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Synthesis of VERCORS and Phebus Data in Severe Accident Codes and Applications

Randall O. Gauntt

Prepared by

Sandia National Laboratories

Albuquerque, New Mexico 87185 and Livermore, California 94550

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Synthesis of VERCORS and Phebus Data in Severe Accident Codes and Applications

Randall O. Gauntt
Reactor Modeling and Analysis Department
P.O. Box 5800
Albuquerque, NM, 87185-0736
USA

ABSTRACT

The Phebus and VERCORS data have played an important role in contemporary understanding and modeling of fission product release and transport from damaged LWR fuel. The data from these test programs have allowed improvement of MELCOR modeling of release and transport processes for both low enrichment uranium fuel as well as high burnup and MOX fuels. The following paper describes the derivation, testing and incorporation of improved radionuclide release models into the MELCOR severe accident code.

TABLE OF CONTENTS

List of Tables	6
List of Figures	7
1 Fission Product Release and Speciation	11
2 MELCOR Release Models.....	13
2.1 CORSOR-M.....	13
2.2 CORSOR-Booth	13
2.3 Limitations of MELCOR Release Models.....	15
3 Assessment of MELCOR Default Release Models	17
3.1 Recommended Modifications to MELCOR Booth Release Modeling ...	17
3.2 Assessment of Modified ORNL-Booth Model Against Phebus FPT-1 ..	21
3.3 Comparison to ORNL VI Tests and VERCOR Tests [12]	30
4 Evaluation of Fission Product Deposition Modeling	39
4.1 Deposition in FPT-1 Circuit (RCS Deposition)	39
5 MOX and High Burnup.....	41
5.1 MELCOR Analysis of RT-2 MOX Experiment Using Fitted Booth Parameters.....	41
5.2 MELCOR Analysis of RT-6 High Burnup Experiment Using Fitted Booth Parameters.....	45
6 Safety Implications of Current Fission Product Understanding	49
7 Summary and Recommendations.....	50
8 References	51

LIST OF TABLES

Table 1. CORSOR-Booth, ORNL-Booth and Modified ORNL-Booth Parameters	18
Table 2. Conditions for selected ORNL VI tests and VERCORS tests.....	31
Table 3. Total release from ORNL VI-2.....	33
Table 4. Total release from ORNL VI-3.....	34
Table 5. Total release from ORNL VI-5.....	34
Table 6. Comparison of fission product release from VERCORS tests RT-1 (LEU) and RT-2 (MOX).	43
Table 7. Diffusion coefficients for MOX and LEU fuel.	44
Table 8. Diffusion Coefficients for HBU and LEU Fuel.....	47

LIST OF FIGURES

Figure 1. Release fractions for different models – release temperature 2000K...	18
Figure 2. Release fractions at constant temperature for the ORNL-Booth and CORSOR-M models.	19
Figure 3. The time derivative of the release fraction.	19
Figure 4. Vapor pressures of selected species.	20
Figure 5. Schematic of the Phebus test facility showing test fuel bundle, heated lines, steam generator tube and simulated containment.	21
Figure 6. FPT-1 Nuclear and chemical heating history.	22
Figure 7. FPT-1 maximum bundle temperature history.	22
Figure 8. Emission gamma tomography of the end state condition of test FPT-1.	23
Figure 9. Comparison of ORNL-Booth versus CORSOR-M for Xe release (Class 1).....	24
Figure 10. Comparison of ORNL-Booth versus CORSOR-M for Cs release (Class 2).....	24
Figure 11. Comparison of ORNL-Booth versus CORSOR-M for Ba release (Class 3).....	24
Figure 12. Comparison of ORNL-Booth versus CORSOR-M for I release (Class 4).....	25
Figure 13. Comparison of ORNL-Booth versus CORSOR-M for Te release (Class 5).....	25
Figure 14. Comparison of ORNL-Booth versus CORSOR-M for Ru release (Class 6)	26
Figure 15. Comparison of ORNL-Booth versus CORSOR-M for Mo release (Class 7)	26
Figure 16. Comparison of ORNL-Booth versus CORSOR-M for Ce release (Class 8).	27

Figure 17. Comparison of ORNL-Booth versus CORSOR-M for La release (Class 9).....	27
Figure 18. Comparison of ORNL-Booth versus CORSOR-M for UO ₂ release (Class 10)	28
Figure 19. Comparison of ORNL-Booth versus CORSOR-M for Cd release (Class 11).	28
Figure 20. Comparison of ORNL-Booth versus CORSOR-M for Sn release (Class 12).....	29
Figure 21. Schematic of VERCORS test facility for measuring fission product release from small fuel samples.....	30
Figure 22. Comparison of Cs release for ORNL Booth modified with CORSOR-M for VI-2 run under steam oxidizing conditions.	32
Figure 23. Comparison of Cs release for ORNL Booth modified with CORSOR-M for VI-3 performed under steam oxidizing conditions.	32
Figure 24. Comparison of Cs release for ORNL Booth modified with CORSOR-M for VI-5 performed under reducing conditions.	33
Figure 25. Comparison of Cs release for ORNL Booth modified with CORSOR-M for VERCORS-2.....	35
Figure 26. Comparison of Cs release for ORNL Booth modified with CORSOR-M for VERCORS-4.....	35
Figure 27. Comparison of Xe release for ORNL Booth modified with CORSOR-M for VERCORS-4.....	36
Figure 28. Comparison of iodine release for ORNL Booth modified with CORSOR-M for VERCORS-4.....	36
Figure 29. Comparison of Te release for ORNL Booth modified with CORSOR-M for VERCORS-4.....	37
Figure 30. Comparison of Ba release for ORNL Booth modified with CORSOR-M for VERCORS-4.....	37
Figure 31. Comparison of Mo release for ORNL Booth modified with CORSOR-M for VERCORS-4.....	38
Figure 32. MELCOR-predicted fission product deposition in FPT-1 circuit using default CORSOR-M release modeling	40

Figure 33. MELCOR-predicted fission product deposition in FPT-1 circuit using modified ORNL-Booth release modeling..... 40

Figure 34. RT-2 release of cesium as a function of test sample temperature. ... 42

Figure 35. Plot of RT-2 Cs release data as a function of time using inverted Booth solution. 44

Figure 36. Comparison of the MELCOR Predicted Release of Cs Class for VERCORS Test RT 2 to the Experimental Measurement of Cs 137..... 45

Figure 37. Instantaneous RT-6 diffusion coefficient and Booth model fits. 46

Figure 38. RT-6 release measurements compared to Booth model predictions for HBU and LBU fuels..... 47

1 Fission Product Release and Speciation

This report presents currently recommended MELCOR specifications for modeling the release of fission products from reactor fuel under severe accident conditions. The present recommendations modify the default specifications in MELCOR. Separate specifications are provided for use in either in-vessel fission product release or spent fuel pool release because of differences in the oxidation potentials of steam and air environments. The motivations for these recommended changes are described. Predictions of fission product releases are compared to results of recent experiments.

The current MELCOR default settings for calculating fission product release specify the CORSOR-M release model, described in the MELCOR Reference manuals and in a Battelle report [1].” Also described in these references are the CORSOR and the Booth diffusion release models, implemented in MELCOR as the CORSOR-Booth optional release model. The CORSOR and CORSOR-M models are classified as *fractional release rate* models, differing only slightly in mathematical form, which specify the fractional release rate of the fission product inventory remaining unreleased. These are empirical models that are based largely on the small-scale HI and VI experiments performed at ORNL [11].

The Booth diffusion model is by comparison a phenomenological model, albeit simplified, that describes the transport of fission products within fuel grains to the grain surface as a diffusion process. In the MELCOR implementation of the Booth diffusion treatment, gas-phase mass transport is included as a process for limiting fission products movement from the grain surfaces to the atmosphere. Elements such as molybdenum that are modeled in MELCOR as having very low vapor pressures are ultimately released at a low rate regardless of the rate of diffusion within the grain. Once released from the fuel, fission product class combinations can be defined, such as CsI, in order to represent fission product chemistry and speciation. In the present code architecture, multiple combination assignments such as CsI and Cs_2MoO_4 were not foreseen and must be approximated. Once assigned to the chemical class on release, generally no additional chemistry is allowed, an exception being CsI chemisorption with subsequent revaporization of iodine, leaving the permanently chemisorbed Cs attached to a deposition surface

Critical assessments of these models and their performance have up to now been few in number, partly due to unavailability of additional quality data. One assessment performed by ORNL with MELCOR 1.8.2 surveyed the performance of the MELCOR default models when applied to predict the results of the VI series of tests [2]. The report observed that while total releases could often be adequately predicted, the predicted release rates were often not in good agreement with the data. Recommendations were made for code modeling improvements, including provision to vary release based on the $\text{H}_2/\text{H}_2\text{O}$ environment. Recently, additional experimental data have come available from international testing programs, in particular the French VERCORS program and

the Phebus integral experiments. A recent user assessment of current MELCOR release models in the prediction of these tests has identified some deficiencies that are partly remedied in the recommendations of this report. The Phebus experiments in particular reveal shortcomings of the empirical CORSOR and CORSOR-M models with respect to release rates during the initial fuel heatup, and have been found to significantly overestimate early release rates even though total integral releases might compare reasonably well. Additionally, the integral Phebus tests provide release data under conditions that are significantly less coherent in terms of temperature and oxidation/reduction conditions than in the small scale tests (HI, VI and VERCORS) where the fuel sample is small, temperatures are uniform and oxidation/reduction conditions controlled and constant. The Phebus experiments provide conditions for release that are more representative of conditions expected in the full-scale reactor accidents, and are used as the principal reference for judging the performance of the MELCOR release models.

2 MELCOR Release Models

The release rate models in MELCOR are briefly summarized as follows. The original CORSOR model correlates the *fractional* release rate coefficient in exponential form,

$$k = A \exp(BT) \quad \text{for } T \geq T_i \quad \text{Eq. 1}$$

where k is the release rate (fraction per minute), A and B are empirical coefficients based on experimental data, and T is the core cell component temperature in degrees Kelvin. Different values for A and B are specified for three separate temperature ranges. If the temperature is below the lowest temperature limit specified, no release is calculated.

2.1 CORSOR-M

The CORSOR-M model correlates the same release data used for the CORSOR model using an Arrhenius form,

$$k = k_o \exp(-Q/RT) \quad \text{Eq. 2}$$

The values of k_o , Q , and T are in units of min^{-1} , kcal/mole , and K , respectively. The value of R is 1.987×10^{-3} in (kcal/mole-K) .

2.2 CORSOR-Booth

The CORSOR-Booth model considers mass transport limitations to radionuclide releases and uses the Booth model for diffusion with empirical diffusion coefficients for cesium releases. Release fractions for other classes are calculated relative to that for cesium. The effective diffusion coefficient for cesium in the fuel matrix is given by

$$D = D_o \exp(-Q/RT) \quad \text{Eq. 3}$$

where R is the universal gas constant, T is the temperature, Q is an activation energy, and the pre-exponential factor D_o is a function of the fuel burn-up. The cesium release fraction at time t is calculated from an approximate solution of the diffusion equation for fuel grains of spherical geometry [3],

$$f = 6 \sqrt{\frac{D't}{\pi}} - 3 D't \quad \text{for } D't < 1/\pi^2 \quad \text{Eq. 4}$$

$$f = 1 - \frac{6}{\pi^2} \exp(-\pi^2 D't) \quad \text{for } D't > 1/\pi^2 \quad \text{Eq. 5}$$

where

$D't = Dt/a^2$ (dimensionless), and

$a =$ equivalent sphere radius for the fuel grain

The release rate of Cs during a time interval t to $t+\Delta t$ from the fuel grain is calculated as

$$\text{Release rate}_{\text{Cs}} = \frac{[f(\sum D'\Delta t)_{t+\Delta t} - f(\sum D'\Delta t)_t] V\rho}{F\Delta t} \quad \text{Eq. 6}$$

where ρ is the molar density in the fuel, V is the fuel volume, F is the fraction of the Cs inventory remaining in the fuel grain, and the summations are done over the time steps up to time $(t + \Delta t)$ and t , respectively.

The release rate formulation in the CORSOR-Booth model is also limited by mass transfer through the gas-phase. The gas-phase mass transport release rate from the fuel rod for species k , \dot{m}_k , is calculated using an analogy from heat transfer as

$$\dot{m}_k = \left[\frac{A_{\text{fuel}} Nu D_{k,\text{gas}}}{D_{\text{fuel}} RT} \right] \cdot (P_{k,\text{eq}} - 0) \quad \text{Eq. 7}$$

where

$D_{\text{fuel}} =$ diameter of fuel pellet

$A_{\text{fuel}} =$ fuel rod flow contact area

$D_{k,\text{gas}} =$ diffusivity of class k in the gas mixture

$Nu =$ Nusselt number

$P_{k,\text{eq}} =$ equilibrium vapor pressure of class k at temperature T .

In the mass transfer term the driving potential is the difference in pressure at the surface of the grain and the pressure in the free stream atmosphere, here assumed to be approximately zero.

The effective release rate for Cs given by Equation 6 is a combination of the rates given by diffusion and by gas-phase mass transport. Therefore, the contribution from diffusion only is taken as

$$DIFF_{\text{Cs}} = \left[\frac{1}{\text{Release rate}_{\text{Cs}}} - \frac{1}{\dot{m}_{\text{Cs}}} \right]^{-1} \quad \text{Eq. 8}$$

The diffusion release rate for species other than cesium is given by multiplying the cesium release rate by an appropriate scaling factor S_k for each radionuclide class k :

$$DIFF_k = DIFF_{\text{Cs}} S_k \quad \text{Eq. 9}$$

The combined mass transport and diffusion release rate $\dot{m}_{tot,k}$ for class k is then

$$\dot{m}_{tot,k} = \frac{1}{DIFF_k^{-1} + \dot{m}_k^{-1}} \quad \text{Eq. 10}$$

Inspection of equations 10 together with equation 7 reveals that the release predicted by the MELCOR models can be mass transfer limited by low vapor pressures even if the diffusive transport is large.

2.3 Limitations of MELCOR Release Models

The default fission product release models implemented in MELCOR are quite simplified and are more than 10 years old. The implemented models base the release of all radionuclide chemical classes on the release predicted for Cs, which in the Booth model is appropriately considered a diffusion process. Scaling factors are used to estimate release of other species based on the data fit to experimentally observed Cs release in spite of the fact that it is recognized that likely not all fission product classes diffuse at the same rate out of the fuel grains, nor are all principal release mechanisms well represented as a diffusion process. Consideration of speciation in MELCOR release models is crude and for the most part fixed at the time of release to represent the predominate speciation. The vapor pressures of the MELCOR release classes are defined to represent the presumed fission product speciation.

A better treatment would be to allow the vapor pressure to be adjusted to account for local speciation as affected by oxidizing or reducing conditions and to then source these species into appropriate chemical classes. Such modifications are probably needed for Ba, Mo UO₂ and Ru. Provision does exist to consider the extent of cladding oxidation to attempt to simulate retention of Te or Ba, but data are needed to use this provision effectively. Separate diffusion coefficients for each of the volatile classes would probably be appropriate, and a UO₂ oxidation model is needed to account for the effect of stoichiometry on diffusion and to predict fuel volatilization. UO₂ volatilization may be responsible for release of UO₃ as well as other non-volatile species by physical stripping away the fuel matrix containing the fission products. A number of more recently evolved release models consider the effect of fuel stoichiometry on the diffusion coefficient as well as the oxidizing/reducing potential of the environment [4,5,6,7]. The VICTORIA code considers a large number of potential fission product species in a thermodynamic equilibrium approach; some simplifications to this numerically burdensome approach may be needed [8].

In the more recent models, often, fission products are classified into three main groups, volatile (Xe, Cs, I, Te) semi-volatile (Ru, Ba, La, Ce) and non volatile (UO₂ and actinides). Volatile fission products are released based on the Booth diffusion model where the diffusion coefficient includes effects of UO₂ hyperstoichiometry. The hyperstoichiometry in turn is determined by a fuel oxidation model. Release of semi-volatile fission products are strongly affected by vapor pressure which in turn is affected strongly by speciation determined by the oxidizing/reducing conditions that arise as air, steam and hydrogen interact

with the metallic cladding on fuel in the release location. Non-volatile release may be dominated by UO_2 volatilization by formation of UO_3 , producing fuel matrix degradation and fuel vaporization. The French Elsa code follows this approach, using models similar to those reported by Lewis *et al.* [4,5].

