Progress Report
DEVELOPMENT
OF
PLUTONIUM-BEARING FUEL MATERIALS

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PROJECT AND FACILITY ADMINISTRATION

Task 1.00

K. H. Puechl

During this period, the major effort was concentrated on the preparation and characterization of fuel materials for the irradiation program. Three lots of PuO_2 were prepared, and these materials will subsequently be mechanically blended with UO_2 to provide UO_2 - PuO_2 fuels. In addition, to allow in-pile behavior comparison, six lots of UO_2 - PuO_2 of four different compositions were also prepared by coprecipitation. Analysis of powder characteristics indicates that all materials except the UO_2 -50 w/o PuO_2 coprecipitated lot are satisfactory for preparation of the irradiation specimens. The high plutonium content lot was not satisfactory due to excessive impurity pickup during hammermilling which resulted from the relatively hard particulate nature of this powder. Sintering trials on all materials verified sinterability as assessed by powder characterization. All materials, except the UO_2 -50 w/o PuO_2 composition sintered to greater than 92% of theoretical density under standard sintering conditions.

Since poor sinterability appears to be related primarily to impurity pickup, the investigation of the effect of controlled SiO_2 additions to coprecipitated UO_2 - PuO_2 powder has been continued. The results show that there is a significant reduction in sintered density over the range of SiO_2 concentration, 0 to 500 ppm; above this concentration the density is consistently low but does not decrease appreciably with increased impurity content. The low concentration range will therefore be further investigated.

It is planned to irradiate fuel samples containing reduced phases in order to assess the behavior of such materials relative to stoichiometric UO_2 - PuO_2 . To prepare suitable samples for this purpose, the oxidation-reduction behavior of PuO_2 and UO_2 - PuO_2 must be determined. Oxidation and reduction studies have therefore been initiated using a variety of furnace atmospheres, temperature and time conditions, and heat-up and cool-down cycles. The results to date are quite erratic but give positive indication that the results are due to complicated interaction of many effects; even the heat-up and cool-down conditions have definite influence.

To supplement these furnace investigations, the oxidation kinetics of UO_2 - PuO_2 powders and pellets are also being assessed using thermogravimetric methods. From these data, oxygen diffusion coefficients are being determined, and the variation of these coefficients with Pu/U ratio is being

ascertained. Data have been generated for pellets over the composition range UO_2 -0 w/o PuO_2 to UO_2 -35 w/o PuO_2 and for powders containing 0.5 and 5 w/o PuO_2 .

The effect of controlled SrO additions on the sinterability of PuO_2 has also been investigated. The data indicate that sinterability is improved with addition of less than 1 mole percent SrO_2 .

Detailed design of the high burnup irradiation capsules has been completed, and negotiations with GETR personnel have been initiated. The short-term irradiation specimens are still at the reactor site. Eleven of the fourteen capsules have been irradiated, but three capsules have been held out until reactor operation is scheduled to allow attainment of the required high flux levels. It is now anticipated that these irradiations will be performed in December 1963.

PREPARATION AND CHARACTERIZATION OF FUEL MATERIALS

Task 2.00

C. S. Caldwell

O. Menis

Preparation and Characterization of Plutonium Oxide
(J. Goodman, T. Potter)

During this period, a total of 0.66 kg of PuO_2 was prepared in three lots to provide feed material for the irradiation specimens. The preparation conditions are listed in Table 2.1, and the resulting powder properties are presented in Table 2.2. Particle size distributions for each of the three lots are shown in Figure 2.1, and these data indicate excellent reproducibility. These materials are currently being blended with UO_2 to prepare the mixed-oxide fuel materials required for irradiation.

Preparation and Characterization of Mixed Plutonium-Uranium Oxides
(J. Goodman, T. Potter)

Six lots of coprecipitated $\text{UO}_2\text{-PuO}_2$ for use in irradiation samples were prepared and characterized during this period. These included UO_2 (7.5% enriched)-0.5% PuO_2 , UO_2 (7.5% enriched)-5% PuO_2 , UO_2 -20% PuO_2 , and UO_2 -50% PuO_2 . Preparation conditions are given in Table 2.3, and powder data are presented in Table 2.4.

