

Summary of Irradiation Tests of Mixed Oxide Fuel Prepared with Weapons-Derived Plutonium^{1,2}

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Abstract—Mixed oxide (MOX) test capsules prepared with weapons-derived plutonium have been irradiated to a burnup of 50 GWd/MT. The MOX fuel was fabricated at Los Alamos National Laboratory by a master-mix process and irradiated in the Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL). Previous withdrawals of the same fuel have occurred at 9, 21, 30, and 40 GWd/MT. Oak Ridge National Laboratory managed this test series for the Department of Energy's Fissile Materials Disposition Program (FMDP). This paper describes the preparation of the MOX fuel, the equipment design, and the irradiation history of the test capsules and discusses the significance of the more important observations of the post-irradiation examinations (PIEs). Fuel performance has been excellent and consistent with code predictions and with existing U.S. and European experience.

Keywords: Fissile Materials Disposition Program, weapons-derived plutonium, mixed oxide fuel, test irradiation, post-irradiation examinations, fuel-performance codes

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²Research sponsored by Office of Fissile Materials Disposition, U.S. Department of Energy, National Nuclear Security Agency, under Contract DE-AC05-00OR22725 with UT-Battelle, LLC.

I. INTRODUCTION

The United States Department of Energy Fissile Materials Disposition Program (FMDP) is pursuing disposal of surplus weapons-usable plutonium by reactor irradiation as the fissile constituent of MOX fuel¹. A large body of mixed oxide (MOX) fuel irradiation experience exists through the genesis of research, development, and deployment programs primarily in Europe over the last four decades. Most of this experience has been gained with reactor-grade plutonium, as derived from spent low-enriched-uranium fuel. Since 1998, a test irradiation² of MOX fuel prepared with weapons-derived plutonium has been conducted at the Idaho National Laboratory (INL) in the Advanced Test Reactor (ATR).

Weapons-derived MOX fuel differs from the commercial fuel utilized in Europe in that its initial fissile inventory comprises a higher proportion of ²³⁹Pu, with smaller contingents of the higher plutonium isotopes, and because the plutonium may be accompanied by small amounts of gallium as an impurity. The present test irradiation supports the disposition mission by demonstrating that introduction of weapons-derived plutonium does not compromise the applicability of the existing MOX database.

Oak Ridge National Laboratory manages this irradiation demonstration project for the Department of Energy.

II. TEST CAPSULE AND FUEL PIN DESIGN

Figure 1 illustrates the fuel pin and capsule cross section. The pellet size and Zircaloy-4 cladding thickness are typical of commercial pressurized-water reactor (PWR) fuel. The initial gap between pellet and cladding is narrower, however—about 50 microns as opposed to 150–200 microns diametral in typical PWR fuel. The gap between cladding and capsule is also small. The absence of water at the outer surface of the cladding precludes hydriding so that loss of cladding ductility is limited to that caused by irradiation hardening or any effects of fuel impurities such as gallium.

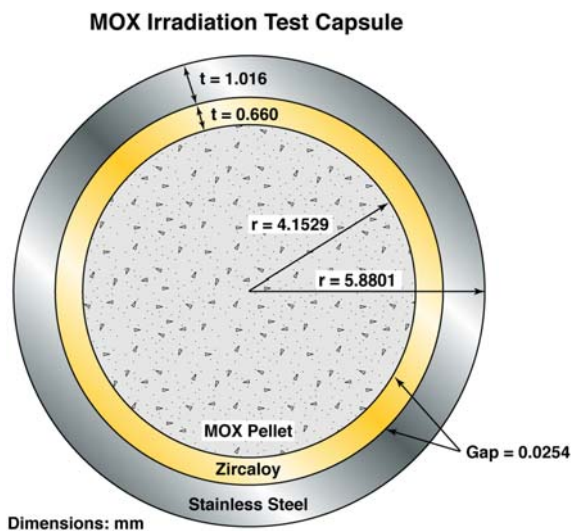


Fig. 1. A stainless steel capsule contains each fuel pin.

