MODELING FAILURE MECHANISM OF DESIGNED-TO-FAIL PARTICLE FUEL

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A model to predict failure of designed-to-fail (dtf) fuel particles is discussed. The dtf fuel under study consisted of a uranium oxycarbide kernel coated with a single pyrocarbon seal coat. Coating failure was assumed to be due to fission gas recoil and knockout mechanisms and direct diffusive release of fission gas from the kernel, which acted to increase pressure and stress in the pyrocarbon layer until it ruptured. Predictions of dtf fuel failure using General Atomics' particle fuel performance code for HRB-17/18 and HFR-B1 irradiation tests were reasonably accurate; however, the model could not predict the failure for COMEDIE BD-1. This was most likely due to insufficient information on reported particle fuel failure at the beginning.

KEYWORDS : Designed-to-fail, Dtf, Fuel Particles, Pyrocarbon, PISA, Failure, Fission Gas

1. INTRODUCTION

The dtf fuel particle consists of a spherical kernel and a single pyrocarbon (PyC) seal coat. As its name suggests, the PyC layer fails very early after irradiation takes place, thus, providing a known source. It simulates failed TRISO fuel with the exposed kernel (i.e., the inner pyrocarbon, SiC, and the outer pyrocarbon layers all fail). Following a failure, radionuclide release commences, and a distinct spike in radioactivity is registered on a detector downstream of the sweep gas.

There has been no model to predict dtf fuel failure as a function of time. It is beneficial to be able to predict how many fuel particles will fail at a certain time so that the time-dependent amount of radioactivity released can be estimated beforehand. In irradiating the dtf fuel, operators would have to wait until a spike in radioactivity is detected. By knowing when the first particle will fail, effort can be concentrated at the right time. Furthermore, knowledge of the time to completion of fuel failure will help in planning the length of the irradiation test. This paper discusses a model to predict the dtf fuel failure probability as a function of time and, correspondingly, burnup and fast fluence. No model of the subsequent fission product release is presented as it is beyond the scope of the present study.

2. DETAILS OF THE DTF FUEL PARTICLES

The General Atomics Company utilized dtf particles to provide a known source term for fission product release in irradiation tests HRB-17/18, HFR-B1, and COMEDIE BD-1. Tests were conducted to generate data for in-pile release of fission gases and metals, plateout of condensable radionuclides, distribution of coolant impurity, corrosion behavior of H-451 graphite, and lift-off of fission products plated out during irradiation. The experiment involved

Table 1. Details of the Designed-to-fail Particles

Kernel	
Composition	$UC_{0.4}O_{1.6}$
Diameter	347 ± 7 μm
Density	10.51 g/cm^3
^{235}U enrichment	19.61 %
Pyrocarbon Seal Coat	
Thickness	22.8 ± 2 μm
Density	1.95 ± 0.01 g/cm^3

irradiating a scaled, pressurized loop with representative fuel, reflector blocks and heat exchanger surfaces. The fuel included a specified number of dtf fuel particles to supply a well-defined quantity of fission products and to assure sufficient release of key fission products. Each of the dtf particles consisted of a UCO kernel coated with a single PyC seal coat. An optimal amount of uranium carbide was present so that all of the oxygen released from fission was taken up by the uranium carbide and metallic fission products. Thus, there was no in-particle carbon monoxide formation. All three irradiation tests used the same batch of dtf fuel. Saurwein [1] summarized details of the dtf particles; information relevant to failure estimation is presented in Table 1.

3. DETAILS OF THE IRRADIATION TESTS

Saurwein [1] gathered details on the three irradiation tests, and a summary of his document relevant to the present study is presented. Full reports for HRB-17/18, HFR-B1, and COMEDIE BD-1 can be found in Refs. 2, 3, and 4, respectively.

4. HRB-17 AND HRB-18

The tests were performed at the High Flux Isotope Reactor at Oak Ridge National Laboratory. Each of the capsules hosted six fuel compacts. Each compact contained 170-180 low-enriched TRISO fuel particles and five dtf particles. The peak particle fuel centerline temperature was 800°C. The fission power was 1.1 W/particle.