Clearly, more detailed (and flexible) release modeling can be adopted in MELCOR in the future. The importance of accounting for speciation and the ensuing effect on species volatility (vapor pressure) is clear. In the present approach, as described in the following section, assumptions are made about the dominant speciation at the time of release and maintaining this assumed speciation globally throughout the core region. A more elegant model would allow variation of release speciation as conditions in the core change locally and temporally with respect to steam and hydrogen concentrations. In the case of air exposure, such as in spent fuel pool accidents, different assumptions about speciation, especially concerning Ru release, are needed. [9]

3 Assessment of MELCOR Default Release Models

The HI-VI ORNL tests provided the original basis for development of the MELCOR fission product release models. The Phebus FPT-1 integral experiment is used as the principal basis for evaluation of release modeling options. In previous assessment exercises, in particular the ISP-46 (International Standard Problem 46 [10]), the MELCOR default CORSOR-M release model was found to predict reasonable total release fractions for many fission products. On the other hand, the empirical model was found by many MELCOR ISP participants to over predict the initial release rates. Similar, rapid, early release is also found when the CORSOR option of MELCOR is used. The Booth diffusion treatment for release was thought to be a superior release model since it has some basis in physical transport processes, however, investigation of the MELCOR CORSOR-Booth option using the default Booth release parameters was found to produce inferior results, with total release of Cs and other fission products being significantly underpredicted for test FPT-1. A review of the literature revealed numerous more recent parameter-fits to the Booth solution.

3.1 Recommended Modifications to MELCOR Booth Release Modeling

A number of alternative models are described in an ORNL report that recommends updated values for the previously discussed models [11]. Shown in Figure 1 are release fractions predicted at a constant temperature of 2000K by the various release models discussed in the ORNL report. From this, it can be seen that CORSOR-M predicts the largest release. This trend is consistent with observations from analyses considering measured releases from FPT-1. Similarly, the CORSOR-Booth diffusion model predicts the lowest release rate of all of the models. This too is consistent with MELCOR analyses of FPT-1 using these modeling parameters. Judging that a best fit might lie somewhere in between these extremes, the ORNL-Booth parameters were subsequently investigated in MELCOR analyses of FPT-1, wherein significantly improved release predictions were obtained. The ORNL-Booth parameters were recommended over the CORSOR-Booth parameters in the 1995 ORNL report. The ORNL-Booth model is specified by the parameters in Table 1. Figure 2 shows other comparisons between the ORNL-Booth and CORSOR-M release behaviors. The fractional release rates shown in Figure 3 were derived by differentiating the release fractions predicted by the two models and shown in Figure 2.

Table 1. CORSOR-Booth, ORNL-Booth and Modified ORNL-Booth Parameters.

	CORSOR-Booth	ORNL-Booth	Adjusted ORNL-Booth
Diffusion coeff. D_0	$2.5 \times 10^{-7} \text{ m}^2/\text{sec}$	$1 \times 10^{-6} \text{ m}^2/\text{sec}$	$1 \times 10^{-6} \text{ m}^2/\text{sec}$
Activation Energy Q	3.814×10^5 joule/mole	3.814×10^5 joule/mole	3.814×10^5 joule/mole
Grain radius, a	6 μm	6 μm	6 μm
Class Scale Factors	---	---	---
Class 1 (Xe)	1	1	1
Class 2 (Cs)	1	1	1
Class 3 (Ba)	3.3×10^{-3}	4×10^{-4}	4×10^{-4}
Class 4 (I)	1	0.64	0.64
Class 5 (Te)	1	0.64	0.64
Class 6 (Ru)	1×10^{-4}	4×10^{-4}	0.0025
Class 7 (Mo)	0.001	0.0625	0.2
Class 8 (Ce)	3.34×10^{-5}	4×10^{-8}	4×10^{-8}
Class 9 (La)	1×10^{-4}	4×10^{-8}	4×10^{-8}
Class 10 (U)	1×10^{-4}	3.6×10^{-7}	3.2×10^{-4}
Class 11 (Cd)	0.05	0.25	.25
Class 12 (Sn)	0.05	0.16	.16

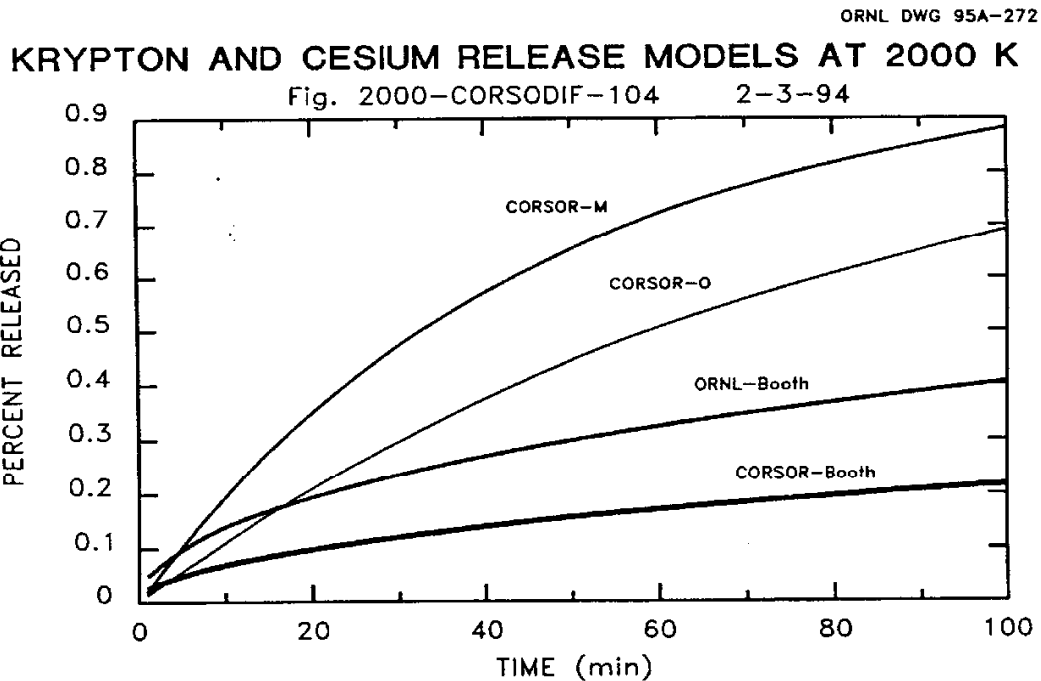


Figure 1. Release fractions for different release models – release temperature 2000K. Note CORSOR-M predicts the largest release whereas CORSOR-Booth predicts the smallest release.

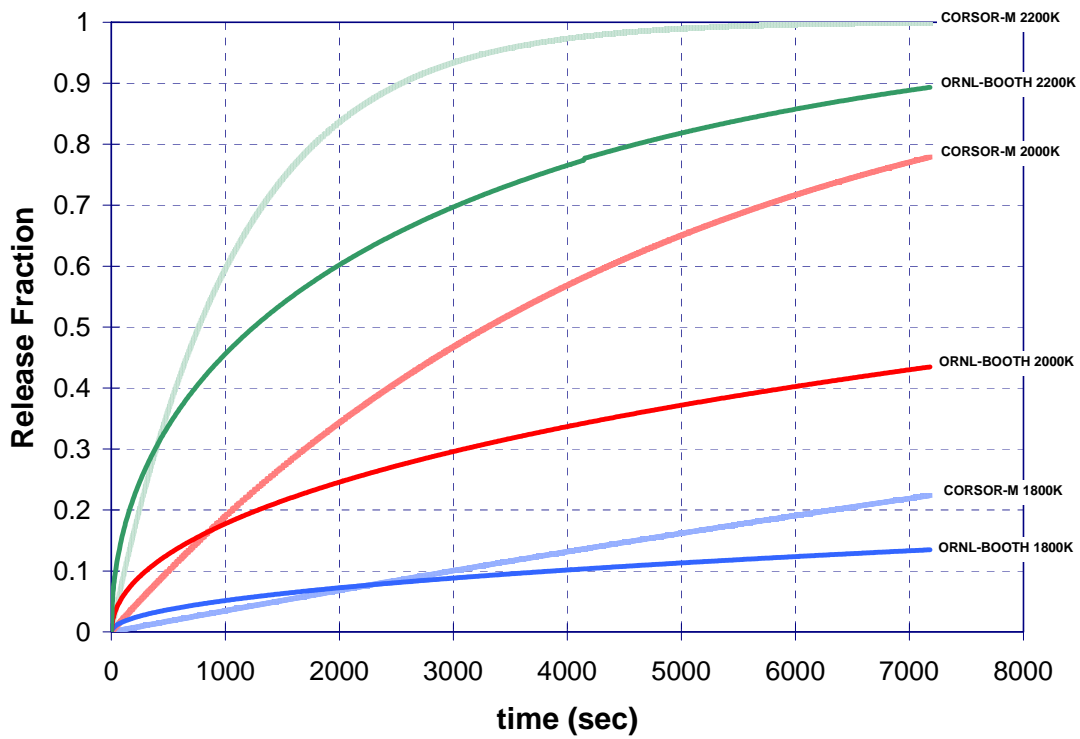


Figure 2. Release fractions at constant temperature for the ORNL-Booth and ORSOR-M models.

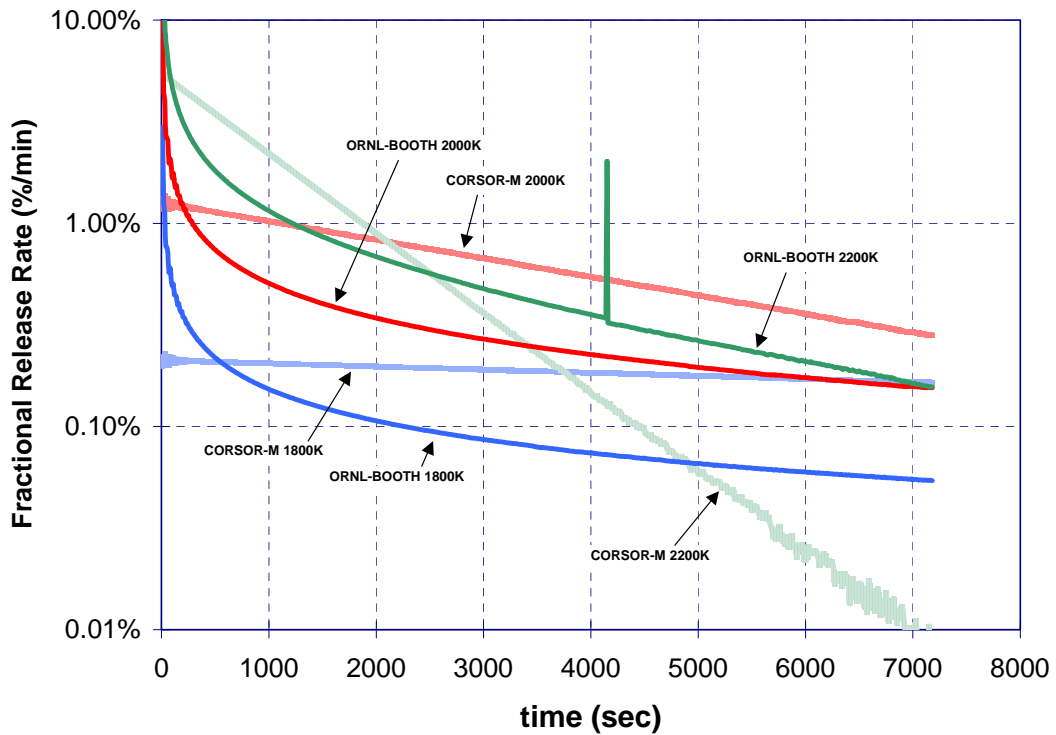


Figure 3. The time derivative of the release fraction.

While significant improvements in release behavior were obtained for the analysis of the FPT-1 test with the as-reported ORNL-Booth parameters, some additional modification to the MELCOR release model was pursued. Evidence from the Phebus experiments increasingly indicates that the dominant chemical form of released Cs is Cs_2MoO_4 . This is based on deposition patterns in the Phebus experiment where Cs is judged to be in aerosol form at 700C. Aerosol deposition is used to explain cesium deposits in the hot upper plenum of the Phebus test section, and deposition patterns of cesium in the cooler steam generator tubes. In recognition of this, the vapor pressure of both Cs and Mo classes were defined to be that of Cs_2MoO_4 . While having little effect on the net release of Cs, this change had a significant effect on the release of Mo. In MELCOR, by default the Mo vapor pressure is so exceedingly low that the net release is limited by the vapor transport term, as expressed in Eq. 7 and Eq. 8. Vapor pressures for selected fission product species are shown in Figure 4. Defining the Mo vapor pressure to be that of Cs_2MoO_4 produced significantly improved predictions of Mo release rate observed in the FPT-1 test, as will be shown in the next section of this report.

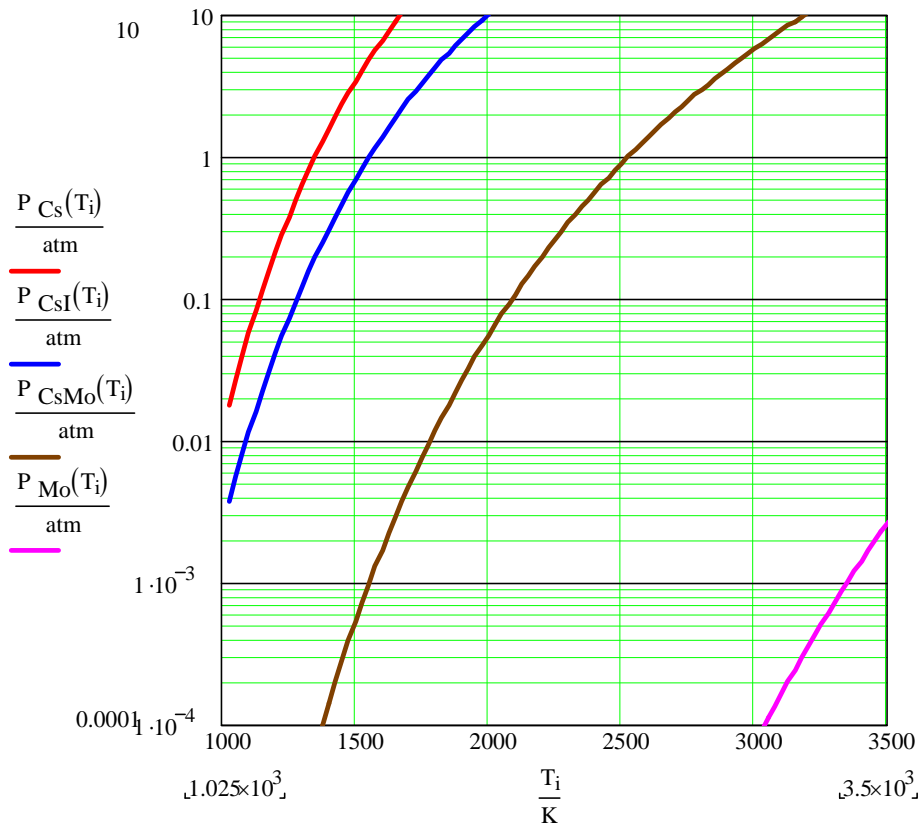


Figure 4. Vapor pressures of selected species.

3.2 Assessment of Modified ORNL-Booth Model Against Phebus FPT-1

The Phebus program provides the best source of prototypic data on fission product release from irradiated fuel. The Phebus tests benefitted from many lessons learned from earlier, similar, experimental efforts and from advances in testing technology, instrumentation, etc. A schematic of the Phebus test facility is shown in Figure 5. A previously irradiated fuel bundle of about a meter in length is situated in the irradiation cavity in the Phebus test reactor. The irradiated fuel undergoes severe damage from nuclear heating and oxidation by injected steam. Fission products released from the test bundle flow through a heated section representing the reactor coolant system (RCS), through a simulated steam generator tube where extensive deposition of fission products can occur, and into a simulated containment where fission product fallout occurs.

