

# MASTER

ALTERNATIVE OXIDE FUEL  
PELLET FABRICATION FOR  
IRRADIATION TESTING

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D. E. Rasmussen  
W. R. Jentzen  
R. B. McCord

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ALTERNATIVE OXIDE FUEL PELLETT FABRICATION FOR IRRADIATION TESTING

by

D. E. Rasmussen, W. R. Jentzen, and R. B. McCord

Hanford Engineering Development Laboratory  
Richland, Washington

operated by

Westinghouse Hanford Company

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ABSTRACT

*Fabrication of experimental breeder reactor fuel pellets by the common cold-press-and-sinter technique for irradiation testing in EBR-II is discussed. Fuel types include mixed thoria-plutonia,  $UO_2$  enriched with 22 weight percent  $^{233}U$  in  $U$ ,  $UO_2$  enriched with thirty-four weight percent  $^{235}U$  in  $U$ , and mixed urania-plutonia.*

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1



## INTRODUCTION

The Hanford Engineering Development Laboratory (HEDL) fabricates mixed oxide fuel pellets and insulator fuel pellets as part of the national breeder reactor development program.<sup>1-4</sup> This presentation (See Figure 1) discusses the fabrication of four nuclear fuel pellet batches made for irradiation testing in Experimental Breeder Reactor II in Idaho Falls, Idaho. The objective of the test is to produce fuel pin performance data for alternative oxide fuel designs with advanced alloy cladding materials (See Figure 2). The four fuel types fabricated for this test are listed in Figure 3 and include a  $\text{UO}_2$  fuel enriched with 21.85 weight percent (w/o)  $^{233}\text{U}$  in U, a mixed 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  fuel, a  $\text{UO}_2$  fuel enriched with 34 w/o  $^{235}\text{U}$  in U, and a reference 15 w/o  $\text{PuO}_2$ -85 w/o  $\text{UO}_2$  fuel enriched with 15 w/o  $^{235}\text{U}$  in U. Target fuel pellet density (88.6% of theoretical density (% TD)) was based on a planar smear density of 85% TD, which is the reference value for Fast Flux Test Facility fuel. The advanced alloy cladding materials include Inconel 706, PE 16 and a reference 316 stainless steel. The test is designed to operate in EBR-II to simulate hot channel temperature and power conditions anticipated near the top end of an alternative mixed oxide fuel assembly.

FIGURE 1. ALTERNATIVE OXIDE FUEL PELLETT FABRICATION FOR IRRADIATION TESTING

DE Rasmussen/AR Jentzen/AS McGraw  
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FIGURE 3. Alternative Oxide Test Design

P-20-2 Alternative Oxide Fuel Specifications

	Fuel Type			
	$(^{233}\text{U}, ^{235}\text{U})\text{O}_2$	$(^{235}\text{U}, ^{238}\text{U})\text{O}_2$	$\text{PuO}_2\text{-}15\text{UO}_2\text{-}85\text{UO}_2\text{-}x$	$\text{PuO}_2\text{-}35\text{ThO}_2\text{-}65\text{UO}_2\text{-}x$
Uranium Enrichment (wt% U in U)	22	34	21	--
Number of Pins	2	12	15	22
Pellet Diameter (in.)	0.2505	0.2605	0.2505	0.2605
Pellet Geometry	Flat Ends	Flat Ends	Flat Ends	Flat Ends
Pellet Density (%TD)	88.6	88.6	88.6	88.6
Fuel G/M	2.000	2.000	1.965	1.930

FIGURE 2. ALTERNATIVE OXIDE TEST OBJECTIVES

- Alternative Oxide Fuel Performance Data
  - $\text{UO}_2/\text{UO}_2$  OXIDE
  - $\text{Pu}/\text{Th}$  OXIDE
  - $\text{UO}_2/\text{UO}_2$  OXIDE
  - $\text{Pu}/\text{U}$  OXIDE
- Advanced Alloy Cladding Performance Data
  - Inconel 706
  - PE 16
  - 316 Stainless Steel

2

## FABRICATION OF $\text{UO}_2$ FUEL CONTAINING 21.85 WEIGHT PERCENT $^{233}\text{U}$ IN $\text{U}$

### FEED POWDERS

Natural  $\text{UO}_2$  was blended with an enriched  $\text{UO}_2$  powder lot containing 98.03 weight percent  $^{233}\text{U}$  in  $\text{U}$  to achieve a final enrichment of 21.85 weight percent  $^{233}\text{U}$  in  $\text{U}$ . The natural  $\text{UO}_2$  powder lot used was an Eldorado powder produced by the ammonium diuranate process and had a surface area of  $6.2 \text{ m}^2/\text{gram}$ . Particle size distribution is shown in Figure 4.

Enriched uranium dioxide containing 98.03 weight percent  $^{233}\text{U}$  in  $\text{U}$  was obtained from the Hanford Engineering Development Laboratory's Materials Engineering Department by permission of the Department of Energy. Uranium-232 concentration associated with this uranium-233 oxide was  $1.5(10)^{-4}$  weight percent, which was relatively low for a uranium-233 powder. Uranium-232 is formed by radioactive decay daughter product reactions and is the source of high gamma radiation activity. Gamma activity from  $^{232}\text{U}$  increases quickly with time, as shown in Figure 5.

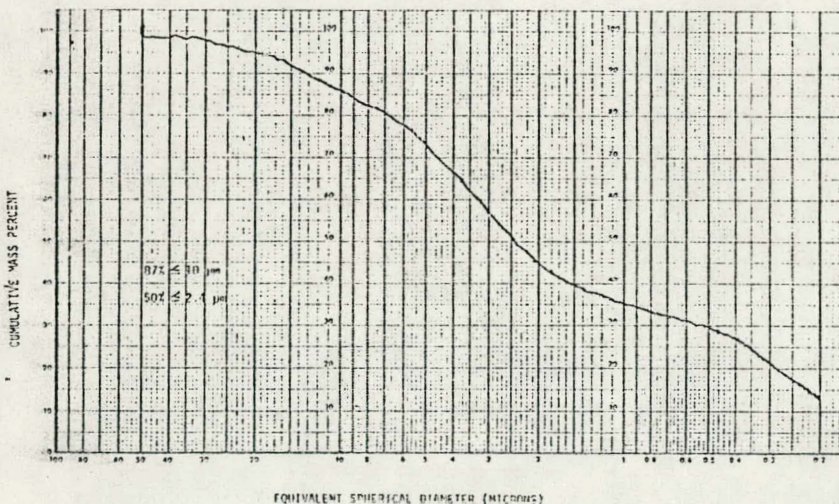


FIGURE 4. Particle Size Distribution of Natural  $\text{UO}_2$  Powder Lot US-374

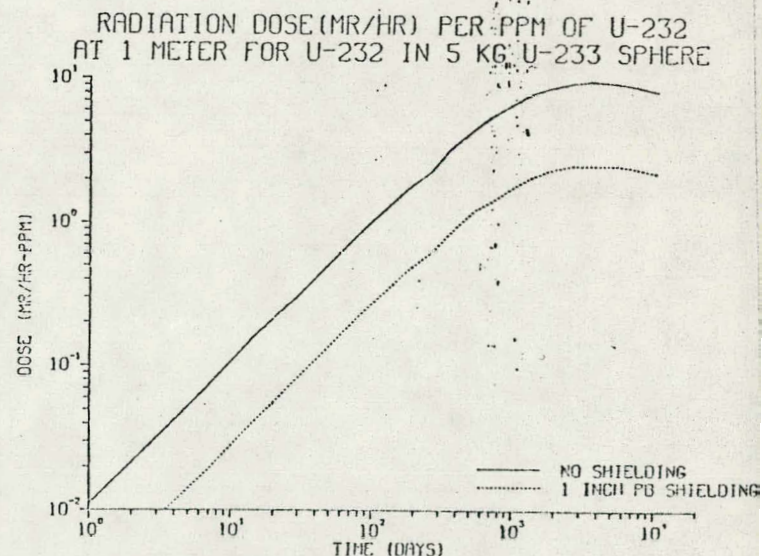


FIGURE 5. Gamma Activity of  $^{232}\text{U}$  As A Function of Time



## PROCESS DEVELOPMENT

Four trial pellet batches were fabricated from the straight natural  $UO_2$  lot prior to fabrication of the final fuel pellet batch. The trial batches were to obtain processing data such as pressing behavior, sintering shrinkage, and sintered pellet density as a baseline for fabrication of the actual fuel batch. Trial pellets containing four different levels of organic binder/additive were all final pressed at 28,700 lbs/in<sup>2</sup> (14.4 tons/in<sup>2</sup>) and sintered together. Results are shown in Figures 6-8. The decrease in sintered density followed by an increase in sintered density with increasing organic binder concentration seen in Figure 6 is attributed to the relatively low pressure used for this natural  $UO_2$  powder. The final batch was pressed at 57,400 psi to stabilize pellet density.