The dtf particles all failed within three days after irradiation started. The ionization chamber registered 30 distinct activity spikes in the sweep gas. For both HRB-17 and HRB-18, the first spike occurred approximately 20 hours into the irradiation and the last one at ~ 68 hours. Thus, the failure probability at about one day is approximately 3.3%[1] and reaches 100% at ~ 3 days. The sweep-gas pressure was maintained at 0.1 MPa. Estimates of burnup and fluence at the times of the failures are summarized in Table 2.

5. HFR-B1

The test was carried out in HFR Petten in The Netherlands. Each of the three independently-purged capsules contained twelve compacts with low-enriched UCO TRISO fuel particles and ThO_2 particles. Each compact contained 87 dtf particles. The approximate time-averaged particle fuel temperature for capsules 1, 2, and 3 were 830-900°C, 800-1100°C, and 940-1000°C, respectively. The fission power was 0.08 W/particle. The R/B profiles for each capsule increased after about five effective-full-power days (EFPD) and saturated at approximately 22 EFPD. Thus, the failure probability at ~ 5 EFPD is approximately 0.1%[2], and reaches 100% at about 22 EFPD. The sweep-gas pressure was maintained at 0.1 MPa. Estimates of burnup and fluence at the times of the failures are summarized in Table 2.

6. COMEDIE BD-1

The irradiation work was performed at the COMEDIE loop in the SILOE materials test reactor in Grenoble, France. Each of the compacts in three capsules contained 845 dtf particles, while compacts in one other capsule

[1] Calculated by dividing the number of failed particles, which is 1, by the total number of dtf particles in all compacts, which is 30.
[2] Calculated by dividing the number of failed particles in each capsule, which is assumed to be 1, by the total number of dtf particles in each capsule, which is 1044.

Table 2. Summary of Dtf Fuel Performance During Irradiation Tests

Parameters	Irradiation Test [1]		
	HRB-17/18	HFR-B1	COMEDIE BD-1
Time to failure (EFPD)	1 - 3	5 - 22	20 - 45
Fission power (W/particle)	1.1	0.08	0.055 - 0.073
Time-average fuel temperature (°C)	800	800 - 1100	> 1193
Burnup at failure (% FIMA)	0.3 - 1	0.2 - 1	0.9 - 2
Fast neutron fluence at failure (10^{24} n/m^2, E > 0.18 MeV)	0.3 - 1	0.3 - 1.4	1.5 - 3.3
Sweep gas pressure (MPa)	0.1	0.1	6

contained 1050 dtf particles. They were irradiated for 63 EFPD over three cycles. The minimum dtf temperatures during the first, second, and third cycles were 1282°C, 1193°C and 1248°C, respectively. Only the minimum dtf particle fuel temperature was reported; the actual fuel temperature was unknown. The fission power was 0.055 - 0.073 W/particle. The sweep-gas pressure was maintained at 6 MPa. The detector indicated that particle failure started at about 20 EFPD and was complete at ~ 45 EFPD. Thus, the failure probability at about 20 EFPD is approximately 0.1%[3], and reaching 100% at ~ 45 EFPD. Estimates of burnup and fluence at failures are summarized in Table 2.

7. DTF FUEL FAILURE MECHANISM

Richards [5] and the author believed that the mechanism that caused PyC seal coat failure was an accumulation of fission-gas pressure within the layer. Fission-gas pressure caused a tensile stress in the PyC, and continued gas release finally resulted in layer rupture. Fission gas atoms were released by the direct recoil and knockout process. Although direct diffusion of gaseous fission products out of the kernel at these very early irradiation times was small, it was also included in the model.

8. DIRECT RECOIL

Direct recoil takes place when energetic fission fragments generated near the kernel surface leave the kernel and become embedded in the PyC layer. Olander [6] derived the recoil fraction as

$$f_{rec} = \frac{1}{4}\left(\frac{S}{V}\right)\mu \qquad (1)$$

where

f_{rec} = fraction of recoil rate to the production rate,
S = surface area of the kernel sphere (μm^2),
V = volume of the kernel sphere (μm^3),
μ = range of fission product in the kernel, 6 μm [7].

