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THERMAL REACTOR MIXED-OXIDE
FUEL PERFORMANCE

OCTOBER 1971

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Conf- 711009--15

THERMAL REACTOR MIXED-OXIDE FUEL PERFORMANCE*

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October 1971

*Paper prepared for presentation at the 1971 Winter Meeting of the American Nuclear Society at Miami Beach, Florida, October 17-21, 1971.

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Thermal Reactor Mixed-Oxide Fuel Performance

by

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The satisfactory irradiation performance of mixed-oxide fuels in thermal reactors has been demonstrated through the success of numerous experimental programs (Table I).^(1,2,3,4) The success of these and other experimental programs is attested to by the universal acceptance of plutonium utilization in thermal reactors. These efforts have been culminated by the initiation of demonstration programs in commercial power reactors (Table I) with the rapid full-scale implementation of plutonium utilization in commercial plants anticipated.^(5,6,7,8,9)

The US-AEC sponsored fuel evaluation program conducted in PRTR was characterized by testing a variety of different mixed-oxide fuel types, some of which may be considered to be advanced concepts, to significant burnups (over 18,000 MWd/MTM) at linear heat ratings (about 20 kW/ft peak) higher than those currently employed in commercial power reactors.^(10,11,12) In addition to the Plutonium Utilization Program at Pacific Northwest Laboratories (PNL), the AEC has supported experimental irradiation testing programs in the Experimental Boiling Water Reactor⁽³⁾ and the Saxton Reactor.⁽⁴⁾ Experiments in the Saxton Reactor are being conducted to evaluate the performance of mixed-oxide fuel at linear power and burnup levels consistent with modern PWR technology.

The different mixed-oxide fuels tested in PRTR included Zircaloy-clad, vibrationally compacted, heterogeneously and homogeneously enriched types as well as pellet fuels produced by hot-pressing and the more conventional cold-press-sinter technique.

TABLE I.

Significant Domestic Mixed-Oxide Irradiations

Reactor	No. of Rods	Fuel Type ^(a)	Peak Burnup, ^(b) MWd/MTM	Peak Rod Power, kW/ft	Peak Heat Flux, BTU/hr-ft ²
PRTR	~1860	Swage	13,500	17	390,000
PRTR	~ 700	Vipac	18,500	16	370,000
PRTR (High Power Density)	~1730	Pellet (cold-press-sinter & hot-press)-Vipac-Swage	13,000	21.5	500,000
EBWR	1296	Vipac	3,000	8	200,000
Saxton-Core II	638	Pellet-Vipac	29,000	16	530,000
Saxton-Core III	~ 250	Pellet	44,000	19	630,000
Dresden-I ^(c)	4	Pellet (hot-press)	13,000	10	230,000
Dresden-I ^(c)	99	Pellet	14,800	14.9	345,000
San Onofre ^(c)	720	Pellet	8,500	13	460,000
Big Rock Point ^(c)	32	Pellet	19,500	14	325,000
Big Rock Point ^(c)	204	Pellet	14,600	16	370,000
Vermont-Yankee ^(c)	48	Pellet	---	17.3	---

(a) Pellets are cold-press-sinter type unless otherwise noted.

(b) Megawatt-days per metric tonne of metal as of September 1971.

(c) Commercial power reactors.

Although small PuO_2 concentrations in mixed-oxide thermal reactor fuel have an insignificant effect on most irradiation performance characteristics, one of the objectives of the mixed-oxide fuel development program at PNL was to investigate important considerations related to performance which are unique with plutonium-enriched fuels.

CLADDING DISTORTION

Cladding distortion during irradiation, which gives rise to fuel/cladding mechanical interaction phenomena, is sensitive to fuel type and design and should not be affected by PuO_2 additions to UO_2 except as the PuO_2 additions may affect fuel swelling behavior.⁽¹³⁾ The effects of fuel/cladding mechanical interaction are more severe in pellet type fuels than in particle fuels.

