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***Mound Laboratory Isotopic
Power Fuels Programs:
April-June 1972***

August 31, 1972

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AEC Research and Development Report

MOUND LABORATORY

Miamisburg, Ohio
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MONSANTO RESEARCH CORPORATION

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U. S. ATOMIC ENERGY COMMISSION

U. S. Government Contract No. AT-33-1-GEN-53

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PRINTED IN THE UNITED STATES OF AMERICA

Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.95

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Foreword

This report presents the technological achievements of Mound Laboratory in its isotopic fuels programs during the past three months. The separate reports compiled here represent the work of the following sections of the Nuclear Operations Department (Director W. T. Cave): Nonweapons Programs (Manager R. E. Vallée), and Quality and Reliability (Manager D. P. Kelly).

This work is supported by the AEC Division of Space Nuclear Systems, Division of Isotopes Development, Division of Reactor Development and Technology, and Division of Military Applications.

Summaries of all sections are compiled at the front of this report. Brief introductions to the work are included with each section.

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MLM-1827
MLM-1887
MLM-1905

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Plutonium-238 Fuels

FUEL PREPARATION

Multihundred Watt Development Five PPO (pressed plutonium oxide) spheres (SPO-22 through SPO-26) were fabricated during this report period. All the spheres were vacuum hot pressed at 1480°C at ~2500-lb load. The feed for all pressings was hydroxide-precipitated $^{238}\text{PuO}_2$ particles that had been oxygen-16 exchanged and fired at 1200°C for 2-4 hr. All batches of feed particles passed through a U. S. Standard Sieve, series No. 50 (297 μm). Four of these spheres, SPO-23 through SPO-26, were fabricated for encapsulation as part of the safety sequential test program for the Multihundred Watt program. SPO-22 is serving as a control sphere for this production sequence. The PPO sphere property data on these units are given in Table 1.

The spheres were given an oxygen-16 treatment after fabrication. In this treatment, after the sphere has reached a temperature of 600°C in a ceramic liner, oxygen-16 is passed over the sphere for 1 hr. The temperature of each sphere ranges from 600 to 750°C during this operation. All the spheres were subjected to an acoustical emission analysis during the cycle of heating, oxygen-16 flow, and cooling. The acoustical emission data are being accumulated and hopefully will be correlated to physical or chemical changes occurring with each sphere during the oxygen-16 treatment. All of the spheres were calorimetered, neutron counted, outgassed at 1200°C for 30 min at 1×10^{-5} torr, and gaged with a set of micrometers. These data are included in Table 1. The spheres (SPO-23 through SPO-26) were encapsulated in iridium shells on May 9, 1972. SPO-22 was subjected to a neutron spectral analysis to characterize the radiation properties of this fuel form. SPO-22 was transferred to another laboratory for an extended thermal ramping and soak test (to be performed in mid-July) and a drop test, and then will be sampled for metallographic, carbon, and impurities analyses.

The SPO-19 analytical sphere was sampled for analysis. Six samples, taken for carbon analysis, all indicated carbon levels <100 ppm (0 to 51 ppm). The total impurities by emission spectroscopy of the destructed sphere was <2010 ppm. Table 2 shows the emission spectroscopy results on SPO-19. Table 3 shows the results of metallographic analysis of SPO-19. The maximum grain sizes ranged from 30 to 100 μm , and the densities ranged from 74 to 96% theoretical density. The

densities were obtained by image analysis on a Classimat Quantitative image analyzer. The maximum grain sizes were measured from etched mounted samples using a standard scale photograph at a known magnification.

Table 1

PPO SPHERE PROPERTY DATA

<u>Sphere</u>	<u>Diameter at 20°C (in.)</u>	<u>Geometric Density^a (g/cc)</u>	<u>Power (W)</u>	<u>Specific Neutron Emission Rate n/sec/g-Pu</u>
SPO-22 ^b	1.471 - 1.479	9.3	100.34	4810
SPO-23	1.447 - 1.472	9.7	100.16	4680
SPO-24	1.452 - 1.473	9.6	99.76	5060
SPO-25	1.458 - 1.475	9.4	99.83	5110
SPO-26	1.472 - 1.479	9.1	100.11	4860

^a Based on pole-to-pole diameter corrected to 20°C.

^b Analytical Sphere

Table 2

EMISSION SPECTROSCOPY RESULTS FOR SPO-19

<u>Element</u>	<u>Impurity (ppm, sample weight)</u>
Al	<50
Ca	<250
Cd	Not Detected (<20)
Co	50
Cr	400
Cu	<50
Fe	600
Mg	<20
Mn	<20
Na	Not Detected (<250)
Ni	<50
Pb	Not Detected (<20)
Si	70
Sn	Not Detected (<10)
Ta	Not Detected (<100)
Zn	Not Detected (<50)

Table 3

SPO-19 METALLOGRAPHIC SAMPLES

<u>Mount No.</u>	<u>Location</u>	<u>Grain Size (μm)</u>	<u>Density (% T.D.^a)</u>
424	Equator & above	40	80
425	Midway (center to surface)	100	74
426	Pole	80	76
427	Core	100	79
428	Exterior Surface (1/4 in. inward)	30	74
429	Equator	90	96

^a Theoretical Density.

Table 4 indicates the grain sizes for SPO-16 which ranged from 25 to 110 μm . (SPO-16 was fabricated before the SST spheres and was destroyed for complete metallographic analysis.) (W. A. Zanolli, W. D. Pardieck, and B. R. Kokenge)

Table 4

SPO-16 GRAIN SIZES

<u>Mount No.</u>	<u>Location</u>	<u>Grain Size (μm)</u>
303	Pole	25
304	Core	110
305	Equator	35

Coating of PMC Discs With Molybdenum and Tungsten A study is under way to develop a method for coating PMC (plutonium-molybdenum cermet) discs with either of the refractory metals molybdenum or tungsten. At first the hydrogen reduction of the metal hexafluorides was studied. Seven simulant (molybdenum discs were used as simulants for PMC throughout this study) discs were coated with tungsten and 12 with molybdenum by tumbling a simulant disc inside a rotating graphite barrel.