The release fraction was calculated to be 2.6% for all three irradiation tests. The total Xe and Kr fission gas yield of 32% was taken from Ref. 8. This number was required to calculate the total number of fission-gas atoms leaving the kernel by this mechanism. Although the 6-μm fission product range applies for ^{135}Xe in a UO_2 matrix, the 20% decrease in oxygen atom concentration replaced by carbon atoms probably increases the fission product range slightly. The smaller atomic mass of carbon compared to oxygen reduces the fission product moderation efficiency. This effect was, however, neglected.

The range of recoil fission-gas atoms in a carbon target was determined using the TRIM software, which simulated the distribution of incident ions in the target material by employing all the kinetic phenomena necessary to predict the ion's energy loss [9]. The maximum birth energy of Xe is 67 MeV [7]. From a simulation of 5000 Xe atoms, all injected perpendicular to the carbon surface, the range of Xe in a carbon sheet with 0.5 g/cm^3 density was ~ 10 ± 1 μm. Note that this is the maximum range, as gaseous fission products may leave the kernel with lower energies and may make any angle of incident prior to arrival at the kernel-PyC interface.

9. KNOCKOUT

Fission-product atoms near the kernel surface can be knocked out by energetic fission fragments. The knocked-out atoms have considerably less energy than the direct recoil [5]. Following derivations by Olander [6], the rate of knockout of short-lived fission products at steady-state can be expressed as:

$$f_{ko} = \frac{\alpha_U \mu S \dot{F}}{4N_U V\lambda} \qquad (2)$$

where

f_{ko} = fraction of knockout rate to the production rate,
α_u = ratio of the current of uranium atoms leaving the surface by knockout to the current of all fission fragments leaving the surface,
μ = range of fission product in the kernel, 6 μm [7],
S = surface area of the kernel sphere (μm^2),
$\dot{F}$ = fission rate density (fissions/μm^3-s),
N_u = number density of uranium atoms, 2.4×10^{10} atoms/μm^3,
V = volume of the kernel sphere (μm^3),
λ = radioactive decay constant for each of the short-lived fission product species (s^{-1}).

Table 3. Knockout Yield for each of the Irradiation Tests

Irradiation Test	$\sum$ (f_{ko} × fission yield of each short-lived gaseous species)
HRB-17/18	7.1×10^{-4}
HFR-B1	5.1×10^{-5}
COMEDIE BD-1	4.2×10^{-5}

[3] Calculated by dividing the number of failed particles in each capsule, which is assumed to be 1, by the total number of dtf particles in each capsule, which is taken to be 845.

Olander calculated α_u to be about 10; however, the measured value was ~ 5 [6]. A value of 5 was used in the present study. Table 3 displays the calculated knockout yield multiplied by fission gas yield. f_{ko} is different for each case because the fission rate density, which is directly related to fission power (W/particle) is different for each case. For COMEDIE, the fission power was assumed to be 0.065 W/particle. The quantity, knockout yield × fission yield, for all cases is at least an order of magnitude lower than that due to direct recoil.

10. DIFFUSIVE RELEASE

Fission gas atoms undergo diffusive release from the kernel provided enough time and temperature are given. The Booth Sphere model was used to calculate the release. The model assumes a spherical grain with radius "a" corresponding to the radius of the dtf kernel. Any fission gas atoms arriving at the surface are assumed to immediately release to the PyC coating. Because fission gases are thermodynamically insoluble in the fuel, their concentration at the fuel surface is zero. By solving the diffusion equation subject to appropriate boundary and initial conditions, the fraction release is given by:

$$f_{diff} = \frac{4}{\sqrt{\pi}}\sqrt{\frac{Dt}{a^2}} \quad (3)$$

where

f_{diff} = fraction of fission gas atoms undergoing diffusive release,

D = diffusion coefficient of Xe in a UCO matrix (cm^2/s),

t = time (seconds),

a = radius of the kernel (0.017 cm).

The diffusivity of Xe in a UCO matrix is not known. To allow the computation to be made, the diffusion coefficient of Xe in the UO_2 matrix given by Davies and Long [10] was used:

$$D = 7.6 \times 10^{-6} \exp(-293 \times 10^3 / RT) \quad (4)$$

where

R = Universal gas constant (8.314 J/mol-K),

T = Temperature (K).