Postirradiation length measurements and diameter profiles were obtained on vipac fuel rods and rods containing hot-press and cold-press-sinter pellet fuel irradiated in PRTR to compare the dimensional behavior of the three fuel types (Figure 1).⁽¹⁴⁾ The vipac rods contained crushed, pneumatically impacted UO_2 - PuO_2 fuel vibrationally compacted to a smear density of nominally 86% TD. The hot-press and the cold-press-sinter pellet density was approximately 93% TD and the rods were assembled with a nominal 0.011 in. diametral gap.

Observations related to the rod dimensional data are summarized as follows:

- Cladding ovality and length changes, which seem to be related, are more pronounced in the cold-press-sinter pellet rods than in the hot-press pellet and vipac rods.

The fuel in the vipac rods provides cladding support and thereby reduces the amount of ovality. Considerably less ovality occurred in the hot-press pellet rods than in the rods which contained cold-press-sinter pellets although both were assembled with a nominal 0.011 in. diametral gap. It is suggested that the hot-press pellet fuel, because of its high stoichiometry ($\text{O/M} = \sim 2.15$)

FIGURE 1. MIXED-OXIDE FUEL ROD DIAMETER PROFILES

and reduced thermal conductivity, operated at higher temperatures than the cold-press-sinter fuel. The increased expansion during irradiation resulted in closure of the fuel-clad gap and prevented cladding collapse and distortion.

- Although the Zircaloy cladding may be calculated to be freestanding, ovality results from creep which occurs early in the irradiation. The creep rate may be enhanced during neutron irradiation.
- There was no evidence of general or local clad swelling in any of the rods.
- There was no evidence of ridging at pellet interfaces in either the hot-press or cold-press-sinter pellet fuel rods.

FISSION GAS RELEASE

Measurements made on mixed-oxide rods during irradiation show that the release of sorbed gases and moisture from the fuel does not contribute to internal pressure.⁽¹⁵⁾ The released gases apparently react rapidly with the Zircaloy cladding and/or the lower temperature fuel. Internal gas pressure resulting from the release of fission gases increases in a stepwise manner during reactor shutdown-startup power cycles with no significant changes occurring during steady power operation. This behavior, which is the same for UO_2 and UO_2 - PuO_2 fuels, is consistent with the diffusion-trapping release mode⁽¹⁶⁾ and the postulated containment of released fission gases in the central cavity of the rod until the fuel is cooled and cracking occurs.⁽¹⁷⁾

The fission gas release curve (Figure 2), which is described by a two term polynomial yielding a straight line on linear coordinates, represents a calculated least squares fit to the data for the vipac rods.⁽¹⁴⁾ Each data point was given equal weighting in the calculation. The data indicates a gas release of under 5 percent in vipac rods for maximum fuel temperatures up to about $1200^{\circ}C$, i.e., volumetric average temperatures less than $650^{\circ}C$. At maximum fuel temperatures ($1500 - 1700^{\circ}C$) sufficient to cause equiaxed grain growth, which produces a corresponding

FIGURE 2. FISSION GAS RELEASE

volumetric average fuel rod temperature of about 800°C, the gas release increases rapidly to about 15 percent. At maximum temperatures (~2800°C) sufficient to cause the onset of fuel melting, the volumetric average fuel temperature in vipac PRTR rods is about 1350-1400°C with a corresponding fission gas release of about 50 percent. There is no abrupt change in gas release behavior indicated at this point associated with fuel melting and the release fraction continues to increase; approaching 100 percent at the highest volumetric average fuel rod temperature of about 2200°C attained in PRTR fuel rods. The lack of an abrupt change in gas release associated with the melting transition is not unexpected because oxide fuel releases essentially 100 percent of the fission gases formed at fuel temperatures about 1800°C which is considerably below melting.

Within the peak burnup range included for the PRTR vipac rods, i.e., <500 to 13,000 MWd/MTM, burnup level does not appear to have a noticeable effect on gas release. Although the gas release values are expected to increase with increasing burnup, particularly at the lower volumetric average fuel temperatures, the high fractional release values should not change as significantly with burnup because the rate of gas release from the high temperature fuel is more rapid.

