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THE MECHANISTIC PREDICTION OF IODINE AND CESIUM RELEASE FROM LWR FUEL*

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SUMMARY

A theoretical model (FASTGRASS) has been used for predicting the behavior of fission gas and volatile fission products (VFPs) in UO_2-base fuels during steady-state and transient conditions.[1-3] This model represents an attempt to develop an efficient predictive capability for the full range of possible reactor operating conditions. Fission products released from the fuel are assumed to reach the fuel surface by successively diffusing (via atomic and gas-bubble mobility) from the grains to grain faces and then to the grain edges, where the fission products are released through a network of interconnected tunnels of fission-gas-induced and fabricated porosity.

Models[1-3] are included for the effects of the following key variables: production of gas from fissioning nuclei, bubble nucleation and coalescence, bubble migration, irradiation induced re-solution, gas-bubble/channel formation on grain faces, temperature and temperature gradients, interlinked porosity on grain edges, microcracking, experimentally derived steady-state bubble mobilities, and phenomenological modeling of bubble mobilities during transient nonequilibrium conditions. These models are used to calculate

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fission gas release and the swelling due to retained fission-gas bubbles in the lattice, on grain faces, and along the grain edges for steady-state and transient thermal conditions.

As the noble gases have been shown to play a major role in establishing the interconnection of escape routes from the interior to the exterior of the fuel,[4,5] a realistic description of VFP release must a priori include a realistic description of fission gas release and swelling. In addition, as the VFPs are known to react with other elements to form compounds, a realistic description of VFP release must include the effects of VFP chemistry on VFP behavior. A mechanistic description of VFP behavior was developed by modifying the FASTGRASS fission gas analysis to include theoretical models for the effective production rates of the relevant VFPs, the chemical interactions between the various VFPs, the interaction of the VFPs with the fission gas bubbles, and the migration of the VFPs through the solid UO_2 fuel. In the present treatment, the VFPs I, Cs, and their major reaction products (CsI, Cs_2MoO_4, and Cs_2UO_4) have been included. The formation of Cs_2MoO_4 and Cs_2UO_4 can have a crucial effect on the reactions involving CsI, which are of major concern for deducing the form of the iodine released in LWR power plant accident scenarios.

As the available data are not sufficient to describe the kinetics of VFP chemical reactions, the approach to modeling VFP chemistry used here is to assume that the kinetics of the relevant VFP chemical reactions occur fast enough that chemical equilibrium is maintained. The model calculates the density and chemical form of retained fission products as a function of fuel morphology as well as the amount and chemical form of the released fission

products. At present, no attempt is made to describe the behavior of fission products after release from the fuel (e.g., the behavior of the fission products in the fuel-cladding gap).

In the present theory, atomic iodine can diffuse through solid UO_2 via the following two mechanisms: (a) it diffuses intragranularly through the solid UO_2 (in solid solution), and (b) it diffuses with CsI in fission gas bubbles (in the vapor phase). As a reliable calculation (and measurement) of iodine solubility is not currently available, the model for iodine solubility is characterized by the equation

$$I_{TOT} = \alpha(I)_a + (1 - \alpha)(I)_b, \tag{1}$$

where I_{TOT} and I refer to total iodine and atomic iodine concentrations, respectively, and the subscripts a and b refer to iodine residing in solid solution and in fission gas bubbles, respectively. Equation (1) is more general than the approach used in Ref. 2 in that α can have a value between 0 and 1. As in Ref. 2, CsI can migrate only within fission-gas bubbles. In principle, the solubility coefficient α in Eq. (1) can be determined by comparison of model predictions with experiment.

