Progress Report **MASTER**
DEVELOPMENT
OF
PLUTONIUM-BEARING FUEL MATERIALS
For Period April 1 through June 30, 1962
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Nuclear Materials and Equipment Corporation
Apollo, Pennsylvania

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PROJECT AND FACILITY ADMINISTRATION

Task 1.00
K. H. Puechl

Summary of Development Activities

During this reporting period, particular emphasis has been placed on the fabrication and characterization of samples scheduled for initial in-pile screening studies. Fabrication of all pellets for these irradiation tests had been completed during the last reporting period, and complete pre-irradiation evaluation is now under way. Sintering studies on PuO_2 and UO_2 - PuO_2 mixed oxide pellets have also been continued, and the results are being evaluated in the light of powder characterization data, especially oxygen to metal ratios. In preparation for direct precipitation of dense PuO_2 containing materials, prototype studies on the preparation of UO_2 by homogeneous (urea) precipitation from uranyl nitrate solutions have been continued. Also, continued effort has been placed on the production of high density spherical PuO_2 particles by both mechanically forming and sintering and plasma torch fusion. Analytical chemistry effort has been concentrated on the development of techniques to allow utilization of x-ray emission as a routine tool for analysis of plutonium and uranium. Reactor physics studies on the potential of plutonium as a fuel in near-thermal reactors have also been continued. Results indicate that dilution of UO_2 - PuO_2 fuel with inert ZrO_2 allows for increased core power density, hence reduction in capital costs without any significant increase in fuel cycle cost.

Pre-irradiation characterization of all samples scheduled for in-pile irradiation is now being performed. Photomicrographs indicate that the grain size of the mechanically mixed component powder samples is smaller than that of the samples produced from coprecipitated UO_2 - PuO_2 . Specified pellet densities were obtained for all mixtures ranging from UO_2 -0.5 w/o PuO_2 to UO_2 -20 w/o PuO_2 .

Sintering studies on PuO_2 pellets have shown that greater density is achieved by sintering in air than by sintering in a N_2 -6% H_2 atmosphere. Also, the results indicate that increased compaction pressure yields higher sintered densities when sintering in air, which was not the case when sintering in N_2 -6% H_2 .

Sintering studies on UO_2 - PuO_2 coprecipitated materials have shown that optimum sintering conditions are dependent upon the uranium to plutonium ratio. Further, it appears that sintering to high density is generally difficult when the oxygen to metal ratio is greater than 2.3 although the adverse sintering characteristic may not be due to this high ratio per se.

Re-reduction of material having a high oxygen to metal ratio was found to be satisfactory treatment for improving attainable density.

Studies on the densification of UO_2 produced by homogeneous (urea) precipitation from uranyl nitrate solutions have shown that over the temperature range (740-1580°C) considered, both densification and surface area reduction vary linearly with temperature. Microscopic examination further showed that the final particle size and shape is strongly dependent upon the corresponding ADU precursor characteristics.

Mechanically formed PuO_2 spheres were sintered in an N_2 -6% H_2 atmosphere at 1700°C for two hours and exhibited a particle density of 10.36 g/cc. More generally, over the sintering temperature range of 1450-1600°C it was found that sintering in air resulted in greater densification than sintering in N_2 -6% H_2 .

Plasma torch fused PuO_2 spherical particles have been produced and microscopically examined after sectioning and cathodic vacuum etching. This etching technique brings out the grain boundaries which are not discernable after chemical etching.

X-ray emission studies have been continued to develop this method into a routine procedure for the determination of uranium and plutonium. Using potassium pyrosulfate as a flux, a satisfactory pellet casting method has been developed which eliminates the previously used grinding and briquette forming operations. The accuracy of this method for the determination of plutonium content was found to be comparable to that obtainable with amperometric titration.

Additional chemistry studies have been initiated to determine the feasibility of removing plutonium from filtrate wastes by coprecipitation with calcium phosphate, to allow determination of oxalate concentration by an indirect CO_2 determination, and to evaluate proposed Cs-137, Sr-90, and Ar-95 separation techniques for the determination of burnup in irradiated specimens.

The apparatus for the measurement of thermal conductivity at elevated temperatures has been completely assembled, and final adjustments are being made prior to initial verification runs on UO_2 samples. A temperature of 1725°C has been obtained without difficulty.

Reactor physics studies on near-thermal converter reactor systems have been continued in order to allow evaluation of the merits of using ZrO_2 as a fuel diluent to permit operation at a higher power density. The results indicate that such dilution is possible without a significant increase in fuel cycle costs and has appreciable merit since operation is possible with relatively tight lattices thereby affording savings in capital costs. Since such dilution offers the reactor designer another degree of freedom without incurring a significant cost penalty, plutonium fuels with added diluents appear to be worth investigation from the materials standpoint.

General Plant and Project Aspects

During this reporting period, the decision was made to perform, at least the initial short-term in-pile screening tests, in the General Electric Testing Reactor (GETR). The preliminary design of the irradiation capsule assembly has therefore been submitted to the GETR for approval.

In support of the irradiation program, considerable effort is being placed on the outfitting of the alpha boxes for the hot cell and the hot metallography cell. Decontamination of this latter cell still presents a major obstacle.

Other equipment necessary to the development program that is being installed includes the swager and the high temperature dilatometer. Installation of the centerless grinder has been completed, and the glove box is undergoing final leak testing.

To eliminate division of responsibility, separate reporting of the progress under Task 11.00, "Preparation and Coating of Spherical Oxide Particles" has been discontinued; however, the effort along these lines is continuing and is now reported under Task 3.00, "Fabrication and Evaluation of Fuel Shapes".

PREPARATION AND CHARACTERIZATION OF FUEL MATERIALS

Task 2.00

C. S. Caldwell

O. Menis

Preparation and Characterization of Mixed Plutonium-Uranium Oxides

(A. Biancheria, G. Ehrlich, J. Goodman, M. D. Houston)

In continuation of the mixed oxide process development program, three lots of $\text{UO}_2\text{-PuO}_2$ ceramic-grade mixed oxides containing 0.5, 5, and 18% PuO_2 were prepared and characterized to provide information on process controllability during the transient and steady-state periods of the precipitation process. The mixed oxides were prepared by the furnace reduction of high surface area ammonium uranate-plutonium hydroxide which were coprecipitated continuously by the addition of concentrated ammonium-hydroxide to an acidified uranyl nitrate-plutonium nitrate solution. Processing data are listed in Table 2.1. The conditions used were standardized with respect to previous work to permit final property data intercomparison. These powder characterization data are presented in Table 2.2. The salient features of each preparation run are discussed below. Detailed sintering results are presented in Table 3.1 of the Section, "Fabrication and Evaluation of Fuel Shapes".

The $\text{UO}_2\text{-0.5 w/o PuO}_2$ powder prepared in Run 297Pu15 was found to have sub-standard sintering characteristics despite repeated hammer milling; however, re-reduction at 740°C for one hour lowered the oxygen to metal ratio from 2.39 to 2.06, and satisfactory pellets of 95-96% T.D. were then obtained after sintering at 1600°C . The high initial oxygen to metal ratio of this material was probably due to incomplete reduction during the furnace conversion step and to some reoxidation during hammermilling.