The results of these runs indicate that high quality tungsten coatings can be deposited on molybdenum simulant discs over a rather broad range of conditions. Also, suitable molybdenum coatings can be deposited on simulant discs with careful control of all conditions.

Following the runs discussed above, the barrel coater was modified for use with MoCl_5 . This modification consisted of (1) the design, fabrication, and installation of a chloride vaporizer; (2) repiping of the reactant gas lines to minimize their length and allow their being heated to at least 200°C to ensure transport of MoCl_5 ; (3) conversion of the water-cooled inlet tube to a controlled-temperature gas-cooled inlet tube; (4) procurement and installation of temperature controllers for the chloride vaporizer, gas lines, and inlet tube; and (5) installation of a two-staged scrubber for removal of HCl and MoCl_5 from the exhaust gases.

These modifications were completed and seven simulant discs were coated with molybdenum using this system with MoCl_5 . The best coatings obtained by this method were porous and somewhat nodular.

An alternate system for coating with MoF_6 was suggested by Battelle Memorial Institute. Their system consists of a dual reactant gas inlet system with one inlet tube directed at each surface of the disc, while the disc is rolled vertically in a graphite barrel. The MRC system was modified to incorporate these features.

Three simulant discs were coated with molybdenum from MoF_6 using this modified system. The discs were coated at stoichiometric H_2/MoF_6 ratios of 1.8, 8.5, and 6.0, respectively.

During the first run, the disc was severely etched by the MoF_6 and lost 2.4 g. Although the second disc obtained a 5-g coating, the coating did not adhere well to the back surface. Photomicrographic analysis indicates that the surface received a 6-mil coating, while the front surface received a 3-mil coating even though identical amounts of reactant gases were introduced through each tube. The third coated disc also exhibited a nonadherent coating especially on the back surface. Again the coating on the back surface was thicker (4.5 vs. 1.2 mil).

Operational modifications were necessary before using the dual inlet system to deposit tungsten. One disc was coated with tungsten by introducing WF_6 and a near stoichiometric quantity of H_2 through the front inlet tube and a large excess of H_2 through the back inlet tube. The front surface of the disc received only a light <1-mil coating while the back surface and chamfer received a dense uniform 5.5-mil coating (Figure 1). This coating, typical of those deposited from WF_6 , was deposited at the rate of 0.09 g/min.

A second disc was coated with tungsten by introducing the reactant gases first through the front tube and then through the rear tube. A uniform, dense, 2-2.5 mil coating was deposited on all surfaces. This coating was deposited at a rate of 0.08 g/min or approximately 2 mil/hr.

The findings derived from the above may be summarized as follows:

- Tungsten is much easier to deposit than molybdenum with either system.

- Tungsten coatings are generally of a higher quality than molybdenum coatings.

FIGURE 1 - Photomicrographs of 5.5-mil tungsten coating on molybdenum discs; a) chamfer, b) surface. 100X

- With present technology PMC discs could most probably be satisfactorily coated with tungsten from WF_6 using either the single inlet-tumbling barrel coater or the dual inlet-rolling barrel coater.
- The single inlet-tumbling design is significantly easier to load and should not require any major modifications for coating the tapered PMC end pieces.
- A suitable molybdenum coating can be deposited from MoF_6 by coating at a slow rate ($<1/2$ mil/hr) and tumbling.
- No suitable molybdenum coating was obtained from the three runs carried out with the dual inlet system using MoF_6 .
- No suitable molybdenum coating was obtained from several runs using the H_2 reduction of $MoCl_5$ and tumbling.

It is felt that with present knowledge, a consistently satisfactory molybdenum coating on PMC discs could not be obtained. (R. R. Jaeger)

Encapsulation of Plutonium in Molybdenum Cups The Modular Heat Source concept is being considered for RTG systems. This concept has created interest in the encapsulation of plutonium inside molybdenum cups via hot pressing techniques. Initial pressings were made using ThO_2 powder. Three discs were hot pressed and will be used for impact testing of capsule units.

The first disc (nominal diameter 2.128 in.) was pressed using a graphite die (Figure 2) lined with 0.001 in. tantalum and 0.001 in. molybdenum foils. The punches were lined with 0.010 in. Grafoil, 0.001 in. tantalum, and 0.001 in. molybdenum foils. Five-mil molybdenum was used to fabricate the cup and lid. The molybdenum cup was charged with 140 g of 53-105 μm ThO_2 powder. The disc was pressed at 1850°C using 4000 psi pressure. The die closed using these conditions producing a disc having a density of 8.4 g/cc. The molybdenum cup was extremely brittle. The molybdenum around the circumference of the disc broke loose and fractured into several pieces. However, the molybdenum adhered to the disc faces.

In an effort to reduce the embrittlement of the molybdenum cup the second die was lined with 0.005 in. tantalum and 0.005 in. molybdenum foils. The punches were lined with 0.010 in. Grafoil, 0.005 in. tantalum, and 0.005 in. molybdenum foils. The molybdenum cup size was not changed. The cup was charged with 140 g of ThO_2 powder. The disc was pressed at 1450°C using 2600 psi pressure. The die did not close under these conditions. The density of the disc was 7.0 g/cc. The molybdenum came loose from both the circumference and faces of the disc. The exposed ThO_2 tended to crumble.

FIGURE 2 - Graphite die used for hot pressing ThO_2 powder inside a molybdenum cup.

The third disc was pressed using die and foils similar to the second. The disc was pressed at 1850°C using 3700 psi pressure. The die was closed producing a disc having a density of 9.3 g/cc. The molybdenum adhered to both the circumference and faces of the pressed ThO_2 . Also, the molybdenum did not appear as brittle as the molybdenum on the first disc. The results of this pressing were promising in that the ceramic was enclosed within the molybdenum shell after removal from the graphite punch and die assembly.

