FREEDOM pour modéliser le combustible à combustion nucléaire (massique) élevée éprouvé lors des essais VI-2 et VI-3 du Oak Ridge National Laboratory. Pour ces essais, on a constaté que le programme de calcul prédit à l'excès la libération pendant les premières phases des essais et la prédit insuffisamment pendant les phases ultérieures. La vitesse de libération, pour les deux essais, a été bien prédite. Ceci étant, il est probable que les résultats de la prédiction, par FREEDOM, de la libération cumulative finale, pour les deux premiers essais PHEBUS FP, sont des valeurs de limite inférieure. Toutefois, la différence importante d'ordre dans le temps, prédite, pour les deux essais, indique ce qui se produira dans la réalité. Par conséquent, on devrait tenir compte de cette différence en planifiant et exécutant les deux essais, particulièrement les points intéressant les mesure effectuées lorsque les essais sont en cours.

Communication présentée
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PHEBUS FP Release Analysis Using a Microstructure-Based Code

L.N. Carlucci

ABSTRACT

The results of pre-test fission-product (FP) release analyses of the first two PHEBUS FP experiments, FPT0 and FPT1, indicate that the FREEDOM microstructure-based code predicts significant differences in both the timing and percent of gaseous FP releases for the two tests. To provide an indication of its predictive capability, FREEDOM was also used to model the high-burnup fuel tested in the Oak Ridge National Laboratory experiments VI-2 and VI-3. For these, the code was found to overpredict releases during the early stages of the tests and to underpredict releases during the later stages. The release kinetics in both tests were reasonably predicted, however. In view of the above, it is likely that the FREEDOM predictions of the final cumulative releases for the first two PHEBUS FP tests are lower-bound estimates. However, the significant difference in the predicted timing of initial releases for the two tests is felt to be indicative of what will occur. Therefore, this difference should be considered in the planning and conduct of the two tests, particularly aspects related to on-line measurements.

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1. INTRODUCTION

The overall objective of the PHEBUS Fission Product (FP) program [1,2] is to conduct a series of integrated, in-pile experiments to improve our understanding of the physical and chemical behaviour of FP's, including their release from a degraded core as well as their transport and deposition in both the primary circuit and containment. The results of these experiments will be used to establish data bases for both the validation and improvement of computer codes for Source Term evaluation and severe accident analyses.

Currently, it is planned to conduct six tests to cover a representative range of phenomena expected to occur during postulated severe accidents. The first experiment, FPT0, which is being conducted as both a scientific and a technological test, will be conducted with a bundle of 20 trace-irradiated fuel rods, each having a burnup of 0.3 MWd/kgU. The second experiment, FPT1, will be conducted at the same thermal-hydraulic conditions as for FPT0, but with pre-irradiated fuel having a pre-transient burnup of 27.3 MWd/kgU. Both experiments will be conducted in an oxidizing environment. Thus these two tests will prove useful in studying the effect of burnup on not only bundle degradation phenomena but also on the timing and release of FP's and of their interaction with structural materials.

FPT0 and FPT1 FP release calculations done to date have been done with versions of the CORSOR code [3], which employs an Arrhenius-form, temperature-based correlation to calculate fractional release rates for a large number of FP species and structural materials. CORSOR predicts the same timing and similar fractional release rates for the two tests because both are assumed to be subject to the same temperature transient. However, differences are expected since, unlike high-burnup fuel, trace-irradiated fuel will have neither a significant amount of FP accumulation on grain boundaries nor the associated bubble interlinkage that would accelerate releases to the free void during the early phase of the test transient. Further, it is likely that release rates for both tests are overestimated, since CORSOR has been found to generally overpredict measured release rates [4].

To provide an alternative estimate of FP release for tests FPT0 and FPT1, a series of analyses has been done using FREEDOM, a more fundamental microstructure-based code, which is under development [5]. As well, to provide an indication of its predictive capability, FREEDOM was used to model FP release tests VI-2 and VI-3, conducted at the Oak Ridge National Laboratory (ORNL) [4,6,7].

This paper provides a brief description of the FREEDOM code and presents the results of both the PHEBUS and the ORNL FP release analyses.

2. CODE DESCRIPTION

2.1 FREEDOM

FREEDOM uses microstructure-based fission-gas and swelling models to predict the steady-state and transient release behaviour of 13 stable and radioactive fission-product nuclides, all assumed to be non-reacting and non-condensing. Radioactive chain decay and neutron-induced transmutation effects are modelled for the radioactive species.