Figure 1 shows predicted fractional release of iodine as a function of irradiation time, and compares these results with the data of Turnbull and Friskney.[6] To reflect the experimental uncertainty in temperature reported in Ref. 6, three predicted curves are given, corresponding to irradiation temperatures of 1733 ± 40 K. The circles in Fig. 1 represent the fractional release of iodine ($^{131}I + ^{133}I$) calculated from the data by taking into account the respective fission yields of ^{131}I and ^{133}I. Figure 1 was generated using a value of 0.6 for the solubility coefficient, α, defined in

Eq. (1). Thus, ~40% of the atomic iodine is assumed to be within the fission gas bubbles, with the other 60% existing in solid solution within the UO_2 lattice. The predictions of the theory for this value of α are in reasonable agreement with the data. In addition, it is predicted that the majority of the iodine released during this experiment occurred as CsI. Unfortunately, no experimental information on the chemical form of the released iodine is available at this time.

FASTGRASS-VFP utilizes an effective generation rate for the VFPs which takes into account precursor effects as well as radioactive decay. The use of an effective generation rate is reasonable after the attainment of quasi steady-state conditions in the fuel such that the VFP concentrations are relatively stable. It is estimated that for (^{131}I + ^{133}I), steady state would occur after several months of irradiation. The fact that the FASTGRASS-VFP-predicted iodine release agrees with the data (as shown in Fig. 1) and the experimental observation[7] that iodine release is qualitatively similar to noble gas release provide additional support for the hypothosis that the noble gases create the majority of the escape paths for VFPs such as I and Cs.

Figure 2 shows the predictions of the theory for total iodine release at 1733 K as a function of irradiation time for various values of the iodine solubility coefficient, α. As in Fig. 1, the circles in Fig. 2 represent the fractional release of iodine (^{131}I + ^{133}I) calculated from the data. The results shown in Fig. 2 demonstrate that the predicted iodine release is a strong function of the iodine solubility. Because of the experimental temperature uncertainties for the data shown in Figs. 1 and 2, it is difficult to suggest a value for α based on agreement between theory and experiment. Clearly, additional experimental and thoeretical work is needed in order to further resolve the question of iodine solubility in UO_2 fuels.

Current improvements in FASTGRASS include a grain-growth/grain-boundary sweeping model. This model has been developed as part of an effort to address the behavior of fission products in UO_2 fuel during steam oxidizing conditions. FASTGRASS calculations have been performed for out-of-pile transient tests on irradiated fuel in a flowing steam environment carried out at Oak Ridge National Laboratory, and for the in-pile severe fuel damage tests SFD-ST and SFD 1-1 carried out in the power burst facility at the Idaho National Engineering Laboratory. The results of the calculations are discussed in relation to fission product behavior during these tests, and to the relative contributions of fuel liquefaction and grain boundary sweeping to fission product release.

REFERENCES

1. J. Rest, "The Prediction of Transient Fission-Gas Release and Fuel Microcracking Under Severe Core-Accident Conditions," Nucl. Technol. 56, 553 (1982).

2. J. Rest, "Evaluation of Volatile and Gaseous Fission Product Behavior in Water Reactor Fuel Under Normal and Severe Core Accident Conditions," Nucl. Technol. 61, 33 (1983).

3. J. Rest, "An Improved Model for Fission Product Behavior in Nuclear Fuel Under Normal and Accident Conditions," J. Nucl. Mater., to be published.

4. J. Rest and S. M. Gehl, "The Mechanistic Prediction of Transient Fission-Gas Release from LWR Fuel," Nucl. Eng. Des. 56, 233 (1980).

5. J. A. Turnbull and M. O. Tucker, "Swelling in UO_2 Under Conditions of Gas Release," Philos. Mag. 30, 47 (1972).

6. J. A. Turnbull and C. A. Friskney, "The Release of Fission Products from Nuclear Fuel During Irradiation by Both Lattice and Grain-Boundary Diffusion," J. Nucl. Mater. 58, 331 (1975).

7. A. D. Appelhans and J. A. Turnbull, "Measured Release of Radioactive Xenon, Krypton, and Iodine from UO_2 at Typical Light Water Reactor Conditions, and Comparison with Release Models," NUREG/CR-2298, U. S. Nuclear Regulatory Commission (1981).