All of the above-described, hot-pressed assemblies were welded into stainless steel capsules for impact testing. (W. D. Pardieck and B. R. Kokenge)

Pellet Fabrication A study was undertaken to ascertain the feasibility of cold pressing PuO_2 fuel material for use in the Milliwatt heat source program. The present procedure requires the pouring of 6 W(th) of high-fired $^{238}\text{PuO}_2$ particles into the primary capsule, then welding the primary. A pellet form would have the advantages of being a safer form in which to handle the fuel material and of reducing the volume required in the present fueled capsule.

Preliminary pressings were made using a Carver bench press and a 0.5 in. diameter infrared pellet die. Fuel simulant materials consisted of ThO_2 and UO_2 powder $<53 \mu\text{m}$. Pressings were performed to determine what pressing parameters would be required to obtain a pellet of sufficient green strength and density to enable safe handling of the material after pressing. Results of these preliminary pressings are shown in Table 5. The ThO_2 pellets pressed at pressures less than 30 kpsi did not meet the above criteria of sufficient green strength. A plot of pressure as a function of green density is shown in Figure 3. The UO_2 was found to require much lower pressures to yield a suitable pellet.

Table 5

ThO_2 PELLET PRESSINGS

<u>Charge (g)</u>	<u>Pressure (psi)</u>	<u>Time at Pressure (min)</u>	<u>Green Density (%)</u>	<u>Remarks</u>
13.9	20,000	15	60	Poor green strength. Fell apart after sintering.
13.9	30,000	15	62.4	Top came off after sintering. Fair green strength.
13.9	30,000	15	65	Good green strength.
13.8	51,000	15	67.1	Good green strength.
13.8	60,000	15	67.1	Good green strength.

FIGURE 3 - Green density of ThO_2 pellets as a function of pressure.

Once the pressure was established, a punch and die specially designed with a chamfered top punch and hemispherical bottom punch was fabricated. A drawing of this die is shown in Figure 4. This design was chosen in an attempt to eliminate any sharp edges on the fuel pellet because of their unfavorable reentry and safety characteristics. All attempts to cold press a pellet into this configuration failed. The pellets invariably broke at the top edge of the hemispherical punch during removal. An example of this is shown in Figure 5. Various techniques were employed in an attempt to overcome this problem but each was unsuccessful. Since the top chamfered punch yielded a well formed top, a new design incorporating a chamfered bottom punch might produce an acceptable product. Figure 6 shows the modified die configuration.

FIGURE 4 - Single chamfered hemispherical milliwatt die.

FIGURE 5 - UO_2 hemispherical bottom punch.

FIGURE 6 - Double chamfered milliwatt die.

Preliminary cold pressing with ThO_2 using this die was generally unacceptable due to the cracking at the chamfered edge as shown in Figure 7. The overall problem with this particular configuration as well as with the previous one was due to the large l/d (length-over-diameter ratio) associated with pressing 13 to 14 g of material into an acceptable pellet size. Pressing this material into a 0.5 in. diameter die results in a pellet with an $l/d \approx 2$. Normally pressings with $l/d > 1$ are considered a deviation from ideal pressing conditions. Therefore, it was decided to cut the charge in half and press two separate UO_2 pellets thus considerably reducing the l/d ratio. These could then later be stacked.

FIGURE 7 - ThO_2 double chamfered die.

This technique proved to be successful. Rejection rate was extremely low.

The UO_2 pellets were sintered in an NRC vacuum furnace for 1 hr at 1400°C at 1×10^{-5} torr. The sintered pellets appeared to have excellent strength and exhibited no surface defects or distortions. Figure 8 and 9 show typical double-chamfered pellets after pressing and sintering. Figure 10 is a typical cross-section of a sintered pellet. The internal stress cracking observed is probably due to sintering. It is felt this can be eliminated with a more controlled thermal cycle. Figure 11 is a high magnification composite photomicrograph indicating uniform density throughout the pellet.

Table 6 gives all the pertinent data collected on the sintered UO_2 pellets.

FIGURE 8 - Sintered UO_2 double chamfered stacked.

FIGURE 9 - Sintered UO_2 double chamfered pellets.

FIGURE 10 - UO_2 cross section; $11\frac{1}{2}\times$ magnification.

FIGURE 11 - Composite photograph of sintered UO_2 pellet; 500X magnification (reduced to 85%).

Table 6

UO₂ <53 μm FEED

No.	Charge (g UO ₂)	Green Density (%)	Sintered Density ^a (%)	% Shrinkage		Binder	Pressure (psi)	Comments
1	7.0	58.8	83.5	Diam.	11.7	Carbowax	18,000	Good green strength
				Hgt.	10.5			
2	7.0	57.6	83.3	Diam.	12.2	Carbowax	15,000	Good green strength
				Hgt.	11.0			
3 ^b	7.0	57.8	84.1	Diam.	12.4	Carbowax	17,000	Good green strength
				Hgt.	11.0			
1A	6.88	57.8	85.6	Diam.	12.8	H ₂ O	15,000	Good green strength
				Hgt.	12.1			
2A	6.90	57.4	85.1	Diam.	12.8	H ₂ O	17,000	Good green strength
				Hgt.	11.9			
3A ^b	6.80	57.2	85.8	Diam.	13.2	H ₂ O	18,000	Slight lamination after sintering
				Hgt.	12.6			
1C	7.40	59.1	85.7	Diam.	11.9	H ₂ O	12,000	Good green strength
				Hgt.	11.3			
2C	7.0	59.0	87.0	Diam.	11.7	H ₂ O	15,000	Good green strength
				Hgt.	11.0			
3C	14.5	61.8	90.2	Diam.	12.0	H ₂ O	30,000	Good green strength
				Hgt.	11.1			

^aAll UO₂ pellets sintered at 1400°C - one hour under vacuum.^b3 and 3A were double chamfered configurations.