## FABRICATION PROCESS

The process flowsheet used for fabrication of the  $UO_2$  fuel containing 21.85 weight percent  $^{233}U$  in  $U$  is shown in Figure 9. Processing equipment are illustrated in Figures 10-13. The  $UO_2$  powders were manually pre-mixed in one kilogram increments in

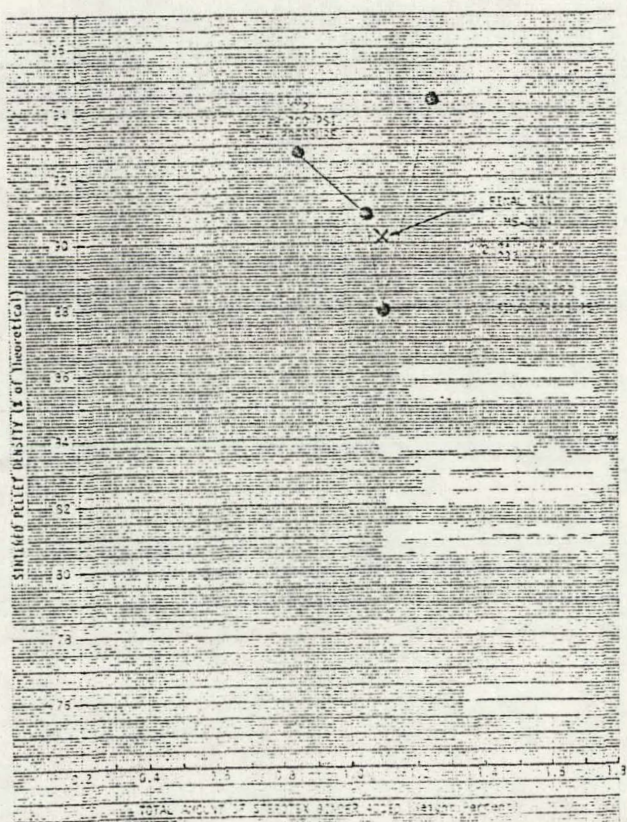


FIGURE 5. Natural  $UO_2$  Sintered Pellet Density As A Function of Total Sterotex Organic Additive Concentration

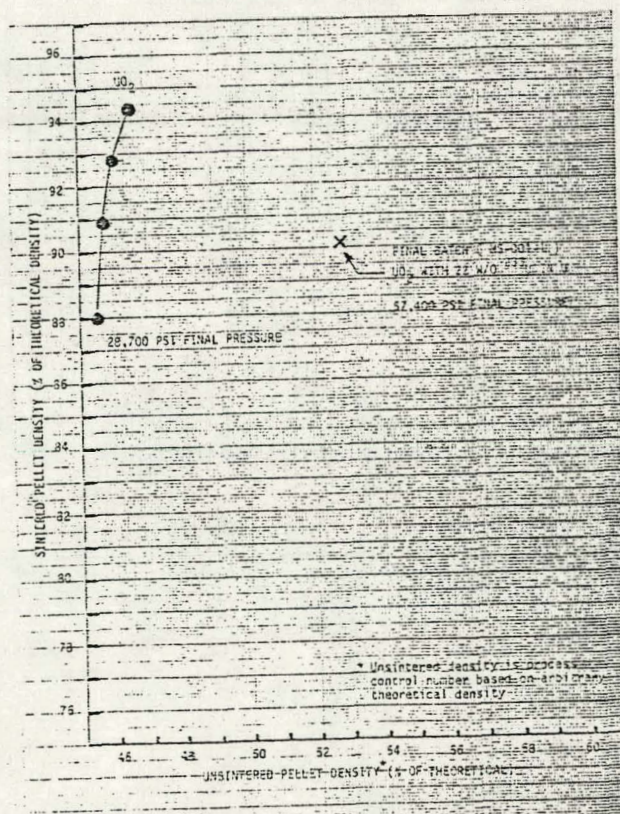


FIGURE 7. Natural  $UO_2$  Sintered Pellet Density As A Function of Unsintered Pellet Density

4



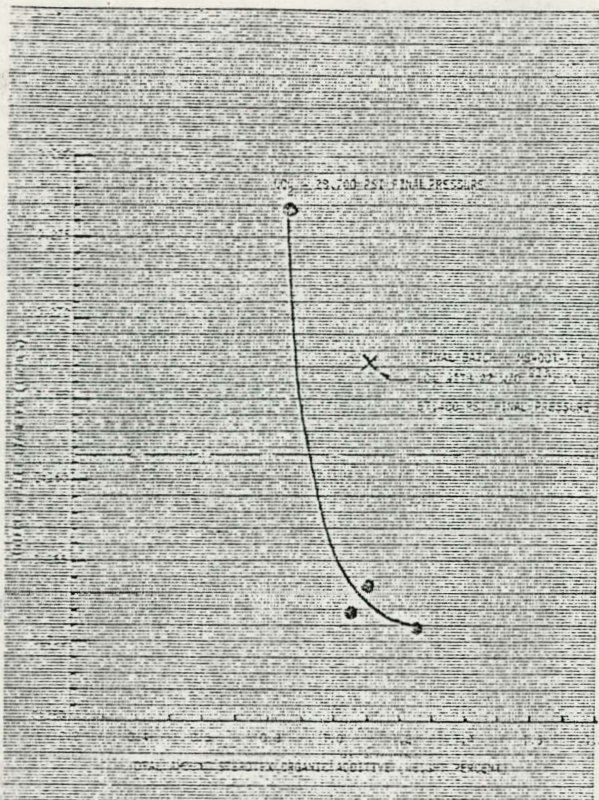


FIGURE 8. Natural  $UO_2$  Sintered Pellet Diameter As A Function of Total Sterotax Organic Additive Concentration

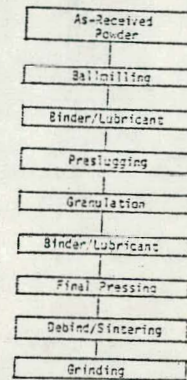


FIGURE 9. Process Flowsheet for Fabrication of  $UO_2$  Fuel Containing 21.85 Weight Percent  $^{235}U$  in  $U$ .

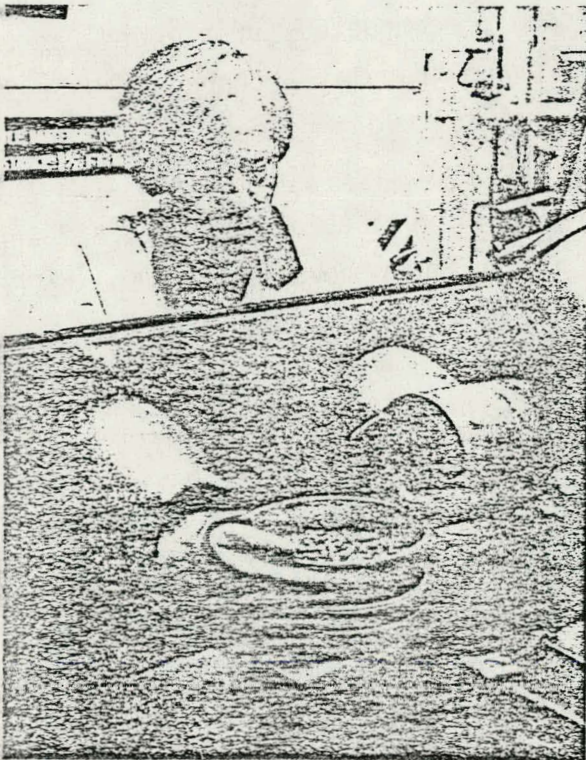


FIGURE 10. Powder Ballmilling Vials and Tungsten Carbide Grinding Media.

