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RESEARCH
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REPORT

CERAMICS RESEARCH AND DEVELOPMENT
QUARTERLY REPORT
OCTOBER-DECEMBER, 1965

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PACIFIC NORTHWEST LABORATORY
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INTRODUCTION

The work reported in this and subsequent reports is a continuation of reactor fuels studies previously discussed in reports issued by the Fuels Development and the Plutonium Metallurgy Operations (prior to October 1962) and by Ceramics Research and Development Operation, General Electric Company, Richland, Washington. Recent reports in this series are:

HW-76300	(Unclassified)	October-December 1962
HW-76301	(Unclassified)	January-March 1963
HW-76302	(Unclassified)	April-June 1963
HW-76303	(Unclassified)	July-September 1963
HW-76304	(Unclassified)	October-December 1963
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HW-81600	(Unclassified)	January-March 1964
HW-81600A	(Confidential)	January-March 1964
HW-81601	(Unclassified)	April-June 1964
HW-81601A	(Confidential)	April-June 1964
HW-81602	(Unclassified)	July-September 1964
BNWL-91	(Unclassified)	January-March 1965
BNWL-150	(Unclassified)	April-June 1965
BNWL-198	(Unclassified)	July-September 1965

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Specifications were written to define the chemical and physical property requirements of mixed UO_2 - PuO_2 powders for densification by pneumatic impaction and use as fast reactor fuels.

Materials and Information Exchange - H. J. Anderson 1.26

UO₂ single crystals and pneumatically impacted UO₂-PuO₂ are being prepared to meet requests from foreign and domestic sites for use in basic studies and information exchanges. In addition, the US/Japan research newsletter number 4 was compiled and exchanged for the information of the co-operating agencies.

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- PRTR Fuel Element Internal Gas Pressure Measurement -
 D. R. Doman. 3.6
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- High Power Density Fuel Element Irradiations in PRTR -
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- Several important observations have been made during the preliminary postirradiation examination of UO_2 -4 wt% PuO_2 FE-6504.
- The fuel structures formed in the ruptured rod and the nondefected rods indicate that, for some as yet unknown reason, the ruptured rod operated at a higher fuel temperature than the adjacent nondefected rods. This fact could possibly be the most important single observation related to the rupture mechanism. It appears that the molten fuel may have been rapidly ejected from that portion of the fuel rod above the rupture, probably when the pressure tube failed. There is evidence of molten fuel contacting the cladding in the rupture region. Although the precise failure mechanism is still unknown, the rupture appears to be more directly related to the defect in the rod than to molten fuel operation per se.
- Molten-Core Fuel Capsules Test - F. E. Panisko, D. A. Bitz and L. A. Pember 3.25
- Four capsules (GEH-14-738 through -741) designed to demonstrate the stability of molten-core fuel element operation were shipped to Battelle-Northwest for examination following a successful irradiation in MTR.
- PRTR Failed Fuel Elements - F. E. Panisko 3.26
- Some of the discolored areas on the Zircaloy-2 cladding of fuel rods which failed in PRTR were metallurgically examined and found to contain zirconium hydride.
- Thermal Cycling of Defective Cladding - R. J. Lobsinger 3.26
- Capsule tests were performed in a steam autoclave to determine the propagation tendency of Zircaloy tubing cracks.
- PRTR-HPD Fuel Fabrication - R. L. Gulley and R. D. Reid. . . 3.27
- Since the start of fabrication on June 8, 1965, a total of 64 PRTR-HPD fuel elements have been fabricated.

PRTR Fuels - Quality Control - J. P. Keenan 3.27

Fuel performance through ultrasonic testing has been highly satisfactory this quarter.

A balance and scale calibration program was initiated.

Building Addition and Facilities Modification -

L. G. Merker 3.29

The Chemical and Materials Engineering Laboratory was accepted from the contractor on December 20, 1965.

The Plutonium Fuels Fabrication Building, Office Addition, will be ready for occupancy during January, 1966.

PART IV - JOINT FUEL DEVELOPMENT PROGRAMS

Irradiation Testing of EBWR Prototype Fuel Rods -

W. J. Bailey 4.1

Three capsules, still under irradiation, have achieved a maximum burnup of 20,050 MWd/MT of fuel (5×10^{20} fissions/cm³). Comparison of flux-wire data and isotopic analysis results indicates the former tends to underestimate actual fuel burnup.

SAP End Closures - Atomics International - R. F. Boolean 4.2

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Phoenix Fuels - J. P. Keenan and R. D. Reid 4.8

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Ultrasonic Examination of Saxton Tubing - R. J. Lobsinger . . . 4.8

An internal void was found during examination of Saxton tubing subjected to thermal cycling.

PART V - DEVELOPMENT OF ADVANCED FUEL CONCEPTS

Materials Joining Tantalum - R. F. Boolean 5.1

Thick walled tantalum tube was joined by magnetic force welding.

Metal Joining Studies - D. E. DeWitt, R. F. Klein, and R. C. Smith 5.1

Welding studies using the tungsten-inert-gas process (TIG) have been underway on Haynes-25, tantalum, molybdenum, and AISI 347 SS.

- Spheroidization of UO_2 - PuO_2 and PuO_2 - R. E. Bardsley. 5.1
- The agglomeration process is being investigated for producing UO_2 - PuO_2 and PuO_2 "spherical" particles. A glove box line was set up and several batches of PuO_2 processed.
- Coining of Cermet Extrusion Billets - E. A. Snajdr 5.6
- A preliminary study was initiated to investigate coining as a method for producing cermet extrusion billets.
- FFTF - Driver Fuel Evaluation - E. A. Snajdr 5.13
- An evaluation program was initiated to establish the physical properties of UO_2 -SS cermet fuel pins.
- Pressure Measurement of UO_2 Capsules at Elevated Temperatures - R. J. Lobsinger and D. R. Doman. 5.23
- Metallic cladding may have to be used for the experimental fuel in the HTLTR. Selection of a suitable material will depend in part upon its ability to withstand the buildup of internal pressure. Capsule tests were performed to measure internal pressure experimentally.
- HTLTR Driver Fuel Elements - R. E. Sharp and R. J. Lobsinger 5.24
- Procurement of both graphite cladding and 5% enriched UO_2 pellet has been initiated.
- Pneumatic Impaction of Tungsten Powders - P. L. Farnsworth . . 5.25
- Experiments to improve the strength and ductility of pneumatically impacted tungsten powders (-5 and -100 μ) by postimpaction heat treatments and by better control of vacuum during preimpaction heating indicated that atmospheric impurities (oxygen, nitrogen) picked up during preheating of the impaction cans may be responsible for the observed low strengths and absences of ductility.
- Fiber Reinforced Metal Matrix Composites - R. K. Robinson. . . 5.26
- Strengthening fuel element core and cladding materials is important to the development of efficient, economical and advanced fuel concepts. The physical properties of a number of fiber-reinforced metal matrix composites have been investigated.

- Tungsten Fiber Reinforced Nickel Composites - R. K. Robinson
and P. L. Farnsworth 5.28
- Significant increases in both the yield strength and the
ultimate tensile strength were obtained in the continuous,
axially aligned fiber reinforced nickel composites.
- "Whisker" Reinforced Metal Matrix Composites -
R. K. Robinson 5.31
- Single crystal aluminum oxides and silicon carbide "whisker"
reinforcements were added to metal matrices and tested.
- Randomly Oriented Fiber - Metal Composites -
R. K. Robinson 5.32
- Chopped, randomly oriented tungsten fibers did not signifi-
cantly improve the mechanical properties of metal matrix
composites.

PART I - CERAMIC FUEL RESEARCH STUDIES

Thermal Diffusivity of MTR-ETR Type Fuel Plates - J. L. Bates

The thermal diffusivity of aluminum clad MTR-ETR fuel plates* was determined from room temperature to 560 °C using a single pulse, transient method.^(1, 2)

Description of Fuel Plates

The fuel plates were approximately 0.13 cm thick (0.05 in.) with a 0.051 cm (0.020 in.) thick fuel core clad on both sides with 0.038 cm (0.015 in.) thick aluminum. A description of the fuel plates and core material are described in Table 1.1.

TABLE 1.1. Fuel Plates and Core Material Descriptions

Sample Number	Thickness		Fuel Core
	cm	in.	
P-1-1048	0.130	(0.051)	52.6 wt% UAl_3 + 0.19 wt% B_4C + X8001 Al
P-2-555	0.130	(0.051)	41.6 wt% U_3O_8 + 0.19 wt% B_4C + X8001 Al
O-4-833	0.128	(0.0505)	63.15 wt% Nb Coated UO_2 - ZrO_2 + X8001 Al
P-5-576	0.130	(0.051)	57.7 wt% UAl_3 + 0.19 wt% B_4C + X8001 Al
P-1-1047	0.132	(0.052)	52.6 wt% UAl_3 + 0.19 wt% B_4C + X8001 Al
79-11	0.082	(0.032)	50 wt% U + 3 wt% Sn + Al P. A APM-257

Samples for thermal diffusivity measurements were 0.635 cm (0.25 in.) in diameter. No changes were made in the thickness of plates. Some samples were coated with a water suspension of graphite to improve transient temperature measurements.

*Measurements were made for and samples were furnished by Phillips Petroleum Company, NRTS, Idaho Falls, Idaho, under related services Purchase Order AEC-74953.

1. Ceramics Research and Development Quarterly Report, April-June, 1965, BNWL-105, pp. 1.8-2.0, Battelle-Northwest, Richland, Washington.
2. W. J. Parker, R. J. Jenkins, C. P. Butler, and G. L. Abbot. "Flash Method of Determining Thermal Diffusivity, Heat Capacity, and Thermal Conductivity," J. Appl. Phys. vol. 32, no. 9, pp. 1679-84. 1961.

Thermal Diffusivity Apparatus

A single pulse, transient method⁽¹⁾ was used to measure the thermal diffusivity. The technique was similar to the "flash" method originally described by Park, et al.⁽²⁾ A laser produces the pulse of radiant energy to the specimen and the temperature transient on the back surface of the specimen was measured with solid state, infrared detectors.

The pulse was supplied by a ruby crystal [1.27 cm (0.5 in.) in diameter and 15.25 cm (6.0 in.) long with TIR end] laser. Measurements were made using pulse energies between 2 and 5 J and a wavelength of 6943 Å. The duration of the laser pulse was 0.54×10^{-3} sec.

Temperature transients on the back surface of the specimens were measured with a liquid nitrogen cooled, indium antimonide detector. The signal from the detector was displayed on a cathode ray oscilloscope and recorded on Polaroid film.

The wafer specimens were inserted into a lavite holder having a tapered center hole into which the specimens are lightly pressed. The holder was positioned in a tantalum tube which was heated by electrical resistance. Temperatures were measured by platinum versus Pt-Rh and chromel versus alumel thermocouples inserted into the specimen holder through a hole in the tantalum tube.

The atmosphere during the measurements was purified argon containing <1 ppm O_2 and <10 ppm H_2O .

Correction for Heat Losses and Finite Pulse Time

Corrections to the thermal diffusivity data for radiation losses and finite pulse time were made using methods of Cape and Lehman⁽³⁾ and Taylor, et al.⁽⁴⁾

References 1 and 2 - see previous page.

3. J. A. Cape and G. W. Lehman. "Temperature and Finite Pulse-Time Effects in the Flash Method for Measuring Thermal Diffusivity," J. Appl. Phys., vol. 34, no. 7, pp. 1909-13. 1963.
4. R. E. Taylor, J. A. Cape and M. M. Makata. Proceedings of the 4th Conference on Thermal Conductivity, October 13-16, 1965. USNRDL, San Francisco, V-C.

Because of the low temperatures $< 560^{\circ}\text{C}$, no radiation loss corrections were required. However, corrections were made for finite pulse time effect.

When the heat pulse (τ) is of infinitesimal duration, the thermal diffusivity (α) is given by

$$\alpha = 1.37 a^2 / \pi^2 t_{1/2};$$

where $t_{1/2}$ is the time required for the back surface to reach half the maximum temperature, a is the sample thickness and

$$t_c = \frac{a^2}{\pi^2 \alpha};$$

where t_c is the characteristic rise time of the back surface of the sample.

When τ is much smaller than t_c , $\frac{t_{1/2}}{t_c} = 1.37$. However, when $\tau \approx t_c$,

it is apparent that $t_{1/2}/t_c > 1.37$ and α should be corrected for the finite pulse time effect.

The shape of the pulse is important. Since the laser pulse more closely approximates a square wave, Taylor's method for finite pulse time correction was used. Estimated error for the thicker specimens (0.130 cm thick) was approximately 5% where $t_{1/2}/t_c = 1.44$.

Results

The results are summarized in Figures 1.1 and 1.2.

An estimate of the accuracy of the thermal diffusivity data was obtained using published thermal conductivity data for wrought aluminum alloys.

The thermal diffusivity is defined as

$$\alpha = k C_p^{-1} \rho^{-1},$$

where

k = thermal conductivity,

C_p = heat capacity, and

ρ = density

FIGURE 1.1 Thermal Diffusivity of Aluminum Clad MTR-ETR Fuel Plates

FIGURE 1.2 Thermal Diffusivity of Aluminum Clad MTR-ETR Fuel Plates

Assuming nominal values of 0.223 cal/g for C_p ; 2.7 g/cm³ for ρ and 1.2 W/cm °C for k for wrought aluminum at room temperature,

$$\alpha_{\text{cal}} = \frac{1.2 \text{ W/cm } ^\circ\text{C}}{0.223 \text{ cal/g/}^\circ\text{C} \times 4.186 \text{ W/cal/sec} \times 2.7 \text{ g/cm}^3}$$

$$\alpha_{\text{cal}} = 0.475 \text{ cm}^2/\text{sec}$$

Although higher than the experimental values, differences in density and heat capacity could explain these variations. It is interesting to note that the sample cores which contained a ceramic, (P-2-555 and P-4-833) exhibited higher thermal diffusivity values than the all metallic cores.

Electrical Conductivity of Uranium Dioxide - J. L. Bates

The electrical conductivity of nearly stoichiometric uranium dioxide has been measured between room temperature and 2710 °C by three different methods.

The first was a direct current, four probe apparatus utilizing a tungsten tube heater.⁽¹⁾ Electrical conductivity was measured between 575 and 1650 °C. The UO₂ specimens were approximately 2.5 to 3.0 cm long and 0.63 cm in diameter.

The second method used direct resistance heating of the UO₂.⁽¹⁾ Thin tungsten probes were clamped between the tungsten electrodes at each end of the 0.65 cm long and 0.35 cm diameter specimen. The specimen was heated to approximately 1500 °C with an auxiliary tungsten heater. Further heating was by direct resistance heating of the specimen. The maximum UO₂ surface temperature was 2710 °C. A significant temperature gradient existed within the specimen at the higher temperatures limiting the accuracy of the measurements.

The results obtained using these two methods have been reported previously.⁽¹⁾

1. Ceramics Research and Development Quarterly Report, April-June, 1965, BNWL-150. Pacific Northwest Laboratory, Battelle Memorial Institute, Richland, Washington pp. 1.21-1.23, 1965.

A four probe conductivity apparatus was used to determine the electrical conductivity of UO_2 from room temperature to 2300°C . Although the furnace is capable of reaching significantly higher temperatures, changes in specimen geometry occurred at the higher temperatures due to sample loss by sublimation.

The specimens were 0.63 cm in diameter and 1.90 cm long. Tungsten probes were set into small holes (spaced 0.63 cm apart). A tungsten holder held the specimen at each end. These holders were also used as the other electrical probes. The holder and sample were placed into a tungsten mesh cylindrical heating element which was heated by direct current from motor driven generators.