Table 6 (Continued)

<u>Material</u>	<u>Form</u>	<u>Charge Weight (g)</u>	<u>Pressure (psi)</u>	<u>Green Density (% T.D.)</u>	<u>Die Lubricant</u>	<u>Binder</u>	<u>Comments</u>
UO ₂	SC-RCC	7.0	30,000- 70,000		Colloidal Graphite	None	Laminated at high psi
UO ₂	DC-RCC	7.0	30,000- 70,000		Colloidal Graphite	Carbowax	Laminated at high psi
UO ₃	DC-RCC	7.0	15,000	57.7	Colloidal Graphite	Carbowax	Good pressing. Sintered at 1400°C - 1 hr 84.1% T.D.
UO ₂	SC-RCC	7.0	15,000	58.8	Colloidal Graphite	Carbowax	Good pressing. Sintered at 1400°C - 1 hr 83.5% T.D.
UO ₂	SC-RCC	7.0	15,000	57.8	Colloidal Graphite	Carbowax	Good pressing. Sintered at 1400°C - 1 hr 83.3% T.D.

Concern was expressed that oxidation of the UO_2 might be occurring during the handling of the UO_2 pellets. Consequently, several samples were taken for O/U analysis: starting material and interior and exterior samples from sintered pellets. These samples were analyzed by TGA at 850°C in air. Results of these analyses are given in Table 7. As expected, the amount of oxidation was insignificant.

Table 7
O/U RATIOS

<u>Sample^a</u>	<u>O/U Ratio</u>
Alpha Inorganic low-fired UO_2	$\text{UO}_2.07$
1 Pellet (interior)	$\text{UO}_2.03$
1 Pellet (exterior)	$\text{UO}_2.01$
1A Pellet (interior)	$\text{UO}_2.02$
1A Pellet (exterior)	$\text{UO}_2.05$

^aAll samples run in duplicate.

It has been demonstrated that simulant fuel material for use in the Milliwatt heat source program can be successfully pressed into pellets. These pellets when pressed, with a reduced ℓ/d and water binder, exhibit sufficient green strength and density to be handled quite safely prior to sintering. The sintered pellets appear to have excellent strength and no surface defects at densities between 84-89% of theoretical.
(B. R. Kokenge and R. L. Deaton)

FUEL CHARACTERIZATION

O/Pu Ratio in PPO Fuel Two series of tests were performed to determine optimum conditions for lowering the O/Pu ratio of PPO (pressed plutonium oxide) fuel samples. These experiments were carried out in a micro high-temperature vacuum furnace. The material used in these tests consisted of residue collected from Multihundred Watt sphere SPO-7 after destructive testing had been completed. This material consisted mainly of small chips weighing between 100-200 mg.

The first series of tests was run in an attempt to determine whether rapid reduction (less than 1 hr) was possible without regard to the temperature requirements. Samples were heated for 30 min to between 1650-1950°C at a pressure of 5×10^{-5} torr. The samples were cooled at the same constant pressure and then analyzed by TGA correcting for actinides. It was determined that, under these conditions of pressure and time, a minimum temperature of 1700°C is required to effect any reduction in the particular sample size used. It should also be noted that sample vaporization became significant when the temperature was increased to 1900°C. This vaporization obviously would eliminate temperatures this high when the process is scaled up to a full size PPO sphere due to the large ΔT across such a sphere.

The second series of tests was initiated to determine whether, by increasing the time cycle, the same degree of reduction could be accomplished at a lower temperature. This decrease is desirable because of the large ΔT associated with a sphere containing 100 W (thermal) of fuel. Calculated values have shown this gradient could be as much as 150-250°C in excess of the furnace temperature.

For these tests, time at temperature was increased from 30 min to 5 hr while the pressure remained the same. The results of the second test series suggest that 1650°C is the minimum temperature required to effect any degree of reduction.

Figure 12 shows the data plot from both series of tests. Since the physical size of the apparatus used in this study permitted only small portions of a Multihundred Watt sphere to be tested, it is difficult to determine whether the entire sphere, under the same conditions, would yield a similar average O/Pu ratio; e.g., large thermal gradients across the sphere could cause variations of O/Pu ratios across the sphere. Other factors to be considered are the inherent density gradients that could influence the diffusion of oxygen. Thus, without running a series of tests on full sized PPO spheres, these potential difficulties would be hard to assess. (R. L. Deaton and B. R. Kokenge)

FIGURE 12 - Stoichiometry as a function of temperature for SPO-7.

Effect of Outgassing Procedures In an attempt to better characterize the simulant fuel forms and establish ideal outgassing procedures of these fuel forms, tests are being conducted using residual gas analysis as the primary mode of investigation. Such testing will be expanded to the actual plutonium-238 fuel forms at a later date when the proper facilities are completed.

In the present study four samples of simulant fuel have already been tested with two samples remaining to be studied. The tested samples consist of molybdenum-coated thoria particles, surface samples of unoutgassed and outgassed thoria spheres, and finally a sample from the core of an outgassed thoria sphere. The two samples remaining to be tested consist of pieces removed from the center of hemispherical portions of a thoria sphere that have been air outgassed. The hemispheres will be vacuum outgassed using different thermal profiles prior to testing.

The results of the completed tests are presented below:

Molybdenum-Coated Thoria Particles This sample consisted of 2 g of thoria particles, coated with 22 wt % molybdenum by the hydrogen reduction of MoCl_5 at 750°C .

This sample was analyzed using an Aero-Vac mass spectrometer. The sample was contained in a stainless steel vessel mounted in a "line-of-sight" fashion to the mass spectrometer. The sample was evacuated to a total pressure of 3×10^{-4} torr prior to the initiation of the test. Also prior to the start of the test, background runs (scans) were made on just the mass spectrometer itself and then on the entire sampling system. The background in both cases was measured at a partial pressure of $\sim 1 \times 10^{-8}$ torr. Other than some residual organic fractions that were observed in the mass spectrometer background, nothing of significance was observed. The actual test consisted of heating the sample tube by resistance heating and monitoring the offgassing products at various times during the thermal profile. The sample was heated from ambient temperature ($\sim 25^{\circ}\text{C}$) to 755°C during the course of the test. Mass spectrometer traces were made at approximately 50°C intervals. The partial pressure of the test was $\sim 1 \times 10^{-8}$ torr. This partial pressure was accomplished by throttling down the sampling port.