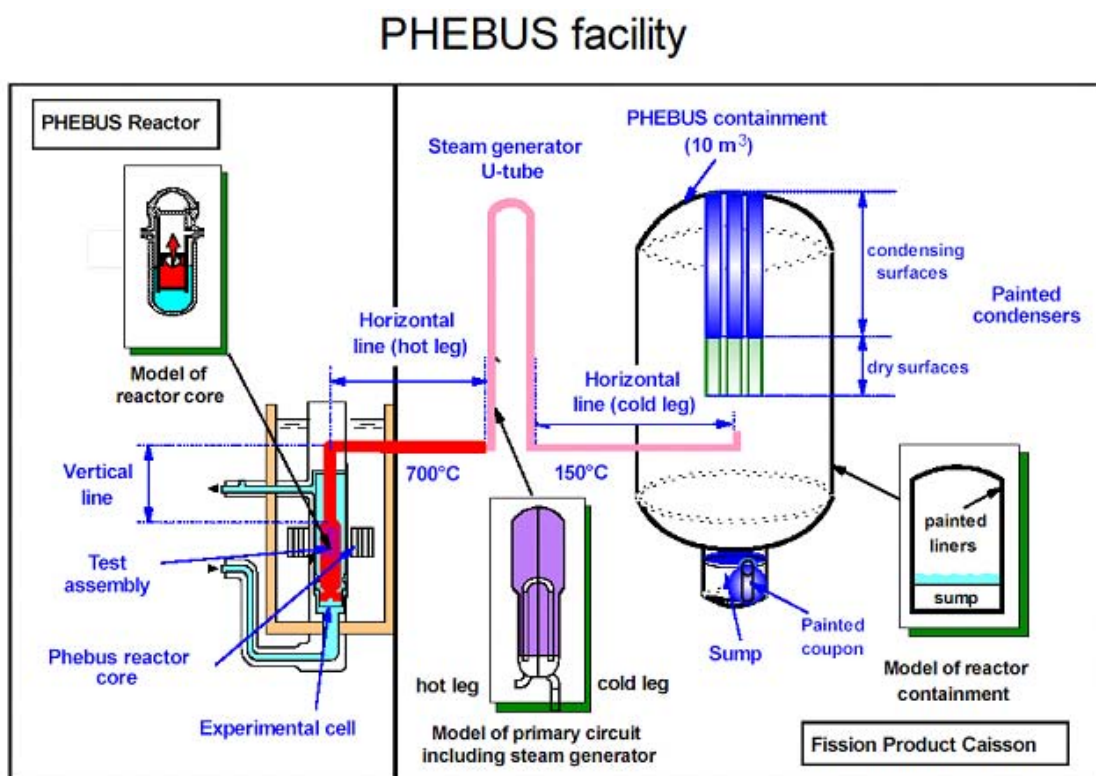


Figure 5. Schematic of the Phebus test facility showing test fuel bundle, heated lines, steam generator tube and simulated containment.

Shown in Figure 6 is the nuclear heating history that was used in test FPT-1 to heat the bundle to simulate severe accident decay heating conditions. The chemical heating produced by steam-Zr oxidation is also shown in the figure. The temperature response of the test fuel is shown in Figure 7 where the temperature transient caused by the additional oxidation heating is clearly evident. During this time, fission products are also released where oxidation conditions vary from oxidizing to more reducing, depending on elevation in the test bundle. Figure 8 shows the end state of the test bundle at the conclusion of the experiment.

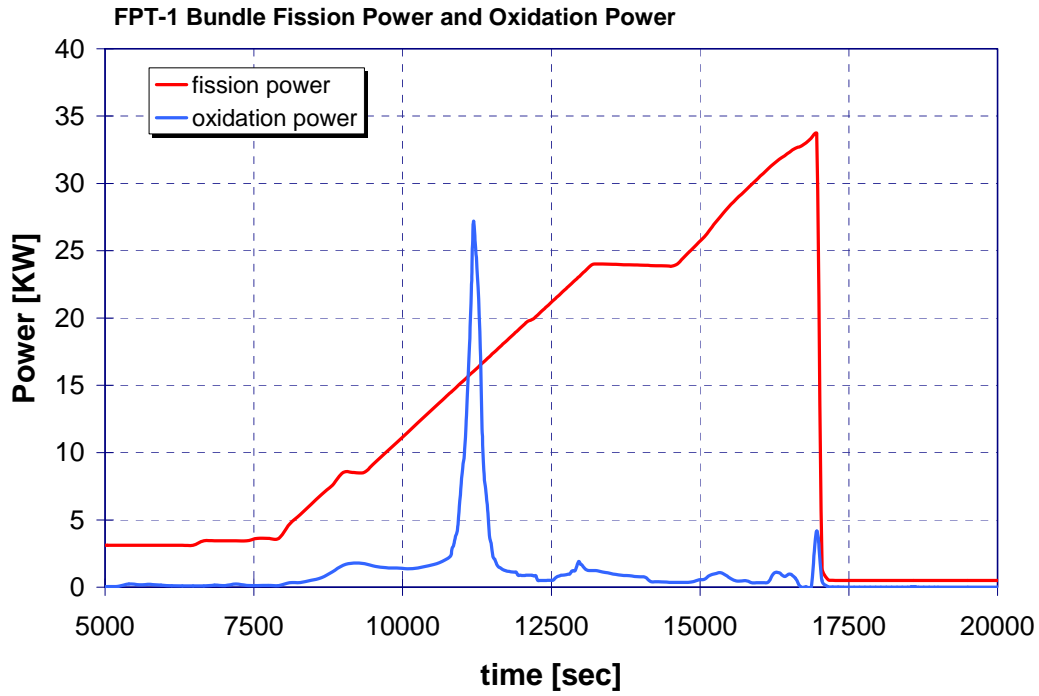


Figure 6. FPT-1 Nuclear and chemical heating history.

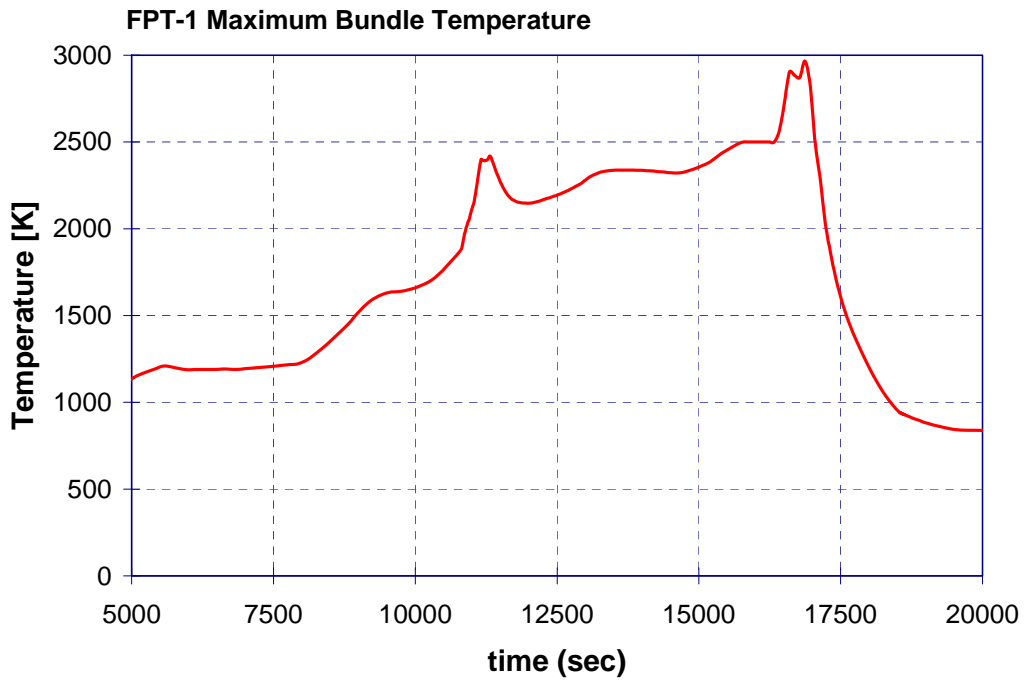


Figure 7. FPT-1 maximum bundle temperature history.

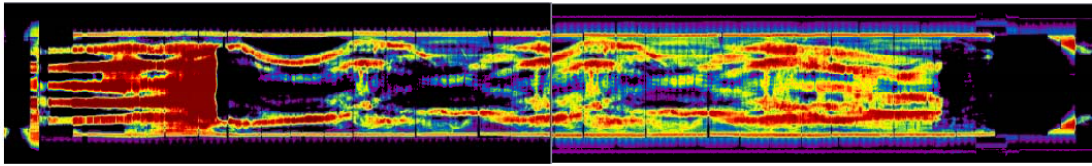


Figure 8. Emission gamma tomography of the end state condition of the test FPT-1 fuel bundle.

Figures 9 through 20 show the results of using the modified ORNL-Booth model to predict fission product release in the FPT-1 test. In most cases, significant improvement is realized in both the early release rate as well as for total predicted released. Where available, Phebus data are shown in the Figures. The release for the Barium class predicted by the ORNL-Booth model is low relative to the data, whereas the release predicted using the CORSOR-M model is high. Improvements to the predictions of this release proved illusive and it is believed that some adjustments to the vapor pressure for barium to account for some not yet identified barium species could produce some improvement. Adjustments to both vapor pressure and scaling factors were rationalized for Mo release based on Phebus program findings, producing good agreement with experiment. The Ru vapor pressure was increased by a factor of 10 to account for some greater volatility attributed to formation of oxides under moderately oxidizing conditions, and the Booth scaling factor was adjusted to get agreement with experimental observations. The Booth scaling factor for UO_2 was increased significantly in order to get agreement with test observations. This also is rationalized as due to effects of fuel oxidation and greater volatility of fuel oxides. Ce and La release parameters were not adjusted owing to lack of experimental basis, however, one could reason that their releases ought to roughly follow UO_2 release if fuel matrix stripping follows from fuel volatilization.

Figure 9. Comparison of ORNL-Booth versus CORSOR-M for Xe release (Class 1)

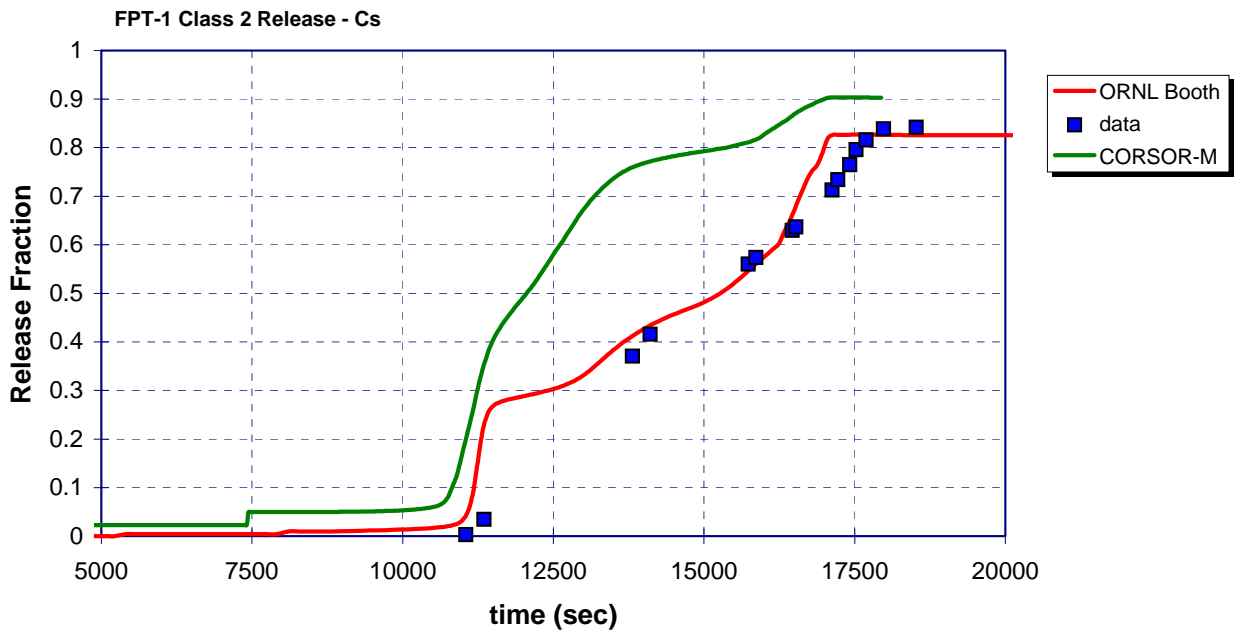


Figure 10. Comparison of ORNL-Booth versus CORSOR-M for Cs release (Class 2).

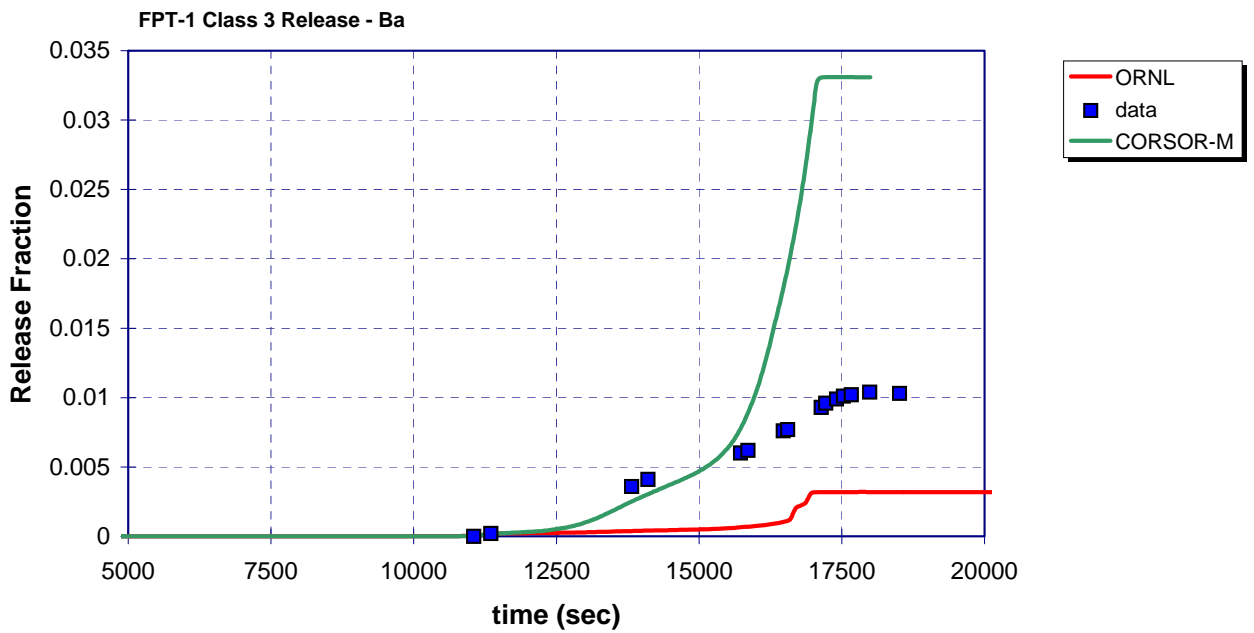


Figure 11. Comparison of ORNL-Booth versus CORSOR-M for Ba release (Class 3).

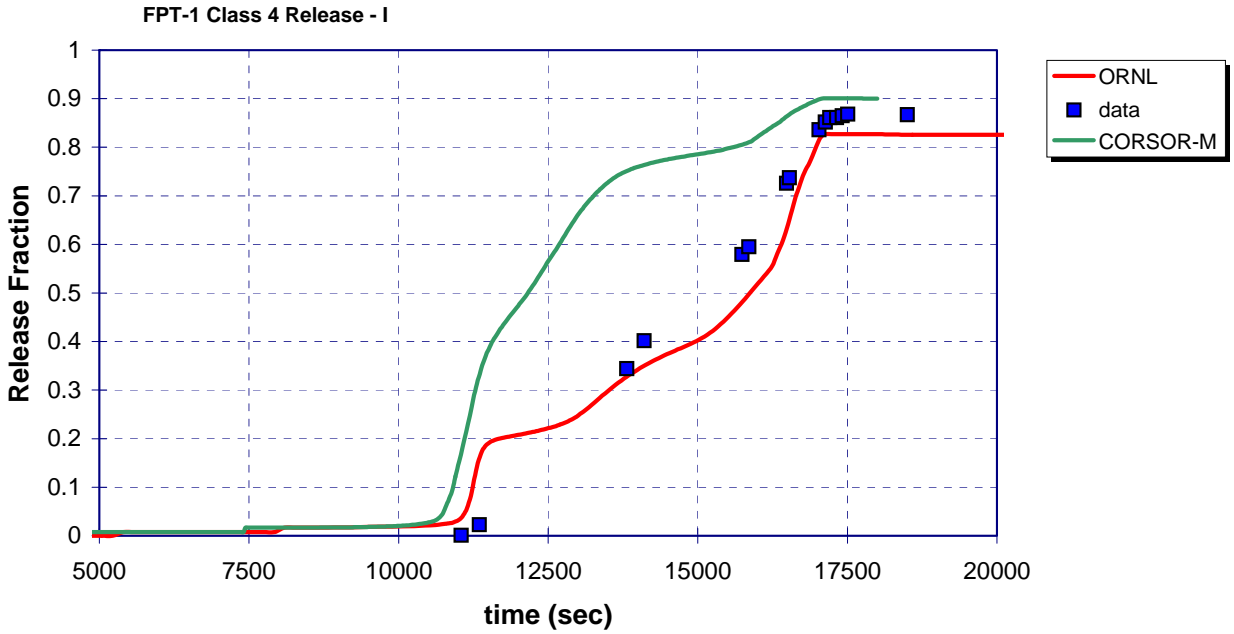


Figure 12. Comparison of ORNL-Booth versus CORSOR-M for I release (Class 4).

