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CERAMICS RESEARCH AND DEVELOPMENT
OPERATION

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INTRODUCTION

This report is the second of a series to be issued by the recently organized Ceramics Research and Development Operation, Reactor and Fuels Laboratory, Hanford Laboratories. The Ceramics Research and Development Component (E. A. Evans, Manager) comprises the following groups:

Ceramics Research	D. R. deHalas, Manager
Advanced Fuel Development	K. Drumheller, Manager
Fabrication Development	R. D. Widrig, Manager
Fuels Testing and Analysis	W. E. Roake, Manager
Special Projects	L. E. Mills, Manager

The work reported in this and subsequent reports is a continuation of reactor fuels studies reported in the first report issued by the Ceramics Research and Development Operation and in previous reports issued by the Fuels Development and the Plutonium Metallurgy Operations. Previous reports in those series are:

<u>Ceramics Research and Development Operation</u>	<u>Date</u>
HW-76300 (Unclassified)	October - December, 1962
<u>Fuels Development</u>	<u>Plutonium Metallurgy</u>
HW-72347 (Secret)	HW-73318 (Unclassified)
HW-74377 (Secret)	HW-74162 (Secret)
HW-74378 (Secret)	HW-74718 (Secret)
HW-72346 (Secret)	HW-72161 (Secret)
	January - March, 1962
	April - June, 1962
	July - September, 1962
	October - December, 1962

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X-ray powder pattern studies have indicated that the region between $\text{PuO}_{1.0}$ and $\text{PuO}_{1.5}$ in the Pu-O phase diagram is a two-phase field of PuO and $\beta\text{-Pu}_2\text{O}_3$.

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An increase in the lattice parameters of several oxide and carbide compounds of plutonium has been observed during the course of standing at room temperature. The rates of expansion range from 0.085×10^{-3} to 1.16×10^{-3} ($\Delta a/a_0$) per 100 days.

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Samples of PuC, PuN, and PuO₂ will be irradiated at a power density of about 900 watts/gram and a surface heat flux of over 3×10^6 Btu/(hr)(sq ft).

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- Particle Size Distribution for Vibrational Compaction - J. O. McPartland and J. J. Hauth. 3.22
 Particle size distribution for vibrational compaction of a 1-3/8-inch OD x 1/4-inch ID annular fuel capsule was calculated from the mathematical model developed by Applied Mathematics. This distribution gave a 93% theoretical density.
- PRTR UO_2 - PuO_2 Swaged Fuel Fabrication - C. H. Bloomster and R. E. Bardsley 3.22
 A pilot fabrication run of 50 swaged PRTR fuel clusters was essentially completed.
- EBWR Plutonium Fuel Loading - R. E. Sharp. 3.28
 Design of the vibration compaction facility for the EBWR plutonium fuel loading was approved and construction will begin in April, 1963.
- Powder Fuel Loading Studies - R. E. Bardsley 3.28
 A uniform physical mixture of UO_2 - PuO_2 in a fuel rod can be achieved by physically mixing the core material for each rod in an individual container.
- Nondestructive Test for Plutonium Distribution - R. E. Bardsley and S. T. Field 3.30
 A gamma-scanning, nondestructive test was developed to obtain both qualitative and quantitative data on the distribution of plutonium in fuel rods.
- Coextruded Al-Pu Fuel Cut to Short Lengths - L. C. Lemon. . 3.31
 Fuel elements free of external contamination were made from a long coextruded rod of Al-Pu cut into short pieces using machining techniques developed to eliminate contamination at the cut ends.
- Special Fuel Element Fabrication - R. E. Bardsley, C. H. Bloomster, and L. C. Lemon 3.32
 Six UO_2 thermocouple capsules, 18 eight-foot UO_2 - PuO_2 pellet rods, and 550 three-foot Al-Pu rods were made for various special tests.

FUELS TESTING AND ANALYSIS

- Inspection of PRTR Fuel Elements - M. D. Freshley
and W. J. Flaherty 4.1
- The general appearance of eight carefully inspected
irradiated PRTR fuel elements was good and no serious
cladding defects were observed.
- Postirradiation Examination of a Swage-Compacted
UO₂-PuO₂ PRTR Fuel Element - M. D. Freshley. 4.3
- Local high fuel temperatures in a swaged, incrementally
loaded, UO₂-PuO₂ PRTR fuel rod were sufficient to cause
center void formation and columnar grain growth.
- Postirradiation Examination of a Vibrationally Compacted,
UO₂-PuO₂ PRTR Fuel Element - M. D. Freshley. 4.7
- Only minor fuel washout and contamination release
occurred following the failure of a vibrationally
compacted, UO₂-PuO₂ fuel element in the PRTR. The
cause of the failure has not been determined.
- Examination of PRTR Nested Tubular Fuel Element No. 1501 -
W. J. Flaherty 4.9
- Preliminary gas release data from PRTR tubular fuel
element (No. 1501) indicate that less than 10% of the
fission gases and less than 50% of the sorbed gases were
released during irradiation.
- Extended Surface, PRTR Fuel Element Wear Pads -
M. D. Freshley, W. J. Flaherty, and R. E. Sharp 4.10
- Process tube wear was reduced by increasing the con-
tact surface area of irradiated PRTR fuel elements.
- Irradiation Testing Program for EBWR Prototypic Fuel Rods -
W. J. Bailey 4.11
- Irradiation tests of prototypic, vibrationally compacted,
EBWR, UO₂-PuO₂ fuel rods were initiated.
- Irradiation Performance of MgO-PuO₂ and ZrO₂-PuO₂ Fuels -
M. D. Freshley and D. F. Carroll 4.11
- Migration of fission products and/or PuO₂ occurs during
irradiation at high fuel temperatures in both MgO-PuO₂
and ZrO₂-PuO₂ fuel pellets.
- Irradiation of Uranium-Plutonium Oxide - W. J. Bailey and
T. D. Chikalla 4.17
- Autoradiographs showing marked plutonium and/or fission
product migration aided microsampling of irradiated
UO₂-PuO₂ pellets. A six-fold difference in specific gamma
activity was found in samples removed from the same pellet.

- Irradiation of Pneumatically Impacted $\text{UO}_2\text{-PuO}_2$ -
 G. R. Horn 4.25
 Capsules containing pneumatically impacted $\text{UO}_2\text{-PuO}_2$
 (0.505 inch diameter fuel) were irradiated at a surface heat
 flux of approximately 900,000 Btu/(hr)(sq ft).
- Irradiation of UO_2 Single Crystal Pellets - G. R. Horn. . . . 4.25
 The thermal conductivity of large grain (single and bi-
 crystal) UO_2 was apparently greater than that of small
 grain UO_2 during irradiation.
- Irradiation of $\text{UO}_2\text{-W}$ Cermets - G. R. Horn and
 W. J. Flaherty 4.25
 $\text{UO}_2\text{-W}$ cermet were chemically and dimensionally stable
 during irradiation for 2 hours with a surface temperature
 greater than 2100 C.
- Phoenix Fuel Experiment - M. D. Freshley 4.26
 The gross reactivity of an Al-Pu fueled test capsule made
 of high exposure (27.17% Pu^{240}) plutonium was 38% greater
 than that of a similar capsule incorporating low exposure
 (6.25% Pu^{240}) plutonium after irradiation to 1.2×10^{20}
 fissions/cc (92% burnup of original fissionable atoms).
- $\text{ThO}_2\text{-PuO}_2$ Studies - M. D. Freshley and H. M. Mattys . . . 4.26
 $\text{ThO}_2\text{-PuO}_2$ irradiation test pellets (95-96% TD) containing
 2 - 5 wt% PuO_2 were made by dry pressing calcined
 powder mixtures and sintering in hydrogen at 1600 C for
 6 hours.
- Low Temperature Sintering of UO_2 - W. J. Flaherty. 4.27
 No bonding of fuel particles was observed in cold-swaged
 UO_2 heated out-of-reactor for more than 1000 hours at
 550 C confirming the hypothesis that sintering observed
 in PRTR fuel rods is an irradiation effect.
- Short Duration Irradiations of UO_2 and $\text{UO}_2\text{-PuO}_2$ -
 W. J. Bailey and T. D. Chikalla 4.27
 Flux monitor measurements show that the four capsules
 reported in the previous Quarterly Report generated rod
 powers between 18.3 and 23.7 kw/ft. Problems associated
 with postirradiation examination of low exposure plutonium
 fuels are discussed.

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 A pilot fabrication run of 50 swaged PRTR fuel clusters was essentially completed.
- EBWR Plutonium Fuel Loading - R. E. Sharp. 3.28
 Design of the vibration compaction facility for the EBWR plutonium fuel loading was approved and construction will begin in April, 1963.
- Powder Fuel Loading Studies - R. E. Bardsley 3.28
 A uniform physical mixture of UO_2 - PuO_2 in a fuel rod can be achieved by physically mixing the core material for each rod in an individual container.
- Nondestructive Test for Plutonium Distribution - R. E. Bardsley and S. T. Field 3.30
 A gamma-scanning, nondestructive test was developed to obtain both qualitative and quantitative data on the distribution of plutonium in fuel rods.
- Coextruded Al-Pu Fuel Cut to Short Lengths - L. C. Lemon. . . 3.31
 Fuel elements free of external contamination were made from a long coextruded rod of Al-Pu cut into short pieces using machining techniques developed to eliminate contamination at the cut ends.
- Special Fuel Element Fabrication - R. E. Bardsley, C. H. Bloomster, and L. C. Lemon 3.32
 Six UO_2 thermocouple capsules, 18 eight-foot UO_2 - PuO_2 pellet rods, and 550 three-foot Al-Pu rods were made for various special tests.

FUELS TESTING AND ANALYSIS

- Inspection of PRTR Fuel Elements - M. D. Freshley
and W. J. Flaherty 4.1
- The general appearance of eight carefully inspected
irradiated PRTR fuel elements was good and no serious
cladding defects were observed.
- Postirradiation Examination of a Swage-Compacted
UO₂-PuO₂ PRTR Fuel Element - M. D. Freshley. 4.3
- Local high fuel temperatures in a swaged, incrementally
loaded, UO₂-PuO₂ PRTR fuel rod were sufficient to cause
center void formation and columnar grain growth.
- Postirradiation Examination of a Vibrationally Compacted,
UO₂-PuO₂ PRTR Fuel Element - M. D. Freshley. 4.7
- Only minor fuel washout and contamination release
occurred following the failure of a vibrationally
compacted, UO₂-PuO₂ fuel element in the PRTR. The
cause of the failure has not been determined.
- Examination of PRTR Nested Tubular Fuel Element No. 1501 -
W. J. Flaherty 4.9
- Preliminary gas release data from PRTR tubular fuel
element (No. 1501) indicate that less than 10% of the
fission gases and less than 50% of the sorbed gases were
released during irradiation.
- Extended Surface, PRTR Fuel Element Wear Pads -
M. D. Freshley, W. J. Flaherty, and R. E. Sharp 4.10
- Process tube wear was reduced by increasing the con-
tact surface area of irradiated PRTR fuel elements.
- Irradiation Testing Program for EBWR Prototypic Fuel Rods -
W. J. Bailey 4.11
- Irradiation tests of prototypic, vibrationally compacted,
EBWR, UO₂-PuO₂ fuel rods were initiated.
- Irradiation Performance of MgO-PuO₂ and ZrO₂-PuO₂ Fuels -
M. D. Freshley and D. F. Carroll 4.11
- Migration of fission products and/or PuO₂ occurs during
irradiation at high fuel temperatures in both MgO-PuO₂
and ZrO₂-PuO₂ fuel pellets.
- Irradiation of Uranium-Plutonium Oxide - W. J. Bailey and
T. D. Chikalla 4.17
- Autoradiographs showing marked plutonium and/or fission
product migration aided microsampling of irradiated
UO₂-PuO₂ pellets. A six-fold difference in specific gamma
activity was found in samples removed from the same pellet.

- Irradiation of Pneumatically Impacted UO_2 - PuO_2 -
 G. R. Horn 4.25
 Capsules containing pneumatically impacted UO_2 - PuO_2
 (0.505 inch diameter fuel) were irradiated at a surface heat
 flux of approximately 900,000 Btu/(hr)(sq ft).
- Irradiation of UO_2 Single Crystal Pellets - G. R. Horn. . . . 4.25
 The thermal conductivity of large grain (single and bi-
 crystal) UO_2 was apparently greater than that of small
 grain UO_2 during irradiation.
- Irradiation of UO_2 -W Cermets - G. R. Horn and
 W. J. Flaherty 4.25
 UO_2 -W cermets were chemically and dimensionally stable
 during irradiation for 2 hours with a surface temperature
 greater than 2100 C.
- Phoenix Fuel Experiment - M. D. Freshley 4.26
 The gross reactivity of an Al-Pu fueled test capsule made
 of high exposure (27.17% Pu^{240}) plutonium was 38% greater
 than that of a similar capsule incorporating low exposure
 (6.25% Pu^{240}) plutonium after irradiation to 1.2×10^{20}
 fissions/cc (92% burnup of original fissionable atoms).
- ThO_2 - PuO_2 Studies - M. D. Freshley and H. M. Mattys . . . 4.26
 ThO_2 - PuO_2 irradiation test pellets (95-96% TD) containing
 2 - 5 wt% PuO_2 were made by dry pressing calcined
 powder mixtures and sintering in hydrogen at 1600 C for
 6 hours.
- Low Temperature Sintering of UO_2 - W. J. Flaherty. 4.27
 No bonding of fuel particles was observed in cold-swaged
 UO_2 heated out-of-reactor for more than 1000 hours at
 550 C confirming the hypothesis that sintering observed
 in PRTR fuel rods is an irradiation effect.
- Short Duration Irradiations of UO_2 and UO_2 - PuO_2 -
 W. J. Bailey and T. D. Chikalla 4.27
 Flux monitor measurements show that the four capsules
 reported in the previous Quarterly Report generated rod
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CERAMICS RESEARCH

The Plutonium-Carbon System - R. E. Skavdahl and J. B. Burnham

Analysis of X-ray powder pattern films of a 48.2 at.% carbon alloy indicate the possible growing in of the zeta phase of the Pu-C system while standing at room temperature for a period of 400 days.