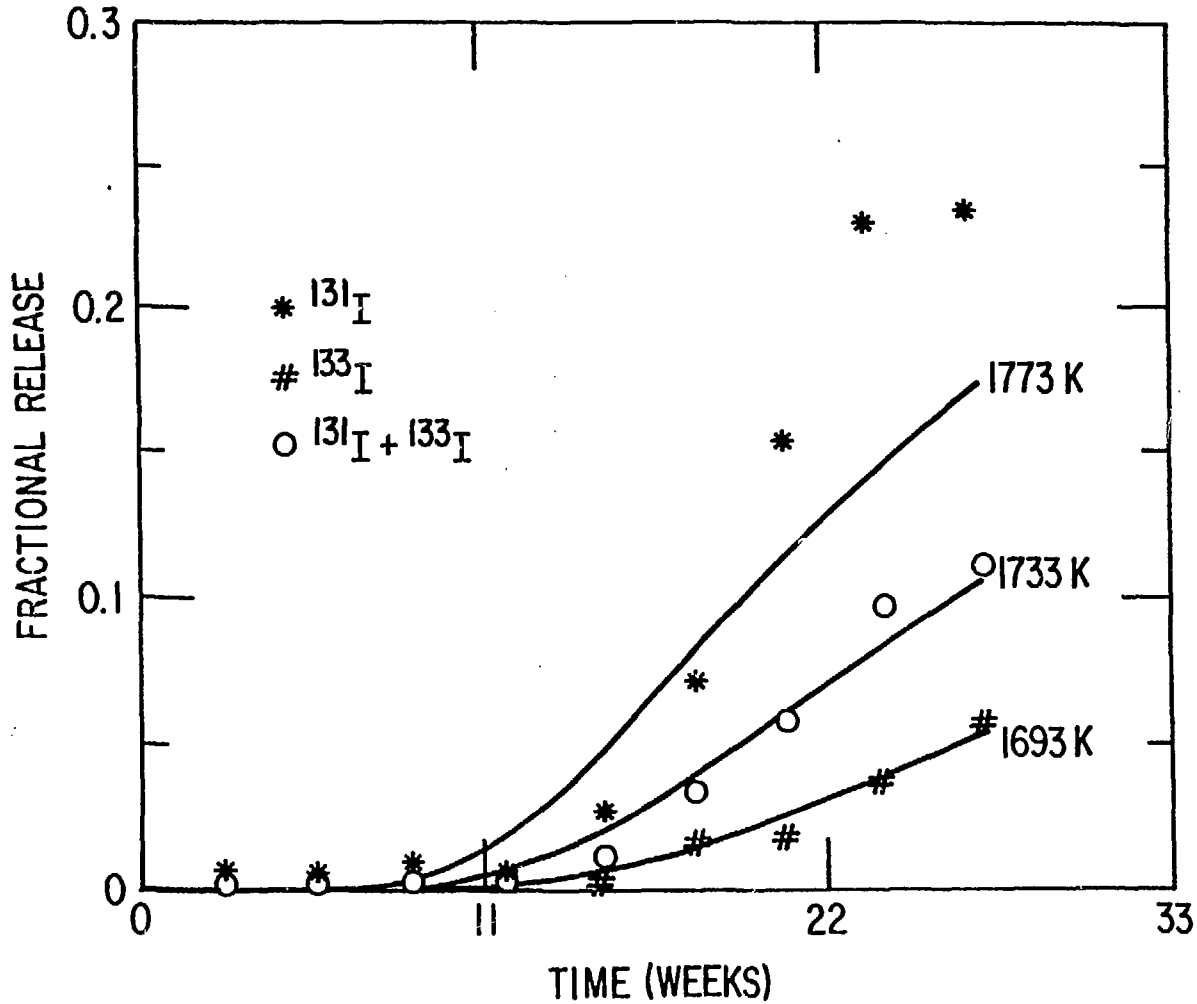

Fig. 1. Predicted Fractional Release of $^{131}I + ^{133}I$ at 1733 ± 40 K (Solid Curves), Compared with Data of Turnbull and Friskney[6] (Symbols).

