A MODEL FOR RELEASE OF FISSION GASES AND VOLATILE FISSION PRODUCTS FROM IRRADIATED UO$_2$ IN STEAM ENVIRONMENT

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Information concerning the release of fission gases and volatile fission products from irradiated UO$_2$ is important for modeling their behavior under accident conditions. There is evidence that fission gas release is enhanced by the presence of steam in the atmosphere in which UO$_2$ is heated. Thus fission gas release from a defected rod in a light water reactor (LWR) under accident conditions would be larger than anticipated from data obtained on fission gas releases in intact fuel rods whose environment does not contain steam. A model that is proposed in this Letter provides an estimate of the fractional release of volatile fission products from UO$_2$ heated in a steam environment and should be applicable to releases from defected rods under LWR accident conditions.

The basis for the model is the well-established fact that the rate of sintering of UO$_2$ is significantly greater in a steam atmosphere than in an inert or reducing atmosphere.[1-4] It has been shown that the release of volatile fission products in irradiated UO$_2$ fuel rods occurs concurrently with grain growth in the UO$_2$ (Refs. 5, 6, and 7). Therefore, it is proposed that the rate of release is given by the rate of sintering in steam.

The enhanced release of fission gases from UO$_2$ heated in steam as compared to the inert or reducing atmosphere in intact fuel rods is documented in two literature reports. Kurka et al.[8] found an approximate 100-fold increase in the fractional fission gas released from a defected rod operated under conditions for which the fuel was exposed to steam. They suggested the increase was caused by oxidation of the UO$_2$. Lorenz et al.[9] reported enhanced fission gas release from punctured fuel rods heated in steam. In this Letter, we propose a quantitative model for fission gas release in steam compared to inert gas conditions to provide a basis for applying data from intact fuel rod releases to defected rods.

The enhanced rate of sintering of UO$_2$ in steam is a well-known phenomenon.[1-4,10] It has been shown that the basic parameter influencing the rate of sintering and grain growth is the oxygen potential of the system. In particular, the rates are related to the oxygen/uranium ratio of the UO$_2$. A recent study[3] showed that the sintering rate is proportional to the excess oxygen content, X, in UO$_{2+X}$. Thus the rate for UO$_{2.1}$ was about 50 times that for U$_{2.002}$. Another study[4] showed that the sintering rate of UO$_{2.1}$ at 800°C was the same as that of stoichoimetric UO$_{2.0}$ at 1320°C and the measured activation energy showed that this corresponded to a 100-fold increase for constant oxygen/uranium.

Blackadder et al.[5-7] have found that the release of volatile fission products from irradiated fuel is correlated with grain growth of the UO$_2$. Malén[11] has presented a quantitative correlation of iodine release with grain growth kinetics for UO$_2$ fuel in experimental fuel rods, subject to a power ramp.

Bittel et al.[10] have measured the rate of oxidation of pellets of stoichoimetric UO$_2$ in steam. In their work, they showed that grain growth accompanied oxidation. We assume that release of fission gas and volatile fission products occurs with oxidation and grain growth. Bittel et al.[10] used an approximate solution for diffusion in cylindrical pellets that is valid to ~1% for ~94% conversion, and we utilize the same equation, namely,

$$F = 1 - \left[1 - 4\left(\frac{\tau_H}{\pi}\right)^{1/2}\right]\left[1 - 4\left(\frac{\tau_\rho}{\pi}\right)^{1/2} + \tau_\rho\right] , \qquad (1)$$

in which

F = fractional release of volatile fission product

$\tau_L = D_c t/L^2$

t = time (s)

L = height (H) or radius (ρ) of a fuel pellet

D_c = chemical diffusion constant representing penetration of oxidant into the UO$_2$.

Bittel et al. obtained the following expression for D_c for UO_2 in steam:

$$D_c \ (m^2/s) = 9.9 \times 10^{-3} \exp(-28\ 600/T) \ , \qquad (2)$$

with T in Kelvins.

A further extension of the model can be included to account for the intrinsic volatility of the fission product considered. The factor $[1 - \exp(-P_i/P_T)]$, in which P_T is the total pressure in the system and P_i is the vapor pressure of the volatile fission product, can be used for that effect. The fission gases, of course, all have large values for P_i; therefore, this factor is unity for the fission gases. For less volatile substances, the factor depends on the vapor pressure of the chemical form in which the fission product exists in the system. For iodine, the equilibrium chemical form is expected to be cesium iodide, so its vapor pressure should be used for P_i. For the present Letter, we restrict our discussion to the very volatile fission gases, for which the volatility term is unity.

With Eq. (1), one can estimate the fractional fission gas released from irradiated UO_2 fuel after various times at constant temperature in a steam atmosphere. Typical values are shown in Table I.

For an inert gas environment, i.e., for undefected fuel rods, Malén[11] has proposed the following equations for volatile fission product release based on grain growth rates:

$$F = 1 - \left(1 + \frac{2kt}{d_0^2}\right)^{-3/2} \qquad (3)$$

and

$$k \ (m^2/s) = 1.46 \times 10^{-8} \exp(-32\ 100/T) \ , \qquad (4)$$

in which d_0 is the initial grain diameter. We have used $d_0 = 10 \ \mu m$ as a likely value. The fractional releases calculated from this model are also presented in Table I.

The results show that release in steam is much more rapid than release in inert atmosphere. The ratios of the fractions released in the two atmospheres vary with time

and temperature because the two models have somewhat different dependence on those parameters. In general, however, the ratios of releases lie in the range from 50 to several hundred. Those ratios are in keeping with the only quantitative estimate available, namely Kurka et al.,[8] who gave an approximate value of 100. Lorenz et al.[9] indicated an increased release of fission gas when fuel was heated in steam compared with the amount released in the intact rod during reactor operation. However, there is not sufficient information about time and fuel temperature during reactor operation to make a quantitative estimate of the ratio.

The model is obviously in a preliminary stage of development. However, it is useful for estimating values for accident sequences for which no other model is presently available.

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TABLE I

Fractional Release of Fission Gases from UO_2 Fuel Heated Isothermally
in Steam Atmospheres and in Inert Atmospheres

Time (s)	Fraction Released									
	1300 K		1400 K		1500 K		1600 K		1700 K	
	Steam[a]	Inert[b]	Steam[a]	Inert[b]	Steam[a]	Inert[b]	Steam[a]	Inert[b]	Steam[a]	Inert[b]
10^3	0.04	– – –	0.06	– – –	0.12	– – –	0.22	– – –	0.34	3×10^{-3}
10^4	0.09	8×10^{-5}	0.19	5×10^{-4}	0.36	2×10^{-3}	0.58	8×10^{-3}	0.82	3×10^{-2}
10^5	0.27	8×10^{-4}	0.53	5×10^{-3}	0.83	2×10^{-2}	– – –	8×10^{-2}	– – –	0.2
10^6	0.69	8×10^{-3}	– – –	0.046	– – –	0.19	– – –	0.49	– – –	0.8
Time (s) for $F = 0.5$	4.3×10^5	1.1×10^8	8.8×10^4	1.8×10^7	2.3×10^4	4×10^6	7×10^3	1×10^6	2.5×10^3	3×10^5

[a]Calculated from Eq. (1) with $H = 13$ mm and $\rho = 6.4$ mm.
[b]Calculated from Eq. (3) with $d_0 = 10 \ \mu m$.

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