TEST IRRADIATION OF MIXED-OXIDE FUEL PREPARED WITH WEAPONS-DERIVED PLUTONIUM[*†]

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ABSTRACT

This paper describes the equipment arrangement and reports the current status of an ongoing test irradiation of mixed uranium-plutonium oxide (MOX) fuel comprising depleted uranium oxide powder with 5% weapons-derived plutonium oxide. The Oak Ridge National Laboratory manages this test irradiation for the U.S. Department of Energy Fissile Materials Disposition Program. The fuel was prepared at Los Alamos National Laboratory by a master-mix process and is irradiated in the Advanced Test Reactor at Idaho National Engineering and Environmental Laboratory. The pellet size is typical of commercial PWR (pressurized water reactor) fuel.

Reactor irradiation of MOX fuel is proposed for disposal of surplus weapons-usable plutonium (Reference 1). A large body of experience generated through research, development, and deployment programs since the mid-1950s supports MOX fuel utilization. Most of this experience, however, has been gained with reactor-grade plutonium in Europe. This test demonstrates that the unique characteristics of weapons-derived plutonium (such as the presence of small amounts of gallium) do not compromise the applicability of this MOX experience base.

There are 15 MOX pellets in each six-inch fuel pin pellet stack. A stainless steel capsule surrounding each Zircaloy fuel pin provides outer containment. This MOX irradiation is carried out under conditions more severe than will be experienced by similar fuel in the actual disposal Mission.

Irradiation began in February 1998 for a total of eleven capsules. In general, these are withdrawn in sets of two for postirradiation examination (PIE). To date, eight capsules have been examined, at burnups of 9-, 21-, 30-, and 40-GWd/MT. The PIE includes visual and dimensional inspection, gamma scanning, metallography, fuel and cladding gallium analyses, fission gas release determination, and burnup estimation.

The remaining three capsules will achieve burnups near 50 GWd/MT in the fall of 2003, when they will be withdrawn for PIE. The final test summary will include results for residual clad ductility as determined by mechanical testing that affords direct measurement of the irradiated cladding hoop strain. All test results are expected to be available by December 2004.

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INTRODUCTION

The United States Department of Energy Fissile Materials Disposition Program (FMDP) is pursuing reactor irradiation for the disposal of surplus weapons-usable plutonium as the fissile constituent of MOX fuel. MOX fuel utilization is supported by a large body of fuel irradiation experience generated primarily in Europe through research, development, and deployment programs during the last four decades. Most of this experience has been gained with reactor-grade plutonium, as derived from spent low-enriched uranium fuel. Accordingly, a test irradiation of fuel prepared with weapons-derived plutonium supports the disposition mission by demonstrating that the unique properties of this constituent do not compromise the applicability of the existing MOX experience base.

This paper reports the current status of an on-going test irradiation of mixed-oxide fuel prepared with weapons-derived plutonium. Oak Ridge National Laboratory manages this project for the Department of Energy. The test fuel was prepared by a master-mix process at Los Alamos National Laboratory. Idaho National Engineering and Environmental Laboratory supplies the irradiation services at the Advanced Test Reactor (ATR).

FUEL

The MOX fuel for this test irradiation was fabricated during 1997 at Los Alamos National Laboratory. This mixed-oxide comprises five percent PuO_2 and 95% depleted UO_2, the latter converted by the ammonium diuranate (ADU) process. All of the PuO_2 was introduced as 31% of the master mix. Stated another way to give a better feel for the numbers, included within each 100 grams of the test MOX fuel are five grams of PuO_2 and 11.11 grams of UO_2 that were milled together to form the master mix. This milling reduced the PuO_2 and UO_2 particle size while promoting a homogeneous dispersion of the PuO_2 particles in the UO_2. Employing a mixer/blender that does not alter particle characteristics (as does milling), these 16.11 grams of master mix were then diluted and dispersed into the remaining 83.89 grams of depleted ADU powder.

It is important to note that only a portion of the UO_2 (that in the master mix) was milled. The physical characteristics (especially flowability) of the UO_2 powder introduced to surround the PuO_2-rich agglomerates are not altered by milling. Thus, a free-flowing press feed without precompaction or granulation was achieved for automated die fill.

