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NUMEC P-60

PROGRESS REPORT

For Period January 1 through March 31, 1961
AEC R&D Contract AT(30-1)-2389

DEVELOPMENT
OF
PLUTONIUM BEARING FUEL MATERIALS
NUMEC P-60

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by authority of *Patent Group* *dtel 6-13-61*
by *J. C. Rideman* TISOR, date *8-10-61*

Nuclear Materials and Equipment Corporation
Apollo, Pennsylvania

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Previous Quarterly Progress Reports issued in this series are:

<u>Number</u>	<u>For the Period Ending</u>
NUMEC P-10	December 31, 1959
NUMEC P-20	March 21, 1960
NUMEC P-30	June 30, 1960
NUMEC P-40	September 30, 1960
NUMEC P-50	December 31, 1960

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PROJECT AND FACILITY ADMINISTRATION

Task 1.00
H. J. Garber

Equipment Installation

Revisions and checking out of the ventilation system for the facility and glove-box lines were completed during the middle of this quarter. Immediately following this, the fourteen higher-priority boxes and the transfer tunnel were committed to operations with plutonium. These include the equipment for carrying out fuel material preparation and characterization, wet and instrumental analytical chemistry, powder feed preparation, and pellet pressing, ceramic evaluation, pellet sintering, and metallography.

Completion of installation of apparatus in a number of intermediate-priority boxes was resumed later this quarter. By the end of June 1961 it is expected that 25 boxes will be in operation on plutonium. Currently, work is in progress on completion and commitment of the boxes for particle size distribution by gravitational sedimentation, thermogravimetric balances, carbon-hydrogen-oxygen analysis, chemical reprocessing, mechanical packing and plasma-flame spheroidization, Stokes pellet press, swager, high uranium-low plutonium powder preparation, and contaminated equipment maintenance machine shop.

A number of conceptual designs of alpha-boxes, to be used in hot-cell examination and processing of irradiated materials were completed and studied with the goal of attaining maximum operational flexibility and full plutonium containment. This work, augmented by a model study, is now serving as a basis for detailed design of the alpha-boxes. Solicitation of bids for construction of the alpha-boxes is scheduled for mid-May.

Summary of Experimental Activities

Three plutonium preparations were made and partially characterized. These included pure PuO_2 via the batch-oxalate route and $\text{PuO}_2\text{-UO}_2$ via the continuous co-precipitation ADU-hydroxide routes in the 0.5 a/o Pu to 20 a/o Pu range, using metal nitrate solutions as the feed materials. Operating conditions for precipitation, digestion, filtering, calcination, and reduction for these preparations were based on the results of the UO_2 preparation studies previously discussed in NUMEC P-40 and NUMEC P-50. Overall yields for these initial studies were in excess of 95 per cent. The resulting products were relatively fluffy, having surface areas in the range 9 to 29 m^2/gm , sub-sieve average particle sizes ranging from 0.92 to 5.4 microns, bulk densities ranging from 1.24 to 1.80 gm/cc , and tap densities ranging from 1.91 to 2.24 gm/cc .

The PuO_2 preparation (surface area $28.7 \text{ m}^2/\text{gm}$) displayed a strong tendency for weight increase upon exposure to the box atmosphere, presumably due to moisture and oxygen pickup. Further elucidation of this behavior will be attempted upon commitment of the thermogravimetric balance box to plutonium.

Dry pressing and sintering studies were conducted on the pure PuO_2 preparation. Die binding difficulties due to fines wedging between the punches and die-body were overcome by preliminary dry-slugging at 6500 psi in a 0.467 inch diameter die followed by hand granulation through a 20-mesh screen. The green strength of these pellets (0.19 inch diameter, L/D ratio 1 to 2.4) pressed in the range 10 to 90 tsi was excellent. For a given pressing pressure, the green density is virtually independent of the L/D ratio. Green pellet densities (4.5 to 7.0 gm/cc) increased essentially linearly with the pressing pressure in the range 10 to 90 tsi.

Sintering trials were conducted at 1400°C and 1600°C in a 5% hydrogen-95% nitrogen environment. The four-hour heat-up and cool-down steps were held to a temperature change rate no larger than 10°C per minute. A four-hour hold time at temperature was used in all cases. Densities of the sintered pellets, as determined by the weight in air-weight in water method, ranged from 10.2 to 11.0 gm/cc, corresponding to percentage theoretical densities of 88.9 to 95.9 respectively. The small size of the pellets used in these first trials militated against the precise density determination needed for determination of definitive sintering characteristics. Consequently, subsequent trials will be made with larger specimens. These first tests show that the sintered density of PuO_2 pellets is relatively independent of green density above 5.6 and 6.0 gm/cc (corresponds to pressing pressures of 50 tsi) for pellets sintered at 1600°C and 1400°C respectively. The only apparent advantage of dry pressing at pressures above 50 tsi is a reduction in shrinkage associated with sintering.

The degree of solid solution formation and atomistic homogenization which accompanies sintering of $\text{PuO}_2\text{-UO}_2$ mixtures has important consequences in the performance of these materials as reactor fuels. In order to throw light on this problem, an investigation of the influence of the key variables - sintering temperature and time, and history and character of the feed materials - has been started. The tools to be employed to determine the degree of homogenization and solid solution formation include x-ray diffraction, differential thermal analysis, and selective metallography.

The mechanical packing studies described previously in NUMEC P-40 and NUMEC P-50 were deferred temporarily in favor of installing and checking out the plasma-torch spheroidization equipment and preparing the spherical particles needed to conduct the packing studies. This work has demonstrated the need for revisions in the torch feed system and methods for simplifying and lowering the cost of the gas-recycle system, and the necessity for using high density feed materials free of gas if essentially theoretical density product is required.

Summary of Reactor Physics Studies

Using the NUSURP procedure, as programmed for the LPG-30 computer, based on a revised formulation of the epithermal index (r_{28}), fuel burnout and variation of isotopic composition with time has been computed for a number of near-thermal reactors in order to ascertain the potential of plutonium as a reactor fuel. The computations made to date indicate that very long reactor lives are attained with a fuel having an initial composition of 78 a/o Pu-239, 12 a/o Pu-240, and 10 a/o Pu-241, and that the reactivity variation during core life is nominal. This study will first be completed, as applied to several existing power and military reactors, and then will be extended to highly enriched systems.

PREPARATION AND CHARACTERIZATION OF FUEL MATERIALS

Task 2.00

C. S. Caldwell

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Prior Processing Studies on UO₂ Preparation and Characterization

During previous reporting periods and prior to committing glove box equipment to plutonium processing, the influence of precipitation temperature, initial nitrate acidity, intermediate product milling, reduction furnace atmosphere, and method of sintering evaluation were examined in a series of batch UO₂ preparation runs. The primary objective was to determine the influence of several variables on final product characteristics for which literature data were not available. Prime emphasis was placed on high surface area UO₂ powder types capable of being sintered to high density without requiring particle size reduction prior to cold-pressing. Experience gained in preparation and evaluation of these types of UO₂ powders combined with existing knowledge of methods already in use for production of acceptable grades of UO₂ having lower surface area and higher bulk density currently serves as a guide for the selection of suitable conditions for preparing solid solution PuO₂-UO₂ powders by co-precipitation (low Pu/U ratio) and for preparing UO₂ for direct blending with PuO₂. The results of these studies were reported previously in NUMEC P-20, NUMEC P-30, NUMEC P-40, and NUMEC P-50. Evaluation studies on these UO₂ preparations were continued and completed during this quarter. The results of these are being employed to prepare batches of UO₂ which are to be blended and mechanically mixed with PuO₂ in future studies for preparation of reference radiation specimens.

Initial Plutonium Oxide Preparation Studies

Early in the quarter, 14 glove boxes were committed to operations with plutonium. By the end of the next quarter, this number will be increased to 25.

The initial plutonium preparations are centered on pure PuO₂ and PuO₂-UO₂ in the composition range $0.005 \leq \text{Pu/U} \leq 4$. To date three lots of plutonium oxide and plutonium-uranium oxide have been prepared and partially characterized. A summary of the preparation details and characteristics observed is presented in the subsequent sections under Task 2.00. Pellet pressing and sintering evaluations of the powders prepared are discussed under Task 3.00.

Plutonium Dioxide Preparation, Batch Method

Figure 2.1 shows the equipment hook-up used for the preparations.

Starting with a concentrated plutonium nitrate solution, as received, the required amount of stock solution was metered and diluted with sufficient acid to yield approximately one liter of feed solution (100 gm Pu/l; 3 M HNO_3). After preheating the feed to 55°C in a 4 liter, 4 inch diameter cone bottom precipitator, a solution of 1.0 M oxalic acid (containing 0.8 M hydrogen peroxide for Pu IV valence stabilization) was added at a constant rate of 23 ml/minute to precipitate plutonium oxalate $\text{Pu}(\text{C}_2\text{O}_4)_4 \cdot 6\text{H}_2\text{O}$. The 2.5 inch diameter turbine blade agitator tip speed was maintained at 3.3 fps, in order to provide adequate mixing of the liquid phase but sufficiently mild to prevent breakdown of the precipitate agglomerates. A 20 minute digestion period at 55°C was used subsequent to the 45 minute precipitation period. The slurry settling rate was observed to be extremely rapid, indicating that a large agglomerate size had been retained. The slurry was then vacuum filtered into a 5 micron porosity stainless steel filter cannister and washed with 520 ml of 2 M HNO_3 -0.05 M $\text{H}_2\text{C}_2\text{O}_4$. The combined filtrates were observed to be free of particulate material. The filtrate and cleanup wastes were retained for subsequent recovery of plutonium.

After vacuum dewatering, the filter cannister and precipitate (on inside of cannister) were transferred to a forced air drying oven and the cake was dried stepwise at 80°C (5 hr), 130°C (5 hr), and 180°C (1 hr) with periodic removals to permit observation of color changes and appearance.

After cooling the dried cake and transferring to a stainless steel furnace flask, the product was calcined at an average temperature of 350°C in air for one hour to yield a finely divided, olive-brown colored PuO_2 powder. The overall yield was approximately 95 per cent. Work at other laboratories with similar processes have shown that the process yield can be expected to vary between 95 and 98 per cent.

The BET surface area of the PuO_2 powder was measured by krypton adsorption and was found to be $28.7 \text{ m}^2/\text{gm}$. The average particle size of the product, as measured by the Fisher Sub-Sieve Sizer (air permeability) method, was 3.1 microns. Bulk and tap densities were 1.8 and 2.2 gm/cc respectively. Chemical analyses of the starting and final materials are in progress, and the results will be presented in the next quarterly progress report.

