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during RIA-Type Transients***

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Effect of Recrystallization in High-Burnup UO_2 on Gas Release during RIA-type Transients

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We recently proposed a model for irradiation-induced recrystallization (grain subdivision) and swelling in UO_2 fuels wherein the stored energy in the material is concentrated in a network of sink-like nuclei that diminish with dose due to interaction with radiation-produced defects [1]. The sink-like nuclei were identified as a cellular dislocation structure that evolves relatively early in the irradiation period. Impurities formed during fissioning of the material diffuse as vacancy-impurity complexes to cell walls where they effectively pin the wall, i.e., dislocation movement to and from the wall is retarded. Not all cell walls are uniformly affected by impurities; the walls containing no impurities continue to undergo subgrain coalescence, which results in viable nuclei for recrystallization. Recrystallization is induced when the energy per nucleus is high enough that the creation of grain-boundary surfaces is offset by the creation of strain-free volumes, with a resultant net decrease in the free energy of the material. This formulation, applied within the context of a mechanistic treatment of fission-gas-bubble behavior, was shown to provide a plausible interpretation of the fission density at which grain subdivision begins, the rate of fuel swelling, and the percentage of gas release in high-burnup UO_2 fuel.

In a recent paper [2], Nogita and Une describe their TEM observations of the formation mechanism of the recrystallized region (the so-called "rim" structure) in high burnup UO_2 fuel. Dislocation density and the volume fraction of small intragranular fission gas bubbles increase with burnup. Low-angle boundaries begin to form above $7.5\text{--}8 \times 10^{26}$ fissions/ m^3 . Analyses of fuel irradiated to 2.1×10^{27} fissions/ m^3 show (a) subdivided grains, 20-30 nm in size, with high-angle boundaries due to the accumulation of an extremely high density of subboundaries; (b) recrystallized grains, 50-200 nm in size and adjacent to the subdivided grain region, which are presumably induced by the stored energy of the matrix; and (c) the absence of bubbles within the recrystallized grains and the appearance of larger bubbles that most likely exist on the new grain boundaries. This is essentially the physical picture that we proposed as the basis of our theory of irradiation-induced recrystallization and swelling [1,3].

It is of considerable interest (4) to explore the consequences of recrystallization on gas release during a reactivity initiated accident (RIA). In the absence of recrystallization, gas release during RIA-type transients is generally limited to gas available on grain boundaries and edges due to the very short heatup times (milliseconds), short cooldown periods (seconds), and relatively long intragranular diffusion distances (on the order of micrometers). However, recrystallization provides grain-boundary surfaces that are substantially closer to the gas retained in the bulk material, and thus the potential

for much higher gas release.

The following expression for the fission density, FDX (fission m^{-3}), at which recrystallization will occur can be obtained from Eq. 22 and Eqs. 3, 13, 14, 15, 16, 26, 35, and 36 in Ref. 1:

$$FDX = \frac{E_{sf} \left[9\Omega\dot{f} + 7a \sqrt{\pi B r_{iv} \xi v_v \Omega \dot{f}} e^{-(\epsilon_{vi} - \epsilon_v/2)/kT} \right]}{168\pi r_{sm} k T c_l \left(\xi_{vi} a^2 v_v e^{-\epsilon_{vi}/kT} + \chi_2 \dot{f} \right)}, \quad (1)$$

where r_{iv} is the radius of the recombination volume; r_{sm} is the annihilation radius of a nucleus/vacancy-solute pair; ϵ_v and ϵ_{vi} are the migration energies of vacancies and vacancy-solute pairs, respectively; v_v is the vibration frequency factor for vacancies; ξ and ξ_{vi} are preexponential factors that account for deviations from diffusion in a pure solvent for vacancies and vacancy-solute pairs, respectively; c_l is the solute concentration; χ_2 is defined by $D_{red} = \chi_2 \dot{f}$ where D_{red} is the radiation enhanced diffusion coefficient; B is a conversion factor ($\dot{f} = BK$, where K is the damage rate in displacements per atom per second); Ω is the atomic volume; $\dot{f}$ is the fission rate; T is the absolute temperature; and $a = \Omega^{1/3}$. The parameters in Eq. 1 and the values used in UO_2 are listed below in Table 1. Eq. 1 (or Eq. 22 in Ref. 1) is the basic result of the analysis and is the expression that should be used for all calculations. The other expressions for FDX given in Ref. 1 are approximations valid only in a restricted range and were derived to demonstrate the limiting behavior of the theory.

