CONF-890546--7

CORRELATION OF RECENT FISSION PRODUCT RELEASE DATA*

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CONF-890546--7

DE89 012972

To Be Presented
at ICHMT INTERNATIONAL SEMINAR
"Fission Product Transport Processes In Reactor Accidents"
Dubrounik, Yugoslavia, May 22-26, 1989

*Research Sponsored by Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission under Interagency Agreement DOE 1886-8084-4B with the U.S. Department of Energy under contract DE-AC05-84OR21400 with the U.S. Department of Energy.

DISCLAIMER

CORRELATION OF RECENT FISSION PRODUCT RELEASE DATA*

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INTRODUCTION

For the calculation of source terms associated with severe accidents, it is necessary to model the release of fission products from fuel as it heats and melts. Perhaps the most definitive model for fission product release is that of the FASTGRASS computer code [1] developed at Argonne National Laboratory. FASTGRASS contains mechanistic models for such phenomena as: diffusive transfer of species within grains to internal bubbles and to grain boundaries; diffusive transfer of bubbles to grain boundaries; diffusion of fission product species along grain boundaries and through interlinked cracks; growth of the grain size with irradiation and with time at temperature, and the effects of such grain growth on the transport. There is persuasive evidence that these processes, as well as additional chemical and gas phase mass transport processes, are important in the release of fission products from fuel. Nevertheless, it has been found convenient to have simplified fission product release correlations that may not be as definitive as models like FASTGRASS but which attempt in some simple way to capture the essence of the mechanisms. One of the most widely used such correlation is called CORSOR-M [2] which is the present fission product/aerosol release model used in the NRC Source Term Code Package. The CORSOR type of modelling had its formal beginning as an expedient in the NUREG-0772 exercise [3] (actually it was patterned after the technique being used at KfK to correlate their SASCHA experimental data [4]). CORSOR has been criticized as having too much uncertainty in the calculated releases and as not accurately reproducing some experimental data (notable PBF, ACRR, and the recent ORNL HI and VI series of tests). For example, Figure 1 shows cesium release data from some recent ORNL tests [5] that indicate release coefficients as much as an order of magnitude lower than CORSOR and that these coefficients systematically decrease with time. It is currently believed that these discrepancies between CORSOR and the more recent data have resulted because of the better time resolution of the more recent data compared to the data base that went into the CORSOR correlation. The release rate coefficients for CORSOR and CORSOR-M had been derived on the basis of

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*Research sponsored by the Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission under Interagency Agreement DOE 1886-8012-7B with the U.S. Department of Energy under contract DE-AC05-840R21400 with the Martin Marietta Energy Systems, Inc.

