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A LIFE-III ISOTOPIC GAS-RELEASE MODEL

by

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ABSTRACT

A spherical diffusion model for isotopic gas release from nuclear-reactor fuel elements is incorporated into the LIFE-III multiregion fuel-behavior code. The predictions of the isotopic gas-release-to-birth ratios model are compared with data from two nuclear fuel elements irradiated in different neutron fluxes. Reasonable agreement between model predictions and experiment is demonstrated.

Also, an argument is given wherein the LIFE-III isotopic gas-release model can be used as an aid in locating failed nuclear-reactor fuel elements.

INTRODUCTION

The measurement of the isotopic content of fission gas released from the fissioning of uranium and plutonium in nuclear reactor fuel elements can lead to information about the physical condition of the fuel element.¹ In particular, the monitoring of fission-gas changes may be effective in identifying fuel-element failures. The present report describes the results obtained from a model incorporated into LIFE-III² to calculate isotopic fission-gas release. The model was applied to two fuel elements: GA-21 irradiated in a thermal flux and F9A-43 irradiated in EBR-II. The vented element GA-21 is typical of the fuel-element design for a gas-cooled fast-breeder reactor. It was irradiated in a thermal flux in sweep-gas capsule GB-10 in the Oak Ridge research reactor poolside facility beginning in August 1972. The purpose of the test

was to obtain information on the release, transport, and trapping of volatile fission products. The element consisted of 0.305-in.-diam (nominal) solid fuel pellets clad in Type 316 stainless steel and cooled with NaK. A detailed description of the GA-21 fuel element in capsule GB-10 can be found in Ref. 3. The terminals of the fission-gas-sampling lines at the various axial locations in the capsule are shown schematically in Fig. 1. Sampling line 2, which enters the capsule at the top, passes down through the annular blanket pellets, and terminates at the top of the fuel column, directly samples the fission-gas release from the top of the fuel column. The data obtained from this sampling line were used for comparison with the LIFE-III gas-release model, since these data most closely approximate the fission-gas content at the surface of the fuel element.

Element F9A-43 was irradiated in subassembly X143 in EBR-II beginning in January 1972. The fuel elements discharged from EBR-II were examined with a gamma scanner located in the hot fuels examination facility adjacent to the reactor. The determinations of ^{133}Xe activities in the fuel-element plena were based on a nondestructive gamma-spectrometric technique for measurement of radioactive fission-gas release from nuclear reactor fuel elements.⁴

DESCRIPTION OF LIFE-III ISOTOPIC GAS-RELEASE MODEL

The method used for computing the release of gaseous and volatile fission products from mixed-oxide fuels is essentially a solid-state diffusion model for spherical fuel particles of a fixed radius; the gas moves to the grain boundaries by diffusion only, and, once on a grain boundary, the gas is instantaneously released. This model, originally developed by Booth,⁵ has been used with the SOLOC computer code³ and has also been adapted to run with the LIFE-II code.⁶ The model assumes a steady-state situation in which the concentration $c(r)$

Fig. 1. Schematic of Capsule GB-10 Showing Sweep-gas Lines. Neg. No. MSD-60276.

of a particular nuclide at any point r along the radius of the grain is obtained by solving the differential equation

$$D \frac{d^2 c(r)}{dr^2} + \frac{2D}{r} \frac{dc(r)}{dr} + B - \lambda c(r) = 0, \quad (1)$$

subject to the boundary condition $c(a) = 0$, where B is the rate of production of the isotope, D is the diffusion coefficient, λ is the rate of decay, and a is the radius of the spherical grain. The solution to Eq. (1) is

$$c(r) = \frac{B}{\lambda} - \frac{B}{\lambda} \frac{a \sinh(r\sqrt{\lambda/D})}{r \sinh(a\sqrt{\lambda/D})}. \quad (2)$$

Integrating the amount of fission gas passing the boundary and dividing by B/λ gives R/B , the fraction of the total equilibrium amount of gas outside the solid after steady state has been attained

$$R/B = 3(D'/\lambda)^{1/2} \left[\coth\left(\frac{\lambda}{D'}\right)^{1/2} - (D'/\lambda)^{1/2} \right], \quad (3)$$

where R is the amount of fission gas released, and $D' = D/a^2$. The radius a of the spherical grain is chosen to be equal to the average grain size of the fuel. However, it is only the ratio D/a^2 that appears in Eq. (3). The diffusion coefficient D is assumed to have an Arrhenius behavior, that is

$$D = D_0 \exp(-Q/kT),$$

where Q is the activation energy in eV, k is the Boltzmann constant = 4.7841×10^{-5} eV/°R, and T is the temperature in °R. The activation energies for krypton and xenon have been taken from Findlay et al.,⁷ i.e.,

	<u>Q(eV)</u>
Kr	1.62
Xe	1.89,

and the preexponential terms are the same as those used by Campana⁶

	<u>D_0 (cm²/sec)</u>
Kr	4.439×10^{-11}
Xe	3.917×10^{-12} ,

where $a = 8 \times 10^{-4}$ cm.

