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EFFECT OF HIGH TEMPERATURE IN-PILE IRRADIATION UPON HIGH DENSITY UO_2 BODIES*

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ABSTRACTEffect of High TemperatureIn-Pile Irradiation Upon High Density UO₂ Bodies

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As a part of the ML-1 fuel evaluation program, high density bulk UO₂ pellets were irradiated to burnups equivalent to 8500 hours of ML-1 reactor operation (3.0 a/o U-235) under temperature conditions approximating those expected in the reactor (1750°F peak cladding surface temperature). Post irradiation examination revealed severe fuel cracking although fission product gas release measurements agreed well with calculated values. No structural changes were revealed by metallography and X-ray diffraction except for a small unidentified phase observed at the fuel grain boundaries. This phase is considered to be the result of impurities or fission product gas sites.

The data obtained on UO₂ fuel is in agreement with that determined in irradiation experiments performed at other facilities. The performance of UO₂ fuel was sufficiently good to permit use in the ML-1 reactor.

I. INTRODUCTION

ML-1 is a gas cooled, mobile power reactor system being designed by Aerojet-General Corporation. The project is supported jointly by the AEC Division of Reactor Development and the U.S. Army.

Reactor fuels selected for the prototype ML-1 were BeO-70 wt% UO_2 and solid UO_2 , the former in the outer ring of fuel pins and the latter in the inner ring (see Figure 1). It was considered desirable to obtain irradiation data on these fuels at near cladding hot-spot temperatures (estimated to be 1750°F). A capsule irradiation was performed to obtain 10,000 hour equivalent burnup data (3.2 a/o U-235) within a relatively short time. To reduce acceleration effects which might obscure results, such as unusually high central temperatures, the specimen diameters were reduced to 0.146-in. O.D. from the reference 0.177-in. O.D.

The irradiation was performed under the joint supervision of personnel from Battelle Memorial Institute (BMI) and Aerojet-General Nucleonics (AGN). Capsule design and fabrication was performed by BMI, specimen design and fabrication by AGN, irradiation was carried out at the Battelle Research Reactor (BRR); post irradiation examination at the Battelle Hot Cell.

of power by reactivity problems. This operating difficulty resulted in the largest portion of the spread observed.

D. POST-IRRADIATION EVALUATION

Post-irradiation examination of the specimens from this capsule commenced in the Battelle Hot Cell Facility shortly after the capsule was discharged.

The examination consisted of these steps: (1) measurement of fission product gas release from the capsule (2) measurement of the dimensions of the irradiated specimens (3) measurement of fission product gas release from the various specimens (4) metallographic evaluation of the fuel and cladding, and (5) determination of burnup and X-ray diffraction analysis of the irradiated fuel.

The dimensions were measured at five points along the specimen length using a standard friction-thimble micrometer and the results compared to the pre-irradiation value. Each specimen was also weighed individually and the results compared with its pre-irradiation value.

After removal of the outer capsule shell (see Figure 3) the atmosphere over the solid NaK bath within the inner capsule was sampled for fission product gas by puncturing the inner capsule in an evacuated chamber. The same procedure was followed on all irradiation specimens. Aliquots of the gases collected were analyzed for the characteristic 0.51 mev gamma ray emitted by Krypton 85 using a gamma ray spectrometer.

Fuel and cladding specimens from specimen numbers 6, 8 and 0 were prepared for microscopic examination by remote metallographic techniques. X-ray diffraction analysis was performed on powder separated from specimen 6 using a shielded General Electric X-ray diffractometer.

III. RESULTS

A. VISUAL AND DIMENSIONAL EXAMINATION

Macroscopic examination of all specimens was performed through a stereo-periscope viewer. All specimens were observed to be intact with little significant change to the cladding, aside from a surface dulling due to prolonged immersion in liquid metal. Figure 4 depicts pre-and post-irradiation views of a typical irradiation specimen, and pre-post cross sections of these fuel specimens.

No dimensional variation was detectable by visual observation. Subsequently, each specimen was measured at five points along its length for diametral variation with a standard, remotely operated micrometer. Specimen length changes were also measured, utilizing similar techniques. The dimensional variations measured on the specimens were considered to be within the experimental error of the equipment used.

B. FISSION PRODUCT GAS RELEASE

The inner capsule was punctured to determine the presence of fission product gases, indicating leaking specimens or diffusion through the specimen cladding. No fission product gases were detected. The major constituent of the gas in the inner capsule was helium, as was to be expected since the capsule had been sealed under a helium atmosphere. All specimens were later gamma-scanned to determine relative activity. The six

The figures illustrate the fuel conditions found in two of the six specimens and show that the structures found in these specimens are greatly similar. A pronounced variation in grain size was apparent in all specimens, a not unexpected condition since central fuel temperatures of 3000 to 3100^oF were predicted for the peak temperature specimen. However, note the striking difference between the post irradiation structures and those of the unirradiated control specimens.