From Table 2.4 it is seen that the metallic impurities of the hammermilled Pu47 product are inordinately high. The feed solution and precursor were therefore also analyzed to determine the source of contamination. These data together with the product analysis are shown in Table 2.5. It is seen that the major contaminants are cobalt, chromium, and nickel and that these are not present in the precursor. It is, therefore, concluded that the contamination is due to hammermilling, wherein the relatively large, high density particles cause excessive wear on the hammers and screens. It is believed that this situation can be remedied by modifying the preparation procedure in the direction of more rapid precipitation and shorter digestion. This should result in the formation of softer particles.

Table 2.1

Conditions for PuO₂ Preparation
by Continuous Oxalate Process

Run Identification	297Pu42
Composition	PuO ₂
Precipitation Conditions	
Method	Continuous
Temperature, °C	35
Feed Composition	
gm Pu/liter	228.3
H ⁺ , molarity	3
Feed Flow Rate, ml/min	4.2
Precipitant Composition	
Oxalic Acid, molarity	1.0
H ₂ O ₂ , molarity	0.8
Precipitant Flow Rate, ml/min	10.8
Precipitator Average Holdup, min	30
Drying Temperature, °C	150
Drying Time, hr	16
Calcination Temperature, °C	760
Calcination Time, min	28
Charge Weight, gms:	
Lot I	198.5
Lot II	232.0
Lot III	232.2

Table 2.2

PuO₂ Powder Characterization Data

<u>Run Identification</u>	<u>Surface Area M²/gm</u>	<u>Bulk Density gm/cc</u>	<u>Tap Density gm/cc</u>	<u>Subsieve Size microns</u>	<u>Average MSA Particle Size for -200 Mesh Fraction microns</u>	<u>Total Metallic Impurity ppm</u>
297Pu42 - Lot I	8.66	1.45	2.59	1.60	2.5	450
297Pu42 - Lot II	3.99	1.72	2.92	1.70	1.9	375
297Pu42 - Lot III	3.69	1.71	2.97	1.90	2.5	320

Table 2.3A
Process Conditions
for Coprecipitated UO₂-PuO₂ Materials

Run Identification	297Pu41	297Pu43	297Pu44
Composition	UO ₂ -0.5 w/o PuO ₂	UO ₂ -50 w/o PuO ₂	UO ₂ -20 w/o PuO ₂
Precipitation Conditions			
Method	Continuous	Continuous	Continuous
Temperature, °C	55	55	55
Feed Composition			
gm Pu/liter	0.5	50.0	20.0
gm U/liter	99.5	50.0	80.0
H ⁺ , molarity	1.0	1.0	1.0
Feed Flow Rate, liters/hour	2.0	1.2	1.2
Precipitation Composition, NH ₃	Gas	Gas	Gas
Precipitant Flow Rate, cc/minute	350	375	285
	24.7 psia	24.7 psia	24.7 psia
Precipitation Average Holdup, minutes	30	30	30
Total Number Throughputs	14.5	6	14
Drying Temperature, °C	240	240	240
Drying Time, minutes	30	45	45
Gas Atmosphere	N ₂ -6% H ₂	N ₂ -6% H ₂	N ₂ -6% H ₂
Gas Flow, SCFH	9.5	6	12
Conversion Temperature, °C	900	840	840
Time at Temperature, minutes	105	80	702.3
Furnace Charge, grams	1171.7	398.8	803.6

Table 2.3B
Process Conditions
for Coprecipitated UO₂-PuO₂ Materials

Run Identification	297Pu45	297Pu46	297Pu47
Composition	UO ₂ *-5 w/o PuO ₂	UO ₂ *	UO ₂ -50 w/o PuO ₂
Precipitation Conditions			
Method	Continuous	Continuous	Continuous
Temperature, °C	55	55	55
Feed Composition			
gm Pu/liter	5.0	-	50
gm U/liter	95.0	150.0	50
H ⁺ , molarity	1.2	1.0	1.0
Feed Flow Rate, liters/hour		1.2	1.2
Precipitation Composition, NH ₃	Gas	Gas	Gas
Precipitant Flow Rate, cc/minute	325	500	375
	24.7 psia	24.7 psia	25.7 psia
Precipitation Average Holdup, minutes	30	30	30
Total Number Throughputs	6	8	5
Drying Temperature, °C	240	240	240
Drying Time, minutes	45	45	45
Gas Atmosphere	N ₂ -6% H ₂	N ₂ -6% H ₂	N ₂ -6% H ₂
Gas Flow, SCFH	6	12	12
Conversion Temperature, °C	840	720	740
Time at Temperature, minutes	80	80	80
Furnace Charge, grams	400.0	1026.4	314.1