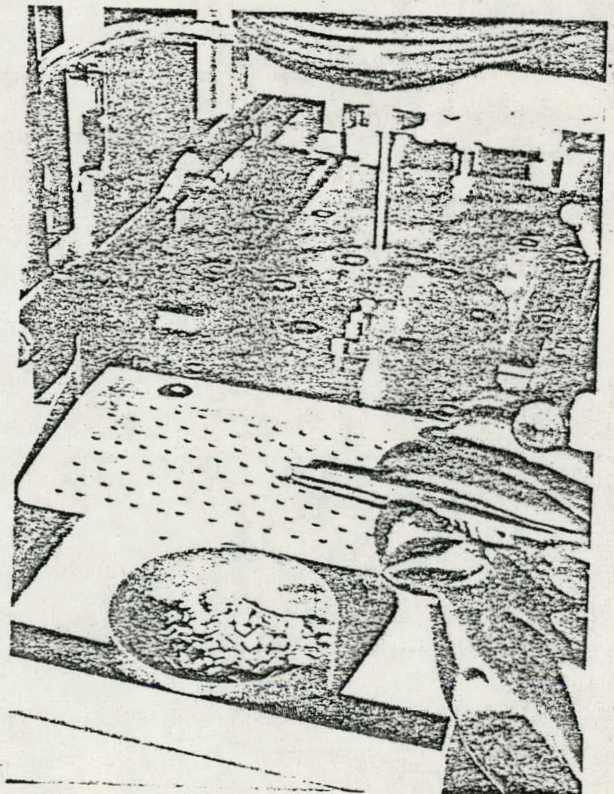


FIGURE 11. Fuel Pellet Hydraulic Press



## SINTERING FURNACE

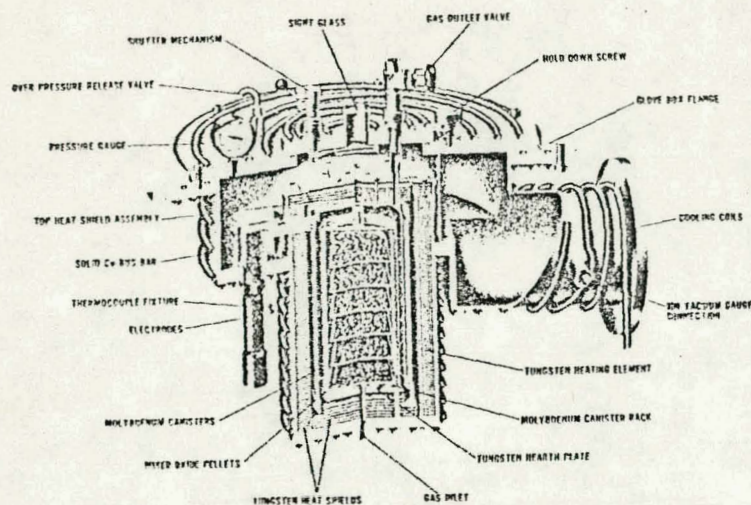


FIGURE 12. Fuel Pellet Sintering Furnace

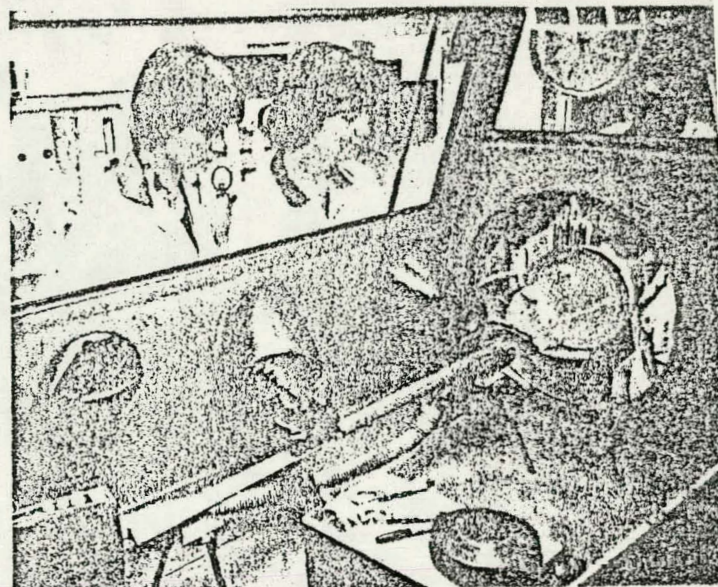


FIGURE 13. Fuel Pin Loading Station

a stainless steel pan and were then ballmilled for 20 hours in a rubber-lined balljar containing tungsten carbide grinding media (with a ratio of 1 kg powder/8 pounds grinding media). After ballmilling (to achieve a high degree of homogeneity as is normal practice for HEDL irradiation test fuels), 0.35 weight percent Sterotex organic additive was manually mixed with the powder. The  $UO_2$  was then precompact (preslugged) at 23,200 lbs/in<sup>2</sup> (11.6 tons/in<sup>2</sup>) in a 0.500 inch diameter die and granulated through a U.S. Standard 2--mesh sieve. After granulation, an additional 0.75 weight percent Sterotex was manually mixed with the powder and it was final pressed at 57,400 lbs/in<sup>2</sup> (28.7 tons/in<sup>2</sup>) in a 0.318 inch diameter die. Final pressed pellets were approximately 0.322 inch in diameter and 0.38 inch in length. Organic binder was removed by heating to 650°C for two hours in argon-8% hydrogen flowing at 8 cfh. Heating rate was 200°C/hour and cooling rate was 300°C/hour. Pellets were then sintered at 1690°C for six hours in argon-8% hydrogen flowing at 6 cfh.<sup>5-7</sup> Heating rate was 200°C/hour and cooling rate was 300°C/hour down to 1100°C. The sintering furnace was switched over to vacuum after the pellets cooled to 100°C so as to combine the usual vacuum offgas cycle with the sintering run in order to reduce the number of process steps for this fuel. After switching to vacuum, cooling rate was approximately 150°C/hour. All pellets were centerless ground to approximately 0.260 inch diameter and loaded into two 316 stainless steel cladding tubes with a 0.266 inch inside diameter and 0.290 inch outside diameter for irradiation testing in Experimental Breeder Reactor II in Idaho Falls, Idaho.



## FUEL PELLET CHARACTERISTICS

Sintered fuel pellet density for the  $\text{UO}_2$  fuel enriched 21.85%  $^{233}\text{U}$  in U was 9.88 g/cc (90.2% of theoretical density) after centerless grinding. Theoretical density (TD) was calculated using the empirical formula:

$$\text{TD} = \frac{(5.8755 + 2.54M)[(232)(F_{232}) + (233)(F_{233}) + (234)(F_{234}) + (235)(F_{235}) + (236)(F_{236}) + (237)(F_{237}) + (238)(F_{238})]}{237.98}$$

WHERE: TD = Theoretical Density in g/cc  
M = Oxygen-to-Metal Ratio  
 $F_{232}, \dots, F_{238}$  = Weight Fraction of  $^{232}\text{U}$ , ...,  $^{238}\text{U}$ , etc.  
Isotopes in U

237.98 = Average Atomic Weight of Natural Uranium  
5.8755 = Empirically-Determined Constant  
2.54 = Empirically-Determined Constant

Smear fuel density as-loaded into the cladding was 9.36 g/cc (86.2% of theoretical) and was calculated as follows:

$$\text{PSD} = \rho \frac{(d)^2}{(D)^2}$$

WHERE:

PSD = Planar smear density expressed in either g/cc or % TD, respectively  
 $\rho$  = Fuel pellet density expressed in either g/cc or % TD, respectively  
d = Fuel pellet diameter  
D = Fuel pin cladding inside diameter

Pellet density results are plotted in Figures 6-8. Average sintered pellet length was 0.3225 inch. Average sintered diameter (before centerless grinding) was 0.2672 inch which represented a sintering shrinkage of 17.0% based on unsintered pellet diameter. Photomicrographs of typical pellet polished cross sections are shown in Figures 14-15. The relatively large pores are attributed to the Sterotex and powder compaction behavior. More detailed microstructure analysis is in progress and will be discussed in a future report. <sup>12-13</sup>

8-11

7



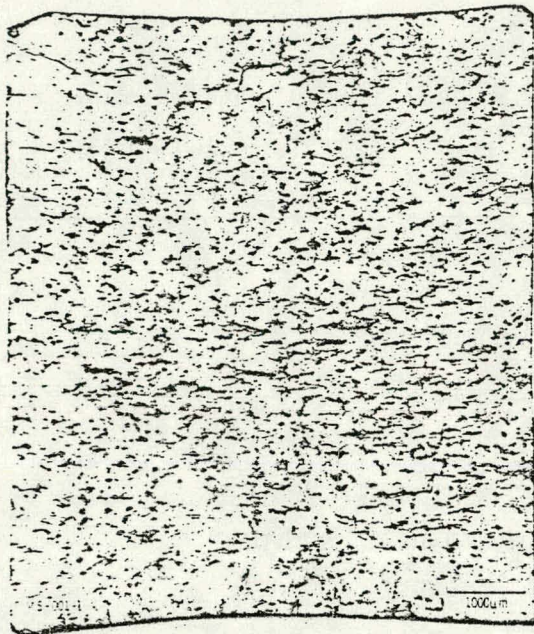


FIGURE 14. Photomicrograph of Polished Cross Section of  $UO_2$  Fuel Pellet Containing 21.85 Weight Percent  $^{233}U$  in  $U$  (Original 30x magnification)

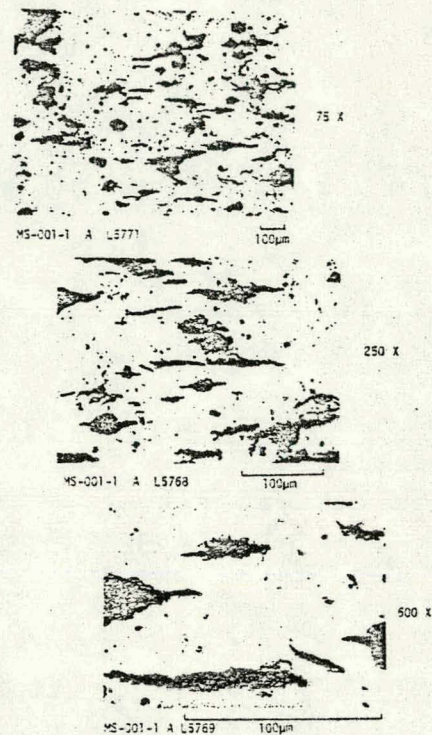


FIGURE 15. Photomicrographs of Polished Cross Section of Typical  $UO_2$  Fuel Pellet Containing 21.85 Weight Percent  $^{233}U$  in  $U$  (Originals 75x, 250x, and 500x Magnifications)

## RADIATION EXPOSURE

Radiation exposure was monitored closely during production of the  $UO_2$  fuel enriched 22% in  $^{233}U$  in  $U$  to assure exposure was maintained within Department of Energy guidelines. For this project, the fuel batch size was small (300 grams), so glovebox operations were permissible and radiation exposure was well within the HEDL guidelines of 4 rems per year whole body dose. The gloveboxes were temporarily lined with plastic to assure easy material cleanout.

Similarly, radiation exposure for fabrication of the other fuels fabricated in this report were well within normal limits.



FEED POWDERS

Plutonium dioxide powder produced by the oxalate precipitation process was blended with thorium dioxide derived by an oxalate precipitation process to achieve a blend of 35 weight percent  $\text{PuO}_2$ , 65 weight percent  $\text{ThO}_2$ . The  $\text{PuO}_2$  powder had a surface area of  $17.1 \text{ m}^2/\text{gram}$ .  $\text{PuO}_2$  particle size distribution is presented in Figure 16.

Thorium dioxide powder used for this project exhibited a surface area of  $20.9 \text{ m}^2/\text{gram}$  and was obtained from the Pacific Northwest Laboratory by approval of the Department of Energy.  $\text{ThO}_2$  particle size distribution is shown in Figure 17. Typical scanning electron micrographs of the  $\text{ThO}_2$  are shown in Figure 18, which show the discreet, rosette-type particle morphology characteristic of  $\text{PuO}_2$  powders.

