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February 2, 1984

Mr. W. M. Jankowski
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Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, D. C. 20555

Dear Mike:

Here are a few comments that I have about the material presented at the last peer review meeting, as well as at previous meetings.

- (1) The methyl iodide problem has still not been addressed, nor has the effect of a strong radiation field on the chemistry of the various species.
- (2) In treatment of flow in the upper plenum at Surry, it was assumed that the domed region above the upper plenum was essentially isolated from the upper plenum. Is this treatment justified in all PWR's?
- (3) The intact RCS loops can serve as effective heat sinks (and fission product sinks) as degradation of the PWR core proceeds. Is this effect significant in reducing the release of fission products from the RCS?
- (4) How sensitive are the releases in an AB sequence to the size of the assumed hole in the piping? I understand BCL assumed a hole of 8-in. diameter.
- (5) Warman's point about the crucial nature of timing of the resuspension of fission products and the failure of the containment was interesting. This point needs to be studied at length.
- (6) In Volume IV, how are the DF values in Table 7.5 calculated? The foot-note is ambiguous and the values for 145 and 160 minutes seem anomalous. For Table 7.7, the DF should be defined in a foot-note. Why the discrepancy between the values of DF for the TMLB' and TML accidents?
- (7) I would like to see a summary of the releases to the environment, as estimated by WASH-1400, NUREG-0772, and BMI-2104 for the spectrum of accidents examined.

I look forward to learning what IDCOR will come out with. I have enjoyed participating in the review meetings.

Yours truly,

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TEMPERATURE DEPENDENCE OF FISSION PRODUCT RELEASE RATES

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In NUREG-0772 [1] and subsequent "source term" reports (e.g., NUREG/CR-2629 [2] and BMI-2104 [3]), the release rates of the fission products from the fuel prior to core slumping were estimated by the Albrecht-Wild model [4]. In this model, a fractional release rate, K , is defined for a fission product such that the amount, M , of the fission product remaining in the fuel after a time, t , is determined by

$$\frac{dM}{dt} = -KM. \quad (1)$$

Defining F as the fractional release of the fission product, so that F is equal to $1 - M/M_0$, where M_0 is the initial amount of the fission product present, Eq. (1) gives

$$F = 1 - e^{-Kt}. \quad (2)$$

In the Albrecht-Wild model, the fractional release rate, K , is a function of temperature, namely,

$$K = Ae^{BT}. \quad (3)$$

Values of A and B have been determined for a number of fission products by fitting Eqs. (2) and (3) to experimental data. The model was further developed in Ref. 1 in which the overall temperature range was divided into three parts ($800^\circ\text{C} < T \leq 1400^\circ\text{C}$, $1400^\circ\text{C} < T \leq 2200^\circ\text{C}$, $T > 2200^\circ\text{C}$) with values of A and B defined for each part. More recent values for A and B are presented in Ref. 2.

In reviewing the fission product release data reported by Albrecht et al. (Kfk) [4-7] and Lorenz et al. (ORNL) [7-15], it seems to us more reasonable that the fractional release rate should exhibit the usual Arrhenius temperature dependence of the form

$$K = K_0 e^{-Q/RT}, \quad (4)$$

instead of the Ae^{BT} form used by Albrecht and Wild. Q may be interpreted to represent the activation energy for the rate-controlling release mechanism. R is the universal gas constant, 8.317 J/mol·K (1.986 cal/mol·K).

To test this idea, and to determine values for K_0 and Q , published release data for each fission product were plotted as $\ln K$ vs $1/T$. For the most part, data from experiments conducted in an air or inert atmosphere were used, although a few of the data were obtained in a steam environment. Albrecht's data were obtained at the SASCHA facility [6]; Lorenz's HI tests were based on fission product release from fuel irradiated in the H. R. Robinson PWR.

The results are presented in Figures 1-13. A single straight line was selected to represent the data for each plot, and in most cases one line correlates the data adequately over the entire temperature range. The corresponding values of K_0 and Q for each fission product are reported in Table I. Based on the indicated Q values, the fission products may be divided into two major categories:

Kr, Sr, Mo, Ag, Sb	:	$Q = \sim 200-300 \text{ kJ/mol}$
Te, I, Cs, Ba		
Zr, Ru, Ce, Nd		$Q = \sim 700-1100 \text{ kJ/mol}$

For purposes of comparing the Arrhenius release rate model with the Albrecht-Wild model, curves corresponding to the latter model are also shown (as dashed lines) in the figures, using values for A and B reported in Ref. 2.