The dc potential across the sample was from a solid-state dc power supply. Voltages were low (less than 2 V, but generally less than 1 V) and a current of less than 100 mA was used. The applied dc potential was reversed as rapidly as possible, and the current-voltage relationship was measured with current flow in both directions.

Measurements were made in purified 8% hydrogen-argon at atmospheric pressure. The water and oxygen content of the gas was <10 ppm and <1 ppm, respectively. (Measurements were also made in purified argon, but since the data were not reproducible, only data obtained in 8% H_2 -argon are reported.)

Results

The electrical conductivity of four single crystals of UO_2 was measured. One specimen (MR-5) was measured by two techniques which required two different sized samples. The initially large specimen that used one, four-probe apparatus was reduced in size for measurements in the second four-probe apparatus. MR-1 and MR-5 were fabricated from the same large, UO_2 crystal boule.

The electrical conductivity values were not reproducible when measured in argon or during the initial measurements of a nonannealed UO_2 specimen. Annealing of a raw specimen was a prerequisite for reproducible data. For example, a specimen (MR-5) was annealed at

850 ± 15 °C in a reducing atmosphere of 8% H_2 -Ar. The electrical conductivity decreased with time as the O:U ratio decreased (Figure 1.3). After approximately 4.5 hr, the specimen was returned to room temperature and exposed to the atmosphere.

At the start of the succeeding heating cycle, the electrical conductivity had increased significantly over the UO_2 conductivity at the termination of the preceeding cycle presumable from a slight increase in O:U of the UO_2 . During the second cycle, the UO_2 conductivity decreased rapidly on further annealing. After approximately 6 hr, no further change was observed in the conductivity. The conductivity value after annealing was 1/5 that prior to the preannealed value.

The electrical conductivity of the UO_2 single crystal between room temperature and 2710 °C by all three methods are in good agreement (Figure 1.4). This was evident from the agreement between the data from specimens MR-1 and MR-5 and the data from MR-5 obtained by different techniques.

At temperatures above 1100 °C, the electrical conductivity of all samples was approximately the same. A plot of $\log \sigma$ versus $1/T$ above 1000 °C is not linear, the slope increasing with temperature. An estimate of the activation energy in this range gives a value between 1.8 and 1.6 which corresponds to a thermal energy gap between 3.6 and 3.2 eV.

The electrical conductivity at temperatures below 1000 °C varies between samples. A plot of $\log \sigma$ against $1/T$ should give a straight line. However above 400 °C, a decrease in slope is observed up to 1000 °C. The energy of the linear portion of the curves at lower temperatures is 0.23 and 0.19 eV.

Thermal Conductivity of $LiAlO_2$ - J. L. Bates and R. L. Gibby

The thermal diffusivity of $LiAlO_2$ (pneumatically impacted at 770 °K) was determined between 300 and 1200 °K using a transient technique. (1, 2)

1. Ceramics Research and Development Quarterly Report, April-June 1965, BNWL-150, pp. 1.8-2.0, Battelle-Northwest, Richland, Washington
2. W. J. C. Parker, R. J. Jenkins, C. P. Butler and G. L. Abbot. "Flash Method of Determining Thermal Diffusivity, Heat Capacity and Thermal Conductivity," J. Appl. Phys., vol 32, no. 9, pp. 1679-84, 1961.

FIGURE 1.3 Change in Electrical Conductivity of Single Crystal UO_2 (MR-5) During Heat Treatment in 8% H_2 -Ar at 1120°K (Specimen was cooled to room temperature, exposed to air and heated again to 1125°K , at times A and B.)

FIGURE 1.4 Electrical Conductivity of Single Crystal $\text{UO}_{2.001 \pm 0.0005}$

The thermal conductivity (k) was determined using the thermal diffusivity (α) data (Table 1.2) and the heat capacity (C_p) data reported by Christensen, et al,⁽¹⁾

$$k = C_p \rho \alpha$$

where ρ is the specimen density. The small expansion coefficient of LiAlO_2 ⁽²⁾ indicates that temperatures are not significant. The thermal conductivity of LiAlO_2 as a function of temperature is shown in Figure 1.5.

TABLE 1.2. Thermal Diffusivity And Thermal Conductivity Of LiAlO_2

Temperature °K	Thermal Diffusivity cm^2/sec	Temperature °K	Thermal Diffusivity cm^2/sec
RUN NO. 1		RUN NO. 2	
297	0.0124	297	0.0146
335	0.0125	297	0.0145
413	0.00967	347	0.0130
421	0.00928	457	0.00976
799	0.00579	459	0.00899
829	0.00543	567	0.00700
829	0.00543	673	0.00623
955	0.00519	765	0.00577
1147	0.00494		
1207	0.00499		
679	0.00683		
653	0.00693		
319	0.0141		
305	0.0153		

1. A. U. Christensen, K. C. Conway, and K. K. Kelly, "High Temperature Heat Contents and Entropies of Aluminates and Ferrites of Lithium and Sodium, and Lithium Titanate." U. S. Bur. Mines Report Investigation 5565, pp. 3-4, 1960.
2. F. A. Hummell, "Thermal Expansion Properties of Some Synthetic Lithia Minerals, J. Am. Ceramic Society, vol 34, no. 8, pp. 235-239. 1951

FIGURE 1.5. Thermal Conductivity of Lithium Aluminate Showing Increase in Low Temperature Conductivity After Heating

The data show a marked deviation from the normal $1/T$ relationship (Figure 1.6); the enhanced thermal conductivity above 800 °K presumably results from radiative heat transfer.

A slight hysteresis in thermal conductivity at the lower temperatures occurred during the heating cycles. This may have been caused by thermally induced compositional changes in the LiAlO_2 .

X-ray diffraction and Debye-Scherrer powder patterns of the impacted LiAlO_2 indicates the presence of $\alpha\text{-LiAlO}_2$ (low temperature form) in almost equal portions, (Table 1.3). Some of the weaker lines in the diffraction pattern correspond to the stronger lines of LiAl_5O_8 and $\alpha\text{-Al}_2\text{O}_3$. The phase composition of the pneumatically impacted material appears identical to the as-received powder. Thermal cycling the impacted material to 900 °C during

thermal conductivity measurements did not change the relative intensities of the diffraction lines of the various phases, indicating immeasurable changes in relative amounts of the phases.

Lithium aluminate powders impacted after a heat treatment of 1470 °K in a vacuum for 15 min contained only two phases: γ -LiAlO₂ and LiAl₅O₈. Diffraction lines, indexed as Al₂O₃ in the as-received material, disappeared after this heat treatment, indicating an Al₂O₃-LiAlO₂ reaction. The probable product is LiAl₅O₈.

FIGURE 1.6. Thermal Resistivity of LiAlO₂ Showing Deviations from 1/T Relationship at High Temperatures

TABLE 1.3. X-ray Diffraction Pattern of Pneumatically Impacted Lithium Aluminate

d	I/I ₀ (obs.)	Phase(1, 2, 3)	hkl	I/I ₀ (actual)
4.81	95	α -LiAlO ₂	---	90
4.03	92	γ -LiAlO ₂	101	100
3.91	20	?	---	---
3.69	11	γ -LiAlO ₂	110	14
3.51	9	LiAl ₅ O ₈	210	17
3.18	24	γ -LiAlO ₂	111	23
2.70	80	γ -LiAlO ₂	102	65
2.61	78	γ -LiAlO ₂	200	73
2.47	15	?	104	---
2.41	33	LiAl ₅ O ₈	311	100
2.18	10	γ -LiAlO ₂	211	8
2.10	18	α -Al ₂ O ₃	113	100
1.97	100	α -LiAlO ₂	---	100
1.87	20	α , γ -LiAlO ₂	212	15, 16
1.83	23	γ -AlO ₂	220	8
1.75	9	α -Al ₂ O ₃	---	45
1.64	13	γ -LiAlO ₂	301	10
1.61	20	α -LiAlO ₂ , α -Al ₂ O ₃	---	60, 80
1.57	27	γ -LiAlO ₂	004	14
1.54	11	LiAl ₅ O ₈	333	29
1.52	40	γ -LiAlO ₂	302	50
1.44	22	α -LiAlO ₂	114	70
1.41	43	α -LiAlO ₂	---	70
1.38	9	γ -Al ₂ O ₃	030	50
1.35	19	γ -LiAlO ₂	204	21
1.31	22	γ -LiAlO ₂	322	42
1.29	10	γ -LiAlO ₂	313	21
1.22	11	γ -LiAlO ₂	411	14
1.19	7	γ -LiAlO ₂	224	13
1.15	11	α -LiAlO ₂	420	55

- (1) F. A. Hummell. "Thermal Expansion Properties of Some Synthetic Lithia Minerals, J. Am. Ceramic Society, vol. 34, no. 8, pp. 235-239. 1951.
- (2) M. Marezio. "The Crystal Structure and Anomalous Dispersion of γ -LiAlO₂, " Acta Cryst., vol. 19, pp. 396-400. 1965.
- (3) Von Hans-Albert Lehmann and Heinz Hesselbarth, "Uber eine neue Modifikation des LiAlO₂, " Zeitschrift fur anorganische und allgemeine Chemie, 313, pp. 117-120.

Phase Studies - T. E. Bauer

Phase studies of PuO_{2-x} by means of reflection electron microscopy have been initiated. Unfortunately, the interpretation of early results was made difficult because of sample oxidation. Consequently, further experiments are planned with provision to reduce oxidation. In an attempt to realize this reduction, two improvements in experimental conditions have been made. First, the operating vacuum of the microscope column has been improved from 5×10^{-3} torr to around 10^{-4} torr. Also, provision has been made to allow a flow of hydrogen over the sample during the experiments.

Particle Crush Strength Measurements - J. B. Burnham

In order to obtain basic information on the forces needed to crush small diameter ceramic fuel particles, a tester was fabricated as shown in Figure 1.7. The system is an adaptation of a Mound Laboratory design and employs a pneumatic-hydraulic force system to deliver a smooth, variable rate loading. The force necessary to crush a selected fuel particle is measured by a load-cell connected to a recording oscillograph. By varying the load-cell excitation voltage and the attenuation factor of the recording oscillograph, the full scale range of the system can be continuously varied from 100 to 2200 g. Both loading and recording devices are actuated by a foot switch, which leaves both hands free for loading particles and cleaning the anvil. Eighty or more particles per hour can be tested.

Since the crush strength of the particles from a single batch can be expected to vary a great deal, a statistical number must be tested in order to obtain any meaningful results. Whenever possible, at least 50 particles in a narrow size range ($-80 + 100$ mesh) are crushed. The mean value is used as the crush strength of the batch. The coefficient of variation (standard deviation divided by mean) is an indication of the regularity of the particles.

FIGURE 1.7 Pneumatic-Hydraulic Load System for Measuring the Crushing Strength of Ceramic Fuel Particles (The electrical lead carries the output of the fully enclosed load-cell to a recording oscillograph.)

Some of the results obtained to date are listed below: (Table 1.4)

Sample Description	Density (% TD)	Mean Crush Strength, g	Standard Deviation, g	Coeff. of Variation
UO ₂ , agglomerated, sintered 1750 °C	76*	482	118	0.245
UO ₂ -0.1 TiO ₂ agglom., sintered 1750 °C	90*	420	95	0.226
UO ₂ particles from 3M	86	565	103	0.182
UO ₂ , dynapaked	97	660	125	0.189
UO ₂ , dynapaked	97	604	104	0.172
PuO ₂ , S-1, dynapaked	95	880	125	0.142
PuO ₂ , S-2, dynapaked	95.5	885	127	0.144
PuO ₂ , plasma fused microspheres	77**	180	58	0.322

*Density measured on large pieces, may not be characteristic of the small particles.

** Microspheres, mostly hollow.

Several conclusions can be drawn from this data: (1) PuO₂ impacted under the same conditions as UO₂ is significantly stronger; (2) strength, as expected, increases with density; (3) hollow microspheres would be the least satisfactory particles (of those tested) for a cermet; (4) the method of determining particle density by measuring the density of large agglomerates from the same batch may not be accurate.

High Frequency Induction Heating of Oxides - J. A. Christensen

Current efforts to correlate oxide fuel heat transfer properties with stoichiometry use in-reactor melt radii as principal temperature indicators. This requires a knowledge of the melting point of the material in question, superstoichiometric UO_{2+x}. These data do not presently exist because of experimental difficulties associated with measuring temperatures in excess of 2500 °C in oxidizing atmospheres. A high frequency (10 m Hz) oxide susceptor induction furnace is being assembled to provide the required experimental environment.

Final design of the furnace was formulated. The susceptor, a 0.95 cm ID ThO_2 tube which is surrounded by packed ThO_2 powder used for thermal insulation, is supported vertically by a quartz tube about which the primary copper core is wound. All the principle components were procured from existing stock and are being assembled. A motor driven movable hearth mechanism was also assembled and attached to the furnace enclosure.

Stoichiometry Gradients in Oxide Fuels - J. A. Christensen, G. R. Horn, and R. R. Lewis

Analytical studies indicate that radial stoichiometry gradients established during irradiation can account for the discrepancy between out-of-reactor measurements of UO_2 thermal conductivity and the lower conductivity values deduced from observations of melt and grain growth radii in irradiated fuel rods.⁽¹⁾ Most simply stated, oxygen probably diffuses down the potential gradient (thermal or electrical) and decreases the thermal conductivity in the cooler fuel regions where the O/U ratio appreciably exceeds 2.00.

The current program is directed toward measuring the oxygen concentration gradients established in UO_2 fuels of various average stoichiometries during irradiation. The results will be correlated with existing data on variation in thermal conductivity with O/U ratio to yield heat transfer parameters which vary with both temperature and bulk fuel stoichiometry. This will be accomplished by radially sampling 3.49 cm diameter UO_{2+x} fuel cores irradiated for short periods and correlating sample analyses with position and temperature in the fuel.

Four capsules, containing $\text{UO}_{1.99}$, $\text{UO}_{2.02}$, $\text{UO}_{2.05}$, $\text{UO}_{2.15}$, are being assembled. The basic fuel material was fabricated by high energy compaction of UO_2 and U_4O_9 blended to these compositions. Blended fuels were loaded into specially designed and procured stainless steel can assemblies preparatory to pneumatic impaction of the integrally clad fuel cores.

Sampling of a $\text{UO}_{2.00}$ fuel core previously irradiated at 30 kW/ft was accomplished. Diamond core drills were used to remove 0.47 cm diameter x 3.3 cm long samples at various radial positions (Figure 1.8). These relatively large samples are required for O/U ratio analyses. Cores were submitted to the Remote Analytical Laboratory.

1. J. A. Christensen, A.N.S. Trans., vol. 7, no. 2, pp. 392, Paper 7.

FIGURE 1.8. Excess Oxygen Distribution in Nominally UO₂ Pellets Irradiated at 30 kW/ft. (Numbers within circles are O/U ratio measured by controlled potential coulometry for that location.)

Attempts to qualitatively discern fuel stoichiometry gradients by radially streak testing irradiated UO₂ fuel cross sections are also in progress. Rotating such a cross section against a porous porcelain surface causes some of the oxide to be abraded and smeared onto the porcelain. The color of this smear or "streak" is indicative of the oxygen concentration in the UO₂. A variation in color across the circular streak indicates a radial oxygen concentration variation in the fuel. Initial efforts were frustrated by the inability to achieve parallel fuel and porcelain surfaces.