The secondary blending (dilution) process that distributed the plutonium-rich agglomerates into the matrix of depleted UO_2 is incomplete, in that residual particles (agglomerates) of the master mix remain intact with equivalent diameters ranging from very small to 200 microns or more within the final blend. Some degree of incompleteness is desirable, in order to maintain the flowability of the UO_2 by limiting the energy input to the powders. The plutonium concentration within the agglomerates is higher than the fuel average, but does not exceed that of the master blend, which is 31% plutonium oxide.

Weapons-derived plutonium includes the alloying agent gallium at approximately one weight-percent. Five of the 11 capsules being irradiated contain pellets prepared from PuO_2 powder that had been thermally treated for gallium removal (Reference 2). Whether the small quantity of gallium carried into the finished MOX fuel will adversely affect irradiation performance is among the technical issues addressed in this test.

TEST CAPSULE CONFIGURATION

The test fuel is irradiated in one of the small I-holes within the ATR reflector. Figure 1 shows one of the simple uninstrumented drop-in capsule assemblies that contain the fuel pellets. Outer containment is provided by the stainless steel capsule that surrounds each Zircaloy fuel pin. There are 15 MOX pellets in each six-inch fuel pin pellet stack. The pellets are held in place by spring loading, the spring being located in an upper gas collection plenum.

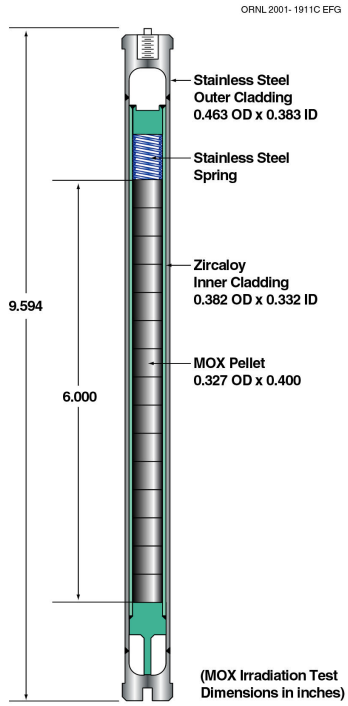

Fig. 1. Each capsule assembly contains one Zircaloy fuel pin with 15 fuel pellets.

The capsule assembly cross-section is shown in Figure 2. The pellet size and Zircaloy-4 cladding thickness are typical of commercial PWR (pressurized water reactor) fuel. The initial gaps between pellet and clad and between clad and capsule are small. The absence of water at the outer surface of the cladding precludes hydriding. This MOX fuel test irradiation is carried out under conditions more severe than will be experienced by similar fuel in the actual disposal Mission. The thermal/hydraulic behavior of these capsule components during irradiation is described in a second paper (Reference 3) of this session.

IRRADIATION EFFECTS IN THE MOX TEST FUEL

Irradiation of the test fuel produces a heterogeneous fission distribution on a microscopic scale, with most fissions occurring within the agglomerates. The local burnup within the agglomerates is much higher than the average for the fuel, and the internal accumulation of fission products (solid and gas) causes the agglomerates to swell against the constraining matrix of UO_2. As burnup increases, the plutonium concentration within the agglomerates decreases because of depletion by fission and a limited diffusion into the surrounding matrix. Simultaneously, the plutonium concentration increases within the UO_2 matrix due both to creation by neutron capture in the U^{238} nuclei and to a minor extent, diffusion from the agglomerates.

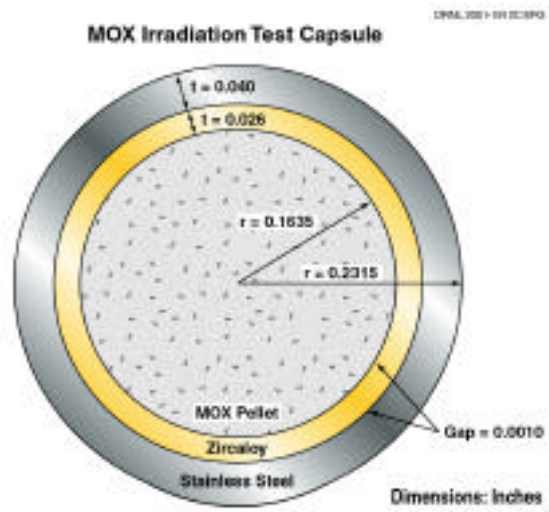

Fig. 2. Test assembly containment is provided by a stainless steel capsule surrounding each sealed Zircaloy fuel pin assembly.

