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# *Preparation of $UO_2$ and $^{238}PuO_2$ Fuel Pellets for a Small Heat Source*

*R. L. Deaton, C. J. Wiedenheft*

*and R. E. Zielinski*

June 14, 1974

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## NOTICE



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ABSTRACT

Pressed and sintered $PuO_2$ fuel pellets were developed as an alternate fuel form for a small radioisotopic heat source. In initial development $UO_2$ pellets were prepared prior to extending the study to $^{238}PuO_2$.

As a result of this study, ideal pellet dimensions, shape, and pressing parameters were determined; 27 $UO_2$ and 18 $PuO_2$ pellets were fabricated.

## INTRODUCTION

In the development phase of the $^{238}PuO_2$ fuel for a small [6 W (th)] radioisotopic heat source for a thermoelectric generator, two fuel forms were considered: 1) a packed bed of $PuO_2$ shards (53-500 μm) and 2) a compacted pellet (small cylinder). Although shards were ultimately selected as the fuel form for this program, the pelletized fuel was developed as described in this report.

In addition to the $PuO_2$ fuel, this particular heat source capsule will contain sufficient yttrium metal to maintain a reduced partial pressure of oxygen. Lowered oxygen partial pressure is required to prevent embrittlement of the refractory metal capsule during the useful life of the generator. The heat source design using a $PuO_2$ pellet is shown in Figure 1.

Two of the major problems anticipated with the pelletized fuel form were:

1. Production of pellets with reproducible dimensions, wattage, and density to the tolerance required.

2. Production of pellets without sharp corners which could rupture the heat source capsule upon high velocity impacts.

In the development of the pelletized fuel form it was first necessary to develop methods for producing the pellets with the $UO_2$ simulant (nonradioactive) fuel. Secondly, the methods and techniques established in the $UO_2$ simulant study were to be used to develop a method for producing $^{238}PuO_2$ fuel pellets of specified dimension and wattage. In addition, it was necessary to produce pellets for testing the characteristics of the radioisotopic heat source.

## SIMULANT STUDY

### $UO_2$ FEED MATERIAL

The $UO_2$ feed material used in this study was commercially produced and had been in stock at Mound Laboratory for several years. (The method

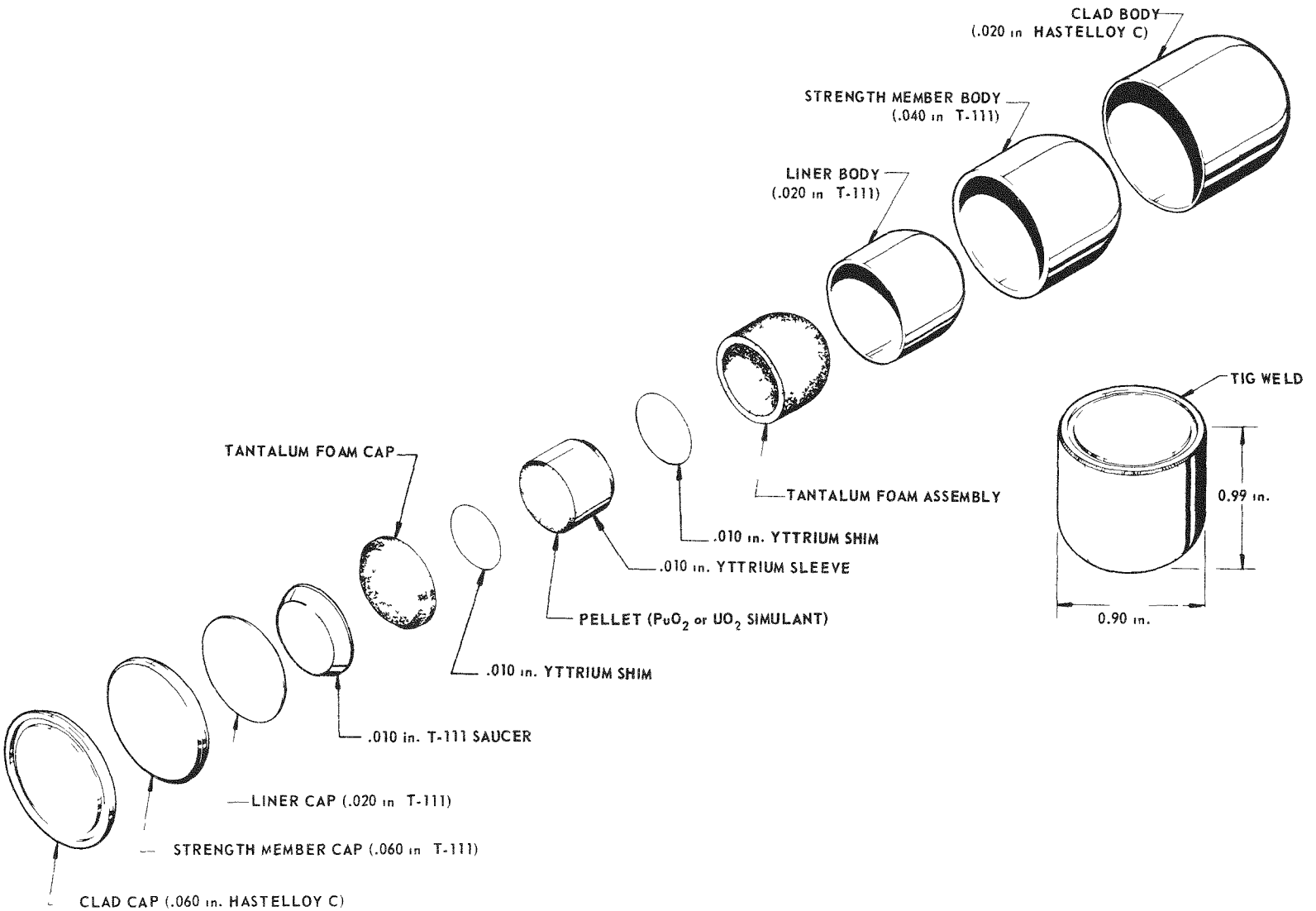


FIGURE 1 - Pellet fuel form capsule.

of preparation and thermal history of this material is unknown.) This $UO_2$ material consisted of very fine particles which had agglomerated into larger spheroidal particles as shown in Figure 2. The properties of this material are summarized in Tables 1 and 2.

## PRELIMINARY FABRICATION STUDIES

Several right-circular-cylinder test pellets were prepared to evaluate the pressing and sintering characteristics of the feed material prior to final die design.

Several pellets were compacted at various pressures. It was found that at pressures as low as 12,000 psi, the $UO_2$ pellets had good green strength and apparent densities of about 57 to 58% of theoretical. Very little increase in green density or green strength occurred as the forming pressure was increased to 30,000 psi. As the forming pressure was increased above ~ 40,000 psi, the pellets tended to laminate and lose their green strength.