The code models a fuel pellet as a set of independent regions of arbitrary volume, each consisting of voidless grains of UO_2 fuel. A moving-boundary, finite-difference method is used to model diffusion in individual fuel grains and grain-boundary sweeping resulting from equiaxed and columnar grain growth. Diffusion in the grains is assumed to be dominated by individual gas atoms, and hence the formation and migration of intragranular bubbles are not explicitly modelled.

Gas atoms arriving at grain boundaries by the diffusion and sweeping processes are assumed to form lenticular bubbles that grow until they touch and interlink, thereby venting the fission gases to the free voidage via cracks and surfaces. The dynamics of bubble growth, which is assumed to be dominated by the stable noble gases Xe and Kr, is calculated using the Hayns and Finnis [8] extension of the bubble growth model of Speight and Beeree [9]. The critical touching radius is calculated to be 0.7 microns, based on an assumed bubble density of 6×10^{11} bubbles per square metre of grain boundary area, a value derived from swelling measurements.

Perfect mixing is assumed between the stable noble gases and radioactive species in the grain boundary bubbles, and hence the release rate of the latter is assumed to be proportional to that of the former.

Grain growth, and therefore release by grain-boundary sweeping, is assumed to stop once grain-boundary bubbles have interlinked. All of the FP inventory is assumed to be released when the fuel melting temperature is reached.

Caution should be exercised when comparing FREEDOM results with measured release data. The code assumes that once bubble interlinkage occurs, excess as well as newly-arrived stable and radioactive fission gases are instantly and coherently released out of the fuel via cracks and surfaces that are presumed to intersect the tunnels of interconnected grain-boundary bubbles. In reality, a finite time is required for the fission gases to migrate along the interconnected tunnel network to fuel surfaces and cracks. As well, in the early stages of a transient, after the cladding fails, some additional time will be required for the fission gases to flow/diffuse along the fuel-cladding gap to the point of rupture. The consequences of not taking these time delays into account is that FREEDOM will indicate an earlier and more abrupt onset of release compared to measurement.

FP release is assumed to take place in a non-reactive, inert environment. No account is yet taken of the observed enhanced release due to deviation from stoichiometry that results as fuel is oxidized [10]. Work on the development of this capability is underway. As well, the effects on FP release of fuel liquefaction by the molten Zircaloy cladding and of fuel cracking during rapid cooling [11] are not modelled by the code, but are planned for future development. Hence, in situations where these phenomena dominate, FREEDOM will likely underestimate gaseous FP release.

FREEDOM has been validated against a database of stable-gas release from commercial and experimental fuel irradiated to maximum burnups of 29 MWd/kgU and linear powers of 120 kW/m [5], as well as against transient, short-lived fission-gas release data obtained from sweep-gas experiments [12,13]. In general, there is good agreement between predicted and measured data. Comparison with FP release data from high-temperature experiments simulating postulated accident conditions is so far limited to the ORNL results presented here.

2.2 FP Release in Fuel Rods

To perform a FP release analysis for an entire fuel rod, FREEDOM has been integrated into two codes that simulate the thermo-mechanical behaviour of single fuel elements: ELESIM for application to normal/steady-state operating conditions [14], and ELOCA.Mk5 for high-temperature transients [12,13]. A typical analysis consists of first running FREEDOM/ELESIM to simulate the irradiation history of the fuel to calculate the pre-transient gas pressure, as well as the distribution of fission products in the grains, on the grain boundaries and in the fuel-

cladding gap of an element. This information is then used to provide the initial conditions for FREEDOM/ELOCA, which in turn calculates the fuel-element behaviour and associated FP release during the transient.

3. INPUT PREPARATION AND ANALYSES CONDUCTED

3.1 FPT0 and FPT1

Separate FREEDOM/ELESIM calculations were done for FPT0 and FPT1. In each case, the calculation was for a typical element subjected to average (rather than mid-element peak) linear powers throughout the irradiation history. Information on the power histories and other relevant parameters of the fuel to be used for the two tests is listed in Table 1.

To calculate the fuel-to-cladding heat transfer rate, the version of ELESIM used in these analyses assumes zero radial clearance (normal condition for CANDU fuel); therefore, it was necessary to specify finite values for the calculations reported here. For FPT0 the fuel-to-cladding gap was set to a fixed value of 83 micrometres, whereas for FPT1 it was specified to decrease linearly as a function of increasing burnup, from a maximum of 100 micrometres to a minimum of 30 micrometres.

