Xe Release from UO₂ upon Oxidation

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For estimating fission gas release from fuel under accident conditions, the present authors have previously measured Xe release from U upon oxidation by CO₂ and have observed an Arrhenius type temperature dependence(1). Parker has measured Xe-Kr release from UO₂ upon oxidation by air(2), and reported a small and rather complex temperature dependence for this case. The release percentage also differed between these two cases: In the former experiment 40% of the Xe initially present was released at temperature as low as 700°C, whereas in Parker’s case (400°~1,400°C) the largest release was only 18% (at 1,100°C). The present note is on the results of an experiment on Xe release from UO₂ upon oxidation, not in air, but in O₂ at 600°~1,300°C.

Broken pieces of sintered UO₂ (95% T.D.), were vacuum sealed in quartz tubes and irradiated in the JRR-1 research reactor to an integrated neutron flux of \(1 \times 10^{16} \text{nvt}\). After cooling for 1 to 2 weeks, the specimens were heated in an alumina combustion tube connected to charcoal traps with a stop cock in-between. The apparatus was so arranged that the specimen could be heated to present temperatures within a few minutes. The

\[
\frac{\partial}{\partial t} = \frac{1}{1 - \beta} (1 - \kappa_p) \frac{\partial}{\partial r} g_{v}(r) + \frac{\partial}{\partial r} \left( \frac{\partial g_{v}(r)}{\partial r} \right) + \frac{\partial}{\partial r} \left( \frac{\partial g_{v}(r)}{\partial r} \right)
\]

\( (8) \)

The above discussions have been made regarding the point reactor model. The substance of the results is not altered when we take space-dependence into account by means of the fundamental mode \( (g_{v}(r)) \) approximation\(^{20}\). We need only to multiply the amplitude \( A \) by \( V^2 g_{v}(r) \int g_{v} dr \) where \( V \) is the system volume and \( r \) the detector position; \( X_{e, \text{reacted}} \) and the r.h.s. of Eq. (8) should also be multiplied by \( \int g_{v} dr \left( \int g_{v} dr \right)^3 \) and \( \int g_{v} dr / \left( \int g_{v} dr \right)^3 \) respectively. All of these are small but not negligible numerical corrections.

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extent of oxidation was determined from the decrease observed in O₂ pressure. The oxidation completed in about 1 hr, following a pattern of progress similar at all temperatures, as shown in Fig. 1. Upon completion of oxidation the combustion tube was evacuated through traps cooled in liquid N bath to collect fission gas, and the gathered ¹³³Xe was analyzed by means of γ-spectrometer. Finally the specimen was dissolved in nitric acid to liberate the remaining fission gas, and again ¹³³Xe was analyzed to find the total ¹³³Xe initially present in the specimen.

Table 1 summarizes the experimental data, and the resulting release values are plotted in Fig. 2, in comparison with Parker's data. The agreement is good. It is thus confirmed that fission gas release from UO₂ in burning accident should be limited to 20%, irrespective of burning temperature and that there should be no appreciable difference between air and O₂ atmosphere. If the environmental temperature stays high, however, fission gas will continue to be released by diffusion through the higher oxide formed by oxidation. Even while the BET surface area of the oxidation product showed a tendency to decrease with increasing oxidation temperature (see Table 1), the effect of rising diffusion coefficient with increasing temperature overrides the decrease in BET area. As an example, we measured post-oxidation release at two temperatures. The results are compared in Fig. 3, which shows that the amount of release after oxidation, which corresponds to release by diffusion, is much higher at 750°C than at 600°C, even though the BET area of the powder formed at 750°C (5,410 cm²/g) is about ⅔ of that formed at 600°C (1,860 cm²/g). The small and rather complicated temperature dependence seen in Fig. 2 suggests that the release during oxidation is primarily due to liberation upon pulverization by oxidation instead of diffusion.

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