Colloidal graphite was used throughout this study as a die lubricant. Water and Carbowax were evaluated as binders. Both binders improved the green strength of the pellets, and did not seem to affect the green density.

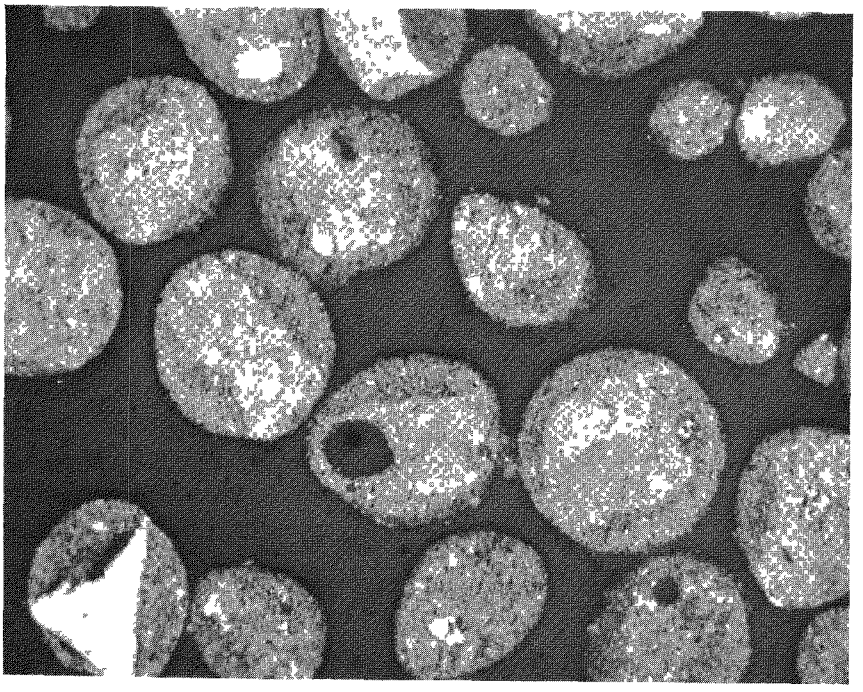

FIGURE 2 - $UO_2$ feed material (100X).

Table 1

PROPERTIES OF $UO_2$ FEED MATERIAL

| | |
|---|---|
| Agglomerated Particle Size | 50-150 μm |
| Apparent Density | 5.8 g/cc (52.9% TD) |
| Tap Density | 4.1 g/cc (37.4% TD) |
| Stoichiometry | $UO_{2.02}$ |

Table 2

IMPURITY AND ISOTOPIC ANALYSIS OF $UO_2$ FEED MATERIAL

| Element | Amount (μg/g) | Isotope | Amount (at. %) |
|---|---|---|---|
| Fe | 450 | U-235 | 0.23 |
| Si | 100 | U-236 | 0.003 |
| Mn | 12 | U-238 | 99.8 |
| B | <2 | | |
| Pb | <10 | | |
| Mg | 10 | | |
| Cr | 30 | | |
| Al | 100 | | |
| Ca | <250 | | |
| Cu | 20 | | |
| Co | 25 | | |
| Ni | 70 | | |
| C | 28 | | |

After being sintered at 1400°C under vacuum for 1 hr, the $UO_2$ pellets exhibited densities of 83 to 86% of theoretical. The pellets were sintered under vacuum to prevent oxidation. Sintering at temperatures higher than 1400°C or for longer than 1 hr resulted in only slightly higher densities.

Pellet measurements before and after sintering indicated that the diameter of the pellet shrank slightly more than did the length.

Properties of the preliminary pellets prepared for this study are listed in Table 3.

Table 3

$UO_2$-CHAMFERED RIGHT CIRCULAR PELLETS

| Pellet | Charge (g $UO_2$) | Green Density (%) | Sintered Density (%) | Shrinkage (%) | Binder | Pressing Pressure (psi) |
|---|---|---|---|---|---|---|
| 1 | 14.1 | 58.8 | 83.5 | Diam. 11.7<br>Hgt. 10.5 | Carbowax | 18,000 |
| 2 | 14.1 | 57.6 | 83.3 | Diam. 12.2<br>Hgt. 11.0 | Carbowax | 15,000 |
| 1A | 14.0 | 57.8 | 85.6 | Diam. 12.8 | $H_2O$ | 15,000 |
| 2A | 14.0 | 57.4 | 85.1 | Diam. 12.8<br>Hgt. 11.9 | $H_2O$ | 17,000 |

FEED MATERIAL AND PELLET OUTGASSING ANALYSIS

Because graphite was used as a die lubricant and water or Carbowax was used as a binder, the vacuum outgassing properties of the feed material and pellets were measured to ensure that outgassed species would not be in sufficient concentration to cause embrittlement of the capsule material. Untreated $UO_2$ feed material and compacted pellets containing both Carbowax and water binder were heated under vacuum, and the gasses evolved at various temperatures were analyzed with a Quadrupole Mass Spectrometer. During this analysis, the presence of $CH_4$, $CO_2$, CO, and $H_2O$ were detected during heating. However, an insignificant amount of gas evolved from both feed material and compacted pellets after a 1-hr sintering at 1400°C. Any carbon in the feed material and compacted pellets after sintering was less than the detectable limit. These analyses indicate that there should not be any deleterious effects on the liner material from evolved gasses or carbon impurity after the 1400°C sintering.

## PREPARATION OF $UO_2$ SIMULANT PELLETS

From the information obtained in the preliminary studies, a die-punch set was designed to prepare pellets with chamfered edges as shown in Figures 3 and 4. Chamfered pellet edges were designed to reduce the possibility that the fuel pellets would rupture the heat source capsule during high velocity impact. Pellet dimensions were established to permit the fuel pellet to be completely surrounded with a collapsible tantalum foam material. This foam holds the pellet immobile within the capsule and helps to absorb the shock that occurs during impact. The free volume within the foam permits the safe storage of helium generated by the alpha decay of $^{238}PuO_2$ during the lifetime of the heat source.

A few test pellets were prepared for destructive analysis using the fabrication parameters listed in Table 3. Two of these chamfered pellets are shown in Figure 5. A photomicrograph of a pellet cross section shown in Figure 6 indicates that the density is uniform throughout the pellet. Pellet impurities are listed in Table 4.

The 27 $UO_2$ simulant pellets shown in Figure 7 were prepared for use in the generator testing program. The properties of these pellets are listed in Table 5.