To perform the calculations for the FPT0 and FPT1 transients, ELOCA.Mk5 was used solely as a source of time-dependent fuel temperatures for FREEDOM. Hence the thermal stress model [12] in ELOCA.Mk5 was suppressed. Further, to eliminate any possible effects of fuel-cladding interaction on the calculated FP release, the cladding was "artificially failed" 900 s into the transient. This was done by setting the internal gas pressure to the external system pressure, which is 1.5 bar for both tests. The 900 s failure time was chosen as being representative of that expected, since calculations done to date indicate that, depending on the failure criteria chosen, clad rupture can occur between 700 s and 1100 s into the transient.

Because of the significant axial variation of temperature along each of the FPT0 and FPT1 bundles, separate FREEDOM/ELOCA analyses were done for each of 8 equal-length segments comprising the inner 80% of a typical interior element of each bundle (the two end segments were not modelled because of insignificant pre-transient FP inventories and releases during the transient). Sheath temperatures for each segment were obtained from the bundle degradation code ICARE2 [15], which was used to analyse the FPT0 reference scenario. With these clad temperatures as input, ELOCA.Mk5 was used to calculate the corresponding fuel temperatures illustrated in Figure 1; these agree with ICARE2 predictions within about 30 K. The same temperature transient was used for both tests.

To calculate the integrated release for the entire element, individual segment fractional release predictions were added using the local element power fraction as a weighting factor. Implicit in this approach is the valid assumption that the pre-transient inventory of a given FP species is directly proportional to the reactor axial power profile. The Belgian BR3 reactor power profile was used for the stable noble gases and the long-lived cesium produced in the FPT1 fuel, while the Phebus power profile was used for the cesium produced in the FPT0 fuel as well as for the short-lived species produced in both fuels. Figure 2 shows the power profiles for both reactors.

3.2 ORNL VI-2 and VI-3

Both of the 15-cm-long fuel segments used in the VI-2 and VI-3 experiments were cut from the same fuel rod, which was irradiated to a burnup of 42 MWd/kgU in the BR3 reactor at a maximum linear power of 25 kW/m, averaged over the 1-m length [6,7]. Because dimensional information of the fuel and cladding was not available, the corresponding FPT1 fuel data was

used. As well, since the detailed, burnup-dependent power history for the fuel was not given, the FREEDOM/ELESIM analysis was done using a single power history point (i.e., constant 25 kW/m from 0 to 42 MWd/kgU).

Each of the two test specimens was modelled as a single segment by FREEDOM/ELOCA. Figures 3 and 4 illustrate the experimental temperature history for the two tests.

4. RESULTS AND DISCUSSION

4.1 FPT0 and FPT1

FREEDOM/ELESIM predictions of the pre-transient internal gas pressure and stable gas FP distribution in the FPT0 and FPT1 fuel rods are summarized in Table 2. Only a small percentage of the total stable gas produced in the trace-irradiated, FPT0 fuel is predicted to have diffused to the grain boundaries; most of it remains within the fuel grains. In contrast, a large percentage of the high-burnup FPT1 fuel is predicted to have diffused and swept to the grain boundaries. The fact that 6.8% is predicted to be released to the gap indicates that some bubble interlinkage occurred in the higher temperature central portion of the fuel rod during the BR3 irradiation.

Figure 5 compares the FREEDOM/ELOCA predictions of element-average cumulative releases of stable noble gases, cesium-137 and iodine-131, for the two tests. Also included for comparison are the corresponding CORSOR-M predictions for cesium, which are virtually identical to those predicted for stable gases and iodine.

FREEDOM is seen to predict significant differences in both the timing and the magnitudes of gaseous FP releases for the two tests. For FPT0, the onset of significant release is not predicted until about 4000 s into the transient, reaching a maximum of about 60% at 7000 s, when temperatures begin to decrease as the neutronic power to the bundle is reduced to zero.

For FPT1, FREEDOM predicts a rather abrupt onset of release at about 1600 s for the stable noble gases and for the long-lived cesium, both largely produced during the irradiation in the BR3 reactor. This time can be interpreted as the onset of significant grain-boundary interlinkage. Release for these two species occurs at a high rate until about 2000 s, after which the release rate drops considerably. By about 3000 s, the cumulative release of the stable noble gases and Cs-137 has reached about 25%, which is close to the pre-transient sum of the gap and grain-boundary inventory.