Figure 2 is an elevation view of one of the simple uninstrumented drop-in capsule assemblies. There are 15 MOX pellets in each 152-mm fuel pin pellet stack. The fill gas for the fuel pins and capsules is helium at atmospheric pressure. A gas collection plenum is provided at the top of each fuel pin (surrounding the stainless steel spring).

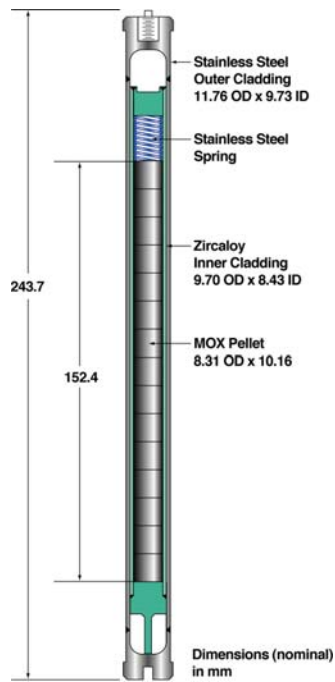


Fig. 2. Each capsule assembly contains one Zircaloy fuel pin with 15 fuel pellets.

III. TEST ASSEMBLY

The test capsules were secured within a test assembly and irradiated initially in the small Northwest I-hole of the ATR reflector. The test assembly provided nine capsule positions, as shown in Figure 3. As the fueled capsules were withdrawn for post-irradiation examination (PIE), solid stainless steel capsule simulators filled any test assembly positions not occupied by MOX test capsules.

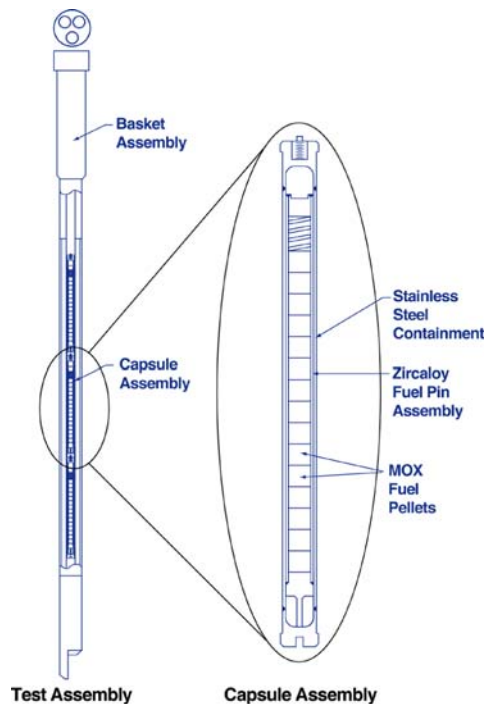


Fig. 3. The test assembly permitted simultaneous irradiation of up to nine MOX test capsules.

As shown in the basket assembly cross section at the top of Figure 3, the capsule columns are arranged with two in front (in the direction of the ATR core) and one behind. The test assembly is aligned vertically such that the midplanes of the middle capsules correspond to the midplane of the ATR core. Thus, the highest thermal fluxes (and axial powers) are found for the capsules at the two front middle positions with descending fluxes for the capsules placed at the front top and bottom, back middle, and back top and bottom positions. As an example, for the initial heatup with all nine positions occupied by fresh fuel, the linear heat generation rates (LHGRs) ranged from 30.2 kW/m at the front middle to 20.3 kW/m in the back top and bottom positions.

The thermal and fast flux profiles along the three capsule positions in each column were determined at intervals of approximately 100 effective full power days (EFPDs) throughout the irradiation by means of flux wires. It was desirable during the course of the irradiation to maintain high LHGRs by increasing the thermal flux at the fuel to counter the effects of plutonium depletion. This was accomplished first by shifting from an Inconel-shielded basket assembly to a basket assembly with an aluminum shield after the first 155 EFPDs. Later, at about 800 EFPDs, the test assembly was shifted to the higher-flux Southwest I-hole position within the ATR reflector.