The fuel-clad interface operates at conditions typical of commercial fuel. Of primary interest are any fuel-clad interaction effects. Residual clad strength will be determined by post-irradiation ductility testing. The stainless steel capsule protects the outer surface of the cladding, so there is no embrittlement by clad hydriding (as normally induced in commercial reactors) to mask any detrimental effects that might be caused by the special nature of this fuel.

Irradiation began in February 1998, for a total of eleven capsules. For record purposes, the ATR irradiation cycles are grouped into “phases,” each phase leading to a capsule withdrawal for post-irradiation examination (PIE). In general, the capsules are withdrawn in sets of two. To date, PIE has been completed for the six capsules withdrawn at 9-, 21-, and 30-GWd/MT burnup, and is in progress for the capsules withdrawn at 40 GWd/MT. Fuel performance has been excellent. A companion paper

(Reference 4) describes the results of these completed PIEs.

The maximum burnup to be achieved in this test was originally set at 30 GWd/MT. It was subsequently decided that the Mission fuel should exceed 40 GWd/MT, and safety analyses confirmed that the current test capsules can be extended beyond this.

The irradiation history for the most recent (March 2002) capsule withdrawals is shown in Figure 3.

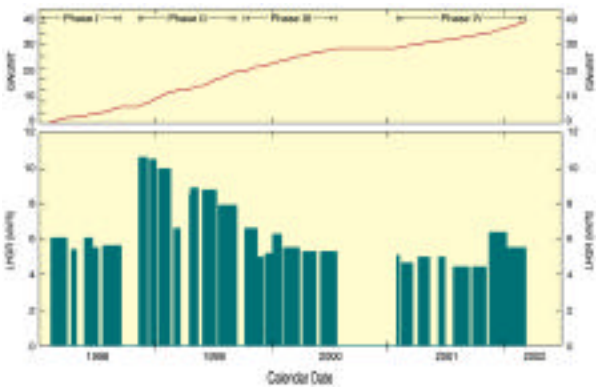

Fig. 3. Irradiation history for the MOX test fuel pins withdrawn at 40 GWd/MT burnup and currently undergoing PIE.

The highest linear heat generation rate (LHGRs) for these fuel pins was about 10.7 kW/ft, reached at the beginning of Phase II of the irradiation as shown in Figure 3. The exposure spanned 30 ATR irradiation cycles, accumulating 904 effective full power days (EFPDs) and 42 thermal cycles. The fissions per initial metal atom value is 4.15%, with integrated fast fluxes (E>1.0 MeV) of 1.5E21 (fuel), 1.1 E21 (Zircaloy cladding), and 1.0E21 (steel capsule). The test assembly was moved to a higher flux location when burnup reached about 35 GWd/MT; hence, the increase in LHGRs during the last two irradiation cycles.

Ongoing PIE involves the two recent capsule withdrawals at 40 GWd/MT; future PIE will address the three capsules to be withdrawn at 50 GWd/MT in October 2003. Irradiated clad specimens from each withdrawal will be subjected to ductility testing to begin in summer 2003.

FISSION GAS RELEASE VS MOX EXPERIENCE IN EUROPE

Figure 4, which is adapted from Reference 5, illustrates literature values for fission gas release of European commercial test fuels plotted against the corresponding average linear heat generation rates (LHGRs) during the second irradiation cycle.