The release pattern of iodine-131 is predicted to be different because it is produced entirely during the 15 days of pre-conditioning in the Phebus reactor and hence, as with FPT0, a very small percentage will have diffused to the grain boundary before the transient. However, the onset of release is predicted to be well before that for FPT0. This is because the early occurrence of grain-boundary bubble interlinkage for FPT1 allows all FP's arriving at the grain boundary to be immediately released to the open voidage.

The maximum cumulative releases for FPT1 are predicted to exceed those for FPT0, reaching 80 to 85% by 7000 s. This is largely a consequence of the earlier onset of release for FPT1. It is emphasized that the predicted cumulative releases for the two tests are likely lower bound values, because currently FREEDOM takes no account of possible enhanced releases due to fuel oxidation, fuel liquefaction and fuel cracking during the cooldown phase, all of which could occur to different extents during the two tests.

The release behaviour of the PHEBUS trace-irradiated and high-burnup fuel predicted by FREEDOM is similar to that observed in SFD 1-1 and SFD 1-4 tests, respectively, conducted at the Idaho National Laboratory at Idaho Falls [16].

CORSOR predicts the same timing and similar cumulative releases for both FPT0 and FPT1. Significant release is predicted to start 2000 s into the transient, reaching maximum values of 85 to 95% at 7000 s. The slight difference in predicted maximum releases is due to the difference in the two power profiles between BR3 and PHEBUS.

4.2 ORNL Tests VI-2 and VI-3

Based on the use of a single power history point, FREEDOM/ELESIM predicted less than 1% of the stable gases to be released to the gap, 30% to be accumulated on the grain boundaries and the balance to remain inside the grains.

Figures 6 and 7 compare measured and predicted releases of cesium by FREEDOM/ELOCA and CORSOR, as reported by Kress et al. [4]. In general, CORSOR overpredicts release. FREEDOM overpredicts release during the initial release phase and underpredicts it during the final phase; however, the release kinetics are reasonably predicted. The overprediction of the initial release by FREEDOM is due in part to the current absence of models to account for the transport of FP's beyond the grain boundaries. As well, FREEDOM likely overpredicts the pre-transient accumulation on the grain boundaries, because it does not model the formation and migration of intragranular bubbles. This could be important to consider for high-burnup fuel such as that used in the VI-2 and VI-3 tests, which was irradiated to 42 MWd/kgU.

The underprediction by FREEDOM during the late phase of both tests could be due to enhanced releases due to possible oxidation of the fuel. There is evidence from the measured hydrogen generation and post-test observations of the cladding that the Zircaloy was largely oxidized in both transients. The FREEDOM curves showing the higher releases in both tests correspond to a multiplication of the FP diffusion coefficient by a factor of two, to simulate the effect of possible enhanced release due to fuel oxidation. The higher diffusion coefficient was used beyond 2800 s for VI-2 and 6800 s for VI-3. The ELOCA code calculated complete oxidation of the cladding at these times.

5. CONCLUSIONS

Pre-test analyses of FPT0 and FPT1, of the planned PHEBUS FP test series, indicate that FREEDOM predicts significant differences in timing and percent of gaseous FP releases for the two tests, unlike CORSOR, which predicts the same timing and similar releases. Similar differences in release trends have also been observed in SFD 1-1 and SFD 1-4, which were conducted with trace-irradiated and high-burnup fuel, respectively.

When applied to model the cesium released from the 42 MWd/kgU fuel used in ORNL tests VI-2 and VI-3, FREEDOM has been found to overpredict the initial releases and to underpredict the final cumulative releases, for both tests. The release kinetics are reasonably predicted, unlike CORSOR, which predicts a continuously high release rate until 100% release is attained.

In view of the above, it is likely that the FREEDOM predictions of the final cumulative releases of gaseous FP's for FPT0 and FPT1 are lower-bound estimates. However, the significant difference in the predicted timing of initial releases for the two tests is felt to be indicative of what will occur. Therefore, this difference should be considered in the planning and conduct of the two tests, particularly aspects related to on-line measurements.