The $\text{UO}_2\text{-5 w/o PuO}_2$ precipitation run (297Pu16) was carried out to allow segregation from the steady-state material of the product produced during startup and shutdown. To allow assessment of the effects of furnace conversion temperature, the steady-state material was then divided into three equal portions and converted into oxide at 640, 740, and 840°C . Further, to allow comparison between startup and shutdown materials, these were both converted to oxide at 740°C . All samples were characterized in the "as-reduced" condition and also after hammermilling. In order to determine the inherent powder behavior with respect to oxidation stability during hammermilling, this operation was performed without a protective atmosphere or supplementary cooling. Referring to Table 2.2, it is seen that hammermilling increased the oxygen to metal ratio by 0.7 to 0.13 for all $\text{UO}_2\text{-5 w/o PuO}_2$ materials processed. The B.E.T. surface area decreased nearly linearly with increasing reduction temperature, and the oxygen to metal ratio followed a similar pattern; however, there was no correspondence between the oxygen pickup during hammermilling and the surface area or the initial oxygen to metal ratio. The oxide powder prepared at 840°C and having the lowest oxygen

Table 2.1

Summary of Preparation Data
for Coprecipitated UO₂-PuO₂ Materials

Run Identification	297Pu15	297Pu16					297Pu17			
Composition	UO ₂ -0.5 w/o PuO ₂	UO ₂ -5 w/o PuO ₂					UO ₂ -18 w/o PuO ₂			
Precipitation Conditions Method Temperature, °C	Continuous 50	Cont. 55	Cont. 55	Cont. 55	Cont. 55	Cont. 55	Cont. 55	Cont. 55	Cont. 55	Cont. 55
Feed Composition g Pu/liter g U/liter H ⁺ , molarity	0.51 103.0 1.0	5.04 96.9 1.0	5.04 96.9 1.0	5.04 96.9 1.0	5.04 96.9 1.0	5.04 96.9 1.0	17.9 80.4 1.0	17.9 80.4 1.0	17.9 80.4 1.0	17.9 80.4 1.0
Feed Flow Rate, liters/hr	1.20	1.22	1.22	1.22	1.22	1.22	1.21	1.21	1.21	1.21
Precipitant Composition NH ₄ OH, molarity	14.8	14.8	14.8	14.8	14.8	14.8	14.8	14.8	14.8	14.8
Precipitant Flow Rate liters/hr	0.35	0.22	0.22	0.22	0.22	0.22	0.26	0.26	0.26	0.26
Precipitation Average Holdup, minutes	29	31.2	31.2	31.2	31.2	31.2	30.4	30.4	30.4	30.4
Total No. Throughputs	8.5	3	4	4	4	3	3	2.15	2.15	3
Run Period	Integral	Startup	Steady State	Steady State	Steady State	Shut- down	Startup	Steady State	Steady State	Shut- down
Oxide Sample Identification	Pu15	TS80-740	SS80-640	SS80-740	SS80-840	RS80-740	TS80-740	SS80-740A	SS80-740B	RS80-740
Drying Temperature, °C	180	180	180	180	180	180	180	180	180	180
Gas Atmosphere	N ₂ -6% H ₂	N ₂ -6% H ₂	N ₂ -6% H ₂	N ₂ -6% H ₂	N ₂ -6% H ₂	N ₂ -6% H ₂	N ₂ -6% H ₂	N ₂ -6% H ₂	N ₂ -6% H ₂	N ₂ -6% H ₂
Gas Flow, SCFH	5	5	5	5	5	5		8.5	8.5	
Rate of Temperature Climb, °C/minutes	12	12	12	12	12	12	12	12	12	12
Conversion Temperature, °C	740	740	640	740	840	740	740	740	740	740
Time at Conversion Temp., minutes	80	80	80	80	80	80	80	80	80	80
Furnace Charge, kg	0.36	0.25	0.49	0.45	0.49	0.09	0.13	0.24	0.25	0.06

1
8
5

Table 2.2

Summary of Powder Characteristics
for Coprecipitated UO₂-PuO₂ Materials

Sample Number	Milling Treatment After Conversion	Oxygen/Letal Ratio	B.E.T. Surface Area M ² /g	Bulk Density g/cc	Tap Density g/cc	Air Perm. Particle Size Microns	Average ISA Particle Size for <200 mesh Fraction microns	w/o Greater Than 74 microns (200 mesh)
297Pu15	None	-	6.3	1.37	1.44	2.5	0.5	30
297Pu15	H ¹ milled	2.39	6.7	0.59	1.44	0.37	0.97	22
297Pu15	H ¹ milled	-	6.4	0.65	1.59	0.37	0.62	<2
297Pu15 Refired	None	2.06	4.6	0.88	1.48	0.54	7.0	22
Pu16TS80-740-III	None	2.12	5.4	1.13	1.73	0.50	1.7	30
Pu16TS80-740-III	H ¹ milled	2.23	6.5	0.78	2.11	0.41	1.2	4
Pu16SS80-640-III	None	2.45	9.5	1.32	1.65	2.86	0.46	38
Pu16SS80-640-III	H ¹ milled	2.52	9.9	0.70	1.89	0.31	2.3	6
Pu16SS80-740-III	None	2.36	7.1	1.25	1.45	0.88	0.3	61
Pu16SS80-740-III	H ¹ milled	2.49	7.1	0.68	1.65	0.32	2.1	<2
Pu16SS80-840-III	None	2.09	3.8	1.56	1.92	2.6	0.73	63
Pu16SS80-840-III	H ¹ milled	2.18	4.1	6.37	2.24	0.5	0.63	7
Pu16RS80-740-III	None	2.13	-	-	-	-	-	-
Pu16RS80-740-III	H ¹ milled	2.30	11.2	0.58	1.55	0.27	1.7	<2
Pu17TS80-740-III	None	-	8.0	1.37	1.90	1.62	0.52	27
Pu17SS80-740A-III	None	2.9	7.2	1.63	2.05	4.55	0.4	82
Pu17SS80-740A-III	H ¹ milled	2.21	7.7	0.93	2.37	0.48	1.0	9
Pu17SS80-740B-III	None	2.23	8.1	1.35	1.93	1.05	0.52	17
Pu17SS80-740B-III	H ¹ milled	2.29	9.0	1.74	2.11	0.42	0.5	15
Pu17RS80-740-III	None	-	13.9	0.93	1.44	0.45	1.3	17

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to metal ratio (2.18) of the series yielded the highest sintered density (96% T.D.) while the 640 and 740°C products with oxygen to metal ratios of 2.52 and 2.30 sintered to only 88-90% T.D. at 1600°C.

The UO_2 -17.8% PuO_2 preparation run (297Pu17) was carried out, as in the previous case, by segregating the startup and shutdown products from the steady-state material. All material was reduced at 740°C. The resulting oxide was also characterized in the "as-reduced" condition and after hammer-milling. In this case, hammermilling increased the oxygen to metal ratio by only 0.02-0.06, yielding final powder having an average ratio of 2.25. Despite the moderate oxygen to metal ratio, this material had substandard sintering characteristics; specifically, sintering at 1600°C yielded pellet densities of only 91-92% T.D. with significant cracking. This result was in contrast to the UO_2 -20% PuO_2 prepared in Run 297Pu13 which sintered to 95-97% T.D. at 1600°C⁽¹⁾.

Several generalized conclusions can be stated on the basis of mixed oxide process development work completed to date:

- 1) Mixed oxide powders containing 5% plutonium oxide exhibit improved sintering characteristics when the furnace reduction processing step is carried out at 840°C in a N_2 -6% H_2 gas atmosphere, rather than at lower temperatures.
- 2) Two interrelated key properties affecting sinterability, namely, surface area and oxygen to metal ratio, are controlled primarily by reduction furnace temperature and time and to a lesser extent by plutonium oxide content. Particle size, another key factor, is controlled by both reduction furnace temperature and precipitation conditions.
- 3) All powder properties are affected to some extent by the plutonium oxide content; therefore, optimum processing conditions must be established for each Pu/U composition investigated.

Preparation and Characterization of Granular Oxide Powders

(A. Biancheria, G. Ehrlich, J. Goodman, J. Miles)

Studies of prototype methods for the preparation of dense granular UO_2 - PuO_2 and PuO_2 materials have continued. During this reporting period, major emphasis was placed on examining the densification of material prepared by homogeneous (urea) precipitation from uranyl nitrate solutions.

(i) NUMEC P-90, Progress Report, "Development of Plutonium-Bearing Fuel Materials", page 29.

