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IRRADIATION PROPERTIES OF HIGH ENERGY RATE
PNEUMATICALLY IMPACTED $\text{UO}_2\text{-PuO}_2$ FUELS

W. J. BAILEY
M. D. FRESHLEY

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By

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Materials Department

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IRRADIATION PROPERTIES OF HIGH ENERGY RATE PNEUMATICALLY IMPACTED UO_2 - PuO_2 FUELS

W. J. Bailey
M. D. Freshley

INTRODUCTION

During the last three years, a significant amount of UO_2 - PuO_2 fuel material prepared by the pneumatic impaction process was fabricated into experimental capsules, prototype elements, and full size fuel elements for irradiation testing in the Materials Testing Reactor (MTR), Engineering Test Reactor (ETR), Plutonium Recycle Test Reactor (PRTR), Experimental Boiling Water Reactor (EBWR), Saxton Reactor, and the Experimental Breeder Reactor II (EBR-II). The PuO_2 concentration in the mixed-oxide fuel ranged from 1 to 20 wt% PuO_2 . Results are presently available on a series of capsule experiments conducted in support of the joint Pacific Northwest Laboratory* - Argonne National Laboratory, Plutonium Recycle Demonstration Experiment in EBWR^(1,2) and fuel element irradiations conducted in the PRTR.^(3,4)

In early packed particle fuel development work using UO_2 , arc fusing was employed satisfactorily to produce the high density particles needed to ensure maximum thermal

conductivity, bulk fuel density, and fission gas retention in the finished swage-compacted or vibrationally compacted fuel rods. Since arc fusion was not considered feasible for use with UO_2 - PuO_2 fuel because of selective vaporization of PuO_2 and the resulting difficulties in controlling composition, pneumatic impaction, a closed die, hot forging technique was developed.⁽⁵⁾ Pneumatic impaction provides a means of consolidating mixtures of UO_2 and PuO_2 particles so as to maintain a uniform distribution of the two materials in particles of near-theoretical density during subsequent processing and irradiation.

SUMMARY AND CONCLUSIONS

Pneumatic impaction is a versatile process for producing high density particles of mechanically mixed UO_2 - PuO_2 fuel material suitable for swaging or vibrational compaction. The PuO_2 concentration and UO_2 and PuO_2 particle sizes can be varied over a wide range to make the fuel suitable for either thermal or fast reactor applications. Engineering proof tests, conducted with capsule irradiations in MTR/ETR and irradiations of full-scale fuel elements under power reactor conditions in PRTR, have demonstrated the satisfactory behavior of the fuel.

*Pacific Northwest Laboratory, Richland, Washington is operated by Battelle Memorial Institute for the U. S. Atomic Energy Commission under contract No. AT-(45-1)-1830.

Irradiation performance of test capsules and PRTR fuel elements containing pneumatically impacted UO_2 - PuO_2 fuel material has demonstrated that mixed oxides prepared by this process are comparable to or better than ceramic fuels prepared by other processes. The satisfactory performance has been demonstrated for linear rod powers to 1276 W/cm (39 kW/ft) and to burnups as high as 5.91×10^{20} fissions/cm³ (23,800 MWd/tonne).

Structures formed during irradiation of pneumatically impacted UO_2 - PuO_2 are similar to those formed during the irradiation of UO_2 . Homogenization of the mechanically mixed UO_2 - PuO_2 occurs at fuel temperatures sufficient to cause in-reactor sintering and grain growth.

Fission products migrate to preferred locations at the higher fuel temperatures. Of the fission products studied, Zr-Nb⁹⁵, Sr⁹⁰, and Ce-Pr¹⁴⁴ migrate the least, if at all; whereas, Cs¹³⁷ and Ru¹⁰⁶ migrate the most. Alpha autoradiographs indicate that, under certain conditions (i.e., molten fuel defect testing), plutonium segregates from the liquid and migrates in the solid in high-performance UO_2 - PuO_2 fuels. This behavior is not considered to be unique with pneumatically impacted fuel and can be expected to occur in fuels fabricated by processes other than pneumatic impaction. Chemical analyses have not substantiated plutonium segregation and migration during irradiation.

There is good agreement between the fission gas release characteristics of capsules and full-sized PRTR

elements. Fission gas release values of ~90% are obtained for volumetric average fuel rod temperatures of ~2200 °C.

PNEUMATIC IMPACTION

Pneumatic impaction provides a means of producing high density UO_2 - PuO_2 fuel particles with any desired PuO_2 composition. The particles are used as feed material for packed-particle fuel elements fabricated by either the swage-compaction or vibrational-compaction processes.⁽⁵⁾

Appropriate particle sizes and proportions of UO_2 and PuO_2 powders are mechanically blended prior to being loaded into a double stainless steel can assembly. After ~6 kg of the powder mixture are loaded into the inner can, the outside surface is decontaminated to remove any plutonium. The outer can lid, which has an attached evacuation tube and porous stainless steel filter, is then welded to the outer can. The assembly is evacuated through the filter and tube while being heated to ~1200 °C. The degree of plasticity that the oxides display during impaction at 1200 °C aids in the densification process. After reaching 1200 °C, the assembly is removed from the furnace and the evacuation tube is pinched off. The assembly is then impacted in the die of the machine. Peak pressures as high as ~250,000 lb/in.² (1.76×10^8 kg/m²) are exerted on the assembly. As much as 145,000 ft-lb of work can be delivered to the assembly within several milliseconds.

After impaction, the assembly is removed from the die, cooled, and grit blasted. The cleaning operation is necessary to prevent the introduction of impurities (such as die lubricant) into the fuel during the subsequent decanning operation. The high density mixed-oxide is then pulverized and mechanically sieved into different size fractions suitable for either swaging or vibrational-compaction.

Control of the oxygen/metal ratio and sorbed gas content of the pre-impacted powders are important factors in obtaining high density fuel material. Slightly hyperstoichiometric UO_2 - PuO_2 feed material is required for the impaction process since the excess oxygen enhances plasticity and produces higher impacted fuel densities. Mixed-oxide particle densities $>98\%$ TD are obtained routinely with material having an oxygen-to-metal (O/M) ratio of 2.02 and a sorbed gas content of $\sim 0.07 \text{ cm}^3/\text{g}$.

REACTOR REQUIREMENTS

The design of fuel elements incorporating mixed-oxides must include factors that affect inherent nuclear safety, such as the Doppler response if a power excursion occurs. Because pneumatically impacted mixed-oxide fuels consist of discrete UO_2 and PuO_2 particles, the size and distribution of the particles as related to their effect on Doppler response is an important consideration.⁽⁶⁾ The versatility of the pneumatic impaction process for mixed-oxides permits a wide range of PuO_2 concen-

trations suitable for either thermal or fast reactor fuel. The particle size of the preimpacted UO_2 and PuO_2 powders may vary depending upon the particular reactor application.