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Table 1: Relevant Input for FREEDOM/ELESIM Analyses of FPT0 and FPT1 Fuel

	FPT0	FPT1
Pre-test Power History	9 days pre-conditioning in PHEBUS at 15 kW/m average linear power to 0.3 MWd/kgU.	Irradiated to 26.7 MWd/kgU in BR3 reactor at 22.1 kW/m average linear power, followed by a 6.75 year storage period before 15 days of pre-conditioning in PHEBUS at 15 kW/m.
Enrichment (%)	4.5	5.0
Initial Grain Size (μm)	8.5	14.0
Fuel Density (kg/m^3)	10 500	10 390
Pellet Diameter (mm)	8.192	8.04
Pellet Length (mm)	13.51	12.34
Fuel Stack Length (m)	1.0	1.0
Effective axial Gap (mm)	37.6	37.6
He Filling Gas Press. (bar)	28	17.7
Diametral Clearance (mm)	0.1655	0.200
Cladding Thickness (mm)	0.57	0.58

Table 2: FREEDOM/ELESIM Element-Average Predictions for FPT0 and FPT1

	FPT0	FPT1
Internal Gas Pressure at 293 K (bar)	27.3	29.8
Predicted Stable Gas Release to Gap (%)	0	6.8
Predicted Stable Gas Release to Grain Boundary (%)	4.3	20.8
Predicted Stable Gas Left in Grains (%)	95.7	72.5