Actinide Oxide Phase Studies - T. D. Chikalla

The study of the oxygen dissociation pressures over americium sub-oxides and phase relations in the Am-O system is nearly complete. Nonstoichiometric compositions have been determined by the measurement of weight change at constant pressure and temperature. Early in the work it had been found that AmO_2 reacted with the platinum container under strongly reducing conditions; it was compatible with iridium under these same conditions. Consequently, all dissociation pressure measurements were made using an iridium container. However, at high oxygen pressures (> 30 mm Hg) and high temperatures ($> 1100^\circ\text{C}$) it was found that iridium reacted with oxygen to form a volatile oxide. A platinum crucible was used for the completion of the measurements at these high pressures. Isothermal $\log p_{\text{O}_2}$ /composition plots made both for oxygen physically added to the system (pressures > 30 mm Hg) and for oxygen supplied by the dissociation of cupric oxide ($p_{\text{O}_2} < 30$ mm Hg) were in good agreement for mutual composition areas. Heat and entropy of solution values were also in accord with earlier results. The total pressure range investigated now extends from below 10^{-6} atm to one atm O_2 . AmO_2 is quite unstable (relative to PuO_2) and at 1175°C and one atm O_2 , reduction to $\text{AmO}_{1.98}$ occurs.

The effects of buoyancy and thermal convection on the observed weight changes were determined as a function of pressure and temperature by measuring the weight change of a sample of high purity ThO_2 under various conditions of temperature and pressure. The sample had been degassed overnight at 800°C to ensure removal of sorbed gases and/or volatile impurities. The apparent weight changes, due to the combined effects of buoyancy and thermal convection on the sample container, amounted to 0.5 to 1.0 mg and necessitate correcting all of the results previously obtained.

Quenched and annealed americium oxide compositions in the interval $1.5 < \text{O}/\text{Am} < 2.0$ have been examined by X-ray diffraction. The phases observed after various thermal treatments are summarized in Figure 1.9. Over the range $1.72 < \text{O}/\text{Am} < 2.00$ the system consists of cubic Am_2O_3 and AmO_2 . The lattice parameter of the AmO_2 phase in this region has been constant in all the samples investigated ($5.378 \pm 0.002 \text{ \AA}$) and agrees with the lattice parameter obtained for pure $\text{AmO}_{2.00}$. This implies that at room

temperature the dioxide does not exist with an appreciable oxygen deficiency. Cubic Am_2O_3 may dissolve excess oxygen and it exists over a rather broad range, $1.5_1 < \text{O}/\text{Am} < 1.7_2$. The lattice parameter of this phase nearest stoichiometry is $11.02 \pm 0.01 \text{ \AA}$. Hexagonal Am_2O_3 was retained by quenching from 1000°C in hydrogen while the cubic form was retained by quenching from 450°C .

FIGURE 1.9. Phases Observed at Room Temperature After Annealing or Quenching from the Temperatures Indicated

The shape of the dissociation pressure isotherms suggested the existence of a single phase above 850 °C and at O/Am >1.80. It was speculated earlier that this was a fluorite phase containing oxygen vacancies. However, it proved impossible to retain such a phase by quenching. This is probably indicative of a very high anion mobility and is similar to that found in the Pu-O and Ce-O systems.

Guinier patterns were run on many suboxide compositions in an effort to determine whether there exists a homologous series of intermediate phases such as reported for the Ce-, Pr-, and Tb-oxygen systems. No evidence for such phases was found. Several samples are currently undergoing a long-time anneal in an additional effort to nucleate such phases.

Substoichiometric Mixed Oxides of Plutonium and Uranium - R. L. Gibby

Substoichiometry on in-reactor fuel behavior, particularly in a mixed plutonium-uranium system, has not been extensively investigated. In-reactor experiments with urania-filled fuel rods have indicated that reduced oxygen contents enhance thermal conductivity. Additional laboratory experiments suggest that other thermal properties and also chemical and physical properties can be improved through reduction of oxygen content to substoichiometric portions.

Substoichiometric mixed plutonium-uranium oxides are being synthesized for investigations of the effects of stoichiometry on their thermal, physical, and chemical properties. Pellets, compounded from mixtures of UO_2 , PuO_2 , and PuH_3 , were formed by dry pressing powders which had been ground and mixed in a ball mill for 24 hr in a N_2 atmosphere. The mixtures were compounded to give sintered products of $\text{U}_{0.8}\text{Pu}_{0.2}\text{O}_{1.75}$ and $\text{U}_{0.8}\text{Pu}_{0.2}\text{O}_{1.80}$. In the first set (O/M = 1.75), the pellets were sintered in a N_2 -5 vol% H_2 atmosphere at 1700 °C for 2 hr. The second set (O/M = 1.80) was sintered for 4 hr at 1600 °C in an Ar-5 vol% H_2 atmosphere.

Following sintering, densities were determined for each pellet, and one pellet from each set was prepared for ceramography and autoradiography. Guinier X-ray powder photographs were taken of one representative sample from each set. Oxygen-metal ratios were not determined on the

sintered pellets; however, oxidation analysis of the ball milled powder used in the first group of pellets indicated that considerable oxidation of the PuH_3 occurred during grinding.

The oxygen content of the nitrogen in the hood was low, but no determination was made of the moisture content. Photomicrographs of both samples indicated the presence of three or more phases: the major phase of uranium-plutonium oxide, unidentified dark phases, and an unidentified finely dispersed metallic-appearing phase (Figure 1.10). The Guinier powder photographs contained only UO_2 lines. Autoradiography indicated a good plutonium distribution but did not suggest good solid solution formation (Figure 1.11). Furthermore, the plutonium-bearing phase was quite coarse in size, denoting insufficient ball milling.

45128B 500X
Sintered 1700 °C 2 hr.
(O/M=1.75)

45129F 500X
Sintered 1600 °C 4 hr.
(O/M=1.80)

FIGURE 1.10. Microstructure of Sintered 20 wt% PuO_2 -80 wt% UO_2 Pellets. Plutonium hydride substituted for PuO_2 to vary stoichiometry. Oxygen-to-metal ratio indicates as-blended stoichiometry.

4652362 10X
Sintered 1700 °C 2 hr.
(O/M=1.75)

466182 10X
Sintered 1600 °C 4 hr.
(O/M=1.80)

FIGURE 1.11 Autoradiographs of Sintered 20 wt% PuO₂-80 wt% UO₂ Pellets

The pellets, under the optical microscope, showed some porosity. Density measurements confirmed this observation; the density values were in the range: 85 to 88% TD, the higher values occurring for the Ar-H₂ sintered pellets.

Work Function Measurements - C. E. McNeilly

A third series of measurements of the work function of high-purity, polycrystalline tantalum has been completed in the retarding-potential-technique apparatus. A value of 4.10 eV was obtained; this compares well with the values of 4.282 and 4.082 eV obtained earlier.

A pumpdown and bakeout cycle has been initiated on the apparatus; a small wafer of sintered ZrC (~3/mm x 3/mm x 0.25/mm) was placed on the emitter. The work function of the ZrC will be measured and compared to values reported in the literature. If it appears that this technique (loose sample supported on separate heater) is capable of accurate work function measurements, then the apparatus will be hooded in preparation

for measuring the work functions of plutonium compounds. A schematic diagram of the apparatus is shown in Figure 1.12.

FIGURE 1.12. Thermionic Emission Apparatus

Differential Thermal Analysis - C. E. McNeilly

Several operational problems have been encountered with the present differential thermal analysis (DTA) apparatus, and a new furnace and sample support have been constructed.

The main feature of the new apparatus is the incorporation of a low thermal-mass, water-cooled furnace and base plate to reduce temperature lags between the sample support and the programmer, particularly at low temperature. This furnace and sample support have been inverted in the new design. The furnace is now located over the sample; formerly, the sample was suspended in the furnace. This should result in much better atmosphere control and reduce sample oxidation.

The other major change involves the use of a larger diameter furnace to even-out temperature gradients in the hot zone and reduce the amount of DT base line drifting. The new furnace has a design temperature capability of 1800 °C.

Thermal Expansion of Plutonium Fuels - R. P. Nelson

The high temperature vacuum dilatometer was modified to increase both the maximum temperature limit and the precision of the instrument. The water cooling system was improved to reduce errors resulting from differential expansion outside the furnace. A tungsten sample holder was fabricated to replace the alumina holder, thereby increasing the accuracy of measurement on a wider range of sample materials and also increasing the temperature limit from 1550 °C to approximately 1800 °C.

Although errors resulting from temperature differentials have not been completely eliminated, accurate expansivities may be obtained by applying small corrections to measured values. Initial measurements using the tungsten sample holder are currently in progress on substoichiometric niobium carbide.

A request has been submitted to the National Bureau of Standards for standard samples to use for calibration of the tungsten sample holder.

Refractory Compounds Research Facility - E. T. Weber

Over the past year, the plutonium ceramics research program has tended toward increased emphasis on the environment-sensitive nitride and substoichiometric oxide compounds. In conjunction with this work, a new glove box facility has been designed, fabricated, and installed. This facility provides work space for high temperature synthesis studies and investigations of physico-chemical stabilities under controlled atmosphere.

A nitrogen atmosphere containing less than 0.3 vol% oxygen and less than 100 ppm water can be maintained in the 76 ft² stainless-steel enclosure. Special shock mounts isolate the glove box from building vibrations; all water, nitrogen, and other service lines are vibration isolated from the glove box. Reactive and inert gases can be introduced to apparatus in the hood through a stainless steel gas system. Four high-vacuum systems

(capable of operation in the 10^{-5} - 10^{-6} torr range) are mounted on the hood for use with research apparatus. Two vacuum-storage bell jars for maintaining sample material integrity over extended periods are provided.

Equipment presently in operation within the enclosure (shown in Figure 1.13) includes: an arc-fusion apparatus used for synthesis and melting of refractory compounds; a tungsten filament or tungsten mesh-element furnace which operates to 3000 °C in inert atmospheres or vacuum; a furnace for high temperature (now: 1000 °C; soon: 1800 °C) heat treatments in high vacuum, static atmospheres, or flowing gas streams; and an analytical balance.

High pressure gas lines, control and safety devices, and a special venting system have been provided for, a high pressure (2000 psi), high temperature (3000 °C) filament furnace yet to be installed. This unit will be used for studies of phase relationships and chemical stabilities as functions of pressure and temperature.

Specifications for Nuclear Fast Reactor Grade Mixed Oxides: Compactible Powder - H. J. Anderson

Proposed specifications were written to define the chemical and physical characteristics and some process requirements of mixed oxides (UO_2 - PuO_2) for use as fuel in fast reactor fuel elements. Specifications were documented as BNWL-CC-319.

Materials and Information Exchange - H. J. Anderson

UO_2 single crystals and pneumatically impacted UO_2 - PuO_2 and PuO_2 materials are being prepared and characterized before shipment to meet requests from foreign and domestic sites for use in basic studies and information exchanges. Current requestors for UO_2 single crystals include: University of California (LASL), Cornell University, Pennsylvania State University, Arizona State University, IBM Research Center, ORNL, Harwell, Japan, and the Hebrew University of Jerusalem, Israel.

FIGURE 1.13 Interior View of the New Refractory Compounds Research Facility [Present Equipment Includes a Vacuum-Atmosphere Muffle Furnace (Bottom), an Analytical Balance (Center, Left), a High Temperature Filament Furnace (Center, Right), Vacuum-Storage Bell Jars (Upper, Left), and an Arc-Fusion Apparatus (Upper, Right)]

To date, more than 380 single crystal UO_2 specimens of various geometries have been prepared and exchanged with domestic and foreign research investigators.⁽¹⁾

Approximately 800 g of 15 wt% PuO_2 - UO_2 (93% enriched) was pneumatically impacted, characterized and shipped in response to a request from AERE-Dounreay. Rocky Flats, Dow Chemical, has requested about 200 g of pneumatically impacted PuO_2 for use in basic studies and information exchanges.

In addition, the US/Japan Ceramic Nuclear Fuels Newsletter, Number 4 was compiled and exchanged under terms of the cooperative US/Japan agreement for the information of the cooperating agencies. The Japan/US Newsletter was received, reproduced, and is being distributed.

1. Ceramics Research and Development Operation Quarterly Report, April - June 1965, BNWL-150, pp. 1.26-1.28, Pacific Northwest Laboratory, Richland, Washington.

PART II - IRRADIATION BEHAVIOR OF CERAMIC FUELS

Irradiation Performance of Fast Reactor Fuel Candidates - UN-20 wt% PuN

J. A. Christensen

Postirradiation examination of UN-20 wt% PuN fuels irradiated to 2.4, 6.0, and 13.0×10^{20} f/cm³ at 2130 W/cm (linear) was continued. Radial drillings from specimen GEH-14-616 (2.4×10^{20} f/cm³) were analyzed for various fission fragments and for plutonium. Results show that significant plutonium redistribution occurred during irradiation (Figure 2.1). A slight decrease in plutonium concentration near the fuel periphery was anticipated because of radial flux depression. This was borne out by the approximately 30% change in concentration across the outer third of the fuel. Contrary to expectations, however, plutonium concentration in the center third of the initially homogeneous fuel was a factor of four to five times less than in the outer two thirds. The major plutonium depletion is associated with the oxide and metal phases which formed near the fuel center; however an appreciable gradient also exists in the surrounding parent mixed nitride. The mechanism whereby the plutonium relocates is uncertain, but may reflect the high dissociation pressure of PuN relative to UN. If plutonium is preferentially precipitated as a product of the dissociation reaction, a plutonium concentration gradient parallel to and in the same direction as the thermal gradient will arise. Dissolved oxygen should react with much of the precipitated plutonium without appreciable plutonium redistribution because of the anticipated high mobility of oxygen relative to plutonium. This is the most likely source of the annular oxide ring near the fuel center. Excess plutonium will then tend to diffuse down the metal concentration gradient toward the fuel exterior. Although this mechanism is speculative, it seems feasible and, if correct, may portend serious, intrinsic problems for nitride fuels.

Analyses for the fission fragments Ce-Pr¹⁴⁴, Zr-Nb⁹⁵, Sr⁹⁰, Ru¹⁰⁶, and Cs¹³⁷ were also performed. With the exception of zirconium, all of these redistributed appreciably during irradiation (Figures 2.2 through 2.7). Cerium and strontium migrated to the outer-most (thus, coolest) regions of the fuel, while cerium and ruthenium tended to concentrate at the fuel center. Nearly all of the ruthenium detected was associated with the central metal

Scale Refers to Following
Six Figures

β - α Autoradiograph

FIGURE 2.1 UN-20 wt% PuN Irradiated to 2.4×10^{20} f/cm³ at 2130 W/cm. Lower Macrograph Shows Location of Samples Analyzed for Plutonium and Fission Fragment Concentration. Analytical Results are Presented in the Following Six Figures.

FIGURE 2.3 Zr-Nb⁹⁵ Distribution in UN-20 wt% PuN Irradiated to $2.4 \times 10^{20} \text{ f/cm}^3$ at 2130 W/cm

FIGURE 2.2 Plutonium Distribution in UN-20 wt% PuN Irradiated to $2.4 \times 10^{20} \text{ f/cm}^3$ at 2130 W/cm

FIGURE 2.4 Ce-Pr¹⁴⁴ Distribution in UN-20 wt% PuN Irradiated to 2.4×10^{20} f/cm³ at 2130 W/cm

FIGURE 2.5 Ru¹⁰⁶ Distribution in UN-20 wt% PuN Irradiated to 2.4×10^{20} f/cm³ at 2130 W/cm

FIGURE 2.6 Cs^{137} Distribution in UN-20 wt% PuN Irradiated to $2.4 \times 10^{20} \text{ f/cm}^3$ at 2130 W/cm

FIGURE 2.7 Strontium Distribution in UN-20 wt% PuN Irradiated to $2.4 \times 10^{20} \text{ f/cm}^3$ at 2130 W/cm

phase which, as discussed previously, appears to consist principally of fission fragment metals.