Precipitation of granular ADU from uranyl nitrate solutions was carried out by slow hydrolysis of urea in a stirred reactor vessel under reflux boiling conditions. The intermediate products were air dried at 25°C (100-200 hrs) or at 60°C (18 hrs), followed by conversion to UO_2 at 740 or 840°C under a flowing N_2 -6% H_2 atmosphere. A summary of the preparation conditions for five similar runs is presented in Table 2.3. Densification characteristics of the granular powders were determined by heating samples under typical presintering and sintering conditions at 1050, 1250, and 1600°C in a reducing atmosphere, followed by measurement of density and total surface area. The resulting data are presented in Table 2.4 together with properties of the as-reduced material. In general, the characterization data indicate that, over the temperature range examined, densification and reduction in surface area vary linearly with temperature. Mercury pycnometer measurements were also made on the sample (60-41-16) that exhibited the highest tap density (3.27 g/cc); the pycnometric results indicate a corresponding apparent particle density of 5.45 g/cc.

Microscopic examination of the final sintered particles showed an inherent resistance to particle agglomeration; only minor bridging was observed, and the material remained free-flowing after each successive furnace treatment. This inherent resistance to agglomeration was further verified by sintering results on a test pellet. Specifically, a cylindrical pellet, pressed at 8.2 tsi from Sample 60-17-32 powder having a surface area of 3 M²/g, sintered to only 70% of theoretical density after one hour at 1600°C.

Microscopic examination further showed that the final particle size and shape were strongly dependent upon the corresponding ADU precursor characteristics. It was concluded therefore that control of final particle size and shape must be accomplished by appropriate control of precursor characteristics. As expected, preliminary results indicate that more rapid stirring during the reflux and cooling period results in smaller ADU particles.

Analytical Chemistry

(O. Menis, E. Garcia, R. Jaroszeski
W. Judd, J. Limpert, P. Rey)

Development effort during this quarter was concentrated on the study of chemical analysis by x-ray emission and evaluation of procedures for the determination of burnup in irradiated samples. Further, a scavenging technique was developed for removal of plutonium from filtrate waste solutions, and preliminary studies were performed to allow determination of oxalate concentration in filtrates and plutonium oxalate materials.

X-Ray Emission Analysis

Using the experimental setup previously described,⁽ⁱ⁾ experiments were continued to evaluate the accuracy of this method for the determination

(i) NUMEC P-100, Progress Report, "Development of Plutonium-Bearing Fuel Materials", page 30.

Table 2.3

Summary of Preparation Conditions
for Uranyl Nitrate-Urea Homogeneous Precipitation Process Trials

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Experiment Number	60-17-32	60-20-9	60-32-1	60-41-16	72-14-2
<u>Reaction Conditions for Uranyl Nitrate-Urea Homogeneous Precipitation</u>					
Moles UO_2^{2+}	0.75	0.25	0.75	0.50	0.50
Initial Volume of UO_2^{2+} Solution, l	1.05	0.35	1.05	1.00	1.00
Moles Urea	2.25	0.75	2.25	1.00	1.00
Initial Volume of Urea Solution, l	0.42	0.15	0.45	0.40	0.40
Total Final Volume of Solution, l	1.47	0.50	1.50	1.40	1.40
Urea/ UO_2^{2+}	3	3	3	2	2
Reaction Temperature	Reflux	Reflux	Reflux	Reflux	Reflux
Reaction/Digestion Time, hrs	18	22	23	22	22
<u>Drying Conditions for ADU</u>					
Temperature, °C	60	25	25	25	25
Atmosphere	Air	Air	Air	Air	Air
Time, hrs	18	100	200	200	200
<u>ADU $\rightarrow$ UO_2 Conversion Conditions</u>					
Weight of Dry ADU Charged, g	250.0	86.6	200	140	115
Gas Composition	$\text{N}_2-6\% \text{H}_2$	$\text{N}_2-6\% \text{H}_2$	$\text{N}_2-6\% \text{H}_2$	$\text{N}_2-6\% \text{H}_2$	$\text{N}_2-6\% \text{H}_2$
Gas Flow Rate, SCFH	4	4	4	4	4
Rate of Temperature Rise, °C/min	4	6.5	6.8	6.8	6.8
Conversion Temperature, °C	740	840	840	840	840
Time at Temperature, min	85	60	60	60	60
Weight of Product, g	212.6	58.2	161.9	119.8	96.4
Weight UO_2 /Weight ADU	0.854	0.830	0.810	0.860	0.840

Table 2.4

Effect of Furnace Treatment on Properties of UO₂
Prepared by Homogeneous Urea Precipitation Process

Sample Identification	Surface Area M ² /g	Bulk Density g/cc	Tap Density g/cc
60-17-32 UO ₂ Powder from Reduction at 740°C	3.03	0.98	1.80
60-17-32 UO ₂ Powder Sintered at 1050°C for 2 hours	0.71	1.69	3.26
60-17-32 UO ₂ Powder Sintered at 1500°C for 4 hours	0.27	1.96	3.25
60-29-9 UO ₂ Powder from Reduction at 840°C	7.86	1.68	2.26
60-32-1 UO ₂ Powder from Reduction at 840°C	1.59	1.59	2.77
60-32-1 UO ₂ Powder Sintered at 1250°C for 2 hours	0.83	2.12	2.94
60-41-16 UO ₂ Powder from Reduction at 840°C	1.77	1.51	1.76
60-41-16 UO ₂ Powder Presintered at 1050°C for 2 hours	1.31	1.81	2.11
60-41-16 Presintered UO ₂ Sintered at 1580°C for 2 hours	0.22	2.48	3.27
72-14-2 UO ₂ Powder from Reduction at 840°C	4.08	1.19	1.49

of uranium and plutonium in mixtures with and without a thorium internal standard. Further, various flux preparations and pellet casting methods were evaluated relative to the pyrosulfate pressed briquette technique⁽¹⁾ in order to simplify sample preparation.

Using the pressed briquette technique, intensity ratio measurements were carried out on a number of standard samples containing a range of plutonium-uranium-thorium mixtures. Precision was evaluated by examining the relative intensities at the emission peaks as well as by comparing intensities with the matrix background at higher and lower wavelengths; for example, for uranium the $L_{\alpha 1}$ line was measured at 26.15° , and measurements relative to matrix background were also taken at 24.18 and 29.18° . The results are presented in Table 2.5 for the standard samples containing thorium and in Table 2.6 for samples without thorium. It is seen that the calibration curve is more nearly linear for the samples containing no thorium. This is illustrated further in the reproducibility data presented in Table 2.7. From these data, it is apparent that the use of thorium as an internal standard would be desirable primarily if the samples contained relatively high concentrations of unknown interfering elements.

Several fluxes were studied in order to simplify sample preparation. The results are presented in Table 2.8. In general, these techniques showed little promise. However, it was discovered that the pyrosulfate method could be used without grinding and briquette preparation by allowing the molten flux to cool slowly and to solidify into a uniform pellet. Increased precision was obtained with subsequent surface polishing as shown in Table 2.9. Calibration data on standard samples prepared by this method are presented in Table 2.10. Unfortunately, these data are not as precise as the corresponding data presented in Table 2.5 for the briquette method. However, based on the statistical variance ratio ("F" Test), the difference between methods is not significant. This cast pellet method is now being used for routine analysis. Comparison of results with those obtained by amperometric titration is shown in Table 2.11.