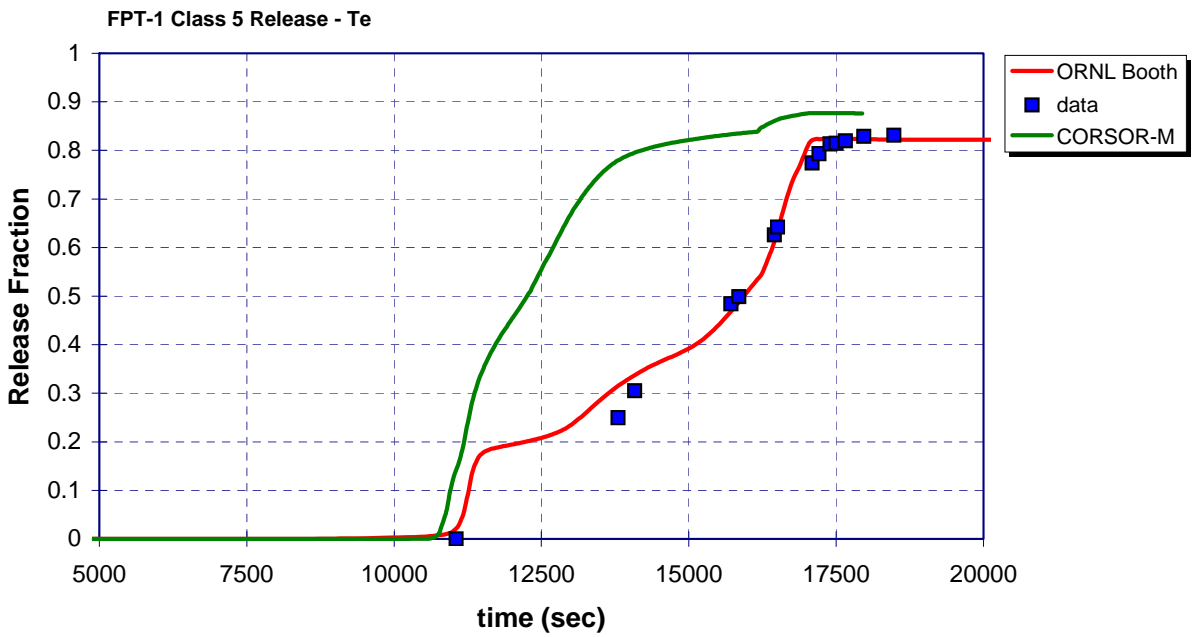


Figure 13. Comparison of ORNL-Booth versus CORSOR-M for Te release (Class 5)

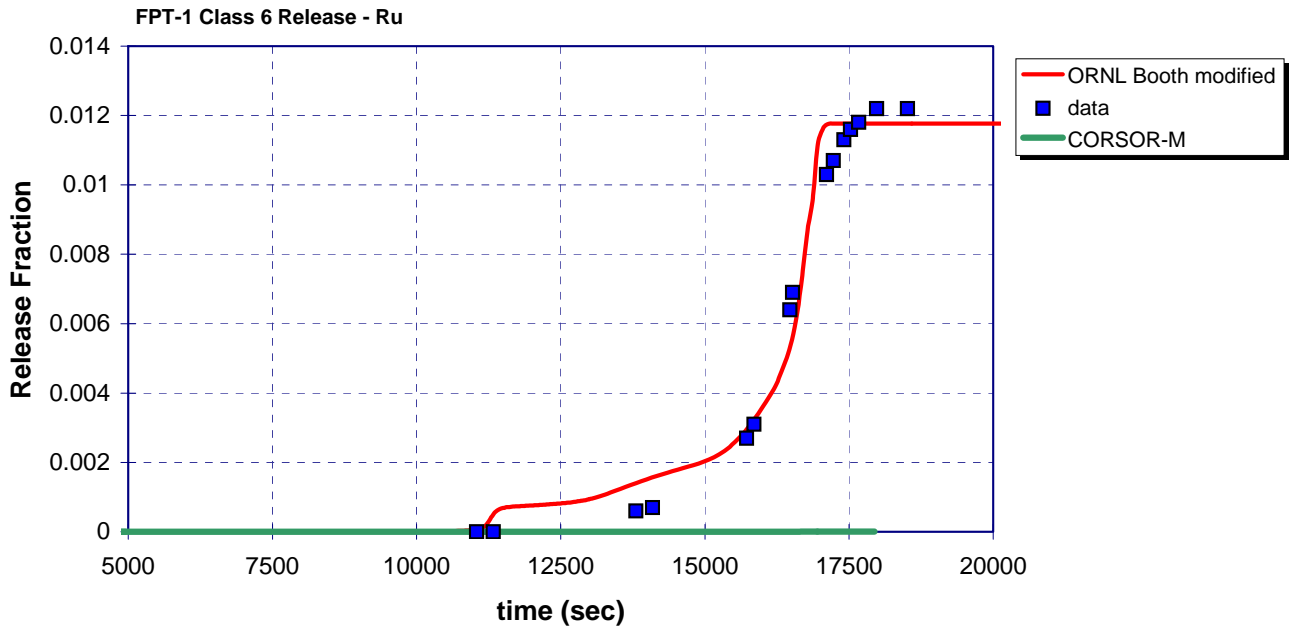


Figure 14. Comparison of ORNL-Booth versus CORSOR-M for Ru release (Class 6)

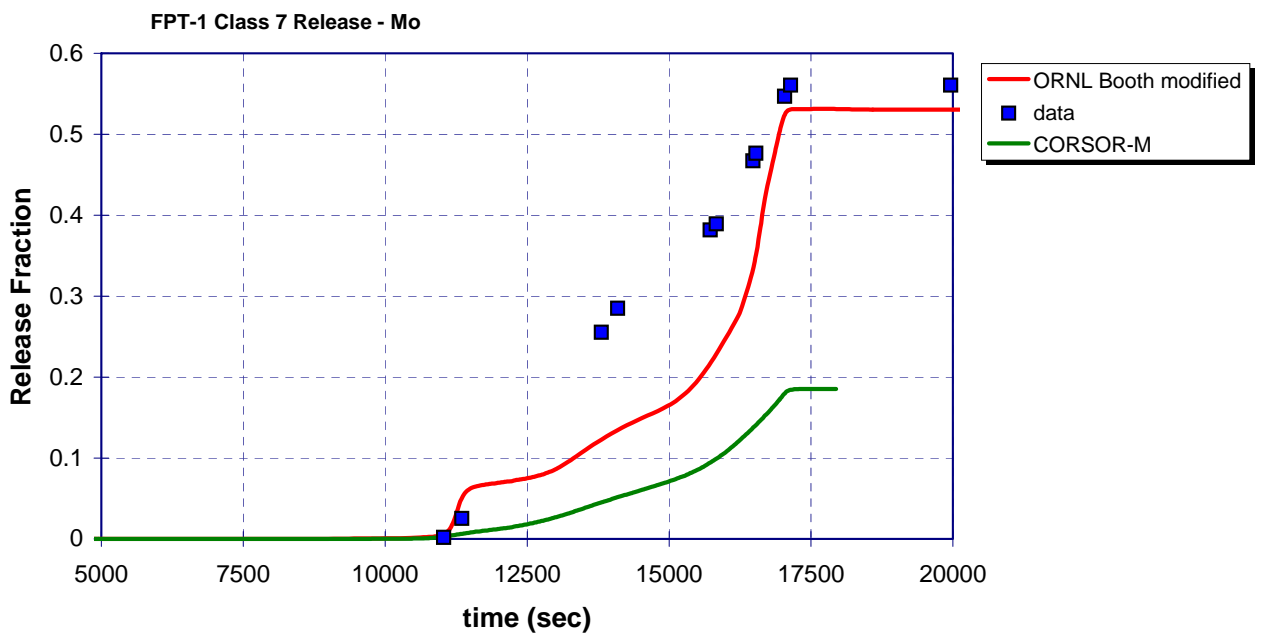


Figure 15. Comparison of ORNL-Booth versus CORSOR-M for Mo release (Class 7). The Mo vapor pressure was set to correspond to Cs_2MoO_4 .

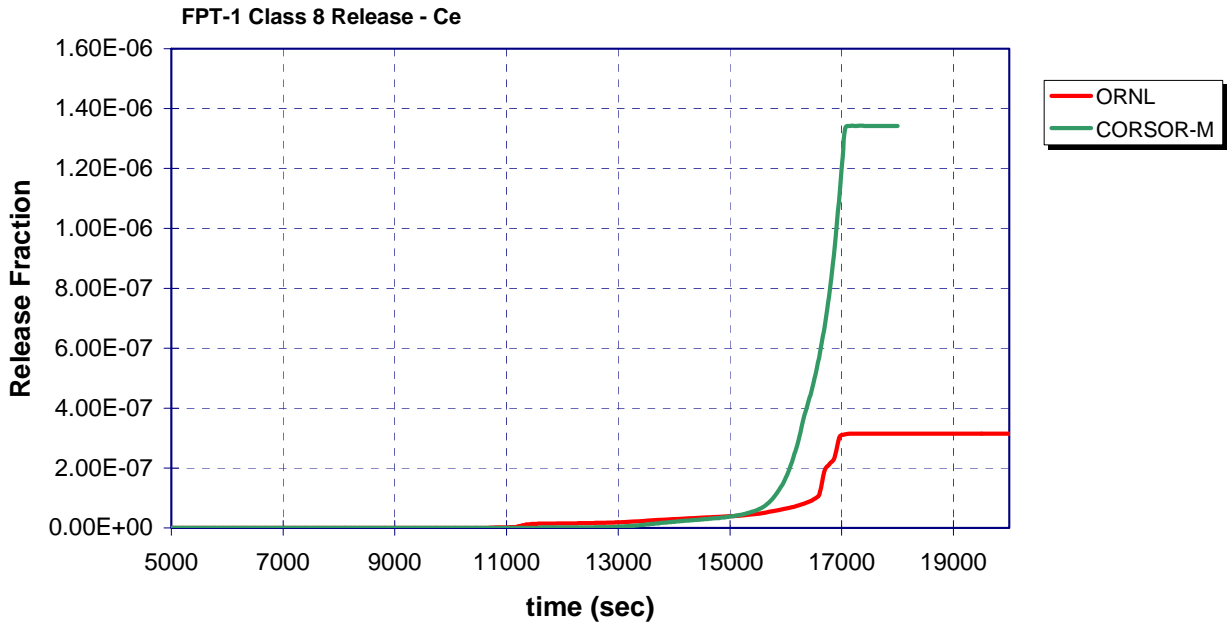


Figure 16. Comparison of ORNL-Booth versus CORSOR-M for Ce release (Class 8).

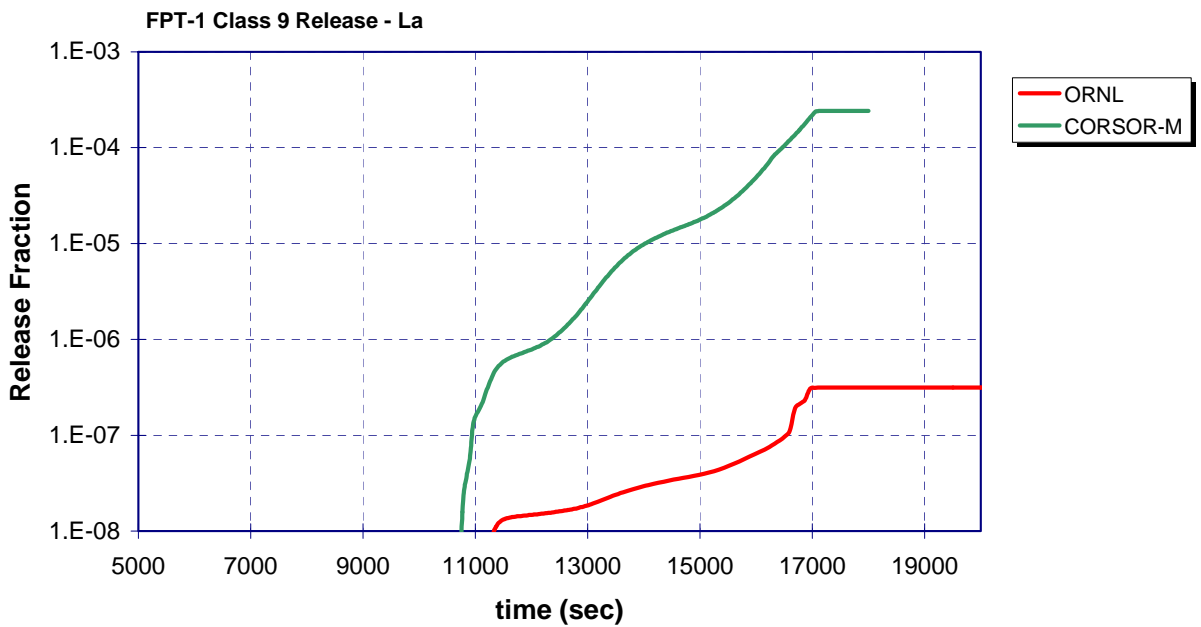


Figure 17. Comparison of ORNL-Booth versus CORSOR-M for La release (Class 9).

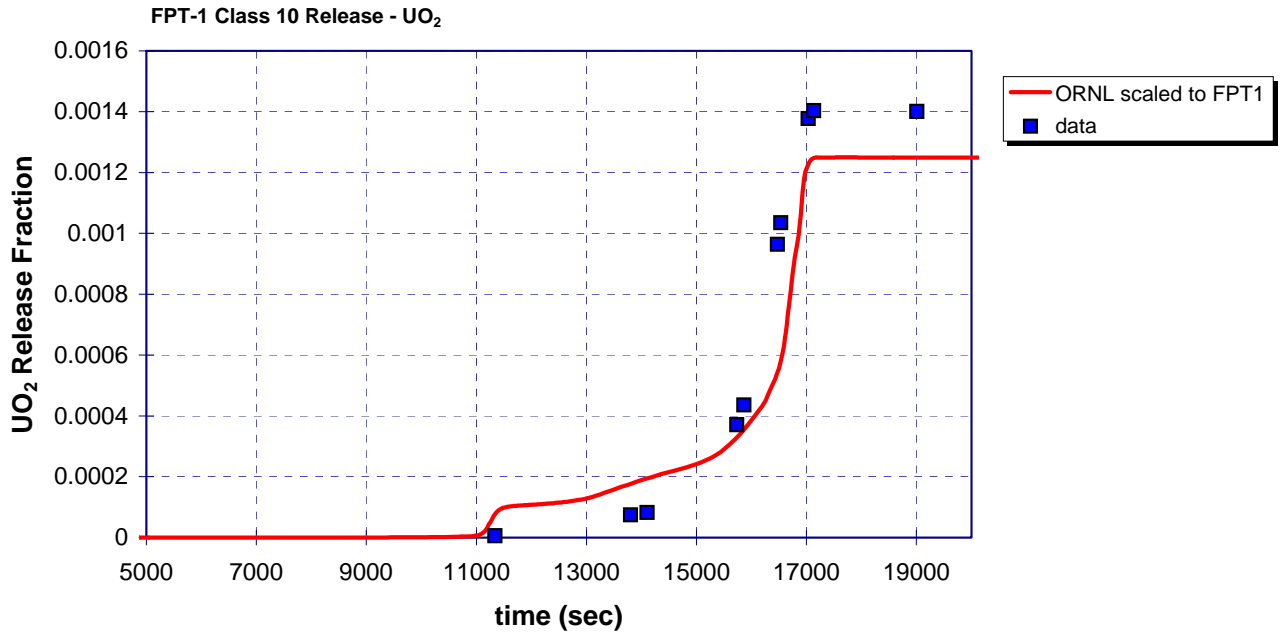


Figure 18. Comparison of ORNL-Booth versus CORSOR-M for UO₂ release (Class 10). *The UO₂ scaling factor was adjusted to match observed releases. La and Ce releases are not expected to be greater than UO₂ release, but may be less owing to lower volatility.*

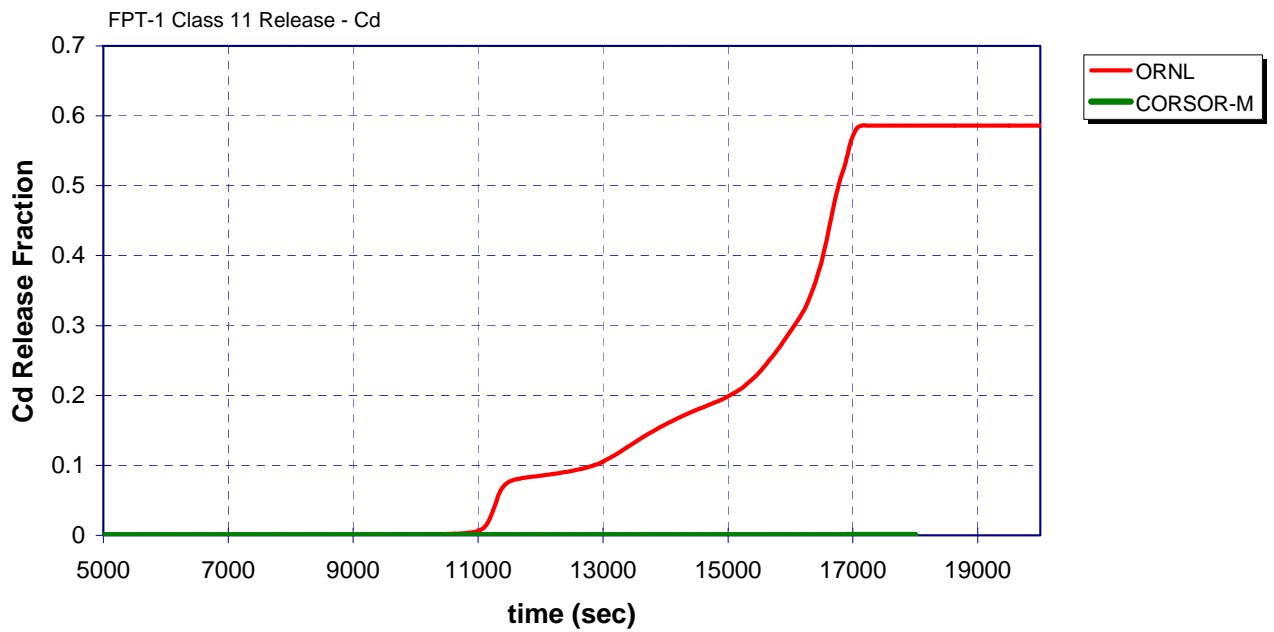


Figure 19. Comparison of ORNL-Booth versus CORSOR-M for Cd release (Class 11).