Because the upper and lower ends of the pellet stack are unshielded, the end pellets receive more thermal flux and hence there is end peaking in the fuel stack

IV. PREPARATION OF THE MOX TEST FUEL

The MOX fuel for this test irradiation was fabricated during 1997 at the Los Alamos National Laboratory via a master mix process. This mixed oxide is comprised of 5% PuO₂ and 95% depleted UO₂, the latter converted by the ammonium diuranate (ADU) process. All of the PuO₂ was introduced as 31% of the master mix.

The secondary blending (dilution) process by which the master mix was distributed into the depleted UO₂ matrix was incomplete, in the sense that residual particles (agglomerates) of the master mix remain intact within the final blend. The equivalent diameters for the test fuel agglomerates range from very small to quite large, with about 1.5% of the fuel cross-sectional area occupied by particles with equivalent diameters of 400 microns or more.

Weapons-derived plutonium includes the alloying agent gallium at approximately 1 wt %. Of the 11 capsules irradiated, five were loaded with pellets prepared from PuO₂ powder that had been thermally treated for gallium removal³. Whether or not the small quantity of gallium carried into the finished MOX fuel has the potential to adversely affect fuel and cladding performance was among the technical issues addressed by this test.

V. IRRADIATION HISTORIES

Irradiation began in February 1998 and continued through to April 2004 for a total of 11 MOX test capsules. The predicted operating envelope (LHGRs, fuel temperatures) for the MOX test irradiation is prototypic of commercial light water reactors (LWRs) with fuel of similar dimensions.

The ATR irradiation cycles have been grouped into "phases," as indicated in Table 1. In general, each irradiation phase defines a different arrangement of capsules (and capsule simulators) within the test assembly. Table 1 also provides the MOX capsule withdrawal schedule and the associated burnups

The average as-run LHGRs are listed in Table 2 for the test MOX capsules withdrawn at burnups of 9-, 21-, 30-, 40-, and 50-GWd/MT.

TABLE 1
The Paired MOX Test Capsules (Normal and TIGR^a-Treated) Have
Been Withdrawn Sequentially

Irradiation Phase	Date Completed	Effective Full-Power Days	Capsules Withdrawn	Burnup (GWd/MT)
I	September 13, 1998	154.9	1 and 8	8.8
II	September 12, 1999	227.7	2 and 9	21.0
III (Part 1)	July 23, 2000	232.4	3 and 10	30.2
III (Part 2 ^b)	January 14, 2001	113.1	—	—
IV (Part 1)	March 9, 2002	289.1	4 and 13	39.8
IV (Parts 2 and 3)	April 18, 2004	443.6	5, 6, and 12	50.0

^aTIGR = thermally induced gallium removal³.

^bPhase III (Part 2) provided catch-up irradiation for Capsules 5, 6, and 12 only.

TABLE 2
Average As-Run LHGRs (kW/m) for 9-, 21-, 30-, 40-, and 50-GWd/MT Withdrawn Capsules

Irradiation Phase	EFPDs	Burnup Capsule	9 GWd/MT		21 GWd/MT		30 GWd/MT		40 GWd/MT		50 GWd/MT		
			1	8	2	9	3	10	4	13	5	6	12
I	154.9		27.0	27.1	26.0	26.5	25.8	26.5	19.2	19.4	20.0	—	—
II	227.7				26.9	27.1	26.5	27.2	29.5	29.9	23.1	25.0	25.3
III—Part 1	232.4						17.7	18.3	18.6	18.8	17.9	19.0	19.2
III—Part 2	113.1								—	—	13.3	20.8	21.2
IV—Part 1	289.1								17.0	17.1	13.6	17.8	17.9
IV—Part 2	110.2										16.6	19.0	19.3
IV—Part 3	333.4										12.6	13.7	14.0
		FGR (%)			1.32	1.88	1.47	2.30	8.37	9.51	3.37	7.22	8.61

VI. PIE SCHEDULE

As indicated in Table 1, ten capsules have been withdrawn for PIE in sequential sets of symmetrically loaded pairs (TIGR—non-TIGR) at burnups of 9-, 21-, 30-, 40-, and 50-GWd/MT. The 11th capsule (non-TIGR Capsule 5) was also withdrawn at 50 GWd/MT.