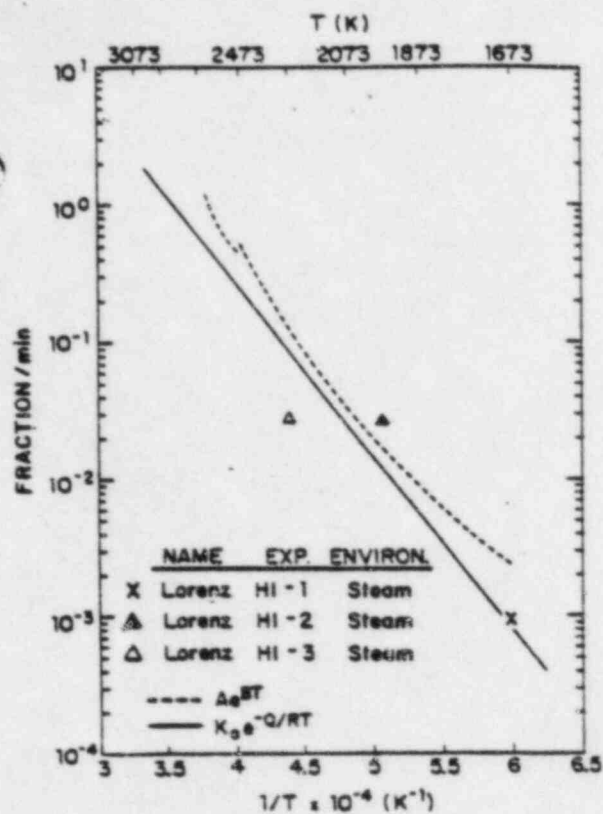


Fig. 1. Release Rate for Krypton

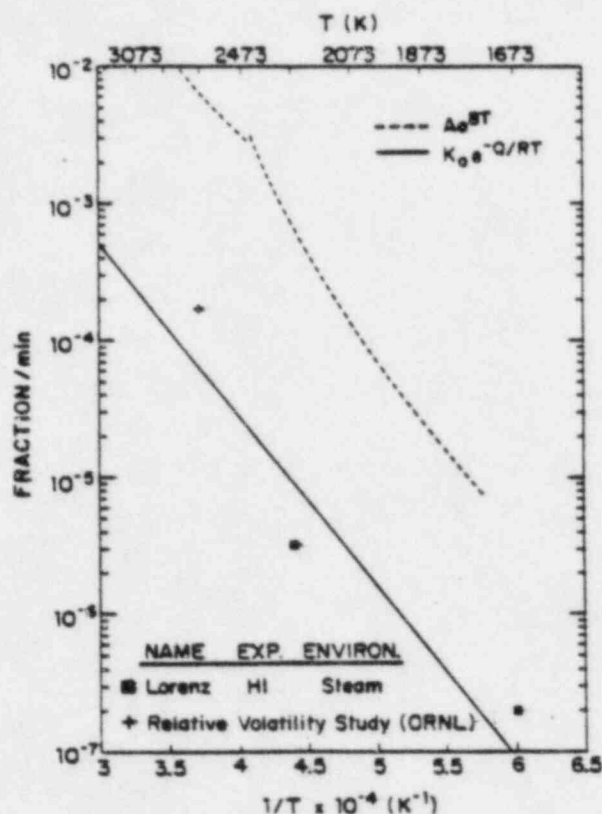


Fig. 2. Release Rate for Strontium

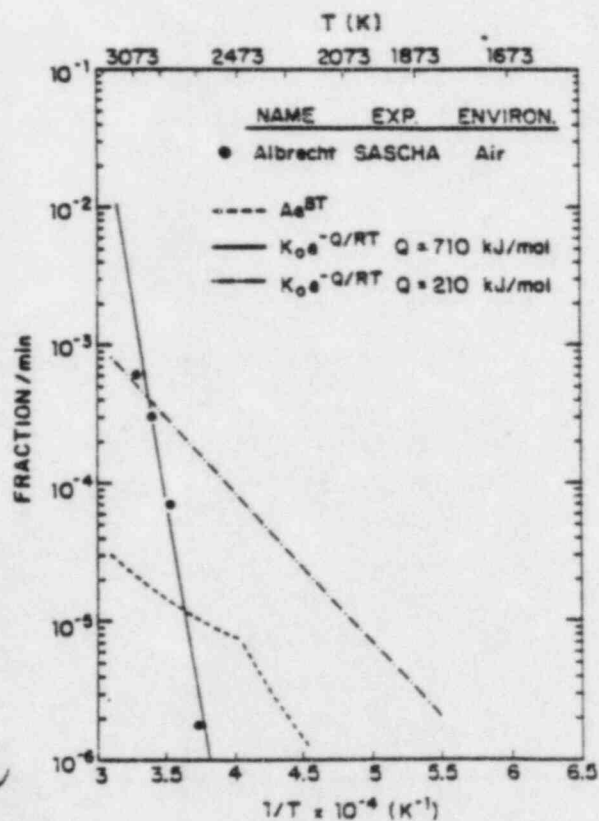


Fig. 3. Release Rate for Zirconium

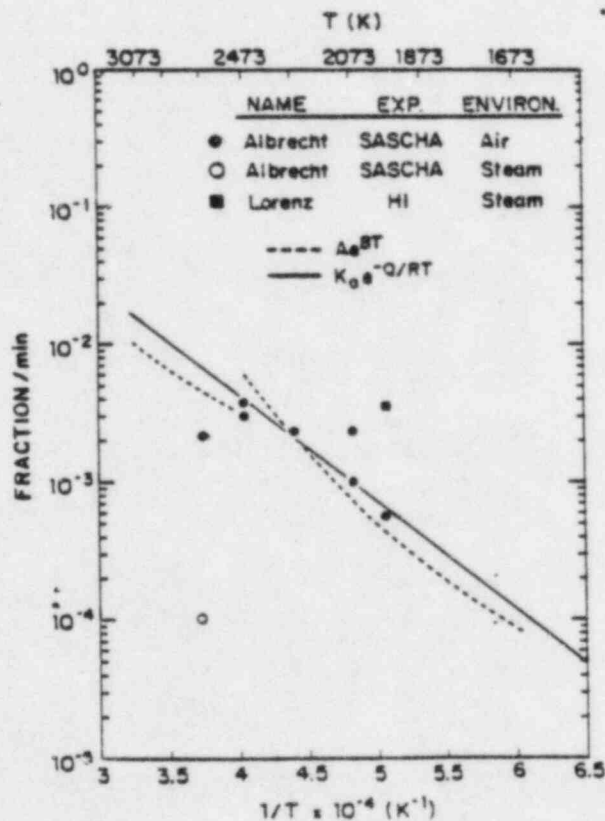


Fig. 4. Release Rate for Molybdenum

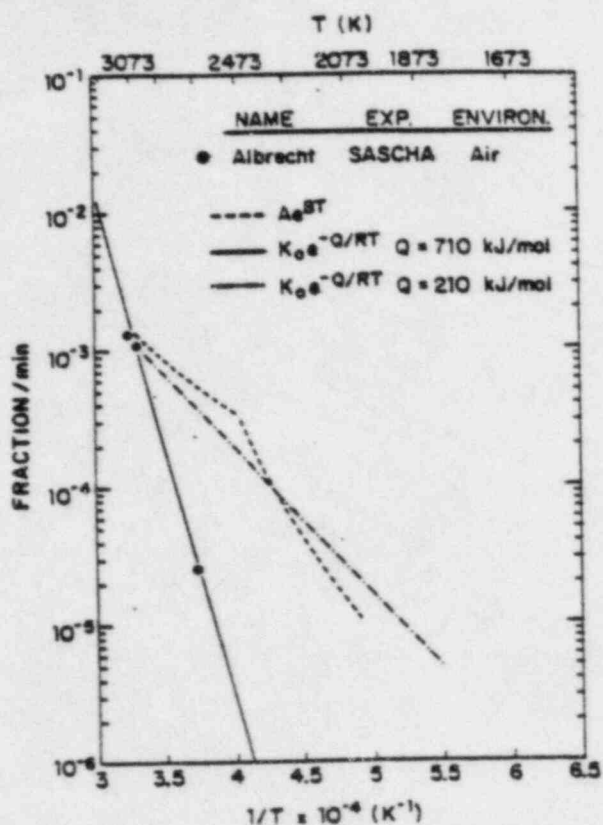


Fig. 5. Release Rate for Ruthenium

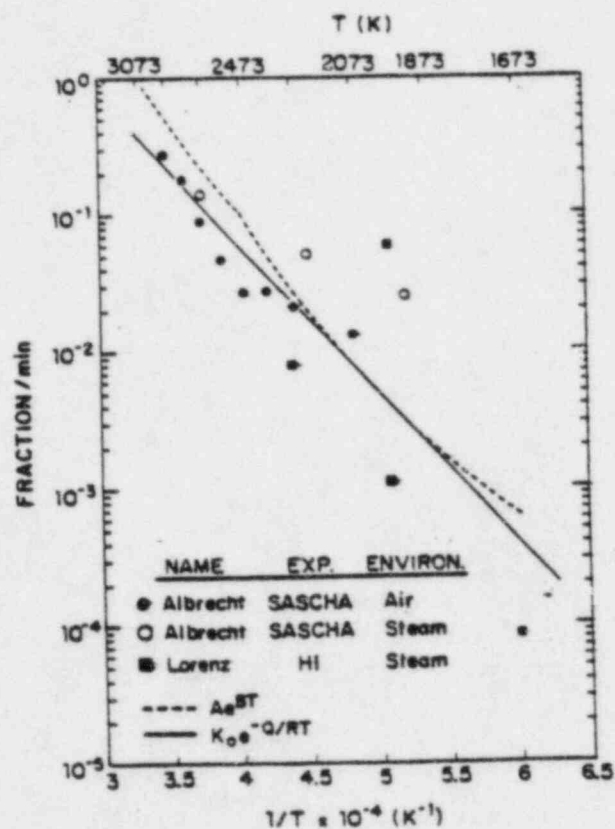


Fig. 6. Release Rate for Silver

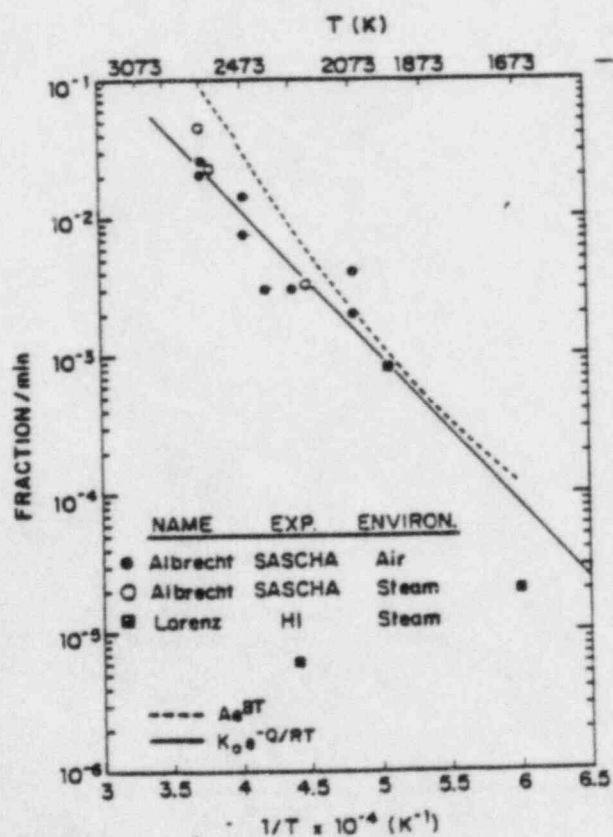


Fig. 7. Release Rate for Antimony

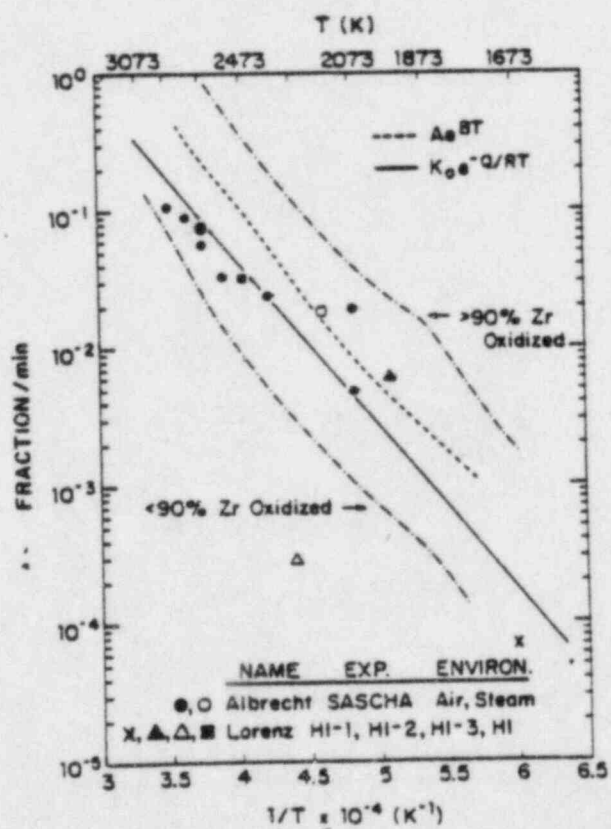


Fig. 8. Release Rate for Tellurium