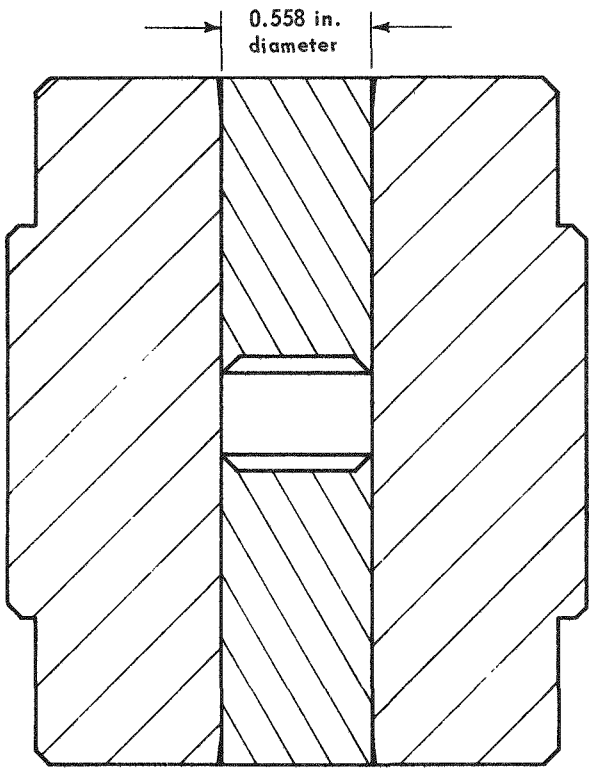


FIGURE 3 - Double chamfered die.

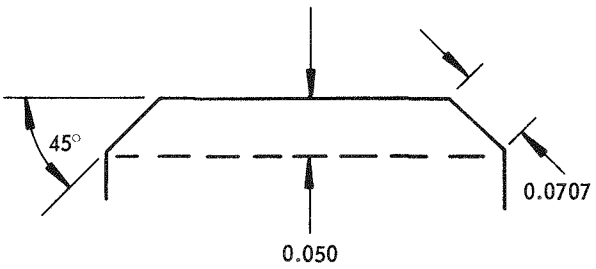


FIGURE 4 - Description of chamfered top.

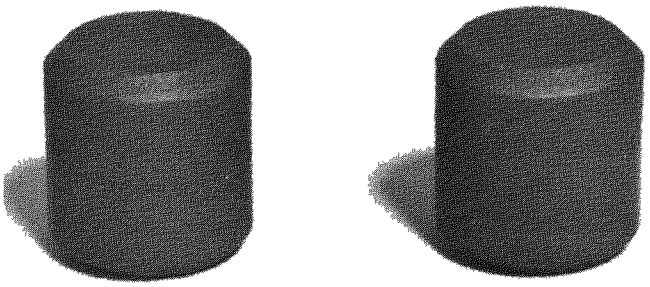

FIGURE 5 - Typical cold-pressed and sintered $UO_2$ pellets (double chamfer configuration).

Table 4

IMPURITY ANALYSIS OF SELECTED $UO_2$ SIMULATED FUEL PELLET

| Element | Amount (µg/g) in Exterior | Amount (µg/g) in Interior |
|---|---|---|
| C | 26 | 32 |
| Fe | 450 | 450 |
| Si | 100 | 100 |
| Mn | 12 | 12 |
| Pb | <10 | <10 |
| Mg | 20 | 20 |
| Cr | 30 | 30 |
| Al | 150 | 150 |
| Ca | 200 | 200 |
| Cu | 10 | 10 |
| CO | 20 | 20 |
| Ni | 90 | 90 |

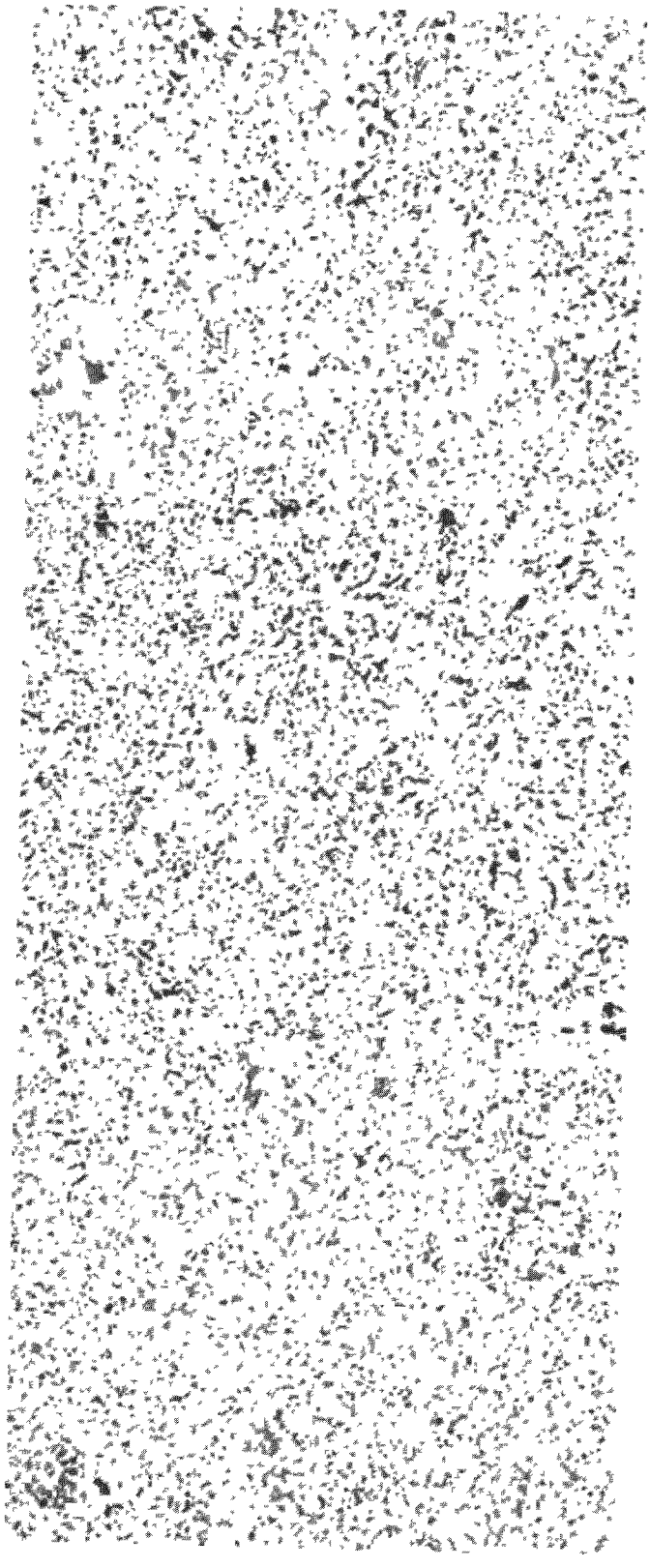

FIGURE 6 - Composite photomicrograph of sintered $UO_2$ pellet; 500X magnification (reduced to 85%).