20 w/o PuO_2 -80 w/o UO_2 Preparation, Continuous Co-precipitation

A 20 per cent plutonium-80 per cent uranium nitrate feed solution, approximately one molar in free nitric acid, was prepared by transferring a measured volume of concentrated plutonium nitrate stock solution into a cone-bottom polyethylene feed tank containing a measured quantity of

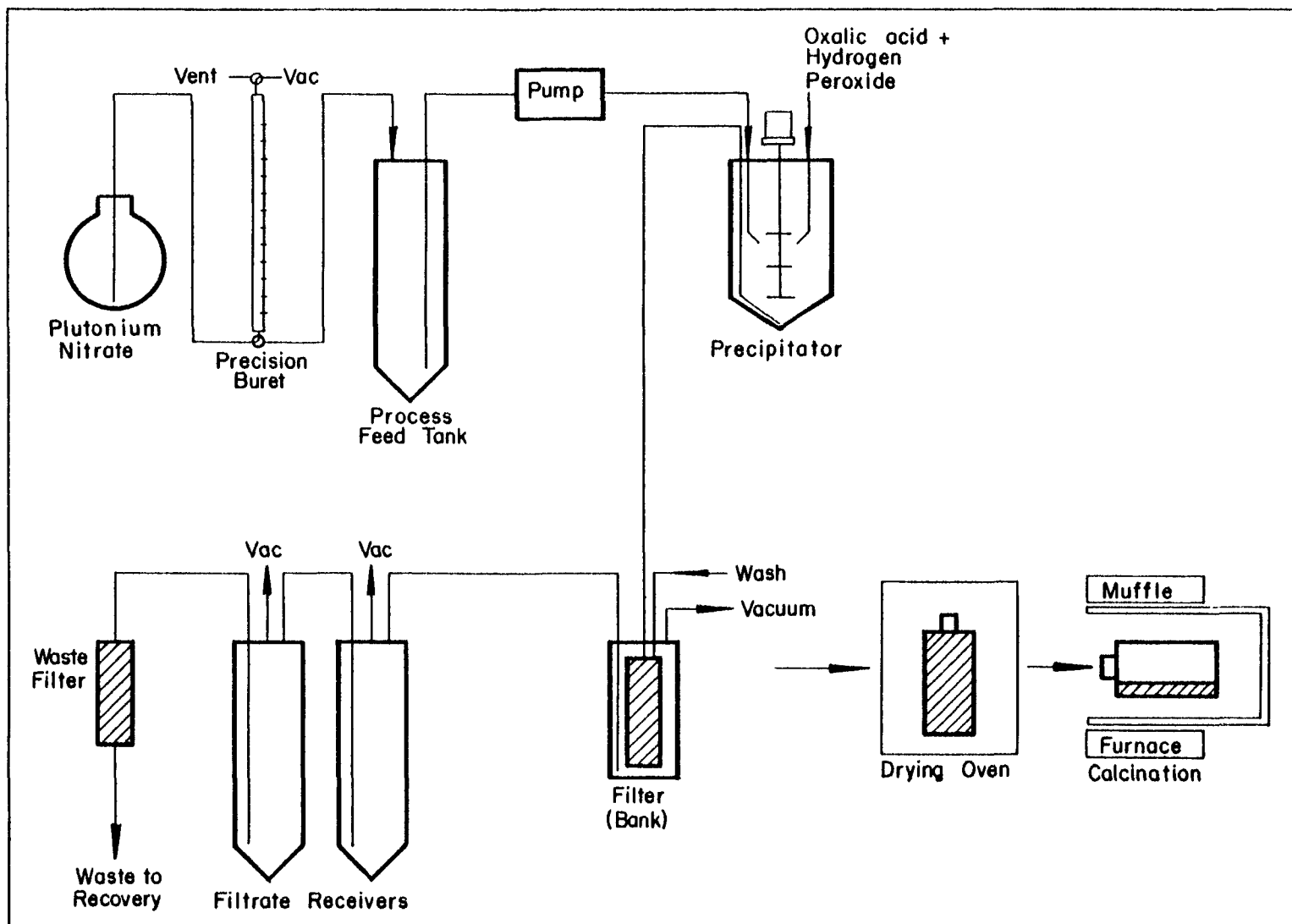


Figure 2.1
PLUTONIUM OXIDE PREPARATION EQUIPMENT

uranyl nitrate-nitric acid solution. The solution was mixed by recycling using a teflon-bellows positive displacement pump. The 4-liter cone-bottom precipitator tank was filled to the 500 ml level with a pH 8 heel solution, synthesized to match the composition of the expected ammonium nitrate filtrate. After preheating the heel to 65°C, the metal nitrate and the concentrated ammonium hydroxide solutions were pumped into the precipitator at predetermined rates calculated to maintain the filtrate pH above 8.5. After stabilizing conditions at pH 10, continuous vacuum filtration of the slurry was maintained for the remaining 30 minutes of the run in order to hold a constant level in the precipitator. The turbine agitator tip speed was maintained at 3.3 fps in order to duplicate conditions used in previous ADU studies. Control of the filtration rate was easily maintained, and a clean filtrate was obtained, having an estimated residual plutonium concentration of 30 μ gm/ml.

After dewatering the precipitate contained in a porous stainless steel filter cannister, it was transferred to a forced-convection oven where the cake was dried at 180°C for 4 hours. After transferring to an Inconel furnace flask, the product was heated in nitrogen and held at 540°C for 70 minutes, followed by reduction in 5.5% hydrogen-94.5% nitrogen at 540°C for 90 minutes. The product was cooled rapidly in the reducing gas mixture to room temperature, and was then contacted with a 7% oxygen-93% nitrogen mixture for 30 minutes to insure a small amount of surface oxidation and prevent later spontaneous oxidation. The product was inspected first under a nitrogen atmosphere and was then transferred to a storage container under an air atmosphere.

The resultant PuO₂-UO₂ powder surface area was found to be 17.8m²/gm. Other properties determined were the bulk density (1.68 gm/cc), tap density (2.15 g/cc) and "subsieve" particle size (5.4 microns). The particle size distribution curve for the product (No. 297-Pu-3) as determined by the MSA centrifuge method is shown in Figure 2.2.

0.5 w/o PuO₂-99.5 w/o UO₂ Preparation, Continuous Co-precipitation

Utilizing a precision volumetric dispensing buret, a small quantity of concentrated plutonium nitrate stock solution was measured and transferred to a polyethylene feed tank containing a predetermined quantity of acidified 0.42 M uranyl nitrate solution. The stock solution was mixed by recycling via an external pumping circuit. A 500 ml filtrate heel was first preheated to 65°C in the precipitation vessel, and subsequently the metal nitrate and ammonium hydroxide streams were pumped in at constant rates to yield a co-precipitated product. After stabilizing the pH at 8.6, vacuum filtration was initiated to remove slurry from the precipitation vessel, thus maintaining a constant level while adding fresh reactants continuously.

The average reaction holdup time for the precipitation step was maintained at 20 minutes, and the turbine impeller tip speed was maintained at 2.2 fps. The filtrate pH increased slowly from 8.6 to 9.2 during the course of the run, and filtration progressed smoothly, although a relatively high wet cake density was observed. Several attempts were made to wash the cake with dilute NH_4OH solutions; however, peptization occurred readily, in contrast with previous experience with normal ADU precipitation. Additional laboratory scale runs are in progress to establish satisfactory washing conditions for this type of precipitate.

The filter cake was then transferred to a forced convection drying oven and held at 180°C for 6 hours. After drying, the product was transferred to an Inconel flask which was inserted into a horizontal tube furnace. The charge was heated in nitrogen, held in nitrogen at 540°C for 70 minutes, reduced in 5.5% hydrogen-94.5% nitrogen at 540°C for 90 minutes, then cooled rapidly in the reducing gas mixture. The product was unloaded in contact with air, and it was observed that a small amount of surface oxidation had occurred. The product BET surface area was measured by krypton adsorption and found to be $9.5 \text{ m}^2/\text{gm}$. The bulk and tap densities were 1.24 and 1.91 gm/cc respectively, and the average particle size by the air permeability method was 0.9 microns. By way of comparison, the properties of pure UO_2 prepared from a uranyl nitrate feed solution having the same initial HNO_3 acidity are seen to correspond generally with those given above. The UO_2 properties were as follows: BET surface area - $9.3 \text{ m}^2/\text{gm}$; bulk density - 1.34 gm/cc; tap density - 2.1 gm/cc. The particle size distribution curve for this sample (No. 297-Pu-2), as determined by MSA centrifugal method, is shown in Figure 2.3.

A summary of the pertinent preparation details and properties of the three plutonium bearing products synthesized to date are presented in Table 2.1.

The following additional information is also being determined on the three types of oxides prepared during this reporting period.

- 1) Chemical and spectrographic analysis of final product, including Pu/O and Pu/U/O ratios.
- 2) Stability with respect to air oxidation.
- 3) Stability with respect to moisture pickup.
- 4) Nitric acid solubility.

Subsequently, thermogravimetric curves for the intermediate products will also be mapped out.

In addition to gaining valuable experience in the operation of process equipment with several types of plutonium materials, the runs carried