Studies Related to Burnup Analysis

With in-pile materials testing due to start in the near future, preliminary studies related to post-irradiation examination have been initiated. Radiochemical methods which appear most promising for estimation of burnup are the determination of Kr-89, Cs-137, Sr-90, and Zr-95. The Zr-95 may be particularly desirable for the examination of the short-irradiation rabbit tests. For the high burnup tests, Sr-90 results can be correlated to Cs-137 data, and hence determination of both radioisotopes will give greater assurance that the data are valid. To obtain estimates of possible errors inherent in the various separation schemes, studies are being initiated using "cold" synthetic samples containing only tracer quantities of radioactivity. Flame photometry will be primarily relied upon for evaluation of the strontium and cesium separations. Preliminary studies have been

Table 2.5

Calibration Data for Pressed Briquettes
Containing Uranium, Plutonium, and Thorium

Standard Number	U mg	Pu mg	Th mg	Pu/Th Intensity Ratio Wt. Ratio	U/Th Intensity Ratio Wt. Ratio	Pu/U Intensity Ratio Wt. Ratio	Pu/LB* Intensity Ratio mg Pu	Pu/HB* Intensity Ratio mg Pu	U/LB Intensity Ratio mg V	U/HB Intensity Ratio mg V
1	30.0	0	40.0	0	1.20	0	0	0	0.138	0.083
2	29.0	1.0	40.0	1.20	1.18	1.03	0.139	0.084	0.137	0.082
3	28.0	2.0	40.0	1.18	1.19	1.00	0.137	0.082	0.137	0.082
4	27.0	3.0	40.0	1.28	1.22	1.05	0.145	0.088	0.138	0.083
5	26.0	4.0	40.0	1.26	1.16	1.08	0.146	0.087	0.135	0.081
6	25.0	5.0	40.0	1.28	1.18	1.08	0.147	0.089	0.136	0.082
7	23.0	7.0	40.0	1.33	1.21	1.10	0.149	0.089	0.136	0.081
8	20.0	10.0	40.0	1.34	1.21	1.10	0.151	0.091	0.137	0.083
Average				1.27	1.19	1.06	0.145	0.087	0.137	0.082
Standard Deviation				± 0.06	± 0.02	± 0.04	± 0.005	± 0.003	± 0.001	± 0.0008
Coefficient of Variation				$\pm 4.7\%$	$\pm 1.7\%$	$\pm 3.5\%$	$\pm 3.4\%$	$\pm 3.4\%$	$\pm 0.7\%$	$\pm 1.0\%$

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* LB designates matrix background at low side of Pu or U emission peak.
 ** HB designates matrix background at high side of Pu or U emission peak.

Table 2.6

Calibration Data
Briquettes Pressed
Containing Uranium and Plutonium without Thorium

Standard Number	U mg	Pu mg	Pu/U Intensity Ratio It. Ratio	Pu/LB* Intensity Ratio mg Pu	Pu/HB** Intensity Ratio mg Pu	U/LB Intensity Ratio mg U	U/HB Intensity Ratio mg U
1	10.0	11.3	1.131	0.155	0.096	0.132	0.033
2	10.0	22.5	1.123	0.152	0.094	0.136	0.034
3	10.0	33.8	1.129	0.150	0.094	0.133	0.033
4	20.0	11.3	1.122	0.152	0.095	0.137	0.034
5	30.0	11.3	1.123	0.152	0.095	0.136	0.034
Average			1.123	0.152	0.095	0.135	0.034
Standard Deviation			± 0.004	± 0.002	± 0.0003	± 0.002	± 0.0007
Coefficient of Variation			$\pm 0.4\%$	$\pm 1.3\%$	$\pm 0.8\%$	$\pm 1.5\%$	$\pm 0.3\%$

Table 2.7

Reproducibility of Duplicate Determinations of Plutonium and Uranium
(Standard Relative Error Between Duplicates at a 95% Confidence Level)

Sample Composition	Pu/Th	U/Th	Pu/U	Pu/LB*	Pu/HB**	U/LB	U/HB
Pu, U, Th	$\pm 2.5\%$	$\pm 2.1\%$	$\pm 2.6\%$	$\pm 2.7\%$	$\pm 2.3\%$	$\pm 0.3\%$	$\pm 1.3\%$
Pu, U	-	-	$\pm 2.1\%$	$\pm 0.2\%$	$\pm 0.4\%$	$\pm 1.0\%$	$\pm 0.3\%$

* LB designates matrix background at low side of Pu or U emission peak.

** HB designates matrix background at high side of Pu or U emission peak.

Table 2.8

Properties of Several Fluxes Used for X-Ray Emission Studies

Mixture	Composition	Approximate Melting Point °C	Casting Properties	Observations
1	Fused Borax (Borax Glass)	250	Good	Viscous mass was difficult to stir. Excellent borax glass beads obtained but showed inhomogeneity and segregation
2	Borax + 10% Boric Acid	950	Good	No advantages over 1.
3	Borax + 10% $K_2S_2O_7$	950	Good	No advantages over 1.
4	Potassium Dichromate	400	None	Crystallized when cooled.
5	$K_2Cr_2O_7 + 5\% K_2S_2O_7$	400	Pellets Crack	
6	$K_2Cr_2O_7 + 10\% K_2S_2O_7$	400	Good	Poor reproducibility due to incomplete dissolution. Dark color masked the completeness of fusion
7	$K_2Cr_2O_7 + 50\% K_2S_2O_7$	No definite melting point	-	Flux decomposed
8	Ammonium Bifluoride	65	-	Attacked UO_2 only at temperatures above $100^\circ C$ with sublimation sufficient to preclude matrix reproducibility
9	$NH_4HF + 10\% K_2S_2O_7$	< 400	-	High fusion temperature affected teflon ware
10	Lithium Borate	840	-	Attacked platinum ware.

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Table 2.9
Effect of Surface Irregularities
on the Precision of Measurements Using Cast Pellets

Ratio	As-Cast Pellets				Polished Pellets			
	Number of Determinations	Average	Standard Deviation	Coefficient of Variation $\%$	Number of Determinations	Average	Standard Deviation	Coefficient of Variation $\%$
Pu/U	7	1.29	± 0.04	± 3.1	5	1.27	± 0.01	± 0.87
Pu/High Background	7	1.07	± 0.06	± 5.4	5	1.03	± 0.02	± 1.6
U/High Background	7	1.03	± 0.04	± 3.8	5	0.82	± 0.02	± 2.0

Table 2.10

Calibration Data for Cast Pellets
Containing Uranium, Plutonium, and Thorium

Standard Number	U mg	Pu mg	Th mg	Pu/Th Intensity Ratio Wt. Ratio	U/Th Intensity Ratio Wt. Ratio	Pu/U Intensity Ratio Wt. Ratio	Pu/LB* Intensity Ratio Wt. Ratio	Pu/HB* Intensity Ratio Wt. Ratio	U/LB Intensity Ratio Wt. Ratio	U/HB Intensity Ratio Wt. Ratio
1	30.0	0	40.0	0	1.25	0	0	0	0.143	0.085
2	29.0	1.0	40.0	1.20	1.24	0.94	0.141	0.086	0.147	0.090
3	28.0	2.0	40.0	1.20	1.20	1.03	0.148	0.090	0.145	0.089
4	27.0	3.0	40.0	1.28	1.18	1.09	0.146	0.089	0.148	0.089
5	26.0	4.0	40.0	1.37	1.21	1.14	0.172	0.103	0.142	0.088
6	25.0	5.0	40.0	1.29	1.21	1.08	0.154	0.093	0.142	0.086
7	23.0	7.0	40.0	1.30	1.17	1.10	0.157	0.095	0.141	0.086
8	20.0	10.0	40.0	1.38	1.16	1.08	0.154	0.093	0.142	0.085
Average				1.29	1.20	1.07	0.153	0.093	0.144	0.087
Standard Deviation				± 0.07	± 0.03	± 0.06	± 0.010	± 0.005	± 0.003	± 0.002
Coefficient of Variation				$\pm 5.4\%$	$\pm 2.5\%$	$\pm 6.0\%$	$\pm 6.5\%$	$\pm 5.4\%$	$\pm 1.8\%$	$\pm 2.3\%$

* LB designates matrix background at low side of Pu or U emission peak.

** HB designates matrix background at high side of Pu or U emission peak.

Table 2.11

Comparison of Cast-Pellet X-Ray Emission Results
with Amperometric Titration

	X-Ray Emission		Amperometric Titration
	Th Internal Standard	No Internal Standard	
Number of Determinations	4	8	8
Determined Pu Content, %	16.02	15.67	15.82
Standard Deviation	0.13	0.13	0.12

carried out to determine the effects of uranium in various acid media on the flame emission of strontium. Perchloric acid seems to increase sensitivity and large variations in nitric acid concentration do not appear to affect the results.