The PIEs⁴ included visual and dimensional inspection, gamma scanning, metallography, fuel and cladding gallium analyses, fission gas release determination, and burnup analyses. Of particular interest were any fuel-cladding interaction effects. Residual cladding strength is determined by post-irradiation 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 introduced by the special nature of this fuel.

To date, PIE has been completed for the 11 capsules withdrawn with burnups of 9-, 21-, 30-, 40-, and 50-GWd/MT. Fuel performance including cladding integrity has been excellent.

VII. OVERVIEW OF SIGNIFICANT PIE OBSERVATIONS

This mixed oxide test irradiation was carried out under conditions more severe than will be encountered by the mission fuel in the U.S. commercial reactors participating in the FMDP. Individual capsule LHGRs and fuel temperatures depend upon the location of the capsule within the test assembly during irradiation but in general were higher than those expected for mission fuel. All of the PIEs have been conducted at the Irradiated Fuels Examination and Radioactive Materials Analysis Laboratories at Oak Ridge National Laboratory. The following discussions provide an overview of the more significant PIE observations.

Agglomerates and Fission Gas Release

For the MOX fuel prepared for this test irradiation, all of the PuO₂ was introduced as 31% of the master-mix. After dilution into the remainder of the UO₂, the equivalent diameters of the residual master-mix particles (agglomerates) in the final blend ranged from very small to 400 microns or more. The presence of a few large agglomerates confirms that the secondary blending (dilution) process was incomplete in this test fuel but is beneficial from the standpoint of insights as to the effects of agglomerate size to be gleaned from the post-irradiation analyses.

Sections of fuel and surrounding cladding have been examined by both scanning electron microscopy (SEM) and electron probe microanalysis (EPMA). Areas of particular interest include the nature of the large agglomerates and their immediate surroundings.

High burnup within the plutonium-rich agglomerates is accompanied by considerable local swelling induced by the accumulated solid and gaseous fission products. Whereas the solid fission products stay with an agglomerate throughout fuel life, the fate of the fission product gases depends upon the temperature during irradiation of the region in which the agglomerate is located. (Even the largest of the agglomerates are still sufficiently small that their internal temperatures only slightly exceed that of the immediately surrounding UO₂ matrix.)

Agglomerates become highly visible when they have transformed into a "high-burnup structure." In general, a high-burnup structure (small grains with a few large pores) evolves during irradiation when the local temperature is less than 1000°C and the local burnup exceeds about 60 GWd/MT. Prior to transformation, much of the fission gas is stored in nanometer-size cavities within the approximately 10-micron fuel grains. Subsequent to transformation, the grains are in the 0.5 to 1.0 micron range, in a structure interspersed with relatively large gas storage pores. Much of the gas displaced from the very small intergranular cavities is collected (at high pressure) in the faceted pores in the recrystallized microstructure.

Agglomerates in the outer region of the MOX test fuel mounts are clearly visible due to their high-burnup structure. The halos around these agglomerates are fission gas in the surrounding UO₂ grains, which either diffused (primarily athermally) prior to transformation or was displaced when the high-burnup structure was formed. Faint agglomerate outlines can be discerned in the central regions of these fuel mounts at a fuel-average burnup of

40 GWd/MT. These are the beginnings of the transformation to high-burnup structure—transformation was delayed for the central region agglomerates because local temperatures remained greater than 1000°C during the early phases of the irradiation.