Table 2.4

Summary of Powder Characteristics
for Coprecipitated UO₂-PuO₂ Materials

<u>Sample Description</u>	<u>Sample Number</u>	<u>PuO₂ Content w/o</u>	<u>Oxygen/Metal Ratio in Uranium Fraction</u>	<u>B.E.T. Surface Area M²/gm</u>	<u>Bulk Density gm/cc</u>	<u>Tap Density gm/cc</u>	<u>Air Permeability Particle Size microns</u>	<u>Average MSA Particle Size for -200 Mesh Fraction microns</u>	<u>Total Metallic Impurities ppm</u>
Hanford Pu(NO ₃) ₄ Feed Solution									
Pu(OH) ₄ -ADU Di Precursor	Pu41	0.5	2.03	0.70	1.64	3.14	1.5	2.7	1,650
	Pu43SS80-840	50	2.17	0.51	3.59	5.57	4.6	2.5	50,000
Hammermilled UO ₂ -50 w/o Pu	Pu44SS80-840	20	2.16	5.31	0.75	1.75	0.4	0.6	700
	Pu45SS80-840	5	2.08	5.24	0.88	1.93	0.4	0.4	500
	Pu46SS80-720	0	2.13	5.78	0.70	1.69	0.4	0.6	250
	Pu47SS80-740	50	2.19	1.66	3.78	5.36	5.3	-	4,500

PARTICLE SIZE DISTRIBUTION OF PuO_2

FIG. 2.1

Analytical Chemistry
(O. Menis, B. Conroy, J. Limpert)

During this period, the majority of effort was devoted to routine service analyses in support of materials preparation for the irradiation specimens.

The thermogravimetric equipment, described previously⁽ⁱ⁾, scheduled to be used for the determination of oxygen-to-metal ratios has been installed in a glove box, and this equipment is now ready to be committed to plutonium.

(i) NUMEC P-105, Progress Report, "Development of Plutonium-Bearing Fuel Materials", page 16.

FABRICATION AND EVALUATION OF FUEL SHAPES

Task 3.00

E. K. Halteman

I. D. Thomas

PuO₂ Sintering Studies
(M. D. Houston)

Since initial data⁽ⁱ⁾ indicated that strontium additions tended to increase the sinterability of PuO₂, a more detailed investigation was undertaken to observe the effect of SrO concentrations using PuO₂ produced by both the oxalate and peroxide routes. Compacts were prepared by blending PuO₂ and SrCO₃ powders and compacting at 40 tsi. Sintering was performed at 1600°C for one hour in both flowing O₂ and N₂-6% H₂ atmospheres. The results are presented in Table 3.1, and these are graphically illustrated in Figure 3.1. These data show that sinterability can be improved by small additions of SrO; the optimum concentration of SrO appears to be less than 1 mol/o.

Mixed Oxide Sintering Studies
(J. Brennan, M. D. Houston, J. Roth)

During this period, sintering trials were run on the materials to be utilized for the irradiation samples and sintering studies were continued to determine the effects of impurities and sintering atmosphere. Oxidation and reduction studies were also initiated to supplement the oxidation kinetics studies being performed by thermogravimetric analysis and to determine a means for controlling the reduced-phase composition of sintered pellets. Such pellets are desired for the irradiation program to assess the effects of the presence of reduced phases on irradiation stability and fission gas retention capability.

The various feed materials scheduled for utilization in irradiation specimens have been compacted and sintered. The coprecipitated materials Pu44, 45 and 46, which will be utilized as is, all sintered extremely well to final densities in excess of 95% of theoretical. The Eldorado UO₂ and the in-house prepared

(i) NUMEC P-105, Progress Report, "Development of Plutonium-Bearing Fuel Materials", pages 20-22.