Removal of Plutonium from Filtrate Waste

In the process of coprecipitating uranium and plutonium from ammonical solutions, a large amount of filtrate is produced. Generally, this filtrate contains activity of 2000-5000 counts/min/ml and cannot be readily disposed of. Attempts have therefore been made to scavenge the alpha emitters from the solution by coprecipitation with calcium phosphate. Specifically, the procedure tested to date consists of adding one gram of trisodium phosphate to each liter of filtrate and then adding, while continuously stirring, a solution containing 200 mg of calcium. The solution is then filtered after 15 minutes of stirring. In five tests using laboratory scale equipment, the activity level was reduced to less than 15 counts/min/ml. The procedure therefore appears to offer sufficient promise to warrant further experimentation with gallon quantities.

Determination of Oxalate Concentration

Because of the presence of plutonium, a direct oxidation method cannot be used for the determination of the oxalate concentration in filtrate and in plutonium oxalate samples. An attempt is therefore being made to determine oxalate content indirectly by determining the amount of CO_2 liberated during refluxing in a sulfuric acid, potassium permanganate solution. Initial trials with sodium oxalate yielded precision and accuracy within 1%.

FABRICATION AND EVALUATION OF FUEL SHAPES

Task 3.00

E. K. Halteman

L. J. Jones

Mixed Oxide Sintering Studies
(M. D. Houston)

Investigation of the sintering characteristics of UO_2 - PuO_2 compositions in the range 0.5 to 20 w/o PuO_2 by the cold pressing and sintering method has been continued. Powder preparation data were presented in Table 2.1 and powder characterization data in Table 2.2. As shown in Table 3.1, sintered densities in the range of 90-96% of theoretical density were observed after sintering at 1600°C except for compositions Pu 15-HM, Pu 16SS80-640, and Pu 17SS80-740A. Failure of the first two compositions to sinter was probably due to the high oxygen to metal ratio. After the oxygen to metal ratio had been reduced by further reduction firing, the sinterability of Pu 15-HM improved, as evidenced by the resulting density of 94.5% theoretical. The reason for the fracturing of Pu 17SS80-740A compacts has not been fully established. Since the associated weight loss for this material was in a satisfactory range (1.25%), gas evolution should not have been the cause of fracturing.

 PuO_2 Sintering Studies
(M. D. Houston, J. Miles)

The sintering characteristics of PuO_2 have been further examined by utilizing a number of different oxide preparations in combination with various compaction and firing parameters.

As reported previously,⁽ⁱ⁾ the final sintered density of PuO_2 after air-firing was higher than that for equivalent samples fired in N_2 -6% H_2 . This study was extended to observe the effects of compaction pressure when coupled with an oxidizing sintering atmosphere. The data in Table 3.2 indicate an increase in density with an increase in compaction pressure over the range employed (7-45 tsi). This is in direct contrast to the results obtained previously with pellets pressed at different pressures and fired in a nitrogen-hydrogen atmosphere; in these earlier studies it was observed that compaction pressure had little, if any, effect upon the final fired density.⁽ⁱⁱ⁾ The sintered density of 10.7 g/cc was the highest observed for powder lot

(i) NUMEC P-100, Progress Report, "Development of Plutonium-Bearing Fuel Materials", page 40.

(ii) NUMEC P-100, Progress Report, "Development of Plutonium-Bearing Fuel Materials", pages 37 and 39.

Table 3.1

Sintering Data for Coprecipitated Mixed Oxides UO_2 - PuO_2

Composition Designation	PuO_2 Content w/o	Precompaction Treatment*	Green Density g/cc	Sintering Temperature $^{\circ}C$	Sintering Time hrs	Firing Weight Loss %	Sintered Density g/cc	Theoretical Density %
Pu15-HM	0.5	S	5.10	1600	1	2.50	Fractured	
Pu15-HM-Re	0.5	N	5.42	1400	1	1.10	10.39	94.5
Pu15-HM-Re	0.5	N	5.42	1500	1	1.15	10.50	95.5
Pu15-HM-Re	0.5	S	5.68	1600	1	1.20	10.50	95.5
Pu15-HM-Re	0.5	S	5.60	1650	1	1.20	10.55	96.0
Pu14-A-IT	5.0	S	5.58	1600	1	1.20	10.56	96.0
Pu14-B-HM	5.0	S	5.55	1600	1	1.10	10.63	96.7
Pu16SS80-640-HM	5.0	N	4.48	1600	1	2.84	9.66	87.8
Pu16SS80-640-HM	5.0	N	4.49	1600	16	2.95	9.68	88.0
Pu16SS80-740-IT	5.0	N	4.92	1600	1	2.51	9.85	89.5
Pu16SS80-740-HM	5.0	N	4.92	1600	16	2.30	10.26	93.3
Pu16TS80-740-HM	5.0	N	5.35	1600	1	1.40	10.18	92.5
Pu16TS80-740-IT	5.0	N	5.37	1600	16	1.50	10.33	93.9
Pu16RS80-740-IT	5.0	N	5.40	1600	1	1.60	9.96	90.5
Pu16RS80-740-HM	5.0	N	5.39	1600	16	1.90	10.03	91.1
Pu16SS80-840-HM	5.0	N	5.52	1600	1	1.00	10.56	96.0
Pu17SS80-740A-IT	17.8	N	5.35	1600	1	1.20	Fractured	
Pu17SS80-740A-HM	17.8	S	5.51	1600	1	1.25	10.07	91.5
Pu17SS80-740B-IT	17.8	N	5.41	1600	1	1.50	10.15	92.4
Pu17SS80-740B-HM	17.8	S	5.52	1600	1	1.20	9.99	90.8

* N - Non-slugged

S - Slugged and granulated

Table 3.2

PuO₂ Sintering Trials
(Sintering Time - 1 Hour)

Material Designation	Calcining Temperature °C	Compaction Pressure TSI	Green Density g/cc	Sintering Atmosphere	Sintering Temperature °C	Firing Weight Loss %	Compact Shrinkage %	Sintered Density g/cc	Theoretical Density %
Pu-9-3	760	7.2	6.53	Air	1600	1.0	13.4	10.03	87.5
	760	18.1	7.05	Air	1600	0.6	12.2	10.40	90.2
	760	28.9	7.38	Air	1600	0.8	11.0	10.73	93.8
	760	38.0	7.51	Air	1600	0.9	11.0	10.60	92.6
	760	45.2	7.70	Air	1600	0.9	10.9	10.79	94.2
Pu-4-1	350	38.0	5.77	Air	1550	6.0	19.0	10.37	90.6
	350	38.0	5.83	N ₂ -H ₂	1600	4.6	20.1	10.46	91.4
	760	38.0	7.13	N ₂ -H ₂	1600	0.7	13.0	10.76	94.0
Pu-4-2	420	38.0	6.00	Air	1550	4.5	18.4	10.63	92.9
	420	38.0	6.00	N ₂ -H ₂	1600	3.2	18.1	10.66	93.1
	760	38.0	7.10	N ₂ -H ₂	1600	1.0	13.0	10.62	92.9
Pu-4-4	560	38.0	6.36	Air	1550	3.1	17.2	10.94	95.6
	560	38.0	5.92	N ₂ -H ₂	1600	2.2	20.1	10.34	90.4
Pu-5-1	350	38.0	5.30	Air	1550	7.1	20.9	10.14	88.6
	350	38.0	4.71	N ₂ -H ₂	1600	8.8	28.5	9.14	79.4
	420	38.0	5.78	Air	1550	4.5	19.8	10.90	95.2
Pu-5-2	420	38.0	5.76	Air	1550	4.5	19.8	10.87	95.0
	420	38.0	5.85	N ₂ -H ₂	1600	3.6	17.7	10.35	90.5
	760	38.0	7.07	N ₂ -H ₂	1600	0.8	13.0	10.64	93.0
	760	11.2	6.86	Air	1600	0.7	12.5	10.30	90.0
Pu-Nit	740	11.2	6.80	N ₂ -H ₂	1600	0.7	10.5	9.32	81.4