The 48.2 at.% carbon alloy was prepared by arc-melting plutonium metal and graphite powder. A small portion of the arc-melted button was broken off, powdered, and placed in a double sealed glass capillary for X-ray analysis. A film was run immediately after arc-melting and capillary preparation (December 11, 1961) and showed a major phase of PuC ($a_o = 4.971 \pm 0.001A$) and a small amount of Pu_2C_3 . As part of a study on the lattice expansion of plutonium compounds due to self-damage by alpha decay, the same capillary was run at various time intervals through February 26, 1963. The last four films are shown in Figure 2.1 and are listed in Table 2.1.

TABLE 2.1
POWDER PATTERNS OF 48.2 at.% CARBON ALLOY

<u>Film Number</u>	<u>Date Run</u>	<u>Phases Observed</u>	<u>Lattice Parameter for PuC Phase</u>
851	April 20, 1962	PuC, Pu_2C_3	$4.974 \pm 0.001A$
1058	October 15, 1962	PuC, Pu_2C_3 , Zeta (?)	$4.976 \pm 0.001A$
1183	January 7, 1963	PuC, Pu_2C_3 (?), Zeta (?)	$4.986 \pm 0.002A$
1272	February 26, 1963	PuC, Pu_2C_3 , Pu-oxides (?)	$4.985 \pm 0.002A$

Assuming that it is zeta-phase that is growing-in, the films show that the rate is such that even after 4 months following initial synthesis, there was not enough present to be detected by X-ray diffraction. However, in the relatively short space of time between October 1962 and January 1963, about 3 months, the amount had increased significantly. On the most recent Film 1272, zeta phase had disappeared.

April 20, 1962
Film 851

October 15, 1962
Film 1058

January 7, 1963
Film 1183

February 26, 1963
Film 1272

FIGURE 2.1

X- Ray Powder Pattern Films of Arc-Melted Pu-48.2 at.% Carbon Alloy

The data which support the assumption that it is zeta phase are three-fold. First, the d-spacings which would correspond to zeta-phase on the films agree very well with the d-spacing observed on samples in which metallography has definitely shown the presence of zeta-phase. Second, these same zeta d-spacings are increasing at a rate quite consistent with the rate of increase of Pu-C d-spacings due to radiation self-damage. Third, the increase in the lattice parameter of PuC during the 3 month interval between Films 1058 and 1183 is much greater than could be accounted for by self-damage. This large increase is to be expected if zeta-phase is being formed, since zeta-phase (nominally Pu_3C_2) would require the PuC phase to give up some of its plutonium atoms, thus enriching the carbon content of the remaining PuC, and hence increasing its lattice parameter.

A listing of the d-spacings of zeta-phase is given in Table 2.2. The data from Film 1183 were run on the IBM 709/7090 computer in an attempt to index zeta on a tetragonal, hexagonal, or orthorhombic basis. The computer results are shown in Table 2.3. The indexing programs were supplied by J. B. Goebel of Applied Mathematics Operation.

TABLE 2.2
ZETA-PHASE d-SPACINGS

Film No. 1085		Film No. 1183					
d, A	Intensity	d, A	Intensity	d, A	Intensity	d, A	Intensity
7.462	M	9.448	VVW	1.950	W	0.9338	VW
6.992	M	8.706	VVW	1.900	MW	0.9216	VW
4.435	MS	7.830	VW	1.883	MW	0.8965	VW
4.315	MS	7.338	S	1.841	MW	0.8602	VW
4.110	MS	7.138	S	1.781	VW	0.7991	VW
3.542	MS	4.853	W	1.733	VW		
3.085	MS	4.488	S	1.687	W		
2.743	W	4.370	S	1.667	M		
2.672	MW	4.175	S	1.622	MW		
2.324	MW	3.870	W	1.587	W		
2.292	W	3.751	MW	1.542	W		
1.897	MW	3.613	MS	1.490	MS		
1.620	MW	3.564	S	1.458	VW		
1.486	W	3.391	W	1.393	VW		
1.233	W	3.178	W	1.366	W		
1.202	W	3.075	MW	1.341	W		
1.103	W	2.793	W	1.307	W		
1.083	W	2.690	MW	1.287	MW		
0.9105	W	2.566	M	1.265	W		
		2.346	MS	1.236	W		
		2.295	M	1.204	VW		
		2.219	W	1.086	VW		
		2.078	W	0.9741	VW		
		1.993	M	0.9452	VW		

TABLE 2.3
COMPUTER ANALYSIS OF ZETA-PHASE d-SPACINGS

<u>Structure</u>	<u>Lattice Parameters</u>	<u>Standard Deviation of $(\sin \theta)^2$</u>
Hexagonal	$a_o = 26.791 \text{ \AA}$ $c_o = 18.768$	0.000121
Tetragonal	$a_o = 22.485$ $c_o = 18.967$	0.000109
Orthorhombic	$a_o = 9.81$ $b_o = 21.75$ $c_o = 7.92$	0.00094

Although zeta-phase should not appear in an alloy of 48.2 at. % carbon, it is possible that either (1) the phase diagram for Pu-C is in error on this point, or, more likely, (2) the arc-melted button was not homogeneous and the portion taken for the capillary had a carbon content compatible with the presence of zeta phase.

The growing-in of zeta phase could be the result of a slow establishment of equilibrium in the system and its disappearance could be the result of its selective susceptibility to the presence of a small amount of oxygen which has diffused into the capillary. The latter point was suggested by Mulford.⁽¹⁾

Electrical Resistivity Measurements of PuC Compounds - C. A. Hinman

The electrical resistivity of two compounds within the PuC system was measured as a function of temperature and time to study solid state phase transformations. The measurements were made using a four-probe direct current method on machined, arc-cast rods of 37.2 and 39.1 at. % carbon. The electrical resistivity of the samples was measured during vacuum heat treatment below 630 C. Absolute accuracy of the measured resistivity values

1. R. R. R. Mulford. Los Alamos Scientific Laboratory. April 4, 1963.
(Private Communication)

was calculated to be $\pm 8 \mu$ ohm cm and the experimental deviational error was observed to be $\pm 2 \mu$ ohm cm. The error in temperature sensing and control was estimated to be ± 2 C.

Figure 2.2 shows the electrical resistivities of the two samples as a function of temperature after vacuum annealing at 500 C for 2 days and furnace cooling. Both compositions showed a reproducible decrease in their electrical resistivities between 500 and 550 C after declining only slightly up to this temperature. Electrical resistivity versus time at various constant temperatures between 500 and 550 C are shown in Figure 2.3.

FIGURE 2.2

Electrical Resistance of PuC as a Function of Temperature

FIGURE 2.3

Change of Electrical Resistance of PuC with Time

The abrupt decrease in resistivity starting at approximately 500 C is due to interphase reactions in the samples. The exact nature of these reactions have not yet been determined. The resistivity versus time curves indicate that the solid state reactions are completed in a relatively short period of time.

Plutonium Nitride - D. F. Carroll

Plutonium nitride decomposes during arc melting under one atmosphere of nitrogen in the following manner:

The PuN, formed by nitriding plutonium hydride, was arc melted under one atmosphere of nitrogen on a water-cooled copper hearth. Metallographic and

X-ray diffraction data show that approximately 95 vol% of this arc-melted material consists of PuN dendrites in a matrix of alpha plutonium (Figure 2.4). The small bright spots are probably particles of tungsten; occasionally, during melting, the tungsten electrode will touch and contaminate the melt. Figure 2.5 shows the solid PuN surface layer formed during the cooling of the melt.

FIGURE 2.4

Arc-Melted PuN (center of button)

As Polished
150 X

α -Pu

As Polished
500 X

PuN

FIGURE 2.5

Arc-Melted PuN (edge of button)

R-898); R-899

AEC-GE RICHLAND, WASH.

Determination of Carbon in Plutonium Carbide - H. J. Anderson and
R. E. Skavdahl

In cooperation with Chemical Laboratory, Hanford Laboratories, a combustion gravimetric method for determination of carbon in plutonium carbides was reduced to routine practice in glove boxes. The method was found to be better than 1% precise for 1 to 5 wt% carbon content in specimens weighing 200 to 1000 mg.

PuN-UN Phase Studies - D. F. Carroll

X-ray diffraction analyses of helium sintered PuN-UN pellets show complete solubility between the components. Random deviations from Vegard's Law, i. e., linear variance of lattice parameter with composition, were observed at several compositions and are believed to be caused by specimen oxidation. The pure sintered PuN exhibited a 0.25% lattice expansion, perhaps resulting from an oxygen addition to the lattice or a nitrogen loss, or both. A trace of PuO_2 was present as a second phase.

Pure UN sintered under identical conditions also showed a lattice expansion, 0.23%, but in contrast to the PuN, there was considerable conversion to the oxide. A strong UO_2 phase, approximately 25 vol%, was detected in the sintered UN material by X-ray diffraction. Evidently, under these conditions, one atmosphere of flowing helium at 1600 C and some small partial pressure of oxygen, UN is less stable than PuN.

The Plutonium-Oxygen System - R. E. Skavdahl

Several samples of plutonium sub-oxides in the range of $\text{O}/\text{Pu} = 1.0$ to 1.5 , produced by carbon reduction of PuO_2 , have shown that the phase diagram in that region is probably represented by a two-phase field of PuO and $\beta\text{-Pu}_2\text{O}_3$. Accordingly, Figure 2.6 is a revised version of the previously published⁽¹⁾ plutonium-oxygen phase diagram. The melting point of 1825 C for PuO and the eutectic temperature of 1750 C and eutectic composition of $\text{PuO}_{1.25}$ are only estimates at this time.

1. T. D. Chikalla, C. E. McNeilly and R. E. Skavdahl. The Plutonium-Oxygen System, HW-74802. September, 1962.

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AEC GE RICHLAND WASH

FIGURE 2. 6

Revised Plutonium-Oxygen Phase Diagram

The indicated congruent melting of PuO is also assumed, but is supported by X-ray powder patterns of samples that had inadvertently melted during synthesis.

PuO₂-Carbon Reactions - R. E. Skavdahl

Experiments involving the synthesis of plutonium sub-oxides by carbon reduction of PuO₂ have indicated that the path leading to the formation of PuO is:

Several samples which were intended to have compositions in the range of PuO_{0.9} to PuO_{1.2} after completion of reacting, proceeded only to 75-85% completion after reaction in flowing helium at 1450 ± 50 C for 16 hours. Table 2.4 summarizes a portion of the data for those samples.

TABLE 2.4

PuO₂-CARBON REACTION SAMPLES

<u>Sample No.</u>	<u>Initial wt% Carbon</u>	<u>Desired Product</u>	<u>Final wt% Carbon</u>	<u>Phases Present as Determined by X-ray Analysis</u>
1-52	3.42	PuO _{1.2}	0.42	-Pu ₂ O ₃ , PuO/PuC
1-53	3.84	PuO _{1.1}	0.82	-Pu ₂ O ₃ , PuO/PuC
1-54	4.24	PuO _{1.0}	1.00	-Pu ₂ O ₃ , Pu ₂ C ₃ , PuO/PuC
1-55	4.65	PuO _{0.9}	1.12	-Pu ₂ O ₃ , Pu ₂ C ₃ , PuO/PuC

The fcc phase which has been listed as PuO/PuC was observed to be about 30-50% of the total sample composition, as estimated from the X-ray powder pattern films. Even assuming all the carbon to be contained in the fcc phase, the residual carbon contents of the samples cannot account for that phase being 100% PuC. It is therefore proposed that it is a solid solution of PuO and PuC.

The lattice parameters of the cubic phases for the above samples are listed in Table 2.5.

TABLE 2.5
RESULTS OF X-RAY ANALYSES

<u>Sample No.</u>	<u>Lattice Parameter ($\pm 0.001\text{\AA}$)</u>	
	<u>PuO/PuC</u>	<u>Pu₂C₃</u>
1-52	4.968	- -
1-53	4.968	- -
1-54	4.968	8.132
1-55	4.968	8.134

Preliminary evidence suggests that the reduction reaction proceeds better in the liquid state than in the solid state. Several pellets of PuO₂-C mixtures which were reacted at 1675 ± 50 C for 8 hours in flowing helium exhibited a black outer crust and an inner core with a shiny, metallic appearance. This inner core, which apparently melted, had a higher residual carbon content than the outer crust but also showed a greater degree of reduction. There was probably a higher partial pressure of CO in the confined inner core than in the outer crust. This could result in a slower rate of release of CO and a higher residual carbon content. The latter could cause a lowering of the solidus temperature of the center portion of the pellets to below the operating temperature, thereby causing the shiny, metallic appearance.