Analyses of radial drillings from the highest burnup specimen (13×10^{20} f/cm³) are in progress. To date, the only results available are the average plutonium isotope distribution data shown in Table 2.1. Results for the lower burnup specimens are also shown in Table 2.1.

TABLE 2.1 Irradiation Effect on Plutonium Isotope Distribution in UN-20 wt% PuN

Specimen	Exposure		MTR Cycles	Isotope Concentration (%)			
	fissions/cm ³	nvt (thermal)		Pu ²³⁹	Pu ²⁴⁰	Pu ²⁴¹	Pu ²⁴²
Nonirradiated	--	--	--	90.46	8.18	1.31	0.05
GEH-14-616	2.4×10^{20}	5.3×10^{20}	3	87.25	11.40	1.28	0.08
GEH-14-617	6.0×10^{20}	14.6×10^{20}	6	81.08	16.68	2.04	0.20
GEH-14-618	13×10^{20}	34.8×10^{20}	18	68.78	26.41	4.05	0.76

Plutonium Ceramics Irradiations - J. A. Christensen

Irradiation of wafer specimens of various plutonium compounds continued. Status of specimens currently in-reactor is summarized in Table 2.2.

TABLE 2.2 Current Plutonium Ceramics Irradiations

GEH-14	Material	Goal	Exposure as of 12/1/65 Fissions/cm ³ x 10 ⁻²⁰	Discharge Date
405	PuN, impacted	172.5	106.0	5/67
407	PuN, impacted	77.5	47.4	5/67
409	PuC, arc cast	150.0	81.5	1/68
411	PuO ₂ , 95% TD sintered	166.0	110.0	1/67
413	Pu ₂ O ₃ , 95% TD sintered	116.0	78.5	1/67
538	PuN-50 vol% W, impacted	124.0	66.0	8/67
540	PuN-50 vol% W, impacted	62.5	37.3	8/67
549	PuN-24 vol% Pu, arc cast	67.0	37.7	8/67
551	PuN-15 vol% Pu, arc cast	32.0	17.2	8/67

Diffusion Test Element Irradiations - M. K. Millhollen and L. A. Pember

The second diffusion test element (GEH-4-117) has now been examined exactly as was the first element.⁽¹⁾ This element was a four-rod cluster of Zircaloy-2 clad sintered oxide pellets. Fuel length in each rod was 40 cm. The test rod examined contained UO_2 -2 wt% PuO_2 pellets interspersed between 1-inch long increments of 2.8 wt% U^{235} enriched UO_2 pellets. Nominal cladding OD was 1.5 cm with a 0.76 mm wall. Pellets were ground to a diameter of 1.3 cm.

The test rod was removed from the element (Figure 2.8) and was sectioned through a UO_2 -2 wt% PuO_2 pellet which had been located in the maximum flux zone. In addition to the transverse section, a 5 cm long longitudinal section was taken from the rod directly above the transverse section. Each of the two specimens was polished and examined visually after which macrographs, beta-gamma autoradiographs, and alpha autoradiographs were made (Figures 2.9 and 2.10).

FIGURE 2.8 Diffusion Test Element GEH-4-117 After Irradiation

(1) Ceramic Research and Development Quarterly Report, July-September, 1965, BNWL-198

Alpha Autoradiograph

Beta-Gamma Autoradiograph

Macrograph

FIGURE 2.9 Transverse Cross Section Through CO_2 - 2 wt% PuO_2 Pellet (GEH-4-117)

FIGURE 2.10 Longitudinal Cross Section (GEH-4-117)

4X

This test element was irradiated for 34.3 effective full power days in the B-3 position in the MTR. Calculations based on flux monitors indicate that a maximum surface heat flux of $\sim 160 \text{ W/cm}^2$ ($\sim 500,000 \text{ Btu/hr-ft}^2$) was reached in the $\text{UO}_2 - 2 \text{ wt\% PuO}_2$ pellets examined. The macrographs in Figures 2.9 and 2.10 show no evidence of central melting although normal central voids were formed.

The beta-gamma autoradiographs of the samples showed normal fission product distribution patterns. The slightly cooler operation of the 2.8 wt% U-235 enriched UO_2 pellets was evident in the beta-gamma autoradiograph fission product distribution patterns in Figure 2.10. The alpha autoradiograph of the longitudinal section shows that very limited PuO_2 migration occurred along long columnar grains. There is also some slight evidence of vapor phase deposition of PuO_2 on the cooler cavity surfaces of the 2.8 wt% U²³⁵ enriched UO_2 pellets. No significant migration of PuO_2 occurred under the test conditions imposed.

Defect Fuel Element Test - F. E. Panisko and L. A. Pember

In order to study the irradiation behavior of a defective fuel rod operating with a molten core, a seven-rod cluster was irradiated in the ETR. Calorimetric data indicated that the power generation of the element was insufficient to meet the test objectives so the test was discharged until the following ETR cycle.

The test element (GEH-12-39) contained one fuel rod with UO_2 (5% U²³⁵) and six rods with $\text{UO}_2 - 3 \text{ wt\% PuO}_2$. A 0.16-inch diameter hole was drilled through the cladding in the fuel region of the UO_2 rod. Each rod contained a 75 cm fuel column including a 1.0 cm long depleted UO_2 pellet at both ends. A 8.9 cm long gas plenum was provided at the top end, and the cladding in this region was internally supported by an Inconel spring. A photograph of the fuel element and stainless steel basket is shown in Figure 2.11. The defected UO_2 fuel rod is scheduled to operate at a power generation of approximately 914 W/cm at the defect.

FIGURE 2.11 Photograph of Defective Fuel Element (GEH-12-30) and Stainless Steel Basket.
Top Photograph Shows a Close-up of the 1/16-inch Diameter Hole Through the Zr-2 Cladding.

PART III. PRTR FUEL ELEMENT PERFORMANCE AND FABRICATION

High Pressure Encapsulation - R. F. Boolen

A method to effect high integrity seals on capsules containing gas to pressures of 102 atm was investigated and applied in fabrication of TREAT (Transient Reactor Experiment) capsules

During the initial study, two approaches were used to effect the seal on 0.32 cm x 0.79 mm wall helium pressurized zircaloy tubes.

The first approach consisted of making a temporary mechanical seal by compressing the tube wall onto a specially designed mandrel with "swageloks", cutting off the feed end of the tube, and forming a fusion weld as the final seal. This approach was successful, however, satisfactory closures could only be made by using a fine finish on the tube ID and on the mandrel.

The second approach consisted of mashing the pressurized tube flat with spot welding electrodes and effecting a mash-spot weld, in a single operation, as the final closure. This approach was successful and offers significant advantages over the first. In addition to the less stringent requirements on tube ID finish (no mandrel is used for this method), mash-spot welding requires less stringent atmosphere (inert gas) control. A purge of inert gas is sufficient to guard the localized heated area from contamination, whereas in arc welding the final closure must be made in a high purity inert atmosphere (eg. a welding box).

For these reasons, the mash-spot resistance weld method was applied to four TREAT capsules, typified by one shown in Figure 3.1. These capsules were filled with enriched UO_2 after one end was closed by usual methods (TIG-box), common to PRTR fabrication. The second end cap was a thin tube with a special high pressure fitting on one end and the standard cap on the other. Following pressurization to 102 atm He, three mash-spot welds were made on each tube (starting nearest the capsule). Each weld was checked immediately after welding by immersion in water. Since optimum welding parameters were not determined prior to welding (due to lack of time)

FIGURE 3.1 TREAT Capsule - Ready for Mash-Spot Welding

some leakage, as expected, was detected after the first and second welds. All welds were made using the 200 kVA combination spot and seam welder shown in Figure 3.2. Helium leakage from the total surface of all four capsules was determined to be approximately 4×10^{-6} atm cm³/sec (detection level 3×10^{-7} atm cm³/sec).

The electrodes were fabricated from RWMA Class 12 resistance welding alloy (a hard tungsten-copper), and are shown in Figure 3.3.

Typical mash-spot welds are shown in Figure 3.4.

This method should be applicable to many types of encapsulation in which the gas or pressure (or both) make fusion welding impractical.

A special plexiglas tube served the dual purpose of introducing inert gas (at one end) to purge the weld area, and reducing the danger of injury or contamination to the welder in the case of a leak or rupture.

FIGURE 3.2 200 kVA Combination Spot and Seam Welder

FIGURE 3.3 Electrodes Used in Mash-Spot Welding

466200

6X

FIGURE 3.4 Mash-Spot Welds on TREAT Capsule Endcap

TREAT Irradiations in Support of the PRTR High Power Density Core Experiment - M. D. Freshley

A joint program has been initiated between Battelle-Northwest and Argonne National Laboratory to investigate the transient irradiation behavior of vibrationally-compacted UO_2 capsules in TREAT. The irradiations will furnish information of interest to the Reactor Safety Program at ANL as well as needed data directly related to the safeguards analysis of molten UO_2 - PuO_2 fuel element irradiations. The eight capsules were fabricated by BNW for irradiation in TREAT by ANL.

The 1.44-cm diameter Zircaloy-2 clad capsules contain a vibrationally-compacted impacted UO_2 (81-83% TD) fuel column 10.7-cm long capped on each end by a depleted UO_2 pellet 1.0-cm long. The UO_2 fuel contains 15% U^{235} . There is a 5.0-cm long gas plenum at one end of the fuel column to simulate the PRTR fuel design.

Two transient irradiations, each involving four capsules, will be performed in TREAT during the first part of January 1966. The four capsules included in one of the transient irradiations contain He at atmospheric pressure.

The four capsules in the second irradiation are pressurized with He at 102 atm to approximate conditions that exist in a high burnup fuel element. The capsules will be contained in a water-filled pressure vessel and subjected to a transient burst of neutrons in TREAT. A transient energy of 460 cal/gm of UO_2 will guarantee melting of the UO_2 , reaction with the Zircaloy cladding, and fragmentation of the capsules. From past experience Argonne National Laboratory has found that 280 cal/g melts UO_2 .

After irradiation, the hydrogen evolved (measured by mass spectrometer) provides a means of computing the extent of clad-water and/or UO_2 -water reaction which occurred. The particle size distribution of the residue from the fragmented capsules will indicate the nature of the UO_2 -water reaction. These data will permit evaluation of the consequences of injecting molten UO_2 - PuO_2 fuel into the water coolant in the event of a catastrophic fuel element failure.

PRTR Fuel Element Internal Gas Pressure Measurement - D. R. Doman

Knowledge of operating fuel rod gas pressures will permit better evaluation of the performance of fuel elements irradiated during the Batch Core Experiment. To determine pressures as well as temperatures in the gas plenums, two rods in each of two UO_2 - 2 wt% PuO_2 and one ThO_2 - 5 wt% PuO_2 Mark I-R fuel elements have been instrumented. Pressure measurement is accomplished by applying a controlled gas pressure on one side of a diaphragm to equalize the fuel rod released gas pressure; balance being indicated by opening an electrical contact.

The reactor nozzle lead-out for this test has been redesigned to eliminate shortening the nozzle. This has resulted in considerable cost savings.

High Power Density Fuel Element Irradiations in PRTR - M. D. Freshley and F. E. Panisko

The fuel element testing program in PRTR has been expanded to include an evaluation of the irradiation performance of vibrationally-compacted impacted mixed oxide fuels operating at high specific powers. One of the major fuel testing objectives is to determine if there are factors that limit operation of plutonium-bearing oxide fuels to powers less than those attainable with uranium-only oxide fuels.

HPD elements were irradiated in the PRTR core under the maximum operating conditions expected during the Batch Core Experiment, i.e., maximum rod power generations on the order of 620 to 655 W/cm with fuel temperatures (2600 to 2800 °C) near melting. The average maximum burnup of the eleven HPD elements currently in the reactor is 1.21×10^{20} fissions/cm³ (4950 MWd/MT fuel). Under these conditions a central void forms over essentially the full length of the element, high density columnar grains form, and homogenization of the mechanically-mixed and impacted UO_2 - PuO_2 occurs near the thermal center.

Nondefected and defected fuel element tests are conducted in the Fuel Element Rupture Test Facility (FERTF) at significantly higher specific power generations, i.e., with considerable fuel melting. This report provides current information obtained from the irradiation of intentionally defected UO_2 -2 wt% PuO_2 HPD FE-6004, nondefected UO_2 -2 wt% PuO_2 HPD FE-6502, and the rupture of intentionally defected UO_2 -4 wt% PuO_2 HPD FE-6504.

I. Irradiation of HPD FE-6004:

A transverse section at the midplane of pre-irradiated, vibrationally-compacted UO_2 -2 wt% PuO_2 HPD FE-6004, which was defected with a 1.6 mm diameter hole is shown in Figure 3.5. The element operated successfully with melting at the plane of the defect (approximately 54% of the radius molten) for approximately 60 hours, when the test was terminated as planned. The defected test had been in the FERTF for a total of 287 operating hours. Prior irradiation of this rod at approximately 620 W/cm to over 0.9×10^{20} fissions/cm³ (3500 MWd/MT_{fuel}) was sufficient to cause considerable in-reactor sintering of the fuel. Burst type activity releases characteristic of leaker defects occurred during incremental power increases. The steady state gamma activity in the loop during irradiation of this element was approximately 15 times greater than for any defected elements previously tested. The rod power, however, was approximately three times greater than for any previous experiment. Activity bursts occurred when the reactor was shut down and also when the loop was depressurized.

FIGURE 3.5 Transverse Section Through a Rod from HPD FE-6004 at the Point of Defect

There was essentially no change in the physical appearance of the defect, no dimensional changes in the rod, no significant fuel washout, and no evidence of increased hydride concentration in the cladding after irradiation in the FERTF. An observable effect of irradiation was a grey discoloration of the cladding downstream from the defect due to the evolution of material and/or gases from the defect. The alpha and beta autoradiographs* indicate that the symmetry of the fuel structure and fission product distribution patterns were affected by the presence of the defect. The concentration distribution patterns of the alpha and beta-gamma emitters as indicated on the autoradiographs do not coincide except in the once-molten fuel region. Some nonuniformity of alpha emitters is indicated on the autoradiograph suggesting possible plutonium segregation from the liquid. This phenomenon has been evident thus far only in defected HPD fuel rods. A source of fission product activity at the fuel periphery (dark spot on the beta-gamma autoradiograph and white spot on the alpha autoradiograph) is apparently an unexplained enriched UO_2 particle.

Examination of the fuel structure formed during irradiation at the plane of the defect indicates a greater degree of melting than anticipated from the maximum rod power generations calculated. Although the appearance of the fuel structure in an adjacent nondefected rod differed in some respects from the fuel structure in the defected rod at the same plane, slightly less fuel melting was indicated in the nondefected rod (50% vs 54% of the radius molten) (Figure 3.6). There was less porosity associated with the once-molten fuel region in the nondefected rod and the fission product distribution patterns (particularly as indicated on the alpha autoradiograph) were different for the defected and nondefected rods. The alpha autoradiograph from the nondefected rod showed no evidence of plutonium segregation. Sixty-four percent of the fission gases were released from the fuel under these operating conditions.

*The alpha autoradiographic technique was developed by J. H. Davies and R. W. Darmitzel at the Vallecitos Atomic Power Laboratory, General Electric Company.