No evidence of recrystallization (“rim effect”) has been found in the fuel matrix around the pellet circumference. Although the rim area experienced low temperature and local burnups higher than the average for the depleted UO₂ matrix, rim area burnup did not reach 60 GWd/MT.

Gallium and Fuel Swelling

Metallographic and chemical analyses of individual unirradiated test fuel components showed gallium concentrations in the range from 1–5 ppm in the fuel (more than 20 times greater than that expected in the mission fuel) and from 0.3 to 0.5 ppm in the cladding. If all of the gallium in the fuel were transferred radially outward, the cladding concentration would increase to about 9 ppm. As part of each PIE, samples of irradiated fuel and cladding were sent to the Radioactive Materials Analytical Laboratory at ORNL for determination of the gallium content. None of the cladding samples has shown any increase over the preirradiation gallium concentration. Correspondingly, there has been no evidence of any attack of the inner cladding surface by gallium. Finally, each fuel sample has indicated, within the limits of analytical accuracy, that the gallium initially present has been retained. Post-irradiation fuel and cladding concentration measurements indicate that any movement of this gallium has been insignificant.

Comparison of the cladding and pellet dimensions as determined for successively higher burnups establishes the history of pellet swelling and cladding creep as experienced during this test irradiation. For each of the PIEs completed to date, the capsule and fuel pin metrological results were combined with measurements made directly (imaging software) from photographic enlargements of the metallographic mounts to determine the cladding thickness and internal diameter, the pellet outer diameter, and the effective gap between pellet and cladding. Fuel behavior (cracking, densification, and swelling) is found to be normal and prototypic of commercial MOX fuel.

The fuel densification and swelling models within the Frapcon-3⁵ code accurately reproduce the densification/swelling history for this fuel.

Cladding Outward Creep and Primary Ridging

The fuel pin cladding experienced an irradiation-assisted outward creep under the impetus of a tensile wall (hoop) stress that increased from 0 to about 5 MPa as fuel pin internal pressure increased during the irradiation. (This cladding movement differs from normal PWR behavior, where cladding creep down is imposed by high external coolant pressure.) The outward cladding creep (about 0.2%) observed at 40-GWd/MT burnup is normal for the test fuel operating conditions and accumulated fast fluence of 1.4×10^{21} n/cm² ($E > 1.0$ MeV) and is compatible with the experience documented in the literature.

The Fuel Pin Measuring Apparatus developed at ORNL for this project provides a precise determination of the fuel pin outer diameter profile. At all burnup levels, the cladding profiles exhibit local ridging (average radial height about 3.6 micron) over the pellet-to-pellet interfaces. This type of local cladding deformation (denoted “primary ridging”) is commonly observed in commercially irradiated PWR fuel. The cause is differential thermal expansion within the fuel—the pellet centerline is much hotter than the outer cylindrical surface and expands axially to a greater extent. The pellets crack into pie-shaped segments, and the differential expansion in the axial direction causes these segments to warp into hourglass shapes.

The formation of ridges in the MOX test fuel pins is somewhat different from that in commercial fuel. The high coolant pressure in commercial PWRs causes inward creep of the cladding, which eventually comes into hard contact with the fuel over the pellet interfaces, where the hourglassing produces the largest (deformed) pellet diameters. The cladding primary ridges are therefore artifacts of the hourglass (or saddle) shape of the underlying pellets.

For the MOX test fuel pins, the pressure differential is outward across the cladding, which creeps outward, not inward. Localized contact between pellet and cladding still occurs, however, because the fuel pins were designed to have initial pellet-clad radial gaps (25 microns) much smaller than those found in commercial fuel (75–100 microns). Here the pellet differential thermal expansion is sufficient to cause hourglass-enhanced local contact with the cladding at initial heatup. This hard contact over the pellet-to-pellet interfaces occurs before any fuel densification or swelling.