Pu-9-3 under any conditions and was significantly higher than the highest density (10.33 g/cc) observed after firing in a reducing atmosphere.⁽¹⁾

Previously prepared batches 297-Pu-4 and 297-Pu-5 prepared by the oxalate route were utilized to study the effect of sintering atmosphere and to explain the variations observed in the final microstructures. As expected, weight losses were higher with material calcined at the lower temperatures, indicating incomplete oxalate decomposition during the low temperature calcinations. In general, an air sintering atmosphere resulted in higher densities and higher firing weight losses. Where lower densities occurred, the weight loss during firing was also excessive. The lower weight losses resulting from sintering in a nitrogen-hydrogen atmosphere were unexpected since, in addition to the losses associated with oxalate decomposition, a further loss of oxygen due to the reduction of PuO_2 to Pu_2O_3 was anticipated. The results indicate that a considerable portion of the decomposition products were retained in the structure, presumably as a carbide or an oxycarbide. The microstructures as revealed by cathodic vacuum etching of the low and high temperature calcines of Pu-4-1, Pu-4-2, and Pu-5-2 show that the texture changes from a fine grained two-phase structure to a massive two-phase structure as shown in Figures 3.1 and 3.2.

In addition to using the standard materials, a sintering study was also performed with a small amount of PuO_2 prepared by evaporating plutonium nitrate solution to dryness in air at 200°C and then calcining at 750°C for one hour. The air atmosphere sintering results for compacts of this material (Pu-nit) are similar to these obtained with Pu-9-3; however, a lower density was obtained when sintered in a nitrogen-hydrogen atmosphere. Metallographic examination of these compacts is currently in process.

Rabbit Test Fuel Fabrication (M. D. Houston)

The fabrication of samples for the short-term screening irradiation program has been completed. All pellets are still being characterized prior to their encapsulation into fuel elements. Compaction and sintering data for the various oxide compositions are given in Table 3.3. X-ray diffraction data show that a single-phase fluorite structure has been formed in all compositions. As shown in Figure 3.3, microstructural differences in the fired pellets were quite noticeable between coprecipitated and mechanically mixed compositions. The grain size of the mechanically mixed samples is smaller than that of the coprecipitated samples, and the grain size tends to decrease with an increase in plutonium concentration. Changes in the microstructure

(i) NUMEC P-100, Progress Report, "Development of Plutonium-Bearing Fuel Materials", page 39.

Table 3.3

Compaction and Sintering Data for Oxide Pellets
(Sintered in N_2 -6% H_2 Atmosphere)

Composition	Preparation	Compaction Pressure TSI	Green Density g/cc	Sintering Temperature $^{\circ}C$	Sintering Time hr	Sintered Density g/cc	Theoretical Density %
UO_2 -9.08% U_{235}	Coprecipitated (Slugged)	15.5	5.69	1600	1	10.4	93-95
UO_2 -21.3% U_{235}	Coprecipitated (Slugged)	12.6	5.45	1400	1	10.3-10.4	93-95
UO_2 -7.56% U_{235} -0.5% PuO_2	Mechanical Mix	11.7	5.80	1600	16	10.3-10.4	93-95
UO_2 -7.56% U_{235} -0.5% PuO_2	Coprecipitated (Slugged)	14.4	5.50	1600	1	10.4	93-95
UO_2 -5% PuO_2	Mechanical Mix	14.4	5.84	1600	16	10.2-10.3	93-95
UO_2 -5% PuO_2	Coprecipitated	11.7	5.55	1600	1	10.4-10.6	95-97
UO_2 -5% PuO_2	Coprecipitated	11.7	5.55	1400	1	10.2-10.4	93-95
UO_2 -20% PuO_2	Mechanical Mix	16.2	6.04	1600	16	10.2-10.4	93-95
UO_2 -20% PuO_2	Coprecipitated (Slugged)	12.0	5.12	1600	1	10.2-10.3	93-95
UO_2 -20% PuO_2	Coprecipitated (Slugged)	11.7	5.10	1300	2	9.7-9.9	88-91
UO_2 -20% PuO_2	Coprecipitated (Slugged)	11.7	5.10	1300	1	8.9-9.3	82-85

Photomicrograph of PuO_2 - 297Pu-4-1
Calcination Temperature - 350°C
Compaction Pressure - 41.4 TSI
Sintered in N_2 -6% H_2 at 1600°C for 1 hour
Neg. 111 400X

Figure 3.1

Photomicrograph of PuO_2 - 297Pu-4-1
Calcination Temperature - 760°C
Compaction Pressure - 41.4 TSI
Sintered in N_2 -6% H_2 at 1600°C for 1 hour
Neg. 106 400X

Figure 3.2

UO₂-5% PuO₂
Coprecipitated

Neg. 98A

400X

UO₂-20% PuO₂
Coprecipitated

Neg. 99A

400X

UO₂-5% PuO₂
Mechanically Mixed

Neg. 96A

400X

UO₂-20% PuO₂
Mechanically Mixed

Neg. 97A

400X

Photomicrographs of Mixed Oxide Pellets
Compaction Pressure - 9 TSI
Sintered in N₂-6% H₂ at 1600°C for 16 hours

Figure 3.3

of PuO_2 samples fabricated from coprecipitated material (oxalate route) sintered for long firing times (up to 72 hours) at 1600°C were insignificant compared with the changes noted during relatively short-term sintering (4 hours). Thus, grain growth after achievement of maximum density appears to be insignificant.

Preparation of Spherical PuO_2 Particles
from Ceramic Grade Powders
 (A. Biancheria, J. Miles)

Investigation of dry-agglomeration pretreatment methods for forming spherical PuO_2 particles for dispersion-type fuels was continued during this period. In addition, densification during sintering was examined as a function of furnace temperature and gas atmosphere.

In order to maintain process flexibility, the agglomeration methods were developed using two different types of PuO_2 powder as starting materials. Both types of powder were prepared by 760°C calcination of plutonium (IV) oxalate; however, one lot (297 Pu-11A/B) was derived from plutonium oxalate precipitated continuously from dilute (~ 20 g Pu/l) plutonium nitrate solution while the other material (297 Pu-9-Lot 4) was precipitated continuously from a concentrated (100 g Pu/l) plutonium nitrate solution. The surface areas of both powder types were nearly equivalent; however, pressing characteristics and dispersion properties were dissimilar. Characteristics of these powders were presented in a previous report.⁽ⁱ⁾

Previous results had shown that the spheroidizing characteristics of PuO_2 powder were markedly improved by wet ball milling. Therefore, the study of pretreatment methods was extended to include ultrasonic attrition as a possible substitute. The PuO_2 powders were treated ultrasonically in a water slurry for two hours at a frequency of 20 kc. Preliminary results indicate a 25 per cent improvement in spheroidization yield for Sample 297Pu-9-Lot 4 over that attained with the normal 4-hour wet ballmilling treatment. However, ultrasonic agitation was relatively less effective in the case of sample 297Pu-11A/B. This result is in agreement with earlier findings when the effectiveness of ultrasonic dispersion was studied for the preparation of PuO_2 samples for particle size distribution measurements.⁽ⁱⁱ⁾ The conclusion is drawn that ultrasonic attrition methods for powder pretreatment prior to

(i) NUMEC P-90, Progress Report, "Development of Plutonium-Bearing Fuel Materials", page 7.

(ii) NUMEC P-90, Progress Report, "Development of Plutonium-Bearing Fuel Materials", page 10.

spheroidization may be attractive for selected materials; however, further trials involving higher power density, varying treatment periods, and varying frequencies are required before the results can be generalized.