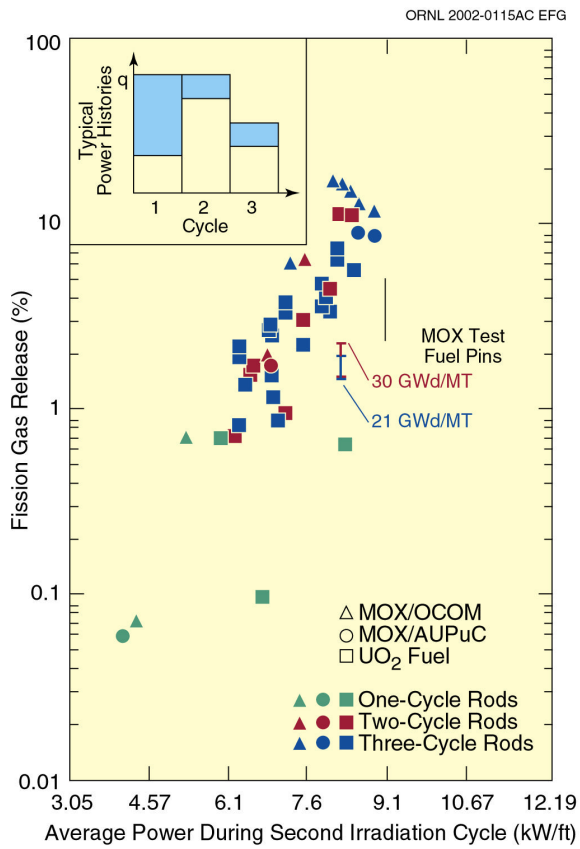

Fig. 4. The MOX test fuel pins exhibit gas release fractions proportional to their linear heat generation rate experience. (Basic plot is taken from Reference 5.)

This Figure also presents, in the upper left-hand corner, a bar chart representing the relative ranges for the axial powers (LHGRs) typically experienced during each of the three commercial reactor irradiation cycles. (In contrast to the much shorter ATR cycle length, a commercial reactor cycle normally comprises 250–300 EFPDs.) The blue portions of the bar chart illustrate the extents of the LHGR variations for the first, second, and third irradiation cycles. In

general, the LHGRs increase slightly in proceeding from the first to the second cycle, and in all cases, decrease from the second to the third cycle.

Because the highest powers are experienced during the second irradiation cycle, the average LHGR during that cycle is chosen as the abscissa parameter for the fission gas release plot. (The exception is when the fuel was irradiated just one cycle—in these cases, the fission gas release is plotted against the average LHGR for that single cycle.)

Since fuel temperatures are proportional to LHGRs, the points plotted on Figure 4 can be considered to indicate the linear relation (on a logarithmic scale) between the accumulated gas release at the end of the irradiation and the highest temperature experienced by the fuel during the irradiation. This demonstrates that it is the highest temperature experienced by the fuel (normally occurring during the second irradiation cycle) that determines the fission gas release fraction, not the extent of the accumulated burnup. (The amount of gas release does, of course, increase directly in proportion to burnup.)

Superimposed on the Fig. 4 plot are the fission gas release ranges obtained for both the 21 and 30 GWd/MT withdrawals of the MOX irradiation test. These four “MOX test fuel pins” were symmetrically located with respect to the ATR core and hence had similar irradiation histories. It is clear that the fission gas releases obtained for the current test are low in comparison to the literature values for the same LHGR history. It is also evident that there has been little change in the percent fission gas release as the burnup of these test rods has increased from 21 to 30 GWd/MT.

GALLIUM ANALYSES

The gallium initially present within the unirradiated fuel (1–5 ppm by individual pellet analyses) is sufficient to produce levels as high as 9 ppm within the cladding if all of this gallium were transferred. The transport of gallium from fuel to cladding can be detected by metallographic and chemical analyses. As part of each PIE, a portion of the irradiated cladding is sent to the Radioactive Materials Analytical Laboratory at ORNL for determination of the gallium content. None of the cladding samples examined to date indicates any increase over the approximately one-half ppm initially (preirradiation) present.

CLAD TESTS (PLANNED)

Determination of residual (post-irradiation) clad ductility is important to the material behavior investigation for the test MOX fuel. The stainless steel capsule protects the cladding from embrittlement by hydriding, but the accumulated fast flux reduces the ductility of the Zircaloy-4 at high burnup levels. It is desirable to determine if there has been any additional reduction in cladding ductility for this MOX test fuel. Accordingly, a simple procedure has been developed to determine the residual ductility by mechanical loading in a manner that provides direct measurement of the irradiated cladding hoop strain.

The loading concept for the irradiated cladding is illustrated in Figure 5. An expanding polyurethane plug is compressed axially, causing radial swelling into the surrounding Zircaloy ring specimen. This simple approach expands the cladding in the same manner as would fuel pellet swelling under reactor accident conditions.