Radiation Self-Damage of Plutonium Compounds - R. E. Skavdahl and
J. B. Burnham

An increase in the lattice parameters of several plutonium compounds has been observed during the course of standing at room temperature in double-sealed, glass X-ray capillaries. It is believed that these increases are due mainly to the radiation self-damage caused by alpha decay of the plutonium atoms. Some of the observations are tabulated in Tables 2.6 and 2.7.

TABLE 2.6INCREASE OF LATTICE PARAMETER FOR ARC-MELTED ALLOYS

<u>Species</u>	<u>Composition (at. % C)</u>	<u>$\Delta a/a_0$ (10^{-3})</u>	<u>Total Time (days)</u>	<u>$\Delta a/a_0/100$ days (10^{-3})</u>
PuC	29.1	2.40	508	0.47
PuC	33.0	1.13	274	0.41
PuC	35.4	2.72	559	0.49
PuC	48.2	1.33	276	0.49
PuC	48.2	1.51	273	0.55
PuC	46.7	0.40	180	0.22
Pu ₂ C ₃	62.0	0.09	115	0.085

TABLE 2.7

INCREASE OF LATTICE PARAMETER
FOR COMPOUNDS PREPARED BY CARBON REDUCTION OF PuO₂

<u>Species</u>	<u>Composition</u>	<u>$\Delta a/a_0$ (10^{-3})</u>	<u>Total Time (days)</u>	<u>$\Delta a/a_0/100$ days (10^{-3})</u>
PuC	50 at. % C	1.01	133	0.76
PuO	50 at. % O	1.01	135	0.75
β -Pu ₂ O ₃	60 at. % O	0.84 (a-axis)	120	0.70
		1.39 (c-axis)	120	1.16

A sample of solid-solution PuO₂-10 at. % UO₂ exhibited a lattice sion of $1.02 \times 10^{-3} \Delta a/a_0$ after 233 days of self-damage, which corresponds to a rate of $0.44 \times 10^{-3} \Delta a/a_0/100$ days.

High Burn-up Irradiations of Plutonium Compounds - J. B. Burnham
and J. L. Bates

Samples of PuC, PuN, and PuO₂ will be irradiated at a power density of about 900 watts/gram and a surface heat flux of over 3×10^6 Btu/(hr)(ft²). In order to irradiate specimens at these levels, without causing excessive temperatures, it is necessary to use thin samples and an extended heat

transfer surface. Plutonium carbide specimens, $1/4 \times 1/4 \times 1/16$ inch, are prepared by cutting and grinding arc-cast buttons. These are double canned in aluminum capsules as shown in Figure 2.7. The inner capsule includes a "split die" section which is accurately machined to fit the individual samples to assure very good contact between the specimen and aluminum under operating conditions. The inner capsule after being evacuated and welded in helium is shrunk-fit into an outer can. The surface heat flux at the outer can is slightly over 100,000 Btu(hr)(ft²), resulting in a maximum aluminum temperature of less than 200 C. The first capsule containing PuC is awaiting irradiation in the Materials Testing Reactor (MTR).

FIGURE 2.7

High Burn-up Capsule Showing "Split-Die" Assembly, Hollow End Spacers, and Double Can (Specimen is black wafer between aluminum spacers in one quadrant of die assembly)

0630708-2
REC'D WICHMAN, WASH.

Preparation of High Density Plutonium Dioxide - H. J. Anderson,

L. C. Lemon and R. J. Anicetti

High density (> 99% TD) PuO_2 was produced by arc melting pressed pellets under one atmosphere of argon followed by oxidation in air at 800 C for 8 hours. Pellets were initially pressed without binder to a density of 6.9 g/cm^3 (mensuration). The arc-melted material had a density of 10.91 g/cm^3 (by displacement) (Figure 2.8). Based upon the percent weight gain after oxidation, it is calculated that the as-formed arc-melted material has an O/Pu ratio of 1.77, i. e., contains approximately 1- to 1-mole ratio of $\alpha\text{-Pu}_2\text{O}_3$ and $\text{PuO}_{1.98}$.⁽¹⁾ X-ray diffraction studies are now in progress.

A PuO_2 -2.5 wt% UO_2 pellet was also arc-melted to a density of 10.89 g/cm^3 .

FIGURE 2.8

Arc-Melted Plutonium Dioxide
with an O/Pu Ratio of 1.77
($\alpha\text{-Pu}_2\text{O}_3$ and $\text{PuO}_{1.98}$) and a Density
of 10.91 g/cm^3

-
1. T. D. Chikalla, C. E. McNeilly and R. E. Skavdahl. The Plutonium - Oxygen System, HW-74802. September, 1962.

PuO₂-MgO Phase Studies - D. F. Carroll

PuO₂-MgO mixtures appear to form a simple binary eutectic system. It must be noted, however, that during the melting experiments PuO₂ and MgO both lost oxygen, and there was some reaction with the tungsten ribbon on which the specimens were heated.

Although not detected by preliminary melting point determinations, high temperature reactions between PuO₂ and MgO under one atmosphere of static helium are evident from metallographic examinations.

Figure 2.9 shows white MgO dendrites dispersed in the PuO_{2-x} phase. The photomicrographs are of melted specimens quenched at a rate of nearly 20,000 C/min. The structure shown in Figure 2.10 suggests a eutectic structure. Although melting point studies gave no indication of a eutectic reaction, it is possible for such a reaction to escape detection. Reactions between ceramic components at temperatures greater than 2000 C are exceedingly difficult to detect visually.

FIGURE 2.9
Melted PuO₂ -22 mole % MgO Mixture
(As Polished)
150 X

FIGURE 2.10

Melted PuO_2 -43 mole % MgO Mixture
(As Polished)
250X

Figure 2.11 shows what are thought to be plutonium oxide dendrites in a matrix of MgO , a tungsten reaction area, and a large crack in the tungsten ribbon. The frequent tungsten ribbon failures at high temperatures were due to reactions with the PuO_2 - MgO mixtures. X-ray diffraction analyses of melted specimens showed traces of WO_2 , WO_3 , and MgWO_4 .

FIGURE 2.11

Melted PuO_2 -96 mole % MgO Mixture
(As Polished)
150 X

Preparation and Properties of UOS - H. J. Anderson, F. A. Scott
and J. L. Bates

A method was developed for preparation of dense uranium oxysulfide (UOS) powder by precipitation from a fused chloride melt at low temperatures (550 C). US_2 and UOS- US_2 mixtures have also been prepared by the proper adjustment of the precipitation conditions.

The UOS powder was heated under an argon atmosphere in a tungsten wedge. During initial heating and at low temperatures the odor of sulfur was detected. Upon melting (mp 1900 ± 50 C) the UOS was

decomposed. The solidified sample was made up of three phases (Figure 2.12). Etching with 5 normal HCl removed the white phase (probably US_2) located at the grain boundaries (Figure 2.13a). Further etching with 30% H_2O_2 -10% H_2SO_4 solution attacked the light grey matrix phase, presumably the original UOS (Figure 2.13b). The unaffected dark grey phase appears to be UO_2 . Preliminary X-ray diffraction data support the identity of these phases.

FIGURE 2.12

Microstructure of UOS After Melting
Sample was Held in Molten State for 15 Minutes at 1930 C
(As Polished)
250 X

4K613F; 4K613D
AEC-GE RICHLAND, WASH.

(a) After Etching for 15 Seconds in 5N HCl

(b) After Etching for 30 Seconds in 90% H_2O_2 -10% H_2SO_4

FIGURE 2.13

Microstructure of UOS After Melting
Sample was Held in Molten State for 15 Minutes at 1930 C
250 X

4K6131I; 4K613H; 4K613M; 4K613K

AEC-GE RICHLAND, WASH.

During heating between 1900 to 2200 C, whiskers (presumably UO_2) grew from cooler UOS surfaces. These whiskers were 0.005 to 0.010 cm in diameter and 0.5 to 1.5 cm long (Figure 2.14).

FIGURE 2.14

Whiskers (Presumably UO_2) Grown During Heating of UOS
in a Tungsten Ribbon Furnace Under an Argon Atmosphere (250 X)

Uranium Monosulfide - J. L. Bates

Significant changes in the physical properties of US were observed after a low neutron exposure. A sintered $\text{US}_{0.98}$ sample* (approximately 1 gram) was irradiated twice to a cumulative exposure of approximately 1.5×10^{16} fissions/cc. A 0.09% dilation of the lattice parameter resulted from the first irradiation (7×10^{15} fissions/cc), measurement of lattice parameter after the second irradiation has not been completed. A large increase in microhardness (75%) resulted from the first irradiation with 22% further increase during the second irradiation. (Table 2.8) No detectable changes were observed in the melting point or microstructure after irradiation.

* The US used in these studies was obtained from the Ceramics Research Group at Argonne National Laboratory.

TABLE 2.8PROPERTY CHANGES IN URANIUM MONOSULFIDE DURING IRRADIATION

	Neutron Exposure (fissions/cc)		
	0	7×10^{15}	1.5×10^{16}
Lattice Constant (A)	5.4897 ± 0.0002	5.4947 ± 0.002	- -
Knoop Hardness Number	$197 \begin{smallmatrix} + 24 \\ - 35 \end{smallmatrix}$	$341 \begin{smallmatrix} + 40 \\ - 38 \end{smallmatrix}$	$386 \begin{smallmatrix} + 32 \\ - 36 \end{smallmatrix}$
Melting Point (C)	2390 ± 30	2390 ± 30	- -

Thermal Reactions of UO_2 -W Cermet - J. L. Daniel

Time-lapse cinemicrography has disclosed further details of thermal reactions occurring during heating of impacted UO_2 -50 wt% tungsten nonsintered cermets in the reflection electron microscope. Peripheral crystallite growth on the UO_2 surface began at 800 C, extending rapidly to the entire UO_2 surface with increasing temperature (Figure 2.15). Tungsten recrystallization occurred above 1200 C, but UO_2 -W interaction was not evident.

Electron Microscopy of UO_2 -W Cermet Autoradiographic Replicas -

N. D. Dockum

UO_2 -W cermet surfaces have been replicated with an alpha sensitive nuclear emulsion for transmission electron microscopy. Shadowed replica preparations reveal surface structure details at the point of origin of the alpha tracks, and in surrounding areas. Figure 2.16 shows a micrograph obtained by this technique. The method is applicable to polished metallographic specimens containing alpha emitters within a nonradioactive matrix.

Before
Reaction

780 C

Above 1200 C

FIGURE 2.15

Thermal Reaction of UO_2 -50 wt% Tungsten Cermet
(Cinemicrographs by reflection electron microscopy) 800 X

FIGURE 2.16

Transmission Electron Micrograph
of Shadowed Autoradiograph from UO_2 -W Cermet

Oxidation of UN-W Cermets - J. L. Daniel and J. O. McPartland

Oxidation of impacted UN-50 wt% tungsten cermets was studied by continuous observation during reaction in the reflection electron microscope. A controlled low-pressure oxygen jet impinging on the sample surface produced rapid oxidation of the UN at temperatures near 1000 C (Figure 2.17). Slower tungsten oxidation occurred simultaneously, but UN-W interaction was not evident, and the specimens remained coherent during the reaction. Severe thermal etching occurred in the absence of oxygen.

Before Reaction

W

UN

W

After
In-Microscope
OxidationFIGURE 2.17

Oxidation of UN-50 wt% Tungsten Cermet
(Reflection electron microscopy)

1500 X

Thinning of UO₂ Crystals for Electron Microscopy - N. L. Dockum
and J. L. Daniel

Sections of UO₂ crystals suitable for electron microscopy experiments were produced by chemical thinning in an apparatus based on a design by Amelinckx at Mol⁽¹⁾ (Figure 2.18). Additional thinning occurred in the electron beam during examination, particularly in cleavage platelets displaying hexagonal breakthrough holes characteristic of (111) crystal planes.

Basic Materials for Research - H. J. Anderson

Standardized uranium ceramic specimens were prepared for distribution as indicated below.

<u>Recipient</u>	<u>Specimens</u>
Japan Atomic Energy Research Institute TOKAI Laboratory	Twenty UO ₂ single crystals (Figure 2.19)
Pratt and Whitney CANEL	Impacted UN and UN-W cermet
University of California LASL	Electrodeposited UO ₂ single crystals
Union Carbide ORNL	Impacted UN
General Electric Company NMPO	Impacted UO ₂ -W cermet

1. H. B. Kirkpatrick and S. Amelinckx. "Device for Chemically Thinning Crystals for Transmission Electron Microscopy," Review of Scientific Instruments, vol. 33, pp. 488-9. 1962.

(a) Thinning Apparatus

Neg. No. J1475-6
(25,000 X)(b) Typical Thin Section from Cleavage Platelet
and Electron Diffraction PatternFIGURE 2.18
Chemical Thinning of UO_2 Crystals

0630407-1
AECGE RICHLAND, WASH.