The test results showed that at all temperatures greater than 60°C there is a large increase ($>50\text{X}$ at 1×10^{-8} torr) in the intensity of masses 35, 36, 37, and 38. This implied that elemental chlorine and hydrogen chloride were being emitted from the sample. Since chlorine and hydrogen chloride are the byproducts from the thermal decomposition and hydrogen reduction of the molybdenum pentachloride during the coating operation and since the sample had not been exposed to any elevated temperature after the coating operation, the presence of these products was expected. There was no increase above background of any other observable products.

Total test time was approximately 2.5 hr, with a 1.5 hr soak at 755°C . Masses 35, 36, 37, and 38 showed no decrease from their maximum values during the entire test run.

Surface Pieces of Sphere STO-2 This sample consisted of a 2 g surface sample of thoria sphere, STO-2. The sphere was produced by pressing approximately 222.5 g of thoria particles (53-246 μm in diameter) with a ram force of 9800 psi at 1850°C for 15 min. The sphere was later air treated at 1100°C for 30 min.

The sample consisted of powder material scraped off the surface of the sphere. The sample was placed in the same test configuration as the previous sample, and the total and partial pressures were the same as in the previous test. The test temperature ranged from ambient to 550°C . As in the previous test, backgrounds at ambient temperatures were observed.

During the test, masses 12, 24, and 28 showed a very definite increase while the other species showed very little increase above background. Water was also present and increased to a maximum at approximately 200°C after which its intensity decreased. The masses 12 and 24 increased approximately 65X and 10X, respectively, above background. Mass 28 increased 18X above background. The significance of this study is that

it indicates that some elemental carbon is still present on the surface of the sphere after the air treatment. The dies and rams used in the pressing process are undoubtedly the source of the carbon impurity.

Surface Pieces of Unoutgassed Sphere STO-2 When thoria sphere STO-2 was pressed and removed from the die, it was discovered that a portion of the outside surface of the sphere had broken away due to the longer hold time at such an elevated temperature. This portion was set aside and was not exposed to the air treatment.

Approximately 2.1 g of this untreated material was mounted in the sampling system of the quadrupole mass spectrometer. Ambient temperature background scans were again made, then the sample was heated from ambient to 480°C, the maximum temperature being limited by the experimental setup. At all temperatures above 50°C, masses 12, 28, 41, 42, 43, 57, and 77 were detected along with the water group. The intensity of water reached a maximum at 235°C then decreased to a level slightly above background. Mass 12 (C^+) increased 4X above background while the remaining masses showed sizable increases, ranging from 10X for mass 77 to >100X for mass 28. At approximately 350°C the higher order organic masses (>28) started to decrease in intensity, and masses 40, 41, 42, and 55 were slightly above background by 475°C. Mass 28 remained intense throughout the test period. This mass number would indicate the possible presence of N_2^+ , CO^+ , and $C_2H_4^+$. When the sample was removed from the test it had lost 0.1 g of its weight.

When these data are compared with the data on the outgassed surface of the sphere, it is obvious that the outgassing in air does have some "cleaning" effect on the surface of the sphere.

Core Sample of Sphere STO-2 Since sphere STO-2 was damaged during pressing and was of no further use, a core specimen of the sphere was obtained to see what remained in the core after the air treatment. The sphere was broken into two approximate hemispheres and a center disc. The two hemispheres were set aside for vacuum outgassing and the center disc was used in the present study.

The disc weighing 7.16 g was loaded into the sampling system of the quadrupole mass spectrometer. The system was evacuated to a total pressure of 9×10^{-8} torr and again background runs were made. The test temperature ranged from ambient to 700°C and several of these thermal cycles were made.

As soon as heat was applied to the sample the total pressure rose to approximately 7×10^{-6} torr and offgassing became significant. Initially masses 12, 14, 16, 17, 18, 28, 32, 41, 42, 57, and 77 were observed to have increased in intensity. The rate of increase varied from 3X for mass 12 to >100X for masses 28 and 32. During the heating, mass 32 immediately peaked out and then decreased to the point that it was no longer detected at temperatures above 200°C. At approximately 550°C a drastic change occurred in the outgassing characteristics. Masses 2,

12, 14, and 29 increased substantially. Mass 2 increased 45X; mass 29 increased 100X, and mass 12 increased 8X above its intensity at the lower temperature. Masses 55 and 77 decreased to background while masses 41 and 42 decreased by approximately a factor of 6. Mass 28 remained at maximum intensity throughout the run and water decreased significantly at about 450°C.

During each of the four thermal cycles the same outgassing pattern was evidenced although at the peak of the fourth cycle the total sample pressure had dropped to 2.8×10^{-7} torr, thus indicating that the sample was being "cleaned up".

After the testing period the sample was removed from the test fixture and was observed to have discolored. Instead of the white color prior to testing, the surface of the sample was steel-gray. This indicated the possibility of sample-container reaction. This reaction was later substantiated when the sample was weighed and found to have gained 0.04 g in weight. The sample was then broken and it was noted that the discoloration was the same throughout the material.

The significance of this test is that it indicates the air treatment is not a sufficient process for cleaning up the interior of ThO₂ spheres.

The presence of the hydrocarbons such as acetylene is probably a result of the decomposition of various carbides in air. The air outgassing drives any organic (C₂H₂) + water reactant products "into" as well as "out of" the sphere. The possibility of inward diffusion is verified at the low temperatures of the test where air is observed as an off-gassing constituent. (An air signal is indicated by the masses 32, 28, 16, 14 in the ratio 12:3:4:1, and when mass 32 is observed in significant quantities the ratio of these masses is approximately 12:12:4.5:1. If mass 28, since it also indicates the presence of CO, is disregarded, there is a good indication that entrapped air is diffusing from the material.) The fact that at 550°C an increase of masses 2 and 12 is observed is an indication that decomposition of some of the organic material is occurring. This is further substantiated by the higher order organics decreasing in intensity while the lower order ones increase. Table 8 lists the possible species represented by the mass numbers detected in this series of tests. All the masses listed were not found in every test.