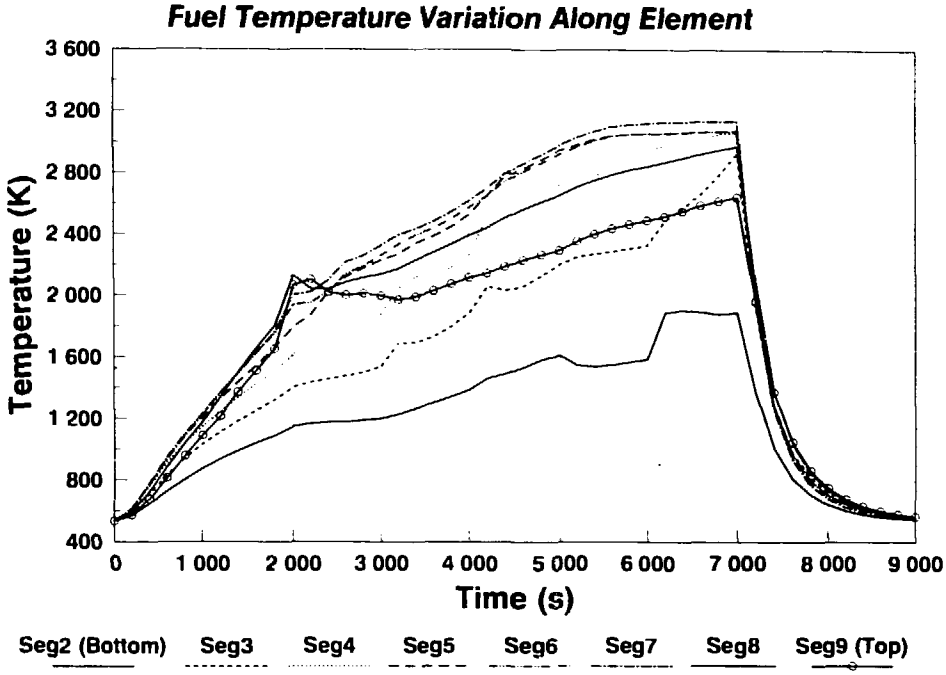

Figure 1: ELOCA-calculated fuel temperature variation along fuel rod

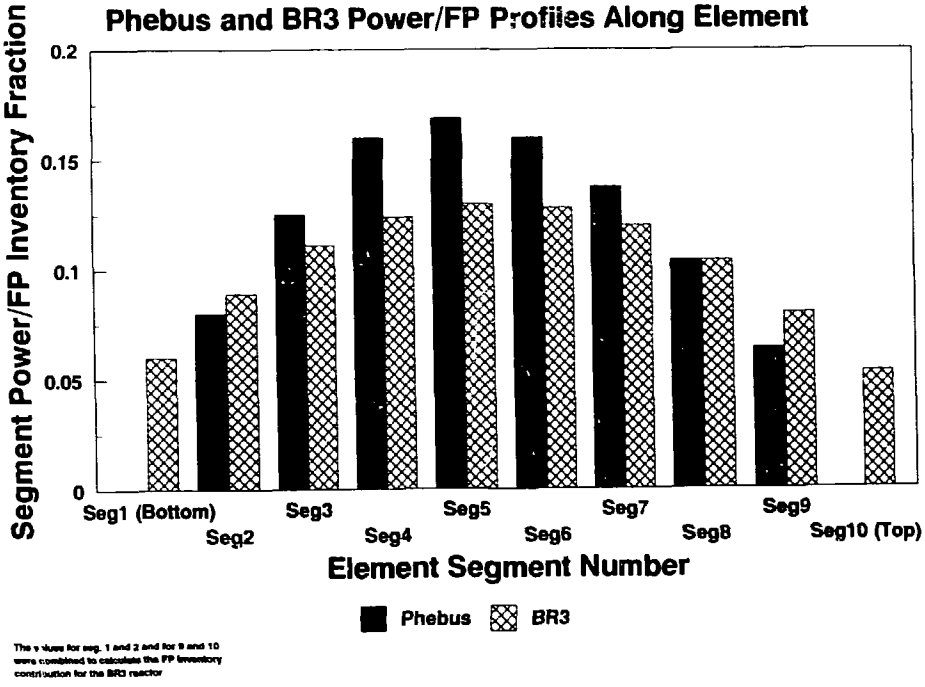

Figure 2: Power profiles along typical fuel rods in PHEBUS and BR3 reactors

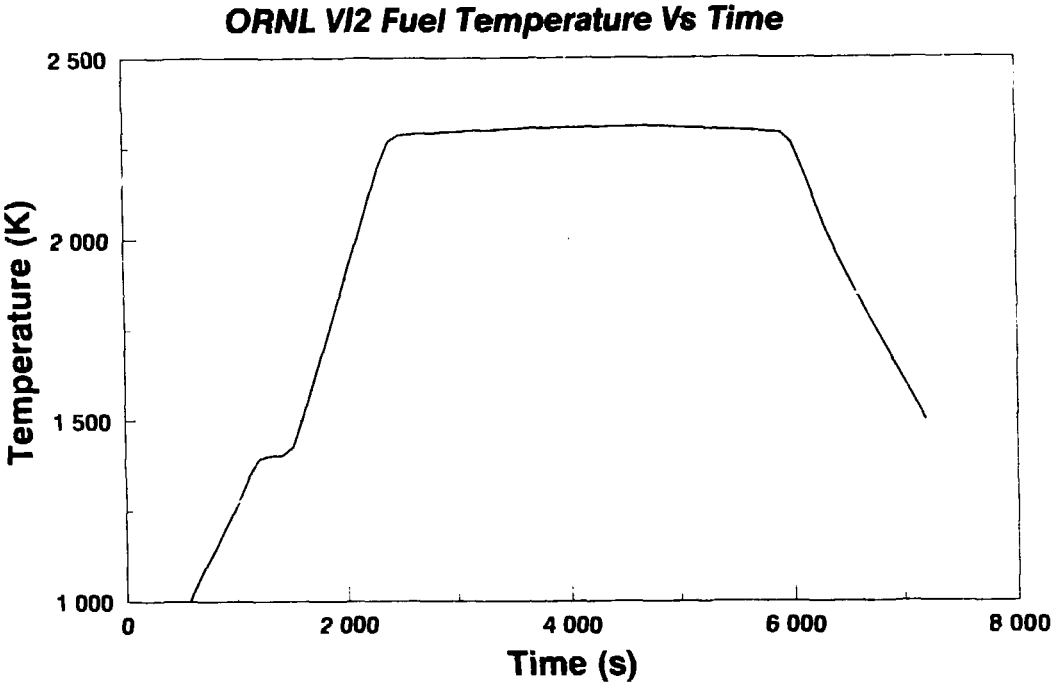