FIGURE 2.19

UO₂ Single Crystals Prepared for Distribution to Tokai Laboratory, Japan

2.28

HW-76301

UO₂ Material Exchange - H. J. Anderson, M. K. Millhollen and
L. P. Murphy

Exchange of UO₂ materials continued with other sites for evaluation and development of new information. Upon request from Savannah River Laboratory for high purity material for testing in Heavy Water Components Test Reactor (HWCTR), a 50 pound lot of -12 +20 mesh arc-fused UO₂ was treated at high temperature in moist hydrogen to reduce impurity content, particularly nitride impurities. Preliminary analyses indicated a high quality product (Table 2.9).

TABLE 2.9
ANALYSIS OF HIGH PURITY UO₂

Density	> 99% theoretical (10.95 g/cm ³ , He displacement) (10.93 g/cm ³ , Hg displacement)
O/U atom ratio	2.000 ⁷
Surface Area	0.05 m ² /g
Nitrogen	< 100 ppm
H ₂ O	5 ppm
Carbon	50 ppm
Fluoride	5 ppm (estimated)
Chloride	20 ppm (estimated)
Spectrographic Analysis	Fe 30 ppm, Si 80 ppm

Free Metal in UO₂ - H. J. Anderson

A new technique was used to reveal nearly 0.1% free uranium metal in UO₂ previously shown by coulometric titration techniques to be hyperstoichiometric (O/U = 2.0039). The technique involved the monometric measurement of the volume of hydrogen gas released during HCl dissolution and oxidation of free metal to an equilibrium condition between the fourth and fifth valence states. The new method revealed 0.092% free metals; a gravimetric oxidation method yielded a value of 0.084% metal.

This new technique was used to analyze several types of UO_2 . Pneumatically impacted UO_2 (micronized) and electrodeposited UO_2 were free of metal. Monometric analysis of a special single crystal of UO_2 ($\text{O}/\text{U} = 1.93$) confirmed free uranium metal; after treatment in wet hydrogen at 1750 C for 16 hours, the crystal contained no free metal as revealed either by this technique or by gravimetric oxidation and coulometric titration techniques.

Thermal Conductivity of UO_2 - J. A. Christensen

Measurements of the thermal conductivity of single crystal UO_2 relative to polycrystal UO_2 are being performed at temperatures to 2300 C using a radial heat flow technique. Preliminary results indicate a substantial increase in single crystal conductivity above 1600 C which appears to arise from radiant energy transfer.

The method uses a stack of 1/4-inch OD x 1/16-inch thick wall, hollow cylinders of UO_2 , internally heated by a 1/10-inch diameter tungsten resistance filament (Figure 2.20). To prevent an IR drop across the specimens, they are insulated from the heater wire by quartz spacers at both ends of the stack of pellets. Each stack of pellets contains a 1/2-inch long single crystal specimen, a 1/2-inch long polycrystal specimen, four 1/4-inch long single crystal end shields, and four 1/4-inch long polycrystal end shields arranged as shown in Figure 2.20. All specimens had an O/U ratio of $2.000^{+0.001}_{-0.000}$, achieved by annealing 12 hours at 1700 C in moist H_2 . Regularly spaced radial slots in the end shields permit monitoring the heater wire temperature during an experiment. Specimen surface temperatures are measured with a brightness pyrometer and with a total radiation pyrometer. The thermal conductivity of the single crystal, relative to the polycrystal (assuming both are opaque to infra-red radiation) is readily obtained from the surface temperatures of the two specimens when their inner surfaces see the same heater temperature. Absolute values of thermal conductivity may also be calculated assuming the spectral emissivity data for UO_2 in the literature is applicable.

FIGURE 2. 20

Radial Flow Thermal Conductivity Apparatus

Preliminary data are given in Figure 2.21. The curve shown for polycrystal UO_2 was determined by Daniel¹, et al.,⁽¹⁾ and represents the most accurate data available. Extrapolation of Daniel's curve, assuming phonon conduction only, yields a constant conductivity of approximately $0.018 \text{ watts cm}^{-1}\text{C}^{-1}$ above 1500 C, the temperature at which the phonon mean free path approximates the lattice parameter. In the present work, temperature measurements using a brightness pyrometer showed that the ratio of the apparent conductivity of single crystals to that of polycrystal UO_2 was 2.0 at 850 C, 1.2 at 1600 C, and 1.8 at 2000 C. Since all specimens had the same composition, the difference in conductivity probably reflects a lower optical density for single crystals, permitting them to more efficiently transmit heat by radiation.

Radiation heat transfer is suggested also by the difference between Daniel's single crystal data (Figure 2.21) and the new radial flow data. Daniel's experiments involved series heat flow through a specimen bounded on both ends by opaque, high reflectivity surfaces, thus relatively little of the radiant flux through the specimen was lost. In the radial flow experiments, no such boundaries were present, and radiation through the specimen was much less inhibited. If significant radiant transfer does occur, the radial heat flow results in Figure 2.21 for the single crystals may be too low since the transmitted energy does not heat the specimen surface and thus passes through undetected. This effect is being investigated by using specimens plated with thin, opaque coatings of tungsten obtained by thermal decomposition of WF_6 . Total radiation pyrometer measurements, now underway, should also provide absolute measurements of heat flux. These may differ appreciably from those calculated from brightness pyrometer (or thermocouple) measurements of the specimen surface if a radiation heat transfer component exists.

1. J. L. Daniel, J. Matolich, Jr, and H. W. Deem. Thermal Conductivity of UO_2 , HW-69945. 1962.

FIGURE 2.21

Thermal Conductivity of UO_2 Single Crystals Relative to Sintered Polycrystals

Thermal Diffusivity Measurements - J. O. McPartland and R. E. Skavdahl

Thermal diffusivity of nuclear ceramics will be determined by a pulsed energy method in a recently assembled apparatus. The equipment (Figure 2.22) is designed to permit measurements at temperatures up to 2500 C.

The energy pulse from a laser is directed to one face of a thin cylindrical specimen. The resultant transient temperature rise on the opposite face is detected by a photovoltaic device. The thermal diffusivity is then computed from the relative temperature versus time plot displayed on a cathode ray oscilloscope.

FIGURE 2.22
Thermal Diffusivity Apparatus

Metallic Inclusions in Irradiated UO₂ - J. L. Bates

Metallic uranium inclusions are formed in UO₂ fuel elements irradiated with central temperatures high enough to produce extensive columnar grain growth. Examination of 15 fuel element sections after irradiation revealed, without exception, the presence of metallic inclusions in the UO₂ which had undergone grain growth. In high exposure fuel there is a tendency for the inclusions to concentrate in a band in the columnar grains (Figure 2.23). The formation of inclusions did not depend on the type of UO₂ and was observed in fuel elements originally containing sintered, swaged, and vibrationally compacted UO₂.

The minimum temperature necessary for forming the inclusions is estimated to be 1500 to 1600 C, this being the approximate temperature for equiaxed grain growth in UO₂ fuel rods. This is also the temperature reported for the formation of metallic uranium during laboratory experiments.^(1, 2, 3) The precipitation of uranium metal is believed to result from the presence of an oxygen-deficient phase above this temperature.

The presence of metallic uranium and the formation of an oxygen-deficient phase above 1600 C could significantly affect the physical properties of the UO₂. For example, there is evidence that the thermal conductivity is increased when metallic uranium is formed.⁽⁴⁾

A concentration of fission products in the region containing the most inclusions was revealed by autoradiographic techniques (Figure 2.24). The greater resistance to etching of the inclusions in the high exposure UO₂ also suggests that the fission fragments have concentrated in, and alloyed with, the metallic uranium.

-
1. J. S. Anderson, J. O. Sawyer, H. W. Werner, G. M. Willis, and M. J. Bannister. "Decomposition of Uranium Dioxide at its Melting Point," Nature. vol. 185, pp. 915-916. 1960.
 2. E. Rothwell. "High Temperature Substiochiometry in Uranium Dioxide," AERE-R-3897. 1961.
 3. R. G. Robins. "The Precipitation of Uranium Metal in Uranium Dioxide Single Crystals," J. Nucl. Mat. vol. 7, pp. 218-219. 1962.
 4. J. E. May, M. J. F. Notley, R. L. Stoute and J. A. L. Robertson. "Observations on the Thermal Conductivity of Uranium Oxide," AECL-1641. November, 1962.

As Polished
75 X

B-4187
B-4188

B-4176
B-4177

FIGURE 2.23

Concentration of Uranium Inclusions in Columnar Grains of Swaged
 UO_2 Fuel Element (GEH-14-237) Irradiated to $\sim 20,000$ Mwd/ton

AE EACH AND WASH

IMAGES ARE INITIAL TO THE

WASH EACH AND AE

FIGURE 2.24

Macrograph and Autoradiograph of Swaged, UO_2 Fuel Element (GEH-14-237)
Irradiated to $\sim 20,000$ Mwd/ton Showing Concentration of Fission Products and
Metallic Inclusions in Same Positions in Columnar Grains

The improved resolution of autoradiographs is obtained by placing the sample as close to the emulsion as possible. A one mil, aluminized plastic sheet is used to cover the plate to prevent exposure to light. The metallographically polished sample is placed directly on the plastic and a weight is used to force the sample surface as close to the emulsion as possible. The emulsion is exposed 15 seconds to 4 days, depending upon the radioactivity of the sample. No bleaching is required.

Release of Fission Products from UO_2 During Transient Heating -

J. L. Bates and J. A. Christensen

A lightly irradiated sample of polycrystalline UO_2 (approximately 1×10^{15} fissions/cc) retained 83 at% of the total fission products and 98.5% of total radioactivity during explosive melting.⁽¹⁾ This UO_2 bar was heated to the melting point in approximately 2 seconds by electrical resistance. A molten ball ejected during heating fell on a cold metal plate and was quenched. The radiochemical analysis of this ejected sample was compared to an as-irradiated portion of the same polycrystalline bar before heating. The radiochemical results are shown in Table 2.10.

1. J. J. Cadwell. Quarterly Progress Report, Fuels Development Operation, July, August, September, 1962. HW-74378, p. 5.1. October, 1962. (Secret)

TABLE 2.10

RADIOCHEMICAL ANALYSES OF UO_2 BEFORE
AND AFTER TRANSIENT MELTING*

Fission Isotope	Percent Released on Melting	Percent of Total Number Fission Product Atoms	
		Before	After
Ru^{103}	21	2.4	2.2
Ru^{106}	27	1.2	1.1
Cs^{137}	70	22.8	8.4
Zr-Nb^{95}	0	59.5	71.8
Sr (total)	0	8.9	10.7
Ce^{141}	4	5.1	5.9
I^{131}	66	0.3	0.1

- * 1. All results are decay corrected to sampling time and date.
 2. U_{wt} was determined by weighing samples before dissolution (88.1 wt% U).
 3. Ru^{103} , Ru^{106} , Sr(total), Co^{137} , Ce^{141} , and I^{131} were determined by chemical separations followed by counting. Zr-Nb^{95} was determined directly from aliquot of sample after dissolution.
 4. Total strontium was calculated as Sr^{89} , a trace of Ba^{140} may be present.

The retention or release of fission products appears to be related to the refractory nature of the fission fragments and their compounds.

UO_2 Microcores - H. J. Anderson

UO_2 microcores, approximately 0.015 inch OD and 0.085 inch long, were extracted from UO_2 specimens by using a core drill and ultrasonic machining techniques (Figure 2.26). Specimens were taken from UO_2 single crystals, high-energy-rate impacted UO_2 , and sintered UO_2 .

FIGURE 2.26

UO₂ Microcore Specimen Showing (a) Size and (b) Detail
Negative No. 0630475

These small cores allow precise sampling of fuel materials for fundamental studies such as those of fission product migration and analysis of impurity phases.

X-Ray Analyses - R. E. Skavdahl

A improved computer program in FORTRAN language for calculating the interplanar spacings of crystals from their line positions on X-ray powder pattern films was written and is operational. The input data are the position of the lines, as read on a film reader. The computer output is a listing, for each line, of the angle θ , $\sin \theta$, $\sin^2 \theta$, d-spacing, and the value of the Nelson-Riley extrapolation function.

UO₂ Relocation and Melting Experiment - G. R. Horn and J. A. Christensen

Continued examination of irradiated fuel pellets (GEH-4-72) that originally contained a spiral array of small tungsten "marker wires" confirmed the previously reported⁽¹⁾ hypothesis that the radial limit of

1. J. J. Cadwell, Quarterly Progress Report, Fuels Development Operation, July, August, September, 1962, HW-74378. October 1962. (Secret)

melting in irradiated ceramic fuels corresponds to the radial limit of observable subgrain structure. A longitudinal cross section through the bottom end of the capsule revealed tungsten in the lower regions of the initially molten zone. As shown in Figure 2.27, the tungsten wires sank in the molten UO_2 and came to rest at a position coincident with the limit of subgrain structure. Subgrain structure, not readily apparent in Figure 2.27, is easily seen in the original 75X photomosaic from which the figure was prepared.

Small, odd-shaped pieces of tungsten in the lower regions of the capsule suggest that the melting point of tungsten (3410 C) was reached during the short irradiation. The test capsule, initially containing 1.25 inch OD sintered UO_2 pellets, was irradiated for less than 1 minute generating $\sim 550,000 \text{ Btu}/(\text{hr})(\text{sq ft})$ maximum surface heat flux.