Table 5

$UO_2$ PELLET DATA

| Charge Weight (g) | Green Dimensions (in.) | Green Density (%) | Sintered Dimensions (in.) | Sintered Density (%) | Shrinkage (%) | Sintered Weight (g) | Weight Loss (%) |
|---|---|---|---|---|---|---|---|
| 14.3784 | diam. 0.558<br>height 0.597 | 56.7 | diam. 0.491<br>height 0.533 | 81.6 | diam. 12.0<br>height 10.8 | 14.1526 | 1.6 |
| 14.3870 | diam. 0.558<br>height 0.599 | 56.6 | diam. 0.491<br>height 0.535 | 81.5 | diam. 12.0<br>height 10.7 | 14.1818 | 1 4 |
| 14.4547 | diam. 0.558<br>height 0.592 | 57.6 | diam. 0.495<br>height 0.531 | 81.9 | diam. 11.3<br>height 10.3 | 14.3862 | 0.5 |
| 14.4732 | diam. 0.558<br>height 0.591 | 57.8 | diam. 0.495<br>height 0.532 | 81.9 | diam. 11.3<br>height 10.0 | 14.4165 | 0.4 |
| 14.4659 | diam. 0.558<br>height 0.5905 | 57.8 | diam. 0.496<br>height 0.532 | 81.6 | diam. 11.1<br>height 9.9 | 14.4146 | 0.4 |
| 14.4667 | diam. 0.558<br>height 0.5885 | 58.0 | diam. 0.4945<br>height 0.529 | 82.2 | diam. 11.4<br>height 10.1 | 14.3403 | 0.9 |
| 14.4804 | diam. 0.558<br>height 0.591 | 57.8 | diam. 0.494<br>height 0.530 | 82.2 | diam. 11.5<br>height 10.3 | 14.3405 | 1.0 |
| 14.4731 | diam. 0.558<br>height 0.591 | 57.8 | diam. 0.494<br>height 0.531 | 81.9 | diam. 11.5<br>height 10.2 | 14.3248 | 1.1 |
| 14.4861 | diam. 0.558<br>height 0.593 | 57.6 | diam. 0.4915<br>height 0.529 | 83.6 | diam. 11.9<br>height 10.8 | 14.4050 | 0.6 |
| 14.4863 | diam. 0.558<br>height 0.592 | 57.7 | diam. 0.492<br>height 0.530 | 83.3 | diam. 11.8<br>height 10.5 | 14.4260 | 0.4 |
| 14.4849 | diam. 0.558<br>height 0.591 | 57.8 | diam. 0.491<br>height 0.528 | 83.9 | diam. 12.0<br>height 10.7 | 14.4108 | 0.5 |
| 14 4865 | diam. 0.558<br>height 0.593 | 57.6 | diam. 0.4915<br>height 0.527 | 83.9 | diam. 11.9<br>height 12.5 | 14.4016 | 0.6 |
| 14.4837 | diam. 0.558<br>height 0.591 | 57.8 | diam. 0.491<br>height 0.5275 | 83.2 | diam. 12.0<br>height 10.7 | 14.2650 | 1.5 |
| 14.4887 | diam. 0.558<br>height 0.593 | 57.6 | diam. 0.490<br>height 0.5285 | 83.3 | diam. 12.8<br>height 10.9 | 14.2591 | 1.6 |
| 14.4860 | diam. 0.558<br>height 0.593 | 57.5 | diam. 0.490<br>height 0.529 | 83.0 | diam. 12.8<br>height 10.8 | 14.2150 | 1.9 |
| 14.4940 | diam. 0.558<br>height 0.591 | 57.9 | diam. 0.494<br>height 0.530 | 82.7 | diam. 11.5<br>height 10.3 | 14.4352 | 0.4 |
| 14.4866 | diam. 0.558<br>height 0.595 | 57.4 | diam. 0.493<br>height 0.533 | 82.5 | diam. 11.6<br>height 10.4 | 14.4281 | 0.4 |
| 14.4882 | diam. 0.558<br>height 0.590 | 57.9 | diam. 0.494<br>height 0.531 | 82.5 | diam. 11.5<br>height 9.9 | 14.4311 | 0.4 |
| 14.4893 | diam. 0.558<br>height 0.593 | 57.6 | diam. 0.494<br>height 0.533 | 82.2 | diam. 11.5<br>height 10.1 | 14.4326 | 0.4 |
| 14.4862 | diam. 0.558<br>height 0.592 | 57.7 | diam. 0.4935<br>height 0.531 | 82.2 | diam. 11.6<br>height 10.3 | 14.3455 | 1.0 |
| 14.4886 | diam. 0.558<br>height 0.592 | 57.7 | diam. 0.493<br>height 0.531 | 82.4 | diam. 11.6<br>height 10.3 | 14.3482 | 1.0 |
| 14.4996 | diam. 0.558<br>height 0.593 | 57.7 | diam. 0.4925<br>height 0.532 | 82.3 | diam. 11.7<br>height 10.3 | 14.3480 | 1.0 |
| 14.4934 | diam. 0.558<br>height 0.591 | 57.9 | diam. 0.492<br>height 0.531 | 82.7 | diam. 11.8<br>height 10.2 | 14.3489 | 1.0 |
| 14.4935 | diam. 0.558<br>height 0.592 | 57.8 | diam. 0.491<br>height 0.530 | 82.8 | diam. 12.0<br>height 10.5 | 14.2798 | 1.5 |
| 14.4941 | diam. 0.558<br>height 0.592 | 57.8 | diam. 0.495<br>height 0.534 | 81.3 | diam. 11.3<br>height 9.8 | 14.3696 | 0.9 |
| 14.4943 | diam. 0.558<br>height 0.593 | 57.7 | diam. 0.492<br>height 0.531 | 82.6 | diam. 11.8<br>height 10.5 | 14.3350 | 1.1 |
| 14.4986 | diam. 0.558<br>height 0.5785 | 59.2 | diam. 0.498<br>height 0.523 | 82.6 | diam. 10.8<br>height 9.6 | 14.4473 | 0.4 |