The fraction of fission gas atoms that undergoes diffusive release at the end of each irradiation test is calculated as follows: 0.027% for HFR-B1, 0.001% for HRB-17/18, and 0.668% for COMEDIE BD-1. The release is negligible for HFR-B1 and HRB-17/18 because of the relatively low temperature and short time to completion of failure. However, for COMEDIE BD-1, due to the relatively high temperature of 1250°C and the long time to failure completion of 45 days, the diffusive release becomes significant as it accounts for 20% of the total fraction release from the three mechanisms.

11. PREDICTION OF FUEL FAILURE USING PARTICLE FUEL PERFORMANCE CODE

General Atomics' proprietary code named PISA (Particle Irradiation Stress Analysis) is a one-dimensional finite-element code used to analyze stresses in the particle layers and calculate failure fraction [11]. The code is written in C++ and incorporates several phenomena necessary for accurate performance prediction: irradiation-induced dimensional changes for PyC materials, irradiation-assisted creep strain, linear elastic and linear viscoelastic (for PyC) material models, thermal expansion, pressure-induced stress, and time-dependent analysis. Although not related to the present study, the code also models fuel failure by kernel migration and SiC corrosion by fission product attack.

Table 4 lists PyC properties necessary for the code to predict coating failure. The irradiation-induced dimensional changes for PyC material were obtained from General Atomics literature [12]. The equation to calculate the creep coefficient was adopted from General Atomics literature [12], and multiplied by 1.8 to better match experimental data according to Ref. 13. Creep and irradiation-induced strains, however, did not affect the stress in the PyC layer because the time and fluence under consideration were very small.

It was assumed that fission-gas atoms (and probably other metallic fission products) stopped in the PyC caused the first ~ 10-μm region of the layer to quickly become porous. The extent of porosity as a function of time was not known. For simplicity in analyzing the problem, all gas atoms were assumed to gather into a hypothetical spherical shell 10 microns away from the kernel surface,

Table 4. Required PyC Properties

Properties	Values [12]
Young's modulus	3.3×10^4 MPa
Poisson's ratio	0.23
Creep Poisson's ratio	0.4
Creep coefficient	2.21×10^{-4} 1/MPa at 1000°C
Thermal expansion coefficient	5.57×10^{-6} 1/K
Characteristic strength (σ_0)	18.53 MPa × $m^{3/9.5}$
Weibull modulus (m)	9.5
Bacon anisotropy factor (BAF)	1.036

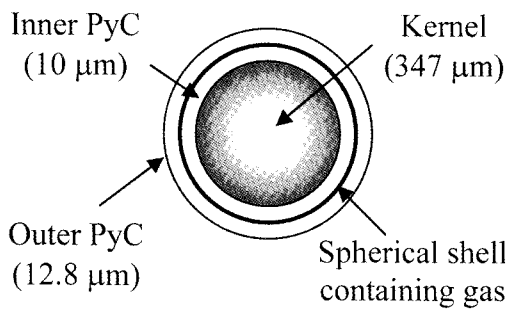

Fig. 1. Representation of the Dtf Fuel with the Assumption of Fission Gas Collecting in the Spherical Shell

occupying a certain fixed thickness that did not change with burnup. For convenience, the PyC region inside this shell was called the "inner PyC," and the PyC outside of this shell was called the "outer PyC." Fission gas arriving at the inner PyC increased pressure in the annulus. Figure 1 shows a drawing of the dft fuel sphere incorporating this assumption.

To model this geometry in PISA, the PyC seal coat was divided into two concentric layers - the inner one being 10-μm thick and the outer one being 12.8-μm thick, as shown in Figure 2. Fission-gas pressure history accumulated in the hypothetical spherical shell was calculated and input into PISA. Gas pressure history was