Thermal Conductivity of Molten UO_2 - G. R. Horn

The heat transfer rate through molten UO_2 was estimated to be 0.05 watts/cm C. This value is considerably less than the thermal conductivity of single crystal UO_2 near the melting temperature. The heat transfer rate was estimated from data obtained during postirradiation examination of a UO_2 specimen containing tungsten marker wires. The estimate is subject to considerable uncertainty and the estimated value could easily be in error by a factor of 2.

15X

FIGURE 2.27

Longitudinal Cross Section of Lower End of Tungsten
Marker Wire Experiment

Negative No. 0630317
AEC GE RICHLAND WASH

FUELS DEVELOPMENT

Pneumatic Impaction of UO_2 - D. W. Brite and K. R. Sump

Pneumatic impaction of UO_2 to 99% TD with punches of conventional design made of tool steel, rather than tungsten carbide, was accomplished. Eight different types of UO_2 , impacted under various conditions to densities greater than 98% TD, are listed in Table 3.1. Microstructures of some of these materials are shown in Figure 3.1. Stress measurements in the punches revealed that the upper stress limit possible without deforming the AISI-SAE class H-13 tool steel punches was 290,000 psi. Further improvements in the capabilities of tool steel punches are expected with the use of improved heat treating and forming methods, such as ausforming or maraging.

Improved mechanical strength and uniform microstructure were produced in UO_2 impacted from pulverized sintered scrap (approximately minus 48 mesh) by roasting the powder to an O/U ratio of 2.01 before impaction. Roasting at 130 C for 16 hours increased the O/U ratio of the smallest particles, without significantly affecting the larger particles. Particle size distribution and O/U ratios are shown in Table 3.2.

One hundred pounds of a different lot of sintered UO_2 scrap, pulverized to minus 20 mesh and roasted to $\text{UO}_{2.016}$, was impacted at 1200 C in cans containing 16 pounds of UO_2 to a density of 10.85 g/cc (98.9% TD) and a final O/U ratio of 2.010.

Heat loss during the 15 seconds required to transfer UO_2 impaction containers from the furnace to the Dynapak machine has no significant effect on the density of impacted UO_2 . Densities of random samples from two types of UO_2 impacted using a hot-wall transfer can were identical to those obtained without using the hot-wall transfer can (Table 3.1).

TABLE 3.1

EFFECT OF PNEUMATIC IMPACTION CONDITIONS ON UO_2 DENSITY

Impaction Experi- ment	Type UO_2	Disk Diameter, Inches	Hot-Wall Transfer Can Used?		Can Wall Thickness, Inches	Tempera- ture, °C	Impact Pressure, psi	Impacted Density,	
			Yes	No				g/cc	%TD
1	Sintered wafers	4.0		X	0.025	1250	Not measured	10.86	99.0
2	Sintered scrap, pulverized, roasted to $UO_{2.01}$	4.0		X	0.025	1200	Not measured	10.85	98.9
3	Micronized powder, $UO_{2.09}$ (from hydrogen reduced, calcined UO_3)	4.0		X	0.025	1200	Not measured	10.84	98.0
4	Ceramic grade powder, $UO_{2.06}$ (from hydrogen reduced ammonium diuranate)	4.0		X	0.025	1200	Not measured	10.89	98.8
5	Sintered wafers	2.5		X	0.025	1200	222,000	10.80	98.4
6	Sintered wafers	2.5	X		0.025	1200	218,000	10.80	98.4
7	Sintered scrap, pulverized to -20 mesh	2.5		X	0.025	1200	200,000	10.76	98.1
8	Sintered scrap, pulverized to -20 mesh	2.5	X		0.025	1200	205,000	10.73	97.8
9	Sintered scrap, pulverized to -48 mesh	2.5		X	0.025	1250	205,000	10.65	97.1
10	Sintered scrap, pulverized to -48 mesh	2.5		X	0.025	1250	215,000	10.68	97.4
11	Sintered scrap, pulverized to -48 mesh	2.5		X	0.025	1250	222,000	10.65	97.1
12	Sintered scrap, pulverized to -48 mesh	2.5		X	0.025	1250	254,000	10.79	98.4

3.2

HW-76301

TABLE 3.1 (contd.)

Impaction Experi- ment	Type UO ₂	Disk Diameter, Inches	Hot-Wall Transfer Can Used?		Can Wall Thickness, Inches	Tempera- ture °C	Impact Pressure, psi	Impacted Density,	
			Yes	No				g/cc	%TD
13	Sintered scrap, pulverized, roasted to UO _{2.01}	2.5		X	0.025	1250	218,000	10.77	98.2
14	Sintered scrap, pulverized, roasted to UO _{2.01}	2.5		X	0.025	1250	230,000	10.81	98.5
15	Sintered scrap, pulverized, roasted to UO _{2.01}	2.5		X	0.025	1250	243,000	10.80	98.5
16	Sintered scrap, pulverized, roasted to UO _{2.01}	2.5		X	0.025	1250	268,000	10.85	98.9
17	Fused UO ₂ , pulverized to -48 mesh	2.5		X	0.025	1250	234,000	10.63	96.9
18	Fused UO ₂ , pulverized, roasted to UO _{2.01}	2.5		X	0.025	1250	260,000	10.81	98.5
19	Impacted scrap, pulverized, roasted to UO _{2.01}	4.0		X	0.025	1200	241,000	10.84	98.8
20	Impacted scrap, pulverized, roasted to UO _{2.01}	4.0		X	0.035	1200	225,000	10.78	98.3
21	Impacted scrap, pulverized roasted to UO _{2.01}	4.0		X	0.063	1200	230,000	10.83	98.7

4K535B 100 X
Pulverized UO_2
Sintered Scrap

4K536B 100 X
Pulverized and
Roasted UO_2 Sintered
Scrap

4K531C 1000 X
Micronized UO_2

4K530C 1000 X
Ceramic Grade
 UO_2

4K599C 250 X
Pulverized, Fused
 UO_2

4K600C 250 X
Pulverized and
Roasted Fused UO_2

4J1522F 250 X
Sintered UO_2 Wafers
Before Impact

4K532F 250 X
Sintered UO_2
Wafers After
Impact

FIGURE 3.1

Microstructure of High-Energy-Rate Impacted UO_2 (Polished and Etched)

TABLE 3.2

O/U RATIOS AND PARTICLE SIZE DISTRIBUTION
OF PULVERIZED UO₂ SINTERED SCRAP
ROASTED 16 HOURS AT 130 C

<u>Particle Size</u>	<u>wt%</u>	<u>O/U Ratio</u>
Average Sample	100	2.003
-20+65 Mesh	32	2.001
-65+100 Mesh	14	2.004
-100 Mesh	54	2.013

A minimum can wall thickness of 0.050 inch for four-inch diameter, Type 310 stainless steel cans containing 17 pounds of UO₂, is sufficient to prevent can breakage during impactation at 1200 C and 230,000 psi. Can breakage occurs occasionally with thin-wall cans upon the high velocity recoil of the impact ram from the compacted can. The can wall thickness required was determined by impacting cans with walls 0.025, 0.035, 0.050, and 0.063 inches thick. An axial cross section of an impacted can containing 17 pounds of UO₂, with an initial wall thickness of 0.063 inches, is shown in Figure 3.2. Density of the UO₂ after impactation was 10.88 g/cc (99.2% TD).

FIGURE 3. 2

Pneumatically Impacted Can of 0.063-Inch Wall Thickness Containing 17 Pounds of UO_2

UO₂ Cermets - D. W. Brite, K. R. Sump and L. O. Slind

Solid cermet discs, approximately 1-inch thick and 4 inches in diameter, were fabricated by pneumatic impaction. The large size of the cermet disks allows the machining of specimens for testing of mechanical properties. Cermets fabricated in this size include UO₂ - 35 vol% niobium, UO₂ - 35 vol% chromium, UO₂ - 35 vol% molybdenum, UO₂ - 35 vol% stainless steel, and UO₂ - 50 wt% tungsten (Figures 3.3 and 3.4).

FIGURE 3.3

Pneumatically Impacted 4-Inch Discs of UO₂ Cermets

0630749-4

AEC GE RICHLAND WASH

FIGURE 3.4

Machined Surface of 4-Inch Diameter
UO₂- 50 wt% Tungsten Cermet

0630749-6

Pneumatically Impacted, Coated UO₂ - D. W. Brite and K. R. Sump

A UO₂-W cermet was formed by pneumatic impaction of tungsten coated -65+200 mesh UO₂ particles (Figure 3.5). Approximately 50% of the particles were coated initially. The cermet was impacted at relatively low pressure (115,000 psi) and, therefore, many voids existed between the particles. Higher pressure and a greater percentage of coated particles would result in a high density (99% TD) cermet having a continuous tungsten

matrix. Cermets fabricated by this technique allow the use of a minimum amount of metal while still retaining a continuous matrix.

4K529B

100 X

4K529C

500 X

FIGURE 3.5

Microstructure of Pneumatically Impacted
Tungsten-Coated UO_2 (As-Polished)

Uranium Nitride - Molybdenum Cermet - D. W. Brite and K. R. Sump

A UN-35 vol% molybdenum cermet was pneumatically impacted to a density of 12.49 g/cm^3 (97% TD) at 1250 C and 225,000 psi (Figure 3.6).

As-Polished

100 X

FIGURE 3.6

Microstructure of UN-35 vol% Molybdenum Cermet

Uranium Monosulfide Cermet - D. W. Brite, K. R. Sump and J. L. Bates

A US-50 wt% tungsten cermet was compacted by pneumatic impact at 1200 C (Figure 3.7). The tungsten matrix was continuous. The density of the cermet was 13.63 g/cm^3 (98.0% TD).

As-Polished

250X

FIGURE 3.7

Microstructure of US-50 wt% Tungsten Cermet

UO₂-PuO₂ Pneumatic Impaction Equipment - D. W. Brite and K. R. Sump

Tool size was increased to make possible the pneumatic impaction of 3.5 pounds of UO₂-PuO₂ at one time. Die diameter was increased from 1.75 to 2.59 inches.

Measurement of stresses in the main tie rods of the model 1200 Dynapak machine revealed that a maximum tie rod stress of 70,000 psi resulted when the impact pressure on the 2.56-inch diameter punch was 275,000 psi. This stress level was reached at an energy release of 80,000 ft-lbs. In the Model 1220B Dynapak machine, 50,000 ft-lbs produced a punch stress of 275,000 psi. The excess energy required using the Model 1200 machine is apparently expended in stretching the tie rods below their elastic limit. The additional 20,000 psi required to reach 275,000 psi with the model 1200 machine is apparently expended in elastic deformation of the smaller tie rods.

UO₂-PuO₂ Pneumatic Impaction - D. W. Brite and K. R. Sump

Uniform plutonium distribution was found in UO₂-2.5 wt% PuO₂ and in UO₂-1.3 wt% PuO₂ formed by pneumatic impaction at 145,000 psi and 1200 C. Four randomly selected 10 gram samples from one container and three samples from the second container were pulverized separately and divided into two parts. The resulting samples were analyzed for plutonium and uranium. Averages are shown in Table 3.3.

TABLE 3.3

PLUTONIUM AND URANIUM DISTRIBUTION
IN PNEUMATICALLY IMPACTED UO₂-PuO₂

<u>Sample</u>	<u>wt% Pu</u>	<u>wt% U</u>
1 A	2.08	86.0
1 B	2.05	86.1
1 C	2.06	85.8
1 D	2.09	86.3
2 A	1.12	86.8
2 B	1.13	87.1
2 C	1.11	86.4

Ceramographic examination of the impacted UO₂-PuO₂ revealed a uniform dispersion of PuO₂ particles in the UO₂ matrix (Figure 3.8). X-ray diffraction analysis indicated the presence of UO₂ and PuO₂ only, with no evidence for the formation of a solid solution of PuO₂ in UO₂.

As-Polished

100 X

As-Polished

500 X

FIGURE 3.8

Microstructure of Pneumatically Impacted UO_2 -2.5 wt% PuO_2
Negative No. R874, R876
AEC-GE RICHLAND, WASH.

Cermet Encapsulation - D. W. Brite and K. R. Sump

A technique for the encapsulation of cermets simultaneously with the pneumatic impaction of the cermet was developed. Cermet powder was loaded in the impaction can and surrounded by a layer of metal powder containing no ceramic. The encapsulating metal was the same as that used for the matrix of the cermet. However, a metal different than that of the matrix could be used.

Macrostructures of encapsulated UO_2 -50 wt% tungsten cermet, UO_2 -35 vol% stainless steel cermet, and UO_2 -35 vol% iron cermet, formed by pneumatic impaction, are shown in Figures 3.9 and 3.10. Characteristics of thin tungsten-coated wafers of UO_2 -80 vol% tungsten cermet formed by high-energy-rate impaction are shown in Figure 3.11.

FIGURE 3.9

Radial Cross Section of a Metal Encapsulated Cylinder of UO_2 -50 wt% Tungsten

SS and UO₂-35 vol% SSFe and UO₂-35 vol% FeFIGURE 3.10

Microstructure of Bond Area (100X - As-Polished) and Axial Cross Section
(2X, As-Polished) of Impact-Encapsulated Cylinders

FIGURE 3.11
Pneumatically Impacted, Metal Clad Wafer
of UO_2 -80 vol% Tungsten

Fission Product Encapsulation - D. W. Brite and K. R. Sump

In cooperation with Chemical Laboratory personnel, simulated fission product oxides were pneumatically impacted in both 2.5- and 4-inch diameter containers. Materials impacted and the densities obtained included: SrTiO_3 , 4.66 g/cc, 91.2% TD; SrO , 4.47 g/cc, 95.0% TD; CeO_2 , 6.09 g/cc, 86.5% TD; Nd_2O_3 , 7.13 g/cc, 98.5% TD; and CsCl , 3.68 g/cc, 92.7% TD.