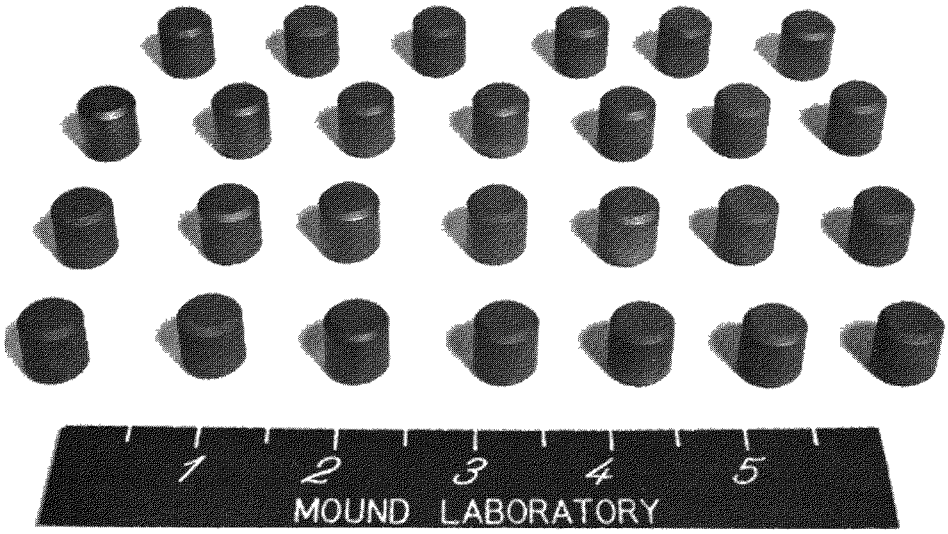


FIGURE 7 - Cold pressed and sintered $UO_2$ pellets for use as fuel simulant.

## PLUTONIUM-238 DIOXIDE STUDY

### FEED MATERIAL PREPARATION

Plutonia shards prepared by the hydroxide process (see Process Flow Sheet, Figure 8) and calcined at 650-700°C for 1 hr were used as feed material for the preparation of the $^{238}PuO_2$ pellets. This material consists of very small crystallites agglomerated into larger particles or shards. Figure 9 shows a typical size distribution of the <297 μm shards used. These particles exhibit low densities (~67% TD) and are quite friable. Pressing characteristics of the $PuO_2$ feed material seem to depend critically on calcination parameters, age, and method of storage. Specifications for $PuO_2$ fuel pellets are given in Table 6.

Table 6

SPECIFICATIONS FOR $PuO_2$ FUEL PELLETS[a]

| | |
|---|---|
| Diameter | 0.494 ± 0.005 in. |
| Height | 0.525 ± 0.015 in. |
| Density | 81 to 85% of theoretical |
| Wattage | 6.0 ± 0.05 W |

[a]Both ends chamfered.

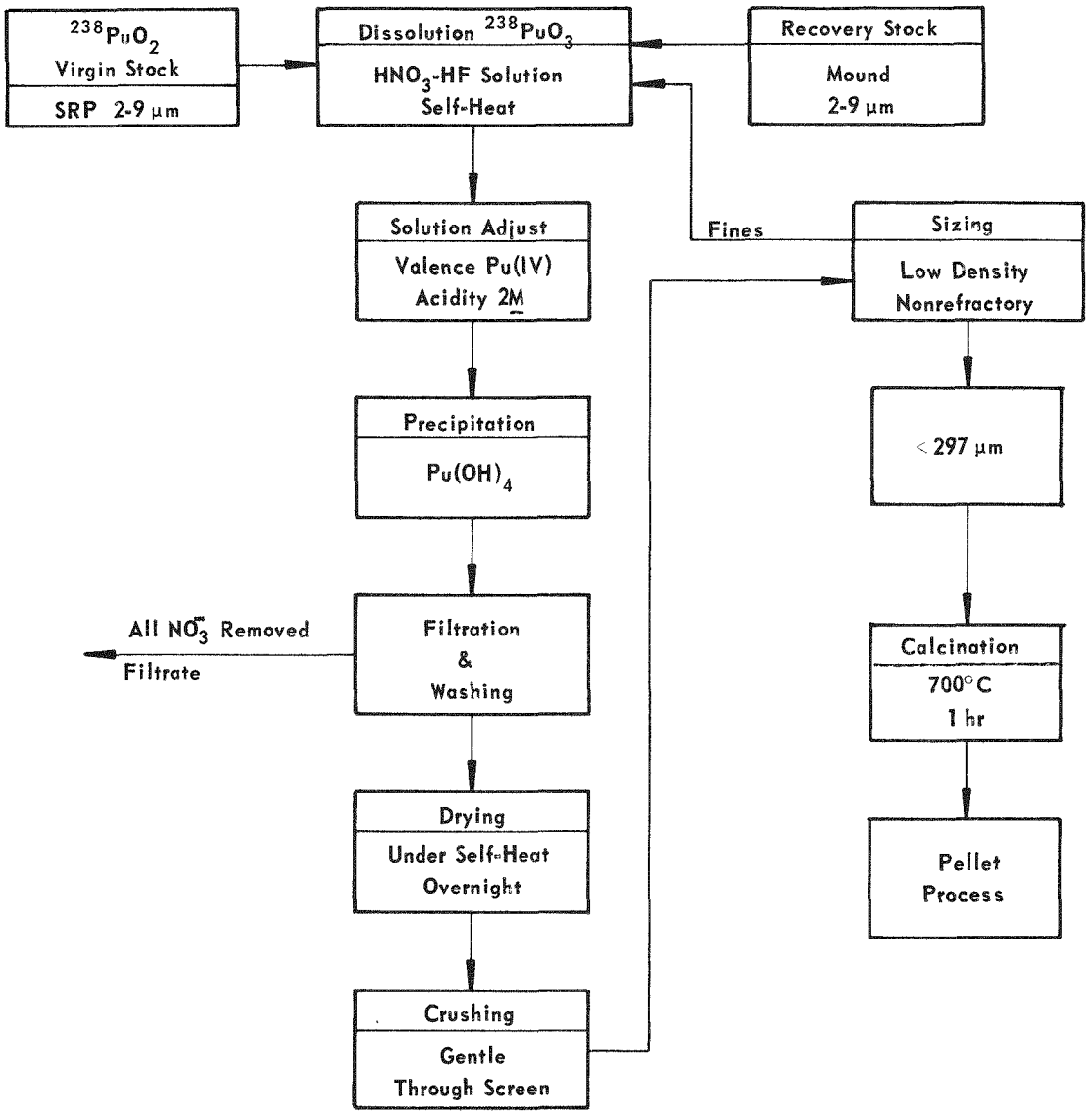


FIGURE 8 - Production process flowsheet for the "hydroxide" shard process.