Because of the different physical characteristics of packed-particle and pellet type fuels, differences in fission gas release characteristics can be expected to occur. Data from the limited number of pellet type mixed-oxide fuel rods irradiated in PRTR indicates that for volumetric average fuel rod temperatures below 1100°C, i.e., a maximum temperature of about 2300°C, the gas release from cold-press-sinter pellet fuel is less than from vipac fuel rods (Figure 2). The higher surface area of vipac fuel particles should result in a larger fission gas release contribution from the recoil-knockout mechanism.⁽¹⁸⁾ A large number of trapping sites in pneumatically impacted fuel operating below the recrystallization temperature has been shown to inhibit diffusional release.⁽¹⁹⁾ However, the total contribution of this release mechanism for fuel operating in this temperature region is small. At higher operating temperatures, the gas release fractions for vipac and cold-press-sinter

pellet fuel should become more nearly the same for comparable operating conditions.

In contrast to cold-press-sinter pellet rods, the gas release from hot-press pellet mixed-oxide PRTR fuel rods operating at estimated maximum volumetric average fuel rod temperatures up to 1400°C, appears to be greater than for vipac fuel (Figure 2). It is conjectured that the enhanced gas release behavior could be caused by increased void migration rates, either by surface diffusion or pore-sweeping evaporation-condensation phenomena in the more plastic hyperstoichiometric hot-press mixed-oxide pellet fuel. The high fission gas release fractions indicated in hot-press pellet fuel could also be explained if the actual fuel operating temperatures were higher than the estimated temperatures because little is known about the effective thermal conductivity of this fuel material.

FUEL RESTRUCTURING

Small PuO_2 additions to UO_2 do not have a significant effect on the thermal conductivity of UO_2 .⁽²⁰⁾ Hence, fuel restructuring kinetics, which are affected by temperature and temperature gradient in the fuel, should not be significantly different for UO_2 and $\text{UO}_2\text{-PuO}_2$ fuels operating under comparable conditions. Restructuring phenomena are more dependent on fuel type.

Sintered mixed-oxide fuel material produced from powder prepared by the coprecipitation process is essentially uniform $(\text{U, Pu})\text{O}_2$ solid solution with small localized PuO_2 nonuniformity.⁽²¹⁾ However, mechanical mixing of UO_2 and PuO_2 powders prior to pressing and sintering, which is the most commonly used method of preparing mixed-oxide fuels, results in the formation of nonuniform solid solution with localized regions of high PuO_2 concentration.⁽²²⁾ Mechanically mixed and heterogeneously enriched mixed-oxide is also used for packed-particle fuels.

Uniform solid solution forms rapidly in regions of mechanically-mixed mixed-oxide fuel operating above columnar grain growth temperatures ($\sim 1700^\circ\text{C}$) by a vaporization-condensation process whereas, homogenization

occurs more slowly in the equiaxed grain growth regions (1400-1700°C) by solid state diffusion (Figure 3). Therefore, localized regions of high plutonium concentration persist in a significant volume of the fuel in a rod operating under normal conditions. The high fission density associated with these localized regions could affect such fuel performance characteristics as swelling and fission gas release although there is no definitive supportive evidence.

The transport processes which affect fission product migration in oxide fuels during irradiation are not affected by small PuO_2 additions in UO_2 .⁽²³⁾ Fission products such as zirconium, niobium, cerium, and praseodymium, whose oxides are thermodynamically more stable than UO_2 and PuO_2 , do not redistribute significantly. Conversely, fission product cesium, ruthenium, and rhodium, whose oxides are thermodynamically less stable than UO_2 and PuO_2 , migrate significantly during irradiation.

PLUTONIUM REDISTRIBUTION

A significant change in radial plutonium concentration can occur rapidly in mixed-oxide fuel rods during irradiation.⁽²³⁾ For mixed-oxide fuel operating above columnar grain growth temperatures ($\sim 1700^\circ\text{C}$), radial plutonium redistribution is coincident with the fuel restructuring phenomena and occurs as a result of the preferential evaporation of gross amounts of uranium oxide from the central hot region of the fuel to the cooler peripheral regions near the cladding.⁽²³⁾ This phenomenon is most pronounced in low density mixed-oxide fuels that undergo significant radial fuel relocation (Figure 4). Thermal diffusion can also cause plutonium enrichment in the high temperature region of the fuel.⁽²⁴⁾