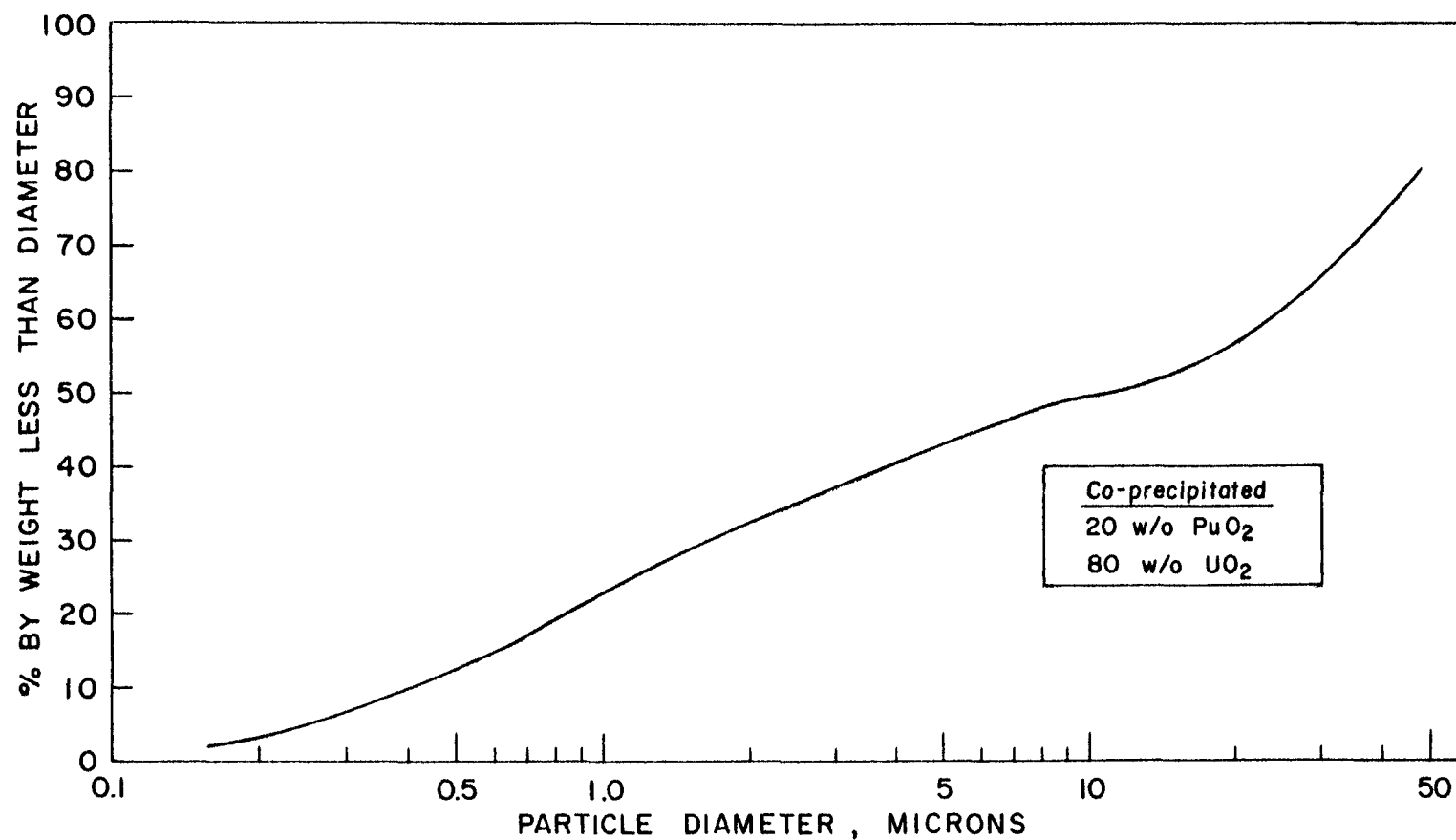


Figure 2.2
PARTICLE SIZE DISTRIBUTION FOR SAMPLE 297-Pu-3

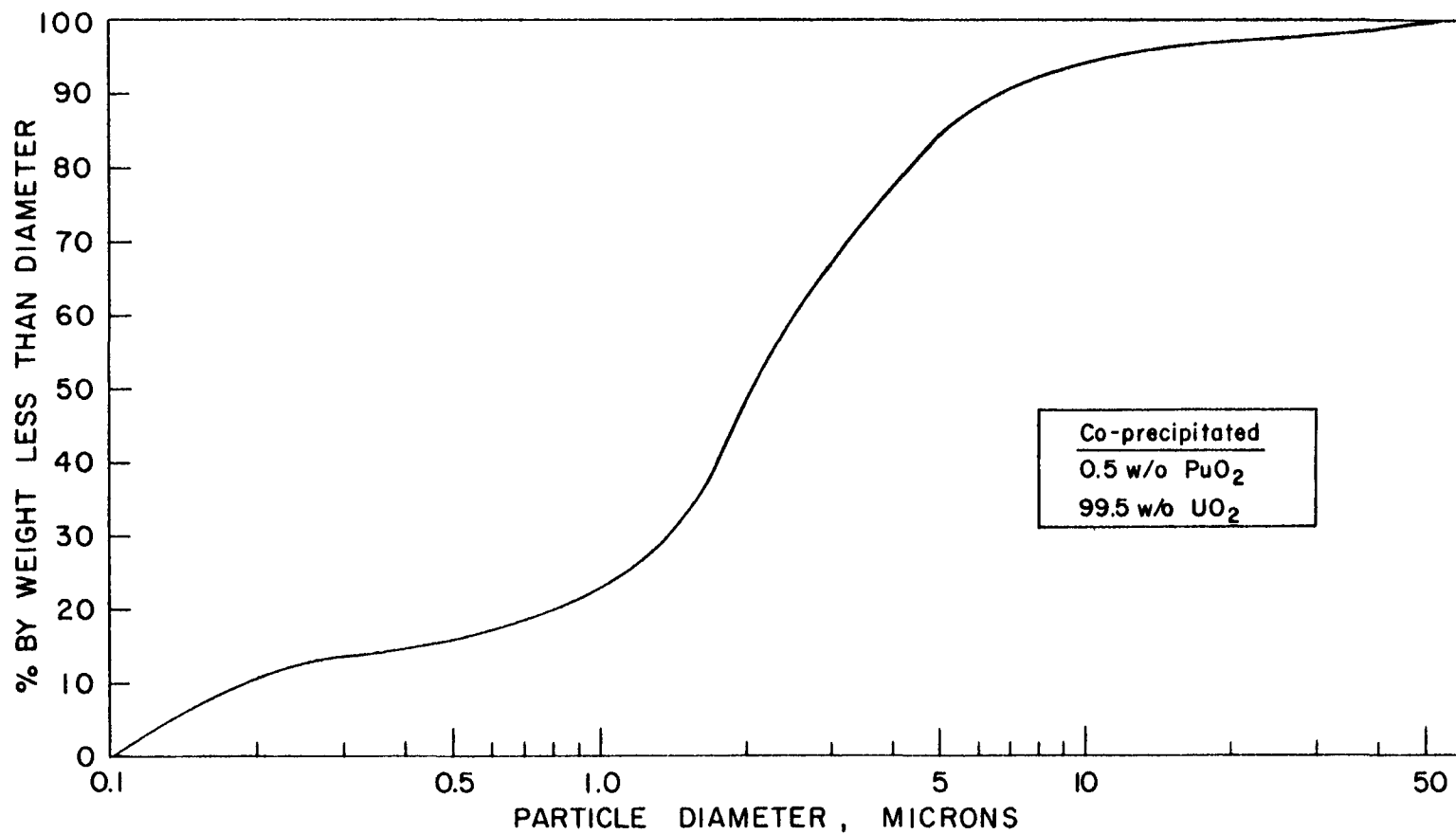


Figure 2.3
PARTICLE SIZE DISTRIBUTION FOR SAMPLE 297 - Pu - 2

Table 2.1

Summary of Conversion Processing and Property Data
Plutonium Oxide Materials

	Run No.		
	297-Pu-1	297-Pu-2	297-Pu-3
End Product	PuO ₂	0.5w/oPuO ₂ - 99.5w/oUO ₂	20w/oPuO ₂ - 80w/oUO ₂
Precipitation Conditions			
Method	Batch	Continuous	Continuous
Temperature, °C	55	65	65
Feed Composition			
gm Pu/liter	100	0.59	20
gm U/liter	-	99.5	80
H ⁺ , molarity	3.0	0.96	1.0
Strike Solution Composition, molarity			
H ₂ C ₂ O ₄	1.0	-	-
H ₂ O ₂	0.8	-	-
NH ₄ OH	-	14.5	14.5
Furnace Conversion Temperature, °C	350	540	540
Powder Characteristics			
Color	Olive-Brown	Dark-Brown	Black-Brown
Surface area, m ² /gm	28.7	9.5	17.8
Sub-sieve average particle size, microns	3.1	0.92	5.4
Bulk Density, gm/cc	1.80	1.24	1.68
Tap Density, gm/cc	2.24	1.91	2.15

out to date provided sufficient starting material for preliminary pressing and sintering trials and for testing a number of methods required for control analysis.

Future Preparation and Characterization Runs

During the next quarter, a comprehensive series of preparation and characterization runs will be completed as outlined in Table 2.2. This work will synthesize sufficient material for blending and sintering trials on a wide variety of powder combinations and for early irradiation specimen fabrication. Characterization of oxide powders and intermediate compounds will utilize all available glove-box facilities. The following experimental equipment has been installed and has been placed in plutonium service since the last reporting period:

- 1) BET (Krypton Adsorption) surface area unit.
- 2) Innes (Nitrogen Adsorption) surface area unit.
- 3) Fisher Subsieve Sizer (Air Permeability)
- 4) M.S.A. (Whitby) centrifuge particle size analyzer.
- 5) Bulk and tap density measuring units.
- 6) Powder density (pycnometer) measuring unit.
- 7) Deltatherm D-2000 D.T.A. apparatus.

Within a short period, the automatic recording Aminco thermobalance will also be in service with plutonium materials.

In conjunction with the aforementioned work, the decomposition, calcination and reduction behavior of the oxalate and ADU-type precipitates will be examined thermogravimetrically. This will aid process control and make for improved reliability. Among the reactions to be evaluated are:

- 1) Decomposition of plutonium oxalate.
- 2) Decomposition of ADU-plutonium hydroxide.
- 3) Stability of $\text{UO}_2\text{-PuO}_2$ re-oxidation.

Analytical Chemistry

The investigation of the spectrographic rotating disk method for the determination of trace elements in plutonium and uranium materials previously described in NUMEC P-50 (p. 25-26) was continued. The other

Table 2.2

Summary of Scheduled Preparation Runs

Number of Replicate Runs	Product Oxide	Grams Product Per Run	Precipitation Conditions	Furnace Processing Temperatures, °C	Objectives
2	PuO ₂	120	Continuous oxalate	350 420 490 560	Determine reproducibility of precipitation and furnace processes; calcination temperature vs surface area and powder properties; provide material for blending and sintering trials to determine compatability of PuO ₂ with UO ₂ .
4	20% PuO ₂ -80% UO ₂	240	Continuous ADU-hydroxide	540 590 640 690	Determine reproducibility of precipitation and furnace processes; reduction temperature vs surface area and powder properties; provide material for blending and sintering trials.
4	5% PuO ₂ -95% UO ₂	240	Continuous ADU-hydroxide	540 590 640 690	Provide reserve material for blending and sintering trials.
2	UO ₂	5,000	Continuous ADU	540	Provide reserve material for blending and sintering trials.
2	UO ₂	5,000	Continuous ADU	640	Provide reserve material for blending and sintering trials.

Table 2.3

Effect of Counter-Electrodes
Rotating Disk Solution Method

Element	Wave Length, °A	Relative Intensity, %T			
		Counter-Electrode		Background	
		Carbon	Copper	Air	Argon
Boron	2497	90	65	-	-
Manganese	2576	40	10	-	-
Lead	2836	90	50	-	-
Silicon	2881	85	40	-	-
Molybdenum	3132	75	15	15	85
Molybdenum	3170	80	35	-	-
Vanadium	3185	85	45	15	85
Cadmium	3261	50	15	15	85

major effort was the completion of the installation and testing of equipment and methods in four glove boxes and the commitment of these to plutonium work.

The spectrographic study was extended to solutions from which uranium and plutonium is separated from microgram quantities of other elements by a thenoyltrifluoroacetone extraction. The effect of carbon versus copper counter-electrode and of an inert atmosphere on the detection limits of boron, cadmium and other elements was investigated. As shown in Table 2.3, marked increase of sensitivity, indicated by the lower %T densitometer values, is attained with the use of a copper counter-electrode. Also, the background due to a continuum in the vicinity of the Mo, V and Cd emission lines was markedly reduced. It is planned to appraise the other variables and to establish optimum conditions for this procedure.