PuO_2 distribution and particle size criteria for low plutonium enrichment mixed-oxide thermal reactor fuels are not as critical as those for high enrichment fast reactor fuels because of the slower neutron kinetics of thermal reactors. Calculations of Doppler response for mixed-oxide fuels have been made by utilizing an effective criterion for the Doppler response—this criterion being that the time constant for heating of UO_2 must be less than the shortest attainable reactor period.⁽⁶⁾ The results show that mechanically blended -65 mesh (Tyler screen size) UO_2 and -325 mesh PuO_2 preimpacted particles provide "safe" fuel for thermal reactors such as the PRTR and EBWR.⁽⁶⁾

UO_2 -20 wt% PuO_2 fuel mixtures, which meet fast reactor criteria of particle size and distribution,^(7,8) have been prepared by pneumatically impacting ball-milled UO_2 and PuO_2 powders.⁽⁹⁾ Alpha autoradiography was used as a qualitative measure of plutonium distribution. At a magnification of 30X, the technique showed a discernible difference, not readily apparent at 500X in plutonium distribution, between coprecipitated-sintered and ball-milled-impacted UO_2 -20 wt% PuO_2 materials. Statistical analysis of electron-microprobe data showed the mean PuO_2 particle size of 64 hr ball-milled and pneumatically impacted fuel material to be $1.9 \mu\text{m}$, considerably

less than the conservatively calculated value of 20 μm which is sufficient to assure a satisfactory Doppler response rate for reactor periods as small as 0.1 msec.⁽¹⁰⁾

TEST CAPSULE IRRADIATION EXPERIENCE

A series of 32 capsules containing pneumatically impacted UO_2 -1.5 wt% PuO_2 fuel is being irradiated in MTR in support of the Plutonium Recycle Demonstration Experiment in EBWR.^(1,2) The capsules have performed satisfactorily and no failures have occurred. Postirradiation examination has been performed on 29 of the capsules, and the remaining three (current maximum burnup is 5.91×10^{20} fissions/cm³ or 23,800 MWd/tonne) are still undergoing irradiation. In addition, full length EBWR fuel rods⁽¹¹⁾ are being irradiated; of 58 EBWR prototype rods irradiated to date, 13 have been discharged (one examined). All rods have performed satisfactorily and no failures have been encountered.

The capsules were fabricated by vibrationally compacting pneumatically-impacted fuel material into the Zircaloy-2 (low-nickel) cladding, 1.08 cm (0.426 in.) OD by 0.064 cm (0.025 in.) wall by 7.6 to 10 cm (3 to 4 in.) long, to densities ranging from 77 to 85% TD. The following UO_2 - PuO_2 particle size fractions were used:

wt%	Mesh Size
55	-6 +10
12.8	-20 +35
12.2	-35 +65
5.5	-200 +325
14.5	-325

Seven of the capsules incorporated prototype fission gas plenums internally supported by a Type-302 SS spring. The spring is separated from the fuel by a 0.64 cm (0.25 in.) thick ZrO_2 pellet. All specimens were sheathed in secondary aluminum jackets (Figure 1), 2.9 cm (1.125 in.) OD, to provide additional plutonium

Neg 5439

FIGURE 1. Typical Appearance of an Irradiated Double-Clad Capsule. The Zircaloy-Clad UO_2 -1.5 wt% PuO_2 Capsule is, in turn, Clad with Aluminum (2.9 cm OD). This Capsule, GEH-14-528, Attained a Burnup of 2.26×10^{20} Fissions/cm³ (9050 MWd/tonne) and had a Maximum Linear Rod Power of 577 W/cm (18 kW/ft). The Removable Cap Allows Changing of the Four, Full-Length, Axial Flux-monitoring Wires in the Aluminum Sheath.

containment and avoid coolant boiling on the surface. The aluminum jackets contain four, full length, replaceable Al-Co flux monitoring wires (Figure 2).

The specimens have been irradiated under a variety of conditions. Linear rod powers ranged from 180 W/cm (5 kW/ft) to 1276 W/cm (39 kW/ft). Burnups ranged from 0.24×10^{20}

fissions/cm³ (1010 MWd/tonne) to 6.7×10^{20} fissions/cm³ (26,800 MWd/tonne). Burnup values are determined from the flux wire data and/or isotopic analysis of the plutonium in conjunction with a computer burnup code. Generally, excellent agreement has occurred in the burnup values obtained by the two methods. A summary of the irradiation conditions and

FIGURE 2. Transverse Section at the Midplane of an Irradiated Double-clad Capsule (GEH-14-528). One of the Four Flux-monitoring Wires was Inadvertently Left in a Peripheral Position in the Aluminum Sheath. This Section Shows the Fission Gas Collection Hole. The Zircaloy Cladding is 1.08 cm OD. ($\sim 3.6X$)

fission gas release data (Section VII) for the 32 capsules (also for one full-size rod, EI-36) is given in Table I.

The linear rod power values tabulated are the initial conditions (average). Burnup samples from two of the 29

TABLE I. Irradiation Testing of Vibrationally Compacted Capsules Containing Pneumatically Impacted, UO_2 -1.5 wt% PuO_2

Capsule GEH-14 No. (a)	Bulk Fuel Density (%TD)	Heat Flux (b) (W/cm ²)	Linear Rod Power (b)		Burnup (b)		Fission Gas Release (%)
			(W/cm)	(kW/ft)	(Fissions/ cm ³) $\times 10^{-20}$	(MWd/tonne of UO_2 - PuO_2)	
517	82	53	180	5.49	0.44	1,840	5
518	82	113	380	11.6	0.40	1,660	14
519	83	340	1,160	35.2	0.61	2,480	Lost
520	82	126	427	13.0	1.73	7,070	38
521	83	139	472	14.4	3.49	14,170	37
522	82	127	431	13.1	3.15	12,930	39
523	84	(158)	(535)	(16.3)	(6.8)	(27,500)	In-reactor
524	83	(158)	(535)	(16.3)	(6.8)	(27,500)	In-reactor
525	80	155	522	15.8	1.25	4,570	54
526	82	125	425	13.0	1.1	4,820	
527	79	129	436	13.3	2.36	10,050	32
528	84	170	577	17.6	2.26	9,050	41
529	84	158	535	16.3	5.5	22,100	
646 (c)	80	147	501	15.3	0.24	1,010	30
647 (c)	77	147	500	15.2	0.72	3,140	22
648 (c)	81	190	635	19.4	3.69	15,400	30
649 (c)	79	188	641	19.5	1.55	6,640	33
650 (c)	80	304	1,037	31.6	0.46	1,950	81
651 (c)	79	374	1,276	38.6	1.6	6,860	84
706	81	298	1,013	30.6	0.55	2,260	51
707	83	298	1,010	30.8	2.25	9,150	94
708	81	125	424	12.9	0.71	2,950	18
709	81	138	465	14.2	1.3	5,440	27
710	81	313	1,070	32.4	1.3	5,180	~100
711	81	219	745	22.7	2.54	10,500	41
712	84	185	630	19.2	3.52	14,200	
713	85	267	905	27.6	2.95	11,800	
714	77	96	327	9.97	0.51	2,220	10
715	78	100	341	10.4	0.82	3,570	
716	78	103	352	10.7	0.81	3,520	
717	80	132	448	13.7	0.48	2,010	14
718 (c)	79	218	745	22.7	0.40	1,730	52
EI-36 (d)	86-89	61	208	6.35	0.52	2,010	6

(a) All capsules contain impacted fuel. Tabular values in parenthesis are proposed or design values. Zircaloy-2 (low-nickel) cladding OD is 1.08 cm (0.426 in.).

(b) Values are based on flux-monitoring wire data and/or burnup analyses. Rod powers listed are the maximum initial values, as calculated by using the average flux over a nominal 15 day run.

(c) Capsules equipped with prototype plenums.

(d) This full-size EBWR rod was irradiated in the PRTR (Fuel Element No. 6501). Rod dimensions were: OD, 1.08 cm (0.424 in.); overall length, 148 cm (58.3 in.); active fuel, 123 cm (48.5 in.); plenum, 14.6 cm (5.75 in.); and nominal Zircaloy-2 (low-nickel) cladding thickness, 0.064 cm (0.025 in.).

discharged capsules were isotopically analyzed, and the results were converted to burnup values by means of computer program (MELEAGER) calculations (Table II). Based on seven samples, the flux wire data tends to underestimate the actual fuel burnup.