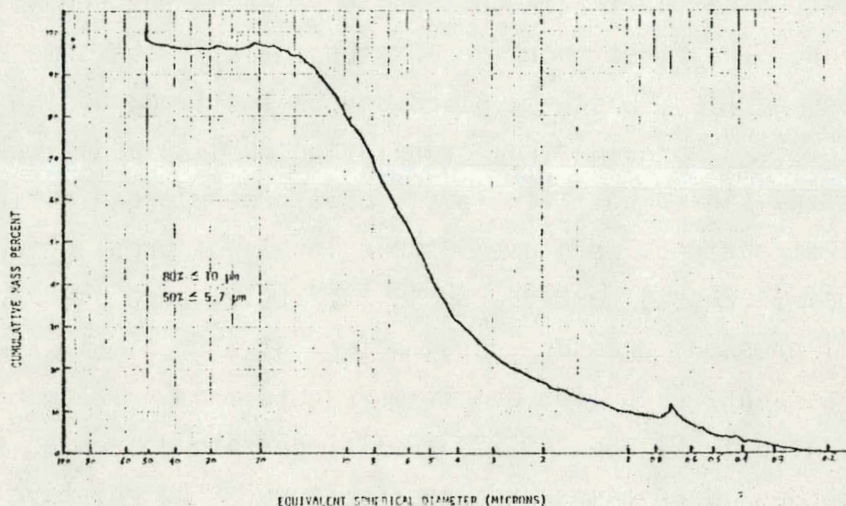


FIGURE 16. Particle Size Distribution of  $\text{PuO}_2$  Powder Lot Pu-114

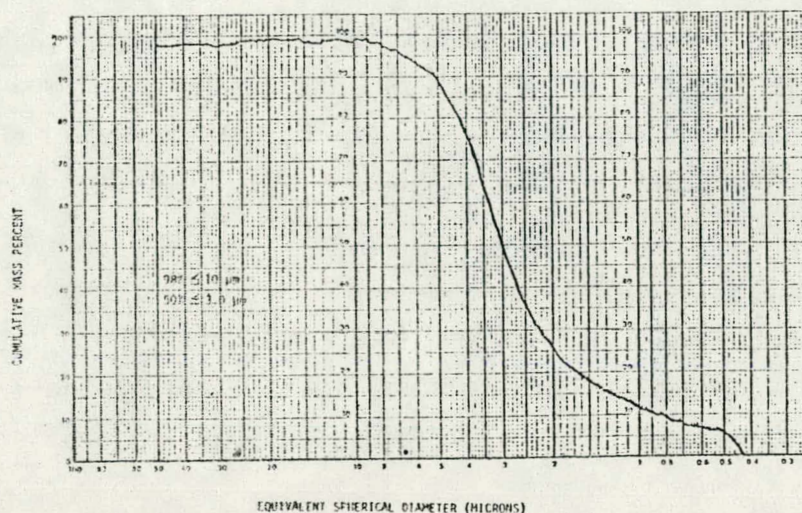


FIGURE 17. Particle Size Distribution of  $\text{ThO}_2$  Powder Lot TH-002



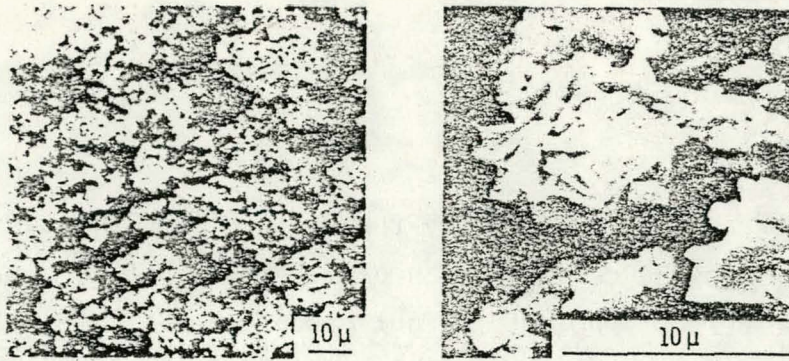


FIGURE 18. Typical Scanning Electron Micrographs of  $\text{ThO}_2$  Powder Lot TH-002

### PROCESS DEVELOPMENT

Two basic processing techniques were used on the trial pressing and sintering tests performed to determine fabrication process parameters for the 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  fuel batch. The first technique was essentially the same method used on the uranium fuel described earlier, which involved dry ballmilling, adding dry organic binder/lubricant (Sterotex), preslugging, granulating, adding additional Sterotex, final pressing, debinding, and sintering. Six levels of Sterotex organic binder, ranging from 0.4 to 1.05 weight percent, were used. Four levels of preslugging pressure, ranging from 8,400 psi (4.2 tsi) to 27,400 psi (13.7 tsi), were used in combination with seven levels of final pressing pressure ranging from 34,300 psi (17.2 tsi) to 59,700 psi (29.8 tsi). Resultant sintered pellet densities ranged from 83.2 to 86.6% TD. Results are graphically presented in Figures 19-22. Pellet density decreased with increasing Sterotex concentration and decreasing pressing pressure. The relatively low sinterability was attributed to the characteristics of the blended  $\text{PuO}_2$ - $\text{ThO}_2$  powder. Straight  $\text{ThO}_2$  pellets made previously using this  $\text{ThO}_2$  powder lot have achieved densities up to approximately 93% TD. Sinterability tests on the  $\text{PuO}_2$  powder lot yielded density values of 95% TD. However, the  $\text{ThO}_2$  powder and mixed  $\text{PuO}_2$ - $\text{ThO}_2$  powders both exhibited poor flowability behavior (as does straight  $\text{ThO}_2$ ) compared with  $\text{UO}_2$  powder or mixed  $\text{PuO}_2$ - $\text{UO}_2$ . Consequently, the mixed  $\text{PuO}_2$ - $\text{ThO}_2$  did not feed into a die cavity as well or compact under pressure as well as other fuel compositions.

The second processing technique used on the 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  material was that of dissolving Carbowax organic additive in water, slurrying that with the powder, and drying the slurry at 70°C for an empirically determined length of time.



The dried powder was then processed the same as described earlier. Higher sintered pellet densities have typically been achieved in the past using this wet binder addition method. Two Carbowax organic additive concentrations 1.5 and 3.0 weight percent, were used in combination with one preslug pressure (10,600 psi (5.3 tsi)) and four final compaction pressures ranging from 18,500 psi (9.4 tsi) to 42,300 psi (21.2 tsi). Different U. S. Standard sieve sizes, ranging from 20-mesh to 200-mesh were used for the granulation operation, but they had insignificant influence on sintered pellet density in this test. Higher sintered pellet densities (up to 89.6% TD) were achieved by the wet Carbowax binder addition technique which met the required specification of 88.6% TD. The relatively higher sintered pellet densities attained by the wet Carbowax addition process was attributed to the uniform binder distribution.

Results are shown in Figures 19-22. Higher densities were achieved with 3.0 w/o Carbowax concentration because of the improved powder compaction (as shown in Figures 19, 21, and 22). Since preslugging pressure had little effect on densities for the Sterotex process (as seen in Figure 20, only one level was tested for the Carbowax process.

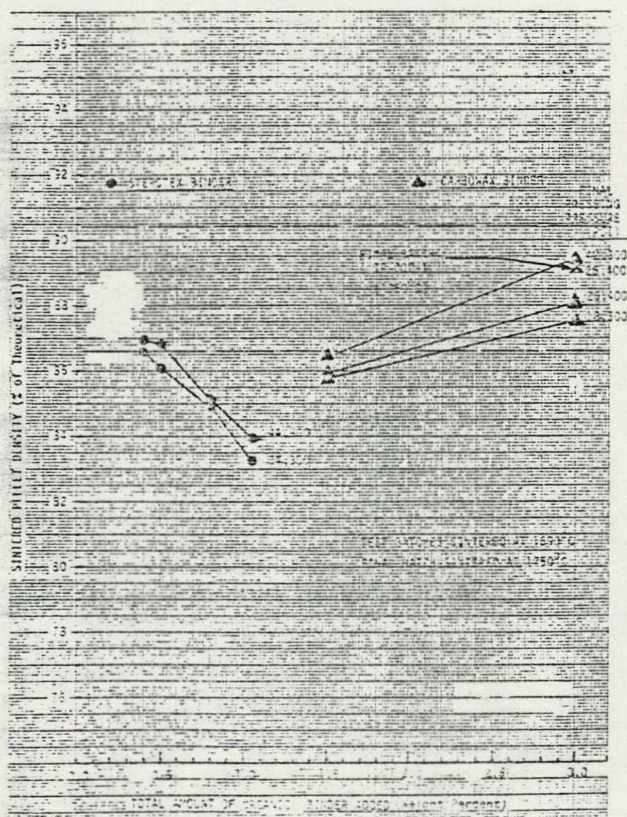


FIGURE 19. Sintered Pellet Density as a Function of Total Organic Additive Concentration for 35 w/o  $PuO_2$ -65 w/o  $ThO_2$  Fuel Pellets

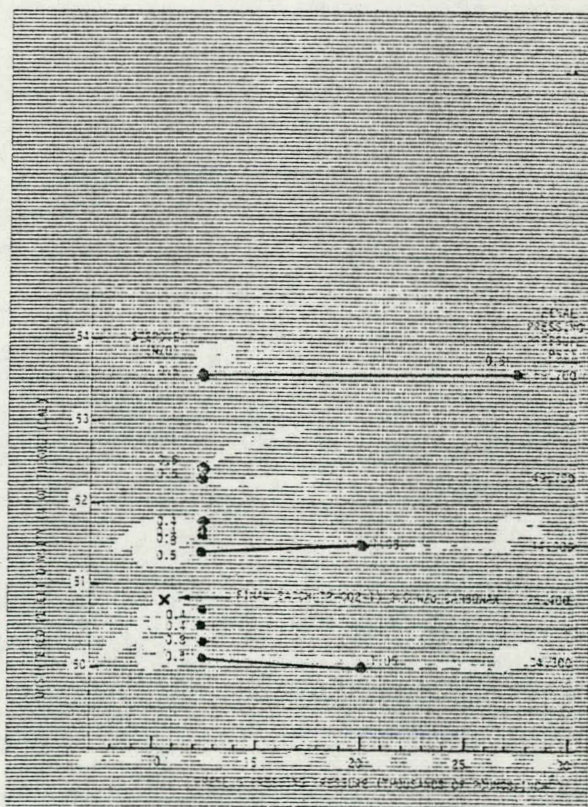


FIGURE 20. Unsintered Pellet Density as a Function of Preslugging Compaction Pressure for 35 w/o  $PuO_2$ -65 w/o  $ThO_2$  Fuel Pellets



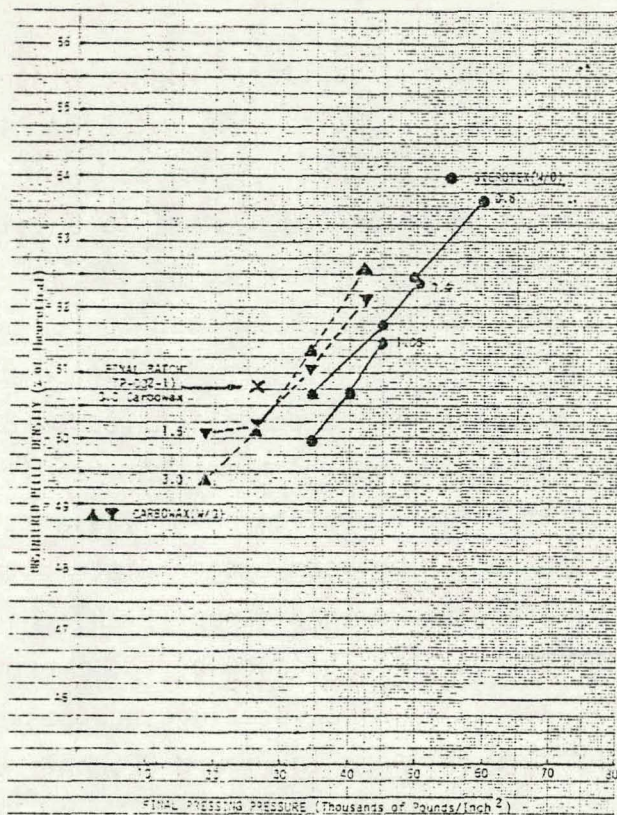


FIGURE 21. Unsintered Pellet Density As A Function of Final Pressing Pressure For 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  Fuel Pellets