Equipment Installation: The analytical chemistry facilities are located in six glove boxes; a few additional tests remain to be completed before the last three can be committed to plutonium work.

In one of the wet chemistry boxes, the space is divided between analytical balances (macro and a micro single-pan balance and a micro electrobalance) and apparatus for carrying out electrochemical analyses. The latter include cells for controlled potential coulometry, polarography, automatic micro-burettes and accessory electrodes for potentiometric titration. Also included is a special inner-box for handling of powder materials to minimize alpha activity spread. Semi-micro or micro techniques are being used to speed up the analysis and reduce handling large quantities of plutonium. Auxiliary chemical preparations are carried out in an adjacent box; transfers of material between the two boxes is via a positive-seal pass-through port. The dissolution of samples and/or adjustment to proper acid environment is accomplished by heating with Glas-col heating mantles and Erlenmeyer flasks which are connected with standard taper inter-joints to condensers for refluxing or distillation. This procedure eliminates the need for fume hoods and the accessory scrubbing systems. Fusion of refractory samples is carried out in a small electric furnace. Dilution of samples to calibrated volumes and aliquoting of samples are likewise performed within this box. Other auxiliary equipment include an infra-red lamp, micro drying oven, suction pump, planchet evaporation oven, small ion exchange columns, and a set of Jones reducers. The severe corrosion and contamination problems that exist in this box, because of the manifold operations conducted therein, were minimized by painting the interior walls with strippable paint to facilitate subsequent decontamination.

The additional analytical chemistry box system which has been committed to plutonium work comprises a set: a large box which houses larger instruments, and a smaller satellite box which is centered in line with the optical bench of the dual grating Bausch and Lomb Spectrograph.

The emission source of the arc-spark stand, which is located within the small box, is focused through a quartz window onto the entrance slit of the spectrograph. The aforementioned larger glove box houses the spectrophotometric equipment, including a flame photometer and a number of accessories. The exhaust fumes from the flame photometer housing pass through a water cooled stack prior to entering into the box ventilation system. The accessories include two alpha-measuring instruments (housed in a rigid plastic tent) connected through cables to scalars outside the glove box. The intended function of the counting equipment is to estimate the relative magnitude of the alpha activity from separation processes.

The remaining analytical chemistry boxes are in the process of completion. In one, whose primary function is to house the x-ray apparatus, liquid-liquid extraction apparatus have been installed. This location was selected in order to eliminate the danger from combustible fumes associated with the use of organic extractants. Commitment of this box to plutonium work is awaiting replacement of the nickel x-ray diffraction window by beryllium.

The remaining analytical chemistry box houses equipment for determining hydrogen, carbon, and oxygen by high frequency induction heating and gas evolution methods. These have been installed together with other physiochemical measuring apparatus (Aminco and Chevenard thermogravimetric balances). Tests are in progress to evaluate the efficiency of heating with extended induction coils and to establish the effect of absolute filters located between the evolution apparatus and the measuring devices which are located outside the glove box.

FABRICATION AND EVALUATION OF FUEL SHAPES

Task 3.0

L. J. Jones

B. M. Cinai	R. M. Horgos	R. K. McGeary
E. K. Halteman	J. K. Koeneman	M. M. Turkanis

In order to evaluate the proper fabrication procedures to be used to form solid solutions of plutonia and urania, the characteristics of each component as they affect the pressing and sintering behavior must be known. This is especially important when considering mechanical mixing, pressing, and sintering of the components. For this reason, initial operations performed in the NUMEC powder box line have been directed toward establishment of the behavior of pure plutonia and the effects of its preparative history on sintering. This information, together with the information garnered from the work with UO_2 summarized in previous quarterly progress reports, will be used to set the specifications for the pure powders to be used in the mechanical mixing and sintering studies.

Water Absorption in Plutonia Powder

It was observed during initial handling of the high surface area powder (No. 297-Pu-1) that an increase in weight was occurring on exposure of the samples to the box atmosphere. A small batch of powder was subsequently vacuum dried for 2.5 hours at 75°C and 27 inches vacuum. Assuming moisture absorption to be the cause, it was expected that the drying treatment would result in a weight loss. However, as shown in Figure 3.1, this treatment resulted in an increase in weight of 0.83 per cent. The reason for this increase is not fully established at this time. One possible explanation is that the original material may not have been the dioxide but rather a mixture of Pu_2O_3 and PuO_2 .

Additional exposure at room temperature caused an increase in weight of 0.94 per cent. A second drying for 12 hours at 75°C and 27 inches vacuum resulted in a decrease in weight to approximately the same weight existing after the first drying operation. As shown in Figure 3.1, further exposure to the box atmosphere at room temperature resulted in a steady increase in weight, presumably due to the pickup of moisture, up to approximately 110 hours when a maximum increase of 1.77 per cent was observed. An additional exposure of 66 hours resulted in no further change. A drying treatment at 130°C , in an attempt to break down higher hydrates, resulted in a decrease of 1.20 per cent. However, this final weight was still 0.57 per cent higher than the original weight of the powder. During sintering at 1400 and 1600°C for four hours in a 5% hydrogen-95% nitrogen atmosphere, the pellets lost $3.88 \pm 0.60\%$ of their weight. There was no significant difference in the

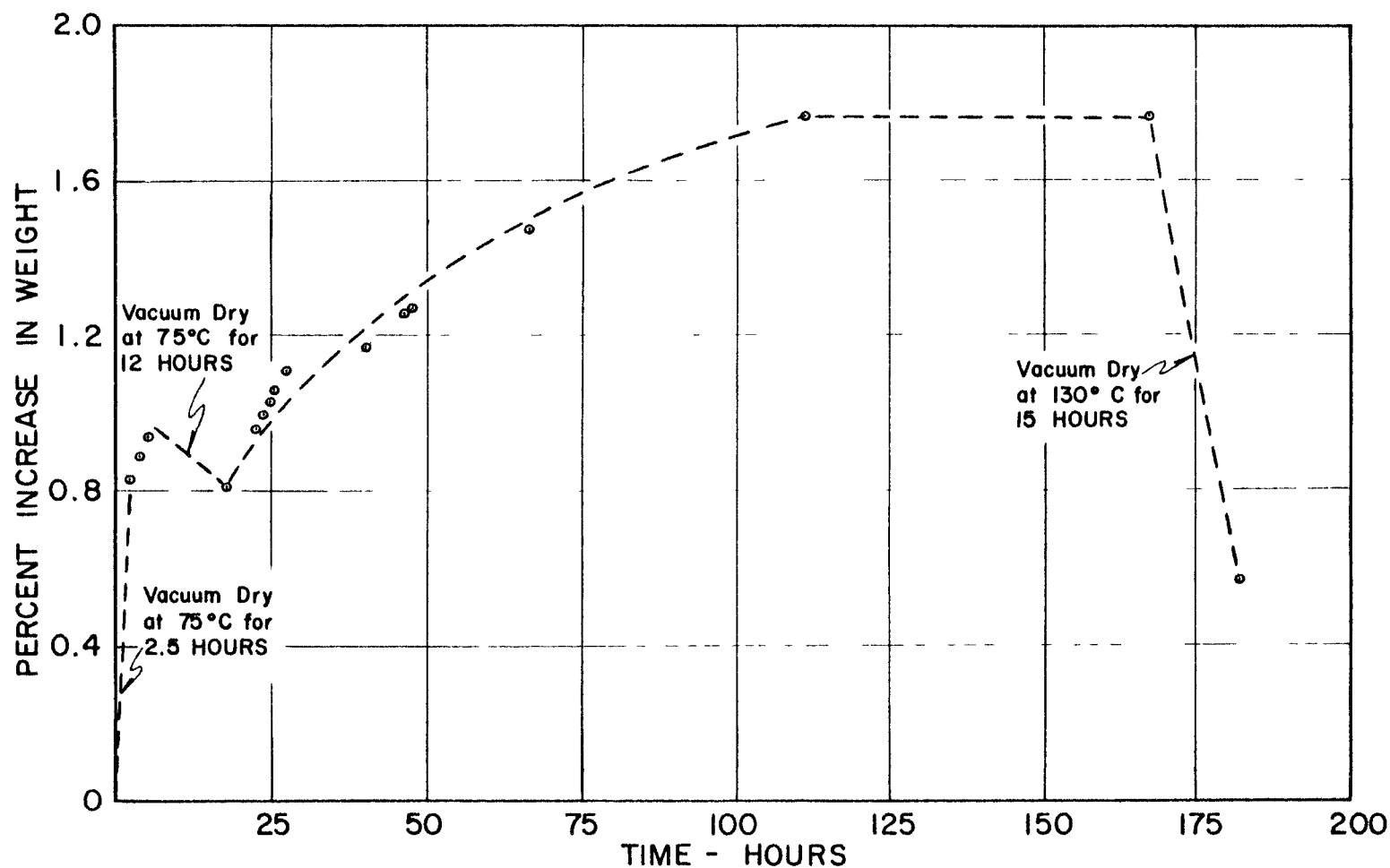


Figure 3.1
MOISTURE ABSORPTION IN HIGH SURFACE AREA Pu O_2
MATERIAL : 297-Pu-1

weight losses between the pellets sintered at 1400°C and those sintered at 1600°C. To be sure, some of the weight loss can be attributed to mechanical abrasion of the green pellets during handling; however, this amount is believed to be small since the consistency of the individual weight losses would preclude chance handling losses.

During the coming quarter, further attempts to explain and establish these phenomena will be carried out. Of special significance to this study will be experiments in which the samples will be exposed to a set of controlled atmospheres in the Aminco thermogravimetric balance. The mass changes to be observed in these experiments should explain the mass increases noted in the preliminary work with high-surface plutonia.

Dry Pressing Characteristics

Attempts to press the as-received powder were unsuccessful in that the punches could not be pressed readily from the die. This was presumably due to the binding of fine powder between the punches and the die body. In order to overcome this difficulty, the powder was dry-slugged at 6500 psi in a 0.467 inch diameter die and hand granulated through a 20 mesh screen. The resultant granules were hand pressed, without the addition of binder, in an 0.187 inch diameter die at pressures ranging from 5 to 95 tsi. Sterotex was used as the die and punch lubricant and was applied as a saturated solution in trichlorethylene. The green strength of all pellets pressed at 10 tsi and over was excellent, and no difficulties were encountered during pellet press-out or punch removal.

The pressing parameters and resultant green densities are shown in Table 3.1 and are plotted in Figure 3.2. It will be noticed that a maximum in green density was not reached over the range of pressures used and that the density increased almost linearly with increasing pressure.

