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The diffusion of rare gases in UO_2 has been the subject of a large number of experimental investigations. Lawrence (1) has reviewed the literature through 1977 and has identified variables which appear to exert large effects on the release of rare gases. These include stoichiometry, density and form of the solid, burnup and conditions under which annealing was accomplished (i.e., in vacuum, or in the presence of reactive or inert gases). At 1400°C, the scatter band of the data reviewed by Lawrence covers over four orders of magnitude. By critical assessment of the data, Matzke (2) has estimated the uncertainty in xenon diffusivity at 1400°C at zero burnup as plus-or-minus a factor of ten.

Very few diffusion data exist on iodine and none for cesium or tellurium. Friskney and Turnbull (3) studied in-pile release of iodine and Peehs et al. (4) examined iodine and cesium behavior in post-irradiation anneal experiments. The amount of iodine released from a fuel sample was evaluated by measuring the activity of ^{131}I or ^{133}I trapped by a charcoal filter in ref. 3 or by a liquid-nitrogen cooled surface in ref. 4.

In our experiments, post-irradiation anneal of lightly-irradiated UO_2 was used to measure the release kinetics of the volatile fission products Xe, I, and Te. As the last three are chemically very reactive, it is quite difficult to insure complete collection of material release by means of a gas-sweeping technique. To avoid this difficulty, we measured the fraction of a particular fission product retained in a specimen by gamma-ray spectroscopy before and following an anneal. The method is based on the assumption (proven experimentally) that once released, no fission products return to the specimen. Thus the fractional release is one minus the fraction retained.

By appropriate selection of reactor irradiation and cooling times, the gamma-ray spectrum obtained with a Ge(Li) detector exhibits well-separated peaks by which each of these fission products can be assayed. Iodine behavior was determined by following the intensity of the 365-keV photopeak from decay of ^{131}I. Xenon removal was obtained from the change in intensity of the 81-keV transition in decay of ^{133}Xe. Tellurium release was followed by measurements of the intensity of the 773-keV photopeak in decay of 2.3-hr ^{132}I after the specimen was allowed to decay sufficiently to attain secular equilibrium with the 78-hr ^{132}Te.

Anneals were conducted in flowing hydrogen at temperatures between 1400 - 1700°C. The water content of the hydrogen was controlled to ∿ 100 ppm (monitored by a moisture meter) to assure perfect stoichiometry of the UO_2. As a test of the reliability of the method, no release of neptunium was detected, as would be expected at these temperatures. Both single crystal and conventional pellet specimens were tested.

The results, which up to now have been interpreted by the Booth equivalent sphere model, indicate that the diffusion coefficient of iodine is about twice as large as that of xenon, and that of tellurium nearly 10 times that of xenon. All three specimens exhibit activation energies of ∿ 130 kcal/mole. The result for xenon at 1400°C is 10^{-13} cm^2/s, which is in the middle of the range of most likely values estimated by Matzke(2). The iodine diffusivities are considerably different from those reported by Friskney and Turnbull(3) obtained by in-pile release tests. They report approximately equal diffusivities of xenon and iodine, and both exhibited very low activation energies. The present data on tellurium is the first diffusivity in UO_2 reported for this element. Tests for two different annealing periods suggests that the fractional release is proportional to the square root of the annealing time, indicating a diffusional process. Comparison of the data from polycrystal and single crystal specimens gave equivalent sphere radii (in the Booth model) of 200 ± 100 μm. This result is considerably larger than the equivalent sphere radius of ∿ 60 μm found by Friskney and Turnbull(3), or the grain size in the polycrystalline specimens (∿ 10 μm), and suggests restricted grain boundary diffusion. However, release from the polycrystals was always nearly an order of magnitude larger than that from single crystals (after accounting for different surface-to-volume ratios), so it is clear that grain boundaries in UO_2 do not simply act as fission product traps which open up only upon linkage of gas bubbles formed on them. This behavior is assumed in most fuel performance codes (5,6) and while such an approach may be adequate at high burnup, it is clearly inappropriate for short irradiations. For the very short irradiations used here, neither inter- or intragranular bubbles are present. The release kinetics are direct measures of combined lattice-grain boundary diffusional processes.

Current efforts include the measurement of concentration profiles of fission products in annealed slabs of UO_2. This is accomplished by assay of the intensity of characteristic gamma ray peaks as successive layers (a few tens of microns each) of the specimen are removed by precision grinding. According to the Booth model, no concentration gradient of the diffusing species should be observed over dimensions large compared to the average grain size. However, more physically realistic migration models which include combined grain boundary and lattice diffusion predict nonuniform concentration profiles. The distribution measurement will thus provide crucial information for modeling the basic release process.

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