Table 3.1

Effect of SrO Addition on Sintering Characteristics of PuO₂
Sintered at 1600°C for 1 Hour
Compaction Pressure - 40 TSI

SrO Content mol/o	Green Density gm/cc	Sintered in Flowing O ₂ Atmosphere				Sintered in N ₂ -6% H ₂ Atmosphere			
		Weight Loss on Firing %	Fired Density gm/cc	Percent Theoretical Pellet Density	Percent Theoretical PuO ₂ Density	Weight Loss on Firing %	Fired Density gm/cc	Percent Theoretical Pellet Density	Percent Theoretical PuO ₂ Density
Pu29-None	6.58	1.7	9.54	83.2	83.2	2.2	10.07	87.9	87.9
Pu29-1.29%	6.57	1.9	10.62	93.3	92.1	2.4	10.23	89.9	88.7
Pu30-None	6.73	2.4	10.51	91.7	91.7	2.7	10.28	89.7	89.7
Pu30-1.29%	6.72	3.2	10.95	96.2	95.0	-	-	-	-
Pu38-None	7.25	1.3	10.55	92.1	92.1	1.5	9.58	83.6	83.6
Pu38-0.4%	7.27	0.8	11.06	96.7	96.3	-	-	-	-
Pu38-0.8%	7.24	0.7	11.02	96.6	95.8	-	-	-	-
Pu38-1.29%	7.25	1.1	10.95	96.3	95.0	2.2	10.23	89.9	88.8
Pu38-2.55%	7.21	1.3	10.70	94.7	92.3	2.3	9.27	82.1	80.0
Pu38-3.78%	7.03	1.4	10.57	94.2	90.7	2.3	9.77	87.1	83.8
Pu38-4.97%	6.96	1.5	10.33	92.7	88.1	2.2	9.41	84.5	80.3

Pu29 - Peroxide Route
 Pu30 - Peroxide Route
 Pu38 - Oxalate Route

EFFECT OF SrO ADDITION ON SINTERED PROPERTIES OF PuO_2
(SINTERED AT 1600°C FOR 1 HOUR IN O_2)
Pu 38 PREPARATION

Fig. 3.1

UO₂(Pu46) which will be mechanically blended with PuO₂ for the preparation of irradiation specimens, also sintered satisfactorily to greater than 91% of theoretical density. The PuO₂(Pu38), which will be blended with the UO₂, exhibited sintering behavior similar to previous preparations; final density of greater than 83% was achieved in a reducing atmosphere and greater than 92% of theoretical in oxygen. This material, therefore, also appears to be satisfactory for the preparation of irradiation specimens when blended with UO₂. Coprecipitated UO₂-50 w/o PuO₂, Pu43 and 47, which was scheduled to be blended with UO₂ to provide material intermediate between completely coprecipitated and completely mechanically blended, showed poor sintering behavior. This behavior is probably due to impurity pickup during hammer-milling as discussed previously. The results of the sintering trials are given in Table 3.2.

Since poor sinterability of mechanically blended UO₂-PuO₂ appears to be related to high impurity content, especially SiO₂ pickup during ball milling, an investigation was continued using controlled SiO₂ additions. For these studies a coprecipitated powder, Pu24 (UO₂-12.5 w/o PuO₂) was blended with powdered quartz. The sintering results presented in Table 3.3 show that the sintered density decreases with increasing SrO₂ content. The most significant density degradation occurs in the range 0-500 ppm SrO₂, and this concentration range will be examined in greater detail to allow appropriate specification limits to be placed on this impurity.

For the irradiation program, it is planned to explore the behavior of UO₂-PuO₂ materials containing reduced phases in order to allow specification of stoichiometry for reactor fuels. Besides resulting in reduced initial pellet density, it is assumed that the presence of such phases will have deleterious effects since the crystal structure will not be of the fluorite type which offers the greatest resistance to radiation. Oxidation and reduction studies have therefore been initiated to allow preparation of these particular irradiation specimens in a reproducible and meaningful manner. The formation of reduced phases in the component PuO₂ was investigated as was the formation of such phases in coprecipitated UO₂-PuO₂.