To determine the effect of L/D ratio on green density, a number of pellets were pressed at 70 tsi with the powder charge being varied. The L/D ratios vary from 1.0 for pellet No. 41 to 2.4 for pellet No. 31. It can be seen in Table 3.1 that the variation in green density is not significantly affected by L/D ratio within this range.

Sintering Studies

Pellets were chosen to represent the spectrum of green density achieved and separated into two groups for sintering at 1400 and 1600°C. The sintering cycle for both sintering runs consisted of a four-hour heat-up period to achieve maximum temperature, a four-hour period at temperature, and a four-hour cooling period. Heating and cooling of the pellets were accomplished by moving the molybdenum boat containing the pellets an inch at a time into or out of the furnace, which was held at constant temperature throughout the run. The maximum rate of pellet heating or cooling was

Table 3.1

Cold Pressing Characteristics

Pellet No.	Pressing Pressure, TSI	Pellet Diam. in	Pellet Length, in	Pellet Weight, gm	Pellet Density, gm/cm ³	% of Theoretical Density
297-Pu-1-4	5	0.188	0.474	0.9704	4.50	39.3
297-Pu-1-5	7.5	0.188	0.422	0.8500	4.43	38.7
297-Pu-1-6	10	0.188	0.414	0.8363	4.44	38.7
297-Pu-1-7	12.5	0.188	0.385	0.8149	4.66	40.7
297-Pu-1-8	15	0.188	0.380	0.8409	4.87	42.5
297-Pu-1-9	17.5	0.188	0.360	0.8197	5.01	43.7
297-Pu-1-10	20	0.188	0.374	0.8599	5.06	44.2
297-Pu-1-11	25	0.188	0.341	0.8064	5.21	45.5
297-Pu-1-12	27.5	0.188	0.293	0.7297	5.48	47.8
297-Pu-1-13	30	0.188	0.344	0.8535	5.46	47.6
297-Pu-1-14	32.5	0.188	0.320	0.8166	5.62	49.0
297-Pu-1-15	35	0.188	0.314	0.8111	5.68	49.6
297-Pu-1-16	40	0.188	0.289	0.7889	6.00	52.4
297-Pu-1-17	45	0.188	0.314	0.8430	5.92	51.7
297-Pu-1-18	50	0.188	0.263	0.6909	5.79	50.5
297-Pu-1-19	55	0.188	0.261	0.7024	5.92	51.7
297-Pu-1-20	60	0.188	0.266	0.7302	6.05	52.8
297-Pu-1-21	65	0.188	0.281	0.7937	6.21	54.2
297-Pu-1-22	70	0.188	0.238	0.6992	6.46	56.4
297-Pu-1-23	75	0.188	0.207	0.6322	6.73	58.7
297-Pu-1-24	80	0.1885	0.242	0.7453	6.79	59.2
297-Pu-1-26	90	0.189	0.275	0.8766	6.96	60.7
297-Pu-1-27	70	0.188	0.262	0.8090	6.80	59.3
297-Pu-1-28	70	0.188	0.333	1.0325	6.83	59.6
297-Pu-1-29	70	0.188	0.431	1.2775	6.46	56.4
297-Pu-1-30	70	0.188	0.392	1.2023	6.76	59.0
297-Pu-1-31	70	0.188	0.450	1.3703	6.64	57.9
297-Pu-1-32	45	0.188	0.310	0.8437	5.98	52.2
297-Pu-1-33	50	0.188	0.284	0.8161	6.32	55.1
297-Pu-1-34	55	0.188	0.288	0.8027	6.12	53.4
297-Pu-1-36	20	0.188	0.329	0.7863	5.24	45.7
297-Pu-1-37	25	0.188	0.275	0.6869	5.50	48.0
297-Pu-1-38	60	0.188	0.277	0.8143	6.46	56.4
297-Pu-1-39	85	0.188	0.233	0.7435	7.01	61.2
297-Pu-1-40	70	0.188	0.201	0.6183	6.44	56.2
297-Pu-1-41	70	0.188	0.187	0.5731	6.74	58.8
297-Pu-1-43	70	0.188	0.201	0.6156	6.69	58.4
297-Pu-1-44	70	0.188	0.212	0.6544	6.75	58.9
297-Pu-1-45	70	0.188	0.106	0.3309	6.89	60.1
297-Pu-1-46	60	0.188	0.260	0.7770	6.61	57.7
297-Pu-1-47	65	0.188	0.216	0.6682	6.57	57.3

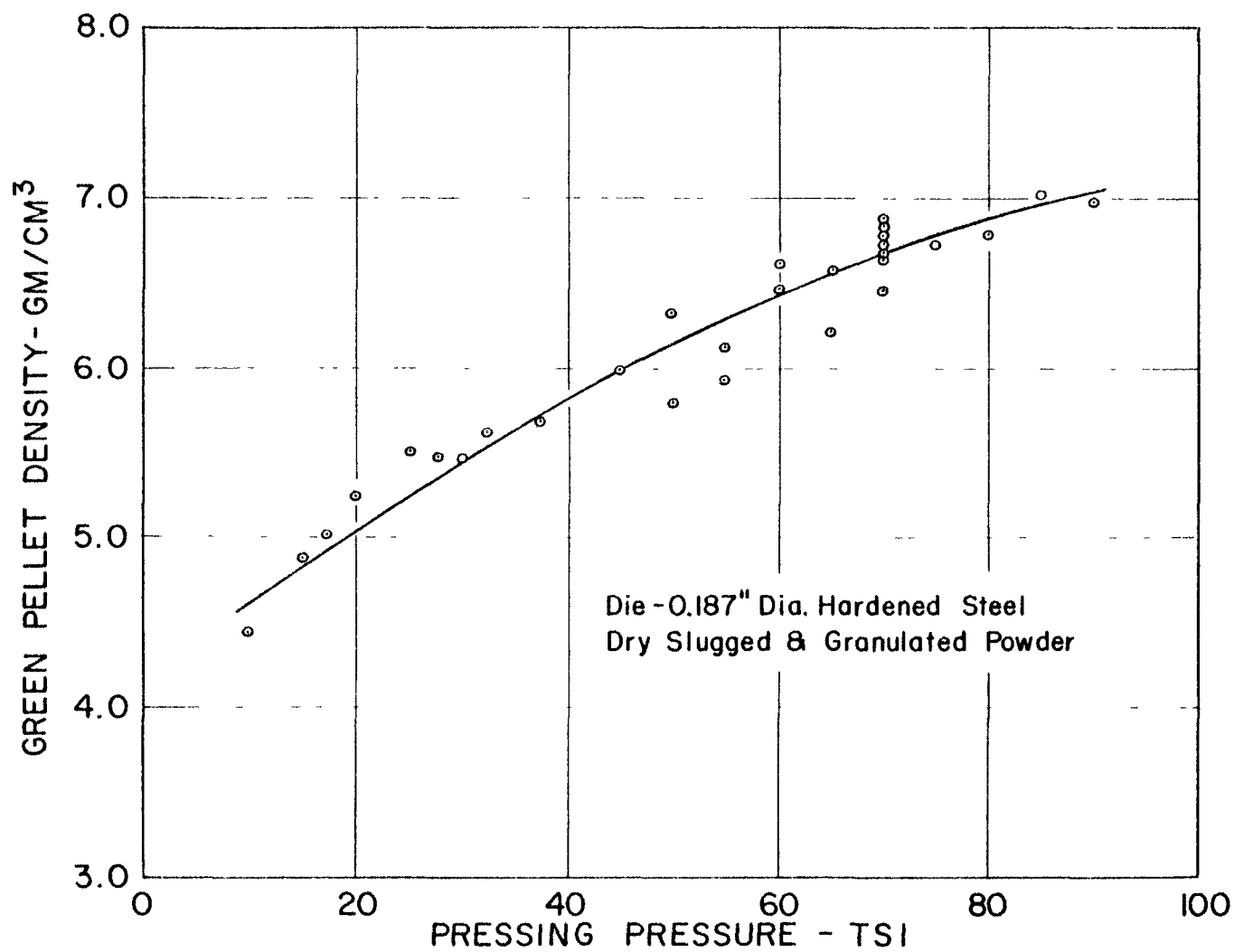


Figure 3.2
GREEN DENSITY OF DRY PRESSED PuO_2
MATERIAL : 297 - Pu - I

approximately 10°C per minute. The furnaces were operated under a non-explosive atmosphere of 5% hydrogen-95% nitrogen over all parts of the sintering runs. The results of these sintering runs are shown in Table 3.2, and the sintered density vs. green density curves are shown in Figure 3.3.

As can be seen in Figure 3.4, the sintered pellets exhibited some hour-glassing. This, coupled with the small size of the pellets, made dimensioning of the diameter to the accuracy demanded for density determinations impractical. Therefore, the density of all pellets was determined by the weight in air-weight in water method. However, the small size and weight of the pellets restricted the accuracy of the density determinations to the third significant figure with a standard deviation of $\pm 0.1 \text{ gm/cm}^3$. This accuracy is not sufficient to fully determine the sintering characteristics of PuO_2 . Future pellets trials will be with larger specimens in order to improve the accuracy of density measurements.

From Figure 3.3 it can be seen that the high surface area PuO_2 sinters to a high theoretical density even from relatively low green densities. It appears that relatively little change in sintered density results from increasing the green density above 5.6 gm/cm^3 for pellets sintered at 1600°C and above 6.0 gm/cm^3 for pellets sintered at 1400°C . Thus, high density PuO_2 can be obtained from dry pressed pellets pressed at pressures of approximately 50 tsi. The only apparent advantage of starting with higher green densities is a reduction in the shrinkage of the pellets. Due to the limitations on the accuracy of the density determinations made to date, a significant difference in sintering behavior at 1400 and 1600°C cannot be claimed. However, if any significant conclusions were to be drawn from these data, it would appear that higher sintered densities are obtained at the lower sintering temperature (1400°C). In this connection, it has been reported by Chikalla* that the density of PuO_2 appears to be completely independent of temperature in the range $1100\text{--}1600^{\circ}\text{C}$. In fact, his data for physical mixtures of $\text{PuO}_2\text{--UO}_2$ indicate that sintering at 1400°C results in higher densities for PuO_2 rich mixtures than sintering at either 1500 or 1600°C .

Future Work: The work during the next quarter will be directed toward establishing:

- 1) The effect of chemical processing variables on the sinterability of pure PuO_2 .
- 2) The effect of lower sintering temperatures on the sinterability of pure PuO_2 .

* T. D. Chikalla, "Sintering Studies on the System $\text{UO}_2\text{--PuO}_2$, Part II - Sinterability", HW-63081, December 17, 1959.