J. H. Davies and R. W. Darmitzel, "Alpha Autoradiographic Technique for Irradiated Fuel," Nucleonics, Vol. 23, No. 7, pp. 86-87, July 1965.

FIGURE 3.6 Transverse Section Through a Nondefected Rod from HPD
Fe-6004

II. Irradiation of HPD FE-6502:

A transverse section through a rod from $\text{UO}_2 - 2 \text{ wt\% PuO}_2$ HPD FE-6502 which operated successfully in the FERTF at an estimated maximum rod power generation of 655 to 690 W/cm to a burnup of 0.12×10^{20} fissions/cm³ (490 MWd/MT_{fuel}) is shown in Figure 3.7. It is estimated that as much as 13% of the radius was molten at the midplane during steady state operation. The once molten fuel, which contains a high concentration of fission products, is delineated by a well defined porous region surrounding the central void. The β - γ autoradiograph indicates that once-molten fuel may have flowed into cracks which existed during operation. This is indicated by the high concentration of fission products in some cracks shown on the cross section. Other cracks, which probably formed during cooldown, show no evidence of penetration by molten fuel. This effect has been observed only under marginal melting conditions as represented in this cross section.

This specimen was sampled at selected locations for chemical analysis to investigate fission product and/or plutonium migration during irradiation (Figure 3.7). The analytical results of 16 samples taken from this transverse surface indicate that the concentrations of Ba-La¹⁴⁰, Zr-Nb⁹⁵, Sr⁸⁹, and Ru¹⁰³ tend to be high in the once-molten fuel region; Cs¹³⁷ and I¹³¹ have migrated to the cooler peripheral regions of the rod. The concentration of Ce¹⁴¹ and Pu is uniform across the cross section. The concentration of Ru¹⁰³ varies by three orders of magnitude in the selected samples taken across the specimen. Ruthenium concentration tends to be high in the once-molten fuel, low in the fission product depleted high density fuel region, high in the band located at the outer edge of the columnar grain growth region, and generally high and uniform in the peripheral region.

III. Irradiation of HPD FE-6504:

A vibrationally-compacted $\text{UO}_2 - 4 \text{ wt\% PuO}_2$ HPD element (FE 6504) was irradiated in the FERTF to a burnup of 0.23 fission/cm³ (920 MWd/MT_{fuel}) at a maximum tube power of 1745 kW (average approximately 1650 kW). This tube power produced a maximum peak rod power of 820 to 850 W/cm. The element successfully withstood a 20% overpower transient lasting approximately 11 seconds during intended steady state operation at 1685 kW.

FIGURE 3.7 Transverse Section Through a Rod from HPD FE-6502.
Maximum Rod Power Generation 655 to 690 W/cm.

After the first irradiation period, two replaceable rods (FR-3 and FR-6) were removed from FE 6504 for examination and were replaced by nonirradiated rods. A third preirradiated replaceable rod was deliberately defected with a 1.6 mm diameter hole near the midplane for continued irradiation in the FERTF. The maximum tube power during the second operating period was 1795 kW. Plans were to irradiate the element for an extended period of time at the estimated maximum rod power of 885 W/cm. The PRTR incident of September 29, 1965, occurred during this test. The element accumulated an additional burnup of 0.03×10^{20} (120 MWd/MT_{fuel}) during the second operating period prior to the rupture. Following the rupture of the element, four additional rods were examined to provide information related to the cause of failure. In addition to rods FR-3 and FR-6 which were removed before the rupture, these include the ruptured rod FR-4, the adjacent rod FB-06, the replaceable rod FR-10, and, recently, the other rod adjacent to the rupture, FB-83. Postirradiation examination data are not yet available on the latter rod. A summary of the current postirradiation examination status is given below.

A. Examination of Rods FR-3 and FR-6:

Two rods (Fr-3 and Fr-6) from FE 6504 were destructively examined after the initial irradiation period in PRTR at a maximum tube power of 1745 kW to a burnup of 0.23×10^{20} fissions/cm³ (920 MWd/MT_{fuel}). This tube power produced a maximum rod power of 820 to 850 W/cm. One rod was sampled for fission gas in the fuel region and the other was sampled in the plenum region. Each yielded approximately the same amount of total gas. There were no measurable changes in rod dimensions or balance point, indicating that no swelling or gross axial fuel movement occurred under these operating conditions.

Examination of the fuel structures formed at the midplane of the rods during irradiation indicates that fuel was molten to as much as 22% of the radius during operation. This is somewhat less than the amount calculated for this rod power generation (to 45% of the radius).

Examination of the bottom end regions of the irradiated rods showed that the depleted UO₂ pellet was essentially unaffected (only minor cracking).

The 1 wt% PuO_2 fuel region in the graded end had undergone sintering with a maximum fuel centerline temperature of approximately 1700 °C indicated. Limited fuel melting occurred in the 4 wt% PuO_2 region with the formation of a uniform central void. Typical structural and fission product distribution patterns were formed. There is evidence of flow of liquid fuel into radial cracks that existed during irradiation.

Fuel structures found in the top regions, however, indicated higher than expected fuel temperatures as a result of flux peaking. This was aggravated by inadvertent mixing of the UO_2 -1 wt% PuO_2 graded enrichment fuel with the 4 wt% PuO_2 fuel in the top end region during fabrication. Melting occurred in the fuel region near the depleted UO_2 pellet at the end of the fuel column. There was no evidence of reaction between the plenum spring and depleted UO_2 pellet in the top end region. In previous irradiations, higher fuel temperatures occurred in the bottom end regions than at the tops. Even though unexpectedly high fuel temperatures were generated adjacent to the gas plenum in the top of the rods, no condition that would compromise fuel element integrity during operation was apparent.

B. Examination of Ruptured Rod FR-4

Postirradiation examination of the ruptured rod (FR-4) from FE 6504 is still in progress.

1. Cladding Examination

A section of cladding approximately 7.6 cm long and almost 1.27 cm wide at the widest point is missing from the ruptured rod (Figure 3.8). The missing area includes the 1.6 mm diameter hole originally drilled through the cladding to defect the rod after its first period of irradiation in the FERTF.

Except for the ruptured region, there were no measurable diameter changes along the length of the rod. Considerable swelling and plastic deformation occurred in the small area of the rupture upstream from the main break. Detailed metallographic examination is being conducted on three transverse sections through the ruptured region. The cladding in the ruptured region appears relatively unaffected except in the immediate vicinity of the break. There is evidence of local overheating of the cladding

0°

90°

180°

270°

Coolant Flow Up

FIGURE 3.8 Burnout Failure of Rod FR-4 from HPD FE-6504
(Rupture occurred in the area of the rod that was previously defected
with a 1.6 mm diameter hole.)

into the beta phase and beyond (temperatures in excess of 850 °C) at the edge of the break indicating that failure was due to burnout. Molten $\text{UO}_2\text{-PuO}_2$ contacted the cladding at the edge of the break and, in some cases, alloyed with the Zircaloy (Figure 3.9).

Photomacrograph (4.8X)

β - γ Autoradiograph (4.8X)

FIGURE 3.9 Transverse Section (sample B-G) Through the Ruptured Region of Rod FR-4 from HPD FE-6504. Note High Activity at the Lip of the Break.

A thick oxide layer was observed on both surfaces of the cladding in the rupture region. The oxide layer is thickest at the edge of the break and tapers off rapidly to the normal thickness away from the break. At one point, the oxide layer continues around the fractured surface suggesting that the cladding may have been fractured for some time in this area before gross rupture occurred.

2. Fuel Examination

As determined by weight difference, more than 700 grams of $\text{UO}_2\text{-PuO}_2$ fuel material, or 40% of the total amount originally in the rod, are missing. Adjacent to the rupture, 12.7 cm of fuel is missing completely. Some $\text{UO}_2\text{-PuO}_2$ fuel has been recovered from the FERTF for examination. Macroexamination of the fuel particles indicates the presence of a few large pores in the fuel (Figure 3.10). Metallographic examination shows that most

of the fuel contains numerous small pores. A relatively large quantity of metal fines is mixed with the fuel particle in the 16-gram sample obtained.

FIGURE 3.10 UO_2 - PuO_2 Fuel Particles Collected from the FERTF System Following the Rupture of HPD FE-6504. The Particles Contain Some Macro and Considerable Micro Porosity Indicative of a Once-Molten Condition. (4.5X)

Examination of the fuel in that portion of the rod below the rupture shows the presence of once-molten fuel and an abnormally large central void in the region near the rupture (Figure 3.11). Graded enrichment was effective in preventing excessive fuel temperatures in the bottom end region of the rod.

Examination of the fuel above the rupture, however, shows a considerably larger central void than expected, indicating that the once-molten fuel is missing completely (Figure 3.12). The large central void extends uninterrupted over the entire fuel region above the rupture. In fact, the void extends

FIGURE 3.11 Transverse Section Through Fuel Rod FR-4 from HPD
FE-6504 42.2 cm (16.625 inches) Below the End of the Rupture (4.8X)

FIGURE 3.12 Transverse Section Through Fuel Rod FR-4 from HPD FE 6504 22.9 cm (9 inches) Above the End of the Rupture. There is no Evidence of Once-Molten $\text{UO}_2\text{-PuO}_2$ fuel. (4.8X)

through the depleted pellet at the top end of the fuel column and into the plenum (Figure 3.13). The UO_2 -1 wt% PuO_2 graded fuel material at the top of the rod became mixed with the UO_2 -4 wt% PuO_2 fuel material during fabrication, allowing higher than expected fuel temperatures in the flux peaked top end region. Apparently the depleted pellet was attacked by the molten fuel column since there is evidence of once-molten fuel on the inner edge of the hole through the depleted UO_2 pellet. Molten fuel material moved into the gas plenum region, and there is evidence of limited reaction between the molten fuel and the Inconel spring.

Photomacrograph

β - γ Autoradiograph α Autoradiograph

FIGURE 3.13 Longitudinal Section Through the top End Region of Ruptured Rod FR-4 from HPD FE-6504. Molten Fuel Column has Penetrated the Depleted UO_2 Pellet and Entered the Plenum Area Attacking the Inconel Spring.

Examination of the fuel structures formed in the defected rod during irradiation show that fuel temperatures were considerably higher than expected from the anticipated power generation. Evidence of abnormally high fuel temperatures in the ruptured rod could possibly be one of the most important observations related to the rupture mechanism. Examinations of an adjacent nondefected rod (FB-06), indicates the expected fuel structures for the calculated power generations. Possible explanations to account for the abnormally high fuel temperatures are being investigated.

C. Examination of Rod FB-06:

Examination of the rod (FB-06) adjacent to the ruptured rod in FE 6504 indicates no diametrical changes in the rod as a result of irradiation with the exception of a localized region near the top where localized necking of the cladding occurred. Although the percent fission gas release has not yet been determined, the total amount of gas collected from the rod (217 ml at STP) was approximately as expected. This indicates that the operating conditions of the rod during irradiation were approximately as anticipated.

Examination of transverse sections at three positions along the length of the rod shows that the expected fuel structures were formed at the maximum power generation of 885 W/cm: i. e., a central void; melting to approximately 50% of the radius; and columnar grain growth representing fuel temperatures in excess of 1800 °C to 80% of the radius at the plane of the defect (Figure 3.14).

A longitudinal section at the bottom of the fuel rod shows that the graded enrichment was effective in preventing excessive fuel temperatures near the end (Figure 3.15). Although the depleted UO_2 pellet adjacent to the bottom end cap was virtually unaffected, fuel melting occurred to within 0.61 cm of the pellet.

Examination of the top region of the rod showed that the 1 cm of UO_2 -1 wt% PuO_2 graded fuel material became mixed with the UO_2 -4 wt% PuO_2 fuel material during fabrication or subsequent handling. Sintering and homogenizing of the mechanically mixed UO_2 - PuO_2 fuel material occurred to the outside diameter of the fuel over a rod length of 1.8 cm

FIGURE 3.14 Transverse Section Through Nondefected Rod FB-06 at the Plane of the Rupture in Adjacent Rod FR-4 in HPD FE-6504. These Structures are Normal for Fuel Rods Operating at 885 W/cm and Indicate Melting to Approximately 50% of the Radius. (4.8X)

Photomicrograph

β - γ Autoradiograph

FIGURE 3.15 Longitudinal Section through the Bottom End of Nondefected Rod FB-06 from HPD FE-6504, (4.8X)

(Figure 3.16). The depleted UO_2 pellet was affected; however, molten fuel did not penetrate it as it did in the defected rod. Local overheating of the Zircaloy cladding occurred which allowed partial collapse of the cladding over a length of approximately 0.76 cm. The maximum diameter decrease is estimated to be 0.5 mm or 4%. Metallographic examination of the cladding, although not complete, shows beta phase zirconium indicative of

Photomacrograph

8- γ Autoradiograph

FIGURE 3.16 Longitudinal Section through the Top of Nondefected Rod FB-06 from HPD FE-6504. Localized Overheating Caused Plastic Deformation of the Cladding. (4.8X)

temperatures in excess of 850 °C. At these temperatures, the inward plastic deformation of the cladding was probably caused by the coolant pressure of 71.4 atm (1050 psi).

Axial flux profile monitors exposed during the irradiation of UO_2 -2 wt% PuO_2 HPD FE-6505 provide an estimate of the power distribution expected in the end regions due to flux peaking. Although these data show an increase in the neutron flux near the end, the degree of associated power peaking could not account for the high fuel temperatures indicated by fuel structures, even if one assumes no fuel grading in the rod ends.

After examination of the nondefected rod adjacent to the ruptured one revealed excessive fuel temperatures in the top end region, and localized cladding deformation, the fuel rod on the other side of the ruptured rod was acquired for examination to determine if the same situation existed in that rod. This examination is in progress.

D. Examination of Rod FR-10:

A minimum examination was performed on the nondefected replaceable rod (FR-10) that was irradiated in FE 6504 during its second period of irradiation in the PRTR of 0.03×10^{20} fissions/cm³ (115 MWd/MT_{fuel}).

There were no changes in the diameter of the rod as a result of irradiation and no measurable change in the balance point, indicating that the operating conditions did not cause swelling or gross axial fuel movement. The rod has not been sampled for fission gas or sectioned for metallographic examination.

Molten-Core Fuel Capsules Test - F. E. Panisko, D. A. Bitz, and L. A. Pember

Four capsules (GEH-14-738 through -741) designed to demonstrate the stability of molten-core fuel element operation were shipped to Battelle-Northwest for examination following a successful irradiation in MTR. The capsules had been charged into the reactor while the reactor was at full power. Each capsule contained a diametral hole in the UO_2 pellets (26% U^{235}) to allow molten fuel to impinge on the Zircaloy-2 cladding. Hole diameters of 0.06, 0.130, 0.19, and 0.25 cm were employed. The melt radius was approximately 60% of the fuel radius.

PRTR Failed Fuel Elements - F. E. Panisko

During 1965 one rod from each of two fuel elements (Numbers 5237 and 5192) leaked fission products into the coolant of PRTR. The defect in each rod was located in a discolored area (white or gray) on the cladding surface. The fuel in each rod was not suspected of being contaminated as F.E. #5237 had reached a burnup of 1.0×10^{20} fissions/cm when discharged on January 17, 1965, and F.E. #5192 had reached 1.7×10^{20} fissions/cm on May 24, 1965. The presence of fission products from the defective rods did not hamper reactor operation. Postirradiation examination of the defect area of the rod from F. E. # 5237 revealed massive hydriding of the cladding. The rod from F. E. # 5192 broke in the area of the defect during handling in the hot cell and the area of the fracture is being examined.