Future Studies The two remaining samples of STO-2 will be vacuum outgassed using different thermal cycles. These samples will then be broken and test specimens will be removed from the centers and tested in the same manner as the previous samples to see what effect the vacuum outgassing has on "cleaning up" the spheres. (R. E. Zielinski and S. T. Cohen)

Table 8

MASS SPECIES IDENTIFICATION

<u>AMU</u>	<u>Possible Species</u>
2	$(H_2)^+$
12	$(C)^+$
14	$(N^{14})^+, (CH_2)^-$
16	$(O^{16})^+, (CH_4)^+$
17	$(OH)^+$
18	$(H_2O)^+$
24	$(C)^+$
28	$(N_2)^+, (CO)^+, (C_2H_4)^+$
29	$(C_2H_5)^+$
32	$(O_2)^+$
35	$(C^{13}H)^+$
36	$(HC^{13}H)^+$
37	$(C^{13}H)^+$
38	$(HC^{13}H)^+$
41	$(C_3H_5)^+$
42	$(C_3H_6)^+$
43	$(C_3H_7)^+$
57	$(C_4H_9)^+$
77	$(C_6H_5)^+$

Creep Studies and Internal Structure Studies of Refractory Metals and Alloys

Introduction The inability to predict accurately the creep of refractory metals and alloys has long been a serious problem. Many investigations on this subject have been made with the purpose of obtaining information on the fundamental metallurgical mechanism and data for design. This present study will encompass the tensile and compressive creep testing of unirradiated and irradiated (10^{22} n-cm⁻² total fluence) specimens of molybdenum and molybdenum-50 wt % rhenium alloy and attempt to correlate these data with substructure studies of the tested material.

Most of the present data on unirradiated and irradiated refractory metals and alloys have been obtained from tensile creep tests, while few data exist on the compressive creep of unirradiated samples of this material and no data exist on the compressive creep of irradiated specimens. Normally, one would expect similar results from tensile and compressive creep studies on the same materials; however, when materials are irradiated, there exists the possibility that they may behave differently in tension and compression since the alignment of the internal damage might be different.

The internal structure will be studied using a Norelco, Model EM 300, Electron Microscope. One of the primary points of interest will be the loop densities of the materials since very little work has been reported on the analysis of loops in bcc metals.

Experimental Set-Up A Satec creep tester with a modified high vacuum Brew furnace attached was set up and is now operational. The unit as it now stands can be used for tensile creep studies in vacuum (2×10^{-6} torr) up to 1600°C. A modified load train was built to enable compressive creep studies to be performed on the same equipment. The creep tester has the ability to handle 3:1, 20:1, and dead weight loadings; therefore, a wide range of loadings can be used in the actual experiments.

Test Specimens The initial molybdenum test specimens were supplied by Dr. John Moteff of the University of Cincinnati. The working history of the samples was as follows: A machine casting, 9.126 in. in diameter, was extruded to 4.125 in. in diameter. The extruded material was rolled to 0.875 in. in diameter, then swaged to 0.250 in. in diameter. The stock material was then stress relieved for 0.25 hr at approximately 870°C. The last recrystallization of the rod occurred when the rod was at 2.0 in. in diameter. The final configuration of the test specimen is shown in Figures 13 and 14. The chemical analysis of the as-received samples is shown in Table 9. The samples received a 1-hr vacuum anneal at the test temperature prior to the initiation of the tests.

Experimental Data Four molybdenum rod specimens were tested in tension at 1400°C in vacuum. Each sample was exposed to a different stress level. In addition, two samples of molybdenum rod were tested in tension at 1150°C. All six of the unirradiated samples are being examined using the electron microscope.

FIGURE 13 - Molybdenum creep specimen.

FIGURE 14 - Molybdenum creep test specimen (dimensions in inches).

Table 9

ANALYSIS IN PPM OF AS-RECEIVED SAMPLES

<u>Element</u>	<u>Quantity (ppm)</u>
C	19
O	5.7
N	1
H	0.5
Fe	16
W	<100
Si	16
Mn	<1
Cu	3
Ca	--
Al	5
Cr	<1
V	--
Zr	<1
Ni	2
Ta	<1000
Ti	<1
Cd	<20
Mg	--
Sn	--
B	<10
Co	<5

Total impurities <1207

Table 10 lists the test conditions and results; the stress-rupture properties of the 1400°C specimens are given in Figure 15 and those for the 1150°C specimens are given in Figure 16.

No data analyses have been performed with the test results, since compression testing and testing at different temperatures have to be done to obtain a complete picture of the creep mechanisms involved.

Table 10

STRESS-RUPTURE RESULTS FOR UNIRRADIATED MOLYBDENUM

Specimen No.	Temp. (°C)	Atm. Pressure (torr)	Stress (kpsi)	Time to Rupture (hr)	Rupture Elongation (%)
Mo-5A4	1150	2×10^{-6}	11.5	47.0	42.56
Mo-7A4		4×10^{-6}	9.5	400.0	35.60
Mo-8A4	1400	2×10^{-6}	10.0	0.433	56.89
Mo-9A4		2.5×10^{-6}	7.5	5.7	34.29
Mo-10A4		6×10^{-6}	5.0	63.7	28.85
Mo-11A4		2×10^{-6}	3.5	211.2	30.32

FIGURE 15 - Stress-rupture properties of molybdenum at 1400°C.

Substructure Analysis In most cases, the problem of rationalizing the creep of metals resolves itself into identifying the movement of dislocations, the obstacles that impede dislocations, and the ways that thermal energy overcomes these obstacles. Plastic strain involves the movement of dislocations gliding over the crystal. When a dislocation moves completely across a crystal, it results in a displacement $\bar{b}$ (the magnitude of the Burgers Vector). The resulting macroscopic shear strain, ϵ , caused by dislocations can be expressed as $\epsilon = \bar{b}\rho d$, where ρ

FIGURE 16 - Stress-rupture properties of molybdenum at 1150°C.

is the dislocation density and d is the average distance moved by the dislocation. Therefore, to completely analyze a creep system a basic substructure analysis is required.