Transverse sections at the mid-plane of the specimens illustrate the effect of different rod powers on the fuel structures formed during irradiation. The specimen (GEH-14-714) shown in Figure 3 operated at a rod power of 327 W/cm (10 kW/ft). The cross-section exhibited little or no sintering as a result of irradiation under these condi-

tions, and the impacted UO_2 -1.5 wt% PuO_2 fuel particles have retained their original character. The multigrain structure of the impacted material is evident in the larger fuel particles. The beta-gamma autoradiograph shows localized concentrations of fission products associated with discrete PuO_2 particles. (NOTE: All beta-gamma autoradiographs described in this report were made by exposing ceramographic samples of the irradiated fuel specimens to high resolution film plates which, when developed, indicate the presence of fission products. The dark areas in the figures correspond to

TABLE II. Comparison of Burnup Determinations by Isotopic Analysis and from Flux-Wire Data

Item	Flux-Wire Data		Isotopic Analysis of Fuel Sample	
	Measured ^(a)	Calculated	Measured ^(b)	Calculated
<u>Capsule GEH-14-521:</u>				
nvt	22.4 x 10 ²⁰			26.7 x 10 ²⁰
%U ²³⁵ remaining in U		0.051	0.042 ± 0.002	
MWd/tonne of fuel		12,450		14,170
Fission/cm ³		3.07 x 10 ²⁰		3.49 x 10 ²⁰
Fission gas release, %		42		37
<u>Capsule GEH-14-648:</u>				
nvt	29.2 x 10 ²⁰			29.5 x 10 ²⁰
%U ²³⁵ remaining in U		0.033	0.031 ± 0.002	
MWd/tonne of fuel		15,120		15,400
Fission/cm ³		3.62 x 10 ²⁰		3.69 x 10 ²⁰
Fission gas release, %		31		30

a) Possible error, ±19%

b) Uncertainties are 95% confidence limits

Photomacrograph

β - γ Autoradiograph

Neg 0643027-2

FIGURE 3. Photomacrograph and Beta-Gamma Autoradiograph of a Transverse Section from Irradiated UO_2 -1.5 wt% PuO_2 Specimen (GEH-14-714). Specimen Operated at a Maximum Rod Power of 327 W/cm (10 kW/ft) to a Burnup of 0.51×10^{20} Fissions/cm³ (2220 MWd/tonne). Cladding OD is 1.08 cm (0.426 in.). (6.4X)

regions of high radioactivity). The autoradiograph also illustrates that the PuO_2 is uniformly distributed within the impacted UO_2 -1.5 wt% PuO_2 fuel since the large impacted fuel particles are not distinguishable on the autoradiograph.

At slightly higher rod powers, fuel temperatures during irradiation are sufficiently high to cause sintering and grain growth. The specimen (GEH-14-525) shown in Figure 4 operated at a linear rod power of 522 W/cm (16 kW/ft). As indicated by a smearing of the radioactivity near the center of the beta-gamma autoradiograph, in-reactor homogenization of the mechanically mixed and impacted UO_2 -1.5 wt% PuO_2 fuel commences at sintering and grain growth temperatures ($\sim 1700^\circ\text{C}$). Localized concentrations of fission products associated with discrete PuO_2 particles are still evident in the cooler peripheral regions of the specimen where homogenization did not take place.

Fuel melting, delineated by the centrally located porous region in the specimen (GEH-14-706) shown in Figure 5, occurs at still higher rod powers. Melting occurred to $\sim 45\%$ of the radius in this specimen that operated at a rod power of 1013 W/cm (31 kW/ft) to a burnup of 0.55×10^{20} fissions/cm³ (2260 MWd/tonne). The central porous region is surrounded by a band of large high density grains which, in turn, are surrounded by a ring of small radially oriented columnar grains. This region is surrounded by a narrow band of equiaxed grains and then by the unsintered fuel in the lower temperature peripheral region near the cladding

Photomicrograph

Photomicrograph

β - γ Autoradiograph

Neg 0643284-1

FIGURE 4. Photomicrograph and Beta-Gamma Autoradiograph of a Transverse Section from Irradiated UO_2 -1.5 wt% PuO_2 Specimen (GEH-14-525). Specimen Operated at a Maximum Rod Power of 522 W/cm (16 kW/ft) to a Burnup of 1.25×10^{20} Fissions/cm³ (4570 MWd/tonne). Cladding OD is 1.08 cm (0.426 in.). (6.4X)

β - γ Autoradiograph

Neg 0643027-3

FIGURE 5. Photomicrograph and Beta-Gamma Autoradiograph of a Transverse Section from Irradiated UO_2 -1.5 wt% PuO_2 Specimen (GEH-14-706). Specimen Operated at a Maximum Rod Power of 1013 W/cm (31 kW/ft) to a burnup of 0.55×10^{20} Fissions/cm³ (2260 MWd/tonne). Cladding OD is 1.08 cm (0.426 in.). (6.4X)

of the specimen. This structure is typical of those formed in specimens operating with molten fuel at the time of shutdown. The beta-gamma autoradiograph shows that certain fission product species have migrated to selected radial locations coincident with structural changes within the fuel. Significantly, even though fuel melting and extensive sintering and grain growth occurred in this specimen during irradiation, the cooler peripheral regions of the fuel are essentially unchanged, thus they provide an effective barrier between the molten fuel and the Zircaloy cladding.

Mixed-oxide fuel structures formed during irradiation are not only affected by power generation, but also by irradiation history. The effect of an initially high but steadily decreasing linear rod power and increased exposure is reflected in the appearance of the specimen (GEH-14-707) shown in Figure 6. The initial maximum power generation in this specimen was the same as for the specimen shown in Figure 5, but the burnup was approximately 3.5 times greater. The maximum rod power value, 1010 W/cm or 31 kW/ft, (Table I), occurred at the beginning of irradiation and decreased gradually with time to approximately 280 W/cm (9 kW/ft) at a burnup of 2.25×10^{20} fissions/cm³ (9150 MWd/tonne). Time temperature dependent solid state diffusion phenomena have erased all evidence of the once-molten fuel structure. The fuel structure with long narrow columnar grains, shown in the photomacrograph, and the fission product distribution pattern, illustrated in the beta-gamma autoradiograph, are typical of

Photomacrograph

β - γ Autoradiograph

Neg 0643284-2

FIGURE 6. Photomacrograph and Beta-Gamma Autoradiograph of a Transverse Section from Irradiated UO_2 -1.5 wt% PuO_2 Specimen (GEH-14-707). The Specimen Operated Initially at a Maximum Rod Power of 1010 W/cm (31 kW/ft) to a Burnup of 2.25×10^{20} Fissions/cm³ (9150 MWd/tonne). The Power Generation Decreased Uniformly to 280 W/cm (9 kW/ft) at the Time of Discharge. Cladding is 1.08 cm (0.426 in.). (6.4X)

ceramic fuels irradiated to high burn-up at maximum fuel temperatures below melting.

High power generation in low density vibrationally compacted fuel rods causes axial fuel relocation or slumping. Fuel slumping occurred during the irradiation of Specimen GEH-14-706 (Figure 7) which had an initial bulk fuel density of 81% TD and operated at a power generation of 1013 W/cm (31 kW/ft) with significant fuel melting (same specimen as in Figure 5). Considerable reaction occurred between the UO_2 -1.5 wt% PuO_2 fuel and the ZrO_2 spacer pellet located at the top of the fuel column. This longitudinal section dramatically illustrates the hazard of evaluating irradiation conditions of capsule specimens based upon a single transverse section because fuel structures can change markedly over relatively short axial distances.

Postirradiation examination of this series of capsules continues to show excellent irradiation behavior of vibrationally compacted, pneumatically impacted UO_2 -1.5 wt% PuO_2 fuel. No failures have occurred. This performance was achieved under conditions of significant fuel melting and, in some instances, under such abnormally severe conditions that the ZrO_2 spacer pellet and stainless steel plenum support spring were consumed by the high temperature UO_2 - PuO_2 fuel.