All of the active PRTR fuel elements with discolored areas on the cladding were visually examined in the storage basin. Twenty-one fuel elements were examined and seven elements had at least one rod with a suspected hydride area on the cladding; Some of the discolored areas will be examined in the hot cells. The remaining 14 fuel elements contained rod with nonuniform oxide films.

The shape of some of the discolored areas is similar to a fingerprint. It is possible that grease was deposited on the cladding during handling of the fuel elements before initial irradiation, consequently, all new fuel elements are now covered with plastic bags to protect them until initial charging.

Thermal Cycling of Defective Cladding - R. J. Lobsinger

On-site ultrasonic inspection of Zircaloy-2 tubing obtained from commercial vendors has occasionally revealed the presence of small cracks. These cracks are usually radially oriented and range in depth from 0.002-0.010 cm and in length from 0.010-0.120 cm. In order to assess the potential problem of even smaller undiscovered cracks becoming larger during reactor operation, testing was initiated to measure the propagation tendencies of small cracks during thermal cycling.

Two test capsules were fabricated using 1.28 cm ID by 0.076 cm wall Zircaloy-2 tubes containing small (0.002-0.006 cm) internal fabrication cracks. These tubes were loaded with depleted UO_2 pellets to provide a means of stressing the cladding during heating. One capsule was purposely contaminated with 40 ppm I_2 to determine possible accelerating effects; the other served as a control. The capsules were then cycled in a steam autoclave from 100 °C and 1 atm to 400 °C and 68 atm.

Ultrasonic examination was performed on the capsules after 20 thermal cycles and compared to data obtained prior to cycling. Crack propagation was observed in the capsule contaminated with I_2 , whereas the control was unchanged. Following further cycling, metallographic examination will be performed to permit interpretation of the results.

PRTR-HPD Fuel Fabrication - R. L. Gulley and R. D. Reid

Since the start of fabrication on June 8, 1965 a total of 64 Mark I-R series fuel elements and 21 replaceable rods have been fabricated. Thirty-nine of these elements were completed within the period of October 25 through December 31, 1965.

Of the 64 elements completed, 55 will be used for the Batch Core Experiment which is scheduled for start up on June 1, 1966. It is expected that 12 additional elements will be fabricated for this experiment.

Eight different PRTR-HPD designs have been fabricated for the Batch Core Experiment and for other PRTR and PRCF experiments. A physical description of these elements is presented in the following table (Table 3.1).

PRTR Fuels - Quality Control - J. P. Keenan

During this quarter, 39 PRTR fuel clusters were fabricated.

Two ultrasonic tests are used to insure clad integrity on all PRTR fuels. One test is used to detect crack-type defects along the entire length of the tube and the other to detect crack-type defects in the weld area. All tester signals which indicate defects greater than 0.038 mm deep are cause for tube rejection unless it can be demonstrated that the signal originates from something other than a propagating-type 0.038 mm or greater defect.

TABLE 3.1 PRTR-HPD Fuel Element Descriptions

Element Type	Core Geometry	Removable Rods	Pressure Measurement Rods	Fuel
Mark I-R ₁	Standard*	0	0	UO ₂ -2 w/o PuO ₂
Mark I-R ₂	Standard	3	0	UO ₂ -2 w/o PuO ₂
Mark I-R ₃	Graded**	3	0	UO ₂ -4 w/o PuO ₂
Mark I-R ₄	Graded	3	0	UO ₂ -2 w/o PuO ₂
Mark I-R ₅	Graded	0	0	UO ₂ -2 w/o PuO ₂
Mark I-R ₆ ***	Standard	0	0	UO ₂ -2 w/o PuO ₂
Mark I-R ₇	Standard	3	2	UO ₂ -2 w/o PuO ₂
Mark I-R ₈ ****	Graded	3	2	ThO ₂ -5 w/o PuO ₂

*A standard core consists of a depleted UO₂ pellet (12.5 g/pellet) at each end of a UO₂-2 w/o PuO₂ fuel column (1793 g).

**With the exception of the Mark I-R₈ element, a graded core consists of a depleted UO₂ pellet (12.5 g/pellet) at each end of a UO₂-PuO₂ fuel column. The UO₂-PuO₂ fuel column contains, from bottom to top respectively, UO₂-1wt% PuO₂ (36.5 g), either UO₂-2 w/o PuO₂ (1743 g) or UO₂-4 w/o PuO₂ (1743 g), and UO₂-1 w/o PuO₂ (12.5 g).

***The Mark I-R₆ differs from the other elements of the Mark I-R series in that three of the 19 rods are made up of three subassemblies. Cadmium and aluminum caps and seals, containing customer supplied foils, are installed between each subassembly. Sixteen of the rods are fabricated with a standard core. Three of the rods consist of a depleted UO₂ pellet (12.5 g/pellet) at each end of a UO₂-PuO₂ fuel column. The UO₂-PuO₂ fuel column contains, from bottom to top respectively, UO₂-2 w/o PuO₂ (770 g), a monitor foil, UO₂-2 w/o PuO₂ (245 g), a monitor foil, and UO₂-2 w/o PuO₂ (770 g).

****Grading of the Mark I-R₈ was accomplished by placing three, 1-cm thick ThO₂ pellets (12 g/pellet) at each end of a ThO₂-5 w/o PuO₂ (1606 g) fuel column.

In past cases, defect indications have been much more of a problem than they were during this run. Less than 5% of the as-received tubes were rejected. After fabrication, no tubes were found with defects over 0.038 mm deep. Typically, no crack-type defects have been found with the weld tester on PRTR-type fuels and this again was the case this time. Optical checking and defect sectioning has verified that the testers are performing acceptably. Improved performance is attributed to the high quality of as-received tubing in the most recent shipment from Reactive Metals, Inc., and to careful handling of the fuel rods throughout the fabrication process.

A balance and scale calibration and checking program has been initiated throughout the operation. It is believed that this will improve fuel element quality attributes in a number of areas by more thoroughly systematizing procedures already in use.

Building Additions and Facilities Modifications - L. G. Merker

Project CAH-916 - Chemical and Materials Engineering Laboratory

The Chemical and Materials Engineering Laboratory (324 Bldg.) was accepted from the contractor on December 20, subject to specific exceptions. Two Commission-owned boom mounted electromechanical manipulators have been converted to bridge style mountings by the manufacturer, and will be installed in the Shielded Materials Facility by the cost plus fixed fee contractor during the first quarter of 1966. Rails will be installed in all cells to allow the manipulators to be used where needed most. Other items received during the quarter for use in the Shielded Materials Facility include seven (7) pair (14 ea.) extended reach Model 8 master slave manipulators, one 17-inch lathe, powder preparation equipment, welding equipment, and a high energy impaction machine which has been modified to increase reliability and accessibility for hot cell use. In-cell maintenance of the high energy impaction machine would be virtually impossible without the modifications that have been made.

It is anticipated that the hot cell complex will become operative and that a hot cell mockup plus most of the existing equipment will be installed within the next six months.

Project CAH-151 - 308 Office Addition

The 308 Building office addition was scheduled for construction completion on December 17, 1965; however, construction delays were experienced and occupancy will not occur until January 1966.

PART IV - JOINT FUEL DEVELOPMENT PROGRAMS

Irradiation Testing of EBWR Prototype Fuel Rods - W. J. Bailey

Three capsules containing vibrationally compacted, pneumatically impacted UO_2 -1.5 wt% PuO_2 continue to be irradiated in the MTR. The capsules, made with short sections of EBWR rods (77-85% TD), have achieved an estimated maximum burnup of 20,050 MWd/MT of fuel (5×10^{20} fissions/cm³) to date. This burnup value is slightly lower (~10%) than that previously reported and is the current and more refined estimate. Burnup samples from two of the 29 previously discharged capsules were isotopically analyzed, and the data are shown in Table 4.1. If only the flux-wire data are used, the trend (based on seven samples) to date has been to underestimate the actual fuel burnup. Isotopic data for other capsules are shown in Table 4.2.

TABLE 4.1 Comparison of Burnup Determinations by Isotopic Analysis and from Flux-Wire Data

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

*Possible error, $\pm 20\%$.

**Confidence level, 95%.

TABLE 4.2. Plutonium Isotopic Data

Capsule GEH-14 No.	Approx. Burnup, Based on Reactor Data:	(Fissions/cm ³) x 10 ⁻²⁰	Burnup, Based on Capsule Flux- Wire Data	(Fissions/cm ³) x 10 ⁻²⁰	Plutonium Mass Spec- trographic Analyses, at. %			
	(MWd/MT fuel)		(MWd/MT fuel)		Pu ²³⁹	Pu ²⁴⁰	Pu ²⁴¹	Pu ²⁴²
--	0		0	0	91.53	7.75	0.69	0.03
526			4900	1.2	72.94	22.90	3.67	0.484
527	12,500	~3.1			55.15	35.04	7.65	2.165
650			1990	0.46	85.48	12.98	1.441	0.100
711	13,000	~3.2			47.47	38.62	9.93	3.98
713	18,600	~4.6			43.22	39.32	10.83	6.62
716	7,150	~1.8			70.74	24.11	4.51	0.639
717			2050	0.48	85.76	12.67	1.476	0.096

Irradiation of the production-run EBWR fuel rods (86-89% TD) in the 21-rod bundle (Fuel Element No. 6501) will continue (current burnup: 0.7×10^{20} fissions/cm³, goal: $\geq 2.7 \times 10^{20}$ fissions/cm³) as soon as the PRTR commences operation. Irradiation of an additional 24 production-run EBWR rods is continuing (goal: burnups to 6.8×10^{20} fissions/cm³ or 27,500 MWd/MT of fuel) in another reactor, and examination of one of the 13 discharged rods is completed (Fig. 4.1). The rods are 1.07 cm OD by 148 cm long including a 14.6 cm long plenum.

A paper, "Irradiation of Impacted UO₂-PuO₂," (BNWL-SA-256) which covers all of the above irradiation testing was presented at the American Nuclear Society Winter Meeting in Washington, D.C., in November.

SAP End Closures - Atomics International - R. F. Bolen

As a result of the successful completion of a contract to supply experimental end closures to Atomics International (see Quarterly Report, July-September 1965), an extension to the contract was agreed upon. Work on this extension to supply Atomics International with an additional 100-200 experimental end closures in the XAP-001 and XAP-005 (aluminum) alloys was begun.

Photomicrograph

 β - γ Autoradiograph

Negative no. D1396, 634

FIGURE 4.1. Transverse Section at Maximum Heat Flux Zone of an EBWR Fuel Rod, Irradiated in the PRTR. Fuel Rod Data; Burnup, 0.52×10^{20} Fissions/cm³; Rod Power, 208 W/cm; Heat Flux, 61 W/cm²; Fissions Gas Release, 6%; Rod Size, 1.07 cm OD by 148 cm Long.

Both XAP-001 and XAP-005 (Alcoa Experimental alloys) contain 6 wt% aluminum oxide as a fine, uniform dispersion. XAP-005 is a nuclear, iron-poor (0.05 wt%) grade. By comparison XAP-001 has an iron content of 0.2 wt%. Tensile tests⁽¹⁾ indicate that parent strength properties of XAP-001 and XAP-005 are practically identical.

One hundred of the magnetic force welded end closures to be provided under the contract extension have already been made. These are XAP-001 finned tubes as shown in Figure 4.2. A limited number of end closures will be made using XAP-001 caps and XAP-005 tubes.

1. Niels Hansen. "Tensile Properties at Room Temperature and at 400 °C of Commercial Sintered Aluminum Products, "Riso Report No. 96, December 1964.

FIGURE 4.2. Magnetic Force Welded SAP Capsules (1 ft. long)

Quality of the welds is maintained by a combination of work piece dimensional control, visual examination, helium leak testing of each completed weld, uniformity of power curves (recorded by a storage oscilloscope), and destructive testing (tensile and burst) of random samples of the completed end closures.

Dye penetrant examination performed at Atomics International on earlier welds showed varying degrees of bleedback at the weld on all samples. This was to be expected since the external upset with unbonded areas in its extremity is sheared off before being withdrawn from the machine electrode. Some indications, however, showed that this unbonded area might extend not only through the upset, but into the original tube OD.

For this reason, seven end closures were dye-penetrant tested at Battelle-Northwest. All seven showed varying degrees of bleedback. Nine welds - two as welded plus seven previously tested - were lathe turned to the tube OD and again dye-penetrant tested. All but two of the nine showed no indications. The two (from those previously tested) showed very minor indications. These two samples were again lathe turned to between 0.075 mm and 0.100 mm under the original 0.69 mm wall diameter and again dye-penetrant tested - after which no indications were found.

Tensile tests of end closure samples showed ultimate strengths varying from 2100 kg/cm^2 to 2800 kg/cm^2 ; joint efficiencies of 81 to 108% (joint area exceeds parent material area by an undetermined amount due to internal upset).

A typical tensile tested is shown in Figure 4.3

Burst test samples, from initial work with XAP-001, are shown in Figure 4.4 a and b.

FIGURE 4.3. SAP Tensile Tested Sample

FIGURE 4.4. XAP-001 Burst Tested Samples

CUSTOMER WORK

Phoenix Fuels - J. P. Keenan and R. D. Reid

Plans have been directed toward fabrication of fuels for critical test experiments during the remainder of this fiscal year and fabrication of reactor grade fuels for irradiation in MTR next fiscal year.

Both fuel groups are to be plate-type elements. The core in each case is 6.3 cm wide x 59.8 cm long x 0.51 cm thick. Nominal enrichment is 20 wt% Pu in aluminum. The Pu²⁴⁰ content of the plutonium to be used in the plates is also approximately 20 wt%. The exact isotopic concentration will be determined based on what can be procured. The fuel matrix and fuel cladding will be aluminum. Critical test plates will be clad, but the cladding will not be bonded to the core. Cladding will be bonded on fuels provided for irradiation testing in the MTR.

Cost and time estimates have been provided for various fuel fabrication alternatives. Pilot size tests of some fabrication steps have been performed. Development work has been initiated on the process which will be used for fabrication of critical plates.

Ultrasonic Examination of Saxton Tubing - R. J. Lobsinger

In order to establish the effect of manufacturing defects on fuel rod performance, a study was initiated to determine the effect of thermal cycling on tubing containing rejectable size internal defects. Three fuel rods were fabricated by vibratory compaction using Saxton tubing rejected at PNWL because of defects in excess of 0.0025 cm depth. Although predominately all of the tubing from a particular lot was rejected, only a limited number of tubes were available for these tests. The remainder of the material was returned to the customer. Thermal cycling of these fuel rods was accomplished by heating in a steam autoclave to 400 °C and 68 atm in one hour, holding at temperature for three hours, and cooling to 100 °C and 1 atm over an 8-hour period.⁽¹⁾ No propagation was observed after 50 thermal cycles, consequently metallographic examination was performed to characterize the defects.

¹BNWL-150, Ceramics Research & Development Quarterly Report

The defect region was localized by precise ultrasonic inspection; however, it was extremely difficult to find the defects during metallographic examination. One which was found is shown in Figure 4.5. The defect appears to be an internal void approximately 0.100 cm long penetrating through 90% of the tube wall. Attempts to estimate the circumferential extent of this void by continued, metallographic polishing indicate it to have been only 0.002-0.003 cm deep. Failure to locate the other defects can be ascribed to the probability of "polishing through" the defect during examination. No appreciable hydrides were found during metallographic examination.

FIGURE 4.5. Zircaloy-2 Defect

PART V. DEVELOPMENT OF ADVANCED FUEL CONCEPTS

Materials Joining, Tantalum - R. F. Boolen

Thick walled tantalum tube was joined by magnetic force welding. Sections of the 1.6 cm OD x 0.17 cm wall tube were joined and examined metallographically. Cross sections of a prototype weld are shown in Figures 5.1 and 5.2. The rapidity with which bonding takes place (less than 14 msec) limits recrystallization to a very narrow band in the weld area.