Figure 3: Fuel temperature versus time for ORNL test VI-2

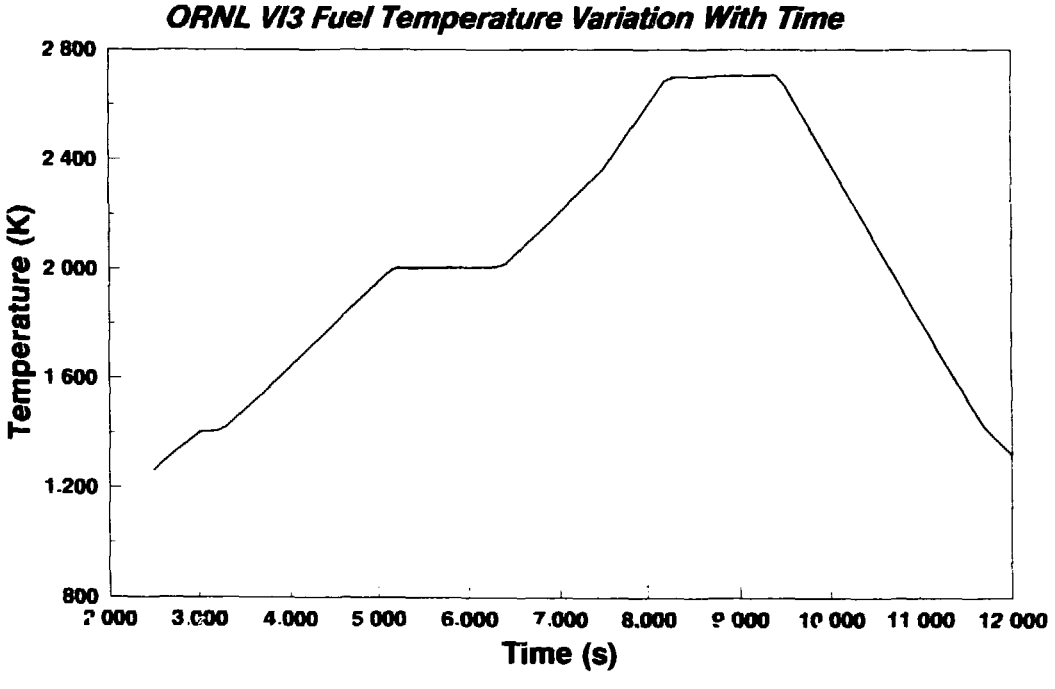

Figure 4: Fuel temperature versus time for ORNL test VI-4

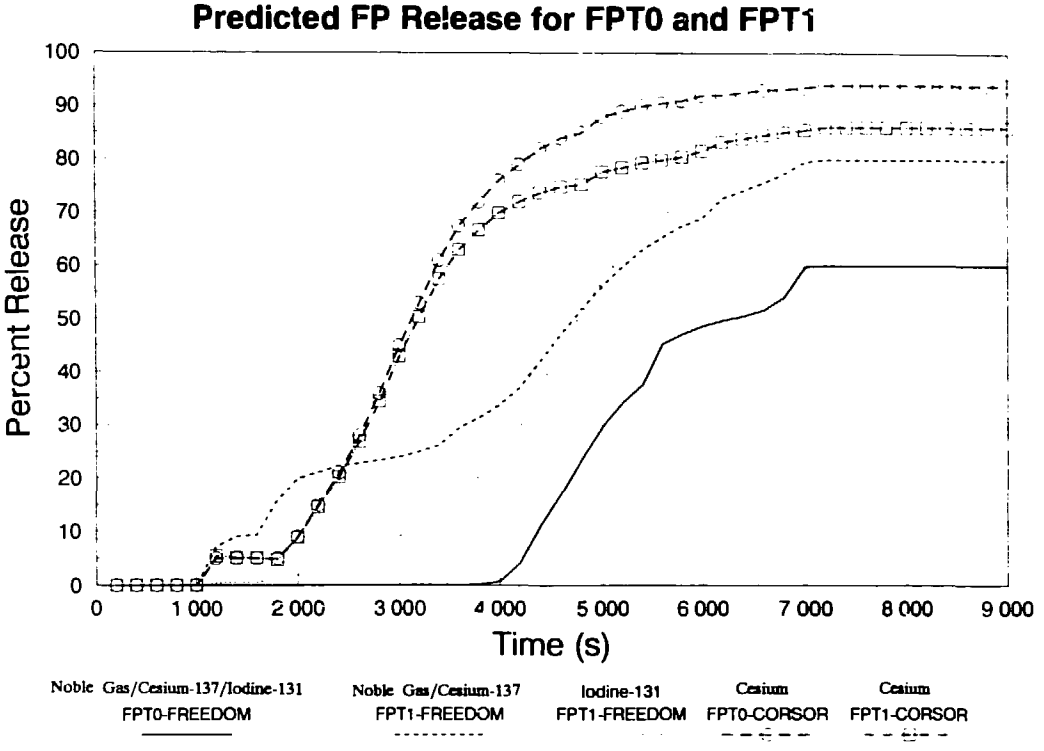

Figure 5: Predicted gaseous fission product releases for PHEBUS tests FPT0 and FPT1