Remote Fabrication Studies - R. C. Smith

A manipulator system, consisting of two boom-crane mounted manipulators, was installed in the remote fuel element fabrication enclosure (Figure 3.12). Equipment shakedown operations are substantially completed and the system is operating satisfactorily. Lighting and camera arrangement studies were initiated to determine optimum conditions for using TV cameras as the only visual link to the manipulator system.

FIGURE 3.12

Remote Fuel Fabrication Manipulator Equipment

Fuel Element Refurbishing - R. C. Smith

A water tank was installed on one side of the remote fabrication facility for experimental fuel element refurbishing studies. Small hand tools built to operate at the end of 9-foot extension rods were constructed and a newly developed replacement rod was successfully assembled in an unirradiated 19-rod cluster fuel element under water (Figure 3.13).

FIGURE 3.13

Fuel Element Refurbishing Tank

Tests of Zircaloy-2 wire used as rod spacing members on PRTR fuel elements indicate a 90° bend does not cause circumferential cracks and has only a small effect on the surface imperfections observed. Twenty-four wires were examined, including three wires randomly sampled from the batch of wire used in the 1000-series UO_2 fuel elements now in the PRTR. Twelve of the wires were bent 90° by the same method used in fuel element assembly. Visual examination at 40 X magnification and photographs at 18.5 X (Figure 3.14) revealed longitudinal pretest microcracks less than 0.001 inches deep in twenty of the wires. Such cracks were widened only very slightly by the 90° bending (Figure 3.15).

FIGURE 3.14

Pretest Microcracks in Zircaloy-2, 18.5X

FIGURE 3.15

Microcracks in Zircaloy-2 Wire After Bending 90 Degrees, 100X
Negative No. 4K 349A, 4K 349B

4E GERCH AN WA 4

Fuel Element Rejuvenation - R. C. Smith

The rejuvenation fuel element (GEH-4-81), successfully irradiated for one cycle in the MTR at the National Reactor Testing Station, was returned to Hanford for re-enrichment.

MELEAGER calculations were made to determine the amount of low exposure plutonium needed to restore an irradiated, mixed oxide fuel element to its initial reactivity. Preliminary calculations show a minimum of 141 grams of low exposure plutonium will be required to rejuvenate a PRTR fuel element exposed to 5000 MWD/ton. Arbitrarily picking a reactivity stop at $K_{\infty} = 1.15$, the second irradiation life would be an additional 4400 MWD/ton with total accumulated exposure of 9400 MWD/ton. These calculations were made for order of magnitude studies only since a fuel element would not necessarily be rejuvenated after 5000 MWD/ton.

Materials Joining, Tungsten - R. F. Klein

Tungsten was bonded without the formation of the large grains that normally form during recrystallization after welding. Small diameter (0.242-inch OD x 0.190-inch ID) tubing was bonded to an end cap through the use of magnetic force resistance welding (Figure 3.16). The rapidity with which the bonding takes place (less than 20 milliseconds) limits recrystallization to an extremely narrow band in the weld area. The recrystallization grains that do form are smaller than the grains of the parent metal, indicating the joint may be stronger than the parent material.

3. 21

HW-76301

10 X

100 X

FIGURE 3.16

Microstructure of Tungsten-Tungsten Bond

Negative No. 5K 250A;5K 250B

AEC-GE RICHLAND, WASH.

Materials Joining, UO_2 -W Cermet - R. F. Klein

Successful joining of pneumatically impacted UO_2 -50 wt% tungsten cermet material was achieved. Figure 3.17 shows cross sections through the bond. The edge of a rectangular 0.050-inch thick x 0.375-inch long bar was bonded to the flat surface of a 0.375-inch diameter x 0.050-inch thick disk using the magnetic force resistance welding process. No detectable UO_2 vaporization occurred during the bonding of the pieces because of the extremely short welding time (10 milliseconds) used in conjunction with a high current density of 184,000 amps per square inch. Pressure applied during welding produced cracking of the base material, but it should be possible to eliminate this in future work.

Particle Size Distribution for Vibrational Compaction - J. O. McPartland and J. J. Hauth

Uranium dioxide was vibrationally compacted to 93% TD in a 1-3/8 inch OD x 1/4 inch ID annular fuel capsule. The particle size distribution required to obtain this high density was accurately predicted from a mathematical model developed by Applied Mathematics. This result, in conjunction with previous results with more conventional geometries, indicates that fuel can be vibrationally compacted to high densities in novel geometries without extended empirical determination of optimum particle size distribution.

PRTR UO_2 - PuO_2 Swaged Fuel Fabrication - C. H. Bloomster and R. E. Bardsley

Five key developments have advanced the swage process for fabricating plutonium fuel elements. A pilot run of fifty fuel elements fabricated by swage compaction is nearly complete. Thirty-eight swaged UO_2 - PuO_2 fuel elements were fabricated and charged in the PRTR. The maximum exposure of the swaged elements to date is 900 Mwd/ton.

The swage process was standardized for the remainder of the pilot run in order to obtain cost estimates on the process. The significant process developments are summarized as follows:

10 X

50 X

FIGURE 3.17

Microstructure of W-UO₂ Cermet Bond

Negative No. 5K 473B, 5K 473A

AEC GE RICHLAND WASH

1. The fuel element is loaded (Figure 3.18) with 1/2-inch increments comprising coarse UO_2 and a UO_2 - PuO_2 mixture.⁽¹⁾ This method restricts gross deviations from the average plutonium concentration in the fuel rod to the short length of one increment. This is particularly desirable in the absence of a nondestructive test for plutonium distribution.

FIGURE 3.18

Incremental Loading of Fuel Rod for Swaging

-
1. C. H. Bloomster, R. E. Bardsley, and W. T. Ross. An Incremental Loading Process to Control Plutonium Enrichment in Mechanically-Mixed Oxide Fuel Systems, HW-SA-2679. November 1962.

2. A simplified procedure was developed for decontaminating the weld zone (Figure 3.19). The procedure takes 7 minutes, including movement of the rod to and from storage.

FIGURE 3. 19

Weld Zone Decontamination of Loaded Fuel Rod

Negative No. 32024-1

AEC-GE HIGHLAND WASH

3. A swageable end cap (Figure 3.20) was developed⁽¹⁾ which simplified loading and completely contained the plutonium through the subsequent operations. Over 1400 rods have been swaged with no failure in either weld or cladding.

FIGURE 3.20
Welded Swageable End Cap

-
1. L. C. Lemon and W. T. Ross. Head-Welded Swageable End Cap for $\text{UO}_2\text{-PuO}_2$ Fuel Elements, HW-74287. July 13, 1962.

Negative No. 0622111-4

AEC GE RICHLAND WASH

4. A blend of particle sizes was developed which permitted the fuel rod to be loaded with fused oxide to a reproducible ($\pm 1\%$) high tap density.
5. The rods are swaged (Figure 3.21) to a prescribed length ($\pm 1/16$ inch). Micrometer adjustment of the swaging dies is an unusual feature of the machine.

FIGURE 3. 21

Diameter Measurement on Swaged Fuel Rod

Negative No. 32024-4

AEC GF R CHLAND WASH

The Zircaloy tubing was categorized by weight per unit length and a relationship between initial tubing length and final length after swaging was established. The combination of reproducible powder loading, categorized tubing, and micrometer control of the swaging dies permitted the length to be controlled to 1/16 inch on the individual rods and aided assembly of the finished cluster.⁽¹⁾

A pilot run of vibrationally compacted $\text{UO}_2\text{-PuO}_2$ fuel elements will follow the completion of the pilot run of swaged elements. Fifteen vibrationally compacted fuel assemblies have been completed and irradiated in the PRTR. The facilities and process for vibrational compaction are being modified to incorporate the latest technology for the pilot run.

EBWR Plutonium Fuel Loading - R. E. Sharp

The EBWR plutonium fuel loading program continued on schedule. Design of a vibrational compaction facility to be installed in the Plutonium Fabrication Pilot Plant was approved by the AEC and construction will start in April, 1963. Equipment and material was ordered for a delivery to meet the overall program schedule. A prototype facility is being set up to check out experimental equipment and develop parts of the process concurrently with the installation of the plutonium process facility.

Powder Fuel Loading Studies - R. E. Bardsley

A promising method now being investigated for fuel loading is the bottle loading of physically mixed oxides. This consists of blending the core material for each fuel rod in an individual container. Preliminary loading and distribution tests were conducted using various blending methods (V-blender, roller blender, end-for-end tumbling) and a range of blending times from 5 minutes to 2 hours. Gamma scans and autoradiographs show that roller blending and end-for-end tumbling provide a more uniform mixture than V-blending. Blending for periods longer than 30 minutes may be desirable. Figure 3.22 shows autoradiographs and

1. R. E. Bardsley and R. E. Sharp. Fabrication of $\text{UO}_2\text{-PuO}_2$ Swage Compacted Fuel Elements, HW-SA-2683. Presented at ANS Meeting in Washington, D. C., November 26-29, 1962.

FIGURE 3.22

Comparative Autoradiographs and Gamma Scan Traces Showing
Plutonium Distribution in Two Bottle Loaded, Mixed Oxide Fuel Rods

(Core material was blended by end-for-end tumbling
for 5 minutes (at top) and 2 hours (at bottom))

Negative No. 0630785

corresponding gamma traces for two fuel rods blended by the same method for different lengths of time. Additional evaluation of these methods is underway.

Nondestructive Test for Plutonium Distribution - R. E. Bardsley and
S. T. Field

A gamma scanning, nondestructive test was developed to obtain both qualitative and quantitative data on the distribution of plutonium in a fuel rod. A prototypic unit (Figure 3.23) was set up in cooperation with the Testing Methods Operation.

FIGURE 3. 23

Left, Test Bed and Crystal Head; Center, Single Channel
Analyzer and Power Supply with Countrate Meter Above;
Right, Strip Chart Recorder

Gamma scan traces of vibrationally compacted fuel rods were correlated in every case with autoradiographs of the same rods. Fuel rods can be scanned in 8 minutes using this method, versus 72 hours for the autoradiographic technique. Standards being fabricated for quantitative calibration of the unit will range from 0.01 wt% PuO_2 to 10 wt% PuO_2 .

Co-Extruded Al-Pu Fuel Elements Cut to Short Lengths - L. C. Lemon

Special machining techniques were developed to permit, for the first time, contamination-free end closures on short pieces cut from a long, co-extruded, Al-clad Al-Pu rod. The cut pieces, 5 inches long by 3/4 inch diameter, are shown in Figure 3. 24.

FIGURE 3. 24

Al-Pu Pieces Cut from
Co-Extruded Rod and
Ready for Counterboring

Negative No. 0630686-2

Significant steps in the procedure include: (1) coating the co-extruded rod with a lacquer to keep the cladding free of contamination; (2) cutting the rod into short lengths on a lathe, making an outside-in cut while avoiding drawing core material back to the cladding; (3) counterboring the ends, (Figures 3. 25 and 3. 26) using a specially ground endmill as the last step to prevent smearing the core material into the cladding; (4) decontaminating

the counterbored surface with alcohol and the cladding end surface with emery cloth; (5) removing the lacquer from the external surface; and (6) installing and fusion welding a plug in the clean counterbored end.

FIGURE 3.25
Counterboring Short
Al-Pu Pieces

FIGURE 3.26
Use of Special End Mill
to Avoid Smearing
Contamination in
Counterbored Area

Special Fuel Element Fabrication - R. E. Bardsley, C. H. Bloomster
and L. C. Lemon

Six, Zircaloy jacketed, stainless steel clad, thermocoupled elements were fabricated for special studies (Figure 3.27). These test elements will be used to evaluate the film deposition characteristics of ammonium hydroxide conditioned coolant. The fused UO_2 fuel core

(5/8-inch OD x 6 inches long) was compacted to 85.5% TD with an electrically operated hammer. The cladding is 3/8-inch thick stainless steel covered by a shrink fitted, 0.015-inch thick Zircaloy outer jacket. Spacer legs, welded to the stainless can, extend through slots in the Zircaloy.

FIGURE 3.27

Assembled Stainless Steel Clad,
Zircaloy Jacketed, Thermocoupled Capsules

Eighteen pellet-type $\text{UO}_2\text{-PuO}_2$ and 16 vibrationally compacted fuel rods were fabricated for physics experiments in PRTR. The pellets were 1/2-inch diameter x 1/2-inch long and contained 0.48 wt% PuO_2 . Rods were wrapped with a Zr-Co alloy wire for flux monitoring and assembled into clusters. Two of the clusters were charged in the PRTR.

A total of 550, 3-foot long, Al-Pu rods containing high exposure plutonium were fabricated for experiments in the PCTR and PRCF.

FUELS TESTING AND ANALYSIS

Inspection of PRTR Fuel Elements - M. D. Freshley and W. J. Bailey

A total of eight irradiated PRTR fuel elements were examined in detail, particularly in regard to condition of cladding. General appearance was good, and there were no serious cladding defects. Some features suggesting possible design improvements were observed.

