A MODEL FOR RELEASE OF FISSION GASES AND VOLATILE FISSION PRODUCTS FROM IRRADIATED UO₂ IN STEAM ENVIRONMENT

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Received October 31, 1980
Accepted for Publication November 18, 1980

Information concerning the release of fission gases and volatile fission products from irradiated UO₂ 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 UO₂ 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 UO₂ 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 UO₂ is significantly greater in a steam atmosphere than in an inert or reducing atmosphere. It has been shown that the release of volatile fission products in irradiated UO₂ fuel rods occurs concurrently with grain growth in the UO₂ (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 UO₂ heated in steam as compared to the inert or reducing atmosphere in intact fuel rods is documented in two literature reports. Kurka et al. 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 UO₂. Lorenz et al. 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 rate of sintering of UO₂ in steam is a well-known phenomenon. 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 UO₂. A recent study showed that the sintering rate is proportional to the excess oxygen content, \( X \), in UO₂. Thus the rate for UO₂, which was about 50 times that for U₂O₃. Another study showed that the sintering rate of UO₂, at 800°C was the same as that of stoichoimetric UO₂ at 1320°C and the measured activation energy showed that this corresponded to a 100-fold increase for constant oxygen/uranium.

Blackadder et al. have found that the release of volatile fission products from irradiated fuel is correlated with grain growth of the UO₂. Malén has presented a quantitative correlation of iodine release with grain growth kinetics for UO₂ fuel in experimental fuel rods, subject to a power ramp.

Bittel et al. have measured the rate of oxidation of pellets of stoichoimetric UO₂ 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. used an approximate solution for diffusion in cylindrical pellets that is valid to \( \sim 1\% \) for \( \sim 94\% \) conversion, and we utilize the same equation, namely
\[
F = 1 - \left[ 1 - 4 \left( \frac{\tau_H}{\tau} \right)^{1/2} \right] \left[ 1 - 4 \left( \frac{\tau_D}{\tau} \right)^{1/2} + \tau_H \right],
\]
in which
\[
F = \text{fractional release of volatile fission product}
\]
\[
\tau_L = \frac{D_c t}{L^2}
\]
\[
t = \text{time (s)}
\]
\[
L = \text{height (H) or radius (ρ) of a fuel pellet}
\]
\[
D_c = \text{chemical diffusion constant representing penetration of oxidant into the UO₂}
\]
Bittel et al. obtained the following expression for $D_c$ for UO₂ in steam:

$$D_c \text{ (m}^2/\text{s}) = 9.9 \times 10^{-3} \exp(-28600/T),$$  \hspace{1cm} (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 UO₂ 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, Malén¹¹ has proposed the following equations for volatile fission product release based on grain growth rates:

$$F = 1 - \left(1 + \frac{2k}{d_0^3/2}\right)^3,$$  \hspace{1cm} (3)

and

$$k \text{ (m}^2/\text{s}) = 1.46 \times 10^{-8} \exp(-32100/T),$$  \hspace{1cm} (4)

in which $d_0$ is the initial grain diameter. We have used $d_0 = 10 \mu\text{m}$ 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.,⁹ 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.

### REFERENCES


### TABLE I

Fractional Release of Fission Gases from UO₂ Fuel Heated Isothermally in Steam Atmospheres and in Inert Atmospheres

<table>
<thead>
<tr>
<th>Time (s)</th>
<th>1300 K</th>
<th>1400 K</th>
<th>1500 K</th>
<th>1600 K</th>
<th>1700 K</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Steam*</td>
<td>Inert*</td>
<td>Steam*</td>
<td>Inert*</td>
<td>Steam*</td>
</tr>
<tr>
<td>10³</td>
<td>0.04</td>
<td>---</td>
<td>0.06</td>
<td>---</td>
<td>0.12</td>
</tr>
<tr>
<td>10⁴</td>
<td>0.09</td>
<td>8 x 10⁻⁵</td>
<td>0.19</td>
<td>5 x 10⁻⁴</td>
<td>0.36</td>
</tr>
<tr>
<td>10⁵</td>
<td>0.27</td>
<td>8 x 10⁻⁴</td>
<td>0.53</td>
<td>5 x 10⁻³</td>
<td>0.83</td>
</tr>
<tr>
<td>10⁶</td>
<td>0.69</td>
<td>8 x 10⁻³</td>
<td>---</td>
<td>0.046</td>
<td>---</td>
</tr>
<tr>
<td>Time (s) for $F = 0.5$</td>
<td>4.3 x 10⁵</td>
<td>1.1 x 10⁶</td>
<td>8.8 x 10⁴</td>
<td>1.8 x 10⁷</td>
<td>2.3 x 10⁴</td>
</tr>
</tbody>
</table>

*Calculated from Eq. (1) with $H = 13 \text{ mm}$ and $p = 6.4 \text{ mm}$.

*Calculated from Eq. (3) with $d_0 = 10 \mu\text{m}$.