Table 1

Values of properties used in Eq. 1 for calculation of irradiation-induced recrystallization in UO_2

Parameter	Value	Parameter	Value
E_{sf}	0.62eV	ϵ_v	2.4eV
B	$6 \times 10^{23} m^{-3} s^{-1}$	r_{sm}	$3 \times 10^{-10} m$
Ω	$3.93 \times 10^{-29} m^3$	c_l	1×10^{-8}
r_{iv}	$2 \times 10^{-10} m$	ξ_{vi}	0.01
ξ	0.1	ϵ_{vi}	2.8eV
v_v	$5 \times 10^{13} s^{-1}$	χ_2	$2 \times 10^{-39} m^5$

Figure 1 shows the calculated burnup (i.e., using Eq. 1) at which grain subdivision will occur as a function of fractional radius (upper horizontal scale) and fuel temperature (lower horizontal scale) for a "generic" pressurized water reactor (PWR) irradiation. Also shown is the fuel burnup equivalent to about 50 MWd/kg M. In principle, the rim region in Fig. 1 would have a higher burnup due to an enhanced fission rate. As shown in Fig. 1, fuel burnup exceeds the critical value given by Eq. 1 from the fuel surface (fractional radius = 1) to a fractional radius of about 0.4. Thus, by 50MWd/kg M, approximately 80% of the fuel volume has undergone recrystallization. As seen in Fig. 1 (and in Fig. 11 in Ref. 1), the amount of fuel undergoing recrystallization is a strong function of the temperature distribution within the fuel. In addition, a recrystallized grain structure would not be formed above about 1000 K. This result is in agreement with the conclusion reached by Nogita and Une [1] based on their study of the thermal recovery of point defects and defect clusters in irradiated UO_2 fuels.

Fig. 1. Calculated burnup (using Eq. 1) at which grain subdivision will occur as a function of fractional radius (upper horizontal scale) and fuel temperature (lower horizontal scale) for "generic" PWR irradiation. Also shown is fuel burnup equivalent to about 50 MWd/kg M.

The FASTGRASS code (5) was used to calculate the fission gas behavior during the in-reactor

irradiation and during the RIA-type transient. Figure 2 shows FASTGRASS-calculated gas release as a function of transient time with and without the occurrence of grain recrystallization (i.e., with 0.5 and 10 μm grain size, respectively). Also shown in Fig. 2 is the temperature scenario used for the calculations: a 10 ms heatup to the maximum temperature (2600 K in Fig. 2) followed by a 10 s cooldown. As is shown in Fig. 2, the percentage gas release from recrystallized fuel reaches 36.5% by the end of the transient compared to 0.02% gas release when no recrystallization occurs. Figure 2 also shows that the calculated gas release occurs in steps. The first burst ($\approx 26\%$ release) is initiated after the fuel temperature has reached its maximum value, during the first 2 s of the transient. Subsequently, no gas release occurs for the next 7 s. The final burst of gas ($\approx 10\%$) occurs during the final second of the transient. The step-like nature of the calculated gas release shown in Fig. 2 is due to the buildup, interconnection and venting, collapse, and redevelopment of gas-filled channels on the grain faces.

Figure 3 shows the calculated total transient gas release as a function of the maximum temperature reached during the RIA-type transient. Essentially no gas release is predicted for maximum temperatures of less than ≈ 2200 K. This result is consistent with experimental studies [6] that relate the burst release of fission from high-burnup PWR fuel during a slow ramp (1.7 K/s) and isothermal annealing at 2073 K to the rapid release of fission gas from grain boundaries that had accumulated during the base irradiation. Figure 3 also shows that gas release of $\approx 90\%$ of that retained in the fuel (which for the case under consideration is equal to that generated during the normal irradiation period) is calculated for maximum temperatures of ≈ 3000 K.

Fig. 2. FASTGRASS calculated gas release as a function of transient time with and without grain recrystallization. Also shown is temperature scenario used for the calculations: a 10 ms heatup to the

maximum temperature (2600 K) followed by a 10 s cooldown.

It is clear from the above discussion that recrystallization of high-burnup UO_2 has implications for the potential consequences of severe accident scenarios such as the RIA type discussed in this letter. Grain subdivision also results in higher fuel swelling rates [1]. Thus, the formation of a porous region within the fuel not only can lead to a substantial increase in fission-product release during a transient, but can also contribute to degradation of the fuel thermal conductivity that could result in substantially higher fuel temperatures.

Fig. 3. FASTGRASS calculated total transient gas release as a function of maximum temperature reached during RIA-type transient.

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