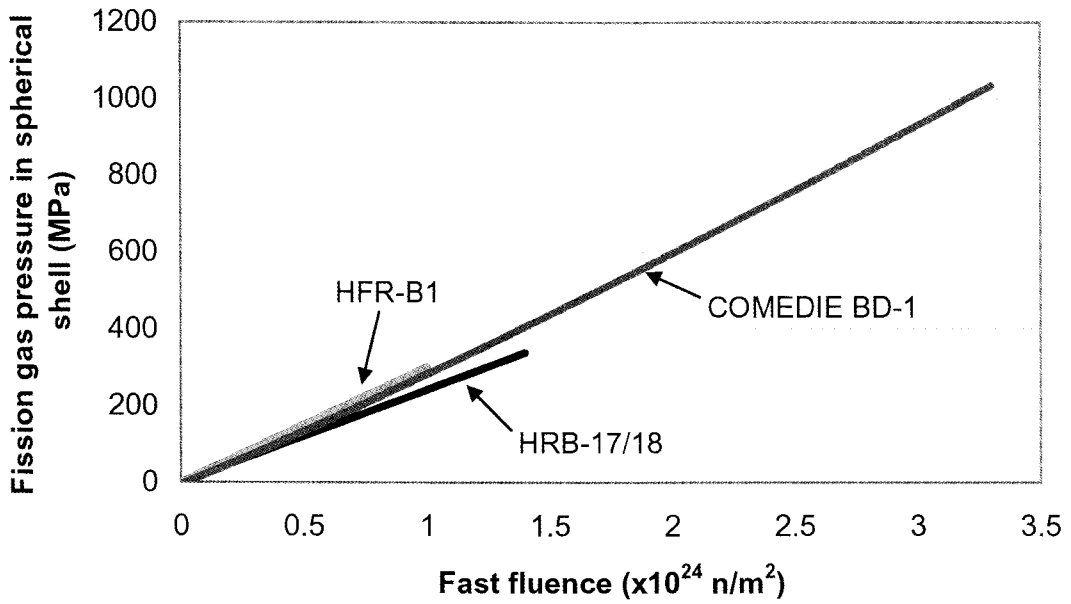

Fig. 2. Fission-gas Pressure History Accumulated in the Hypothetical Spherical Shell with a Thickness of 3.3 Nanometers

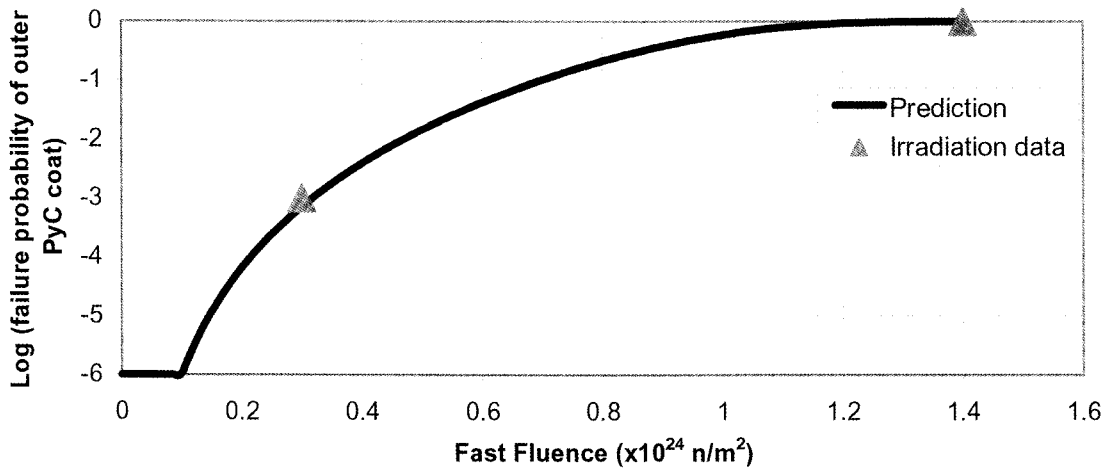

Fig. 3. Failure Probability of the Outer PyC Predicted by PISA for HFR-B1. The Shell Thickness is 3.3 Nanometers