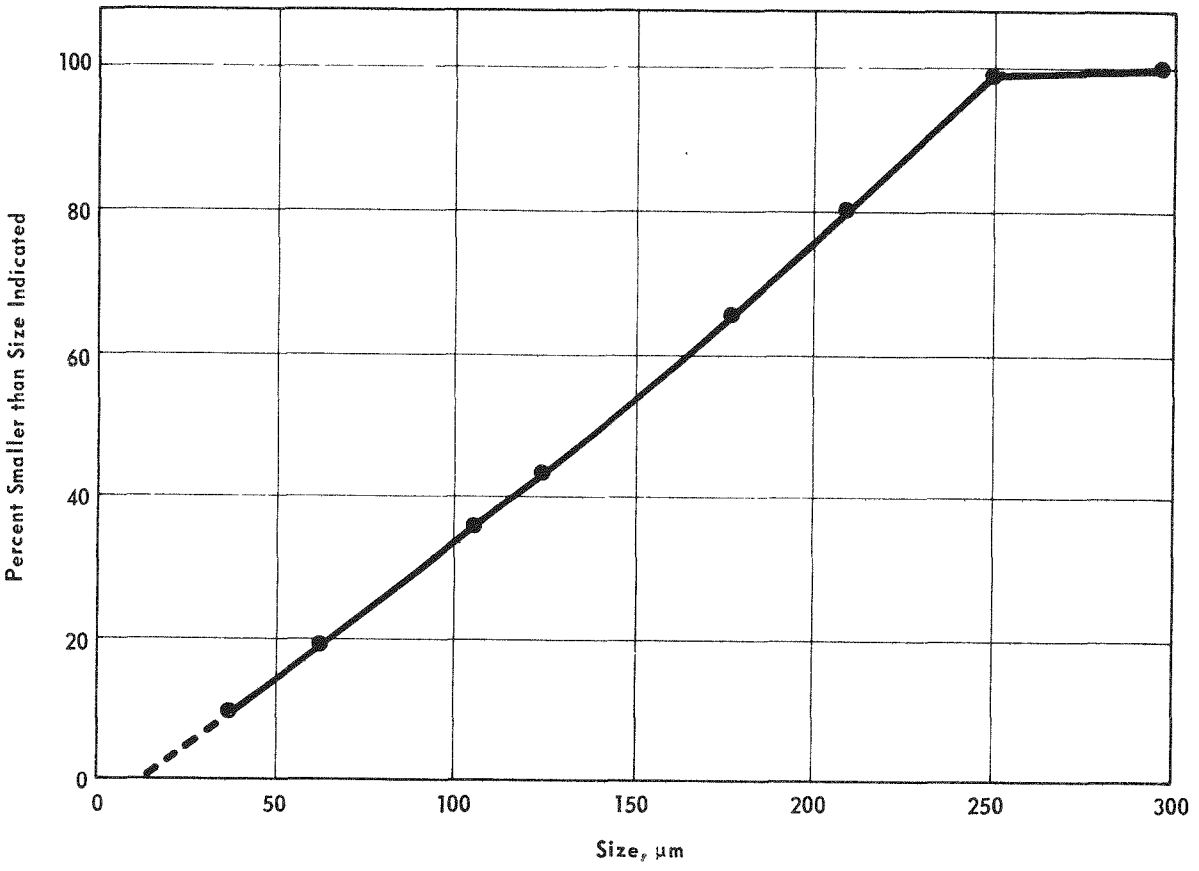


FIGURE 9 - Typical size distribution of <297 μm $^{238}Pu^{16}O_2$ shards produced by the hydroxide process.

The effect of the calcination parameters was evaluated by calcining portions of a single batch of shards at various temperatures and times. Test pellets were prepared from each of these portions as shown in Tables 7 and 8; the green density of the pellets increased as the duration and temperature of calcination are increased. Green pellets prepared from material calcined at or below 700°C for 1 hr sintered to densities of 81 to 85% of theoretical. Calcination at 750°C for 1 hr, however, resulted in pellets with densities less than 80% of theoretical. In order to obtain reproducibility of pellet dimensions while holding the die dimension constant, both the green and sintered density had to be relatively constant from batch to batch.

From Tables 7 and 8, it can be seen that the green density of a cold-pressed pellet increases as the time and temperature of calcining the feed material are increased. In order to reduce the sensitivity of the calcination parameters, the highest temperature (700°C) and the longest time (1 hr) that yielded an acceptable final density (after sintering) were selected as the calcination parameters. It was found also that care must be taken to ensure that the feed material is calcined homogeneously (even from self-heat) or the pressing and sintering properties

Table 7

EFFECT OF FUEL CALCINATION
TIME ON GREEN DENSITY OF PELLET

| Calcination Time at 700°C[a] (min) | Pellet Density (g/cc) |
|---|---|
| 0 | 50.9 |
| 1 | 54.3 |
| 10 | 55.5 |
| 15 | 56.3 |
| 60 | 60.4 |

[a] Previously calcined at 650°C for 1 hr.

Table 8

EFFECT OF FUEL CALCINATION
TEMPERATURE ON GREEN DENSITY OF PELLET

| One-hour Calcination Temperature[a] (°C) | Pellet Density (g/cc) |
|---|---|
| 650 | 50.9 |
| 675 | 57.4 |
| 700 | 57.9 |
| 750 | 61.4 |

[a] Previously uncalcined.

vary widely. In addition, if the material is stored in large quantities for several days after calcination, its cold pressing properties will deteriorate, apparently caused by self-heat (self-sintering). For example, a batch of feed material calcined at 700°C for 15 min yielded

green pellets of 58% and sintered densities of 83.2% of theoretical density. After two weeks of storage, the material produced pellets which exhibited a green density of 61.1% and a sintered density of less than 80% TD. Subsequent measurement of the apparent density of the feed material indicated that the density had increased from 67% to 73% TD during the storage period. Table 9 summarizes the feed material properties.

The effect of compaction pressures was investigated by preparing several pellets at pressures from 33,000 to 50,000 psi. At pressures below 33,000 psi, the green strength of the pellet was too low for normal handling. At forming pressures from 33,000 to 41,000 psi, the pellet green strength was adequate and the green density varied from 57% to 59% TD. As the pressure was increased to about 50,000 psi and above, the pellets tended to laminate and fracture, especially at the chamfered edge. Table 10 summarizes some of the preliminary pellet properties.

Colloidal graphite was used throughout as a die lubricant, and the addition of two drops of water per charge was sufficient to increase the green strength of the pellets without significant change in the green or sintered densities.