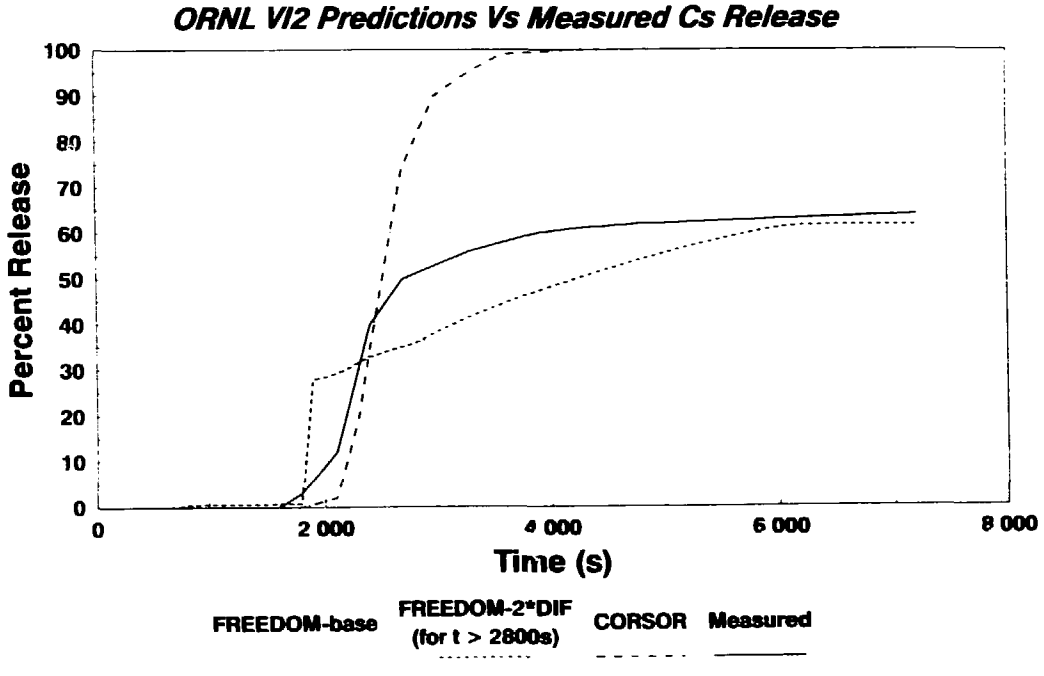

Figure 6: Predicted and measured cesium release for ORNL test VI-2

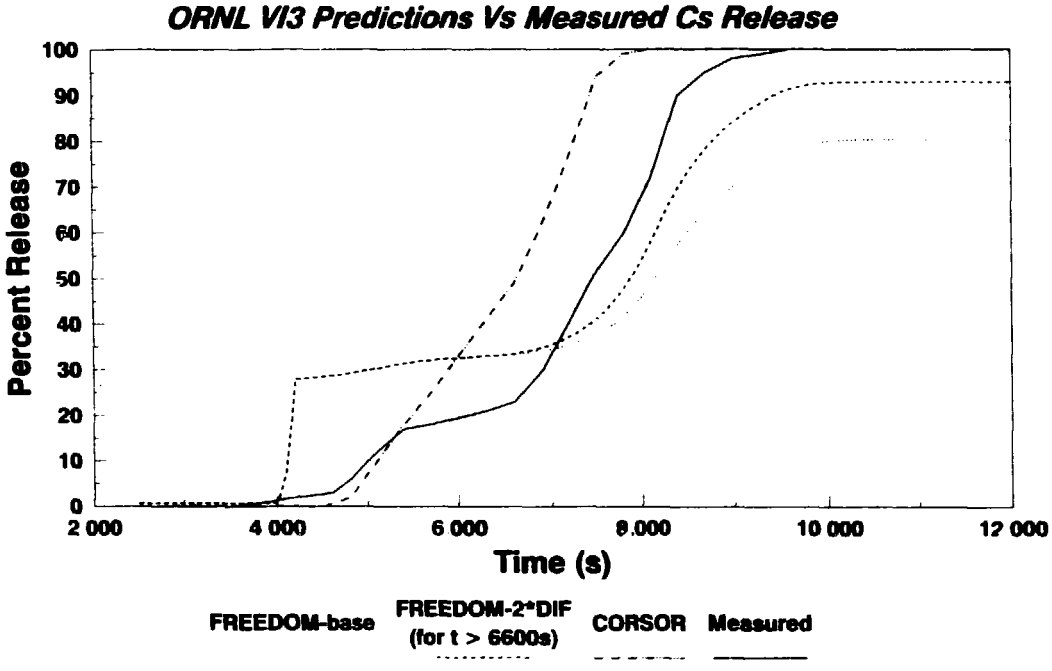

Figure 7: Predicted and measured cesium release for ORNL test VI-3

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