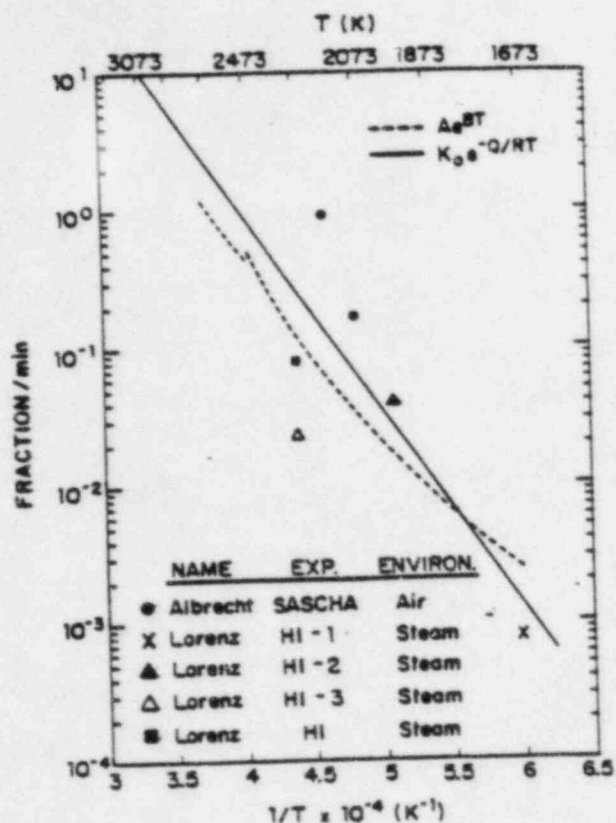


Fig. 9. Release Rate for Iodine

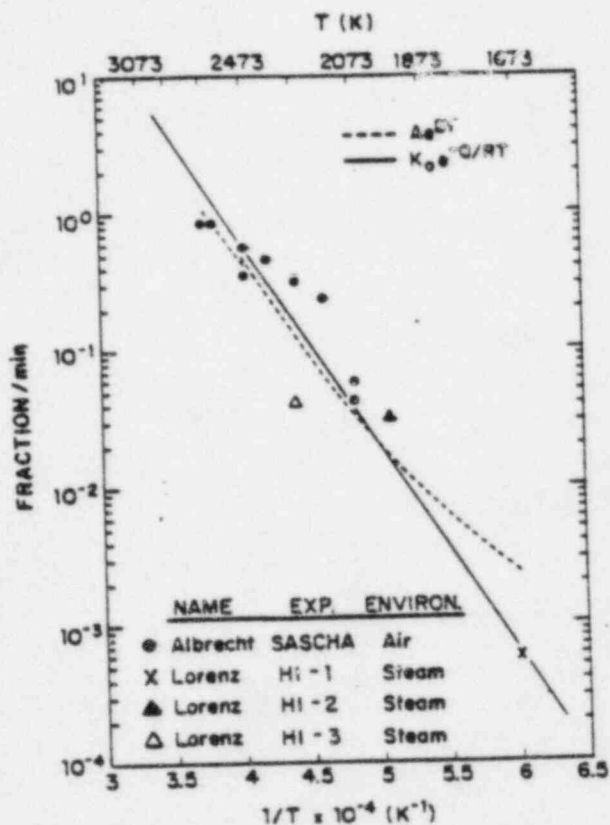


Fig. 10. Release Rate for Cesium

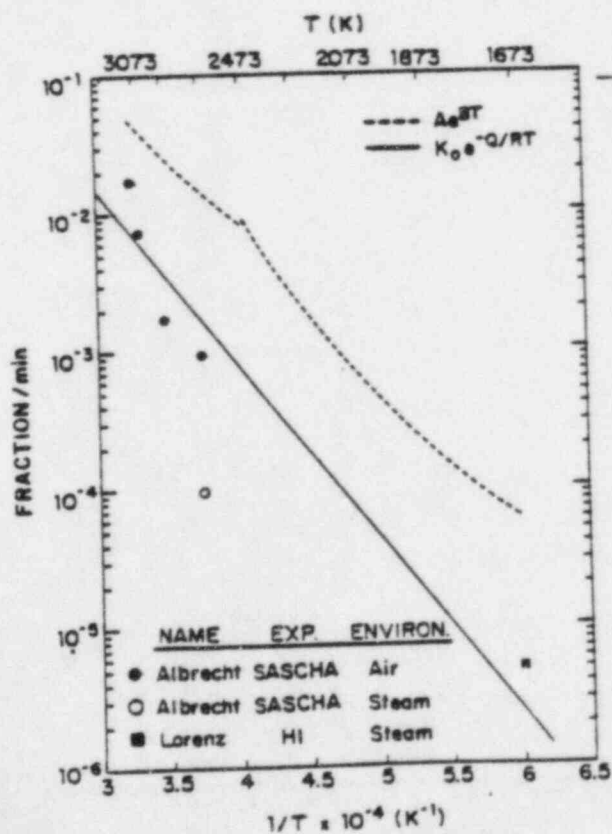


Fig. 11. Release Rate for Barium

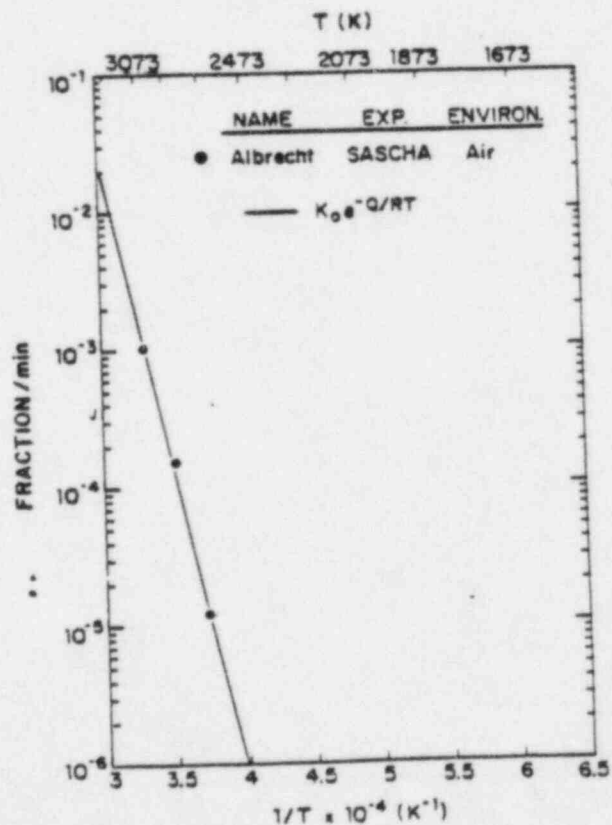


Fig. 12. Release Rate for Cerium

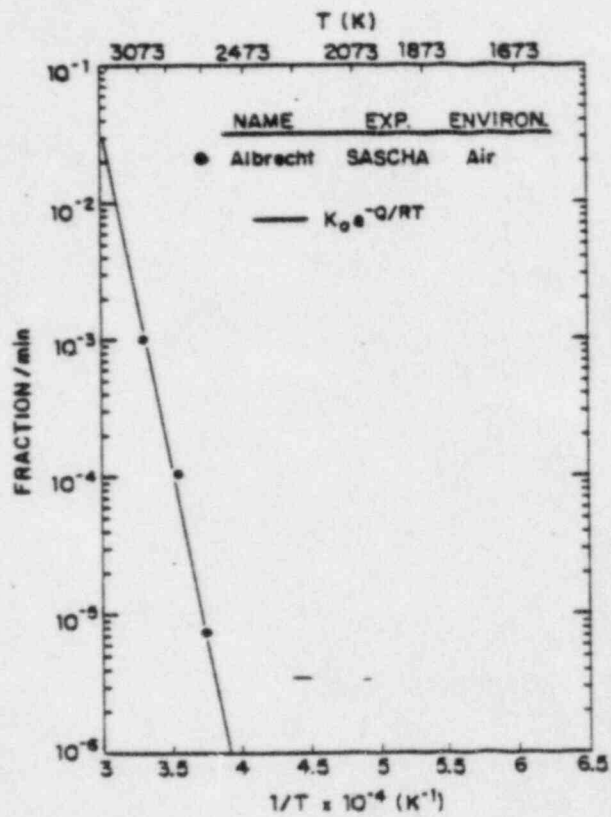


Fig. 13. Release Rate for Neodymium

Table I

Arrhenius Parameters for Fission Product Release

Fission Product	K_0 (min^{-1})	Specific Activation Energy, Q	
		(kJ/mol)	(kcal/mol)
^{36}Kr	3.4×10^4	240	58
^{38}Sr	4.3	250	59
^{40}Zr	3.2×10^6	1100	270
^{42}Mo	6.1	150	36
^{44}Ru	2.0×10^9	710	170