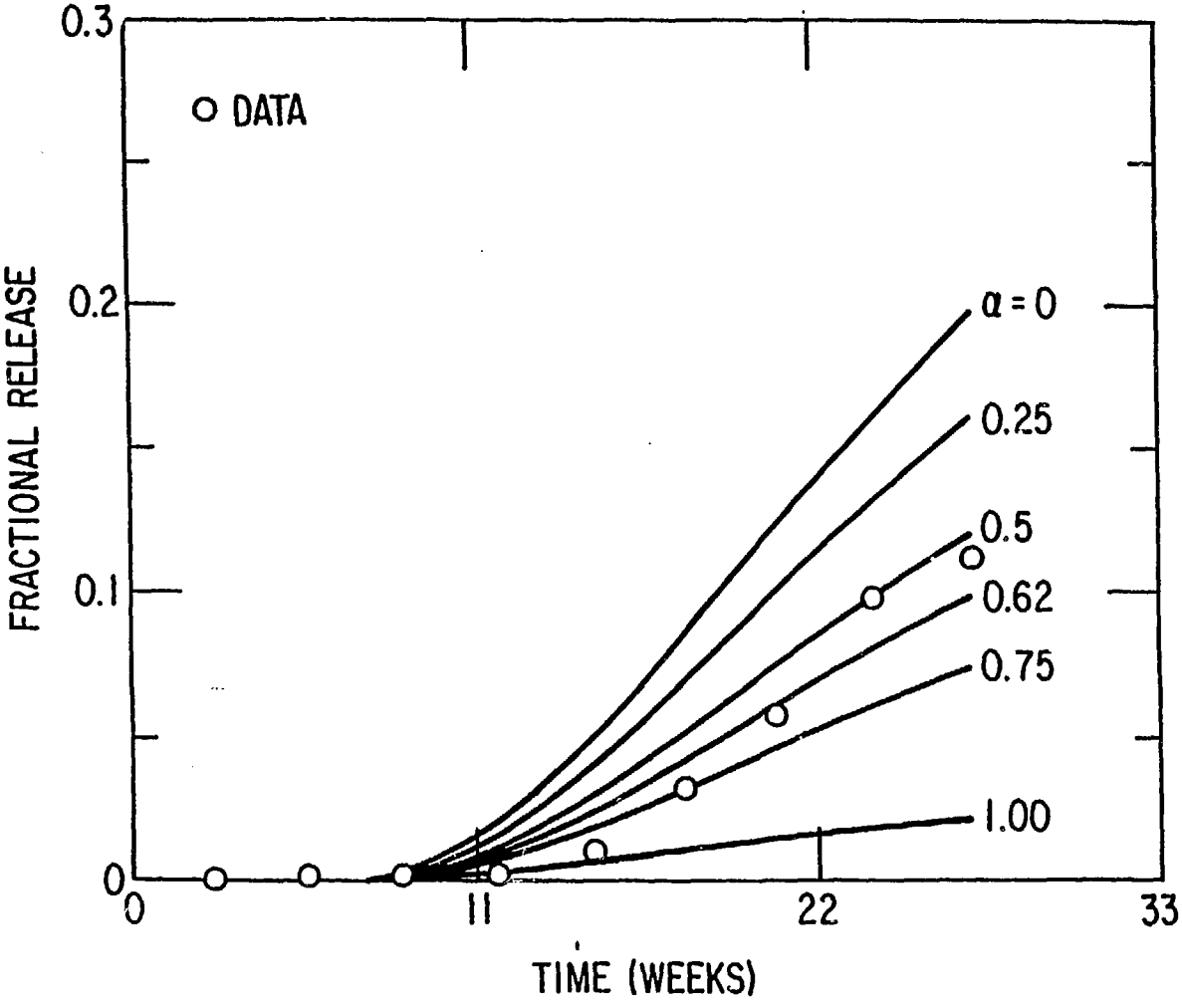

Fig. 2. Predicted Fractional Release of Iodine at 1733 K for Various Values of the Iodine Solubility Coefficient, α (Solid Curves), Compared with the Data of Turnbull and Friskney[6] (Symbols).