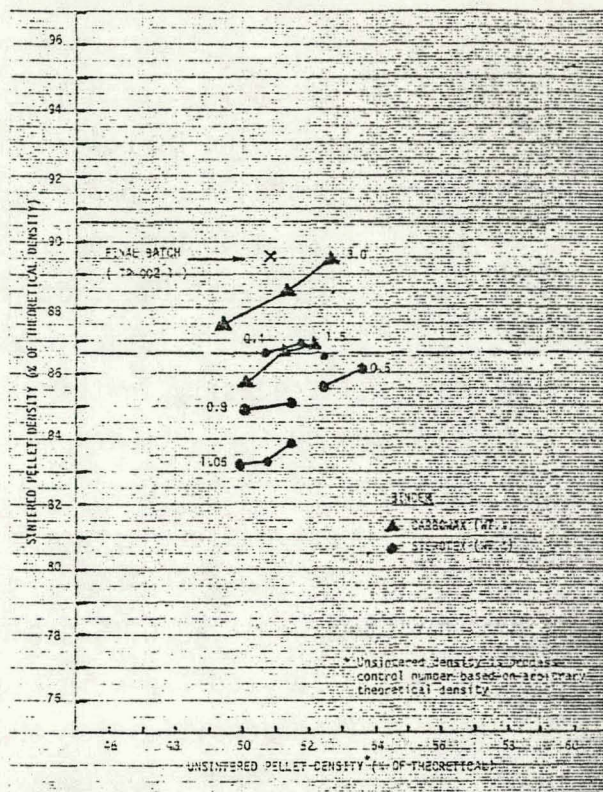


FIGURE 22. Sintered Pellet Density As A Function of Unsintered Pellet Density For 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  Fuel Pellets

## FABRICATION PROCESS FLOWSHEET

Figure 23 shows the process flowsheet followed during production of the 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  fuel pellet batch. The  $\text{PuO}_2$  and  $\text{ThO}_2$  powders were manually blended together to form one kilogram increments which were ballmilled twenty hours in a rubber lined ball jar containing 8 pounds of cylindrical tungsten carbide grinding media per kilogram (2.2 pounds) powder. After ballmilling, the 1 kilogram increments were blended together for 15 minutes in a twin-shelled mechanical blender (vee blender). Carbowax organic binder (in the form of thin flakes) was dissolved in demineralized water to form a 20 weight percent Carbowax solution. The mixed oxide powder was slurred in 500 gram increments with Carbowax solution to achieve a final binder concentration of 3.0 weight percent based on dry powder weight. The slurrying operation was performed in 500 gram increments because of the size of the stainless steel pans and drying oven. All slurries were dried at 70°C for an empirically determined length of time and then granulated together through a U.S. Standard 60-mesh sieve.

12



Preslug pellets were compacted in a 0.500 inch diameter die at 10,600 psi (5.3 tsi) to a green density of 47.7% TD and granulated through a U.S. Standard 20 mesh screen. The granulated powder was final pressed in a 0.316 inch diameter die at 26,400 psi (13.2 tsi) to a green pellet density of 50.8% TD. Organic binder was burned out by heating at 650°C for two hours. Heating rate was 100°C/hour and cooling rate was 300°C/hour. During the sintering cycle, a transistor in the furnace controller system failed which caused the pellets to sinter at 1500°C instead of the setpoint of 1690°C. Therefore, the 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  pellets were resintered at 1750°C for twelve hours, which successfully raised pellet density from 82.6 to 89.5% TD. Heating and cooling rates were the usual, 200°C hour and 300°C hour, respectively. All pellets were dry centerless ground to approximately 0.2595 inch diameter. After grinding, the usual vacuum offgas heat treatment at 850°C for six hours was performed. Vacuum offgassing is performed routinely to assure residual moisture and gases are removed from the fuel pellets. Pellets were then loaded into three types of fuel pin cladding, including 316 stainless steel, Inconel 706, and PE 16. The performance of these advanced alloy cladding in EBR-II is also being evaluated in this test program. Fuel pin diameters were the same as described earlier, 0.266 inch inside and 0.290 inch outside diameter.

#### FUEL PELLET CHARACTERISTICS

Finished, sintered fuel pellet density for the 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  fuel was 9.27 g/cc (89.5% TD) after centerless grinding. Fuel planar smear density (calculated as described earlier) was 8.83 g/cc (85.2% TD). Results are plotted and labeled in Figures 19-22. Sintered pellet density was slightly higher than achieved during the trial runs for the same green pellet density because the final batch was sintered at 1750°C.

Average sintered pellet diameter (before centerless grinding) was 0.2648 inch, which represented a sintering shrinkage of 16.6% based on unsintered pellet diameter. Average sintered pellet length was 0.3114 inch. Photomicrographs of typical fuel pellet polished cross sections are shown in Figures 24-25. The lack of relatively large pores produced by Sterotex binder is apparent for this fuel which indeed did not use Sterotex, but instead used wet Carbowax which produces a very uniform appearance as seen in Figure 24. Photomicrographs of a chemically-etched pellet cross section are shown in Figure 26. An alpha-autoradiograph of a polished fuel pellet cross section is shown in Figure 27. Any plutonium-rich regions would show up as optically dark regions on the



autoradiograph. A high degree of Pu homogeneity is required for HEDL irradiation test fuels and Figure 27 shows the relatively uniform Pu distribution and lack of any large Pu-rich regions. Microstructure characterization measurements of the  $\text{PuO}_2\text{-ThO}_2$  fuel pellets is in progress and will be discussed in a future report.

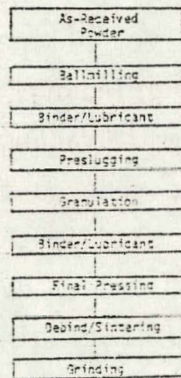


FIGURE 23. Process Flowsheet for Fabrication of 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  Fuel

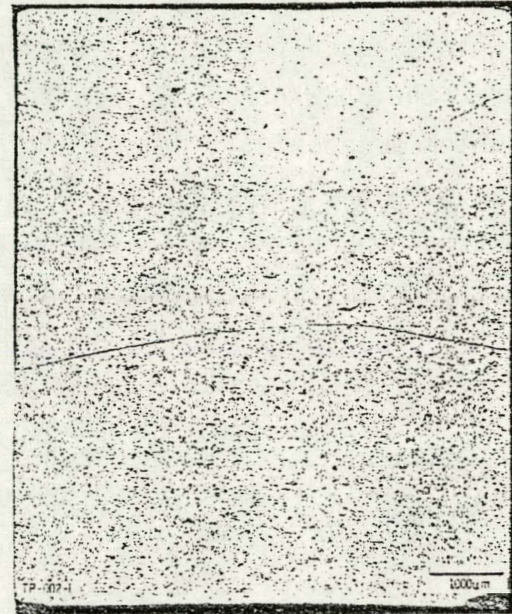


FIGURE 24. Photomicrograph of Polished Cross Section of 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  Fuel Pellet (original 30x magnification)

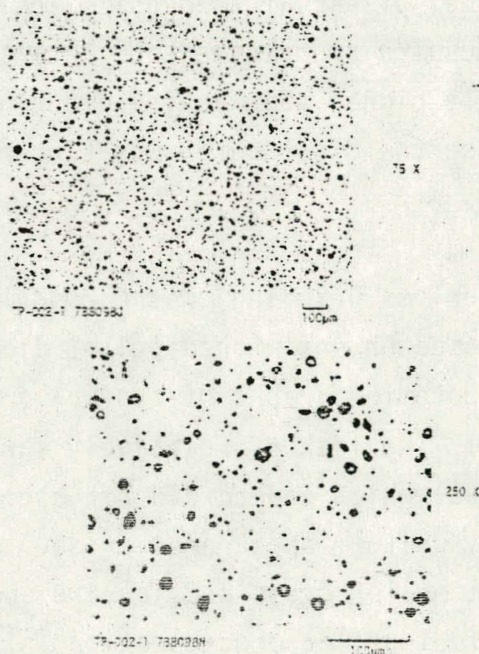


FIGURE 25. Photomicrographs of Polished Cross Section of 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  Fuel Pellet (Originals 75x, 250x Magnifications)