^{47}Ag	1.7×10^3	210	50
^{51}Sb	2.3×10^2	210	49
^{52}Te	2.7×10^3	230	55
^{53}I	6.0×10^5	280	66
^{55}Cs	8.6×10^5	290	70
^{56}Ba	94	240	58
^{58}Ce	6×10^{11}	840	200
^{60}Nd	4.4×10^{13}	960	230

For most of the fission products, the Arrhenius model yields fractional release rates that are fairly close to the Albrecht-Wild model. The most significant differences arise in the cases of Sr, Zr, Ru and Ba. In each of these cases, the Arrhenius model better represents that data but is less conservative, especially at moderate to low temperatures (i.e., at $T < 2200$ C). Release coefficients were not given in References 1-3 for Ce and Nd.

In the case of Ru (Fig. 5), it should be recognized that the value of Q is strongly influenced by the single datum in the vicinity of 2500 C (or $3.75 \times 10^{-4} \text{ K}^{-1}$). Since Ru and its daughter rhodium are important fission products, it is important that the Ru release rate not be underestimated. For this reason, a second line is shown on Fig. 5 which corresponds to an assumed value for $Q = 210 \text{ kJ/mol}$ and passes through the two data that seem to confirm one another. The Arrhenius line for $Q = 210 \text{ kJ/mol}$ gives release rates that are fairly consistent with the Ref. 2 values for the Albrecht-Wild model.

A second line was also drawn for $Q = 210 \text{ kJ/mol}$ for Zr (Fig. 3). The slope of this line is comparable to that for the Ref. 2 values for the Albrecht-Wild model. The location of this line was chosen to pass between the two data at the high end of the temperature range.

With respect to tellurium (Fig. 8), two additional Ae^{BT} curves are shown. These curves were recently recommended by Lorenz et al. [13,14]. Parker et al. [16] and Lorenz noted that Te releases will be low if unoxidized zircaloy metal is present, whereas Te releases will be significant if most (i.e., $> 90\%$) of the zircaloy is oxidized. The upper and lower curves in Fig. 8 correspond to the oxidized and unoxidized cladding conditions. It was also found in Parker's experiments that Te releases are similarly reduced by the presence of stainless steel.

It is concluded that single values for K_0 and Q in the Arrhenius temperature equation fit the release rate data for a specific fission product over the entire temperature range. It is suggested that the latest data be assembled by the experiment teams at KfK and ORNL in order to establish updated values for K_0 and Q for use in source term calculations.

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