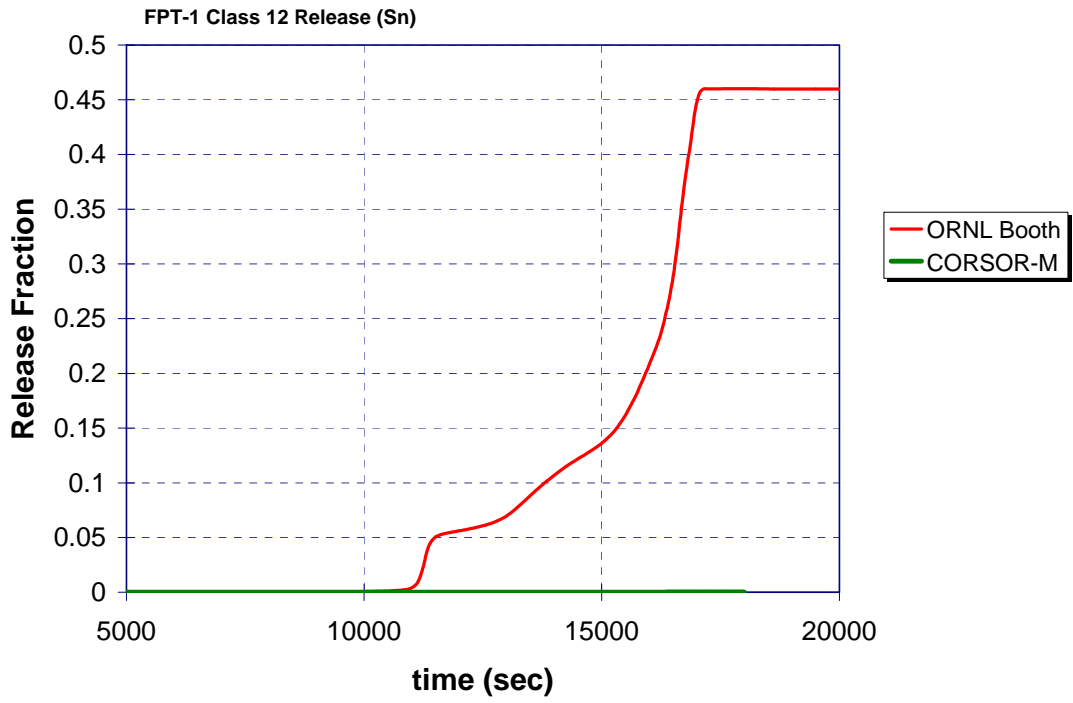


Figure 20. Comparison of ORNL-Booth versus CORSOR-M for Sn release (Class 12).

3.3 Comparison to ORNL VI Tests and VERCOR Tests [12]

After optimizing the ORNL-Booth fission product release parameters for the FPT-1 experiment, it was of interest to compare the modified model to the original ORNL test data upon which the CORSOR-M models was based. The following section explores the application of the modified ORNL-Booth release modeling to selected ORNL-VI test results as well as to the more recent VERCORS test data. The comparisons are made mainly to the Cs release observed in these experiments since all other releases are modeled in the MELCOR implementation of the Booth model by simply scaling to the Cs release. Cesium release data are available for all the tests. In the case of VERCORS 4 test, data on release of other fission products were available and comparisons of model predictions to these releases are included. The MELCOR models were obtained from a recent IBRAE MELCOR Validation exercise [9] investigating the MELCOR default release models. The experimental data are taken from reference [9]. These analyses were performed using a simple MELCOR model of these experiments. The present analyses make use of the modified ORNL-coefficients and compare results with the MELCOR default CORSOR-M release model.

A schematic of the VERCORS testing facility is shown in Figure 21, the general layout is similar in the ORNL VI tests. The tests examined are summarized in Table 2. The tests involved both oxidizing and reducing conditions.

G. Ducros et al. / Nuclear Engineering and Design 208 (2001) 191–203

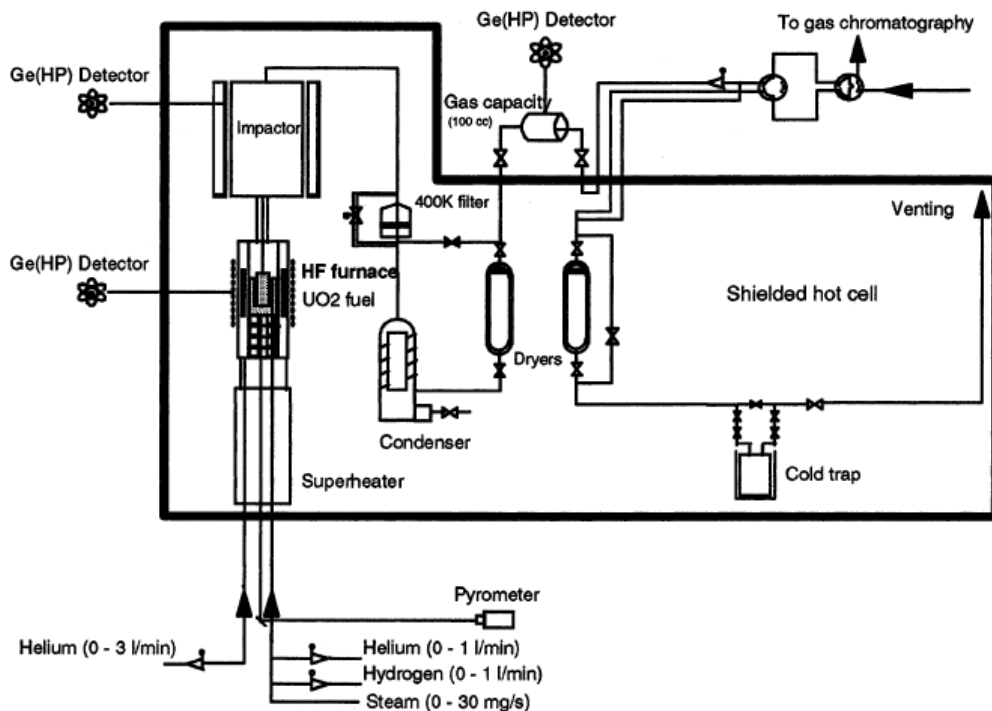


Figure 21. Schematic of VERCORS test facility for measuring fission product release from small fuel samples.

Table 2. Conditions for selected ORNL VI tests and VERCORS tests.

Test	Hydrogen	Steam	Max Temperature
ORNL VI-2	0	1.8 liter/min	2300K
ORNL VI-3	0	1.6 liter/min	2700K
ORNL VI-5	0.4 liter/min	0	2740K
VERCORS 2	0.027 gm/min	1.5 gm/min	2150K
VERCORS 4	0.012 gm/min	1.5 – 0 gm/min	2573K

In almost all cases, the modified ORNL-Booth model yields improved predictions, as shown in Figure 22 through Figure 24 for the VI tests, and in Figure 25 through Figure 31 for the VERCORS tests.

In test VI-2, which was run under steam conditions, the peak temperature attained was ~2300K. Both models overpredicted the Cs release for this test, with the modified ORNL-Booth treatment performing slightly better (Figure 22). Test VI-3 was similar to VI-2 except that higher temperatures were attained. For this test, predictions of both models are closer to the data, and again the modified ORNL-Booth model performs somewhat better (Figure 23). From these two tests, it appears that release rates in the 2300K range are still slightly over-predicted for oxidizing conditions. Results of Test VI-5, which was conducted under reducing conditions, were well predicted by both models, as shown in Figure 24. Table 3 through Table 5 provides total releases predicted by CORSOR-M and ORNL-Booth compared with totals reported for the ORNL VI tests 2, 3 and 5.

Both VERCORS tests 2 and 5 were run in mixed conditions with both steam and hydrogen. In VERCORS 5, the steam flow was reduced to zero in an attempt to produce reducing conditions at the high temperature plateau. Test VERCORS 2, like test ORNL-VI2 was performed at a lower temperature and produced a comparatively lower Cs release (Figure 25). The modified ORNL-Booth model captured this lower release whereas the CORSOR-M model did not. Test VERCORS 4 was performed under completely reducing conditions during the release phase. In this case, CORSOR-M under-predicted release, whereas the modified ORNL-Booth model captured the release behavior reasonably well.

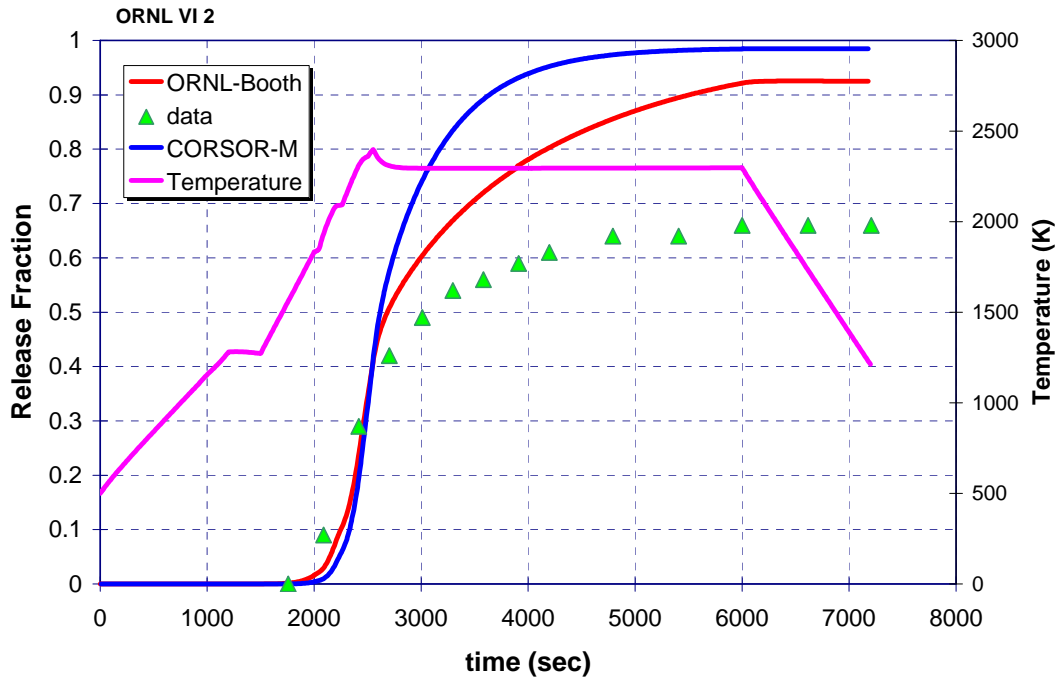


Figure 22. Comparison of Cs release for ORNL Booth modified with CORSOR-M for VI-2 run under steam oxidizing conditions.

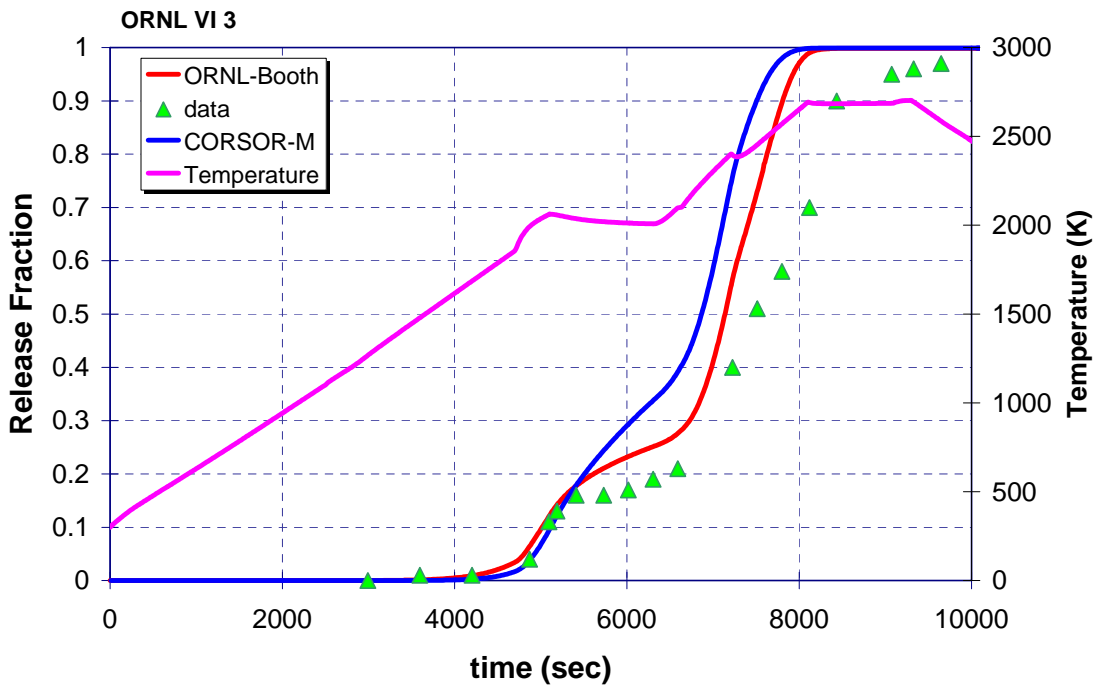


Figure 23. Comparison of Cs release for ORNL Booth modified with CORSOR-M for VI-3 performed under steam oxidizing conditions.

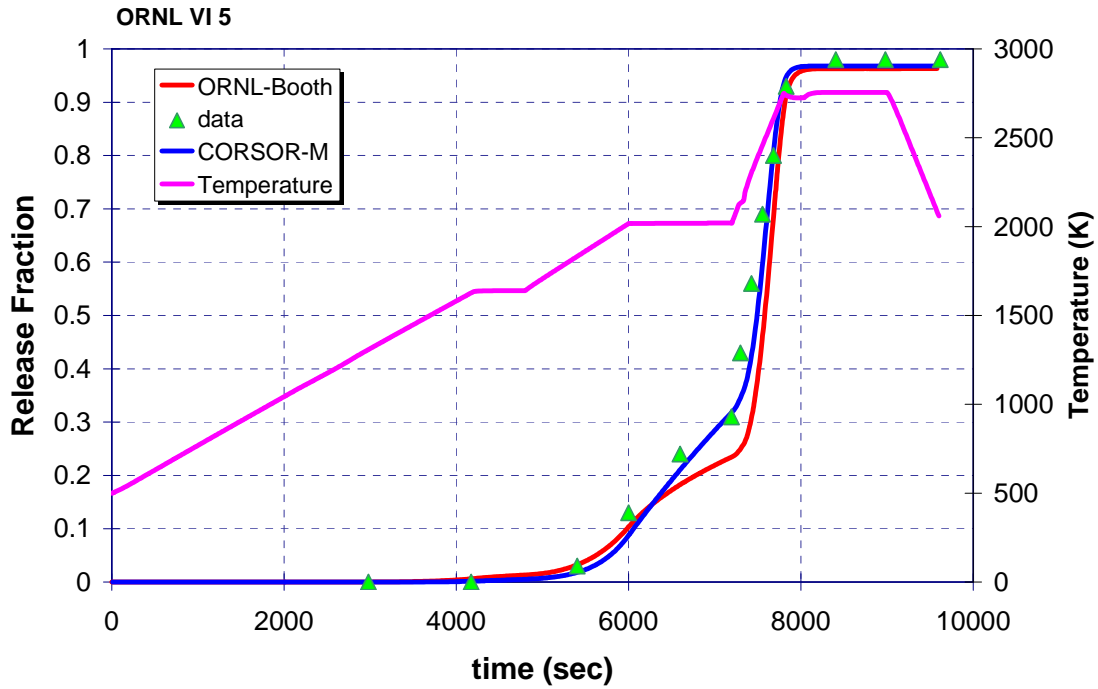


Figure 24. Comparison of Cs release for ORNL Booth modified with CORSOR-M for VI-5 performed under reducing conditions.