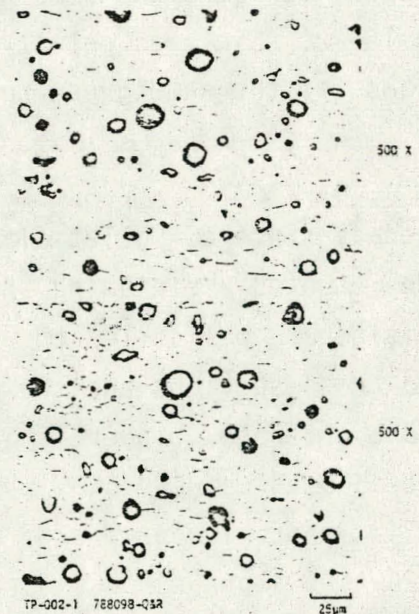
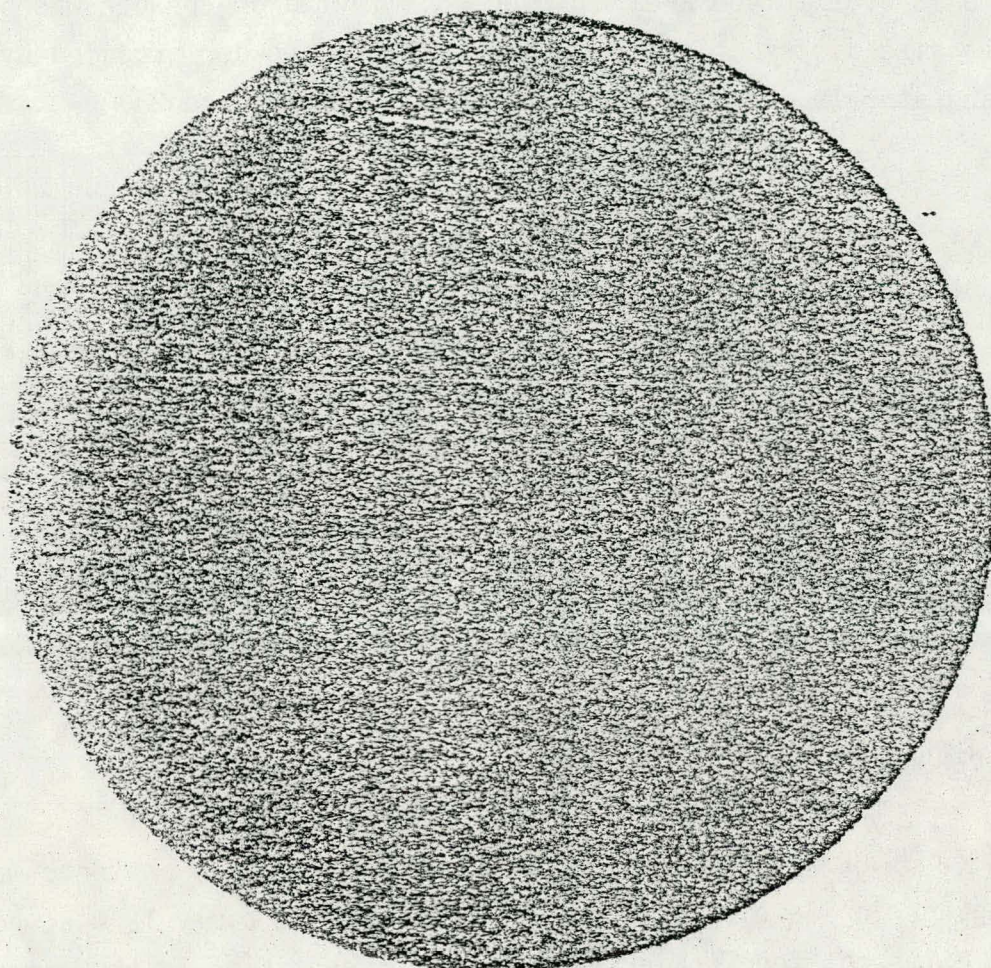


FIGURE 26. Photomicrographs of Chemically Etched Cross Section of 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  Fuel Pellet (Originals 500x Magnification)





TP-002-1 788098-A3

250 $\mu$ m

FIGURE 27. Alpha-Autoradiograph of Polished Cross Section of 35 w/o  $\text{PuO}_2$ -65 w/o  $\text{ThO}_2$  Fuel Pellet (Original 30x Magnification)



FEED POWDERS

Two powders, the same natural  $\text{UO}_2$  powder lot described earlier and a 93% enriched ( $^{235}\text{U}$  in U) $\text{UO}_2$  powder lot were blended to achieve an enrichment of 34 weight percent  $^{235}\text{U}$  in U. Surface area for the 93% enriched  $\text{UO}_2$  powder was  $5.2 \text{ m}^2/\text{gram}$ . The particle size distribution for the 93% enriched powder produced by Oak Ridge National Laboratory's enrichment process is shown in Figure 28.

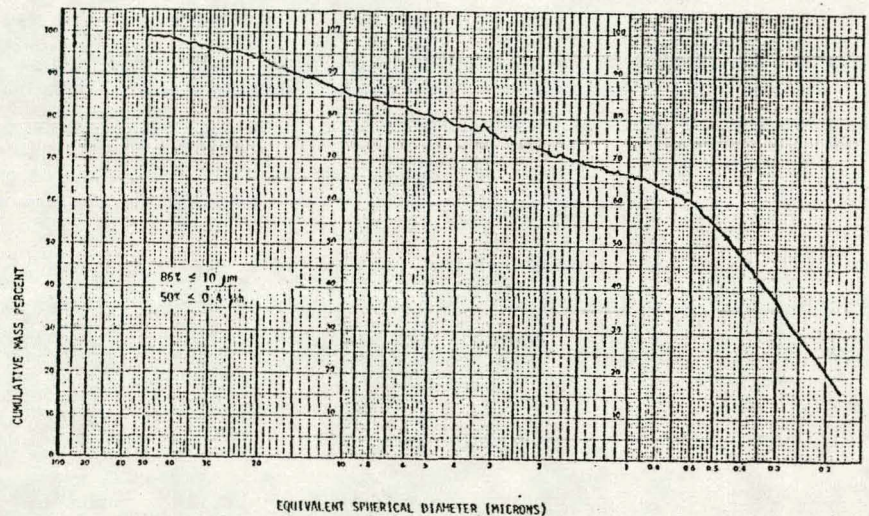


FIGURE 28. Particle Size Distribution of  $\text{UO}_2$  Powder Lot UE-080 Enriched 93%  $^{235}\text{U}$  in U

PROCESS DEVELOPMENT

A number of  $\text{UO}_2$  fuel batches enriched to various  $^{235}\text{U}$  contents were made at the Hanford Engineering development Laboratory during the early 1970's; however, they were made using different feed powders and to pellet densities in the range of 92-94%. Hence, some process development was required to establish fabrication process parameters for the 88.6% dense fuel designed for this test. The dry Sterotex organic additive powder processing described earlier was used because of the known pore-forming ability of Sterotex. Test pellets were fabricated using five levels of organic additive concentration (ranging from 12,700 psi (6.4 tsi) to 29,500 psi (14.8 tsi)) in combination with three final pellet compaction pressures (ranging from 19,700 psi (9.8 tsi) to 39,500 psi (19.8 tsi)). Pellets with low binder content sintered to relatively high densities (94% TD) and pellets with high binder content exhibited low densities (80% TD). Results are presented in Figures 29-32. Preslugging compaction pressure was lower than final pressure in most instances and therefore, had little effect on pellet density as shown in Figure 30. Pellet density increased as expected with increasing final pellet pressing pressures as shown in Figures 31-32. The density specification was within this range so the  $\text{UO}_2$  fuel enriched 34% in  $^{235}\text{U}$  in U was fabricated by the dry Sterotex binder addition technique.



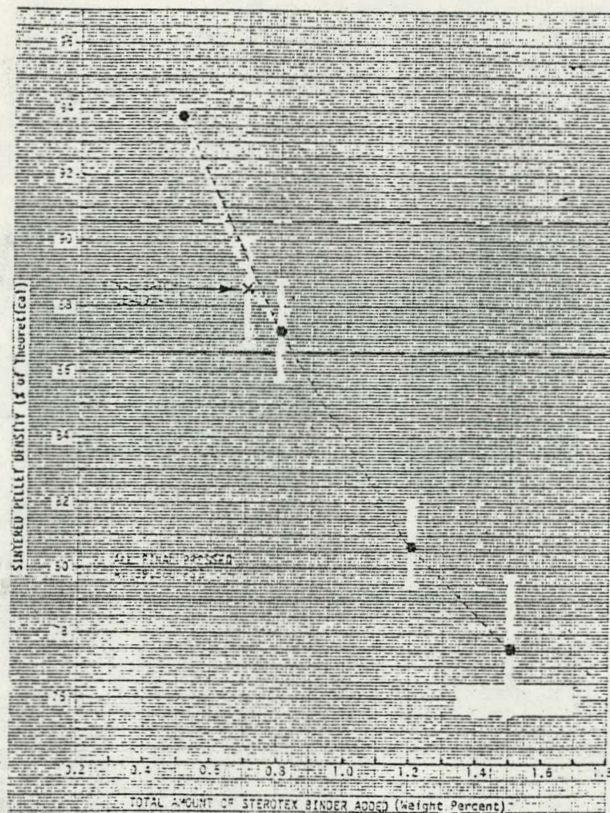


FIGURE 29. Sintered Pellet Density As A Function of Total Sterotex Organic Additive Concentration for  $UO_2$  Fuel Pellets Containing 34 w/o  $^{235}U$  in  $U$

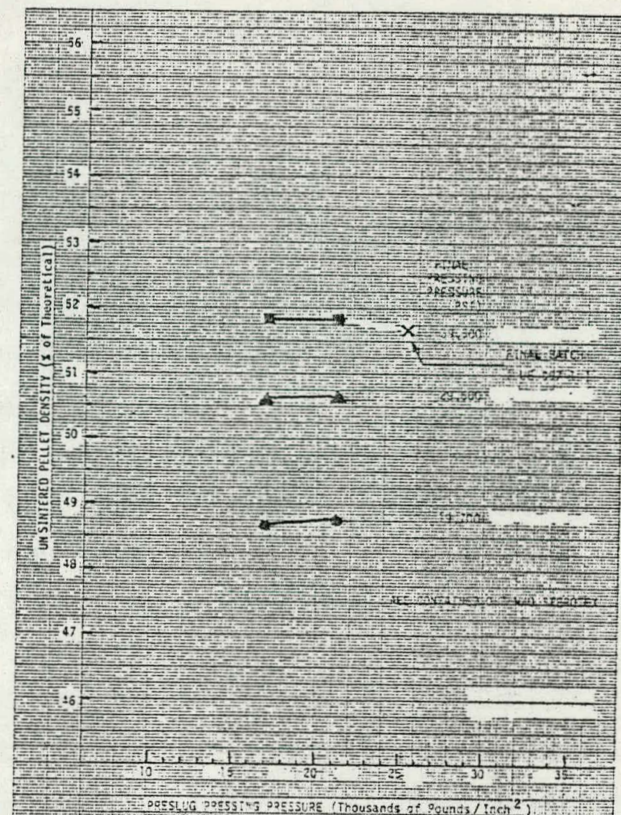


FIGURE 30. Unsintered Pellet Density As A Function of Pressing Compaction Pressure for  $UO_2$  Fuel Containing 34 Weight Percent  $^{235}U$  in  $U$

