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Consolidated Fuel Reprocessing Program

DISSOLUTION BEHAVIOR OF FFTF FUEL

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INTRODUCTION:

This paper describes recent studies that are an extension of prior work with UO_2 fuels irradiated in LWRs and with mixed-oxide fuels irradiated in test reactors or the EBR-2. These new studies investigate the dissolution behavior of irradiated fuels currently being discharged from the Fast Flux Test Facility (FFTF) and shipped to the Oak Ridge National Laboratory. Such fuels are representative of the compositions, thermal histories, and burnups expected for future U.S. breeder fuels and will yield data of use in the design and operation of dissolvers for fuel recycle plants.

The irradiated reactor fuel is commonly dissolved in ~7–10 M HNO_3 to give a final solution containing 250 to 300 g of heavy metals per liter in ~3.5 M acid. Small quantities of residue consisting of plutonium-rich oxide and insoluble fission products remain after this treatment. The thermal history of the fuel is probably of most importance in determining the gross dissolution behavior of the fuel, since the high-temperature operation and thermal cycling may generate a central void (due to densification) and radial cracks (due to differential expansion and contraction). The plutonium content, fabrication history, and burnup of the fuel determine the quantity of plutonium remaining in the residue after treatment with nitric acid.

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FUEL PROCUREMENT:

The FFTF fuels are obtained on the following schedule:

DEA-1 3,000 MWd/Te; Discharged 12/81; Received 3/10/83

DE-1 30,000 MWd/Te; Discharged 11/82; Received 11/83

DE-2 55,000 MWd/Te; Discharged 05/83; Received 11/83

DE-3 80,000 MWd/Te; Discharged 10/83; Shipment to be scheduled.

DE-4 and subsequent irradiation cycles will be available at ~six-month intervals.

Unirradiated and irradiated high-plutonium fuels (ANL-08 pins from the EBR-II reactor) containing up to 40% plutonia are also being obtained to investigate the behavior of breeder follow-on fuels.

FLOWSHEET CONDITIONS:

The current rotary dissolver design for the Breeder Reactor Engineering Test (BRET) Facility introduces the chopped fuel into dissolver solution and transports it countercurrent to the flow of the solution; the hulls are discharged after being rinsed with the incoming fresh acid. Hot-cell tests with irradiated fuel are examining this condition and alternatives where the chopped fuel is initially contacted with fresh acid or intermediate solutions.

RESULTS AND DISCUSSION:

The received FFTF fuel was cut into 1" lengths using a single-pin shear. Dissolution occurred in a glass dissolver with a reflux condenser; the acid volume was adjusted to give ~300 g heavy metal per liter in 3 M nitric acid (after complete fuel dissolution). The dissolution was initiated by pouring hot acid onto fuel preheated in the dissolver, and it was quenched by quickly cooling the dissolver in room-temperature water. The fuel pieces and cladding were then separated from the solution, rinsed with water, dried, weighed, and screened. Subsequent dissolutions of part or all of the same material were performed similarly.

When DEA-1 fuel was dissolved as described, it was found that the fuel was released from the cladding in <30 min under quiescent conditions, and within 8-10 min using physical agitation. The fragments appeared to follow the pattern of cracks always found in the irradiated fuel, and they were freed from the cladding as the cracks were enlarged as fuel dissolved in acid that penetrated the central void and cracks. Typical fragments appear to be particles with one or two dimensions in the 2-3-mm range (pellet radius ~2.4 mm) and one dimension frequently smaller. It was also possible to remove this fuel from the cladding by physical means, such as ultrasonic vibration and shaking in a glass bottle or by simply shaking several pieces of clad fuel in a centrifuge cone for ~15 min. It is not clear that this can always be done, especially if the clad ends are bent inward more than in our samples.

Under the defined chemical conditions, ~50% of the fuel was dissolved in 10 min and ~90% in 30 min. The larger fragments that separated from the cladding decreased in size and became more rounded in shape as dissolution proceeded in the 10-30 min time period. After 30 min, the typical largest particles were rounded and ~1 mm (rarely over 1.5 mm) in the longest dimension. This implies a linear dissolution rate of ~2 mm/h. Dissolution tests with similar but unirradiated whole pellets gave penetration rates of ~0.5 mm/h. The higher value for irradiated fuel could result from the different initial fuel properties, the increased surface area due to roughness, the small cracks and bubbles in grain boundaries, or other factors, but the data agreement is surprisingly good.

As dissolution proceeds, the larger fragments shrink and pass through smaller size ranges. We found 1 to 2% in the 100-300 μm range after 10-30 min. A larger fraction was found in the <100-μm range, suggesting that the smaller particles dissolve more slowly. This is consistent with the existence of a hard-to-dissolve plutonium residue, indicating that separate treatment of the fines with strong nitric acid (instead of dissolver solution) may be advantageous.

Additional data are being obtained under the condition of initial exposure to dissolver solution (as in the BRET design) and using FFTF fuel of progressively higher burnup. Studies of fuels with plutonium concentrations >30% and of insoluble fission product residues are continuing.

SUMMARY:

These tests, using FFTF fuel, show that fuel fragmentation and dislodgement from the cladding occurs rather early in the dissolution. The large surface areas of the fuel fragments will lead to rapid dissolution, certainly more rapid than would be expected if the fuel remained within the cladding and dissolved from the open ends, as is sometimes assumed.