Table 3. Total release from ORNL VI-2.

	Experiment	CORSOR-M	ORNL-Booth
Kr	*	.98	.92
Cs	.67	.98	.92
Ba	.18	.003	0.002
Sr		.003	0.002
I	.4	.98	.81
Te		.97	.81
Ru		1×10^{-7}	0.006
Mo	.86	.06	0.42
Ce		1×10^{-8}	1.1×10^{-7}
Eu		1×10^{-5}	1.1×10^{-7}
U	.003	1×10^{-5}	0.001
Sb	.68	0.04	0.93

Table 4. Total release from ORNL VI-3.

	Experiment	CORSOR-M	ORNL-Booth
Kr	1	1	1
Cs	1	1	1
Ba	.3	.04	0.004
Sr	.03	.04	0.004
I	.8	1	1
Te	.99	1	0.99
Ru	.05	10^{-5}	0.03
Mo	.77	0.15	0.88
Ce	0	2×10^{-6}	4×10^{-7}
Eu	0	0.0005	4×10^{-7}
U	0	0.0005	0.003
Sb	.99	0.2	0.93

Table 5. Total release from ORNL VI-5.

	Experiment	CORSOR-M	ORNL-Booth
Kr	1	.97	.96
Cs	1	.97	.96
Ba	.76	.04	0.005
Sr	.34	.04	0.005
I	.7	.97	.96
Te	.82	.95	0.96
Ru	0	10^{-5}	0.03
Mo	.02	0.11	0.85
Ce	.02	3×10^{-6}	4×10^{-7}
Eu	.57	0.0008	4×10^{-7}
U	0	0.0008	0.003
Sb	.18	0.19	0.89

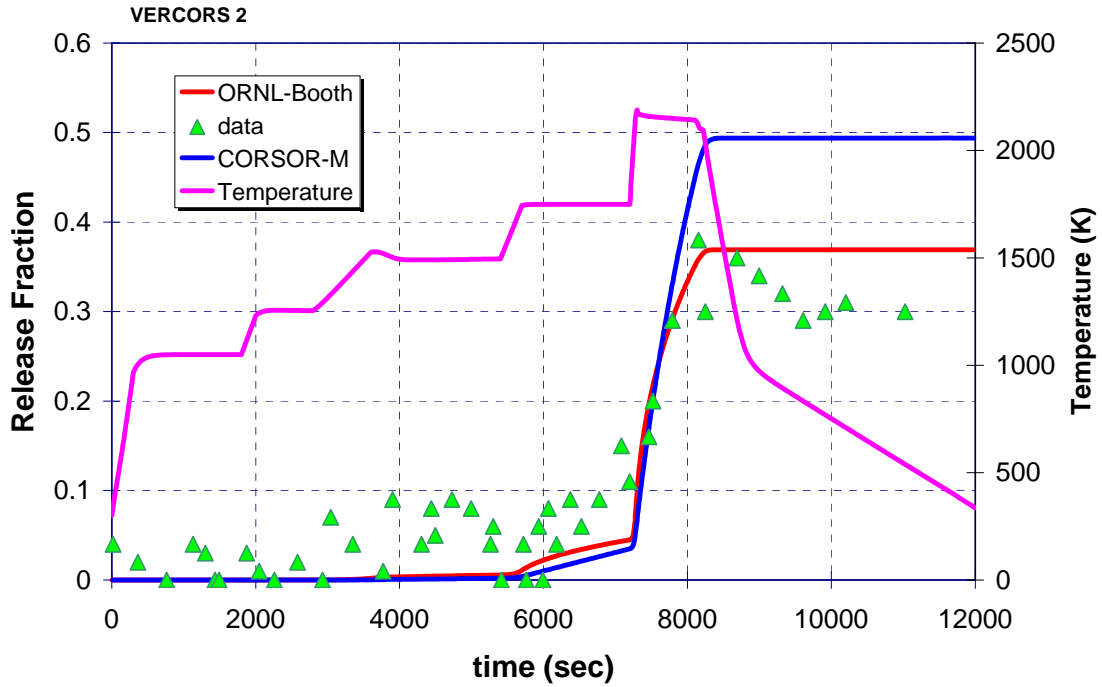


Figure 25. Comparison of Cs release for ORNL Booth modified with CORSOR-M for VERCORS-2.

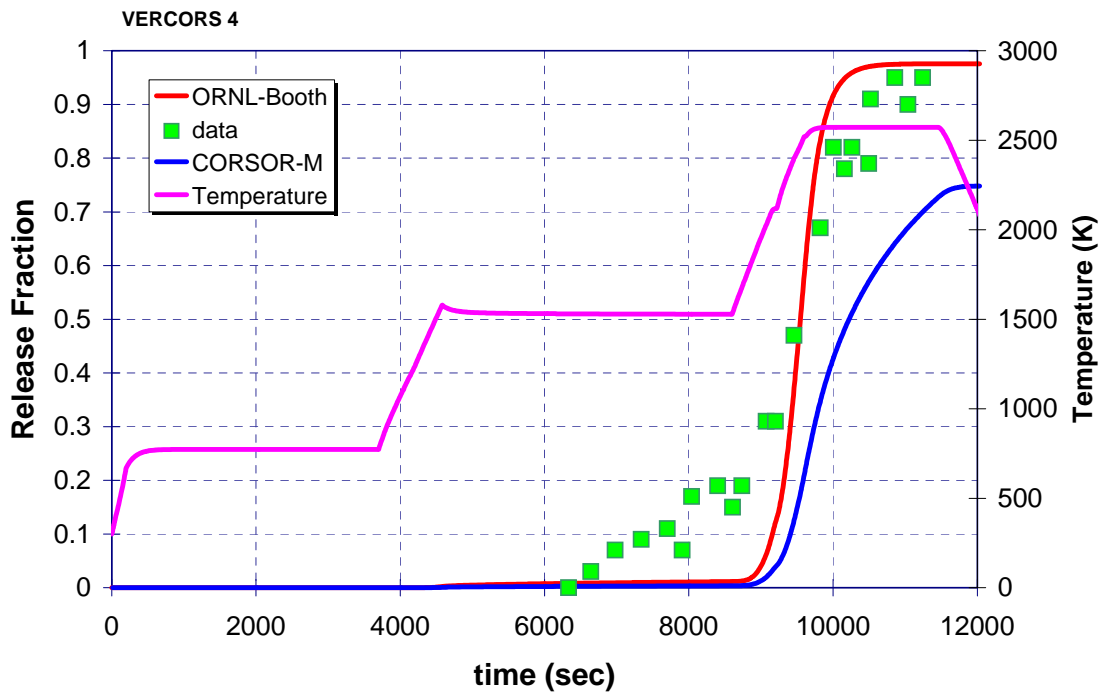


Figure 26. Comparison of Cs release for ORNL Booth modified with CORSOR-M for VERCORS-4.

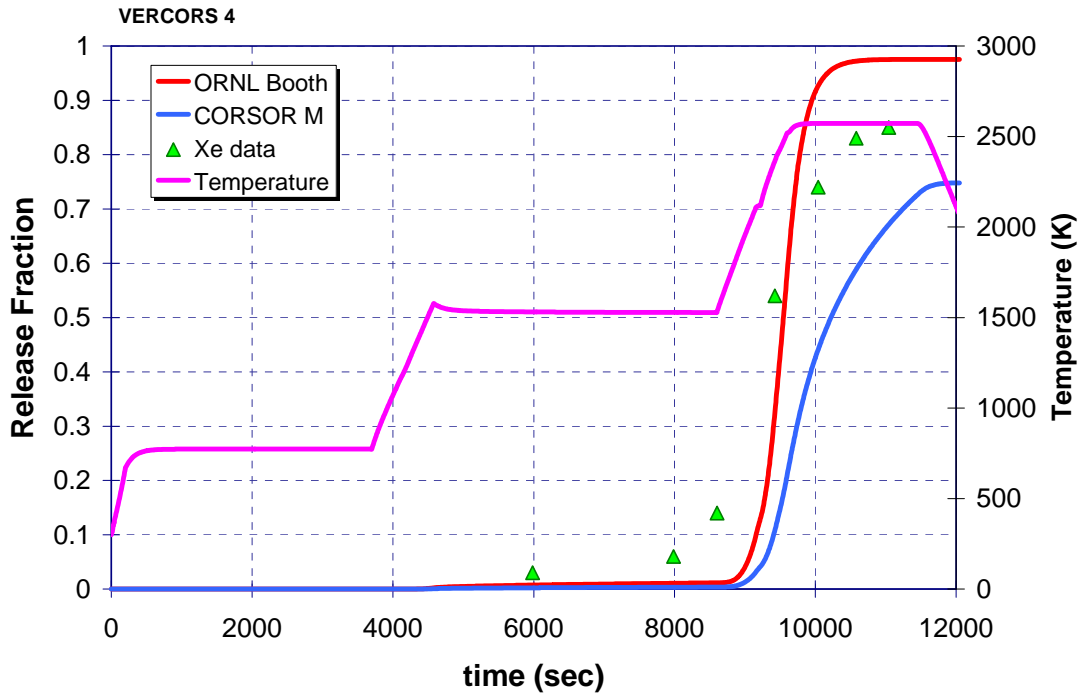


Figure 27. Comparison of Xe release for ORNL Booth modified with CORSOR-M for VERCORS-4.

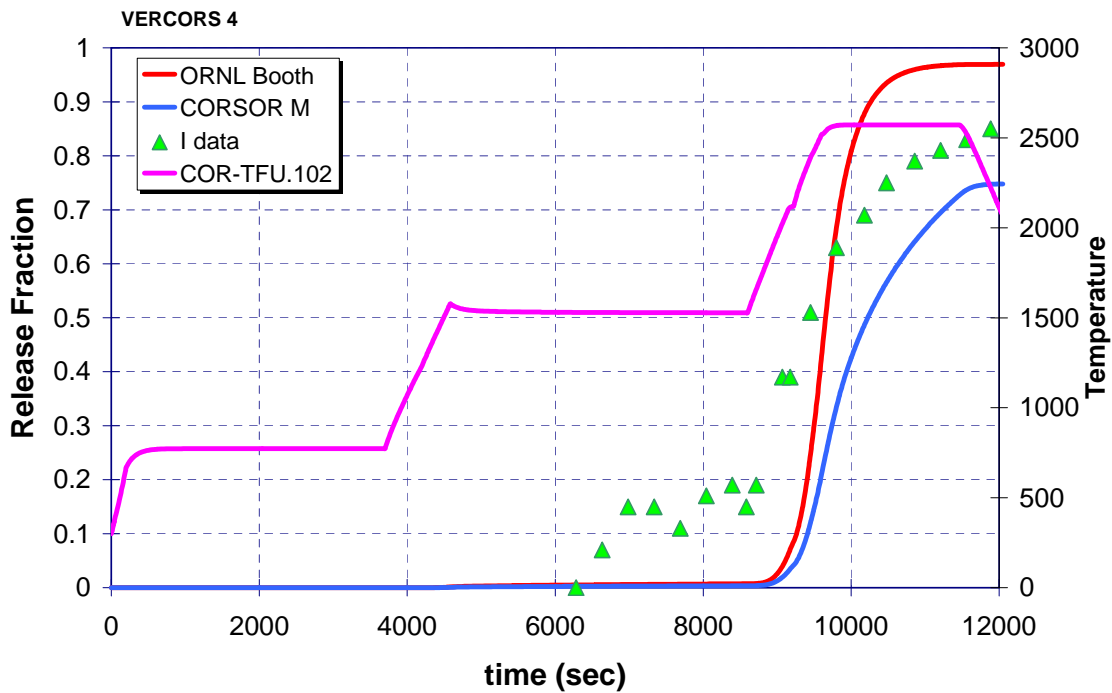


Figure 28. Comparison of iodine release for ORNL Booth modified with CORSOR-M for VERCORS-4.

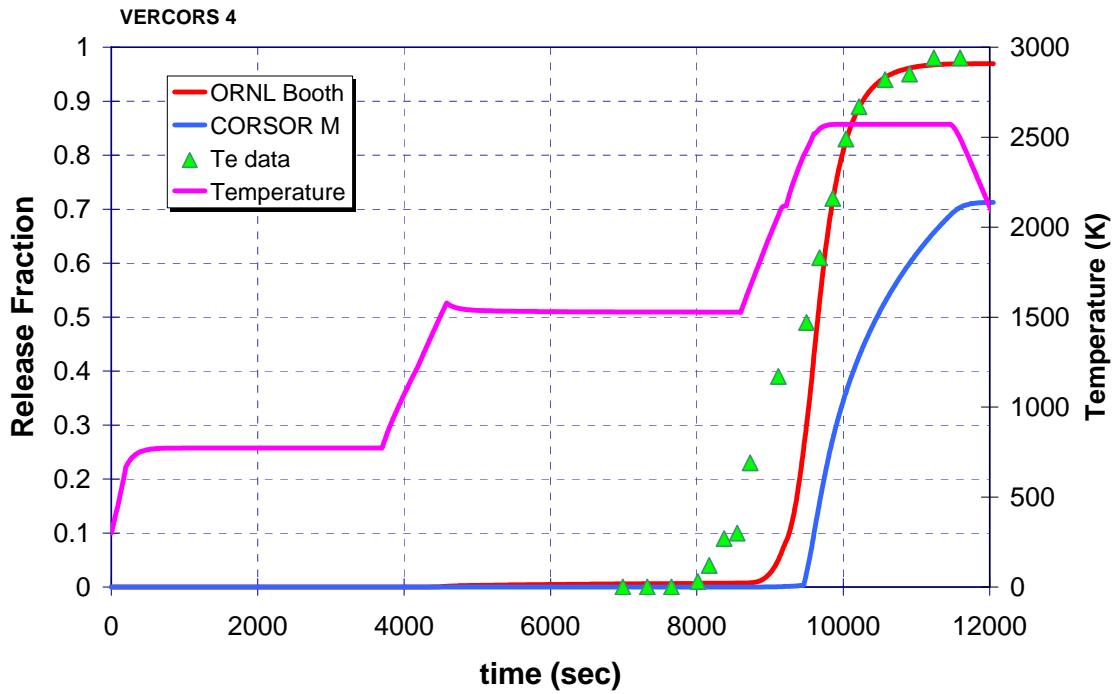


Figure 29. Comparison of Te release for ORNL Booth modified with CORSOR-M for VERCORS-4.

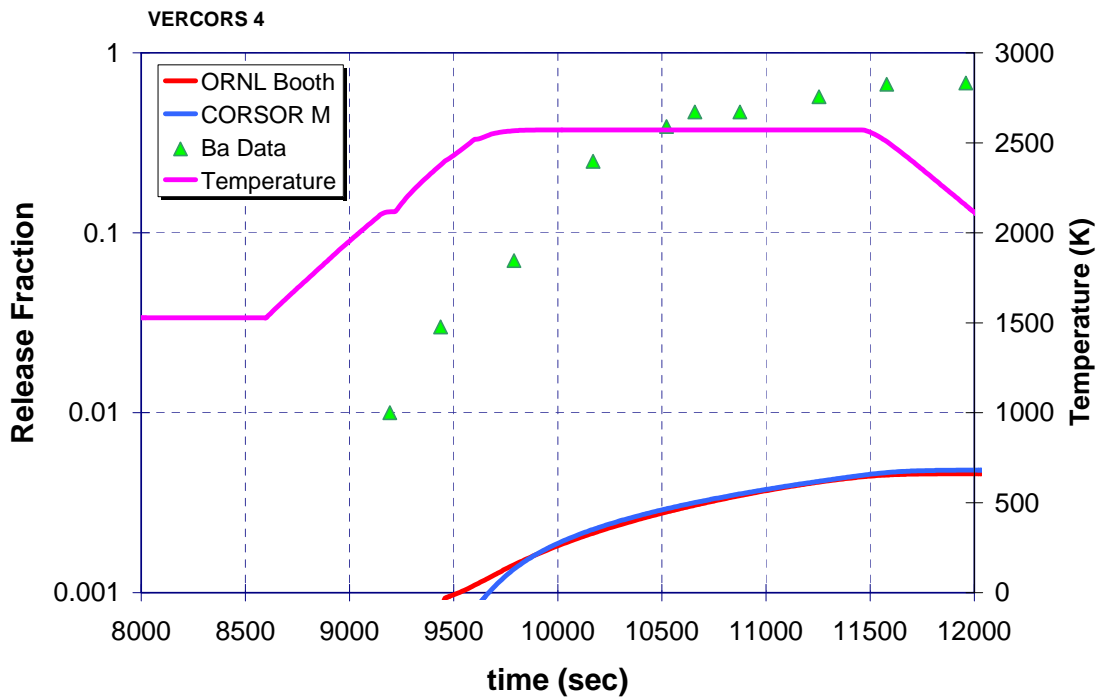


Figure 30. Comparison of Ba release for ORNL Booth modified with CORSOR-M for VERCORS-4.