For the preparation of reduced phases in PuO₂, carbon was mixed with the plutonium oxide in such proportion as to satisfy the equation, $2\text{PuO}_2 + \text{C} \rightarrow \text{Pu}_2\text{O}_3 + \text{CO}$. The mixture was placed in a tantalum cup and heated to 1550°C in vacuum (6.6×10^{-4} mm). A weight loss during firing of 4.67% was observed compared to the theoretically anticipated value of 5.05% if the starting material had been stoichiometric PuO₂. Reoxidation of this product in oxygen at 800°C indicated an O/Pu ratio of 1.53. The reduced material, PuO_{1.53}, was compacted at 10 tsi into pellets, and these were fired at 1600°C in an N₂-6% H₂ atmosphere for

Table 3.2

Sintering Characteristics
of Irradiation Specimen Feed Materials
 (Sintered at 1600°C for 1 Hour)

<u>Composition</u>	<u>Pu Content w/o</u>	<u>Compaction Pressure TSI</u>	<u>Green Density gm/cc</u>	<u>Sintering Atmosphere</u>	<u>Firing Weight Loss %</u>	<u>Fired Density gm/cc</u>	<u>% Theoretical Density</u>
Eldorado UO ₂	0	10	4.72	N ₂ -6% H ₂	1.90	10.05	91.7
Pu46 (UO ₂)	0	10	5.06	N ₂ -6% H ₂	1.50	10.50	95.8
Pu45SS80-840	5	10	5.76	N ₂ -6% H ₂	0.90	10.63	96.5
Pu44SS80-840	20	10	5.62	N ₂ -6% H ₂	1.10	10.74	97.1
Pu44SS80-840	20	20	6.23	N ₂ -6% H ₂	1.10	10.75	97.2
Pu43SS80-840	50	40	8.01	N ₂ -6% H ₂	1.56	9.43	84.1
Pu47SS80-840	50	10	7.02	N ₂ -6% H ₂	1.80	8.17	72.9
Pu38	100	10	5.89	N ₂ -6% H ₂	1.20	10.04	87.6
Pu38	100	40	7.25	N ₂ -6% H ₂	1.50	9.58	83.6
Pu38	100	40	7.24	O ₂	1.30	10.55	92.1

Table 3.3

Effect of SiO₂ Impurity
on the Sintering Behavior of UO₂-PuO₂
 (Sintered at 1600°C for 1 Hour in N₂-6% H₂)

<u>Silica Content ppm</u>	<u>Compaction Pressure TSI</u>	<u>Green Density gm/cc</u>	<u>Weight Loss on Firing %</u>	<u>Fired Density gm/cc</u>	<u>% Theoretical Density</u>
0	10	4.99	1.6	10.56	95.8
0	20	5.71	1.6	10.49	95.2
500	20	5.78	2.0	9.88	89.7
2,000	20	5.83	2.1	9.74	88.4
10,000	20	5.82	2.2	9.64	87.5

periods ranging from 10 to 90 minutes. The results of such firing, and in some cases refiring in oxygen, are presented in Table 3.4. In every instance, a considerable weight gain was observed, even though the atmosphere, N_2 -6% H_2 , had been assumed to be a reducing medium. These data confirm previous results that small quantities of moisture in this atmosphere can cause considerable oxidation. In general, the weight gains indicated partial reoxidation of approximately 50%. X-ray diffraction analysis prior to firing in N_2 -6% H_2 indicated that the $PuO_{1.53}$ was composed primarily of β - Pu_2O_3 with some residual PuO_2 . After firing in N_2 -6% H_2 , larger amounts of PuO_2 were generally observed although the ratio of PuO_2 to β - Pu_2O_3 did not appear to be dependent upon the firing time. This is also indicated by the erratic weight-gain data presented in Table 3.4. α - Pu_2O_3 was detected only in the sample that was fired for 60 minutes. Following x-ray examination, some samples were refired in oxygen at $800^\circ C$ to determine if any additional oxidation had occurred during storage in air at room temperature. The results indicate that some additional oxidation had occurred in this manner.