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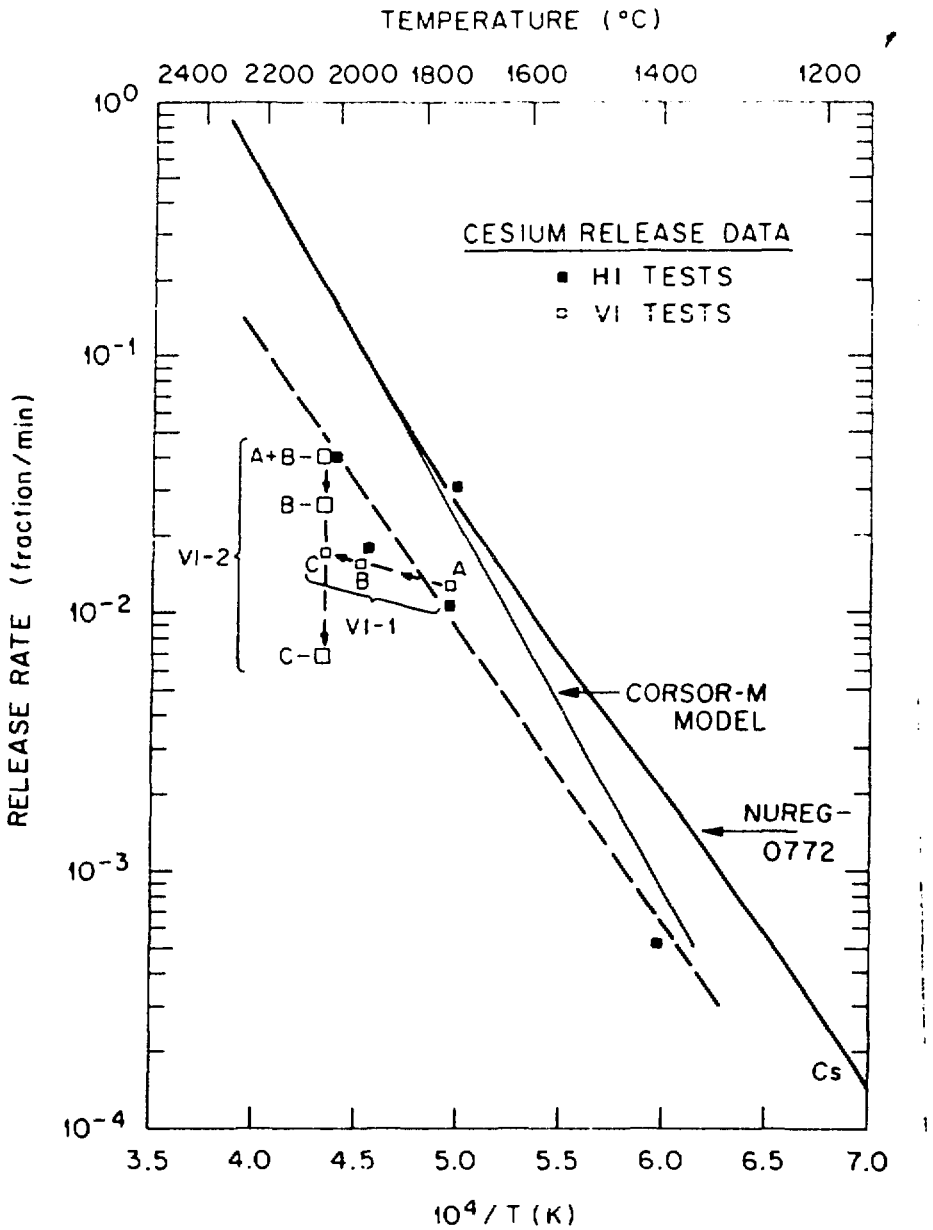

FIGURE 1. Comparison of release rate coefficients from tests VI-2 and VI-1 with earlier work.

relatively short term constant temperature release experiments and on simulated fuel experiments conducted prior to 1983. Both of these conditions tend to bias the results to the high side.

The systematic decrease in the release rate coefficients with time at constant temperature as observed in the ORNL experiment results shown in Figure 1 indicates that the first order rate equation that is the basis for CORSOR may be an inadequate correlational model. (Note that the A, B, and C designations on Figure 1 indicate successively later times in the indicated experiment.) Recently some success has been had in better correlating both the ORNL and the PBF data with a "diffusive-like" correlational model [6]. This is not too surprising because the mechanisms (believed realistic) in FASTGRASS are generally diffusive in nature, and even pure surface evaporation can be described by equations of similar form.

A reasonable choice, then, for a simple correlational model is one that has a diffusive-like character to it. The question then arises, how best can the existing data base that tends to be oriented toward release rate coefficients be utilized in such a model? A technique for doing this was developed by the authors for use in connection with the NUREC-1150 risk uncertainty exercise sponsored by NRC at Sandia National Laboratories and is presented below.

SIMPLIFIED CORRELATIONAL MODEL

A "diffusive-like" correlational model was developed for fission product release from heating fuel:

$$\frac{\partial C}{\partial t} = D \frac{1}{r} \frac{\partial^2}{\partial r^2} (r\ C)$$

where $D = D_o \exp(-Q/RT)$.

It is not implied that this is in any way a true mechanistic description of the governing phenomena - just that it appears to be a convenient empirical correlational scheme. It is intended that the above equation be coupled with the thermal hydraulic calculation of the fuel temperature and be solved in finite difference form using some effective diffusion length parameter. The correlational parameters to be empirically based, then are:

1. an effective spherical length parameter, a, that could be viewed as being related to the mean grain radius,
2. an effective "diffusion coefficient frequency factor," D_o, and
3. an effective "activation energy," Q.