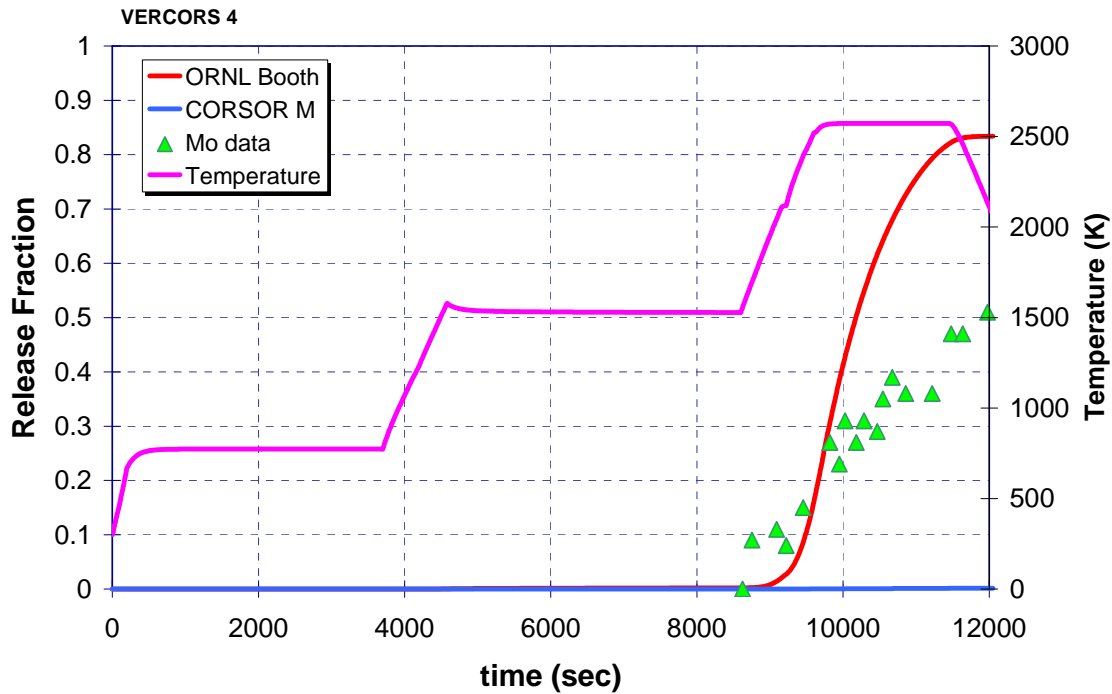


Figure 31. Comparison of Mo release for ORNL Booth modified with CORSOR-M for VERCORS-4.

On balance, the use of the modified ORNL-Booth model produces significantly improved predictions for both the in-pile Phebus FPT-1 test as well as for the original small scale ORNL VI and more recent VERCORS tests. Prediction of barium behavior however remains somewhat problematic. Barium releases observed in small-scale tests are larger than observed in the in-pile, integral tests. It is thought that this is due to the more complete oxidation of cladding in the small scale tests, whereas considerably less coherent conditions are encountered in the in-pile integral tests. It is hypothesized that the Ba speciation in the small-scale tests leads to greater volatility than is possible in the in-pile tests where unoxidized Zr is plentiful.

4 Evaluation of Fission Product Deposition Modeling

4.1 Deposition in FPT-1 Circuit (RCS Deposition)

The modified ORNL-Booth release models have been shown to produce favorable release predictions when examining the Phebus FPT-1 test and produced good comparisons with the ORNL VI and French VERCORS tests. The modifications to the vapor pressures for Cs and Mo, which produced favorable release behavior in FPT-1, will have an effect on the subsequent deposition of these species in the RCS piping. The effect is illustrated in Figures 32 and 33 that show deposition patterns in the Phebus FPT-1 test circuit and model containment.

Figure 32 shows the predicted deposition distribution in the FPT-1 experiment when the default CORSOR-M release model was used. While the total Cs release compares reasonably well with the measured value, and the total Cs transported to the containment is about right, the distribution of Cs deposits in the heated test section above the fuel (upper plenum) and in the steam generator tube do not compare well with the test data. Deposits in the steam generator are overpredicted and deposits in the heated plenum above the fueled region are underpredicted. In fact, deposits of Cs in the plenum were never predicted to exceed about 0.1%. What was predicted to deposit was also predicted to be completely revaporized before the end of the test. Underpredicting deposition in the hot plenum region is a big factor in the over-predicting deposition in the steam generator tube.

Figure 33 shows the Cs distribution predicted for FPT-1 when the modified ORNL-Booth model is used. The lower vapor pressure of the presumed Cs_2MoO_4 results in the prediction that cesium will be in aerosol form in the hot upper plenum region. As a result, Cs deposited in the upper plenum remains for the duration of the test. This together with a slightly lower total Cs release results in half as much predicted to be deposited in the steam generator tubes, considerably closer to the observed deposition in the steam generator tube. The amount reaching the containment remains about the same, which from a “release to the environment” point of view, one can observe that either model retains about the right amount of fission product within the simulated RCS. The changes in Cs deposition within the RCS could of course alter the decay heat distributions throughout the RCS, which in turn could affect revolatilization of other more volatile deposited species, such as CsI, which is transported in addition to the presumed predominant species, Cs_2MoO_4 .

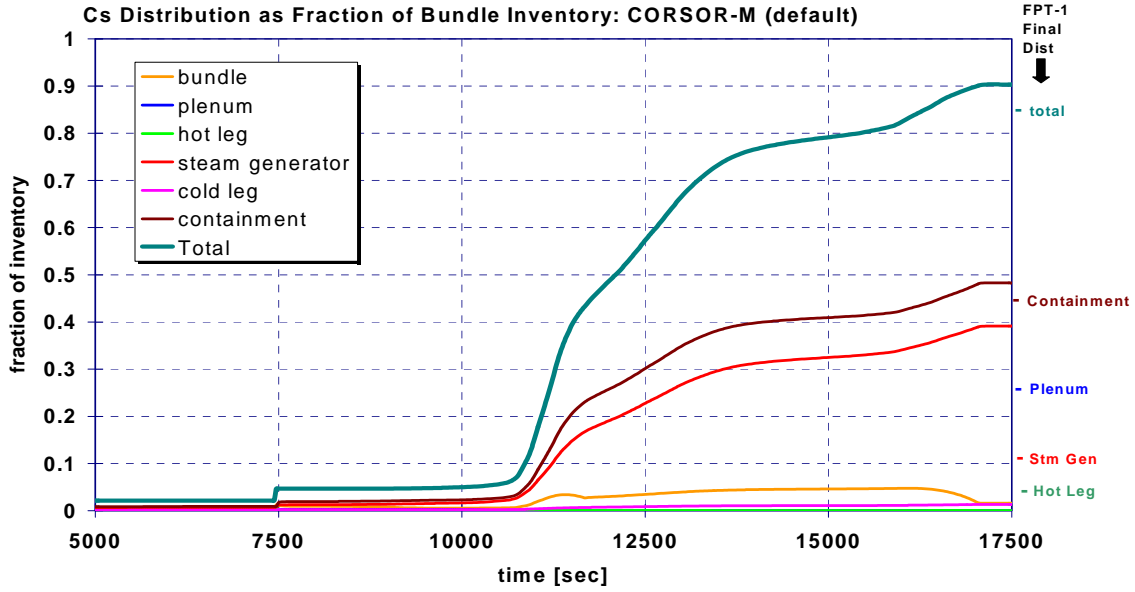


Figure 32. MELCOR-predicted fission product deposition in FPT-1 circuit using default CORSOR-M release modeling. Predicted plenum deposits for this case were less than 0.1%, not visible on this scale, and were subsequently revaporized.

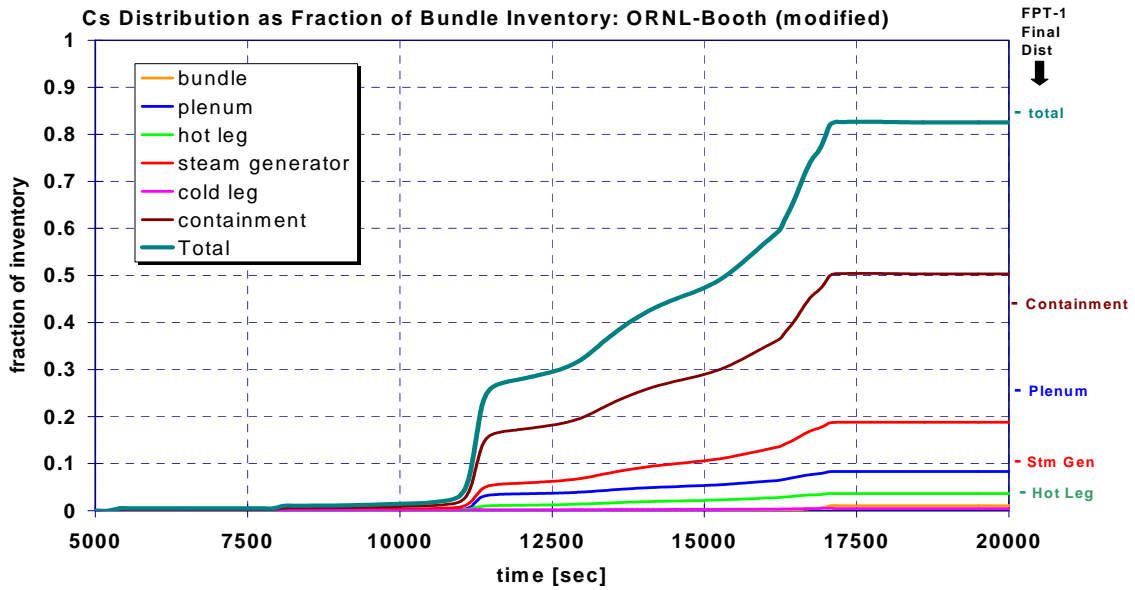


Figure 33. MELCOR-predicted fission product deposition in FPT-1 circuit using modified ORNL-Booth release modeling.

5 MOX and High Burnup

5.1 MELCOR Analysis of RT-2 MOX Experiment Using Fitted Booth Parameters

The VERCORS RT-2 test [13] was performed using mixed-oxide (MOX) fuel from the Gravelines nuclear power plant in France. The burnup was about 47.3 MWd/tonne. The objective of the VERCORS tests is to provide fission product release measurements for use in developing and validating models predicting release under severe accident or off-normal conditions. The RT-2 test was analogous to the RT-1 test, which was performed using low enrichment uranium (LEU) fuel. Both tests were performed under similar conditions with a mixture of steam and hydrogen (0.5:25 mg/s of H₂:H₂O) during fission product release up to temperatures nearing 2500K. Neither test RT-1 or RT-2 involved any re-irradiation of the test fuel samples prior to testing and, because of this, no data were obtained on the releases of iodine or other short-lived fission products. The test measurements focused on releases of krypton, cesium, ruthenium, cerium, and europium, the latter three of which are generally considered to be of low volatility in comparison to cesium and iodine. The principal measurement in test RT-2 was for the time-temperature release of cesium, the results of which are shown in Figure 34. A second MOX fission product release test was performed in the VERCORS program, test RT-7 [14]. This test was performed with release under pure reducing conditions. Since fission product release is expected to take place under conditions with both steam and hydrogen present, the RT-7 data are not considered to be as representative of in-vessel release conditions overall, and for this reason are not considered this study. Total releases for other isotopes measured in tests RT-1 (LEU) and RT-2 (MOX) are summarized in Table 6.

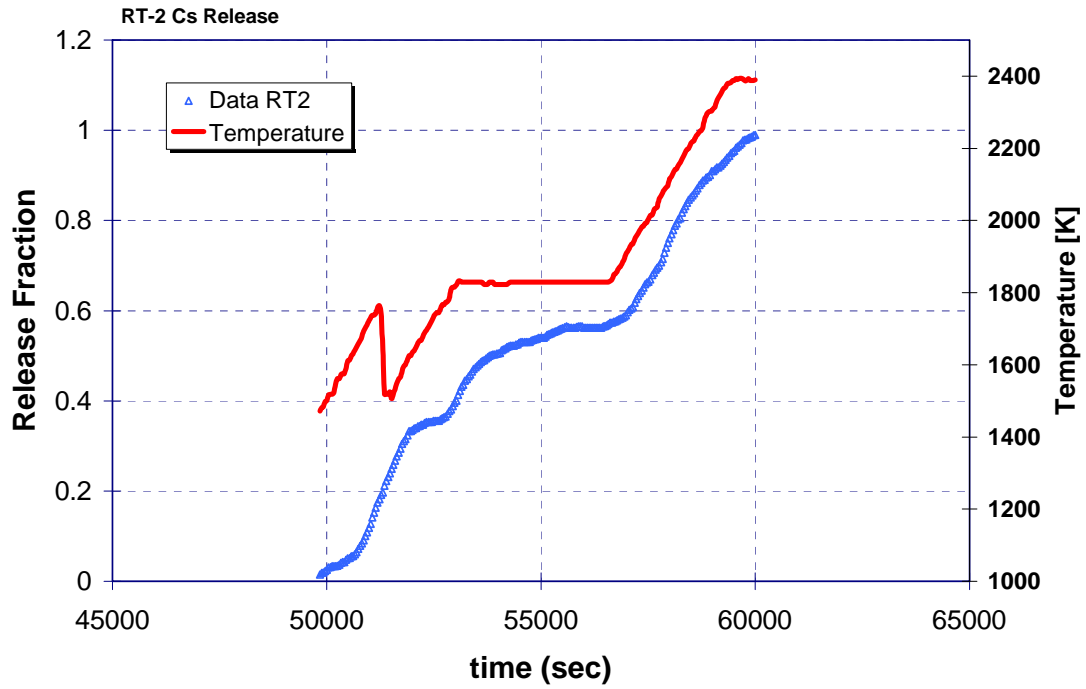


Figure 34. RT-2 release of cesium as a function of test sample temperature.

Table 6. Comparison of fission product release from VERCORS tests RT-1 (LEU) and RT-2 (MOX).

Isotope	MELCOR Release Class	Fraction Released RT-1 (LEU)	Fraction Released RT-2 (MOX)
¹⁰⁶ Ru	Class 6 - Platinoids (Ru) Ru, Rh, Pd, Re, Os, Ir, Pt, Au, Ni	0.09	0.0535
¹¹⁰ Ag	Class 12 - Main Group, less volatile (Ag) Ag, Ga, Ge, In, Sn	0.9	0.97
¹²⁵ Sb	Class 11 - Main Group, more volatile (Cd) Cd, Hg, Zn, As, Sb, Pd, Tl, Bi	0.96	0.77
¹³⁴ Cs	Class 2 – Alkali Metals (Cs) Li, Na, K, Rb, Cs, Fr, Cu	1.0	1.0
¹³⁷ Cs	Class 2 – Alkali Metals (Cs) Li, Na, K, Rb, Cs, Fr, Cu	1.0	1.0
¹⁴⁴ Ce	Class 8 – Tetravalent (Ce) Ti, Zr, Hf, Ce, Th, Pa, Np, Pu, C	0.03	0.02
¹⁵⁴ Eu	Class 9 – Trivalent (La) Al, Sc, Y, La, Ac, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Am, Cm, Bk, Cf	0.01	0.003
⁸⁵ Kr	Class 1 – Noble Gases (Xe) He, Ne, Ar, Kr, Xe, Rn, H, N	0.86	0.87

The parameters of the Booth release model described earlier in Eq. 4 and 5, namely the diffusion coefficient, D_0 and Q , may be determined from the experimental data by a fitting process described by Lorenz and Osborne [15]. In this process, Eq. 4 and 5 are inverted to solve for the product Dt/a^2 , as indicated below.

$$\frac{Dt}{a^2} = \frac{2}{\pi} - \frac{f}{3} - 2\sqrt{\frac{1}{\pi^2} - \frac{f}{3\pi}} \quad \text{for } f < 0.85 \quad \text{Eq. 11}$$

$$\frac{Dt}{a^2} = \frac{-1}{\pi^2} \ln \left[\frac{\pi^2(1-f)}{6} \right] \quad \text{for } f > 0.85 \quad \text{Eq. 12}$$

where f is the release fraction.