Full length EBWR fuel rods,⁽¹¹⁾ ~2000 fabricated by Pacific Northwest Laboratory (PNL), are currently being irradiated (see Appendix) to evaluate the fabrication process variables and the joint PNL-ANL rod design. The rods

Photomacrograph β - γ Autoradiograph

Neg 0661473-3

FIGURE 7. Photomacrograph and Beta-Gamma Autoradiograph of Longitudinal Sections Near the Top and Bottom of Irradiated UO_2 -1.5 wt% PuO_2 Specimen GEH-14-706. The Specimen Operated at a Maximum Rod Power of 1013 W/cm (31 kW/ft) to a Burnup of 0.55×10^{20} Fissions/cm³ (2260 MWd/tonne). Cladding OD is 1.08 cm (0.426 in.). (~2.6X)

are designed for a burnup of 6.8×10^{20} fissions/cm³ (27,500 MWd/tonne). The burnup region of greatest interest for the cooperative PNL-ANL Plutonium Recycle Demonstration Experiment in the EBWR is $\sim 2.5 \times 10^{20}$ fissions/cm³ ($\sim 10,000$ MWd/tonne); however, some of the rods may be irradiated to a burnup of 6.8×10^{20} fissions/cm³ (27,500 MWd/tonne). The physics objectives of the PNL-ANL experiment are to obtain data on reactivity, isotopic, and material changes as a function of core burnup.

Another objective is to obtain a correlation between the short-length capsule performance and the full-size rod performance data. Also the irradiations can provide lead time (in terms of burnup) for corrective action on unanticipated problems prior to the irradiation of the 1296 rod loading in EBWR.

Each vibrationally compacted, Zircaloy-clad rod contains 830 ± 5 g of pneumatically impacted, UO_2 -1.5 wt% PuO_2 fuel. The uranium contains 0.22 wt% U^{235} and the plutonium contains ~ 8 wt% Pu^{240} . The impacted fuel density is $\geq 98\%$ TD, with an associated bulk density for the vibrationally compacted fuel of 86 to 89% TD. The rods are ~ 1.08 cm (~ 0.424 in.) OD by 148 cm (58.3 in.) long.

The EBWR prototype rods are being irradiated in two types of fuel elements: a 21 rod bundle with replaceable rods, and three-rod clusters with nonreplaceable rods. Goal burnups $> 2.5 \times 10^{20}$ fissions/cm³ ($> 10,000$ MWd/tonne) are planned for rods in the 21 rod bundle. One rod (EI-36) was removed from the element and replaced as planned

at a burnup of 0.52×10^{20} fissions/cm³ (2010 MWd/tonne) and examined (Table I). The rod was in good condition and showed no evidence of external corrosion in the bottom end cap region, the only unautoclaved part of the rod. The fuel released 6% of the fission gas (Section VII). The rod operated a maximum heat flux of 61 W/cm^2 ($194,000 \text{ Btu/hr-ft}^2$), which is approximately the same as the currently proposed maximum EBWR conditions*. Four to six additional rods will be inserted (two each time) in the bundle at burnups of $\sim 5,000$, $10,000$, and $> 10,000$ MWd/tonne. The bundle has operated satisfactorily, and no failures have occurred to the present maximum burnup of 0.7×10^{20} fissions/cm³ (2690 MWd/tonne).

Of the 36 rods being irradiated in three-rod clusters, 12 have been discharged. Cluster goal burnups are 0.4×10^{20} fissions/cm³ (1650 MWd/tonne); this group was discharged, 1.4×10^{20} fissions/cm³ (5500 MWd/tonne), and 6.8×10^{20} fissions/cm³ (27,500 MWd/tonne). To date, the clusters have exhibited good performance with no failures. The irradiation data disclosed that the clusters operated initially at predicted linear power ratings, and that the change in linear power with burnup agrees with the calculational model within 6%.

PRTR FUEL ELEMENT IRRADIATION EXPERIENCE

Most of the fuel element irradiation experience with pneumatically impacted

*Originally, the maximum heat flux value was set at 158 W/cm^2 ($500,000 \text{ Btu/hr-ft}^2$), but because of EBWR control problems, the limit was subsequently reduced. The capsule and rod tests were initiated while the original limit was in force.

UO₂-PuO₂ fuels has been obtained in the Plutonium Recycle Test Reactor (PRTR).^(3,4) Nineteen-rod cluster fuel elements containing pneumatically impacted UO₂-PuO₂ fuel are being irradiated under power reactor conditions in the PRTR. The fuel assemblies comprise 19 Zircaloy clad swaged or vibrationally compacted fuel rods 1.435 cm (0.565 in.) OD with an 0.076 cm (0.030 in.) wall thickness. The elements containing UO₂-1 wt% PuO₂ fuel are 224 cm (88 in.) long; whereas, the rods containing UO₂-2 wt% PuO₂ fuel are 167 cm (66 in.) long. The latter fuel rods were designed for the High Power Density Program (HPD) in PRTR and incorporate a 17.8 cm (7 in.) long fission gas plenum at the top of the rods to accommodate the gases released at higher fuel temperatures (Figure 8).

The Batch Core Experiment, to be conducted as part of the High Power Density Program, is a physics-fuels experiment to investigate the burnup-reactivity characteristics of a plutonium fueled reactor, and the irradiation behavior of a statistically significant number of pneumatically impacted mixed-oxide elements. UO₂-PuO₂ HPD elements will operate at maximum rod powers of 650 W/cm (20 kW/ft) to burnups as high as 3.3×10^{20} fissions/cm³ (13,000 MWd/tonne) with maximum fuel temperatures (2600 to 2800 °C) near melting. Special UO₂-PuO₂ nineteen-rod cluster test elements have been irradiated in the Fuel Element Rupture Test Facility (FERTF), a self-contained high pressure light water cooled test loop, at maximum rod powers as high as 885 W/cm (27 kW/ft) with significant

Neg 0653112

FIGURE 8. PRTR High Power Density Nineteen-Rod Cluster Fuel Element

fuel melting. A summary of PRTR fuel element experience with pneumatically impacted UO₂PuO₂ fuel is given in Table III.

Of the 1140 rods containing pneumatically impacted fuel irradiated in PRTR, sixteen failed during irradiation. None of these failures were related to the pneumatic-impaction treatment of the fuel material. All but one of the failures were directly attributed⁽³⁾ to impurities in the fuel material. These impurities were of three types:

- Fluoride contamination in plutonium
- Sorbed moisture
- Traces of hydrocarbons introduced by failure of mechanical processing equipment.

TABLE III. PRTR Fuel Element Experience (as of January 1, 1967) with Pneumatically Impacted $\text{UO}_2\text{-PuO}_2$

	224 cm long fuel columns, $\text{UO}_2\text{-1% PuO}_2$	147 cm long fuel columns, $\text{UO}_2\text{-2% PuO}_2$
Total number of fuel elements irradiated	46	14
Swage compacted	27	2
Vibrationally compacted	19	12
Number currently in PRTR	32	10
Maximum burnup, fission/ cm^3 (MWd/tonne)	1.91×10^{20} (6920)	1.21×10^{20} (4950)
Maximum rod power of in-core irradiations, W/cm (kW/ft)	545 (16.6)	625 (19)
Maximum rod power of FERTF ^(a) irradiations, W/cm (kW/ft)		885 (27)
Maximum heat flux in-core irradiations, W/ cm^2 (Btu/hr/ft ²)	120 (380,000)	137 (435,000)
Maximum heat flux of FERTF irradiations, W/ cm^2 (Btu/hr/ft ²)		196 (620,000)
Maximum fuel temperature of in-core irradiations, °C	2500	2600-2700
Maximum fuel temperature of FERTF irradiations, °C		above melting

(a) Fuel Element Rupture Test Facility

These fuel rod failures characteristically occurred at low burnups (approximately 0.1×10^{20} fissions/ cm^3 or 300 MWd/tonne because of severe internal hydriding and embrittlement of the Zircaloy cladding caused by a gas phase hydriding mechanism. The primary cause of the other failure has not been determined.

Although severe localized embrittlement and loss of cladding fragments occurred in some instances, little or no fuel loss into the coolant resulted and no severe reactor operating difficulties were experienced. Fuel washout or erosion resistance is improved by in-reactor sintering of the packed particle fuel which occurs during the early stages of irradiation at the higher fuel operating temperatures. Water-logging* has not been observed

in fuel elements that either failed during normal reactor operation or were initially intentionally defected. In many instances, the release of fission product activity was first detected during postshutdown depressurization.