Table 3.2

Sintering Characteristics

Pellet No.	Green Density, gm/cm ³	Sintering Temperature, °C	Weight in Air, gm	Weight in Water, gm	Water Temperature, °C	Sintered Density, gm/cm ³	% of Theoretical Density
297-Pu-1-6	4.44	1400	0.7938	0.7204	26.2	10.8	94.2
297-Pu 1-9	5.01	1400	0.7904	0.7158	26.2	10.6	92.4
297-Pu-1-13	5.46	1400	0.8224	0.7455	26.2	10.7	93.3
297-Pu-1-15	5.68	1400	0.8146	0.7390	26.2	10.7	93.3
297-Pu-1-19	5.92	1400	0.6793	0.6173	26.7	10.9	95.0
297-Pu-1-22	6.46	1400	0.6841	0.6207	26.7	10.8	94.2
297-Pu-1-24	6.79	1400	0.7217	0.6546	27.0	10.7	93.3
297-Pu-1-26	6.96	1400	0.8476	0.7710	27.1	11.0	95.9
297-Pu-1-28	6.83	1400	0.9970	0.9064	27.1	11.0	95.9
297-Pu-1-30	6.76	1400	1.1599	1.0541	27.1	10.9	95.0
297-Pu-1-33	6.32	1400	0.7862	0.7138	27.2	10.8	94.2
297-Pu-1-37	5.50	1400	0.6628	0.6002	27.2	10.6	92.4
297-Pu-1-38	6.46	1400	0.7859	0.7144	27.2	11.0	95.9
297-Pu-1-40	6.44	1400	0.5857	0.5327	27.2	11.0	95.9
297-Pu-1-43	6.69	1400	0.5958	0.5417	27.2	11.0	95.9
297-Pu-1-45	6.89	1400	0.3117	0.2834	27.2	11.0	95.9
297-Pu-1-47	6.57	1400	0.6365	0.5786	27.2	11.0	95.9
297-Pu-1-5	4.43	1600	0.8142	0.7349	26.2	10.2	88.9
297-Pu-1-8	4.87	1600	0.8071	0.7302	26.2	10.5	91.5
297-Pu-1-12	5.48	1600	0.7575	0.6869	26.2	10.7	93.3
297-Pu-1-14	5.62	1600	0.7872	0.7135	26.2	10.7	93.3
297-Pu-1-18	5.79	1600	0.6707	0.6083	26.2	10.7	93.3
297-Pu-1-21	6.21	1600	0.7675	0.6962	26.7	10.7	93.3
297-Pu-1-23	6.73	1600	0.6106	0.5547	27.1	10.9	95.0
297-Pu-1-27	6.80	1600	0.7764	0.7047	27.1	10.8	94.2
297-Pu-1-29	6.46	1600	1.2276	1.131	27.1	10.7	93.3
297-Pu-1-31	6.64	1600	1.3192	1.1967	27.2	10.7	93.3
297-Pu-1-32	5.98	1600	0.8098	0.7340	27.2	10.7	93.3
297-Pu-1-34	6.12	1600	0.7683	0.6966	27.2	10.7	93.3
297-Pu-1-36	5.24	1600	0.7555	0.6833	27.2	10.4	90.7
297-Pu-1-39	7.01	1600	0.7123	0.6456	27.2	10.6	92.4
297-Pu-1-41	6.74	1600	0.5480	0.4971	27.2	10.7	93.3
297-Pu-1-44	6.75	1600	0.6227	0.5644	27.2	10.6	92.4

NUMEC P-60

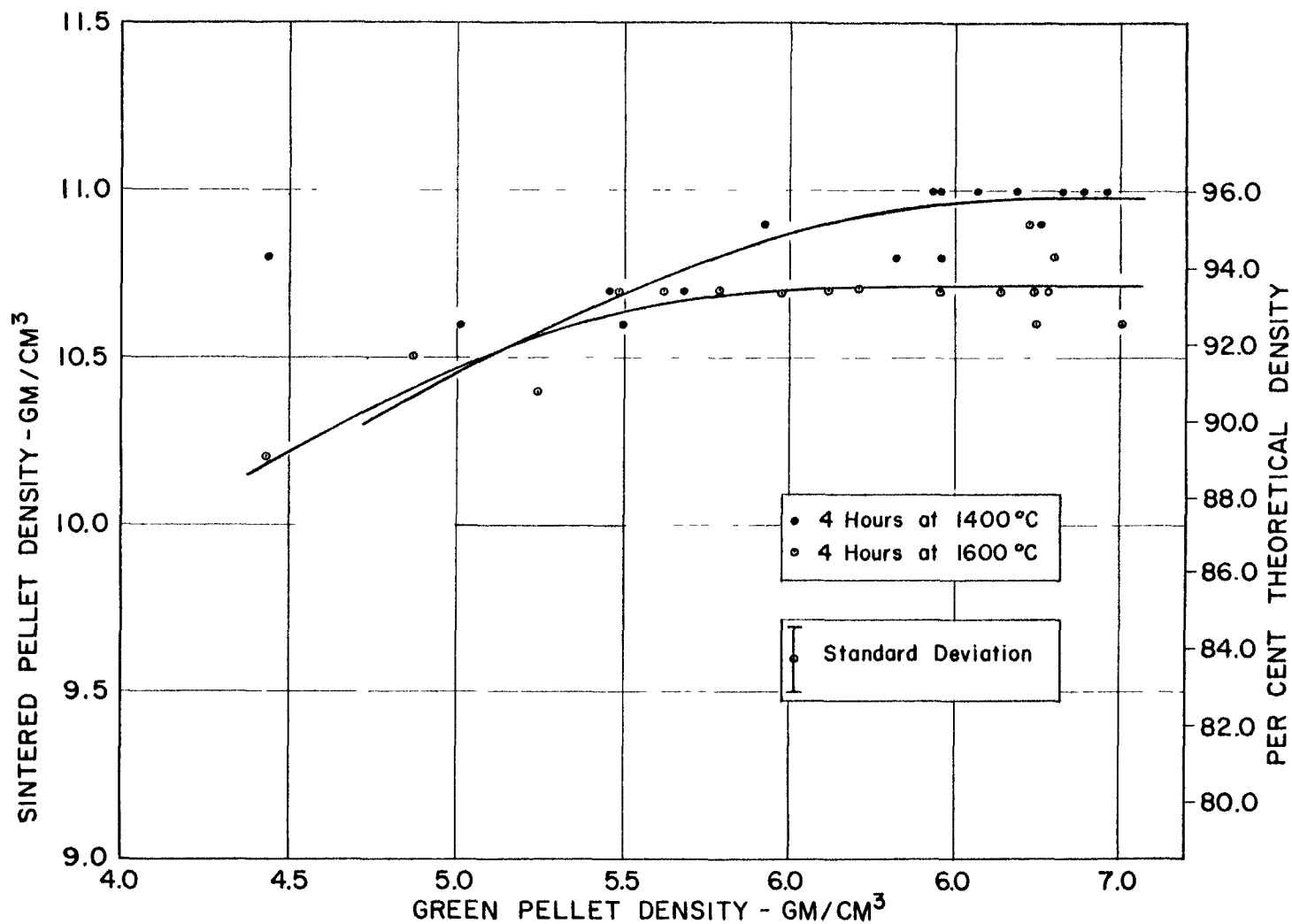


Figure 3.3
SINTERED DENSITY VS GREEN DENSITY
MATERIAL : 297-Pu-1

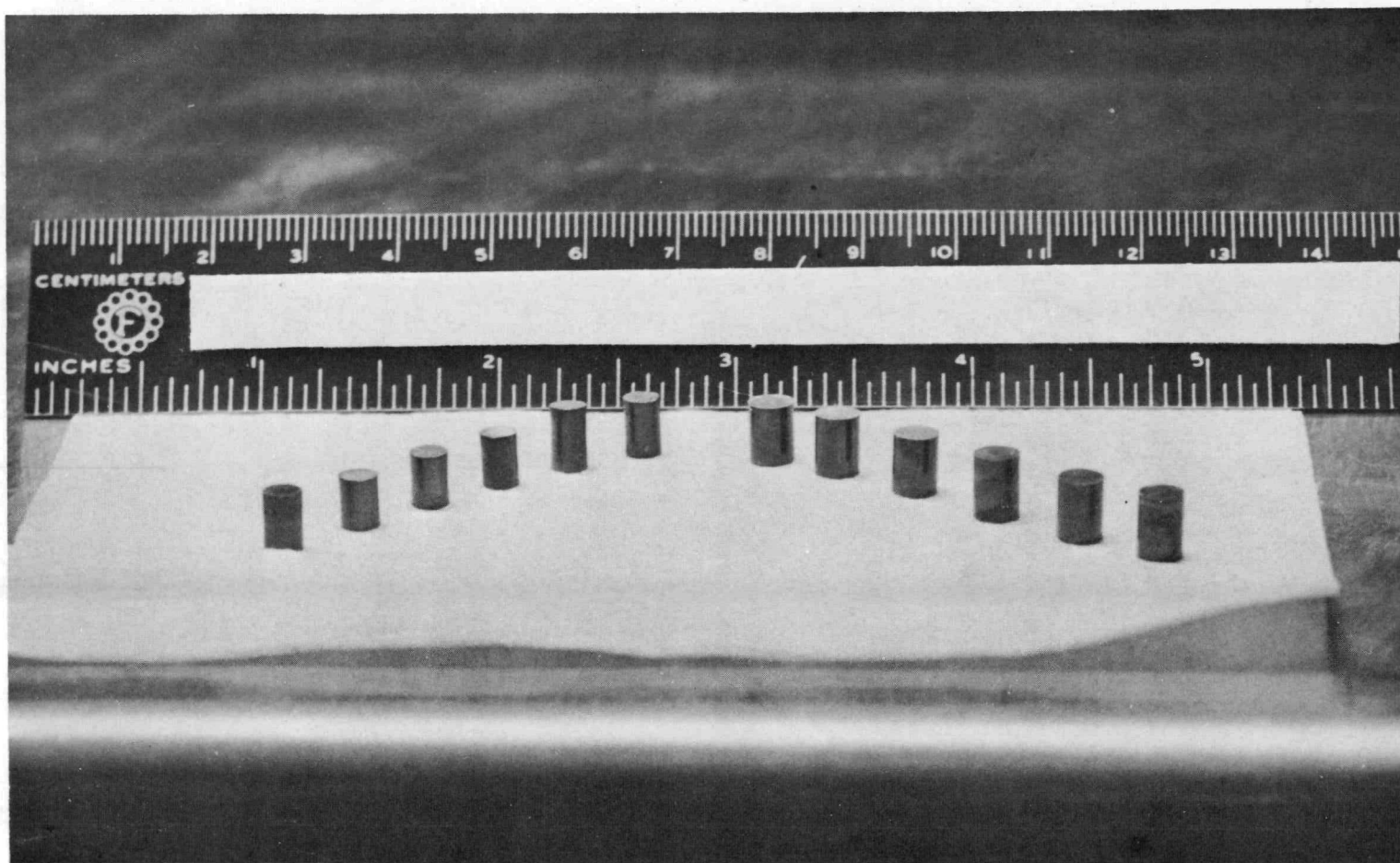


Figure 3.4

Cold Pressed and Sintered PuO_2 Pellets

Approximately 1.3 X

- 3) The pressing and sintering characteristics of co-precipitated $\text{PuO}_2\text{-UO}_2$.
- 4) Procedures for best accomplishing solid solution of PuO_2 and UO_2 mechanical mixtures.