Spheroidized PuO_2 particles (-100 +200 mesh) obtained by roll-agglomeration after wet ballmilling or ultrasonic pretreatment were of uniform shape and had green densities in the range of 3.5 to 3.8 g/cc. Spherical particles from sample 297Pu-11A/B were sintered for two hours in air at 1450 and 1600°C and also for two hours in a N_2 -6% H_2 reducing atmosphere at 1450, 1600, and 1700°C. Pycnometric density data and cross-section photomicrographs of the sintered spheres are shown in Figures 3.4, 3.5, and 3.6. A progressive increase in sintered density from 9.79 g/cc at 1450°C to 10.36 g/cc at 1700°C was observed during reducing-atmosphere sintering. Preliminary results in oxidizing-atmosphere sintering indicate a slight enhancement in sintered density at 1450°C, but the lack of further densification in air at 1600°C cannot be explained at present. Crushing strength measurements of the high-density spherical particles are presently being evaluated.

Production of Spherical Particles by Plasma Torch (R. M. Horgos)

During this period much effort was placed on associated metallographic examination in order to allow more detailed evaluation of the plasma torch product. In addition, efforts on torch operation were directed towards increasing the particle residence time within the extremely high temperature plasma region.

Chemical etching of polished particle sections was found to be unsuccessful in bringing out the grain structure. Subsequently, cathodic vacuum etching was attempted since earlier results⁽¹⁾ on PuO_2 compacts had been successful. However, this etching technique could not be directly applied to spherical particles since the particles are so small that the bombarding ions tend to destroy the standard mounting material. Both aluminum and graphite mounts were therefore tried with limited success. Sphere cross sections using graphite mounts are shown in Figure 3.7. Further refinements of procedures are now being developed.

Preliminary torch runs with UO_2 - PuO_2 and UO_2 - ZrO_2 mixtures have given indication that such mixtures are more difficult to fuse than are the individual components. Attempts are therefore being made to increase particle residence time within the high temperature plasma region. Modified nozzle designs are being tried in conjunction with various gas flow conditions.

(1) NUMEC P-90, Progress Report, "Development of Plutonium-Bearing Fuel Materials", page 59.

Sintered in Air at 1450°C for 2 hours
Density 9.92 g/cc

Neg. 90

200X

Sintered in $\text{N}_2\text{-}6\% \text{H}_2$ at 1450°C for 2 hours
Density 9.79 g/cc

Neg. 92

200X

Typical Cross Sections of PuO_2 Spheres
(-100 + 120 mesh)
(Lot 297Pu-11A/B)

Figure 3.4

Sintered in Air at 1600°C for 2 hours
Density 9.93 g/cc

Neg. 91

200X

Sintered in N₂-6% H₂ at 1600°C for 2 hours
Density 10.22 g/cc

Neg. 93

200X

Typical Cross Sections of PuO₂ Spheres
(-100 +120 mesh)
(Lot 297Pu-11A/B)

Figure 3.5

Sintered in N_2 -6% H_2 at $1700^\circ C$ for 2 hours
Density 10.36 g/cc

Neg. 113A

200X

Typical Cross Section of PuO_2 Spheres
(-100 +120 mesh)
(Lot 297Pu-11A/B)

Figure 3.6

Neg. 81A

600X

Neg. 81B

600X

Cross-Sections of PuO_2 Spheres
Graphite Mount - Cathodic Vacuum Etched

Figure 3.7

Physical Property Measurements
(E. K. Halteman)

The testing of the various systems involved in the apparatus for the measurement of the thermal conductivity of poor thermal conductors at high temperatures is nearing completion. The RF power supply for inductively heating the tantalum susceptor which surrounds the sample has been matched to the load. This was accomplished by removing the step-down transformer originally supplied in the tank circuit and inserting a properly matched work coil directly in the tank circuit. By using a test susceptor without any radiation shields, a brightness temperature of 1600°C was obtained. This corresponds to a true temperature of 1725°C, using an assumed emissivity of 0.36. A complete susceptor assembly has been successfully welded together and installed, although modifications had to be made in the method for holding the radiation shield in order to avoid the completion of loop circuits through the support legs of the susceptor. The magnetically focused electron gun has been installed and partially tested. The magnetic focusing lens and magnetic deflection systems have also been installed. In addition, a standard lamp with associated control circuitry has been installed in the glove box to allow calibration of the optical pyrometer and the measurement of the optical absorption of the various prisms and viewing windows. Test samples of UO_2 for testing prior to committing the equipment to plutonium are currently being fabricated.

A high temperature dilatometer has been designed and fabricated. The test sample will be heated therein by radiation from a resistively heated tantalum cylinder. A tungsten support will hold the sample, and a tungsten rod will be used to transmit the sample dilation to a LVDT transducer. The vacuum hardware is on hand, and the assembly of the apparatus into a glove box will be initiated during the next quarter.

FUEL ELEMENT FABRICATION AND EVALUATION

Task 4.00
L. J. Jones

Box and Equipment Installation
(M. Zambarnard)

Installation of the centerless grinder box and associated equipment has been completed, and the box is currently undergoing leak testing. A Sheffield air gauge has been installed with this equipment to measure ground pellets and to automatically correct for belt wear. Included with the recirculating coolant system is a wash-down system to minimize the build-up of fissionable material at any location within the box. As an added precaution, complete and rigid material accountability will be maintained within the box.

A swager box feed mechanism has been built and is currently undergoing initial testing. The swager proper has been installed in a glove, and final outfitting of the system is under way.

RADIATION TESTING AND EVALUATION

Task 5.00
L. J. Jones

Rabbit Tests
(L. J. Jones, R. M. Horgos)

Both the Materials Testing Reactor (MTR) and the General Electric Testing Reactor (GETR) were visited to evaluate their irradiation facilities for compatibility with the overall proposed testing program. As a result of these visits, the decision was made to negotiate further with the GETR for reactor space for the proposed short-term irradiations tests. It is expected that the Trail Cable Facility in the GETR will be used. A special flow director and capsule holder for insertion in this Facility will be designed and constructed. This flow director will be connected to an out-of-pile water supply by a flexible hose, which will also serve as the "trail cable". The entire assembly containing the capsule can be removed from the reactor through a 45° angle port while the reactor is at power and transferred to the GETR canal. The capsule can then be removed from the flow director and a new capsule inserted in its place. By following the reverse procedure, a new capsule can be placed into the irradiation facility. Thus, all capsules for this short-term irradiations program can be irradiated within a relatively short time period since insertion and discharge of capsules does not depend upon reactor shutdown schedules. A preliminary design of the capsule assembly has been completed (Figure 5.1) and forwarded to the GETR for approval of the general design.

The unperturbed neutron flux available in the Trail Cable Facility is 7.5×10^{13} nv, which will be perturbed by the stainless steel flow director to a flux level of approximately 7.1×10^{13} nv at the capsule surface. In order to perturb the neutron flux to the desired maximum of 3.0×10^{13} nv for certain specimens, the capsule will be constructed of a shrunk-fit boron-stainless steel liner within an aluminum outer capsule. The wall thickness of this liner will be varied to yield fluxes in the range of 3.0×10^{13} to 7.0×10^{13} nv at the specimen surfaces. Detailed drawings of the irradiation capsule will be started as soon as preliminary approval of the capsule design is obtained from GETR.