Elements containing pneumatically impacted $\text{UO}_2\text{-2 wt% PuO}_2$ fuel (Figure 9) have been irradiated in the PRTR core at maximum rod power generations of 655 W/cm (20 kW/ft). Considerable in-reactor sintering occurs under these conditions, and a central void and columnar grains form over ~65% of the fuel radius at an estimated maximum fuel temperature of 2600 to 2700 °C. The central void extends over essentially the full length (147 cm) of the fuel rods. Homogenization of the $\text{UO}_2\text{-PuO}_2$ is evidenced on the beta-gamma autoradiograph by the lack of localized fission product concentrations associated with discrete PuO_2 particles in the columnar grain growth region (Figure 10). High

*Massive clad failure resulting from rapid internal pressure buildup under constrained conditions due to vaporization of coolant water entering a defected fuel rod.

Photomicrograph

β - γ Autoradiograph

Neg 0653144-3

FIGURE 9. Transverse Section Through the Axial Midpoint of a Vibrationally Compacted PRTR Fuel Rod Containing Pneumatically Impacted UO_2 -2 wt% PuO_2 . The Rod Operated at a Maximum Power of 655 W/cm (20 kW/ft) to a Burnup of 0.4×10^{20} Fissions/cm³ (1300 MWd/tonne) with Maximum Fuel Temperatures of 2600-2700 °C. Columnar Grain Growth Extends to 65% of the Radius. ($\sim 3.8X$)

magnification photomicrographs (Figure 11) better illustrate PuO_2 diffusion and homogenization in UO_2 - PuO_2 which commence at fuel temperatures sufficient to cause in-reactor sintering. The photomicrographs show the presence of discrete PuO_2 particles in the pneumatically impacted UO_2 -2 wt% PuO_2 fuel near

the periphery of the transverse section (Figure 11) and the commencement of UO_2 and PuO_2 interdiffusion at sintering temperatures. Such in-reactor homogenization is typical of physically mixed pneumatically impacted UO_2 - PuO_2 fuels.

Irradiation experience in PRTR indicates that melting of pneumatically impacted, vibrationally compacted (86% TD) UO_2 -2 wt% PuO_2 fuel commences at a linear rod power generation of 655 to 690 W/cm (20 to 21 kW/ft) under PRTR coolant conditions (nominally 260 °C). (The power generation required to produce the onset of melting is subject to refinement as more data are acquired). This is equivalent⁽¹²⁾ to an $\int_{T_s}^{T_m} K d\theta$ of 52 to 55 W/cm for UO_2 -2 wt% PuO_2 or, when compensating for the difference in self-shielding, it is comparable to an $\int_{T_s}^{T_m} K d\theta$ of 45 to 48 W/cm for similarly vibrationally compacted natural UO_2 where:

T_s = Temperature of fuel surface
= ~ 530 °C

T_m = Temperature of fuel melting
= ~ 2800 °C

k = Thermal conductivity of fuel
 θ = Temperature.

This value is in good agreement with the value of 49 W/cm published by Lyons, et al.⁽¹³⁾ The results show that the power to produce melting in vibrationally compacted fuels is approximately 20% less than for sintered pellet fuel.

Postirradiation examination of fuel structures shows reasonably good agreement between observed and calculated thermal conditions below fuel melting temperatures. The calculations assume that in-reactor sintering of the initially 86% bulk density fuel causes an

FIGURE 10. Transverse Section of a Vibrationally Compacted PRTR Fuel Rod Irradiated at a Maximum Rod Power of $\sim 625 \text{ W/cm}$ (19 kW/ft). Homogenization of the Discrete PuO_2 Particles in the Periphery of the Pneumatically Impacted UO_2 -2 wt% PuO_2 Fuel Specimen Commences at Temperatures Sufficient to Cause Sintering and Grain Growth.

Neg 0661473-5

Neg 0661684-4

FIGURE 11. Photomicrograph of a Transverse Section From an Irradiated PRTR Fuel Rod Containing Vibrationally Compacted Pneumatically Impacted UO_2 -4 wt% PuO_2 Fuel. (156X)

increase in the effective thermal conductivity to the value for 100% TD fuel at temperatures above 1600 °C.

A transverse section at the midplane of a preirradiated vibrationally compacted PRTR fuel rod containing pneumati-

cally impacted UO_2 -2 wt% PuO_2 , which was defected with a 1.6 mm (0.06 in.) diam hole, is shown in Figure 12. The element operated successfully at a maximum linear rod power of $\sim 790 \text{ W/cm}$ (24 kW/ft) with significant fuel melting

Neg 0653047-3

FIGURE 12. Transverse Section Through an Intentionally Defected Vibrationally Compacted PRTR Fuel Rod Containing Pneumatically Impacted UO_2 -2 wt% PuO_2 Fuel. Fuel was Molten to $\sim 55\%$ of the Radius at the Plane of the 1.6 mm diam Hole Drilled Through the Cladding. Maximum Linear Rod Power was $\sim 24 \text{ kW/ft}$. ($\sim 4.3X$)

(55% of the radius molten) at the plane of the defect for ~60 hr before the test was terminated as planned. The defect test element remained in the FERTF for a total of 287 operating hours. Prior irradiation of the rod in the nondefected condition at a maximum power of ~625 W/cm (19 kW/ft) to a burnup of over 1.0×10^{20} fissions/cm³ (3620 MWd/tonne) was sufficient to cause considerable in-reactor sintering of the fuel. Burst type activity release, characteristic of leaker defects, occurred during incremental power increases at the lower power levels. Incremental power increases at the higher levels produced less severe activity releases and, in all cases, the loop activity approached a steady state value in an exponential manner during continued operation. The steady state gamma activity in the loop during irradiation of this element was ~15 times greater than for any defected elements previously tested. However, the rod power was approximately three times greater than for any previous experiment. Activity bursts occurred when the reactor was shut down and also when the loop was depressurized.

Essentially no change occurred in the physical appearance of the defect--no measurable dimensional changes in the rod, no significant fuel washout, and no evidence of increased hydride concentration in the cladding were observed after irradiation in the FERTF. An observable effect of irradiation was a grey discoloration of the cladding downstream from the defect. The discoloration was caused by the evolution of material and/or gases from the hole.

The alpha and beta-gamma autoradiographs* (Figure 12) indicate that the symmetry of the fuel structure and fission product distribution patterns were affected by the presence of the defect.

The alpha and beta-gamma emitter concentration distribution patterns, as indicated by the autoradiographs of the defected specimen, (Figure 12) do not coincide except in the once-molten fuel region. A source of high fission product activity at the fuel periphery (dark spot on the beta-gamma autoradiograph and white spot on the alpha autoradiograph) is apparently an unexplained enriched UO₂ particle. Examination of the fuel structure formed during irradiation at the plane of the defect indicates a greater

*The alpha-autoradiographs⁽¹⁴⁾ described in this report were made by exposing the irradiated fuel specimens to cellulose acetate, which, after etching in sodium hydroxide, indicated the presence of plutonium or alpha emitting isotopes associated with the presence of plutonium. Thus, the alpha-autoradiographs provide a semiquantitative indication of plutonium concentration. The dark areas in the autoradiograph correspond to regions of high activity. Alpha energy analysis of microdrilled samples from irradiated UO₂-PuO₂ fuel specimens showed that ~32% of the total alpha activity was caused by alpha energies normally associated with Cm²⁴². The remainder of the alpha activity was from plutonium isotopes and Am²⁴¹, although there was no significant amount of Am²⁴¹ present in the samples. Curium²⁴² is associated with the plutonium originally included as enrichment material, plus that formed from U²³⁸ (uranium decay chains modified by neutron reactions do not result in significant formation of curium). Curium²⁴² is formed by neutron capture Am²⁴² formed by neutron capture in Am²⁴¹, a daughter product via beta decay of Pu²⁴¹.

degree of melting than anticipated from the calculated maximum rod power generation.