The tested samples are being examined by transmission electron microscopy for dislocation density, subgrain size, and loop density. These analyses are required, since each of the above play a role in affecting the creep rate of the test material.

The analyses have been completed on all samples except Mo-10A4 and Mo-11A4. Figure 17 is a plot of subgrain size as a function of stress. Figure 18 is a plot of dislocation density as a function of stress, and Figure 19 is a plot of loop density as a function of stress. All of the data follow expected trends but there are not enough data to project any conclusions. One significant point is that the subgrain sizes measured in this study are significantly smaller (approximately 1/10) than those measured by Carvalkinhos and Argent¹ at approximately the same stress levels. They performed compressive creep tests and this may account for the difference; however, most researchers have contended that there is very little difference in substructure in compression and tension. This will be examined when the compressive creep studies are performed. Another explanation for the difference in subgrain size is the difference in the method of analysis. Carvalkinhos and Argent employed x-ray techniques while the present study is employing electron microscope techniques which are more accurate.

FIGURE 17 - Subgrain size as a function of stress for as-tested molybdenum samples.

Figures 20 through 30 show the various structural features present in the samples. All the photomicrographs were made on thinned specimens using the electron microscope as the primary tool of investigation.

Dislocation networks were observed in the tested molybdenum samples. These networks are formed by dislocation interaction, which can occur when the reaction of two dislocations results in a lowering of the energy of the dislocations. The elastic energy per unit length of a dislocation line is proportional to b^2 (to a first approximation), thus a dislocation line tends to acquire the smallest possible Burgers Vector. A typical dislocation interaction in molybdenum would be

$$(a/2) [111] + (a/2) [\bar{1}\bar{1}1] = a[001],$$

where $3/4a^2 + 3/4a^2 > a^2$ and a is a lattice constant. This reaction can be represented as shown in Figure 20, where dislocations are interacting and the result is a nodal portion of a typical network. When families

of dislocation interact the network expands as illustrated by the hexagonal network shown in Figure 21.

FIGURE 18 - Dislocation density as a function of applied stress for molybdenum.

FIGURE 19 - Dislocation loop density as a function of applied stress for molybdenum.

FIGURE 20 - Typical dislocation reaction.

FIGURE 21 - Formation of a dislocation network in molybdenum.

Subgrain formation is a mechanism whereby the grains of a sample become fragmented into a number of smaller ones called subgrains. Subgrains have a misorientation relative to each other and the boundaries between them are composed of walls of dislocations. The driving force is the strain energy, since the energy of a group of dislocations piled up in a glide plane can be considerably reduced if they are rearranged into a small-angle boundary. Subgrains once formed may have an effect on the creep behavior of the metal. It is conceivable that subgrain boundaries can serve as crack initiators and thereby affect the performance of a material in service.

Figures 22 and 23 show the subgrains present in the molybdenum samples after testing. In Figure 22, arrow A shows a subgrain boundary; arrow B points to the slip that has occurred between subgrains; and arrow C indicates a free dislocation located in a subgrain. In Figure 23 arrow A points to the tie up of dislocations at a subgrain boundary and arrow B indicates a dislocation loop present in a subgrain.

FIGURE 22 - Subgrains formed in molybdenum during testing.

FIGURE 23 - Subgrains formed in molybdenum during testing.

Figure 24 shows two triple point subgrain boundaries (arrows A and B). Each of the boundaries consists of networks of dislocation and all three subgrains are misoriented with respect to each other.

Figure 25 is a high magnification photograph of a subgrain. Obvious here is the high dislocation density near the subgrain's boundaries (arrow A). The dislocations have migrated toward the boundaries leaving a low free-dislocation density in the interior of the subgrain. Eventually, the concentration of dislocations can reach a saturation value then begin to annihilate each other. It is obvious, however, that this particular specimen failed before the annihilation of the dislocations.

FIGURE 24 - Triple point subgrain boundaries in molybdenum.

FIGURE 25 - Dislocation density in a subgrain of molybdenum after test.

Figure 26 shows the tie up of dislocations in the tested molybdenum. Dislocations can be pinned by other dislocations, grain boundaries, or precipitate particles. In this photograph the tangles seem to be the result of other dislocations.

Dislocation loops can also occur in the matrix material. These loops enclose stacking faults. The loop exists entirely in the subgrain and separates a limited region of deformation from the underformed material. A loop can be either interstitial or vacancy in nature. If it is a vacancy type, the deformed region is the result of the precipitation of

FIGURE 26 - Dislocation tie-ups in tested molybdenum.

one layer of vacancies in the stacking matrix. If the loop is an interstitial type, the deformed region consists of one layer of interstitials. Figure 27 shows the presence of dislocation loops (arrows) in the tested molybdenum. The nature of the loops has not yet been determined.

Figures 28 and 29 show three dimensional dislocation networks that have formed in the tested molybdenum specimens. Similar phenomena have been observed in niobium.² Both photographs were made from samples of the molybdenum specimen that had been tested at 1400°C and a stress level of 7.5 kpsi. The two networks appear geometrical thus indicating that a uniform interaction of dislocations is taking place. Analyses of the networks will be made in an attempt to identify the dislocation interactions responsible for their formation.

FIGURE 27 - Dislocation loops in post-test molybdenum.

FIGURE 28 - Network of dislocations formed during creep test of molybdenum.

FIGURE 29 - Three dimensional dislocation network in post-test molybdenum.

Figure 30 shows the misorientation that exists between subgrains in tested material. There is a $0^{\circ} 10' 18.8''$ misorientation between subgrains 3 and 4, while the misorientation between subgrains 2 and 3 is only $0^{\circ} 1' 8.76''$.

FIGURE 30 - Misorientation of subgrains in tested molybdenum.