A swage-compacted UO_2 - PuO_2 fuel element (5110) having an average exposure of approximately 600 Mwd/ton was examined in the Fuel Element Examination Facility. The examination revealed: (1) very little wear on the contact points on the top end bracket; (2) severely worn contact points on the bottom end bracket; (3) no loose wires or worn spots on the visible wire wrap surfaces; (4) three damaged circumferential strip bands, apparently caused during charging of the elements (the resulting upward displacement of the bands uncovered discolored spots on the surface of the fuel rods at the band contact points); (5) a limited amount of white corrosion product on wire wrap welds on the top ends of some rods; (6) a corroded wire wrap weld area on the bottom of one rod (weld zone completely covered with white corrosion product); and (7) otherwise satisfactory fuel rod surfaces and no discoloration. The contact points on the bottom end bracket of the element had previously caused excessive process tube wear. Extended surface, clip-on wear pads were attached to the end brackets before reinsertion into the reactor.

Another swaged UO_2 fuel element (1050) was examined in the FEEF to determine the cause of difficulties experienced during an attempt to insert the element into a PRTR process tube. A broken, single rod wire wrap was discovered, as well as other evidence that mechanical damage was incurred during fuel transfer. This element, which had reached approximately 2,600 Mwd/ton, will be refurbished in the reactor basin and recharged for further irradiation.

An irradiated Al-Pu PRTR spike element (5088) with an average exposure of 54 Mwd, which gave indications of a cladding leak during stagnant water tests in the PRTR basin, was examined both in the FEEF and in the storage basin. The general appearance of the rods was good and no defects were observed. A loose spiral wire wrap on one fuel rod indicated that mechanical core-clad interaction had shortened the rod approximately 5/32 inch. The element will be disassembled and each rod inspected individually in an effort to locate the source of fission product activity.

A second Al-Pu spike element (5084) with an average exposure of 74.2 Mwd (approximately goal exposure) was examined in the basin. Fuel rod shortening (approximately 0.28% is evidenced by loose spiral spacing wires. One circumferential strip band had been severely damaged.

Two other suspected leaky UO_2 - PuO_2 elements (swage compacted 5162 and vibrationally compacted 5113) were examined in the FEEF. Their general surface appearances also were good. No serious surface defects were visible. However, a cladding defect was later located on an inner rod of 5113. The extended surface, clip-on wear pads performed satisfactorily and only limited superficial wear was observed. Some of the spot fusion welds at the ends of the spiral spacing wires were corroded, probably as a result of inadequate inert gas coverage during assembly.

The center support stem was accidentally twisted from the gussets of the top end fixture of a UO_2 - PuO_2 vibrationally compacted PRTR element (5120) during manipulation in the FEEF. Subsequent examination of the element in the storage basin revealed that the surface condition of the rods was good with very few scratches and no loose wires. Careful examination of the broken gusset welds showed them to be sound. Subsequent destructive evaluation demonstrated that the tensile strength of nonirradiated end fixtures is more than three times design strength. However, the end fixtures were not designed to resist twisting, and only moderate torsional forces (35-70 foot pounds) were required to break the stems from the gussets. Metallographic examination of a nonirradiated end bracket revealed normal hydride

concentration for autoclaved Zircaloy. Increased hardness values in the immediate vicinity of the fracture indicated a small amount of cold work. No metallurgical weakness was detected. The present end fixture design allows failure in a component not containing fuel when movement of the element is restrained during twisting of the stem. Ordinary manipulations in PRTR equipment transmit no twisting motion of any consequence to the fuel elements.

Postirradiation Examination of a Swage-Compacted UO_2 - PuO_2 PRTR Fuel Element - M. D. Freshley

A transverse cross section of a swage-compacted, incrementally loaded, UO_2 - PuO_2 PRTR fuel rod revealed that localized core temperatures resulting from PuO_2 segregation were sufficient to cause center void formation and columnar grain growth in a limited region (Figure 4.1). Previous postirradiation examinations of similar PRTR fuel rods revealed only low order sintering and equiaxed grain growth in the PuO_2 -rich areas (Figure 4.2). The fuel rod, loaded with 157 increments, was included in an element (5096) that generated 1139 kw maximum power during accumulation of an average exposure of 300 Mwd/ton. A large population of lenticular voids in the columnar grains adjacent to the center void (Figure 4.3) indicate that the center void and columnar grains were formed at temperatures slightly above that required for lenticular void formation. An unidentified white appearing second phase, possible uranium nitride, is dispersed throughout the UO_2 - PuO_2 (Figure 4.3). There is no evidence of unusual core-clad reaction in the region of increased PuO_2 concentrations and no local discoloration indicative of "hot spots" on the external cladding surface. The fuel rod operated satisfactorily under these conditions.

A limited reaction layer of uniform thickness (approximately 0.0005 inch thick) was observed on the inner surface of the Zircaloy cladding (Figure 4.4). During postirradiation study, a reaction layer was observed in a 157 increment rod, but not in an 80 increment rod, even though the constituents of the two rods were thought to be identical. The effect is

FIGURE 4.1

Transverse Cross Section (top) and Autoradiograph (bottom)
of an Incrementally Loaded, Swage-Compacted, $\text{UO}_2\text{-PuO}_2$
PRTR Fuel Rod Showing the PuO_2 -Rich Areas

FIGURE 4. 2

Longitudinal Cross Section (top) and Autoradiograph (bottom)
of an Incrementally Loaded, Swage-Compacted, $\text{UO}_2\text{-PuO}_2$
PRTR Fuel Rod Showing the PuO_2 -Rich Areas (3-1/2 X)

Negative No. B-3655

4 - 0 5 1 1 0 AND W - 4

FIGURE 4. 3

High Population of Lenticular Voids in Columnar Grains
Adjacent to the Central Void in a $\text{UO}_2\text{-PuO}_2$ PRTR Fuel Rod
(White inclusions are dispersed throughout the fuel) (250 X)

FIGURE 4. 4

Transverse Cross Section of a Swaged $\text{UO}_2\text{-PuO}_2$, PRTR Fuel Rod
Showing a Reaction Layer on the Inner Surface
of the Zircaloy Cladding (250 X)

thought to be related to impurities in the fuel material. The inner surface of the cladding having the reaction layer appears black, whereas the inner surface of the cladding without the reaction layer retained its original metallic luster.

Postirradiation Examination of a Vibrationally Compacted $\text{UO}_2\text{-PuO}_2$ PRTR Fuel Element - M. D. Freshley

Destructive examination of a vibrationally compacted, $\text{UO}_2\text{-PuO}_2$ PRTR Mark I fuel element (5113), discharged from the PRTR as a suspected leaker, revealed the first cladding failure discovered and examined in a conventional element in the PRTR. A hole in the Zircaloy cladding (Figure 4.5), approximately 3/16 inch long by 3/32 inch wide, was found approximately 36-1/2 inches from the top of a rod in the six-rod ring. Only minor fuel washout and fission product release occurred and there was no evidence of waterlogging as a result of three pressure and two power cycles of the reactor after a leak was suspected. The average exposure of the element is approximately 370 Mwd/ton and maximum recorded heat generation rate is 1140 kw. An autoradiograph of the irradiated rod shows no high localized PuO_2 concentration in the failure region and only low order sintering of the core occurred during irradiation. There is no evidence of excessive cladding temperature, or of associating the defect with the rod wire wraps or with fretting corrosion. The cladding erupted slightly around the hole and the fracture surface appears brittle. Short radial cracks lead away from the hole where the Zircaloy is severely hydrided. Further postirradiation examination is being conducted to determine the cause of failure.

FIGURE 4.5

Cladding Defect in Vibrationally Compacted, Zircaloy-Clad PRTR Fuel Rod (5113). Transverse Cross Section, A-A, Shows (1) Low Order Fuel Sintering; (2) Minor Washout of Nonsintered Fuel; and (3) Cracks and Hydride in Outer Surfaces of Zircaloy Cladding.

Negative No. B-5224

AEC GE RICHLAND WASH

Examination of PRTR nested Tubular Fuel Element No. 1501 - W. J. Flaherty

Destructive examination of the first PRTR Mark-II concentric, tubular fuel element was conducted to obtain information on internal pressure buildup caused by release of sorbed and fission product gases. The test element (1501) was successfully irradiated to an exposure of 1360 Mwd/ton. Analysis of gas samples taken from the element revealed that less than 10% of the fission product gases formed during irradiation were released from the fuel. Measurements of gases collected during sampling indicate that less than 50% of the sorbed gases were released during irradiation. The calculated collapse conditions for the inner cladding of the outer tube of the Mark II-C PRTR fuel element (Figure 4.6) were based on assumed release of 25% of the fission gases and 75% of the sorbed gases. Information from this test element indicates that the internal pressures predicted are very conservative and that exposures more than twice those shown could be attained without failure by cladding collapse. The internal pressure of the outer component during full power operation was less than 90 psig.

FIGURE 4.6

Calculated Collapse Conditions for PRTR, Mark II-C Tubular Fuel Element Cladding (Support by fuel neglected)

Extended Surface, PRTR Fuel Element Wear Pads - M. D. Freshley,
W. J. Flaherty, and R. E. Sharp

Accelerated wear or fretting corrosion occurred between the 1/16-inch wide fuel element end fixture wear pads and the Zircaloy process tubes in the PRTR. Wear was reduced by increasing the contact area of each of the three pads on each end fixture to 1/4 inch wide x 1/2 inch long. Extended surface, clip-on wear pads which can be remotely attached to irradiated, 19-rod cluster PRTR fuel elements were tested in- and out-of-reactor. A 19-rod cluster element with clip-on pads developed no measurable wear when tested at PRTR coolant temperature and flow conditions for 89 days in the mechanically vibrated, TF-7 (out-of-reactor) test loop. Clip-on pads were subsequently remotely installed on 12 irradiated PRTR fuel elements. The in-reactor performance of the pads is encouraging; no measurable wear was observed on the elements or process tubes after 27 days operation in the reactor. Installation tools and clip-on wear pads were also fabricated for 13 UO_2 fuel elements selected for extended life tests in PRTR.

Fuel element end brackets with ring contact surfaces (Figure 4.7) to alleviate fretting are being tested in the TF-7 test loop at PRTR coolant conditions. After 26 days operation with a superimposed vibration on the test section, only superficial wear occurred on one area of the 3/8-inch wide contact ring on the bottom bracket and the oxide film was not removed from the inner surface of the simulated Zircaloy process tube. No wear occurred on the contact area of the top end bracket. The test is being continued.

FIGURE 4.7
Ring Contact Surface for
19-Rod Cluster PRTR Fuel
Elements. Ring is 3/8
Inch Wide.

Irradiation Testing Program for EBWR Prototypic Fuel Rods - W.J. Bailey

Irradiation tests of prototypic, EBWR, UO_2 - PuO_2 fuel rods were initiated. The prototypic fuel comprises vibrationally compacted, UO_2 (0.22% U^{235}) with 2-3 wt% PuO_2 in which the plutonium contains 8% Pu^{240} . A series of capsule screening tests will be followed by extended tests of prototypic fuel rods in a suitable reactor facility.

Irradiation Performance of MgO-PuO_2 and $\text{ZrO}_2\text{-PuO}_2$ Fuels -

M. D. Freshley and D. F. Carroll

Microstructural features different from those in many irradiated ceramic fuels were observed in irradiated MgO-PuO_2 sintered pellets. High resolution autoradiographs provided better definition of the behavior of fission products and plutonia in irradiated MgO-PuO_2 and $\text{ZrO}_2\text{-PuO}_2$ fuel pellets.

Autoradiographs made from transverse cross sections of irradiated $\text{MgO-3.05 wt\% PuO}_2$ pellets indicate that the fission products and/or the PuO_2 second phase had migrated from a central region of translucent columnar grains (Figure 4.8). Ceramographic examinations suggest that the PuO_2 was rejected from the larger high-temperature columnar grains near the thermal center of the specimens and concentrated at the boundary between large and small columnar grains. The distribution of the PuO_2 second phase was not altered in the smaller columnar grain region in spite of extensive recrystallization. High fission product concentrations on the inner surface of the center void, indicated by the autoradiograph, were also resolved on high magnification micrographs.

Limited migration occurred in the irradiated, $\text{MgO-13.5 wt\% PuO}_2$ pellets (Figure 4.9). However, the maximum operating temperatures were less than those of the $\text{MgO-3.05 wt\% PuO}_2$ pellets mentioned earlier. Two concentric rings on the autoradiograph reveal limited fission product and/or PuO_2 migration in these pellets also. The rings correspond to boundaries demarcating different grain structure regions.

FIGURE 4.8

Transverse Cross Section (top) and Autoradiograph (bottom) of an Irradiated MgO-3.05 wt% PuO₂ Pellet (calculated maximum centerline temperature - 2200 C) Illustrating the Rejection of Fission Products and PuO₂ from the Large, Central, Translucent Grains

6X

6X

FIGURE 4.9

Transverse Cross Section (top) and Autoradiograph (bottom)
of an Irradiated, $\text{MgO}-13.5 \text{ wt\% PuO}_2$ Pellet (calculated
maximum centerline temperature $< 2200 \text{ C}$) Showing Concentration
Rings Indicating Limited Migration of Fission Products and/or PuO_2

Negative No. B-1557, 352

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Transverse autoradiographs and microstructural features of the irradiated ZrO_2 -2.2 wt% PuO_2 pellets were significantly different from those of the ZrO_2 -10.4 wt% PuO_2 material, although both were supposedly irradiated to produce approximately the same maximum centerline fuel temperatures. No center void or columnar grains were formed in the low PuO_2 -containing pellets (Figure 4.10). Short radial cracks developed in the transition region. Autoradiographs show that the fission products and/or PuO_2 are severely segregated into randomly distributed islands. Autoradiographs of sibling, nonirradiated pellets show incomplete homogenization in ZrO_2 -2.2 wt% PuO_2 pellets even though the ceramic grade powders were blended 72 hours before pressing. Limited homogenizing occurred near the thermal center of the irradiation specimens that operated at the higher temperatures.