Hot Laboratory Equipment Fabrication
(R. M. Horgos)

Decontamination of the interior of the steel cell for the metallographic facility has been continued with little success. Sources of high beta-gamma activity have apparently been firmly deposited in the pores of the

ITEM	DESCRIPTION MATERIAL REF DRAWINGS	GROUP No			
		1	2	3	4
1	ITEM 15 1 11 1 31R				1
2	ITEM 16 1 11 1 31R				1
3	ITEM 17 1 11 1 31R				1
4	ITEM 18 1 11 1 31R				1
5	ITEM 19 1 11 1 31R				1
6	ITEM 20 1 11 1 31R				1
7	ITEM 21 1 11 1 31R				1
8	ITEM 22 1 11 1 31R				1
9	ITEM 23 1 11 1 31R				1
10	ITEM 24 1 11 1 31R				1
11	ITEM 25 1 11 1 31R				1
12	ITEM 26 1 11 1 31R				1
13	ITEM 27 1 11 1 31R				1
14	ITEM 28 1 11 1 31R				1
15	ITEM 29 1 11 1 31R				1
16	ITEM 30 1 11 1 31R				1
17	ITEM 31 1 11 1 31R				1
18	ITEM 32 1 11 1 31R				1
19	ITEM 33 1 11 1 31R				1
20	ITEM 34 1 11 1 31R				1
21	ITEM 35 1 11 1 31R				1
22	ITEM 36 1 11 1 31R				1
23	ITEM 37 1 11 1 31R				1
24	ITEM 38 1 11 1 31R				1
25	ITEM 39 1 11 1 31R				1
26	ITEM 40 1 11 1 31R				1
27	ITEM 41 1 11 1 31R				1
28	ITEM 42 1 11 1 31R				1
29	ITEM 43 1 11 1 31R				1
30	ITEM 44 1 11 1 31R				1
31	ITEM 45 1 11 1 31R				1
32	ITEM 46 1 11 1 31R				1
33	ITEM 47 1 11 1 31R				1
34	ITEM 48 1 11 1 31R				1
35	ITEM 49 1 11 1 31R				1
36	ITEM 50 1 11 1 31R				1
37	ITEM 51 1 11 1 31R				1
38	ITEM 52 1 11 1 31R				1
39	ITEM 53 1 11 1 31R				1
40	ITEM 54 1 11 1 31R				1
41	ITEM 55 1 11 1 31R				1
42	ITEM 56 1 11 1 31R				1
43	ITEM 57 1 11 1 31R				1
44	ITEM 58 1 11 1 31R				1
45	ITEM 59 1 11 1 31R				1
46	ITEM 60 1 11 1 31R				1
47	ITEM 61 1 11 1 31R				1
48	ITEM 62 1 11 1 31R				1
49	ITEM 63 1 11 1 31R				1
50	ITEM 64 1 11 1 31R				1
51	ITEM 65 1 11 1 31R				1
52	ITEM 66 1 11 1 31R				1
53	ITEM 67 1 11 1 31R				1
54	ITEM 68 1 11 1 31R				1
55	ITEM 69 1 11 1 31R				1
56	ITEM 70 1 11 1 31R				1
57	ITEM 71 1 11 1 31R				1
58	ITEM 72 1 11 1 31R				1
59	ITEM 73 1 11 1 31R				1
60	ITEM 74 1 11 1 31R				1
61	ITEM 75 1 11 1 31R				1
62	ITEM 76 1 11 1 31R				1
63	ITEM 77 1 11 1 31R				1
64	ITEM 78 1 11 1 31R				1
65	ITEM 79 1 11 1 31R				1
66	ITEM 80 1 11 1 31R				1
67	ITEM 81 1 11 1 31R				1
68	ITEM 82 1 11 1 31R				1
69	ITEM 83 1 11 1 31R				1
70	ITEM 84 1 11 1 31R				1
71	ITEM 85 1 11 1 31R				1
72	ITEM 86 1 11 1 31R				1
73	ITEM 87 1 11 1 31R				1
74	ITEM 88 1 11 1 31R				1
75	ITEM 89 1 11 1 31R				1
76	ITEM 90 1 11 1 31R				1
77	ITEM 91 1 11 1 31R				1
78	ITEM 92 1 11 1 31R				1
79	ITEM 93 1 11 1 31R				1
80	ITEM 94 1 11 1 31R				1
81	ITEM 95 1 11 1 31R				1
82	ITEM 96 1 11 1 31R				1
83	ITEM 97 1 11 1 31R				1
84	ITEM 98 1 11 1 31R				1
85	ITEM 99 1 11 1 31R				1
86	ITEM 100 1 11 1 31R				1

NUMEC			
NUCLEAR MATERIALS & EQUIPMENT CORP.			
APOLLO, PA. USA			
GETR TRAIL CABLE FACILITY			
CAPSULE ASSEMBLY			
SCALE	NONE	DWG	11-C-137
DRAWN	W. TRAVIS/AB 948	NO	OF
CHECKED		REV	
APPROVED			

Ref. Dwg. 11-C-095
Rev. Dwg. 11-C-098

steel walls. Air samples and smear papers indicate a relatively low level of removable contamination, but beta-gamma survey meters show a general radiation field of 250 to 300 mr/hr beta-gamma with localized readings as high as 30 to 50 r/hr. Further attempts will be made to lower the activity before attempting modification of the cell interior to allow acceptance of the alpha enclosure. This alpha enclosure has been fabricated, and the associated radiation-resistant windows have been obtained.

Construction and assembly of the alpha boxes for the hot cell is continuing. A prototype hydraulic lifting mechanism to allow positioning of the alpha box within the hot cell has been constructed and satisfactorily tested.

REACTOR PHYSICS AND ENGINEERING PARAMETRIC STUDIES

Task 8.00
K. H. Puechl

Assessment of Plutonium Potential in Near-Thermal Reactors
(W. Ross, J. Ruzbacki)

Previous studies⁽ⁱ⁾⁽ⁱⁱ⁾ have indicated that in converter reactors, optimum fuel cycle costs with plutonium as the fuel are achieved in lattices that are generally looser than present-day uranium fueled lattices. With identical restrictions on heat transfer and minimum fuel element size, it would therefore appear that plutonium loaded cores must be operated at a lower power density relative to uranium with a concomitant larger core for the same power output, hence, a larger associated capital cost. These same studies also indicated that the major contribution to the fuel cycle costs for plutonium fueled systems is the fuel use charge resulting from the relatively high required plutonium to natural uranium ratio. Further, these studies showed that the core life of plutonium fueled systems based only on physics considerations is appreciably greater than what can be reasonably anticipated in practice due to materials limitations. Consideration of all these aspects leads one to the investigation of the possible effects of the use of an inert fuel diluent such as ZrO_2 . Specifically, if the fuel diluent were entirely without effect on the nuclear characteristics, then the use of such diluent

- 1) would leave the optimum moderation, water to heavy element ratio, unchanged; thereby a tighter lattice would result, and more power could be gotten out of a specified core size;
- 2) would leave the total amount of plutonium required within the core unchanged; hence, would not reduce the core inventory or use charge;
- 3) would leave the reactivity-limited core life in terms of megawatt-days per unit weight of plutonium unchanged, hence would leave the reactivity-limited core life in terms of integrated power output per fuel element unchanged, hence because of the inert nature of the diluent would make this core life more nearly equivalent to that which could be obtained in practice.

(i) K. H. Puechl, "The Potential of Plutonium in Near-Thermal Converter Reactors", Nuclear Science and Engineering 12, 135 ± 150 (1962).

(ii) NUMEC P-100, Progress Report, "Development of Plutonium-Bearing Fuel Materials", page 54.

It is seen, therefore, that by the use of an inert diluent in conjunction with plutonium-uranium fuel, capital costs can probably be reduced, and actual core life can probably be extended.

Since no diluent exists that has absolutely no effect on the nuclear characteristics of the core, calculations were performed to study the effects of ZrO_2 diluent. The results generally confirm the above hypotheses. Specifically, it was found that dilution up to 75% by volume of ZrO_2 is possible without a significant increase in fuel cycle cost, assuming that it costs just as much to fabricate a rod of a particular size no matter what the composition. On this basis, the calculated increase in fuel cycle cost at 75% dilution with ZrO_2 was only 2% above the cost calculated without dilution. Further, at 75% dilution it was found that the optimum water to fuel element volume ratio was reduced to 70% of the corresponding optimum value calculated without dilution. Finally, it was found that the reactivity-limited core life in terms of integrated power output per fuel element was essentially the same for 75% dilution as for no dilution (equivalent to 60,000 MWD/Tonne at no dilution).