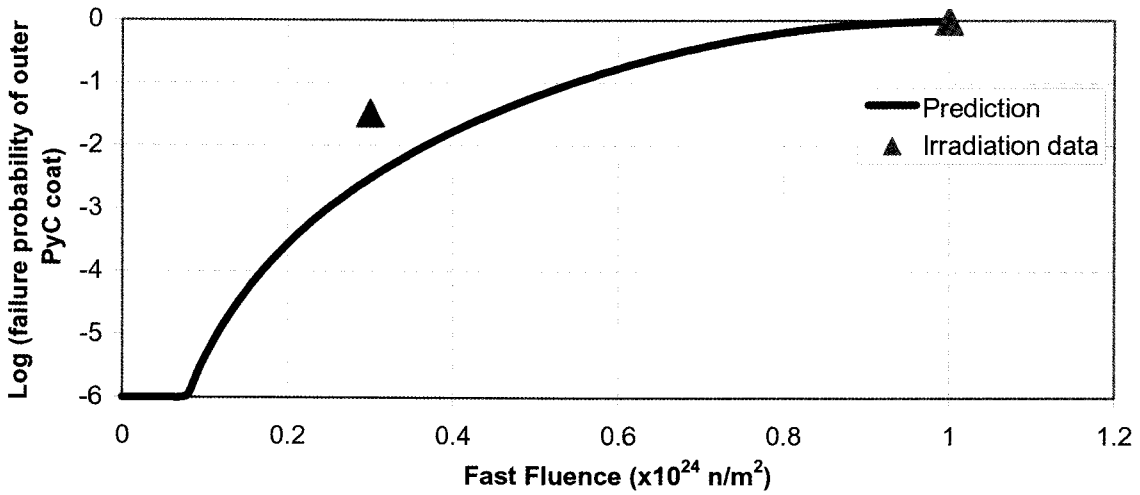

Fig. 4. Failure Probability of the Outer PyC Predicted by PISA for HRB-17/18. The Shell Thickness is 3.3 Nanometers

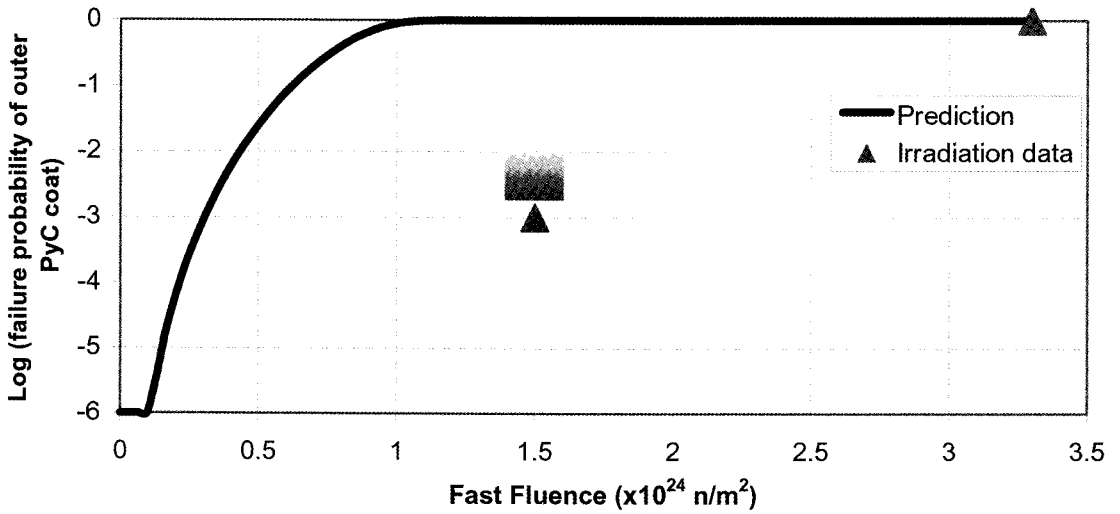

Fig. 5. Failure Probability of the Outer PyC Predicted by PISA for COMEDIE BD-1. The Shell Thickness is 3.3 Nanometers. The Shaded Region Indicates that the Data Point at the Fast Fluence of 1.5×10^{24} n/m^2 could be Placed Higher Due to the Insufficient Data on the Number of Particles that Failed at this Instance

calculated using the Ideal Gas Law. The spherical shell volume was known from an assumed fixed thickness. The number of moles of gas atoms in the shell was known from the total yield and burnup, and the temperature was known. The sweep gas pressure of 0.1 or 6 MPa acted to decrease the tangential stress in the outer PyC layer because it counteracted the fission gas pressure acting on the inner wall of the layer.