In the ORNL Hot Cell Fission Product Release tests using real discharged fuel, the Cs generally dominates the released activity from the test fuel specimens [7]. It has been possible, then, to use the gamma measurements made downstream of the furnace for each of the tests to get continuous measurements of the Cs release. The correlational model proposed above was "best fit" to a significant number of the recent ORNL tests to obtain for cesium release from high burnup fuel:

$$a = 6 \text{ microns},$$
$$D_o = 0.0000763 \text{ cm}^2/\text{s}, \text{ and}$$
$$Q = 74300 \text{ cal/mole}.$$

Typical comparisons of the predicted and experimental release patterns using these parameters are shown in Figure 2 (the present model is referred to as the "Booth" model on Figure 2).

If sufficient time-resolved data existed, it would be possible to similarly establish a set of such parameters for each fission product elemental group (and other released material). Unfortunately such a data base for the various fission product and inert aerosol groups does not presently exist. Even if only total (integral) release data were available from a significant number of release experiments conducted under different temperature conditions, then it would be possible to fit these via regression techniques to determine appropriate values for the parameters. While this is the present approach of the ORNL research, as of now the quantity of high temperature data is still insufficient to fully

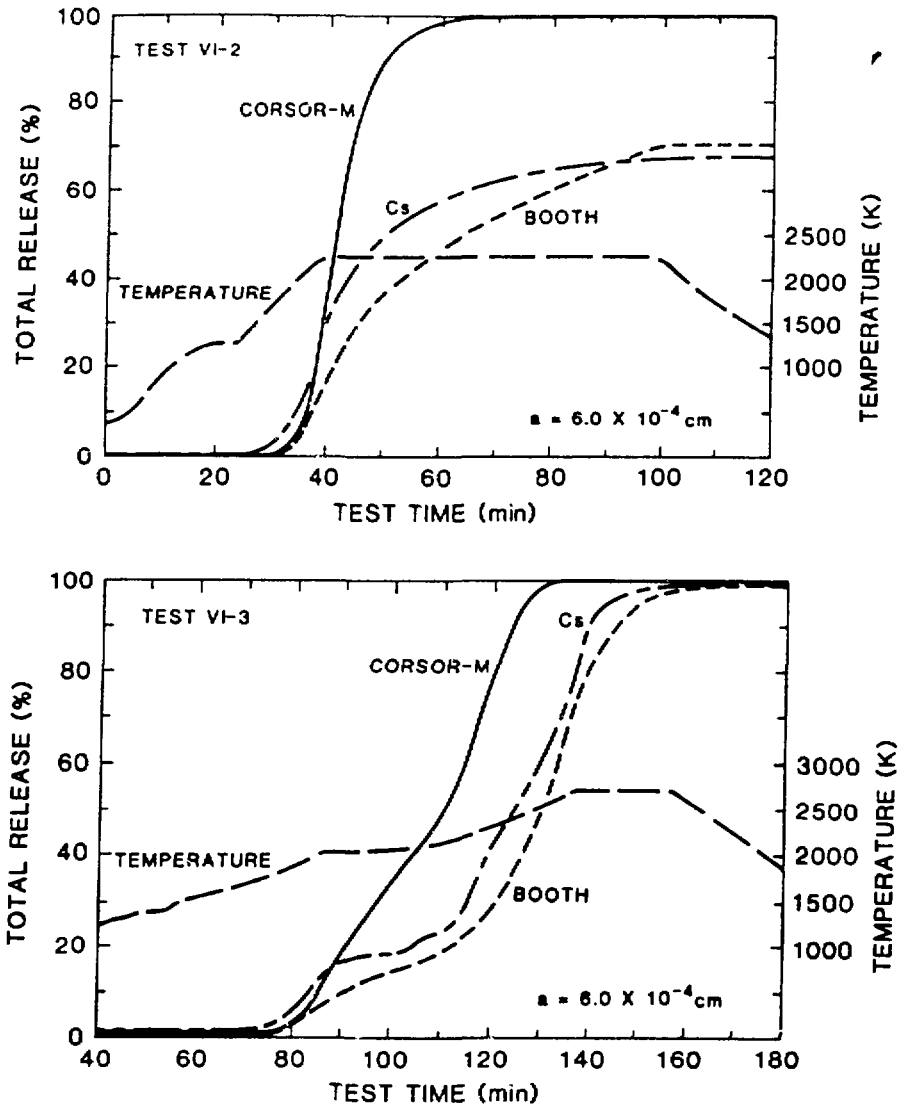

FIGURE 2. Typical comparisons of ORNL Fission Product Cesium Release Data with a "Diffusion" model (Booth).

implement it. There is at present insufficient data to develop values for D_o and Q for each elemental group. Consequently, we are proposing an alternate approach that utilizes past fission product release experience for application with the proposed simplified "diffusion" correlation.

Lorenz [8] has observed that there is a remarkable consistency in the fission product and structural material release data when reported in terms of a release rate coefficient (for small releases, this is essentially the integral release fraction for fuel heated to very high temperatures divided by the release time at temperature). Figures 3 and 4, taken from Reference 8, illustrate this consistency. If any particular tests' results for the measured release rate coefficients are placed on a logarithmic scale ordinate in such a way that they form a straight line, then the resulting positions of the constituents on the linear abscissa defines what Lorenz called a "relative volatility" for each. Lorenz observed that the release rate coefficient data from other tests at markedly different conditions, when plotted on these same scales, also resulted in straight lines. The large difference in slopes for the relative volatility of species in the SASCHA tests (Fig. 3), and other tests