In addition, pellets will be prepared and characterized for insertion in fuel elements for irradiation samples.

Homogenization of $\text{UO}_2\text{-PuO}_2$ Mixtures on Sintering

A research program to establish the degree of solid solution formation and atomistic homogenization in $\text{UO}_2\text{-PuO}_2$ mixtures on sintering as affected by the sintering temperature, sintering time, and history and character of the feed materials has been initiated. A search of the literature for information available on this subject has been conducted and the results incorporated into the research program. The initial phase of the program will be devoted to determining the amount of solid solution formation achieved in co-precipitated plutonium hydroxide-ammonium diuranate and in the $\text{UO}_2\text{-PuO}_2$ following calcining and reduction of the aforementioned feed materials. It is expected that subsequent phases of the program will encompass complete homogenization of co-precipitated materials by sintering, homogenization of mechanical mixtures of UO_2 and PuO_2 as affected by the individual component characteristics, and homogenization of mechanical mixtures of UO_2 and $\text{UO}_2\text{-PuO}_2$ solid solutions. The latter two phases have special significance in the study of an economical preparative route for solid solutions of these two oxides. It is obvious that preparation of the UO_2 (which in most cases represents the major portion of the fuel material) outside of the glove boxes by conventional techniques and introduction into the glove box line at a late stage of the fuel material preparation represents a considerable savings in fuel material fabrication costs.

Among the tools to be used to evaluate the degree of solid solution formation and homogenization are x-ray diffraction and differential thermal analysis. Accordingly, installation and calibration of these experimental facilities has been started. The existing x-ray diffraction equipment for powder samples has been redesigned and reconstructed to permit glancing angle photographs to be taken on solid sintered samples of $\text{UO}_2\text{-PuO}_2$. The supporting framework for the nickel window which serves simultaneously as a filter and primary alpha barrier was redesigned to provide greater safety in use. The optimum glancing angle for UO_2 was found to be between 25 and 35°. Work has been started on the operation of the differential thermal analysis equipment using 13% chromium stainless steel and 18/8 stainless steel as standards prior to work on oxides. This equipment will be committed to work on plutonium bearing materials during the next quarter.

Metallography

The three metallography glove boxes were completed, leak tested, and accepted for plutonium insertion during the latter portion of this quarter. On the basis of safety and operational reviews, a number of additions and modifications were made, including additional electrical outlets, electrical quick-disconnects on all polishers, safety interlock switches on all rotating machinery, and small sample cleaning systems. The cathodic vacuum etcher was completed, tested, and modified slightly; the cathode was replaced with one of newer design.

While it is expected that acid etching of plutonia bearing materials will be held to a minimum, equipment to accomplish this type of sample preparation has been installed in the box line. To allow such operation in safe fashion and to protect other polishing equipment from acid fumes, a special etching box was designed to fit within one of the metallography glove boxes. Construction and installation of this fume box was completed early this quarter. The acid fume exhaust and scrubber system previously described in NUMEC P-21 (p. 3) was also constructed to operate in conjunction with the etching glove box. The entire system has been found to operate successfully with boiling nitric acid in the etching box.

All metallographic boxes have now been subjected to the negative pressure leak test and the leakage rate found to be within the allowable limits. Metallographic operations on plutonium bearing materials, accordingly, will be initiated during the coming quarter.

Fuel Particle Preparation and Mechanical Packing

A technical report entitled "Mechanical Packing of Spherical Particles", which covers the research conducted to date with spherical metal shot packed by mechanical vibration in relatively small transparent containers, has been accepted for publication in the Journal of the American Ceramic Society. Continuation of the packing experiments, using aggregates produced by crushing high density UO_2 pellets has been deferred due to assignment of the available manpower to completion and commitment of the higher priority glove-boxes to plutonium processing. The packing experiments will be resumed during the next quarter.

Preparation of spherical particles for mechanical packing feed materials has been initiated using plasma torch equipment. The plasma torch, powder feed mechanism, gas circulation system, and a collection chamber have been installed in a glove box. Preliminary spheroidization runs, using stabilized zirconia, have been completed. A Thermal Dynamics low gas velocity, high enthalpy #5A, 5/16 inch electrode-nozzle assembly was used for all runs. Helium has been used exclusively thus far; however, it is intended to use helium-argon mixtures, straight argon, and nitrogen-hydrogen mixtures in near-future tests. If sufficient heat energy can be obtained in the plasma

to melt $\text{UO}_2\text{-PuO}_2$ with nitrogen-hydrogen mixtures, a very inexpensive once-through gas cycle system can be used instead of the presently planned closed cycle helium system.

The work with zirconia consisted of separating agglomerated high-fired powder into several size fractions and melting these powders under various equipment operating parameters to determine optimum conditions. The following sizes of stabilized zirconia were used:

- 325 mesh : Metco 201 Spray Zirconia
- 80 + 120 mesh : from Zircoa (Solon, Ohio)
- 60 + 80 mesh : from Zircoa
- 45 + 60 mesh : from Zircoa

A considerable amount of "blow", smoke cloud formation, and deposition on the collection chamber walls were experienced with the -325 mesh powder. Operation with the -80+120 mesh powder was ideal. It appeared to feed into the plasma almost completely, and the melted particles formed a shallow incandescent cone for about two feed from the nozzle at which point they lost their brightness. Over 99 per cent of the particles were as spherical as those shown in Figure 3.5. The surfaces of the spheres were extremely smooth and essentially free of flaws. No difficulty was encountered with spheres sticking together. All zirconia stock material is reported (manufacturer's literature) to have 5 to 10% porosity; it was found that most particles had voids at their centers. Apparently, during solidification of the molten spheres from the outside surface, the gas or porosity is accumulated at their centers. This implies the necessity for using high density feed materials if a high density product is required.

The -45+60 fraction required full power (500 amps) for melting, and a few per cent of non-spherical particles was obtained. However, experience has shown that the powder feed rate is very critical as the particles become larger and heavier, and it is believed that powder feed conditions for this size fraction were not optimum. With such material, major dependence must be on gravity feed with an assist from the carrier gas stream, whereas the finer powders can be fed by the carrier gas stream alone. Tests are planned on the feeding of UO_2 in order to simulate the conditions to be expected for $\text{UO}_2\text{-PuO}_2$ powders.

The zirconia particles ranged in color from their original yellow through various shades of gray to black. The smallest particles which appeared to have reached the highest temperature were the darkest in color. Since the particles were melted in a helium atmosphere contaminated by a small amount of air, it is unlikely that foreign material contamination could account for the color change. X-ray diffraction of the melted material shows two cubic lattices having slightly different lattice parameters while the unmelted material shows only one. Spectrographic analysis will be performed to determine if contamination is occurring. This effect ~~will~~ be examined in greater detail during the next quarter.

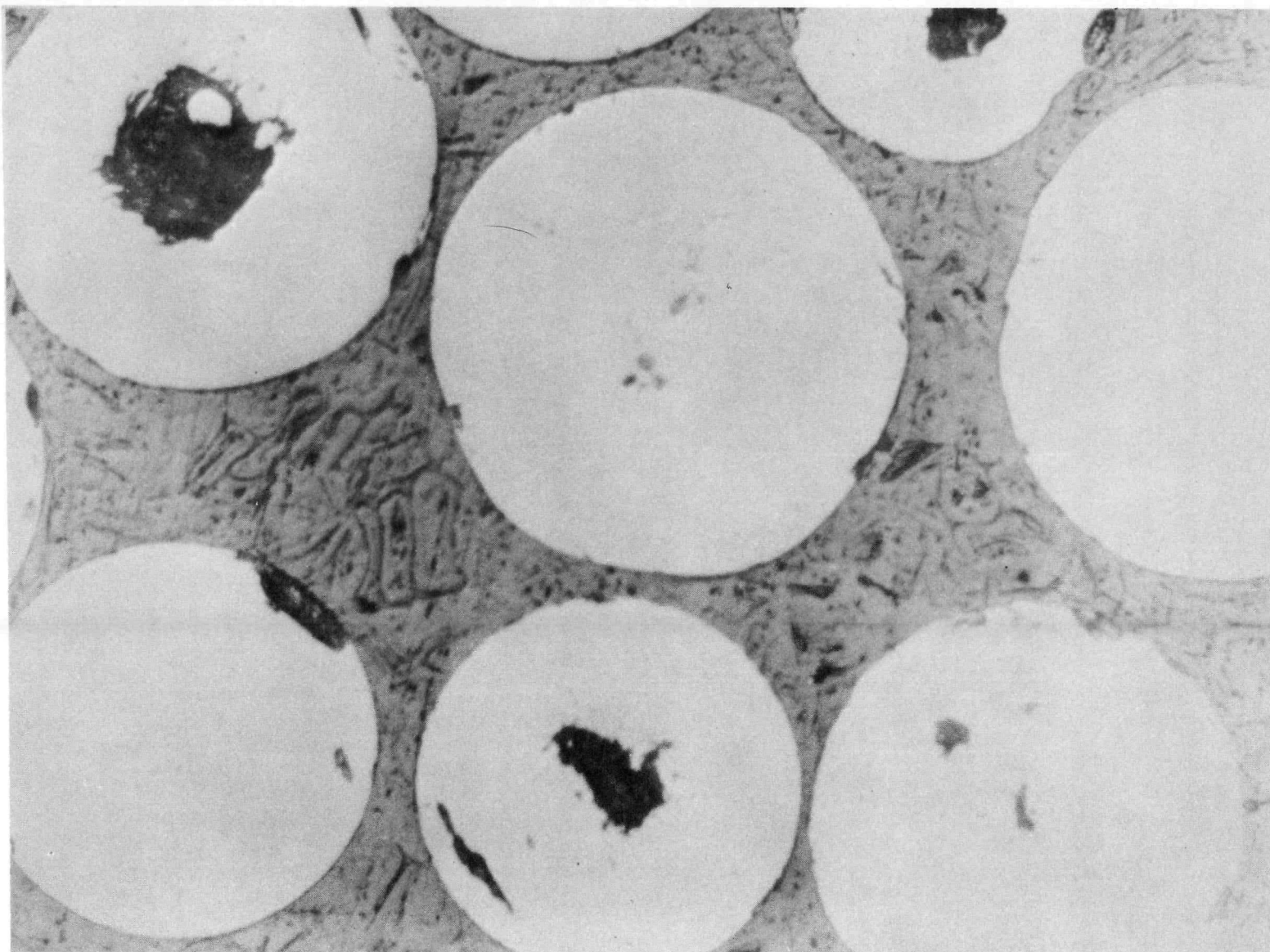


Figure 3.5

-80 +120 Mesh Stabilized ZrO_2 Spheroidized in Plasma Jet

500 X

Thermal Conductivity Measurements

The code for the LGP-30 computer for obtaining the thermal conductivity from experimental data (as described previously in NUMEC P-50, p. 31) has been completed, test problems run, and placed in production. The test equipment for performing the experiment has been designed, and complete working drawings prepared. Bids for constructing the main hardware items have been solicited.