The microstructural features of the irradiated ZrO_2 -10.4 wt% PuO_2 fuel (Figure 4.11) include center voids and columnar grains. Autoradiographs show that fission products and/or plutonia have migrated to preferred radial locations.

Examination of the Zircaloy cladding from the ZrO_2 -10.4 wt% PuO_2 fuel capsules (calculated maximum core temperature above melting) that failed during irradiation revealed severe hydriding and extensive cracking. Six cracks that completely penetrated the cladding (Figure 4.12) are shown in one transverse cross section. A core-clad reaction, possibly indicative of excessive interface temperatures, was observed.

FIGURE 4.10

Transverse Cross Section (top) and Autoradiograph (bottom) of an Irradiated ZrO_2 -2.2 wt% PuO_2 Pellet (in-reactor melting is doubtful) Showing Homogenization of the Fuel Near the High Temperature Thermal Center

Negative No. 340, 87

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FIGURE 4.11

Transverse Cross Section (top) and Autoradiograph (bottom) of an Irradiated ZrO_2 -10.4 wt% PuO_2 Pellet (estimated core temperature - 1700 C) Showing Radial Fission Product and/or PuO_2 Migration

Negative No. B-5031; 342

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FIGURE 4.12

Transverse Cross Section of ZrO₂-10.4 wt% PuO₂ Pellet (estimated temperature above melting) Which Failed During Irradiation. The Severely Hydrided, Zircaloy Cladding Failed at Six Locations.

Irradiation of Uranium-Plutonium Oxide - W. J. Bailey and T. D. Chikalla

Preliminary postirradiation examination of irradiated specimens of uranium-plutonium oxide made by various methods was previously reported.⁽¹⁾ Fission fragment migration and nonuniform plutonium distribution was revealed.

Samples for analyses were removed from sharply-defined zones of markedly different radioactivity levels observed on a cross section of an irradiated (U, Pu)O₂ fuel capsule (GEH-14-86), shown in Figure 4.13. The autoradiograph in Figure 4.13 was prepared by a modified GE-APED method which employs a high-resolution, beta sensitive film plate. Accurately located, 0.5 mg analytical samples were obtained by collecting

1. Ceramics Research and Development Quarterly for October - December, 1962, HW-76300, pp. 29.

microdrillings from 0.020-inch diameter, 0.010-inch deep holes, using a diamond tipped microdrill developed by Radiometallurgy Laboratory personnel. Additional shielding is required for the equipment before other fuel specimens can be drilled.

The average exposure of the 0.5 inch diameter (U, Pu)O₂ sintered pellets is 5600 Mwd/ton of (U, Pu)O₂ or 1.66×10^{20} fissions/cc. Samples from the zones of lowest activity (1/4 inch diameter at center) and of highest activity (1/32-inch-wide annular ring, 1/4 inch ID) provide contact dose rates of 200 mrem/hr (including 15 mr/hr gamma) and 450 mrem/hr (including 100 mr/hr gamma), respectively. The samples were submitted for analyses of plutonium, uranium, Ru¹⁰⁶, Ce-Pr¹⁴⁴, Zr-Nb⁹⁵, and Cs¹³⁷ contents.

Additional specimens were autoradiographed (see Figures 4.14 - 4.18) to establish a microsampling pattern.

Autoradiograph

FIGURE 4.13

UO₂-4.13 mole% PuO₂ Fuel Capsule (GEH-14-86) with an Initial Density of 91% TD Irradiated to an Exposure of 166×10^{18} Fissions/cc (Microsampling unit was used to pinpoint location to drill, and to collect analytical samples.)

Negative No. 0622107-11, B-4986

AEC GE RICHLAND WASH

6X

Autoradiograph

6X

FIGURE 4.14

UO₂-0.0259 mole% PuO₂ Fuel Capsule (GEH-14-20) With an Initial
Density of 90% TD Irradiated to an Exposure of 84×10^{18} fissions/cc

Negative No. 63; A-8168

AEC-GE RICHLAND, WASH.

Autoradiograph

FIGURE 4.15

UO₂-1.02 mole% PuO₂ Fuel Capsule (GEH-14-83) With an Initial
Density of 93% TD Irradiated to an Exposure of 28×10^{18} fissions/cc

Negative No. 64; A-276

AEC-GE RICHLAND, WASH.

Autoradiograph

FIGURE 4.16

UO₂-4.13 mole % PuO₂ Fuel Capsule (GEH-14-87) With an Initial
Density of 91% TD Irradiated to an Exposure of 26×10^{18} fissions/cc

Negative No. 65; A-34

AEC-GE RICHLAND, WASH.

Autoradiograph

FIGURE 4. 17

UO₂-0.0259 mole % PuO₂ Fuel Capsule (GEH-14-22) With an Initial Density of 65% TD Irradiated to an Exposure of 14×10^{18} fissions/cc

Negative No. 95; A-7823

AEC-GE RICHLAND, WASH.

Autoradiograph

6X

FIGURE 4.18

UO₂-0.187 mole % PuO₂ Fuel Capsule (GEH-14-66) With an Initial
Density of 65% TD Irradiated to an Exposure of 79×10^{18} fissions/cc

Negative No. 97, A-7824

A F CH AN W

Irradiation of Pneumatically Impacted $\text{UO}_2\text{-PuO}_2$ - G. R. Horn

Capsules for the first irradiation test of pneumatically impacted $\text{UO}_2\text{-PuO}_2$ were fabricated and transferred to the MTR. The $\text{UO}_2\text{-2.5 wt% PuO}_2$ was vibrationally compacted into 3-1/2-inch-long capsules to a density of $\sim 80\%$ TD. The 9/16-inch diameter capsules were irradiated to generate $\sim 900,000 \text{ Btu}/(\text{hr})(\text{sq ft})$ surface heat flux.

Irradiation of UO_2 Single Crystal Pellets - G. R. Horn

Additional evidence that the thermal conductivity of large grain UO_2 is much greater than that of small grain UO_2 was provided by irradiating a capsule containing 1/2-inch diameter single crystal pellets.⁽¹⁾ Gross structural alteration (other than fracture) did not occur during irradiation under conditions known to cause central melting and extensive columnar grain growth in fine-grained, polycrystalline samples. Eight additional capsules are being irradiated to provide a more direct comparison of the thermal conductivity of large grain and small grain UO_2 . The test capsules contain various geometrical combinations of sintered UO_2 pellets and single crystal, fused UO_2 pellets. Four capsules were irradiated for one ETR cycle (~ 20 equivalent full power days) at a maximum surface heat flux of $\sim 800,000 \text{ Btu}/(\text{hr})(\text{sq ft})$. The other four, generating the same surface heat flux, will be discharged after five ETR cycles.

Irradiation of $\text{UO}_2\text{-W}$ Cermets - G. R. Horn and W. J. Flaherty

$\text{UO}_2\text{-W}$ cermets were chemically and dimensionally stable during short term, high temperature irradiations. Continued postirradiation examination of a $\text{UO}_2\text{-W}$ cermet capsule,⁽¹⁾ irradiated for 2 hours at a cladding surface temperature greater than 2100°C , revealed no evidence of reaction between the UO_2 and tungsten. The postirradiation diameter of the tungsten clad capsule was not changed from the 0.250 ± 0.001 inch preirradiation value.

1. Ceramics Research and Development Operation Quarterly Report, October - December, 1962, HW-76300.

Phoenix Fuel Experiment - M. D. Freshley

A plutonium burn-up experiment* designed to investigate the effect of exposure on the reactivity of plutonium was previously reported.⁽¹⁾ Three aluminum-plutonium alloy specimens were irradiated in the MTR for seven cycles (1.2×10^{20} fissions/cc or 92% burnup of original fissionable atoms) and the resulting reactivity changes were measured in the Advanced Reactivity Measurement Facility (ARMF). These specimens contained plutonium which initially had 6.25, 16.33, and 27.17% Pu²⁴⁰. Preliminary data indicate that after seven cycles of irradiation in the MTR, the gross reactivity of the sample incorporating plutonium that initially contained 27.17% Pu²⁴⁰ is 38% higher than the sample that initially contained 6.25% Pu²⁴⁰. The samples will be returned to Hanford for reactivity measurements in the Plutonium Recycle Critical Facility and subsequent burnup and isotopic analyses.

Two additional specimens were fabricated to determine the effect of an aluminum-plutonium sample on the ARMF neutron energy spectrum. Details of the calibration test were established and the experiment was performed. Data are being evaluated.

ThO₂-PuO₂ Studies - M. D. Freshley and H. M. Mattys

ThO₂-PuO₂ pellets (95-96% TD) containing 2 - 5 wt% PuO₂ were made for irradiation tests. An unexpected sensitivity to powder and die lubricants was observed when pellet surfaces spalled and cracked during sintering. Excellent pellet quality was obtained by dry pressing calcined (900 C) powder mixtures and sintering in hydrogen at 1600 C for 6 hours. Thirty pellets of each composition were made.

*In cooperation with L. C. Schmid, Physics Laboratory.

- 1. O. J. Wick. Quarterly Progress Report, Plutonium Metallurgy Operation, July - September, 1962, HW-74718. October 15, 1962.
(Secret)

Low Temperature Sintering of UO_2 - W. J. Flaherty

Ceramographic examination of a sample of cold-swaged UO_2 PRTR fuel rod heated in the laboratory for more than 1000 hours at 550 C revealed no bonding between UO_2 particles. This supports the hypothesis that low temperature sintering⁽¹⁾ observed in irradiated, swaged UO_2 PRTR fuel rods is an irradiation effect.

Short Duration Irradiations of UO_2 and $\text{UO}_2\text{-PuO}_2$ - W. J. Bailey and T. D. Chikalla

Heat generation rates were determined for four capsules, each containing high density pellets of $\text{UO}_2\text{-0.154 mole \% PuO}_2$ and of UO_2 (1.00 mole % U^{235}), that were irradiated in the VH-4 Hydraulic Rabbit Facility of the MTR.⁽¹⁾ Rod powers were estimated from the activities of Al-0.5% Co alloy and pure cobalt flux-monitoring wires. Capsule GEH-21-16, irradiated for 60 minutes, averaged 18.4 kw/ft in the $\text{UO}_2\text{-PuO}_2$ section and 18.3 kw/ft in the UO_2 section as indicated by eleven alloy and one cobalt pin readings. Cobalt pins removed from the other three capsules were measured on March 13, 1963 at Hanford. These results were converted to thermal neutron flux values by Phillips Petroleum Company personnel at NRTS. The results are summarized in Table 4.1.

TABLE 4.1

ROD POWERS FROM COBALT MONITORS

<u>GEH-20-</u> <u>No.</u>	<u>Fuel Section</u>	<u>In-Reactor</u> <u>Time (min)</u>	<u>Exposure</u> <u>(nvt x 10^{-16})</u>	<u>Thermal</u> <u>Flux (nv 10^{-14})</u>	<u>Rod Power</u> <u>(kw/ft)</u>
13	UO_2	3	2.69	1.46	18.3
13	(U, Pu) O_2	3	3.28	1.79	23.7
14	UO_2	5	4.51	1.47	18.5
14	(U, Pu) O_2	5	4.38	1.43	19.0
15	UO_2	15	13.7	1.49	18.7
15	(U, Pu) O_2	15	14.5	1.58	21.0
16	UO_2	60	lost	----	----
16	(U, Pu) O_2	60	51.9	1.41	18.7

The experiment design, based on a flux of 1.53×10^{14} nv, specified 19.2 kw/ft for the UO_2 sections and 20.3 kw/ft for the (U, Pu) O_2 sections.

-
1. Ceramics Research and Development Operation Quarterly Progress Report, October - December, 1962, HW-76300, p. 4.44.

Some of the problems associated with examining low exposure UO_2 - PuO_2 fuel specimens (i. e. , 10^{17} to 10^{18} fissions/cc) in HAPO facilities are being investigated by computer calculations. The expected β plus γ radiation level for a specific fuel rod (reference case: a single PRTR, Mark I UO_2 - PuO_2 rod) is being determined as a function of initial composition, exposure, time since discharge, and rod power prior to discharge. Postirradiation examination facilities in the Radiometallurgy Laboratory (327 Bldg.) and in the Plutonium Fabrication Pilot Plant (308 Bldg.) are shielded against penetrating radiation, but are not considered to be alpha boxes. Thus, alpha emitters must be associated with β or γ emitters for safe postirradiation examination. Ultimately, it may be possible with the cooperation of the appropriate groups to characterize irradiated samples as:

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2. Safe for 327 Building with specified in-out scheduling,
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4. Combination of (1), and (2) for use in 308 building after sufficient decay.

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