Although the appearance of the fuel structure in an adjacent nondefected rod differed somewhat from the fuel structure in the defected rod at the same plane, approximately the same amount of fuel melting (50 versus 55% of the radius) was indicated (Figure 13). Less porosity associated with the once-molten fuel region was observed in the nondefected rod, and the radioactivity distribution patterns (particularly as indicated on the alpha-autoradiographs) were different for the nondefected and defected rods. The alpha-autoradiograph of the defected specimen (Figure 12) indicates segregation of alpha emitters from the once-molten fuel region and migration into other areas of the specimen, while the nondefected UO_2 - PuO_2 fuel rod (Figure 13) shows no evidence of such segregation or migration. This suggests a difference in the irradiation behavior, i.e., plutonium segregation and migration, between defected and nondefected mixed-oxide fuel rods.

An intentionally defected PRTR fuel rod containing pneumatically impacted UO_2 -4 wt% PuO_2 fuel ruptured while operating in the FERTF at a power generation of 885 W/cm (27 kW/ft) with significant fuel melting at the plane of the defect.⁽¹⁵⁾ A piece of cladding ~7.6 cm (3 in.) long and almost 1.29 cm (0.5 in.) wide at the widest point was missing from the area of the rod containing the defect. Approximately 700 g of fuel material (39% of the total) were released from the rod because of the rupture, and the Zircaloy

β - γ Auto-radiograph

α Auto-radiograph

Photo-macrograph

Neg 0653144-2

FIGURE 13. Transverse Section Through a Nondefected Vibrationally Compacted PRTR Fuel Rod Containing Pneumatically Impacted UO_2 -2 wt% PuO_2 Fuel. Section is at the Same Plane and Operated Under the Same Conditions as the Defected Rod Shown in Figure 12. Melting is Indicated to ~50% of the Radius. (4X)

compacted $\text{UO}_2\text{-PuO}_2$ fuel rods experiencing fuel melting during operation. This would suggest that the balance pressure tube was penetrated by a 1.2 cm (0.5 in.) diam hole adjacent to the rupture in the fuel rod. Postirradiation examination indicated that it was primarily molten fuel that was lost from the rod. The molten fuel would have been quickly and forcibly ejected from the rod under the pressure differential created between the inside of the rod and the decreased FERTF coolant pressure when the pressure tube failed.

The precise mechanism by which failure occurred is not known, but postirradiation examination indicates that the ruptured rod operated at a significantly higher fuel temperature than did adjacent nondefected rods operating at the same power generation. The unexpected large amount of fuel melting in the intentionally defected rod could have been caused by a decrease in the effective thermal conductivity of the fuel, a decrease in the melting temperature of the fuel, a slightly higher power generation due to the venting of fission gas poisons, or a combination of these possible effects. Variations in the fuel-to-jacket thermal contact would also affect the amount of fuel melting.

Analysis of $\text{UO}_2\text{-PuO}_2$ fuel samples from the vicinity of the rupture shows that the O:U ratio increased from 2.01 prior to irradiation to ~ 2.1 after irradiation. This and other very recent data⁽¹⁶⁾ indicate that in the presence of water vapor, UO_{2+x} might exist at higher temperatures than previously considered possible, and that its formation can be very rapid. Increasing the O:U ratio from 2.00 to 2.15 has been reported to decrease the effective thermal conductivity of

UO_2 by as much as 40%.^(17,18) A reduction in the melting temperature of the fuel would further increase the melt radius. It has been reported that increasing the O:U ratio from 2.00 to 2.15 decreases the melting temperature of UO_2 by as much as 400 °C.⁽¹⁹⁾

The large amount of fuel melting in the defected rod may have been the underlying cause of the rupture. Nevertheless, the cause of the rupture is more directly related to the defect in the rod than to operation with molten vibrationally compacted, pneumatically impacted mixed-oxide fuel *per se*.

PRTR experience has shown that the diameter of the central void formed at and above columnar grain growth temperatures in vibrationally compacted, pneumatically impacted $\text{UO}_2\text{-PuO}_2$ fuel rods is not necessarily related to the power generation or the degree of fuel melting in a long fuel rod. So far, no evidence of gross axial fuel relocation exists in the testing program (up to a maximum rod power of 885 W/cm or 27 kW/ft), as determined by rod balance point measurements, although fuel structures formed during irradiation suggest axial movement. As expected, examination of transverse sections from above the midplane of the rods shows that the area of the central void is nearly equal to the initial free volume of the sintered fuel region. However, the size of the central void below the midplane is generally smaller than the initial free volume of the sintered region, thus indicating an apparent mass imbalance. The apparent mass imbalance is not completely understood, but the examination results indicate that axial fuel relocation occurs in vibrationally compacted $\text{UO}_2\text{-PuO}_2$ fuel rods experiencing fuel melting during operations. This would suggest that the balance

point technique for determining axial fuel relocation is insensitive, or that the difficulties experienced in making such measurements in remote facilities preclude obtaining the required accuracy.

FISSION GAS RELEASE

Studies of pneumatically impacted fuel indicate that this method of fuel preparation results in fission gas release values equal to or somewhat less than those resulting from other methods of preparation. Some of the fission gas release data obtained

from the EBWR capsule and rod irradiation testing program are shown in Table I and Figure 14. The trend of increased gas release with increased rod power is apparent. The data include burnups from 0.24 to 3.69×10^{20} fissions/cm³ (1010 to 15,400 MWd/tonne), bulk fuel densities in the range of 77 to 85% theoretical, and fuel enrichment of 1.5 wt% PuO₂ in UO₂. The points aligned vertically above a power generation of ~ 500 W/cm (~ 15 kW/ft) illustrate the effect of fuel bulk density on fission gas release; the lower points have the

Neg 0661473-4

FIGURE 14. Fission Gas Release as a Function of Rod Power for Irradiated, Vibrationally Compacted Capsule Specimens Containing Pneumatically Impacted UO₂-1.5 wt% PuO₂. Maximum Burnup is 3.69×10^{20} Fissions/cm³ (15,400 MWd/tonne of UO₂-PuO₂).

highest bulk fuel density. Lower gas release in the higher density fuel could be the result of lower fuel temperatures caused by a higher effective thermal conductivity. The specimens that operated above 1000 W/cm (~ 30 kW/ft) all underwent fuel-pellet or fuel-pellet-spring interactions, which undoubtedly affected fission gas release values.

The amount of fission gas release has been determined for full-sized vibrationally compacted PRTR fuel rods containing pneumatically impacted UO_2 - PuO_2 fuel. A plot of percent Xe+Kr release versus volumetric average fuel rod temperature ($\bar{T}_v$)* is given in Figure 15. The average fuel element burnup for the points plotted in Figure 15 ranged from 0.025 to 1.3×10^{20} fissions/cm³ (100 to 5000 MWd/tonne). The results show a maximum of 88% Xe+Kr release from fuel rods that operated with volumetric

**Volumetric average fuel temperatures are based on calculated temperature profiles within the fuel using an estimated thermal conductivity of packed particle UO_2 as a function of temperature and the thermal conductivity of UO_2 at 2800 °C as the thermal conductivity of molten fuel. The flux depression within UO_2 - PuO_2 fuel rods is calculated by using the THERMOS computer code. The volumetric average fuel temperature over the entire volume of a fuel rod is defined as:*

$$\bar{T}_v = \frac{T_v}{\phi / \bar{\phi}}$$

where:

$\bar{T}_v$ = Volumetric average fuel temperature within the fuel rod

T_v = Calculated volume weighted average fuel temperature at the maximum flux position

ϕ = Peak neutron flux

$\bar{\phi}$ = Average neutron flux along the length of the fuel rod.

average fuel temperatures of ~ 2200 °C resulting from a maximum rod power of 885 W/cm (27 kW/ft). Different rods from the same element sampled in either the fuel or plenum regions yielded approximately the same quantity of gas, which thus indicated good communication along the entire length of the fuel rods and between plenum and fuel.