Electron microprobe scanning analyses of a vibrationally compacted UO_2 -2 wt% PuO_2 fuel specimen irradiated for only 230 hours in PRTR show that the average plutonium concentration in the region of the fuel that operated at columnar grain growth temperatures and above, i.e., $>1700^\circ\text{C}$, is about 10% higher than the nominal as-fabricated concentration (Figure 4). The plutonium concentration in the once-molten fuel region is about 18% higher than the nominal value. Plutonium redistribution effects in mixed-oxide fuels have been shown to be stoichiometry dependent.⁽²⁴⁾

FIGURE 3. COLD-PRESS-SINTER MIXED-OXIDE PELLET FUEL
15-16 KW/FT - 3290 MWD/MTM

FIGURE 4. PLUTONIUM REDISTRIBUTION IN MIXED-OXIDE FUELS DURING IRRADIATION. LOW DENSITY MIXED-OXIDE PELLET FUEL ABOVE AND VIPAC MIXED-OXIDE FUEL BELWO.

FUEL/CLADDING REACTION

Cold-press-sinter and hot-press mixed-oxide pellet fuel rods irradiated in PRTR were clad in Zircaloy-2 tubes whose internal and external surfaces were pre-filmed (400°C for 16 hours in superheated steam) prior to irradiation.⁽¹⁴⁾ This treatment forms a zirconium-oxide film about 1 μm thick on the cladding surface. There was no significant change in the appearance of the uniform zirconium-oxide film formed on the external surface of rods subjected to this treatment after irradiation to peak burnups as high as 8,150 MWd/MTM. Also, there was no significant change in the zirconium-oxide pre-film layer which formed on the inside surface of the cladding on rods containing cold-press-sinter mixed-oxide pellet fuel (Figure 5). However, the film thickness on the inner surface of the cladding on rods containing hot-press mixed-oxide pellet fuel changed significantly during irradiation. In the peak heat flux regions of hot-press pellet rods, the zirconium-oxide layer on the inner cladding surface increased in thickness to about 8 μm with localized regions of severe oxidation occurring (Figure 5). In many locations, the hot-press mixed-oxide pellet fuel reacted or bonded to the zirconium-oxide layer. Where reaction occurred, there was also a tendency for the zirconium-oxide layer on the inner cladding surface to become porous and separate.

A more uniform zirconium-oxide layer, generally about 9 μm thick, formed on the inner cladding surface in the high power regions of packed-particle mixed-oxide fuel rods irradiated in PRTR.⁽¹⁴⁾ A fuel/clad reaction often occurs in vipac rods in which the mixed-oxide fuel particles react or bond with the zirconium-oxide layer apparently without affecting the Zircaloy-2 base metal (Figure 5). As is the case with hot-press mixed-oxide pellet fuel rods, in regions where fuel/clad reactions occur in mixed-oxide vipac rods, the ZrO_2 layer commonly becomes porous and appears to separate from the base metal. Such a separation of the zirconium-oxide layer on the inner surface of Zircaloy-clad rods could have a significant effect on fuel/cladding heat transfer.

COLD-PRESS-SINTER
PELLET

VIBRATIONALLY
COMPACTED

HOT-PRESS PELLET

FIGURE 5. FUEL/CLADDING REACTION IN ZIRCALOY-2 CLAD MIXED-
OXIDE FUELS

Because the fuel/cladding reaction was observed to occur in both vibrationally compacted and hot-press pellet rods and not in cold-press-sinter pellet rods, its occurrence is postulated to be associated with the initial stoichiometry of the fuel, i.e., the reaction occurs in hyperstoichiometric fuel materials and not in stoichiometric or hypo-stoichiometric fuels irradiated to the burnup levels achieved in PRTR. It is further postulated that the increased propensity for hyperstoichiometric mixed-oxide fuel to react with the zirconium-oxide layer is caused by the increased oxidation state of the uranium-ion which reduces the ionic radius and as a result enhances uranium diffusion into the zirconium-oxide layer.