The magnetic force welder is ordinarily used to effect welds in refractory metals of about 0.323 sq cm. The application described here indicates the process is capable of producing high integrity joints in relatively heavier sections of refractory materials.

Metal Joining Studies - D. E. DeWitt, R. F. Klein, and R. C. Smith

Welding studies using the tungsten inert gas process (TIG) have been under way on Haynes-25, Ta, Mo, and AISI 347 SS. Argon gas was found necessary to produce good clean welds with Haynes-25 and was extremely beneficial in the AISI 347 SS studies.

Molybdenum and tantalum studies were directed toward obtaining deep penetration on "melt-down" type weld joints. Welds in tantalum with 0.178 cm penetration were achieved. Refinement of the welding parameters may be necessary to provide maximum weld confidence. Some microcracking problems were experienced with molybdenum joints. The characteristics of molybdenum, particularly the variability of character in different specimens and the extreme sensitivity to history and purity, make it difficult to obtain consistent welding results with this metal. However, further development should provide good welds up to 0.178 cm in penetration.

Spheroidization of UO_2 - PuO_2 and PuO_2 - R. E. Bardsley

Two kilograms of UO_2 -2 wt% PuO_2 spheres were produced by the agglomeration process. The UO_2 with 0.1 wt% TiO_2 added as a sintering aid was ball milled for 36 hr. The UO_2 was then dry blended with PuO_2 prior to adding carbowax binder. The blended material was agglomerated,

FIGURE 5.1. Magnetic Force Welded Tantalum - 0.172 cm Wall Tube Section (50X)

Neg. 5652307C & D

FIGURE 5.2. Magnetic Force Welded Tantalum 0.172 cm Wall Tube
Section (50X)

Neg. 5652307A & B

dried and sintered in an Ar-8% H atmosphere for 12 hr at 1650 ± 50 °C. Densities (as measured from 6 mesh spheres) were 80-81% of TD. Figure 5.3 shows ceramographic sections of two particles. Figure 5.4 shows alpha autoradiographs of sintered allomerates before final screening.

A 230-g batch of PuO_2^* was agglomerated to determine if low density (68-85% TD) PuO_2 spheres, having relatively high strength, could be produced by this process.

Forty-five grams of -35 + 100 mesh particles (as green particles) were obtained. Approximately half of these particles were -35 + 60 mesh, the remainder being -60 + 100 mesh. These particles were sintered at 1650 °C for 12 hr in an Ar-8% H atmosphere.

A glove box line was set up in Room 131 for developing the agglomeration process and to obtain additional PuO_2 spheres. Three hundred grams of -35 + 200 mesh spheres were produced from PuO_2 that had been previously pneumatically impacted. Approximately a 50% yield (-35 + 200 mesh size agglomerates) was realized for these powder batches compared to approximately 20% for the initial 230 g batch mentioned earlier.

In addition to developing the process, this material was processed to determine if, because of its previous densification, the pneumatically impacted powder might not sinter as completely as high-fired PuO_2 and would produce a lower density particle. The powder was ball milled for 17 hr to reactivate the surface of the pneumatically impacted material, agglomerated, and sintered at a nominal temperature of 1600 °C. The other sintering conditions were the same as previously described.

Varying degrees of sintering were observed in the finished spheres due to the thermal gradient within the furnace. Control of smaller furnace loads within the central heat zone will be required.

*Under current nuclear safety specifications, the agglomeration of PuO_2 is limited to 230 g batches due to the water-carbowax binder acting as a moderator. The process is now being reviewed to determine if a special nuclear specification can be formulated which will allow larger quantities of material to be handled safely.

FIGURE 5.3. Sintered Low Density Agglomerates, UO_2 -2 wt% PuO_2
(50X)

Neg. 45125F & H

FIGURE 5.4. Alpha Autoradiographs of UO_2 -2 wt% PuO_2 Agglomerates
Prior to Final Screening (10X)

Neg. 5652293B, 5652294B

Laboratory analyses indicate that for the initial PuO_2 batch, sphere densities were 81% TD while the particles produced from the impacted PuO_2 had densities ranging from 75 to 81% TD.

Presently there is no method to accurately determine directly the density of small (-35 + 200) mesh particles. Small particle densities must be extrapolated from results obtained from either pellets or large (6-10 mesh) particles that are sintered with the small agglomerates. The chemical analysis group is working in cooperation with us to determine if a method can be developed to give the desired information.

Crush tests will be made early in January on the various agglomerates.

Additional batches of PuO_2 spheres are to be produced using high-fired oxide.

Coining of Cermet Extrusion Billets - E. A. Snajdr

A preliminary study was initiated to investigate coining as a method for producing extrusion billets.

The coining operation is based on three steps: (1) cold pressing a UO_2 -AISI 304 SS powder mixture to form a powder compact, (2) heat treating to either sinter and/or anneal the powder compact, and (3) repressing the heat-treated pellet in the punch and die used in step (1) to increase the density and resize the compact.

Procedure and Results

The procedure outlined above was followed in general; however, variations in compacting pressures and heat treatments were employed. Initially, powder compacts were pressed at varying pressures and sintered as shown in Table 5.1 with the results of final pressing. Sintering was accomplished at 1300 °C for 1 hr.

Pellets 1-6 were approximately 3.8 cm long and 1.82 cm. diam. Pellets 7-12 were approximately 1.9 cm long and 1.82 cm. diam. All density measurements were made by the geometric method, since the pellets were good right circular cylinders.

TABLE 5.1. Data for Coined Cermets

Specimen	Cold Pressed		Sintered		Coined	
	Pressure kg/cm ²	% TD	Atmosphere	% TD	Pressure kg/cm ²	% TD
1	2450	65.8	H ₂	75.2	8790	--
2	2950	68.0	H ₂	78.8	8790	85.9
3	3450	69.3	H ₂	77.1	8790	85.9
4	3940	70.0	H ₂	77.0	8790	86.5
5	4425	71.5	H ₂	76.2	8790	85.6
6	5000	72.1	H ₂	79.0	8790	85.7
7	2450	65.7	He	72.2	8790	85.7
8	3150	69.4	He	76.2	8790	86.9
9	3500	70.6	He	76.2	8790	86.3
10	3875	71.6	He	77.3	8790	92.5
11	4425	74.0	He	79.0	8790	91.0
12	4910	74.3	He	79.2	8790	87.0

The microstructure of a typical pellet after sintering is shown in Figure 5.5. The inclusions present in the voids of the stainless steel matrix are unidentified; however, observations under polarized light show them to be transparent and of a dark brown color indicating the possibility of iron oxide. Figure 5.6 shows the grain structure of the sintered compact's stainless steel matrix. A typical sintered matrix microstructure after repressing is shown in Figure 5.7.

An alternate procedure was examined utilizing an annealing heat treatment at 950 °C for 1 hr. Data for pellets produced by the annealing technique are shown in Table 5.2.

A typical microstructure of the as-annealed compact is shown in Figure 5.8. The powder compacts were annealed in flowing argon (2 ft³/min) which was cold trapped with dry ice and acetone before passing through the fused quartz annealing tube. The grain structure of the stainless steel is shown in Figures 5.9 and 5.10. Grain growth has not occurred across the entire stainless particle as in Figure 5.6. Only preferential etching

FIGURE 5.5. Microstructure of a Coined Pellet, as Polished (75X)

FIGURE 5.6. Grain Structure of Stainless Steel Sintered Matrix, as Etched (250X)

Neg. 465-7384A, 465-7386B

FIGURE 5.7. Microstructure of Coined or Repressed Pellet (250X)

FIGURE 5.8. Microstructure of an Annealed Pellet (75X)
Neg. 465-2289, 465-2312Z

FIGURE 5.9. Grain Structure of Stainless Steel Powder After Annealing, as Etched (250X)

FIGURE 5.10. Grain Structure of Stainless Steel Powder After Annealing, as Etched (1000X)

Neg. 465-2311A and C

TABLE 5.2.

Specimen	Cold Pressed		Annealed		Coined	
	Pressure kg/cm ²	% TD	Atmosphere	% TD	Pressure kg/cm ²	% TD
13	7030	79	Argon	80	9840	87
14	7730	81	Argon	82	9840	88
15	8440	82	Argon	83	9840	88
16	9140	83	Argon	84	9840	88
17	9840	84	Argon	85	9840	89
18	9840	84	Argon	85	9840	90

occurred because the electrolytic etching was difficult to perform with uniform results on the powder compact as a result of poor bonding between the stainless steel particles. The initial size of the stainless steel powder used to fabricate the compacts is shown in Figure 5.11.

FIGURE 5.11. Stainless Steel Powder Before Compaction (250X)

Neg. 465-2329

Discussion of Results

The final repressed densities of compacts 1-12 were higher than compacts 13-18. All compacts were cold pressed and repressed in the same die (1.82 cm diam), and consequently, when pellets 1-12, having a higher shrinkage during heat treatment, were repressed; it was possible to close up some of the remaining porosity by greater deformation of the matrix.

The inclusions were not identified by analytical means but are believed to be an oxide phase because of their transparent nature. Particles of a similar nature but of smaller particle size are present in as-extruded fuel pins.

Conclusions and Future Work

This investigation was intended to be of a preliminary nature; however, it has demonstrated the possibility of utilizing coining as a process for producing extrusion bullets of 90% TD, 30% UO_2 -304 SS cermet. This process appears to have several desirable aspects, namely:

1. Simplicity
2. Low temperature (950 °C)
3. Short time (1 hr) at temperature
4. Low supporting hardware requirements.

From a product standpoint the survival of the fuel particles during all pressure applications (up to 9840 kg/cm²) might indicate the possibility of producing a high density matrix with low density fuel particles with a minimum of fuel particle fracturing.

FFTF - Driver Fuel Evaluation - E. A. Snajdr

An evaluation program was initiated to establish the physical properties of UO_2 -stainless steel cermet fuel pins. Current fabrication procedures consist of powder compaction, encapsulation, and extrusion. Powder compaction is accomplished by cold pressing hand-mixed AISI 304L SS- UO_2 powders in a double acting punch and die system. Pressures can be varied to 9840 kg/cm². Pellets are then slipped into a tube of AISI 304L SS. The resultant assembly is loaded into a 1030 mild steel extrusion can which is evacuated and sealed. The can assembly is heated to 1200 °C and extruded. The can is removed from the pin by acid dissolution.

Currently the effect of volume percent UO_2 in the cermet on yield strength, ultimate tensile strength, and elastic modulus are being investigated. A decision was made to use the current fabrication technology as a base for future physical and microstructural properties evaluation. The volume percent of UO_2 studied will be 10, 20, and 30 vol%; in addition, an unfueled pin will be used as a control sample for the study.

The pins of varying concentrations of UO_2 were fabricated, and the microstructure was examined as a part of the fuel pin characterization prior to mechanical testing. The as-extruded fuel pins are shown in Figures 5.12 through 5.17. The low UO_2 volume concentration cermets, 10 and 20 vol%, show considerable evidence of particle segregation during fabrication. This segregation may occur during the pellet pressing or extrusion operation. The 20 vol% longitudinal cross section shows some evidence of UO_2 alignment and preferential placement at flow laminations of the matrix which results in the UO_2 particles being aligned on a parabolic curve. The 30 vol% UO_2 -cermet shows evidence of being exposed to the atmosphere during extrusion (See Figures 5.16 and 5.17). Figures 5.18 through 5.23 show the same fuel pins after swaging. For most cases the UO_2 fuel particles have been fractured and have pulled away from the matrix during polishing. The 30 vol% stainless matrix did not survive the swaging operation, as evidenced by the gross cracking. Chemical analyses had been performed on the 0 vol% UO_2 -stainless steel fuel pin to determine carbon pickup during extrusion. The following results listed in Table 5.3 were obtained by chemical analysis.

TABLE 5.3.

	<u>Starting Material</u>	<u>Extruded</u>	<u>Swaged</u>
Clad	270 ppm (0.027%)	2418 (0.24%)	2017 (0.2%)
Core	213 (0.0213%)	3462 (0.4%)	1440 (0.14%)

Evidence of the carbon concentration in the as-extruded and as-swaged pins is found in the increased etching at the grain boundaries and the observation of carbide particles in the grain boundaries of the fuel core.

Figure 5.24 shows the grain boundary attack during etching as a result of the decreased corrosion resistance due to localized chromium depletion at the grain boundaries when chromium carbide is precipitated. Figure 5.25 shows the carbide particles in the grain boundaries. The fine stringered particles appear to be the result of both porosity and an included oxide phase (See Figure 5.26). This identification has not been confirmed to complete satisfaction and is subject to further investigation. Figure 5.27 shows the bond between the wrought clad and stainless steel powder metallurgical core. Carbide precipitation has not occurred in the clad; however, chemical analysis shows carbon to be present at the 0.2% concentration level. Carbon is evidently in solution as evidenced by the etching characteristics of the clad in Figure 5.27. Figure 5.27 shows the core to clad bond to be quite good with grain growth across the original interface.

The effect of cold swaging upon the fuel particles is shown in Figure 5.28. The particle fracture planes are parallel to the swaging force. Fuel particles after extrusion are shown in Figure 5.29 for comparison.

Conclusions and Future Work

1. Carbon pickup during extrusion must be eliminated.
2. Lamination and alignment effects are evident in the stainless steel matrix as a result of metal flow during extrusion and fuel particle shape.
3. Cold swaging fractures fuel particles to a high degree.
4. Oxide inclusions were observed in the matrix of extruded pins requiring considerable refinement of present powder metallurgical practices.

FIGURE 5.12. AISI 304 SS-10 vol% UO_2 Fuel Pin As-Extruded, Transverse Cross Section, As-Polished (7X)

FIGURE 5.13. AISI 304 SS -10 vol% UO_2 Fuel Pin As-Extruded, Longitudinal Cross Section, As-Polished (7X)

FIGURE 5.14. AISI 304 SS-20 vol% UO_2 Fuel Pin As-Extruded, Transverse Cross Section, As-Polished (7X)

Neg. 465-2288D, 465-2289D, 565-2319-20.