Table 9

PROPERTIES OF $PuO_2$ SHARD FEED MATERIAL
PREPARED BY THE HYDROXIDE PROCESS AND
CALCINED AT 700°C FOR ONE HOUR

| | |
|---|---|
| Particle Size Range | <297 μm |
| Particle Size Distribution | See Figure 9 |
| Apparent Density | 7.7 g/cc (67% TD) |
| Total Impurities | 0.11% |

| Element | Amount (μg/g) |
|---|---|
| Al | 54 |
| Ca | <250 |
| Co | 96 |
| Cu | 73 |
| Fe | 212 |
| Mg | 20 |
| Pb | 137 |
| Ni | 73 |
| Si | 154 |

Pellet sintering temperatures from 1525 to 1750°C for periods up to 6 hr were investigated. It was found that sintering temperatures above 1550°C for periods greater than 2 hr did not significantly increase the pellet density. Higher temperatures for longer periods of time tended to increase the grain size.

Table 10

PRELIMINARY $PuO_2$ PELLETS

| Pellet | Charge (g $PuO_2$) | Green Density (%) | Sintered Density (%) | Shrinkage (%) | Pressing Binder | Pressing Pressure (psi) | Power (W) | Comments |
|---|---|---|---|---|---|---|---|---|
| 1G | 15.20 | 60.8 | 78.6 | diam 8.6<br>ht. 6.6 | $H_2O$ | 50,000 | 5.96 | Laminated and cracked at chamfered edge. |
| 3A | 15.16 | 58.3 | 83.2 | diam 11.5<br>ht. 10.2 | $H_2O$ | 41,000 | 6.00 | |
| 3B | 15.19 | 57.2 | 81.8 | diam 11.5<br>ht. 10.7 | $H_2O$ | 33,000 | 6.01 | |
| 3C | 15.16 | 57.6 | 82.5 | diam 11.5<br>ht. 10.6 | $H_2O$ | 41,000 | 5.98 | |
| 3D | 15.15 | 57.7 | 82.6 | diam 11.5<br>ht. 10.6 | $H_2O$ | 41,000 | 5.98 | |
| 3E | 15.08 | 59.4 | 84.5 | diam 11.5<br>ht. 10.2 | None | 41,000 | 5.98 | |
| 3F | 15.16 | 59.2 | 83.7 | diam 11.5<br>ht. 9.7 | $H_2O$ | 41,000 | 5.97 | |
| 4A | 15.16 | 56.8 | 82.3 | diam 11.8<br>ht. 10.8 | $H_2O$ | 41,000 | 5.98 | |
| 4B | 15.16 | 56.8 | 82.8 | diam 12.0<br>ht. 11.1 | $H_2O$ | 41,000 | 5.98 | |
| 4C | 15.15 | 57.6 | 83.7 | diam 12.0<br>ht. 10.8 | $H_2O$ | 41,000 | 6.00 | |
| 4D | 15.16 | 57.4 | 83.6 | diam 12.0<br>ht. 11.1 | $H_2O$ | 41,000 | 5.98 | |

## PREPARATION OF $^{238}PuO_2$ FUEL PELLETS

Eighteen $^{238}PuO_2$ fuel pellets were prepared from freshly prepared and calcined (700°C for 1 hr) $^{238}PuO_2$ feed material using the fabrication parameters listed in Table 11.

The properties of the fuel pellets, thus produced, are summarized in Table 12. A photograph of several of these pellets is shown in Figure 10. Selected pellets were destructively analyzed for microstructure and chemical impurities. The cross section of the total specimen shown in Figure 11 indicates a reasonably uniform density distribution within the pellet. The cracks and chips shown in the photomicrograph of the total specimen occurred during mounting. Pellet impurities are listed in Table 13.

Table 11

$^{238}PuO_2$ PELLET FABRICATION PARAMETERS

| | |
|---|---|
| Die Design | See Figure 3 |
| Die Lubricant | Colloidal Graphite |
| Binder | Two drops of water/charge |
| Compacting Pressure | 41,000 psi |
| Sintering Temperature | 1550°C |
| Sintering Atmosphere | Oxygen-16 |
| Time | 2 hr |

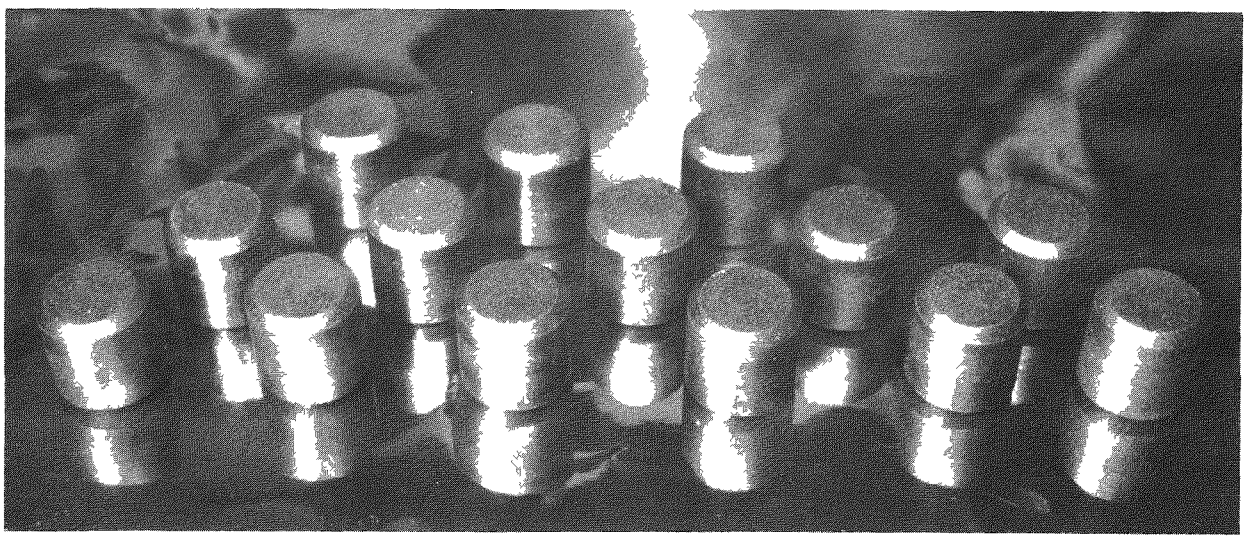

FIGURE 10 - Cold pressed and sintered $PuO_2$ pellets.