FISSION PRODUCT MIGRATION

Beta-gamma autoradiographs of irradiated pneumatically impacted mixed-oxide fuel specimens show that fission product migration commences at fuel temperatures sufficient to cause in-reactor sintering and grain growth. These observations are not unique with pneumatically impacted mixed-oxide fuel because the same phenomena occur during irradiation of UO_2 ⁽²⁰⁾ and other ceramic fuel systems such as MgO - PuO_2 ,⁽²¹⁾ ZrO_2 - PuO_2 ,⁽²²⁾ and ThO_2 - PuO_2 .⁽²³⁾ Recently developed alpha-sensitive autoradiographic techniques⁽¹²⁾ have shown that, under certain operating conditions (i.e., molten core defect), alpha emitter concentrations vary in irradiated mixed-oxide fuel specimens. The variance indicates plutonium segregation from the liquid and migration in the solid. The migration of fission products and plutonium during irradiation at high fuel temperatures could have a profound effect upon burnup analyses, reactivity, and fuel behavior.

Irradiated pneumatically impacted UO_2 - PuO_2 fuel specimens have been analyzed to evaluate fission product migration and plutonium segregation.

Neg 0661473-6

FIGURE 15. Fission Gas Release as a Function of Volumetric Average Fuel Rod Temperature in 167 cm Long Vibrationally Compacted PRTR Fuel Rods Containing Pneumatically Impacted UO_2 - PuO_2 Fuel.

Chips collected from 0.5 mm diam holes drilled into the surface of the irradiated fuel specimens⁽²⁴⁾ are radiochemically analyzed for selected fission products. Analytical results obtained from a pneumatically impacted UO_2 -1.5 wt% PuO_2 fuel specimen (Figure 3) are summarized in Figure 16. Little or no change in the radial fission product distribution were indicated under these moderate operating conditions, and no in-reactor sintering or fission product migration was observed on the beta-gamma autoradiograph.

However, at and above fuel temperatures sufficient to cause in-reactor sintering and grain growth, certain fission products migrate. Figure 17 summarizes radiochemical analytical results for the UO_2 -1.5 wt% PuO_2 specimen shown in Figure 5. This specimen operated at a maximum rod power of 1013 W/cm (31 kW/ft) with fuel melting to ~45% of the radius. Radial fission product migration is indicated on the beta-gamma autoradiograph. The radiochemical results show that Zr-Nb⁹⁵, Sr⁹⁰, and Ce-Pr¹⁴⁴ migrate the least; whereas, Cs¹³⁷ and Ru¹⁰⁶ migrate the most; the Ru¹⁰⁶ concentration varies by more than an order of magnitude across the radius of the sample. The radiochemical results also show that plutonium has not segregated radially. The rapid increase in Ru¹⁰⁶ concentration coincides with the dark band on the beta-gamma autoradiograph at the inner edge of the small columnar grain growth region. This region also corresponds to a well defined area characterized by a high concentration of metallic inclusions (Figure 11).

These inclusions have been identified as metallic beta-uranium in UO_2 ^(20,25) and are probably associated with a high concentration of fission products. Similar appearing inclusions have also been identified as fission-product (molybdenum, ruthenium) oxides.⁽²⁶⁾

Activity distribution patterns indicated by beta-gamma and alpha autoradiographs of some irradiated UO_2 - PuO_2 fuel specimens do not coincide. As shown in the beta-gamma and alpha

Neg 0653160-2

FIGURE 16. Fission Product and Plutonium Distribution in the Irradiated Pneumatically Impacted UO_2 -1.5 wt% PuO_2 Fuel Specimen (GEH-14-714) Shown in Figure 3. Specimen Operated at a Maximum Rod Power of 327 W/cm (10 kW/ft) to a Burnup of 0.51×10^{20} Fissions/cm³ (2220 MWd/tonne).

Neg 0653160-3

FIGURE 17. Fission Product and Plutonium Distribution in Irradiated Pneumatically Impacted UO_2 -1.5 wt% PuO_2 Fuel Specimen (GEH-14-706) Shown in Figure 5. Specimen Operated at a Maximum Rod Power of 1013 W/cm (31 kW/ft) to a burnup of 0.55 $\times 10^{20}$ Fissions/cm³ (2260 MWd/tonne).

sensitive autoradiographs of an intentionally defected specimen (Figure 12), the only area that coincides is the once-molten fuel region near the center of the specimen. Other features of both autoradiographs correspond with structural changes in the irradiated specimen illustrated in the photomacrograph.

The alpha autoradiograph shown in Figure 12 suggests segregation of alpha-emitters from the liquid phase and migra-

tion in the solid during irradiation. This effect has not been observed in every pneumatically impacted mixed-oxide specimen irradiated under molten fuel conditions, as illustrated by the autoradiographs of a nondefected UO_2 - PuO_2 specimen (Figure 13). Because possible plutonium segregation effects have been more clearly indicated by alpha-autoradiographs of defected fuel specimens, a difference (plutonium segregation and migration) in the irradiation behavior between defected and nondefected high performance fuel rods is suggested.

The pneumatically impacted UO_2 -2 wt% PuO_2 PRTR fuel rod specimen shown in Figure 12 was drilled and radiochemically analyzed at selected locations, and the analytical results are summarized in Figure 18. The length of the lines represent the diameter of the drilled samples. It is estimated that this intentionally defected specimen operated at a rod power of ~ 790 W/cm (~ 24 kW/ft) with fuel melting to $\sim 55\%$ of the radius. Again, the results show that Zr-Nb^{95} , Ce-Pr^{144} , and Sr^{90} migrate the least. Cesium¹³⁷ and Ru¹⁰⁶ migrate the most; the Ru¹⁰⁶ concentration varies by over an order of magnitude across the radius of the sample. The sharp increase in Ru¹⁰⁶ concentration at a radius of ~ 0.4 cm corresponds to the region of high activity on the beta-gamma autoradiograph at the inner edge of the small columnar grain growth region. Preliminary analytical results indicate that the plutonium concentration does not vary appreciably across the radius of the irradiated fuel specimen. There is no analytical evidence of plutonium segregation from the liquid or migration in the solid

FIGURE 18. Fission Product and Plutonium Distribution in the Intentionally Defected Pneumatically Impacted UO_2 -2 wt% PuO_2 PRTR Fuel Rod Shown in Figure 12. Specimen Operated at a Maximum Rod Power of $\sim 790 \text{ W/cm}$ (24 kW/ft).

phase as suggested by the alpha-autoradiograph. The slight decrease in plutonium concentration near the periphery of the specimen is possibly caused by nonuniform radial burnup effects; the burnup is highest on the outer surface of the fuel rod. Little significance is attached to the abnormally low plutonium concentration indicated by the one sample closest to the center of the specimen.

DISCUSSION

Pneumatic impaction is a versatile process for producing high density particles of mechanically mixed UO_2 - PuO_2 fuel material suitable for swage-or vibrational-compaction. The PuO_2 concentration and UO_2 and PuO_2 particle sizes can be varied over a wide range to make pneumatically impacted fuel material suitable for either thermal or fast reactor applications. Engineering proof tests, conducted with capsule irradiations in MTR/ETR and irradiations of full-scale fuel elements under power reactor conditions in PRTR, have demonstrated the satisfactory behavior of the fuel.

Fuel structures formed during irradiation of pneumatically impacted UO_2 - PuO_2 are similar to those formed during the irradiation of UO_2 fuels. Irradiation history is an important consideration in the analysis of fuel structures formed during irradiation. Time-temperature dependent diffusion phenomena can erase structural characteristics formed during initial high power generation operation of UO_2 - PuO_2 fuels after extended exposure at a reduced power generation. In-reactor

homogenization and solid solution formation occur relatively rapidly in mechanically mixed, pneumatically impacted UO_2 - PuO_2 fuel material at the higher irradiation temperatures. The high degree of consolidation of the UO_2 and the small (-325 mesh) PuO_2 particles enhance the homogenization process. However, under all normal operating conditions, an unsintered region composed of discrete mechanically mixed UO_2 - PuO_2 particles will persist at the outer periphery of the fuel rods, even though fuel melting may have occurred in the central region.