Reduction-oxidation studies were also carried out for the full range of UO_2 - PuO_2 compositions using both coprecipitated and mechanically blended materials. The powders were compacted into pellets and were sintered at $1600^\circ C$ for one hour using three different atmospheres; namely, (1) flowing N_2 -6% H_2 gettered over calcium chips and dried through a calcium sulfate column, (2) flowing N_2 -6% H_2 bubbled through water for pickup of 1 cc of water per cubic foot, and (3) vacuum at 10^{-4} mm of Hg. Subsequent to sintering, the pellets were comminuted and reoxidized in O_2 at $800^\circ C$ for one hour. Oxygen pickup during comminution was checked and found to be negligible. Initial results from this investigation are presented in Table 3.5, and data from subsequent runs are presented graphically in Figure 3.2. In general, there is appreciable scatter in the data and no significant trends are discernable. From these data, it became apparent that a more thorough investigation is required to yield meaningful and interpretable results.

For two selected compositions, chosen on the basis of low, medium, and high weight gains, compositions were determined by x-ray diffraction following oxidation at $800^\circ C$ for one hour in O_2 . The results are presented in Table 3.6. For the UO_2 -5 w/o PuO_2 composition prepared by coprecipitation, only the $(U,Pu)_3O_8$ pattern was detected following oxidation. In the mechanically blended material of the same composition, the $(U,Pu)O_2$ phase was also observed. The weight gain observed with the coprecipitated material is essentially that required to convert all UO_2 to U_3O_8 (3.75%). For the UO_2 -20 w/o PuO_2 composition both $(U,Pu)_3O_8$ and $(U,Pu)O_2$ phases were detected in both the coprecipitated and mechanically blended samples, and the weight gain on oxidation can be readily correlated to the amount of $(U,Pu)_3O_8$ phase observed.

Table 3.4

Weight Gain of $\text{PuO}_{1.53}$
Due to Firing in N_2 -6% H_2 at 1600°C

Firing Time minutes	Weight Gain, %			X-Ray Diffraction Results (After Firing in N_2 -6% H_2)
	N_2 -6% H_2	O_2	Total	
0	-	2.84	2.84	β - Pu_2O_3 + PuO_2
10	1.35	-	-	β - Pu_2O_3 + PuO_2
20	1.48	1.25	2.73	β - Pu_2O_3 + PuO_2
30	2.13	-	-	β - Pu_2O_3 + PuO_2
45	1.30	-	-	β - Pu_2O_3 + PuO_2
60	1.48	1.33	2.81	α - Pu_2O_3 + PuO_2
90	1.82	0.95	2.77	β - Pu_2O_3 + PuO_2

Table 3.5
Percent Weight Gain on Oxidation
for UO_2 - PuO_2 Materials Sintered under Various Conditions
Oxidation at 800°C in O_2 for 1 Hour

<u>Composition</u>	<u>Sintered in N_2-6% H_2</u> <u>(Gettered)</u>		<u>Sintered in N_2-6% H_2</u> <u>(H_2O Added)</u>		<u>Sintered</u> <u>in Vacuum</u> <u>10⁻⁴ mm Hg</u>	<u>Theoretical⁽²⁾</u> <u>Weight Gain</u> <u>$\text{U}_3\text{O}_8 + \text{PuO}_2$</u>
	<u>1 Hour</u>	<u>16 Hours</u>	<u>1 Hour</u>	<u>16 Hours</u>	<u>1 Hour</u>	
UO_2	3.91	-	3.89	-	3.45	3.95
UO_2 -5% PuO_2 (cp) ⁽¹⁾	3.85	3.32	3.67	3.78	3.60	3.75
UO_2 -5% PuO_2 (mb)	2.94/3.71 ⁽³⁾	3.47	3.56	3.64	3.55	3.75
UO_2 -12.5% PuO_2 (cp)	3.30	-	-	-	-	3.46
UO_2 -20% PuO_2 (cp)	2.76/2.81	2.91/2.67	2.83	2.47	2.75	3.15
UO_2 -20% PuO_2 (mb)	3.41/3.02	2.71/3.05	2.90	2.91	2.97	3.15
UO_2 -35% PuO_2 (cp)	1.91	1.88	-	-	-	2.57
PuO_2	0.54	-	0.43	-	0.19	0

(1) cp = coprecipitated; mb = mechanical blended

(2) Based on no reduction of PuO_2 to α - Pu_2O_3

(3) Second values represent results for repeat experiment.