FIGURE 5.15. AISI 304 SS-20 vol% UO_2 Fuel Pin As-Extruded
Longitudinal Cross Section, As-Polished (7X)

FIGURE 5.16. AISI 304 SS-30 vol% UO_2 Fuel Pin As-Extruded,
Transverse Cross Section, As-Polished (7X)

FIGURE 5.17. AISI 304 SS-30 vol% UO_2 Fuel Pin As-Extruded,
Longitudinal Cross Section, As-Polished (7X)

Neg. 565-2319-20, 565-2223-24, 565-2223-24

FIGURE 5.18. AISI 304 SS-10 vol% UO_2 Fuel Pin Extruded and Swaged
Transverse Cross Section, As-Polished (7X)

FIGURE 5.19. AISI 304 SS-10 vol% UO_2 Fuel Pin Extruded and Swaged
Longitudinal Cross Section, As-Polished (7X)

FIGURE 5.20. AISI 304 SS-20 vol% UO_2 Fuel Pin Extruded and Swaged
Transverse Cross Section, As-Polished (7X)

FIGURE 5.21. AISI 304 SS-20 vol% UO_2 Fuel Pin Extruded and Swaged
Longitudinal Cross Section, As-Polished (7X)

FIGURE 5.22. AISI 304 SS-30 vol% UO_2 Fuel Pin Extruded and Swaged
Transverse Cross Section, As-Polished (7X)

FIGURE 5.23. AISI 304 SS-30 vol% UO_2 Fuel Pin Extruded and Swaged
Longitudinal Cross Section, As-Polished (7X)

FIGURE 5.24. AISI 304 SS Cladding As-Extruded Fuel Pin. Etched in Oxalic Acid Electrolytic (100X)

FIGURE 5.25. AISI 304 SS Cladding As-Extruded. Etched in Oxalic Acid (1000X)

Neg. 465-2289M, 465-2288C

FIGURE 5.26. Fuel Core of Cermet Pin (1000X)

FIGURE 5.27. Core to Clad Bond Etched (500X)

Neg. 465-2133R, 465-2290B

FIGURE 5.28. Core and Clad After Swaging Longitudinal Cross Section, As-Polished (75X)

FIGURE 5.29. Fuel Core After Extrusion Longitudinal Cross Section, As-Polished (75X)

Neg. 465-2293J, 465-2289E

Pressure Measurement of UO_2 Capsules at Elevated Temperatures -

R. J. Lobsinger and D. R. Doman

Although graphite has been selected as the cladding material for the the driver and flux adjusting fuels in the HTLTR, metallic cladding may have to be used for the experimental fuel. Selection of a suitable material will depend in part upon the ability to withstand the buildup of internal pressure caused by the release of sorbed gases and water from the fuel at elevated temperatures. Calculations to determine equilibrium internal pressures are difficult to perform because reactions between the water and sorbed gases with the core and/or cladding will affect the results. Consequently, capsule tests were performed to measure such data experimentally.

Four capsules were fabricated using AISI 304 SS cladding, 1.26 cm ID by 0.089 cm wall, and instrumented with negligible volume, pressure transducers. Two of these capsules were loaded with depleted UO_2 pellets and two were vibrationally compacted to approximately 70% density using impacted, depleted UO_2 powder. These assemblies were heated to 1000 °C and the capsule pressures were measured during heating. Results are shown in Table 5.4.

TABLE 5.4. Pressures Generated by UO_2 Capsules, psig

<u>Temperature, °C</u>	<u>Capsule 1, Pellets</u>	<u>Capsule 2, Pellets</u>	<u>Capsule 3, Powder</u>	<u>Capsule 4, Powder</u>
255	16.4	20.0	63.8	45.7
400	31.2	38.5	85.8	64.8
400 ^(a)	25.5	29.7	66.3	55.1
600	34.4	40.1	90.4	77.9
800	13.5	13.7	48.1	39.5
1000	15.2	14.9	35.5	33.9

(a) Capsules were cooled after heating to 400 °C initially and following 14 hr of cooling reheated to 400 °C.

In general, capsule pressures increased until temperatures in excess of 400 °C were obtained; subsequent heating resulted in decreasing pressures, indicating extensive reaction of the gases with the cladding and/or core. Pressures measured at temperatures other than at 1000 °C do not represent

equilibrium conditions. Calculations were made to determine agreement with the experimental data. An analysis of the fuel used showed:

	<u>Powder</u>	<u>Pellets</u>
H ₂ O	88 ppm	23 ppm
Adsorbed Gases	0.093 cc/g	0.046 cc/g
CO ₂	5.07%	7.5%
Ar	< 0.05	0.2
O ₂	0.06	1.6
N ₂	67.2	52.6
CO	19.1	15.6
He	< 0.01	< 0.1
H ₂	8.45	21.9
CH ₄	0.07	0.1
C ₂ H _x	< 0.1	< 0.5
C _x H _y	< 0.1	< 0.1
SO ₂	< 0.05	< 0.1
NO	< 0.1	< 0.1

Agreement of the experimental data with calculations for the pellet rods could be obtained if it was assumed 28-29% of the adsorbed gases reacted. However, pressures produced in the powder rods were greater than could be accounted for even assuming 100% reaction of the adsorbed gases. This may be due in part to inaccurate analyses; however, it appears that further experimental work is necessary before calculations can be made on pressure buildup with any degree of accuracy.

HTLTR Driver Fuel Elements - R. E. Sharp and R. J. Lobsinger

The area of major concern in the design of the driver fuel elements for the HTLTR has been the selection of a suitable cladding material. The choice of materials is limited because of the extremely severe environment which will be encountered in reactor (nitriding, carburizing atmosphere at 1000 °C). Previous environmental testing and evaluation indicated only graphite and TD nickel would be suitable. Problems in procurement, core compatibility, joint closure and irradiation effects preclude the use of TD nickel at this time. The suitability of graphite cladding under HTLTR

conditions (1000°C , 2×10^{10} nv, gas cooled) has been successfully demonstrated in the BR-1 located at Mol, Belgium. Results from these tests indicate that although gases diffuse through an unimpregnated graphite shell within a few seconds, less than 0.1% of the total I^{131} , I^{133} , and Te^{132} isotopes are released. Consequently, it will be possible to utilize a vented fuel element design to prevent internal gas pressurization and still retain detrimental fission products. However, with graphite cladding the possibility of breakage and resultant spread of contamination will necessitate the use of pelletized fuel material rather than high energy rate impacted particles.

Procurement of both graphite cladding and 5% enriched UO_2 pellets has been initiated.

Pneumatic Impaction of Tungsten Powders - P. L. Farnsworth

Experiments to improve the strength and ductility of pneumatically impacted tungsten powders (-5 and $-100\ \mu$) by postimpaction heat treatments and by better control of vacuum during preimpaction heating were conducted. These experiments are part of a study designed to provide preliminary information on the suitability of pneumatically impacted tungsten as a potential matrix material for dispersed UO_2 fuel elements.

The results of these experiments are as follows:

1. Postimpaction heat treatments do not improve either the strength or the ductility of pneumatically impacted tungsten powders.
2. Improved vacuum facilities for preimpaction heating resulted in apparent increases in strength of the pneumatically impacted tungsten powders; no evidence of ductility was observed.
3. These results indicate that atmospheric impurities (oxygen, nitrogen) picked up during preheating of the impaction cans may be responsible for the observed low strengths and absences of ductility.

The results of strength (modulus of rupture) tests made on these materials are tabulated in Table 5.5.

TABLE 5.5. Properties of Pneumatically Impacted Tungsten Powders: Effect of Preimpaction Heating Atmosphere and Postimpaction Heat Treatments (Impaction temperature: 1600 °C)

Powder Size, μ	Vacuum Apparatus for Preimpaction Heating	Postimpaction Heat Treatment	Modulus of Rupture, kg/cm^2
- 5	Rough pump	None	7520 - 8085
- 5	Rough pump	1750 °C for 10 hr in H_2	4780
- 5	Rough pump	1500 °C for 10 hr in H_2	6400 - 7590
- 5	Rough and diffusion pumps	None	7945 - 9560
-100	Rough pump	None	5485 - 6190
-100	Rough pump	1750 °C for 10 hr in H_2	5695 - 7590
-100	Rough pump	1500 °C for 10 hr in H_2	6330 - 6820
-100	Rough and diffusion pumps	None	7170 - 11,460

Fiber Reinforced Metal Matrix Composites - R. K. Robinson

Strengthening fuel element core and cladding materials is important to the development of efficient, economical, and advanced fuel concepts. The development of stronger cladding materials is required if fuel elements capable of operating for longer times, at higher burnups, and at higher temperatures are to be produced. One of the more promising approaches to increasing the strength (particularly at high temperatures) of cladding materials is the development of fiber-reinforced composites. Numerous methods exist for consolidating fiber-reinforced composites into dense, useful materials having a variety of applications. One rather unique powder metallurgy technique is a high energy-rate forming method known as pneumatic impaction.

Metal powders are added to random or oriented reinforcing fibers. The fiber-powder mixture is placed in a mold which is sealed in a metal heating and evacuation can. This assembly (billet) is evacuated and/or purged with gases and preheated to the desired bulk impaction temperature. The billet evacuation tube is then sealed and the billet quickly transferred to

the pneumatic impaction machine. Consolidation is achieved by the rapid deceleration of the impaction punch against the heated billet contained in a closed die. Pressures (nearly isostatic) generated by the decelerating punch range up to $28,000 \text{ kg/cm}^2$. The densified composite and the surrounding mold and canning materials are ejected from the die. Subsequent machining and/or chemical dissolution of the billet is required to remove the composite material. A flow chart indicating the general process steps used in this work is shown in Figure 5.30.

FIGURE 5.30. Process Flow Chart for High Energy-Rate Forming Fiber-Reinforced Metal Matrix Composites

The shape of the impacted composite is largely dependent upon the initial shape of the metal mold. Direct forming of composite mixtures to a specific shape is sometimes desirable. Pneumatic impaction of composite mixtures into rod or plate stock may be followed by secondary forming (extrusion, rolling, etc.) or machining.

Tungsten Fiber Reinforced Nickel Composites - R. K. Robinson
and P. L. Farnsworth

Composite plates containing nominally 10 vol% continuous, aligned tungsten fibers (0.025 mm diam) in pure nickel matrices were successfully prepared by pneumatic impaction. Good bonding between fibers and matrix and high densities ($\sim 99\%$ of theoretical) were achieved during impaction. Little or no breakage of the fibers occurred during fabrication and, for the most part, the fibers remained straight throughout the specimen length. Transverse sections of the composites indicated a random distribution of the fibers although they were loaded in an ordered array (Figures 5.31 and 5.32).

FIGURE 5.31. Tungsten in Nickel
(36X)

Neg. 465823A

FIGURE 5.32. Tungsten in Nickel
(36X)

Neg. 465823B

Significant increases in both the yield strength and the ultimate tensile strength were obtained in the continuous, axially aligned fiber reinforced nickel composites (Table 5.6). At room temperature, the yield strength (0.2% offset) of the fiber reinforced nickel was approximately 1.7 times that of an unalloyed impacted nickel standard. At 1050 °C (1925 °F), both the yield strength (0.2% offset) and the ultimate tensile strength of the reinforced nickel were about 5.5 times that of unalloyed nickel.

TABLE 5.6. Preliminary Tensile Strength Data for Pneumatically Impacted Tungsten Fiber Reinforced Nickel Composites

Sample No.	Material	Test Temperature, °C	0.2% Yield Strength, kg/cm ²	Ultimate Tensile Strength, kg/cm ²	Elongation, %	vol% Tungsten Fibers, 0.001 mm diam
C-N-1	Ni (unalloyed)	25	2420	3745	40	0
C-N-2	Ni (unalloyed)	1050	220	245	6	0
4D-N-1	Ni-Aligned W Fibers	25	4085	4315	12	0.239
4D-N-2	Ni-Aligned W Fibers	1050	290	290	2	0.018
4D-N-3	Ni-Aligned W Fibers	1050	1335	1335	8	0.231
4D-N-4	Ni-Aligned W Fibers	1050	490	520	3	0.122
1A-NW-2	Ni-Random W Fibers	25	2445	3770	30	0.025
1A-NW-1	Ni-Random W Fibers	1050	230	240	5	0.025

At room temperature, fractures in the tungsten fibers do not propagate through the nickel. Many fractures of the tungsten fibers occur before the composite fails (Figure 5.33). Some localized shear on the nickel-tungsten interface occurs near the tungsten wire fracture area. At 1050 °C (1925 °F), fracture in the tungsten fibers does propagate through the nickel (Figure 5.34). The nickel fails intergranularly. Diffusion between the nickel matrix and tungsten fibers is appreciable during testing at 1050 °C (Figure 5.35).

Small additions (~1 vol%) of randomly oriented, chopped tungsten fibers (0.001 in. diam) did not appreciably affect the strength of pneumatically impacted nickel.

FIGURE 5.34. Tungsten in Nickel (36X)

FIGURE 5.33. Tungsten in Nickel (36X)

FIGURE 5.35. Tungsten in Nickel (500X)

"Whisker" Reinforced Metal Matrix Composites - R. K. Robinson

Alumina whickers 1 to 10 μ diam were incorporated into aluminum, molybdenum, and nickel matrices. No mechanical testing was performed on the aluminum matrix composite. Several molybdenum matrix test specimens were fabricated, but brittle fractures occurred at the (1) pinhole and (2) radius leading to the gage length. Elevated temperature tests on molybdenum were inconclusive due to vaporization of the molybdenum during heating under an inert gas purge.

Alumina reinforced nickel composites were prepared. Several as-impacted specimens were tested (~ 10 and 20 vol%), and specimens were prepared from an impacted-extruded (8:1 reduction) rod. Tensile tests on these materials are summarized in Table 5.7. Note four point bend tests were used in conjunction with tensile tests on several specimens.

TABLE 5.7. Mechanical Properties of Whisker Reinforced Metal Matrices

Composite Fiber-Matrix	Reinforcement		Test Temperature, °F (°C)	Yield Strength, kg/cm ²	Tensile Strength, kg/cm ²	Elongation, %	Effective Strengthening	
	vol%	diam, mils					Yield	Tensile
Al ₂ O ₃ -Ni ^(a)	(10)	1 - 10	Room	3260	5170	22	(1.4)	(1.1)
Al ₂ O ₃ -Ni	(20)	1 - 10	Room	5400 - 6250 ^(b)	5400 - 6250 ^(b)	2	2.4 ^(b)	1.2
SiC-Ni	(10)	1 - 10	Room	3750	4190	--	(1.6)	(0.86)

(a) Subsequently extruded

(b) Range bases on tensile tests and calculated fiber stress from four point bend tests

Silicon carbide whiskers have been added to aluminum, nickel, and titanium. Preliminary evaluation and testing of 10 vol% SiC-Ti indicated laminations in the composite as if preferential segregation of the whiskers had occurred. Also, some silicon carbide whiskers used contained contamination.

Polishing for metallographic examination of these materials is difficult due to whisker fracturing with subsequent pull out and because of the relative hardness of the whiskers and matrices.

Metallography of the Al₂O₃ molybdenum and nickel systems are indicated in Figures 5.36 and 5.37.

Randomly Oriented Fiber-Metal Composites - R. K. Robinson

Initial studies in fiber reinforcement of metal matrices are concerned with 0.025 and 0.125 mm diam W filaments randomly distributed in aluminum, nickel, stainless steel matrices, tungsten, and W-UO₂ cermets. Filament lengths varied from 1/8 to 1/2 in. long. Volume percent loadings ranged from approximately 1 to 10%.

As-impacted microstructures of these early fiber-reinforced components are shown in Figures 5.38, 5.39, 5.40, and 5.41. The extremely soft aluminum reinforced with tungsten filaments is difficult to polish. Metal matrices were formed from powders ranging from <44 to 66 μ . Impaction pressures ranged from 14,000 to 24,500 kg/cm².

Tensile tests on several composite plate specimens with small (~ 1 to 5% U/O) fiber addition indicated little or no strengthening for the nickel and stainless steel composites and considerable loss in strength for the aluminum matrix composite. Mechanical testing was not performed on the W-W, W-W-UO₂ systems. Only a couple of specimens from each composite were prepared and tested.

FIGURE 5.36. α - Al_2O_3 Whisker Reinforced Metal Composites (20 vol% Al_2O_3 Reinforced Molybdenum) (1000X)

FIGURE 5.37. As-Impacted Whisker Reinforced Metal Composites (20 vol% Al_2O_3 Reinforced Nickel) (1000X)

FIGURE 5.39. Tungsten in Stainless Steel
Matrix (500X)

FIGURE 5.38. Tungsten in Nickel
Matrix (500X)

FIGURE 5.41. Tungsten Fibers in Tungsten-UO₂ Cermet (100X)

FIGURE 5.40. Tungsten Fibers in Aluminum (100X)

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