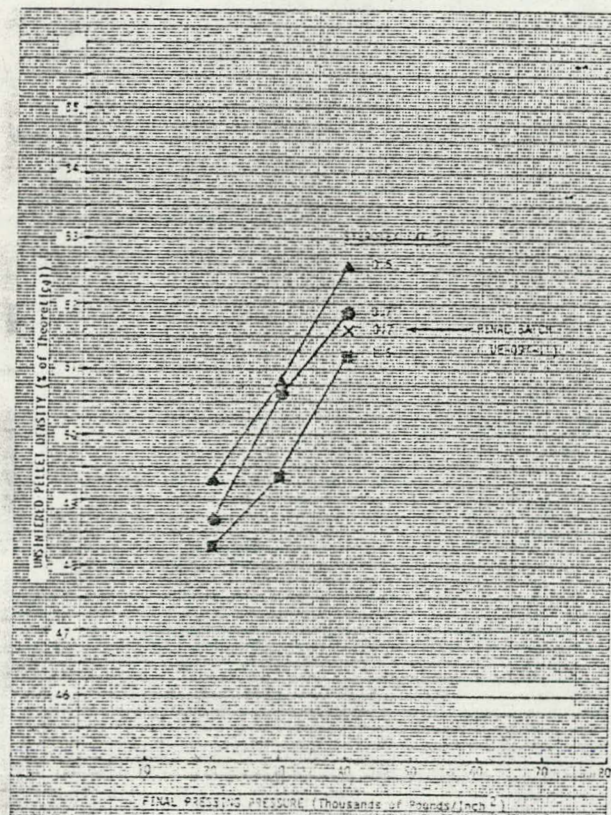


FIGURE 31. Unsintered Pellet Density As A Function of Final Pressing Pressure for  $UO_2$  Fuel Containing 34 Weight Percent  $^{235}U$  in  $U$

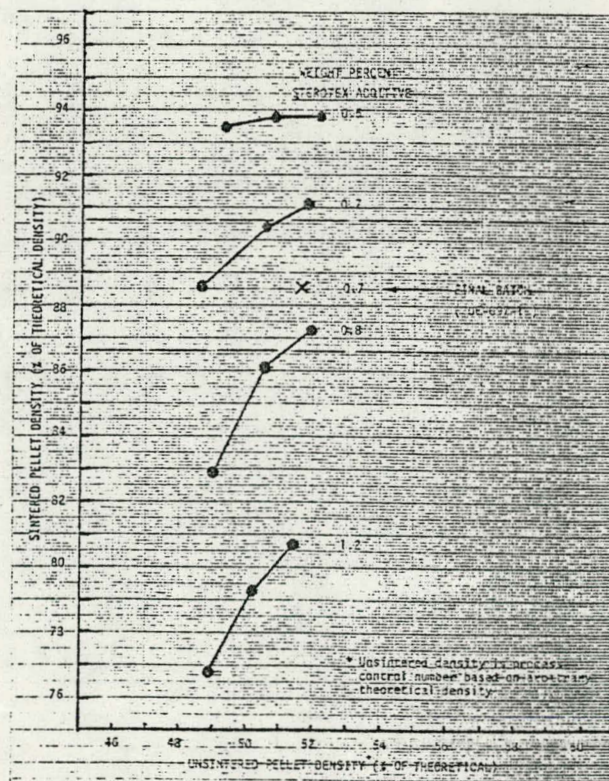


FIGURE 32. Sintered Pellet Density As A Function of Unsintered Pellet Density for  $UO_2$  Fuel Containing 34 Weight Percent  $^{235}U$  in  $U$



## FABRICATION PROCESS FLOWSHEET

The  $\text{UO}_2$  fuel containing 34%  $^{235}\text{U}$  in U was fabricated by essentially the same process described earlier for the  $\text{UO}_2$  fuel enriched with 21.85 weight percent  $^{233}\text{U}$  in U, except the  $^{235}\text{U}$  oxide fuel was vee-blended 15 minutes in a twin-shelled mechanical blender after 20 hours ballmilling and received the usual vacuum offgas treatment at  $850^\circ\text{C}$  for six hours. Sterotex binder additions were 0.3 weight percent (w/o) prior to pre-slugging and 0.4 w/o prior to final pressing, making a total of 0.7 w/o binder. Pellets were pre-slugged in an 0.500 inch diameter die at 25,300 psi (12.6 tsi) to a green density of 48.2% TD and were granulated through a U.S. Standard 20-mesh screen. Final pellet compaction was in a 0.327 inch diameter die at 39,500 psi (19.8 tsi) to a green density of 51.7% TD. Subsequent pellet processing was as described for the earlier  $\text{UO}_2$  fuel (see Figure 23). All pellets were centerless ground to approximately 0.2594 inch and were loaded into the same three types of fuel pin cladding (316 stainless steel, Inconel 706 and PE 16) described previously.

## FUEL PELLET CHARACTERISTICS

Finished pellet densities for the  $\text{UO}_2$  fuel enriched 34% in  $^{235}\text{U}$  in U are plotted and labeled in Figures 29-32. Sintered pellet density was 9.65 g/cc (88.5% TD) after centerless grinding. Fuel planar smear density (calculated as described earlier) was 9.18 g/cc (84.2% TD). Average sintered pellet diameter (before centerless grinding) was 0.2721 inch, which represented a sintering shrinkage of 17.5% based on unsintered pellet diameter. Average sintered pellet length was 0.3090 inch. Photomicrographs of typical fuel pellet polished cross sections are shown in Figures 33-34. The pores characteristic of Sterotex binder additions are evident as seen in Figure 33. A chemically etched cross section is shown in Figure 35. Microstructure characterization is also in progress on this  $\text{UO}_2$  fuel and will be discussed in a future report.



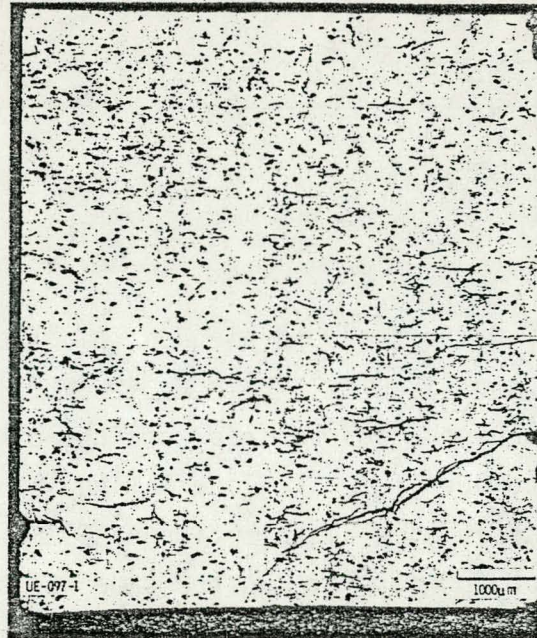


FIGURE 33. Photomicrograph of Polished Cross Section of UO<sub>2</sub> Fuel Pellet Containing 34 Weight Percent <sup>235</sup>U in U (Original 30x Magnification)

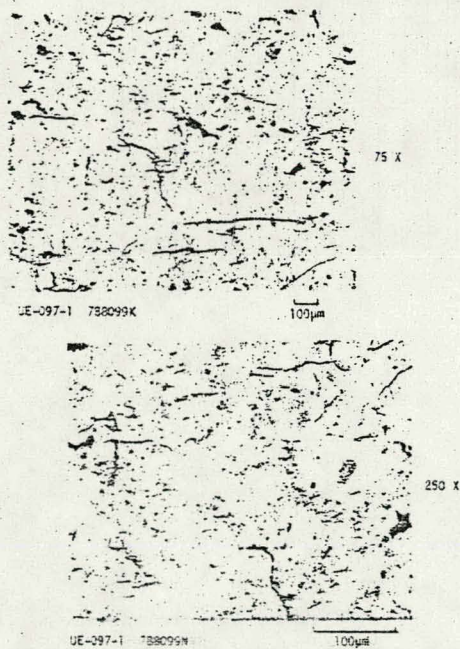


FIGURE 34. Photomicrographs of Polished Cross Section of UO<sub>2</sub> Fuel Pellet Containing 34 weight Percent <sup>235</sup>U in U (Originals 75x, 250x Magnifications)

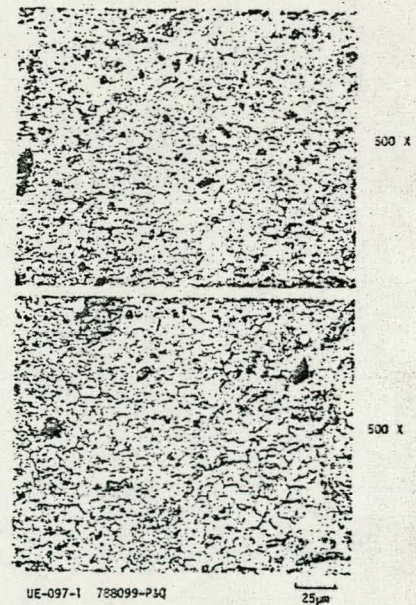


FIGURE 35. Photomicrographs of Chemically-Etched Cross Section of UO<sub>2</sub> Fuel Pellet Containing 34 weight percent <sup>235</sup>U in U (Originals 500x magnification)



## FABRICATION OF MIXED 15 w/o PuO<sub>2</sub>-85 w/o UO<sub>2</sub> FUEL

### FEED POWDERS

Two UO<sub>2</sub> powders and one PuO<sub>2</sub> powder were blended to achieve a PuO<sub>2</sub> content of 15 w/o and a <sup>235</sup>U enrichment of 15 w/o <sup>235</sup>U in U. One UO<sub>2</sub> powder was the natural UO<sub>2</sub> used in the previously discussed fuel batches, and the other was a different 93% enriched (<sup>235</sup>U in U) UO<sub>2</sub> powder from Oak Ridge National Laboratory with a surface area of 6.2 m<sup>2</sup>/gram. The particle size distribution of 93% enriched UO<sub>2</sub> powder lot is shown in Figure 36. The PuO<sub>2</sub> powder lot was the same one as used in the 35 w/o PuO<sub>2</sub>-65 w/o ThO<sub>2</sub> fuel batch discussed previously.