Table 3.6

Oxidation Data for $\text{UO}_2\text{-PuO}_2$
Materials Sintered Under Various Conditions
Oxidization at 800°C in O_2 for 1 Hour

<u>Starting Material</u>	<u>Sintering Conditions</u>	<u>Weight Gain on Oxidation %</u>	<u>Phases Present by X-Ray</u>	<u>Relative Ratio $(\text{U,Pu})_3\text{O}_8:(\text{U,Pu})\text{O}_2$</u>
$\text{UO}_2\text{-5 w/o PuO}_2(\text{cp})^{(1)}$	16 hrs-Wet	3.78	$(\text{U,Pu})_3\text{O}_8$	-
$\text{UO}_2\text{-5 w/o PuO}_2(\text{cp})$	1 hr-Vac	3.60	$(\text{U,Pu})_3\text{O}_8$	-
$\text{UO}_2\text{-5 w/o PuO}_2(\text{mb})$	1 hr-Dry	2.94	$(\text{U,Pu})_2 + (\text{U,Pu})\text{O}_2$	7.56
$\text{UO}_2\text{-20 w/o PuO}_2(\text{cp})$	16 hr-Wet	2.47	$(\text{U,Pu})_3\text{O}_8 + (\text{U,Pu})\text{O}_2$	0.60
$\text{UO}_2\text{-20 w/o PuO}_2(\text{cp})$	1 hr-Wet	2.83	$(\text{U,Pu})_3\text{O}_8 + (\text{U,Pu})\text{O}_2$	1.10
$\text{UO}_2\text{-20 w/o PuO}_2(\text{mb})$	1 hr-Dry	3.41	$(\text{U,Pu})_3\text{O}_8 + (\text{U,Pu})\text{O}_2$	1.20

(1) cp = coprecipitated; mb = mechanical blended

(2) Relative Intensity of $\frac{I_{(101)}(\text{U,Pu})_3\text{O}_8}{I_{(200)}(\text{U,Pu})\text{O}_2}$

With the UO_2 -35 w/o PuO_2 composition, further oxidation studies were performed to determine the effects of temperature and time-at-temperature. The results of oxygen pickup as a function of time-at-temperature are given in Table 3.7. All weight gains are lower than expected and, with two exceptions, fall in the range of 1.84 to 1.93% (theoretical 2.57%). The starting, sintered and oxidized materials were examined by x-ray diffraction, and a U_3O_8 phase was observed only in the oxidized raw batch. Unit cell parameters were calculated by the Nelson-Riley method, and these dimensions are given in Table 3.8. The raw batch material with an obvious O/M ratio greater than 2.00 has a smaller cell length than theoretical while the reduced materials approach this value quite closely. Upon oxidation, the unit cell contracts considerably, analogous to the contraction of $\text{UO}_{2.00}$, when oxidized to $\text{UO}_{2.20}$, which can be explained by the incorporation of excess oxygen with the formation of U^{+6} . The higher valence cation is not only smaller in actual size but also has a stronger binding force. At this time, no significance is given to the differences in the reoxidized lattice constants as a function of reduction time. The relationship between weight gain and oxidation temperature is shown in Figure 3.3. A minimum temperature of 600°C was chosen since at temperatures below this value, the UO_3 phase is considered the most stable, and in oxidation trials at temperatures in the range 400 - 550°C the actual weight gain was always greater than the value calculated for the U_3O_8 end product. As seen in the Figure, the oxygen pickup varied between 1.80 and 1.97% up to 1200°C , then decreased at 1400°C . The results of x-ray diffraction examination revealed a slight amount of U_3O_8 phase after oxidation at 1000 and 1200°C but none at the lower or higher temperatures, only $(\text{U,Pu})\text{O}_2 + x$. The behavior of the oxidized material at the high end of the temperature range is again analogous to pure UO_2 which oxidizes to U_3O_8 , and this in turn undergoes self-reduction at temperatures of about 1400°C . For example, sintered UO_2 was oxidized at 800 and 1400°C for one hour, and weight gains of 3.95% (all U_3O_8) and 3.57%, respectively, were observed. The weight losses due to self-reduction observed in both materials, UO_2 -35% PuO_2 and UO_2 , at the higher temperature are about equal, 0.40-0.50%.