In order to avoid the necessity of disassembling the apparatus to change samples, an air lock has been incorporated into the equipment. The sample is placed in a shouldered hole in a rod which moves horizontally through a set of four "O"-rings so as to prevent the loss of vacuum during insertion. After the horizontal rod is stopped over the axis of the susceptor, a rod is passed vertically through the susceptor, the sample picked up and lowered through the susceptor where it is retained on an internal ring inside the susceptor. In order to reduce the heat transfer out the ends of the susceptor, a tantalum cap is picked up at the same time as the sample and retained on the top of the susceptor as the vertical rod is lowered. A lower cap is also placed on the bottom of the susceptor by engaging the cap in slots in the susceptor support legs by means of the vertical rod. Details of the apparatus are discussed in a technical report which is under preparation and which is scheduled for publication during the next quarter.

Bids have also been solicited, orders placed, and items received for numerous components of the apparatus, e.g., vacuum pump, diffusion pump, vacuum gages, electron guns, etc. Delivery of the constant power, variable voltage supply is expected late in June. The major components for the vacuum system (fore pump line, diffusion pump, cold trap and main vacuum valve) have been assembled and helium leak-tested.

FUEL REPROCESSING

Task 6.00
C. S. Caldwell

R. J. Atkins H. Krake

Installation of low gamma level solvent extraction ion-exchange and related equipment is in progress within a four compartment glove-box unit attached to the transfer tunnel; two of the cubicles are double-layered to accommodate the taller equipment items as ion-exchange columns and feed tanks. This equipment will undergo check runs during the next eight weeks and should be in service by the end of the next quarter.

A preliminary layout of containment box equipment for fuel reprocessing studies has been established as shown in Figure 6.1. This hot cell alpha-box unit will contain an integrated system for fuel sample dissolution, feed preparation, fission product separation, sampling, and auxiliary handling of hot waste and dissolver off-gas. Separation runs using either solvent extraction or anion exchange will be made on a small scale basis as required to establish engineering data suitable for scaleup. Access during hot operation will be via ANL Model 8 manipulators which penetrate the horizontal top panel through a flexible boot section. Access following preliminary decontamination will be through glove ports at eleven locations.

Procurement of high-level solvent-extraction equipment is expected to be completed prior to the end of the next quarter. Assembly of components and cold alpha-box checkout runs are scheduled immediately following completion of the glove-box line reprocessing setup. Prior to the receipt of irradiated $\text{PuO}_2\text{-UO}_2$ fuel samples, several alpha-box "hot" runs will be made with spike amounts of mixed fission products.

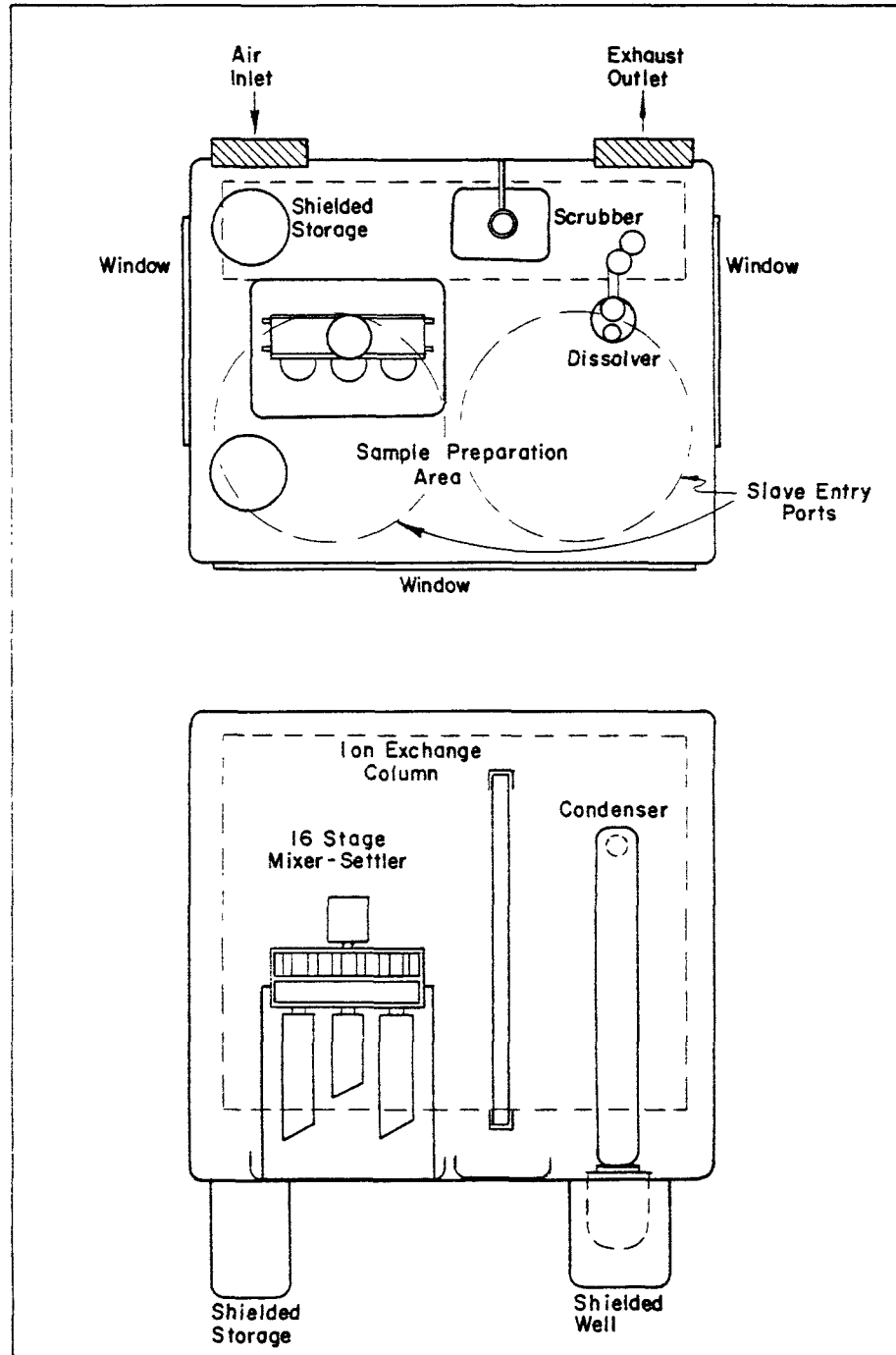


Figure 6.1
ALPHA-BOX EQUIPMENT ARRANGEMENT FOR
IRRADIATED FUEL DISSOLUTION & DECONTAMINATION

REACTOR PHYSICS AND ENGINEERING PARAMETRIC STUDIES

Task 8.0
K. H. Puechl

J. Ruzbacki

The analysis covering the development of the NUMEC Survey Procedure (NUSURP) was published in the February 1961 issue of Nuclear Science and Engineering [2, 241-259 (1961)] under the title "An Approach to Reactor Physics Using Results of Integral Experiments". Since then, further work has raised some doubt concerning the accuracy of the experimental U-238 cadmium ratio data utilized in this development. Specifically, the use of standard calculational techniques to determine this cadmium ratio, does not lead to the spread between metal and oxide points when the epithermal index r_{28} is plotted against the absorption parameter $\frac{\sum_a}{\sum_s}$. Instead, r_{28} for both metal and oxide is given by the simple relationship

$$r_{28} = \left[1.0635 - 0.0245 S/V \right] \frac{\overline{\sum_a}}{\sum_s} \quad (1)$$

High accuracy cadmium ratio measurements and further detailed analysis of critical experiment results are required to resolve this discrepancy between experiment and theory. In the interim, it has been decided to utilize the revised correlation given by Equation (1). Utilization of this correlation will generally be conservative, leading to lower values for k_{∞}/ϵ , and hence lower estimates for core life.

This revised NUSURP procedure has been programmed for the LGP-30 computer. Fuel burnout and variation of isotopic composition with time is being computed by using Gill's modification of the fourth-order Runge-Kutta method for solving the differential equations involved.

This procedure is being utilized to ascertain the potential of plutonium as a fuel in near-thermal reactors. Initially, pressurized water systems are being investigated. The range of lattice parameters and other input data for these initial investigations are given in Table 8.1. Preliminary results indicate that extremely long core lives are attainable with this composition of plutonium and that the reactivity variation during core life is nominal. Detailed analysis shows that this behavior is due primarily to the fact that Pu-240 acts naturally as a combination source material and burnable poison. The monotonic reduction in neutron temperature associated with burnout tends to monotonically reduce the effective cross section of Pu-240; thereby the reactivity tends to rise with burnout. Of course, this tendency is counterbalanced by the grow-in

Table 8.1

Input Data for Primary Calculation Program
Potential of Plutonium in Near-Thermal Reactors

Data Applicable for All Cases:

Average fuel temperature, T_f , $^{\circ}\text{K}$	1589
Average moderator temperature, T_m , $^{\circ}\text{K}$	560
Heavy atom number density in fuel, $\sum_h \text{Ni}$, (atom/cc) $\times 10^{24}$	0.0225

Lattice Parameters:

Surface to Volume Ratio, S/V	0.025 ev Absorption Cross Section in Cladding $(\sum a)_{\text{clad}}/\sum_h \text{Ni}$	Degree of Moderation $\xi \sum_s / \sum_h \text{Ni}$	0.025 ev Absorption Cross Section in Moderator $(\sum a)_{\text{mod}}/\sum_h \text{Ni}$
3.22	1.80	69.1	1.30
		100	1.88
		125	2.35
		150	2.84
		175	3.31
5.36	3.00	69.1	1.30
		100	1.88
		125	2.35
		150	2.84
		175	3.31
7.00	3.92	69.1	1.30
		100	1.88
		125	2.35
		150	2.84
		175	3.31

Initial Pu Isotopic Composition:

<u>Isotope</u>	<u>a/o</u>
Pu-239	78
Pu-240	12
Pu-241	10
Pu-242	0

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of fission products; therefore, the reactivity tends to remain, more or less, constant over a long period of time. Upon completion of this study, some highly-encircled burner systems will be investigated.