The hypothetical shell thickness was varied until the predicted failure curve matched the experimental data from the HFR-B1 irradiation test. The shell thickness is 3.3 nanometers. This makes the model an empirical fit to the data. Although this approach was not physically realistic, this was the only way to utilize the existing PISA code to predict the failure. Failure was assumed when the outer PyC failed. The inner PyC layer was assumed to fail instantly because it would become porous and lose integrity. Figure 3 plots the predicted failure probability of the outer PyC predicted by PISA for HFR-B1.

Data from the HRB-17/18 irradiation test could have been used to determine the shell thickness, or an average value of the hypothetical shell thicknesses that are independently calculated for HFR-B1 and HRB-17/18 could have been employed as well. As will be obvious later, using

the shell thickness determined from HRB-17/18 will result in an accurate prediction of HRB-17/18, but the prediction will slightly overestimate HFR-B1. Using an average value of the shell thickness will somewhat overestimate HFR-B1, while slightly underestimating HRB-17/18.

Figures 4 and 5 display the predicted failure with the same hypothetical shell size of 3.3 nanometers used in HFR-B1. Quite good agreement to the experimental data points was observed for HRB-17/18. For the COMEDIE case, the prediction was considerably off. The temperature for COMEDIE was set at 1250°C; the fission power was assumed to be 0.065 W/particle.

For the COMEDIE BD-1 irradiation test, the irradiation data only stated that the detector indicated that particle failure started at ~ 20 EFPD with a fast fluence of ~ 1.5 $\times 10^{24}$ n/m^2. It does not indicate how many dtf particles failed in this instance. There could be simultaneous dtf fuel failures at ~ 20 EFPD. The shaded region in Figure 5 indicates that the data point at the fast fluence of 1.5 $\times$ 10^{24} n/m^2 could be placed higher. In addition, for all irradiation tests, any manufacturing defect of the dtf particles could have influenced the irradiation performance. Finally, the PyC properties listed in Table 4 may only be fairly representative of the actual PyC properties coated on the dtf particles. Therefore, accurate information on particle fuel properties, irradiation parameters, and results are essential for accurate performance prediction. More irradiation tests are also needed to achieve a universal shell thickness that fits a large number of irradiation tests over a wide range of irradiation parameters. Nonetheless, using the available information, predictions from this model exhibited the right trends and were reasonably correct in two out of three irradiation tests. The model is beneficial for the estimation of the number of fuel particles that will fail at a certain time so that the time-dependent amount of radioactivity released can be estimated beforehand. Furthermore, by knowing when the first particle will fail, effort can be concentrated at the right time. Finally, knowledge of the time to completion of fuel failure will help in planning the length of the irradiation test.

12. CONCLUSIONS

A model to predict failure of dtf fuel particles was discussed. The causes of fuel failure were assumed to be fission gas recoil and knockout mechanisms and direct diffusive release of fission gas from the kernel, which acted to increase stress in the PyC seal coat. Although the diffusive release was negligible for the HRB-17/18 and HFR-B1 irradiation tests, it became significant for COMEDIE BD-1. Fission gas deposited within the first 10 microns of the PyC seal coat (the maximum range of fission gas) was assumed to gather into a hypothetical spherical shell at 10 μm from the kernel surface. Predictions of dtf fuel failure for HRB-17/18 and HFR-B1 irradiation tests were quite accurate; however, the model could not satisfactorily predict failure for COMEDIE BD-1. This was most likely due to the insufficient information on the reported particle fuel failure occurring at ~ 20 EFPD. Accurate information on particle fuel properties, irradiation parameters, and results are essential for precise performance prediction of this model. More irradiation tests will also help achieve a universal shell thickness that fits a large number of irradiation tests over a wide range of irradiation parameters. The model is beneficial for the estimation of the number of fuel particles that will fail at a certain time so that the time-dependent amount of radioactivity released can be estimated beforehand. Furthermore, by knowing when the first particle will fail, effort can be concentrated at the right time. Finally, knowledge of the time to completion of fuel failure will help in planning the length of the irradiation test.

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