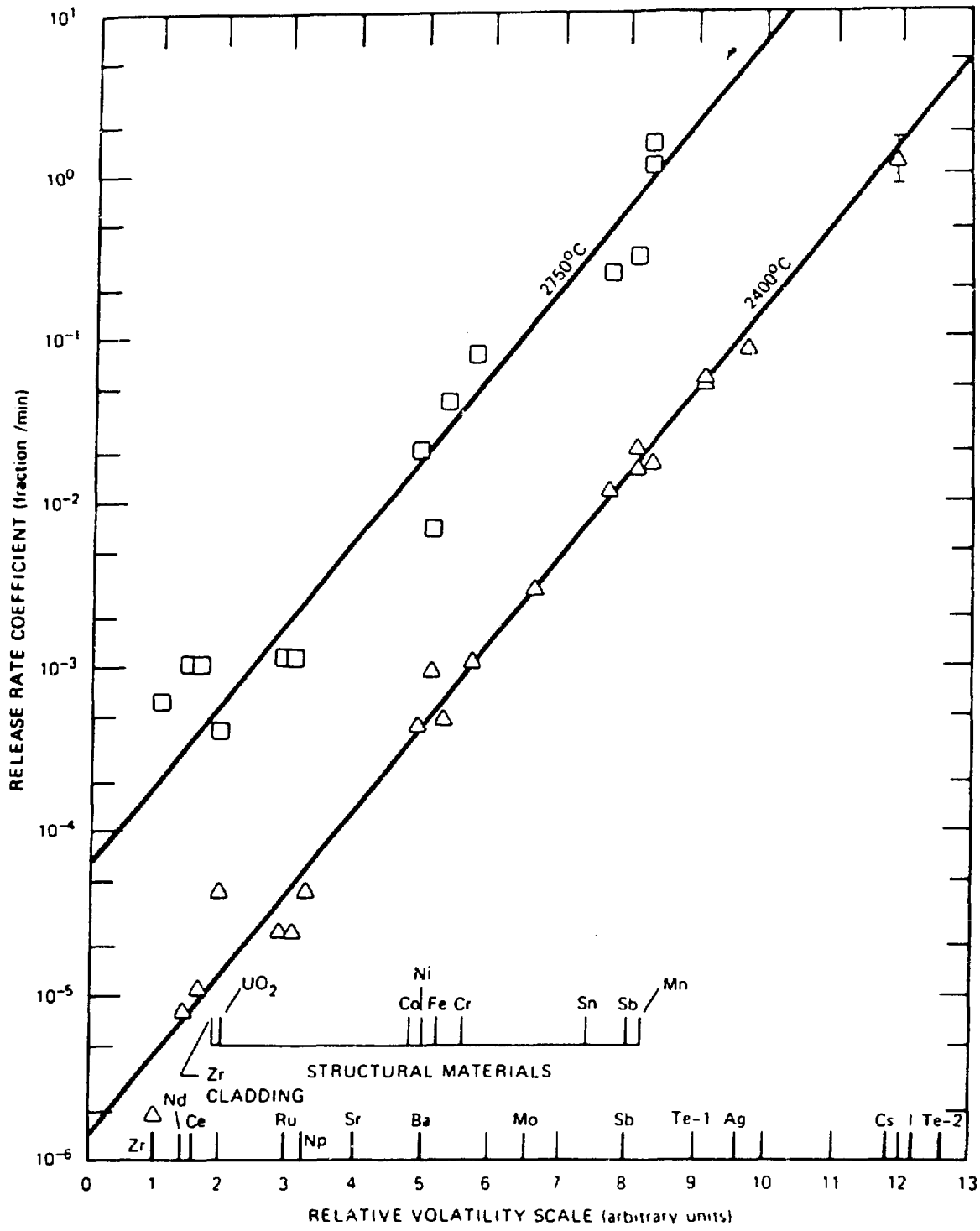

FIGURE 3. Release rate of species in SASCHA tests in air at 2400 and 2750°C.

(Fig. 4) appears to be related to the size of the fuel specimen and the geometry of the tests. The relative release rates for structural material components used in the SASCHA tests were found to be directly proportional to their partial pressures [8].

If the straight line relationship can be considered to be a universally applicable principle (to be proven), then we have a powerful tool,