The present diffusion model uses the temperature and power distribution computed by LIFE-III. The fuel was divided into one axial segment and ten annular rings. The average temperature in each incremental volume was used to compute incremental-volume-release isotopic fractions. The release fractions of the volume increments were then weighted by the associated volume fractions and the fission-product generation distribution to arrive at the overall release from the fuel rod.

Element F9A-43 was irradiated in EBR-II in position 6A2 during Runs 54 and 55 and in position 6A1 during Runs 56 through 58. Table I shows the power history input to LIFE-III and the cladding outside diameter (OD) peak temperatures. The inlet coolant temperature was 700°F. The fuel element was irradiated to 115.5 full-power days, corresponding to 2.54 at. % burnup. Table II shows the power history for GA-21 with the associated cladding outside diameter temperatures. Table III lists some of the materials information for elements F9A-43 and GA-21.

RESULTS

The results of the LIFE-III modeling effort are shown in Figs. 2-5. In Fig. 2, R/B is plotted versus λ for various krypton and xenon isotopes. The ¹³³Xe half-life for GA-21 was modified to an effective half-life = $0.693 / [\lambda(^{135}\text{Xe}) + \sigma_a(^{135}\text{Xe}) \phi_{th}]$ to account for the thermal absorption of neutrons

TABLE I. Power History for Fuel Element F9A-43

Time Interval, h	Reactor Power, MW	Cladding OD Temp., °F	Time Interval, h	Reactor Power, MW	Cladding OD Temp., °F		
<u>RUN 54</u>			<u>RUN 56C</u>				
1	0 → 0.05	899	1	0 → 0.05	909		
12	0.05 → 29.5		10.5	0.05 → 31.8			
14	29.5 → 62.5		47.5	31.8 → 62.5			
578	62.5		575	62.5			
1	62.5 → 0	900	1	62.5 → 0	902		
<u>RUN 55A</u>			<u>RUN 57</u>				
1	0 → 0.05		1	0 → 0.05			
11	0.05 → 34.3		15	0.05 → 34.4			
11.5	34.3 → 62.5		28.5	34.4 → 62.5			
301.5	62.5	530.5	62.5				
1	62.5 → 0	902	1	62.5 → 0	908		
<u>RUN 55B</u>			<u>RUN 58A</u>				
1	0 → 0.05		1	0 → 0.05			
4	0.05 → 29.8		4	0.05 → 29.5			
10.5	29.8 → 62.5		23.5	29.5 → 62.5			
212.5	62.5		241.5	62.5			
1	62.5 → 0	902	1	62.5 → 0	905		
<u>RUN 56A</u>			<u>RUN 58B</u>				
1	0 → 0.05		1	0 → 0.05			
11	0.05 → 11.3		1	0.05 → 29.5			
13	11.3		14.5	29.5 → 62.5			
1	11.3 → 0		120.5	62.5			
<u>RUN 56B</u>			1	62.5 → 0	905		
<u>RUN 56B</u>			<u>RUN 58C</u>				
1	0 → 0.05	1	0 → 0.05				
1	0.05 → 0	7.5	0.05 → 29.5				
		13.5	29.5 → 62.5				
		212	62.5				
		1	62.5 → 0				

TABLE II. Power History for Element GA-21

Time Interval, h	Power, kW/ft	Cladding OD Temp., °F
1	0 → 0.01	
9	0.01 → 12	
6,559	12	1049
10	12 → 13.5	
4,990	13.5	1166
10	13.5 → 14.8	
10,990	14.8	1267
1	14.8 → 0	

TABLE III. Materials Information for Elements F9A-43 and GA-21

Materials Properties	Elements	
	F9A-43	GA-21
Fuel diameter, in.	0.215	0.251
Fuel length, in.	13.49	8.876
Cladding length, in.	41.97	14.18
Inside cladding radius, in.	0.110	0.152
Outside cladding radius, in.	0.125	0.176
PuO ₂ , wt %	0.25	0.1225
Initial pellet density, g/cm ³	10.02	9.718

Fig. 2. Release-to-birth Ratio Calculated by LIFE-III, as a Function of Decay Constant, for Various Xenon and Krypton Isotopes Compared with Actual Data at ~ 2.5 at. % Burnup. Neg. No. MSD-60280.