UO_2 - PuO_2 fuel material prepared by the present pneumatic impaction process is made slightly hyperstoichiometric to aid the densification process. Oxygen-to-metal ratios range from 2.01 to 2.03, with an average of ~ 2.02 . This excess oxygen could affect the irradiation behavior of the fuel by influencing the effective thermal conductivity. It has been reported that UO_2 fuels become hypostoichiometric during irradiation at temperatures in excess of 1500 °C.⁽²⁷⁾ It has also been postulated that UO_2 fuel becomes heterogeneous during irradiation, varying from hyperstoichiometric in the cooler peripheral regions to hypostoichiometric near the center. A decrease in effective thermal conductivity has been reported with increasing O:U ratios above 2.00.^(17,18) If UO_2 - PuO_2 fuels behave similarly to the postulated manner for UO_2 , the excess oxygen in pneumatically impacted material could affect fuel temperatures during irradiation.

The irradiation behavior of intentionally defected vibrationally compacted $\text{UO}_2\text{-PuO}_2$ fuel rods operating under high performance conditions does not seem to be completely consistent. One defected rod behaved satisfactorily while operating under molten core conditions; whereas, another molten core defect test ruptured. A direct comparison of the irradiation results of these two experiments may be invalidated by the fact that the irradiation conditions were not the same. For instance, the rod that behaved satisfactorily did not have a fission gas plenum and operated at a maximum power generation of $\sim 790 \text{ W/cm}$ (24 kW/ft). The ruptured rod contained a fission gas plenum; it operated at a maximum power generation of 885 W/cm (27 kW/ft); and the coolant temperature was cycled during irradiation.

Fuel temperatures in the defected rod that behaved satisfactorily were approximately as expected; whereas, indicated fuel temperatures were unexpectedly high in the ruptured rod. The high fuel temperatures, believed to be the underlying cause of failure in the ruptured rod, are attributed to oxidation of the fuel by the water coolant and the consequent reduction in effective thermal conductivity and/or a reduction in the melting temperature. This behavior is not believed to be unique with pneumatically impacted fuel, but the reason for the apparent difference in the oxidation behavior of the fuel in the two experiments is unknown.

The limited amount of fission gas release data for pneumatically impacted $\text{UO}_2\text{-PuO}_2$ fuel indicate that this type of material exhibits release values

equal to or somewhat less than those resulting from other types of fuel. The data illustrate the trend for increased gas release with increasing rod power or volumetric average fuel rod temperature. The fission gas release data presented in Figure 15 is from $\text{UO}_2\text{-PuO}_2$ PRTR fuel rods with relatively low (100 to 5000 MWd/tonne) burnup. It is expected that the fission gas release values will increase with increasing burnup, particularly for the low fractional release values. The high fractional release values should not change appreciably with irradiation time because the rate of gas release from the high temperature fuel is more rapid.

Fission gas release values can be calculated with reasonable accuracy if it is assumed that 100% of the gas is released from the fuel volume operating above 1800°C (columnar grain growth region) and if 10% is released from the fuel operating below 1800°C (Figure 19). These data were calculated* from the capsule irradiation tests, and they agree well with the gas release values obtained with the long fuel rods irradiated in PRTR.

Comparison of the gas release data obtained from pneumatically impacted $\text{UO}_2\text{-PuO}_2$ fuel with that derived in earlier work using sintered and coprecipitated materials⁽²⁸⁾ suggests possible lower fission gas release values for pneumatically impacted material. However, the comparison, is difficult because of differences in specimen

*Calculations were performed by G. Testa (CNEN) during temporary assignment at PNL.

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FIGURE 19. Fission Gas Release as a Function of the Cross-Sectional Area Above 1800 °C for Vibrationally Compacted Capsules Containing Pneumatically Impacted UO_2 -1.5 wt% PuO_2 Fuel.

characteristics, particularly density, and more definitive data are needed before meaningful conclusions can be drawn.

In a carefully controlled experiment, Carroll and Sisman⁽²⁹⁾ of Oak Ridge National Laboratory compared the fission gas release characteristics of pneumatically impacted UO_2 with those of arc-fused single crystal UO_2 . A considerable difference exists in these two gas release rates: the single crystal UO_2 release rate at 1000 °C is an order of magnitude greater and increases with temperature faster than the release rate from the pneumatically impacted material. Carroll and Sisman⁽²⁹⁾ postulate "that grain boundaries do not act

as paths of rapid escape, as once believed, but rather as traps which anchor migrating fission gas." Pneumatically impacted fuel material being polycrystalline could explain its lower gas release at moderate temperatures. However, at temperatures resulting in sintering and grain growth, the gas release characteristics of the two fuel materials should be the same.

During irradiation at the higher operating temperatures, fission products migrate to preferred locations within pneumatically impacted UO_2 - PuO_2 fuel rods. Analytical results show that the migration behavior of the fission products Zr-Nb^{95} , Sr^{90} , Ce-Pr^{144} , Cs^{137} , and Ru^{106} is similar for UO_2 and pneumatically impacted UO_2 - PuO_2 fuels. This similarity is also demonstrated in the beta-gamma autoradiographs of the two types of fuel.

Alpha-sensitive autoradiographs of pneumatically impacted UO_2 - PuO_2 irradiated with a degree of fuel melting have, in some cases, shown evidence of alpha emitter or plutonium segregation from the liquid phase and migration in the solid. This phenomenon has been particularly evident in transverse sections from high performance fuel rods operating with defected cladding, and suggests a difference in the irradiation behavior between defected and nondefected fuel rods. Evaluation of the alpha-autoradiographs indicates a uniform radioactivity density in the once-molten fuel region surrounded by a narrow, well defined, depleted or enriched band. The surrounding nonmolten region exhibits a higher alpha-emitter

concentration. Radiochemical analyses for plutonium concentration have not confirmed any evidence of radial migration in the solid or segregation from liquid UO_2 - PuO_2 fuel during irradiation. This apparent discrepancy between chemical analysis and autoradiography needs further clarification.

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APPENDIX

UO_2 -1.5 wt% PuO_2 fuel rods⁽¹¹⁾ were fabricated by Pacific Northwest Laboratory for irradiation in the EBWR. An EBWR fuel rod and the fuel element assembly are shown in Figure A-1. Some of the EBWR fuel rods are being irra-

diated by Pacific Northwest Laboratory in two types of experimental fuel test assemblies. One type, a 21-rod bundle, is shown in Figures A-2, -3, and -4. The second type, a three-rod cluster, is shown in Figure A-5.

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FIGURE A-1. EBWR UO_2 -1.5 wt% PuO_2 Fuel Rod and Fuel Element Basket. Each Fuel Rod in the 6 x 6 Array is 1.08 cm (0.424 in.) OD and 148 cm (58.3 in.) Long.

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FIGURE A-2. Fuel Element 6501, a 21 Rod Bundle, Assembled and Ready for Charging into the PRTR. The Prototype EBWR Fuel Rods Contain Vibrationally Compacted, Impacted UO_2 -1.5 wt% PuO_2 Fuel. The Bundle Diameter is 8.1 cm (3.2 in.) and the Length is 164 cm (64.5 in.).

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FIGURE A-3. Close-up View of Lower End of the 21 Rod PRTR Bundle Containing EBWR Fuel Rods. Stainless Steel-Clad, Al-Co Alloy, Flux-monitoring Wires are Installed in the Two Round Protrusions.

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FIGURE A-4. Close-up View of Upper End of the 21 Rod PRTR Bundle Containing EBWR Fuel Rods.

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FIGURE A-5. Twelve, Three Rod Clusters Prepared for Irradiation Testing. Clusters Contain 148 cm (58.3 in.) Long Prototype EBWR Fuel Rods and have Stainless Steel Fittings and Spacers.

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