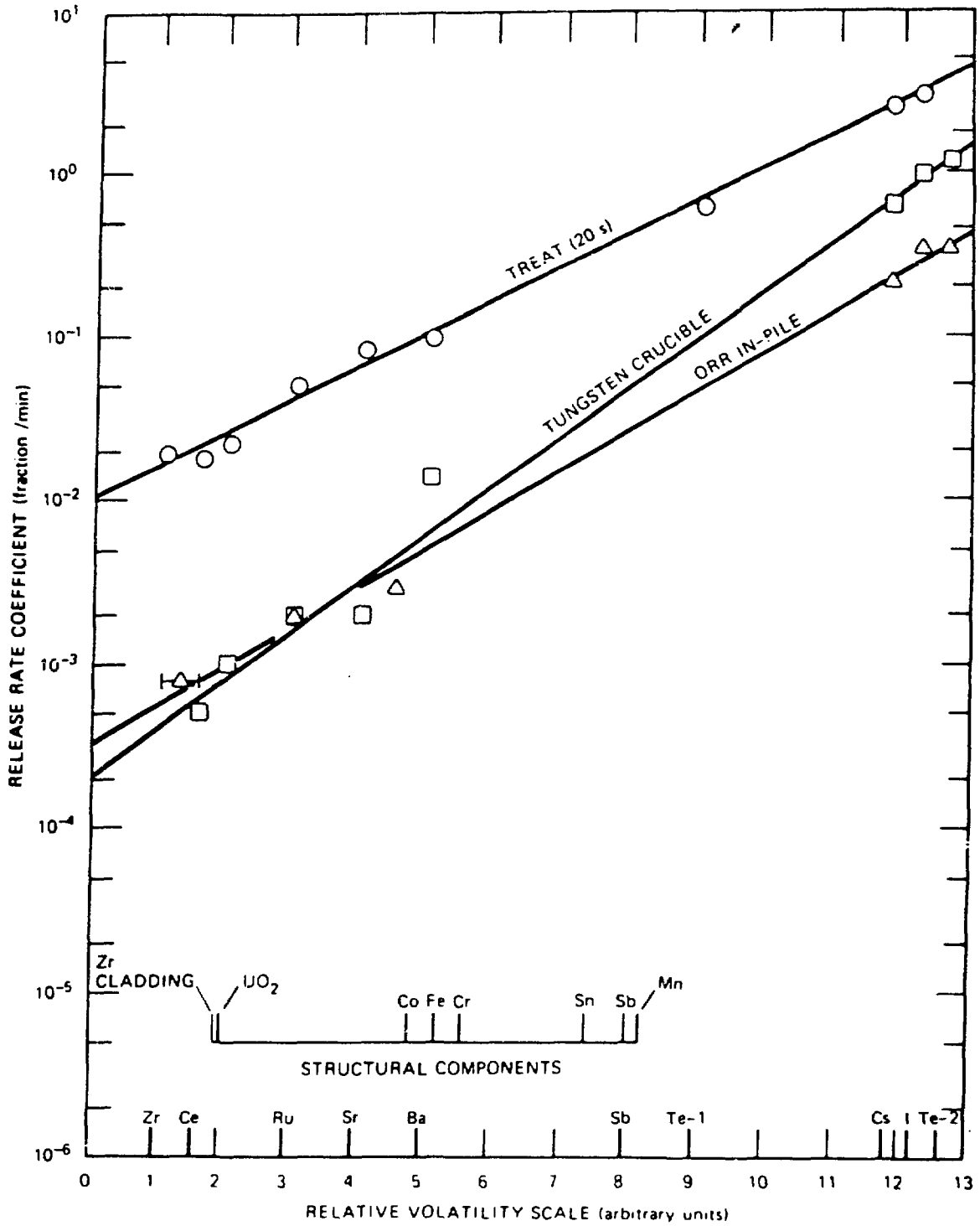

FIGURE 4. Release of fission products from molten UO_2.

indeed. The "relative volatility" relationship permits a semi-quantitative comparison of release results even though the exact time-temperature history may be unknown (as is the case for the TREAT transient heating test data shown in Fig. 4). Because of the way the release rate coefficients were derived for Figs. 3 and 4, it would be expected that plotting the integral release values on the relative volatility scale would also give a straight line. Therefore, we need only to know the release values for two elements (to define the straight line) then we can interpolate to

find the release values for all the other constituents, knowing only their "relative volatility" values.

We already have one good standard in the D_o, Q values for Cs – so we need only to look for one more. Available data from the most recent HI and VI series of tests conducted in the ORNL Fission Product Release Program have been examined to see if they are adequate for developing values for a second standard. Sufficient data exist to develop D_o and Q values for Cs, Sb, and Kr. However, some judicial selection was necessary. The "relative volatility" value for Kr is too near that of Cs for it to be a good second standard. Unfortunately, the "relative volatility" value for Sb on Figures 4 and 5 primarily come from SASCHA data which were developed under conditions for which considerable unoxidized metallic phases (primarily Zr) were present during the release periods. It is known from recent ORNL results that the presence of unoxidized Zr hinders the release of Sb just as it does Te in the CORSOR-M model. The release rate for Sb when the cladding becomes fully oxidized is much higher, probably around 11 on the relative volatility scale of Figs. 3 and 4: much too close to Cs to be a good second standard. Therefore, to use Sb as the second standard in the proposed technique, the D_o and Q values would have to be developed from tests that reflect the unoxidized Zr conditions under which the position of Sb on the "relative volatility" scale was determined. At this time, it appears that only two of the ORNL tests satisfy this criteria. It is recognized that the integral release values from only two tests of different fuel types do not represent an adequate data base on which to place much confidence in the parameters for the second standard for the proposed model. Nevertheless, we have fit the model to these two tests for purposes of illustrating the technique and for providing interim preliminary standards. The values will need to be refined as additional high temperature data become available.