These forms may be used to plot the apparent instantaneous measured diffusion coefficient as a function of the measured temperature as shown in Figure 35. Also shown in this figure are two fits of the diffusion coefficient (Eq 3), one for MOX determined by inspection for the RT-2 data shown in red, and another shown in blue found to represent well Cs release from LEU fuel. As can be seen, there are significant differences in the apparent Cs diffusion coefficient for MOX fuel in comparison to the diffusion coefficient derived from release from LEU fuel.

The parameters used to represent Cs diffusion release from MOX and LEU fuel are summarized in Table 7.

Table 7. Diffusion coefficient for MOX and LEU fuel.

	D_o [m ² /s]	Q [Joule/kg-mole]
LEU Fuel (ORNL-Booth)	1×10^{-6}	3.814×10^5
MOX Fuel (MOX-Booth)	2×10^{-11}	1.664×10^5
Grain radius	6 m	6 m

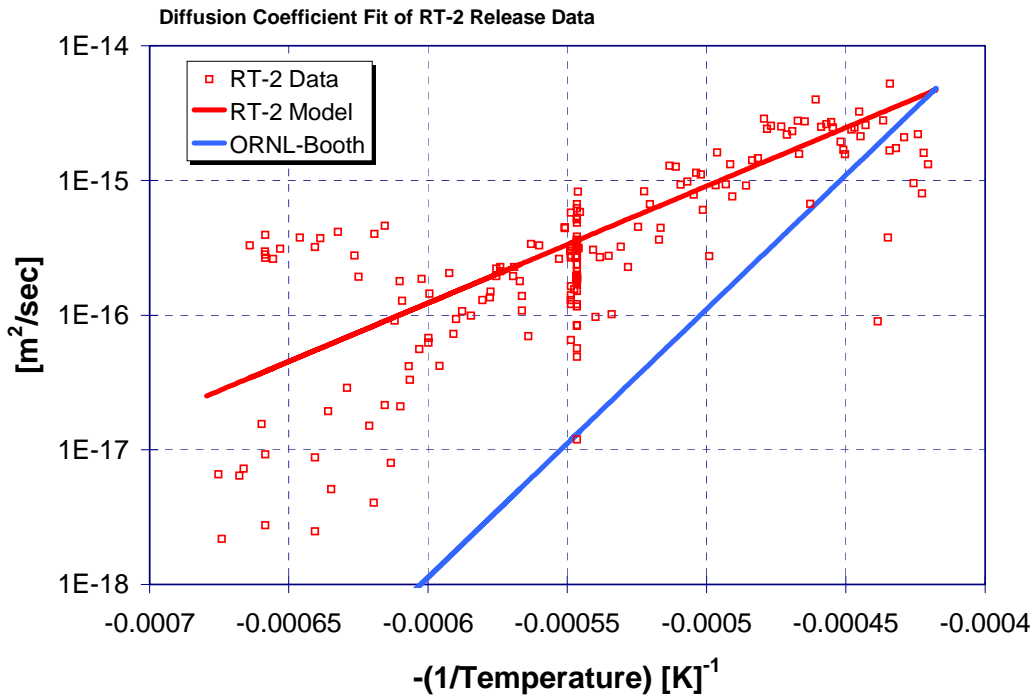


Figure 35. Plot of RT-2 Cs release data as a function of time using inverted Booth solution. Also shown are fits for $D(T)$ determined from the MOX data and the form recommended for LEU use (modified ORNL-Booth coefficients).

The Booth parameters for Cs release from MOX fuel determined from the RT-2 data are used in a MELCOR model of the RT-2 test to assess the predicted release against that observed experimentally. The results of the MELCOR release prediction for Cs release in test RT-2 are shown in Figure 36. As seen in Figure 36, the rates of Cs release are well predicted by the MOX-Booth diffusion parameters, and that the low temperature release rate compared to that of LEU fuel (ORNL-Booth) is greater for the same assumed temperature history. Significant release of Cs is observed to begin at around 1700 K for MOX fuel,

whereas, similar release rate in LEU fuel is not observed until temperatures exceed 2000 K.

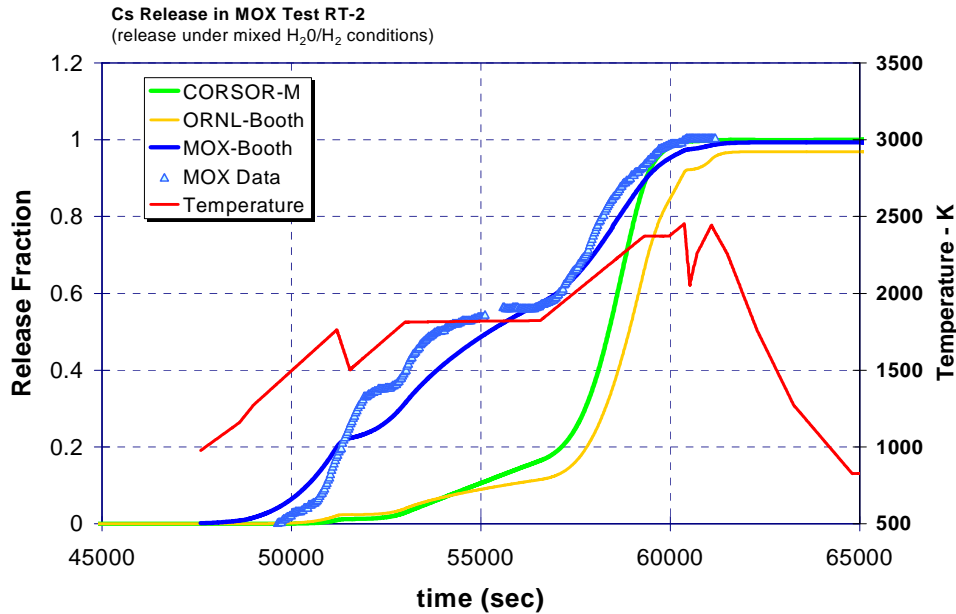


Figure 36. Comparison of the MELCOR Predicted Release of Cs Class for VERCORS Test RT 2 to the Experimental Measurement of Cs 137.

5.2 MELCOR Analysis of RT-6 High Burnup Experiment Using Fitted Booth Parameters

Using the method described in the previous section, the RT-6 experimental data may be cast in terms of the apparent instantaneous diffusion coefficient as shown in Figure 37. Also shown in this figure are expressions for the Booth temperature dependent diffusion coefficients for LBU fuel and the HBU fuel. Notice that the HBU data fit follows the trends of the RT-6 data. The parameters of the diffusion coefficient (D_0 and Q) were adjusted to obtain both a reasonable fit to the data shown in Figure 37 and the release rate versus temperature measurements. The parameters used to represent Cs diffusion release from HBU and LEU fuel are summarized in Table 8. The release prediction obtained using the Booth formula for HBU fuel is compared to the measured RT-6 data in Figure 38 along with the Booth prediction for LBU fuel under the same temperature history.

The predictions obtained using the HBU parameters match the initial release trends very well, underestimate the release rate at intermediate temperatures, and again match the release rate at high temperature very well. Note also that the LBU release model significantly underestimates the RT-6 observations. A more complex model for fission product release would be required to improve the intermediate temperature release rate; however, the HBU Booth model captures

the important observed trends, namely the onset of the release at lower temperatures than typical for LBU fuels and the completeness of release at high temperature.

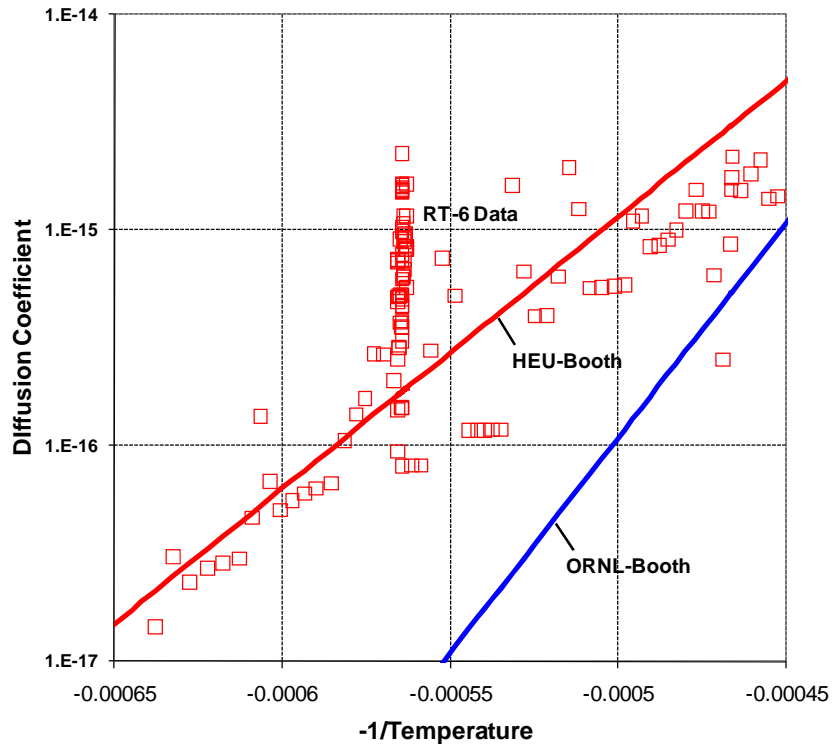


Figure 37. Instantaneous RT-6 diffusion coefficient and Booth model fits.

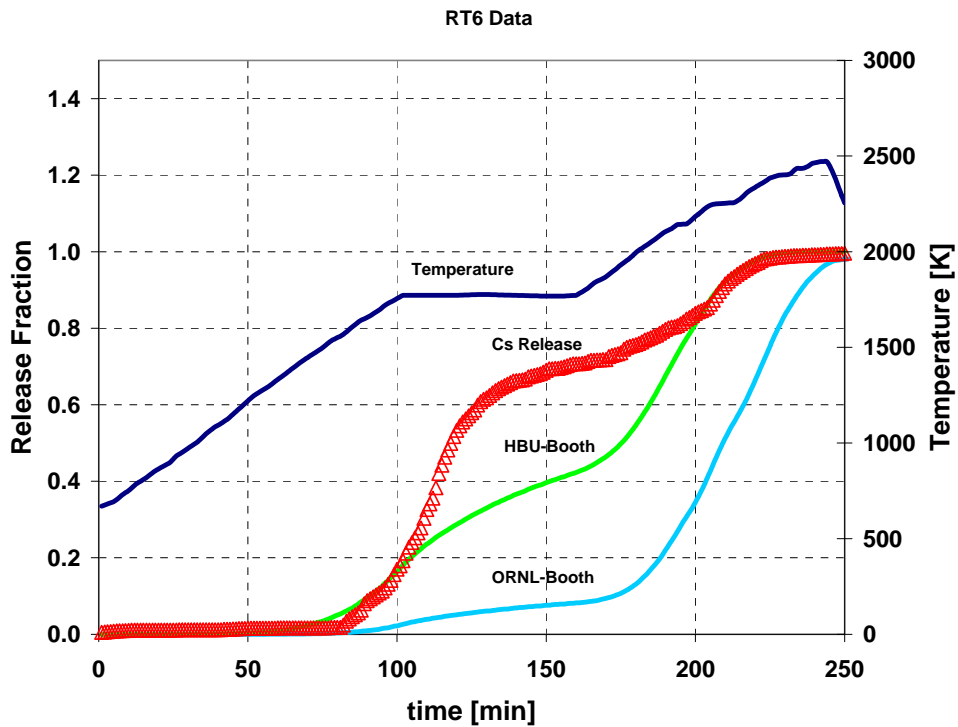


Figure 38. RT-6 release measurements compared to Booth model predictions for HBU and LBU fuels.

Table 8. Parameters for Diffusion Coefficient for HBU and LEU Fuel.

	D_o [m ² /s]	Q [J/kg-mole]
LEU Fuel (ORNL-Booth)	1×10^{-6}	3.814×10^5
HBU Fuel (HBU-Booth)	2.3×10^{-9}	2.411×10^5
Grain radius	6 μ m	6 μ m

The fitted Booth parameters for the available MOX and HBU VERCORS tests may be used in the integral codes such as MELCOR to evaluate the impact on accident consequences of using these fuels types and burnups. Of course fission product inventories of the appropriate isotopes must be specified accordingly to account for the differences in total inventory masses.

6 Safety Implications of Current Fission Product Understanding

The release trends observed in small scale experiments such as the VERCORS tests do not necessarily translate directly to the trends to be expected of the full scale reactor accident. In fact, this is the primary reason for the development of the integral accident codes such as MELCOR. In a reactor accident, there are considerable incoherencies spatially within the reactor core. Some regions of the core are significantly damaged and there is extensive release of fission products while other regions have experienced much less damage and there is much less release of fission products. In addition, the degree of damage associated with different types of reactor accidents may also produce significantly different fission product release patterns both with respect to timing as well as extent of release. Today, as knowledge such as that gained from the Phebus and VERCORS testing programs are incorporated into the severe accident codes, increasingly better estimates of fission product releases from widely varying accident conditions can be rendered.

7 Summary and Recommendations

Based in recent assessments of MELCOR fission product release modeling against the Phebus FPT-1 test and on observations from the ISP-46 exercise [10], modifications to the default MELCOR 1.8.5 release models are recommended. These assessments identified an alternative set of Booth diffusion parameters recommended by ORNL (ORNL-Booth) [11], which produce significantly improved release predictions for Cs and other fission product groups. Some adjustments to the scaling factors in the ORNL-Booth model were made for selected fission product groups, including UO_2 , Mo and Ru in order to gain better comparisons with the FPT-1 data. The adjusted model, referred to as “Modified ORNL-Booth,” was subsequently compared to original ORNL VI fission product release experiments and to the more recently performed French VERCORS tests, and the comparisons are as favorable or better than could be obtained using the CORSOR-M MELCOR default release model. These modified ORNL-Booth parameters, input to MELCOR as “sensitivity coefficients” (i.e. user input that over-rides the code defaults) are recommended for the interim period until improved release models can be implemented into MELCOR¹.

Deposition patterns in the Phebus FPT-1 circuit were also significantly improved by using the modified ORNL-Booth parameters, where retention of lower volatile Cs_2MoO_4 is now predicted in the heated exit regions of the FPT-1 test, bringing down deposition in the FPT-1 steam generator tube and in closer alignment with the experimental data. This improvement in “RCS” deposition behavior preserves the overall correct release of Cs to the containment that was predicted even with the default CORSOR-M model.

In the course of this assessment, a review of MELCOR release models was performed, identifying several areas for future improvements. These include upgrading the Booth release model to account for changes in local oxidizing/reducing conditions and developing a fuel oxidation model to account for the effects of fuel stoichiometry. Models such as implemented in the French ELSA code and described by Lewis [4,5] are considered appropriate for future MELCOR improvements. A model for Ru release under air oxidizing conditions is also needed and should be included as part of a fuel oxidation model as fuel stoichiometry is a fundamental parameter in determining the vapor pressure of Ru oxides over the fuel. There is also a need to expand the MELCOR architecture for tracking fission product classes to allow for more speciation of fission products. An example is the formation of CsI and Cs_2MoO_4 and possibly CsOH, if all Mo is combined with Cs such that excess Cs exists in the fuel.

¹ **Note added to proof:** Refined versions of the release models have been incorporated into the 1.8.6 and 2.1 versions of the MELCOR code. Also, a model of silver release from degrading silver-indium-cadmium control rods has been added.

References

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Copy to:

Mr. Richard Lee
RES/DSA/FSTB
U.S. Nuclear Regulatory Commission
MS C3A07M
Washington, DC 20555-0001

Mr. Jay Lee
NRO/DSER/RSAC
U.S. Nuclear Regulatory Commission
MS 7 F27
Washington, DC 20555-0001

Mr. Michael Salay
RES/DSA/FSTB
U.S. Nuclear Regulatory Commission
MS C3A07M
Washington, DC 20555-0001

Mr. Mark Leonard
Dycoda, LLC
267 Los Lentos Rd. NE
Los Lunas, NM 87031-9390

INTERNAL RECIPIENTS

Copy to:

MS-0736 Dana Powers, 06762
MS-0748 Nathan Bixler, 06762
MS-0748 Randy Gauntt, 06762
MS-0748 Larry Humphries, 06762
MS-0748, Joonyub Jun, 06762
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