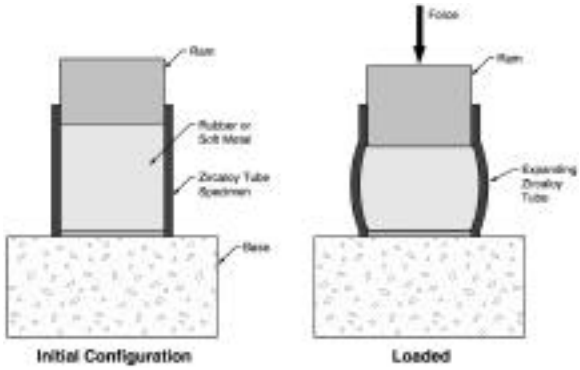

Fig. 5. Loading concept for the ductility testing of the MOX irradiation test cladding specimens.

It should be noted that no special machining of the irradiated test specimens (cladding rings) is required. Furthermore, the hoop strain is

directly proportional to, and can be simply derived from, the change in ring diameter.

Figure 6 shows the assembled test apparatus. Proximity probes are utilized to continuously monitor the hoop strain as it is imposed in the irradiated clad specimen.

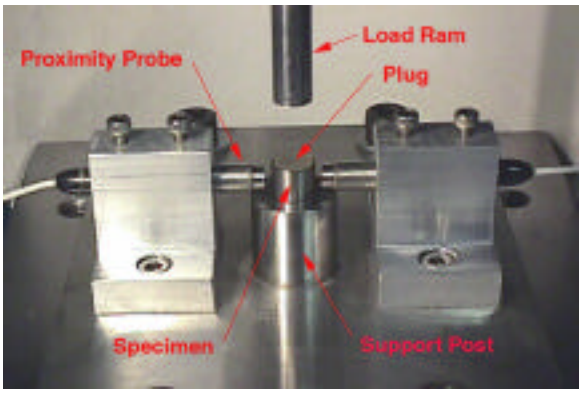

Fig. 6. Test apparatus for determination of residual strain-to-failure for the MOX irradiation test fuel pins.

Trials of this cladding test apparatus have been successfully conducted with both unirradiated Zircaloy-4 and high-strength tool steel ring specimens, the ductility of the latter being more representative of the high-burnup cladding specimens to be loaded in the actual tests.

This test method was introduced in the recent clad ductility round-robin test program sponsored by Argonne National Laboratory for the Mechanical Properties Expert Group on Clad Behavior. Participants from France, Japan, Russia, and the U. S. (Argonne and Oak Ridge National Laboratories) were provided a short length from a single unirradiated Zr-4 tube. Each determined the tensile properties using the specific specimen design/test methodology developed by that participant, and the results were compared. The ORNL measurements agreed very well with those of most other participants, providing further qualification of the expandable plug method.

Although this method can be used to develop a full stress-strain curve for the ring material, it is only necessary for the purposes of the MOX fuel irradiation to demonstrate a residual strain-to-failure in the range of 3 to 5 percent, as normal for irradiated Zircaloy in the absence of hydrides. First tests with irradiated cladding specimens are scheduled to begin in 2003.

SUMMARY

Fuel performance as determined to date for MOX fuel prepared in the United States with weapons-derived plutonium has been excellent. This test fuel has been irradiated under conditions more severe than will be encountered during the fissile material disposition mission. In addition to higher linear heat generation rates and elevated fuel temperatures, this includes the imposition of additional thermal cycling in the presence of very small initial pellet-clad gaps. The plutonium-rich agglomerates are larger, and contain a larger fraction of plutonium. The overall plutonium loading is greater. As a condition of the test, the current fuel contains much more gallium.

Eight irradiated capsules have been withdrawn to date, with burnups ranging from 9 to 40 GWd/MT. Fission gas release is relatively low although higher LHGRs were applied in the presence of agglomerates larger than found in commercial European MOX fuel. No cladding deterioration has been noted. Successful completion of this MOX test irradiation will demonstrate that a gallium concentration of 1–5 ppm in fuel is acceptable; mission levels will be much lower than this.

The final three capsules will reach 50 GWd/MT in the fall of 2003. This will exceed the planned mission fuel peak burnup of 44 GWd/MT. Subsequent PIE results, including the values for cladding strain-to-failure, are expected to be available in December 2004.

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