Fig. 3. Release-to-birth Ratio vs Decay Constant Calculated by LIFE-III for Element GA-21 for Various Xenon and Krypton Isotopes as a Function of Power and Burnup Level. Neg. No. MSD-60279.

Fig. 4. Release-to-birth Ratio vs Burnup Calculated by LIFE-III for ^{133}Xe and ^{135}Xe for Element 6A-21 Compared with Actual Data. The solid curves are to guide the eye. Neg. No. MSD-60277.

Fig. 5. Release-to-birth Ratio vs Burnup Calculated by LIFE-III for ^{133}Xe and ^{135}Xe for Element F9A-43. Neg. No. MSD-60278.

$(\sigma_a \phi_{th} = 1.232 \times 10^{-5} \text{ sec}^{-1})$. As can be seen in Fig. 2, the agreement with the data is quite reasonable, although no quantitative statements concerning this particular model can be made at the present time because the accuracy of the data is not known. Figure 3 shows predicted R/B versus λ for the various krypton and xenon isotopes at three GA-21 power and burnup levels. The sensitivity of the release-to-burnup ratio to reactor power level, as given by this model, will be tested as soon as the 13.5 and 15 kW/ft data for GA-21 become available. In Figs. 4 and 5, $(R/B)^{133}\text{Xe}$ and $(R/B)^{135}\text{Xe}$ are plotted versus burnup for the GA-21 and F9A-43 elements, respectively. The solid curves in Fig. 4 show the approximate shape of the GA-21 data and the LIFE-III predictions. For burnup < 1 at. %, the predictions of the LIFE-III isotopic gas-release model for ^{133}Xe differ markedly from the data. However, for low burnup, one would not expect this model to work well, because the assumptions for the diffusion mechanism included the condition that an equilibrium situation had been attained. Furthermore, the early burnup history of an oxide fuel is characterized by comparatively rapid structural changes with accompanying complex fission-gas behavior. One would expect the isotopic LIFE-III gas-release model to be more realistic at a higher burnup where the fission gas is in a less transient state than at low burnup where the nucleation, growth, coalescence, and migration of fission-gas bubbles is expected to play a dominant role.^{8,9} The agreement between prediction and data for element GA-21 is much better for ^{135}Xe than for ^{133}Xe , as shown in Fig. 4. Although the reason for this phenomenon is not clear at the present time, it is important to note that the measurement of the release rate for ^{135}Xe is more difficult than for ^{133}Xe because of the shorter half-life of ^{135}Xe . An unknown path length exists between where the gas is released in the fuel and where the gas is sampled. The correction to the release rate for radioactive decay of the gas between the release point and the sampling

point becomes more critical as the half-life of the isotope decreases. The release-to-birth ratio predictions for ^{133}Xe and ^{135}Xe for element F9A-43, as a function of burnup, are illustrated in Fig. 5. When the F9A-43 data become available, the sensitivity of the LIFE-III isotopic gas-release model to burnup for this element can be checked. The sensitivity of this model to burnup is an important parameter for establishing the accuracy to which the model can pinpoint failed fuel elements.

At any point in the fuel burnup, the isotopic content of the fission gas across the matrix of the reactor core will be temperature dependent. Once fuel-element failure has occurred, the general location of the failed element may be inferred from a knowledge of the core-temperature profile. Since, in a failure situation, the amount of gas released is contingent on the type of failure and on the path the fission gas takes to the detector, the proper quantity to measure is the ratio of release-to-birth values for different isotopes rather than the release-to-birth ratio for one particular isotope.

CONCLUSIONS

Based on the available data at the time of the present report, the spherical diffusion model incorporated into LIFE-III gives reasonable agreement with experiment. The measurement of the relative release-to-birth ratios for different radioactive isotopes is a possible quantity from which to infer the general location of a failed fuel element. A good test for this LIFE-III gas-release model would be to compare its predictions for relative release-to-birth ratios, as a function of temperature, with the values obtained from a fuel element run to failure in-reactor.

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