Table 12

$PuO_2$ PELLETS

| Charge Weight (g) | Green Dimension (in.) | Green Density (%) | Sintered Weight (g) | Sintered Dimension (in.) | Sintered Density (%) | Shrinkage (%) | Weight Loss on Sintering (%) |
|---|---|---|---|---|---|---|---|
| 15.279 | diam 0.5585<br>ht. 0.587 | 58.6 | 15.1439 | diam 0.499<br>ht. 0.529 | 81.4 | 10.7<br>9.9 | 0.9 |
| 15.263 | diam 0.5585<br>ht. 0.591 | 58.2 | 15.2040 | diam 0.496<br>ht. 0.529 | 82.8 | 11.2<br>10.5 | 0.4 |
| 15.265 | diam 0.5585<br>ht. 0.594 | 57.9 | 15.2011 | diam 0.495<br>ht. 0.531 | 82.8 | 11.4<br>10.6 | 0.4 |
| 15.271 | diam 0.5585<br>ht. 0.595 | 57.8 | 15.1946 | diam 0.494<br>ht. 0.529 | 83.4 | 11.5<br>11.1 | 0.5 |
| 15.270 | diam 0.5585<br>ht. 0.596 | 57.7 | 15.2238 | diam 0.497<br>ht. 0.537 | 81.3 | 11.0<br>9.9 | 0.3 |
| 15.270 | diam 0.5585<br>ht. 0.598 | 57.5 | 15.2151 | diam 0.493<br>ht. 0.529 | 83.9 | 11.7<br>11.5 | 0.4 |
| 15.274 | diam 0.5585<br>ht. 0.594 | 57.9 | 15.2178 | diam 0.496<br>ht. 0.536 | 81.7 | 11.2<br>9.8 | 0.4 |
| 15.272 | diam 0.5585<br>ht. 0.603 | 57.0 | 15.2057 | diam 0.492<br>ht. 0.537 | 82.9 | 11.9<br>10.9 | 0.4 |
| 15.252 | diam 0.5585<br>ht. 0.577 | 59.6 | 15.1430 | diam 0.5005<br>ht. 0.525 | 81.6 | 10.4<br>9.0 | 0.7 |
| 15.247 | diam 0.5585<br>ht. 0.574 | 59.9 | 15.1388 | diam 0.4995<br>ht. 0.520 | 82.7 | 10.4<br>9.4 | 0.7 |
| 15.250 | diam 0.5585<br>ht. 0.572 | 60.1 | 15.1340 | diam 0.4995<br>ht. 0.519 | 82.7 | 10.4<br>9.3 | 0.8 |
| 15.240 | diam 0.5585<br>ht. 0.582 | 59.0 | 15.1820 | diam 0.497<br>ht. 0.523 | 83.3 | 11.0<br>10.1 | 0.4 |
| 15.241 | diam 0.5585<br>ht. 0.585 | 58.7 | 15.1845 | diam 0.497<br>ht. 0.527 | 82.7 | 11.0<br>9.9 | 0.4 |
| 15.241 | diam 0.5585<br>ht. 0.583 | 58.9 | 15.1751 | diam 0.497<br>ht. 0.524 | 83.1 | 11.0<br>10.1 | 0.4 |
| 15.246 | diam 0.5585<br>ht. 0.587 | 58.5 | 15.1414 | diam 0.496<br>ht. 0.528 | 82.6 | 11.2<br>10.1 | 0.7 |
| 15.247 | diam 0.5585<br>ht. 0.587 | 58.5 | 15.1339 | diam 0.496<br>ht. 0.528 | 82.6 | 11.2<br>10.1 | 0.7 |
| 15.242 | diam 0.5585<br>ht. 0.585 | 58.7 | 15.1260 | diam 0.497<br>ht. 0.525 | 82.7 | 11.0<br>10.3 | 0.8 |
| 15.253 | diam 0.5585<br>ht. 0.579 | 59.4 | 15.0985 | diam 0.498<br>ht. 0.521 | 82.7 | 10.7<br>10.0 | 1.0 |

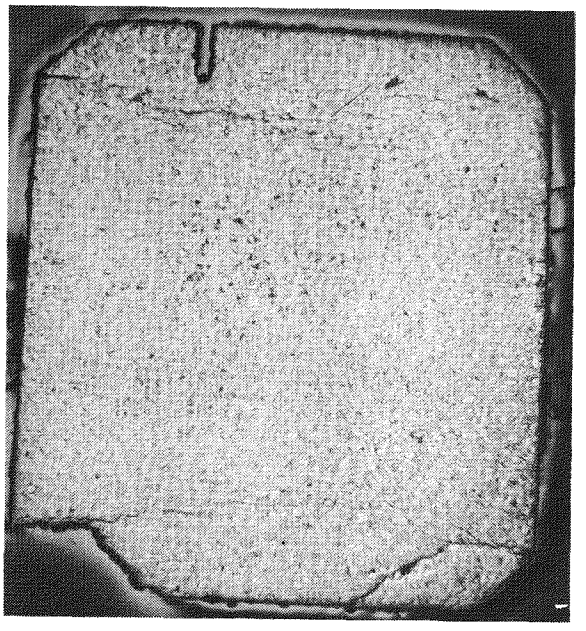


FIGURE 11 - Metallographic cross section of a $PuO_2$ cold pressed and sintered pellet. Total specimen 3X, all other magnifications 125X.

Table 13

IMPURITY ANALYSIS OF A SELECTED $^{238}PuO_2$ FUEL PELLET

| Element | Amount (µg/g) Exterior | Interior |
|---|---|---|
| C | 22 | 34 |
| Fe | 450 | 500 |
| Si | 100 | 100 |
| Mn | 12 | 12 |
| Pb | <10 | <10 |
| Mg | 10 | 10 |
| Cr | 30 | 30 |
| Al | 100 | 100 |
| Ca | <250 | <250 |
| Cu | 20 | <10 |
| Co | 25 | 25 |
| Ni | 70 | 70 |

## SUMMARY

This study demonstrates the feasibility of preparing reproducible $^{238}PuO_2$ fuel pellets from low-fired hydroxide shards by a cold press and sinter technique. During the course of this study 18 $PuO_2$ and 27 $UO_2$ chamfered pellets were prepared to the following specifications: diameter - 0.494 ± 0.005 in., height - 0.525 ± 0.015 in., density - 81 to 85% TD and wattage (for $PuO_2$ pellets) - 6.0 ± 0.05 W.

$PuO_2$ pellet preparation parameters were investigated. Properties of the pellets depend on the $PuO_2$ feed preparation parameters, compaction pressure, and the sintering conditions. Of these parameters, the thermal history of the feed shards is the most critical for control of the dimensional and density properties of the pellets.

The hydroxide-precipitated and dried feed shards must be calcined with careful control of temperature and time. It was also noted that aged feed shards exhibited rather different pressing and sintering behavior. The properties of aged shards appear to be similar to those of shards calcined at much higher temperatures. This aging effect is believed to be due to a reduction of the surface free energy resulting from the self-heat in $PuO_2$ (self-sintering). The kinetics and mechanism of this reaction require further investigation.

The microstructural features of the pellet also depend on the preparation parameters. The amount of porosity depends more on the nature of the feed material and the pressing parameters than on the sintering conditions beyond 1550°C. Under extreme sintering conditions the grain size increases without a significant increase in density.

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## REFERENCE

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