Future Work The substructure analysis of the remaining tensile specimens is in progress. Once it is completed the actual test data and substructure analyses will be correlated in an attempt to get a better understanding of the creep mechanisms. The creep tester was modified to run a set of compressive creep specimens at 1400°C and the same stress levels at which the tensile specimens had been tested. The results of the compressive tests will be compared against the tensile specimens to note any similarities or differences. Testing of irradiated molybdenum specimens should begin shortly after the control specimen tests are completed. After the irradiated specimens are tested, testing will resume on unirradiated specimens at different temperatures and stress levels. (R. E. Zielinski)

PROCESS STUDIES

Evaluation of Graphite Dies A study was initiated to evaluate the graphite-punch knife-edge configurations in an effort to minimize knife-edge breakage during the hot pressing of Multihundred Watt PPO (pressed plutonium oxide) spheres. Reduction of knife-edge breakage would produce a more uniform equatorial band around the spheres than is presently being produced. Initial work was done using ThO_2 powder.

The ThO_2 powder was a 53-246 μm fraction that had been sintered at 1100°C for 20 min. The dies were fabricated from POCO graphite grade AXF. All dies used had nominal diameters of 1.450 in. (The Multihundred Watt sphere diameter specification is 1.465 ± 0.015 in.) The dies were charged with 222.5 g of ThO_2 powder. The spheres were hot pressed at 1850°C using 8000 lb force except sphere STO-2 which required 9800 lb. The temperature was maintained 11 min after the ram movement indicated essentially no compaction was taking place. After removal from the die, the spheres were heated to 1100°C for 1/2 hr in air to remove graphite or carbides from the surface.

The first sphere, STO-1, was pressed with a die similar to those used for PPO sphere fabrication. These dies were designed to have a 0.010-0.012 in. gap between punch edges at closeout of the punches. The knife edges on the punches were 0.004 in. wide. The pressed sphere weighed 221.5 g having a polar diameter of 1.453 in. and an equatorial diameter of 1.467 in. The band around the equator of the sphere appeared similar to the bands observed on the PPO spheres. The punch edges were broken, embedding the graphite in the sphere. The graphite that did not fall out of the sphere during removal from the die was removed by heating to 1100°C in air, producing voids on both sides of the equatorial band. The equatorial band width was irregular varying between 0.050 and 0.190 in. Figures 31 and 32 are photographs of the punches and sphere.

The second sphere, STO-2, was pressed using a die designed to have a 0.240 in. gap between punch edges at closeout. The knife edges on the punches were 0.015 in. thick. A force of 9800 lb was required to close the die. The sphere adhered to the punches especially around the knife edges. The bottom part of the sphere broke off. The sticking problem, which was not observed for STO-1, was most probably a result of the higher compaction force and longer time needed to close the die. The increased knife-edge area of the punches used for STO-2 probably explains the difficulty experienced in obtaining die closeout.

A third sphere, STO-3, was pressed using a die designed to have a 0.162 in. gap between punch edges at closeout. The knife edges on the punches were about 0.008 in. thick. The pressed sphere weighed 221.8 g having a polar diameter of 1.452 in. and an equatorial diameter of 1.470 in. The band around the equator of the sphere appeared uniform and was about 0.175 in. wide. The knife edges of the punches were not broken.

FIGURE 31 - ThO_2 sphere STO-1 and graphite punches.

FIGURE 32 - ThO_2 sphere STO-1 showing equatorial band.

Figures 33 and 34 are photographs of the punches and sphere. A graphite die having a similar knife-edge configuration will be used to press a PPO sphere. (W. D. Pardieck and B. R. Kokenge)

FIGURE 33 - ThO_2 sphere STO-3 and graphite punches.

FIGURE 34 - ThO_2 sphere STO-3 showing equatorial band.

Production of Pellets for PRHU Capsule Fifteen 1-W heat sources are being made for the PRHU program by diamond core drilling of standard production PMC discs with an ultrasonic drill press. Developmental studies showed that there are three essential factors in obtaining acceptable pellets for loading into the PRHU capsule. These three factors are tool design, tool feed, and proper clearance of chips during drilling. An acceptable pellet is defined as one whose diameter is less than 0.191 in. and has a flat on each end.

The standard diamond core drill has an impregnated section on the tip which is a thin-walled cylinder. This type of drill works well with ceramic drilling operations where liquid coolants are forced through the center of the tool. Since argon gas is being used in this glovebox application, it was necessary to relieve the inside diameter of the drill 2° to assist in chip and powder clearance. When no inside relief was used on the core drill, it would not drill effectively after penetrating about 0.100 in. or about halfway through a standard PMC disc. Continued attempts at drilling would then result in the core binding and being broken out.

The second most important factor is tool feed. It was shown that the diamond drills improve in cutting efficiency with high speed. The maximum speed of the ultrasonic drill press is now being used at all times, but the actual rpm of the spindle is unknown. It was also found that a constant pressure of the tool against the drilling surface is imperative. This was accomplished by allowing the drill to feed under its own weight in a operator "hands-off" status until breakthrough is approached.

To obtain the pellets with a minimum of breakout on the bottom, it is necessary to remove the pressure from the drilling face in the last 0.030 in. before breakout. This was done by the operator using his hands on the control lever of the drill press to carefully restrain the drill's forward movement. A dial indicator is used to determine drill depth.

The third factor, clearing the residual PMC chips, is the most important factor in drilling efficiency. It was found that the level of ultrasonic vibration was unimportant for the power level range provided, but that the drills would not drill at all without ultrasonics. Therefore, the ultrasonics are operated at the lowest level since there was some indication that higher ultrasonic power levels increased the incidence of rough breakout.

Another significant factor is that the ultrasonics set up a pattern of vibration on the surface of the disc which causes the chips to vibrate back into the hole being drilled where they interfered severely with the efficiency of the drill. Essentially, excessive chips at the interface between the drill and the PMC disc can act like roller bearings and completely stop the progress of the drill. This problem was eliminated by directing a blast of argon against the tangential escape pattern of the chips as ejected by the drill. the results could be observed by watching

the dial indicator used to measure drilling depth. The feed rate would become very smooth when the argon blast was at the proper angle and direction. (G. N. Huffman and B. R. Kokenge)

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