ABAQUS⁶ code finite-element calculations performed for the zero-burnup initial heatup with as-built dimensions for the pellet, fuel pin, and capsule, and with the actual initial LHGRs clearly predict pellet hourglassing with cladding contact at the pellet ends. The applied stress is sufficient to induce local yielding.

To recap, the observed ridging is predicted to have occurred on initial heatup at zero burnup. Primary ridging is expected for modern PWR fuel and does not constitute a mechanism for failure during normal operation. There is no indication for the current test fuel that such localized contact has had any detrimental effect on cladding integrity.

Cladding Inner Surface Oxidation

The nature of the corrosion layers intermittently located along the pellet-clad interface is of interest to the examination of cladding performance. These layers exist along the portions of the cladding inner surface where the fuel was in contact with the cladding during irradiation.

The uneven and noncontiguous nature of the corrosion observed on the cladding inner surfaces is an artifact of the manner in which the pellet fragments came into contact with the cladding during irradiation. Inner surface oxidation requires that excess oxygen be available from the fission process and that the fuel be in contact with the cladding to provide a path for solid-state athermal diffusion of the oxygen atoms. The thicker oxidation layers in regions where an agglomerate is located at or near the fuel surface follow directly from the narrower local pellet-clad gaps during irradiation when these agglomerates are swollen. The observed corrosion patterns are in accordance with expectations based on U.S. and European experience with both UO₂ and MOX fuels.

Fission Gas Release versus MOX Experience in Europe

Figure 4, which is adapted from Reference 7, illustrates literature values for fission gas release of European commercial test fuels plotted against the corresponding average LHGRs during the second irradiation cycle.

In general, in the first three irradiation cycles in commercial reactors, 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 in Figure 4. The exception is for cases where the fuel was irradiated just one cycle—here, the fission gas release is plotted against the average LHGR for that single cycle.

Since fuel temperatures are determined by the 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 the fission gas release fraction is determined by the temperature history rather than by the burnup extent. (The amount of gas release does, of course, increase in proportion to burnup.)

Superimposed on the plot of Figure 4 are the fission gas release fractions as obtained by krypton-85 activity measurements for the 21-, 30-, 40-, and 50-GWd/MT burnup fuel pins of the MOX irradiation test. The abscissa values for these release fractions are the average LHGRs during Phase II of the MOX test irradiation.

It is clear from Figure 4 that the fission gas release fractions obtained for the MOX test irradiation are consistent with the literature values (European experience) for both MOX and UO₂ fuels with the same LHGR history.