DEFECT BEHAVIOR

Presently, there is no defect experience with mixed-oxide fuels in commercial power reactors. Comparative experiments to investigate the performance of defective and intentionally defected Zircaloy-clad mixed-oxide fuels were conducted in the Plutonium Recycle Test Reactor (PRTR)⁽²⁵⁾ and Engineering Test Reactor (ETR).⁽²⁶⁾

The results of the defect experiments conducted to date show that (1) the defect performance of UO_2 and UO_2 - PuO_2 fuels is essentially the same, although there is evidence that plutonium redistribution may be enhanced in defected mixed-oxide fuels possibly as a result of a change in stoichiometry, i.e., increased O/M,⁽²⁷⁾ (2) defect behavior, i.e., activity release, is most sensitive to fuel type, fuel temperature, and power history,^(27,28) and (3) the defect performance of both pellet and particle oxide fuels is excellent under normal operating conditions. It appears that the defect behavior of ceramic fuels is more sensitive to surface heat flux than to fuel type.^(27,28) Consequently, as more severe operating conditions are imposed on the fuel, defect behavior could become a performance limitation.

A defect test was performed on a mixed-oxide fuel rod in the ETR pressurized loop facility. The pellet-containing rod was nominally 0.565 in. OD clad in 0.030 in. thick Zircaloy-4 cladding with a 26.8 in. active fuel length. The UO_2 -4 wt% PuO_2 pellets were assembled in the cladding with a 0.006 in. diametral gap. Peak burnups of approximately

3000 MWd/MTM were achieved. The system pressure was 2000 psi at an average coolant temperature of 255°C.

The rod was intentionally defected by drilling a 0.020 in. diameter hole through the cladding in the peak power region after the rod was irradiated at a peak linear heat rating of about 20 kW/ft to a peak burn-up of about 1200 MWd/MTM (Figure 6). The defected rod was subsequently irradiated for 17.3 effective full power days at a linear heat rating of about 12.5 kW/ft which produced a maximum fuel temperature of approximately 1650°C. Rotation of the element increased the heat rating at the defect to the range of 20.2 to 22.4 kW/ft with calculated maximum fuel temperatures in the range of 2620°C to >2800°C, i.e., > fuel melting, for an additional 14.1 effective full power days.

During irradiation of the defected rod at the lower heat rating, gross gamma activity levels monitored in the loop piping system increased in proportion with power with no activity bursts but remained well within acceptable limits. At the higher heat rating, activity bursts occurred that were associated with changes in reactor power level, some of which were severe. However, the steady state activity levels for both modes of operation were comparable.

An insignificant amount of $\text{UO}_2\text{-PuO}_2$ fuel washout occurred during irradiation of the defected rod with fuel melting extending to about 23% of the radius at the time of shutdown (Figure 6). As indicated above, the observed fuel melting condition in the defected rod is within the range of calculated maximum fuel temperatures when assuming that no property changes occurred in the mixed-oxide fuel as a result of operating in the defected mode. Fuel oxidation to form U_4O_9 Widmännstätten type precipitates in the mixed-oxide fuel microstructure occurred in the defected rod at distances of 6 inches away from the defect. A high U_4O_9 concentration was present near the thermal center of the fuel as evidenced by the formation of a continuous U_4O_9 network.

Essentially the same amount of fuel melting occurred in both the defected and a companion nondefected rod. Because the two rods were irradiated at comparable peak linear heat rating conditions, it appears that, within the experimental uncertainties associated with this experiment, the oxidation of the fuel in the defected rod had an insignificant effect on the melting heat rating.

FIGURE 6. DEFECT BEHAVIOR OF ZIRCALOY-2 CLAD, COLD-PRESS-SINTER MIXED-OXIDE PELLET FUEL OPERATED WITH FUEL MELTING.

TRANSIENT BEHAVIOR

The behavior of oxide fuels when subjected to accidental high energy, short duration, power excursions is an important safety consideration in both thermal and fast reactors. Experiments were conducted to investigate and compare the transient behavior of the different plutonium-enriched fuel types^(13,29) and to investigate the possible effect of large PuO_2 particles or agglomerates in mixed-oxide fuels.⁽³⁰⁾

The results of comparative transient experiments performed with unirradiated pellet and particle fuel pins^(13,29) show that:

- The cladding failure threshold energy of approximately 270 cal/gm fuel is essentially the same for both pellet and particle fuels.
- For energy inputs near the cladding failure threshold value, the failure mechanisms are different for the two fuel types. In vipac fuel, the cladding fails by contact with molten UO_2 whereas, in pellet fuel, the high temperature and molten cladding rapidly deteriorate due to the reaction with steam without the presence of molten fuel.
- The extent of Zircaloy-water reaction is comparable for both pellet and particle fuels and increases with increasing total energy deposition.