Clearly observable in all photographs ((see arrows)) are grain boundary conditions, some of which may represent fission product gas sites; others, the darker, more angular areas, were probably caused by pull-out which occurred during specimen preparation and etching steps. The bubble-like structure usually associated with UO₂ dispersions that have accumulated sizeable neutron exposures was not noticeable in the specimens examined.

E. X-RAY DIFFRACTION ANALYSIS

Powder samples from specimen 6 were prepared and diffracted to determine the degree of damage incurred in the UO₂ structure. The diffraction analysis indicated very little change in the crystal structure of the UO₂ as a result of irradiation. A lattice contraction of 0.36% and an internal lattice strain of 0.1% was measured, illustrating the minor effects upon structure resulting from this irradiation.

IV. DISCUSSION OF RESULTS

As a result of irradiation there was no apparent effect upon the clad specimen dimensions, indicating that either the fuel swelling was minimal or that the resistance of the cladding to fuel swelling prevented measurable changes. Post irradiation measurement of pellets directly was impossible because of their cracked condition. Radiation-induced UO_2 swelling was not expected at these burnup levels on the basis of work performed by Bettis^{*}. The fuel fracturing observed was not unexpected since such an occurrence has been observed in most irradiations which involved solid UO_2 materials. The thermal shock resistance of this material is relatively low; in the course of its irradiation it was exposed to the shocks and cycling involved in reactor startup and shutdown, and scrams.

The fission product gas release was fairly large, but was within predictions based upon theoretical analysis. The measured Kr-85 gas release in the hottest specimens (6 and 8) were determined as 16.1 and 23.7% of fission product gas released respectively. Calculated values (based upon 10,000 hour operation, 1750°F cladding hot spot temperatures, a .003-in. diametrical gap and a 3007°F fuel center temperature) was found to be 22.8% of fission product gas released. Fuel cracking quite probably influenced fission product gas release. The amount of Kr-85 released by recoil has been estimated as 0.37%, where no cracking was included in the calculations. However, on the basis of the estimated value from the uncracked case, the increase due to cracking is not expected to account for the difference be-

*Bleiberg, Berman et al. WAPD-BT-18

tween this value and that observed in this experiment. Consequently, the gas release is considered to be by diffusion through the fuel itself, probably along grain boundaries. Time at temperature was relatively extended and central fuel temperature were quite high in this experiment, points which support the diffusion contention.

The fuel structure was found to be considerably different than either a pre-irradiated as-fabricated structure or an un-irradiated, heat treated specimen (see Figure 9). There are grain size differences, between pre- and post-irradiation UO_2 materials, which probably account, in part, for the gas releases measured. These increased grain sizes providing longer, uninterrupted paths for diffusion than does the finer grain material. The grain boundary constituents observed are thought to be from three causes, (1) damage to the material as a result of polishing and specimen preparation, and (2) fission product gas pockets which migrated to the grain boundaries, and (3) UO_2 impurities which migrated to the grain boundaries as a result of the extended thermal treatment. The finer voids or particles observed at the grain boundaries have not yet been identified, although chemical etching has a decided effect upon them as Figure 9 shows.

Most of the larger voids probably occurred during specimen preparation. The UO_2 is considerably more brittle after irradiation so that handling which would not cause severe pull-out with un-irradiated material might occur with the irradiated fuel.

The X-ray diffraction analysis was performed to determine whether any damage to the crystal structure could be observed. Bleiberg et al^{*} observed the beginnings of such damage at 16 to 17×10^{20} fissions/cc and suggested that at isolated points the UO_2 crystal structure was destroyed and amorphous areas resulted. From the diffraction results discussed earlier in this report, it appears that such damage has not become noticeable.

* Bleiberg, Berman, et al. WAPD-BT-18

V. CONCLUSIONS

The purpose of this irradiation was to determine the stability and integrity of UO_2 after 10,000 hr equivalent ML-1 reactor operation. Uranium burnup equivalent to 10,000 hr operation was estimated to be 3.52 total a/o U-235, and design specimen surface temperatures of 1750°F were to be attained. On the basis of the post-irradiation examination, it is apparent that the desired goal has been achieved. While fairly extensive fuel cracking occurred and significant quantities of fission gases were released, neither will effect operation of the over-all fuel element. The UO_2 crystal structure was found to be unimpaired by the exposure. The gas pressure exerted will not cause pin failure, neither will fuel swelling be of sufficient magnitude to distort the fuel pin cladding.

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