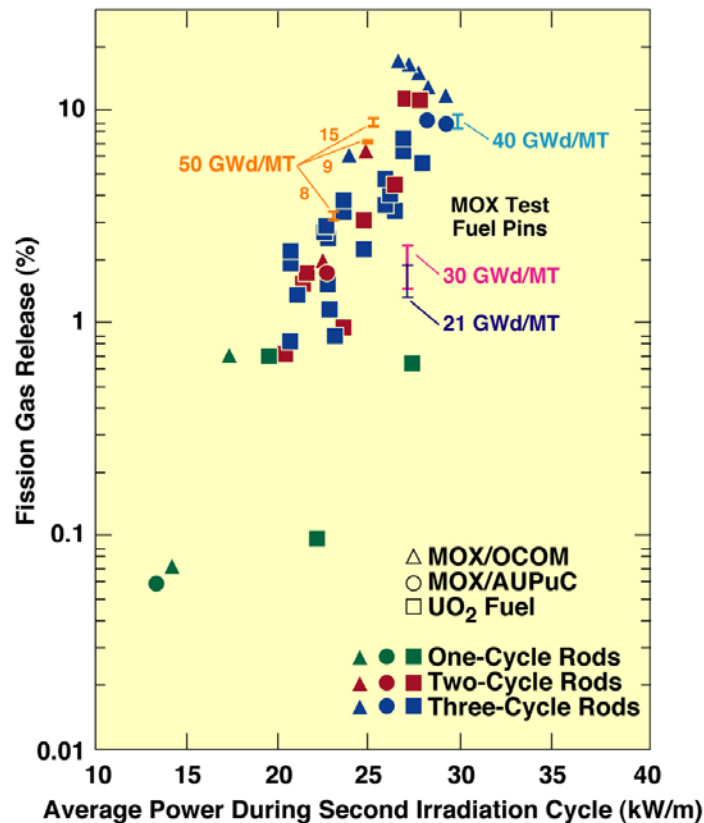


Fig. 4. The MOX test-fuel-pin gas release fractions can be correlated with their LHGR experience. Basic plot is taken from Reference 7.

Synopsis of MOX Test Fuel Behavior

The test fuel has been found to perform well, and in accordance with expectations based on the documented European experience. The presence of a few very large agglomerates has not adversely affected fission gas release. No abnormal behavior has been observed. [With respect to the MOX test fuel, “abnormal behavior” is defined as any deviation from expectations based on the documented MOX fuel irradiation experience in Europe that cannot be explained solely by differences in fuel preparation or test conditions. As an example, the cladding creeps outward in this test as opposed to inward in the commercial MOX experience, but this difference is readily explained by the absence of external coolant pressure on the MOX test fuel pins.]

In addition to demonstrating the applicability of the European database, these PIEs of similar MOX fuels at five burnup intervals offer unique opportunities to study the effects of burnup and irradiation history on MOX fuel performance characteristics.

VIII. SUMMARY

The test MOX fuel prepared with weapons-derived plutonium exhibits normal fuel swelling, densification, and fission gas release. The cladding and fuel behavior has been as expected from the literature and is reasonably predicted by available fuel performance computer codes (such as FRAPCON-3).

The MOX irradiation experimental operating envelope (LHGRs and fuel temperatures) is prototypic of commercial light water reactors with fuel of similar dimensions; however, the test fuel has been irradiated under conditions more severe than what will be encountered during the fissile material disposition mission. Fuel performance for these test irradiations has been excellent.

A weapons-derived MOX fuel benchmark problem (based on the irradiation of Capsules 4 and 5) has been proposed to the OECD/NEA/WPRS/TFRPD (Expert Group on Reactor-Based Plutonium Disposition). Please contact Enrico Sartori (sartori@nea.fr) at the OECD/NEA Data Bank for information on the benchmark.

These test MOX irradiations has supported the FMDP MOX fuel qualification and the insertion of WG-MOX Lead Test Assemblies (LTAs) in the Catawba NPP.

IX. WG-MOX LTAS

The U.S.DOE selected Areva NP in September 2003 to manufacture the LTAs (4 total) for Duke Energy's Catawba NPP. Polished PuO₂ powder (from U.S. weapons components) was produced at the Los Alamos National Laboratory and shipped to France in the fall of 2004. The WG-MOX fuel and fuel rods were produced at Cadarache, France, in late 2004 with fabrication of the fuel assemblies at Melox in early 2005. Subsequently the assemblies were transloaded at La Hague and shipped to the United States in March of 2005. Four assemblies were inserted in Catawba-1 in May 2005.

The first cycle of irradiation in Catawba ended in November 2006; performance of the LTAs was as expected (normal behavior and no leaks). Poolside examination of the LTAs again yielded as-expected results. The LTAs were re-inserted in Catawba-1 for their second cycle in December 2006.

The end of the second cycle of irradiation for the four LTAs will be late spring of 2008. The LTAs will again be inspected poolside, and at that time five (5) fuel rods will be withdrawn from the assemblies to be shipped to ORNL for destructive PIE (starting in the fall of 2008). The four LTAs will then be reconstituted and re-inserted in Catawba for a third irradiation cycle (starting in the summer of 2008).

The third (and last) cycle of irradiation of the LTAs is expected to end in the fall of 2009. The assemblies will again be inspected poolside, and additional fuel rods will be withdrawn for PIE (to be started at ORNL in the spring of 2010).

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