Transient tests were conducted on unirradiated oxide pellet-containing thermal recycle fuel pins to investigate the possible effects of large single 550 μm diameter PuO_2 particles, which could possibly be present in the fabricated fuel, on the transient behavior.⁽³⁰⁾ The results are compared with the results of similar tests conducted on pins containing enriched UO_2 and UO_2 - PuO_2 pellet fuel which did not contain abnormally large PuO_2 particles.

These tests demonstrate that below the energy deposition range of 200 to 213 cal/gm of fuel the energy generated in a single 550 μm diameter PuO_2 microsphere is readily absorbed by the surrounding UO_2 matrix and no external effects occur. Above this energy range, large PuO_2 particles near (0 to 800 μm) the pellet surface can penetrate the cladding (Figure 7).

FIGURE 7. EFFECT OF LARGE PuO_2 PARTICLES ON THE TRANSIENT BEHAVIOR OF MIXED-OXIDE FUELS

The cladding perforation threshold for thermal recycle pellet fuel containing 550 μm diameter PuO_2 agglomerates is in the energy deposition range of 200 to 213 cal/gm of fuel. Perforation occurs by localized melting caused by the expulsion of PuO_2 particles located near the pellet surface through the cladding. Fuel loss to the water is limited to only the PuO_2 particle. Other tests have shown that the prompt cladding failure threshold due to melting for unirradiated pins containing enriched UO_2 and UO_2 - PuO_2 pellet fuel is in the range of 225 to 274 cal/gm of fuel. Therefore, the presence of large 550 μm diameter particles in the fuel has slightly reduced the threshold energy deposition required for cladding perforation and the subsequent release of fission products. However, there were no indications of the effects of prompt fuel dispersal caused by the expulsion of the PuO_2 particles into the surrounding water when tested at these energy levels.

Because the presence of single 550 μm diameter PuO_2 particles in mixed-oxide fuels does not appear to significantly affect the cladding failure threshold energy from that of mixed-oxide fuels with the normal PuO_2 particle size and distribution, product specifications which limit the maximum PuO_2 particle size below 550 μm diameter do not appear to be warranted from the standpoint of transient fuel performance considerations. Specifications should be developed which limit the maximum PuO_2 particle size. These experiments have shown that such a limit is greater than 550 μm diameter.

GENERAL

The results of these experimental programs have culminated in the initiation of demonstration programs in commercial power reactors (Table I) with the rapid full-scale implementation of plutonium utilization in commercial plants anticipated in a timely manner. Because of the current state of the industry, cold-press-sinter pellet fuel is presently the reference mixed-oxide fuel type for commercial application. However, vibrationally compacted fuel is considered to be the most promising alternate fuel form. Under the operating conditions presently employed or projected for commercial power reactors in the near future, neither

pellet nor packed particle mixed-oxide fuels appear to be confronted with inherent performance limitations.

Although there do not appear to be any performance limitations of significance that are unique to mixed-oxide fuels, for ease of licensing, the first mixed-oxide fuel designs introduced into commercial plants on a reload scale will be based as much as possible upon proven technology and plutonium enrichment will be simply substituted for ^{235}U . Currently, most attention related to the introduction of commercial plutonium recycle is focused on environmental, safeguards, shipping, handling, safety, and licensing considerations.

There are significant fuel design and fabrication considerations unique with plutonium-containing fuels. For instance, one of the primary considerations is associated with fuel fabrication quality assurance as related to PuO_2 particle size and distribution requirements in the absence of experimentally determined limits. Neutronically, special considerations are related to control worth and power distributions among rods or plutonium-containing fuel assemblies. In order to better optimize plutonium utilization, future fuel designs must be developed which exploit the unique characteristics of plutonium to improve power sharing among rods in fuel assemblies, extend reactivity lifetime, and further improve fuel operating characteristics.

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