Therefore, the preliminary values for the use of Sb as the second standard are:

$$D_o = 2.81 \times 10^{-6} \text{ cm}^2/\text{s}$$

$$Q = 82{,}500 \text{ cal/mole.}$$

The simplified fission product release algorithm, then, is as follows:

1. For the given temperature transient, input an average value for the grain radius (6 microns is recommended), or some correlation for grain size growth, and solve the basic model equation for two "standard" species to obtain their fractional release values. Here, for illustration, we recommend Cs and Sb, for which:

Species group	D_o (cm²/s)	Q (cal/mole)
Cs	7.63×10^{-5}	74300
Sb	2.81×10^{-6}	82500

2. Use the "relative volatility", RV, scale as taken from Figure 4 and shown in Table 1 (note that the arbitrary scale originally selected on Figure 4 has been adjusted in Table 1 to give a value of 1.00 for the "relative volatility" for Cs).

TABLE 1. Relative volatility values

Species group (i)	RV(i)
Fission Products	
NG	1.1
Te	1.07 (oxidized cladding)
I	1.03
Cs	1.00
Sb	0.68 (unoxidized cladding)
Ba	0.42
Sr	0.34
Ru	0.25
La	0.14
Ce	0.085
Structural Materials	
Mn	0.69
Sb	0.68
Sn	0.63
Cr	0.47
Fe	0.44
Co	0.41
UO_2	0.17

3. With the release value for Cs, (RCs), and for Sb, (RSb), calculated in (1) and the RV(i) values listed in (2), use the following interpolation formula to calculate the fractional release R(i) for any other species group:

$$R(i) = \frac{RCs}{\exp\left[\frac{\ln(RCs/RSb)\ RV(Cs)}{RV(Cs) - RV(Sb)}\right]} \exp\left[\frac{\ln(RCs/RSb)\ RV(i)}{RV(cs) - RV(Sb)}\right]$$

It is noted here that, in addition to Sb, certain other species, (notably Te and possibly Ru) are held up by unoxidized Zr clad. The above release calculation for such species represents only the release from the fuel to the clad. At some higher clad oxidation level these will then get re-released from the clad to the gas space. In addition, special provisions may have to be made to include the effects of highly oxidizing conditions on other species (particularly Ru and Mo) as well as for the effects of burnup and for conditions in which the resistance in the gas phase controls.

To fully implement this proposed correlation, additional data are needed to establish a better second "standard" and, perhaps, additional refinement of the RV scale. Because of the data sources, the suggested technique is most appropriate for high temperature release.

REFERENCES

1. Rest, J., An Improved Model for Fission Product Behavior in Nuclear Fuel Under Normal and Accident Conditions, *J. Nucl. Mater.*, 120, 195 (1984).

2. Kuhlman, M. R. (BCL), Lehmicke, D. J. (BCL), and Meyer, R. O. (NRC), "CORSOR User's Manual," NUREG/CR-4173, BMI-2122, Battelle's Columbus Laboratories, March 1985.

3. Technical Bases of Estimating Fission Product Behavior During LWR Accidents, NUREG-0772, U.S. Nuclear Regulatory Commission (June 1981).

4. Albrecht, H., and Wild, H., Review of the Main Results of the SASCHA Program on Fission Product Release Under Core Melting Conditions, *Proc. ANS Topl. Mtg. Fission Product Behavior and Source Term Research*, Snowbird, Utah, July 15-19, 1984, Electric Power Research Institute (1985).

5. Osborne, M. F., Collins, J. L., and Lorenz, R. A., Experimental Studies of Fission Product Release from Commercial LWR Fuel Under Accident Conditions, *Nucl. Technol.* 78(2), 157–69 (August 1987).

6. Nakamura, T., and Lorenz, R. A., Effective Diffusion Coefficients Calculated From ORNL Fission Product Release Tests, ORNL Technical Report (in progress).

7. Collins, J. L., Osborne, M. F., Lorenz, R. A., and Malinauskas, A. P., Fission Product Iodine and Cesium Release Behavior Under Severe LWR Accident Conditions, *Nucl. Technol.* 81(1), 78–94 (1987).

8. Lorenz, R. A., The Vaporization of Structural Materials in Severe Accidents, *Proceedings of the International Meeting on Thermal Nuclear Reactor Safety*, NUREG/CP-0027, Vol. 2, August 29 - Sept. 2, 1982.