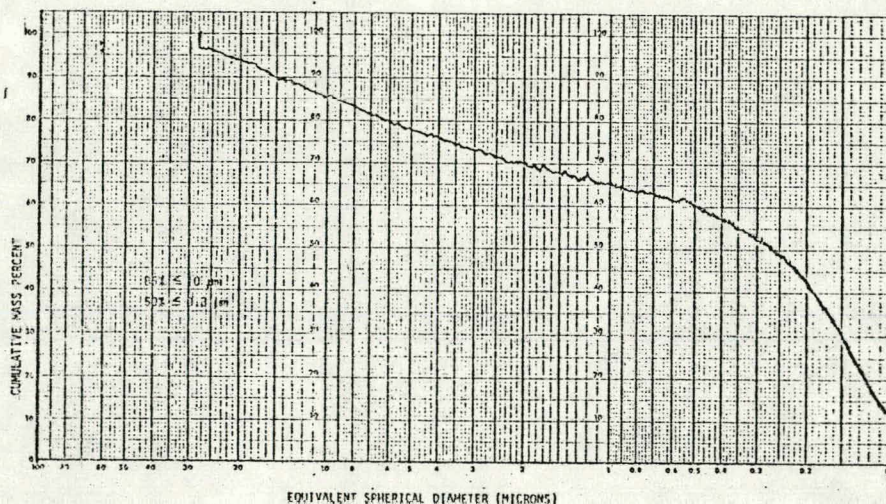


FIGURE 36. Particle Size Distribution of UO<sub>2</sub> Powder Lot UF-079 Enriched 93% <sup>235</sup>U in U

### PROCESS DEVELOPMENT

Even though the PuO<sub>2</sub> powder lot used for the 15 w/o PuO<sub>2</sub>-85 w/o UO<sub>2</sub> fuel batch had not been used previously for PuO<sub>2</sub>-UO<sub>2</sub> fuel fabrication, enough baseline experience existed from previous 15 w/o PuO<sub>2</sub>-85 w/o UO<sub>2</sub> fuel fabricated at HEDL, that one trial test batch was required to establish fabrication process parameters.

20



## FABRICATION PROCESS FLOWSHEET

The 15 w/o  $\text{PuO}_2$ -85  $\text{UO}_2$  fuel (containing 15 w/o  $^{235}\text{U}$  in U) was fabricated by basically the same dry Sterotex organic additive technique described previously for the  $\text{UO}_2$  fuel enriched with 34 w/o  $^{235}\text{U}$  in U. Sterotex binder additions were 0.75 w/o before preslugging and 0.4 w/o before final pressing, making a total of 1.15 w/o organic additive. Pellets were preslugged in a 0.500 inch diameter die at 20,000 spi (10.0 tsi) to a green pellet density of 46.7% TD and were granulated through a standard 20-mesh screen. Final pellet compaction was in a 0.316 inch diameter die at 38,500 psi (19.2 tsi) to a green pellet density of 51.6% TD. Subsequent pellet processing was the same as described for the  $\text{UO}_2$  fuel enriched 34 w/o in  $^{235}\text{U}$  in U (See Figure 23). All pellets were centerless ground to approximately 0.2606 inch diameter and were loaded into the same three fuel pin cladding types (316 stainless steel, Inconel 706 and PE 16).

## FUEL PELLET CHARACTERISTICS

Sintered pellet density for the 15 w/o  $\text{PuO}_2$ -85 w/o  $\text{UO}_2$  after centerless grinding was 9.80 g/cc (89.9% TD). Fuel planar smear density was 9.41 g/cc (86.3% TD). Average sintered pellet diameter (before centerless grinding) was 0.2624 inch, which represented a sintering shrinkage of 17.6% based on unsintered pellet diameter. Average sintered pellet length was 0.3109 inch. Photomicrographs of typical fuel pellet polished cross sections are shown in Figures 37-38. Once again, the relatively large pores formed by the Sterotex organic additive are evident, as seen in Figure 37. A chemically etched pellet cross section is shown in Figure 39. An alpha-autoradiograph of a typical pellet is shown in Figure 40, which shows that the Pu distribution is homogeneous and no large Pu-rich regions are visible. The light regions in Figure 40 are caused by pores and by the relatively higher amount of  $\text{UO}_2$  in this 15 w/o  $\text{PuO}_2$ -85 w/o  $\text{UO}_2$  fuel.

## ACKNOWLEDGEMENT

The following persons are acknowledged with thanks: R. D. Henry, for pellet processing and fabrication coordination; G. X. Beard, for data compilation and presentation; and Don Sybolt, for sample preparation and photomicroscopy.



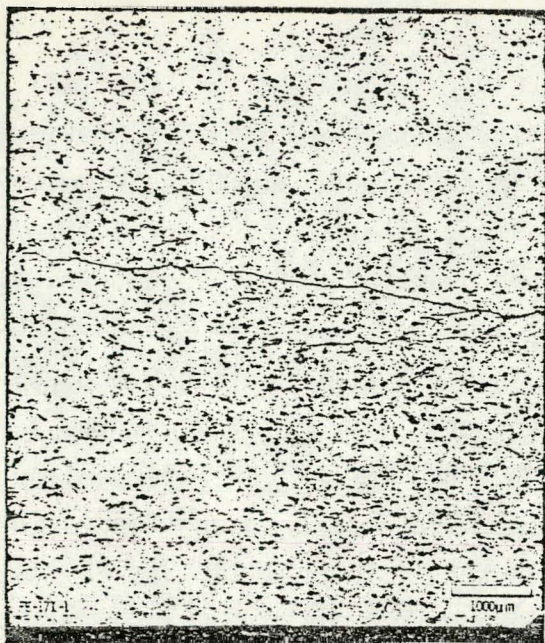
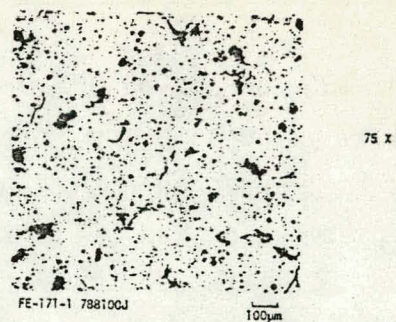
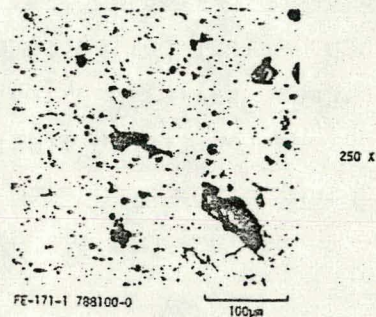


FIGURE 37. Photomicrograph of Polished Cross Section of 15 w/o  $\text{PuO}_2$ -85 w/o  $\text{UO}_2$  Fuel Pellet (Original 30x Magnification)



75 x



250 x

FIGURE 38. Photomicrographs of Polished Cross Section of 15 w/o  $\text{PuO}_2$ -85 w/o  $\text{UO}_2$  Fuel Pellet (Originals 75x, 250x magnifications)

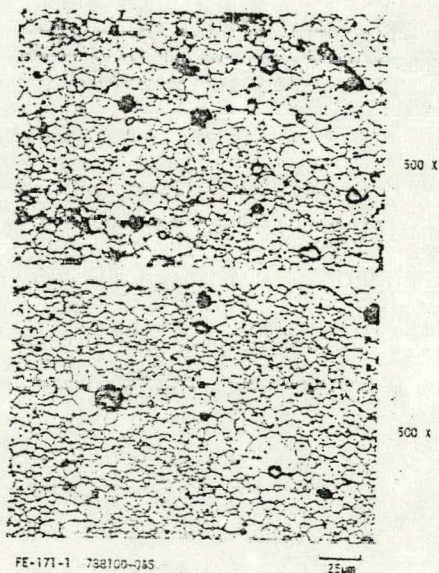


FIGURE 39. Photomicrographs of Chemically-Etched Cross Section of 15 w/o  $\text{PuO}_2$ -85 w/o  $\text{UO}_2$  Fuel Pellet (Originals 500x Magnification)

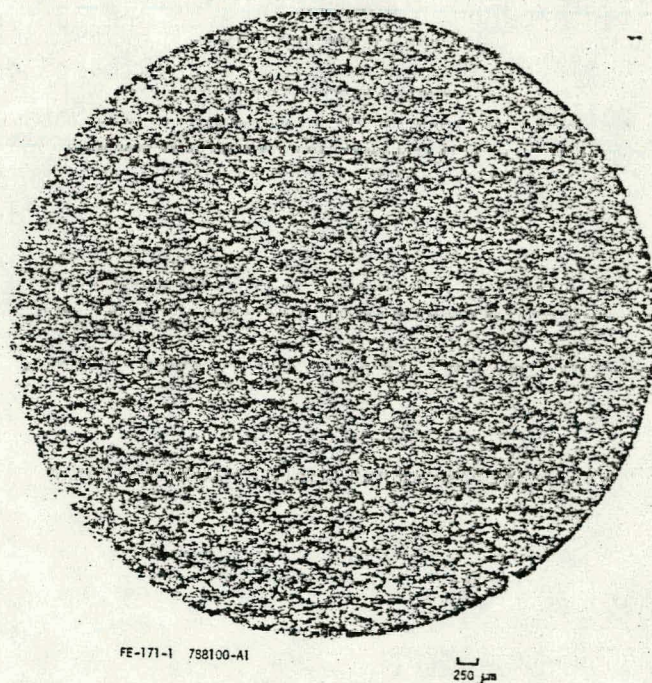


FIGURE 40. Alpha-Autoradiograph of Polished Cross Section of 15 w/o  $\text{PuO}_2$ -85 w/o  $\text{UO}_2$  Fuel Pellets (Original 30x magnification)



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