Since the reduction-oxidation results were not always consistent, various aspects of the oxidation process were investigated. For expediency, most of the oxidation runs described above were performed by placing the crushed materials into platinum boats and then inserting the boats into a hot furnace. After oxidation, the boats were removed from the furnace and allowed to cool under ambient conditions prior to weighing. With this "in cool-out hot" process, good reproducibility was achieved; however, it was suspected that the degree of oxygen pickup may be due to the heating and cooling cycle since excessive oxygen pickup has been

Table 3.7

Percent Weight Gain vs Oxidation Time
for UO₂-35% PuO₂ Sintered in N₂-6% H₂
Oxidation at 600°C in O₂
 (Theoretical Weight Gain = 2.57%)

<u>Material</u>	<u>10 min</u>	<u>20 min</u>	<u>30 min</u>	<u>45 min</u>	<u>60 min</u>	<u>90 min</u>
Raw Batch	-	-	-	-	1.78	-
Sintered 1 Hour	1.84	2.65	1.92	1.93	1.91	1.91
Sintered 16 Hours	2.46	1.85	1.91	1.89	1.88	1.89

Table 3.8

Oxidation-Reduction Properties
of UO₂-35 % PuO₂
Reduced at 1600°C in N₂-6% H₂
Oxidized at 600°C in O₂

<u>Reduction Time</u> <u>hours</u>	<u>Oxidation Time</u> <u>minutes</u>	<u>Weight Gain</u> <u>on Oxidation</u> <u>%</u>	<u>Lattice Constant</u> <u>(Theoretical = 5.4435)</u>
0	-	-	5.4399
1	-	-	5.4455
16	-	-	5.4438
1	10	1.84	5.4243
1	90	1.91	5.4219
16	10	2.46	5.4199
16	90	1.89	5.4196

**WEIGHT GAIN ON OXIDATION vs. OXIDATION TEMPERATURE
FOR UO_2 -35% PuO_2 SINTERED AT 1600°C IN P_2 -6% H_2**

UMEC P-106

Fig. 3.3

observed by other investigators working with UO_2 in the temperature range 100-500°C. UO_2 -20 w/o PuO_2 was therefore utilized under different heat-up and cool-down conditions to ascertain the dependence of oxygen pickup on these process variables. The results are presented in Table 3.9 for weight gain on oxidation as well as weight loss on subsequent reduction. From these data oxygen-to-metal ratios were calculated, and the values ranged from 1.929 for the in-hot, out-hot process to 1.983 for the in-cool, out-hot process. Since the oxidation-reduction procedure is utilized as a general method for determining the oxygen-to-metal ratio of UO_2 - PuO_2 materials, these data are most significant and shed doubt on the validity of reported O/M data.

Oxidation Kinetics of UO_2 - PuO_2
(R. W. Gerrish)

To aid in the interpretation of the gross oxidation-reduction data presented previously, studies of the oxidation kinetics of coprecipitated UO_2 - PuO_2 powders with surface areas of 0.7 M^2/gm to 7.0 M^2/gm and of sintered pellets are continuing using thermogravimetric methods.

Re-evaluation of the preliminary data reported previously⁽ⁱ⁾ for the oxidation of the UO_2 powder was also performed. From such re-evaluation, the diffusion equation was found to be

$$D = 2.12 \times 10^{-3} \exp \left[(- 25,380 \pm 1,900) / RT \right]$$

The determined activation energy is still in close agreement with values reported by Anderson and Alberman⁽ⁱⁱ⁾ (27 Kcal/mole) and the values reported by Aronson, et al,⁽ⁱⁱⁱ⁾ (26.3 Kcal/mole), although it is higher than the values reported by Smith^(iv) (21.9 Kcal/mole), and by Blackburn et al,^(v) (21.7 Kcal/mole). The UO_2 powder that was studied was slightly

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- (i) NUMEC P-104, Progress Report, "Development of Plutonium-Bearing Fuel Materials", page 40.
 - (ii) K. B. Alberman and J. S. Anderson, J. Chem. Soc. (London), Supplement 5303, 1949.
 - (iii) S. Aronson, R. B. Roof, Jr., and J. Belle, "Kinetic Study of the Oxidation of Uranium Dioxide", J. Chem. Physics, 27, 137, 1957.
 - (iv) T. Smith, "Kinetics and Mechanism of the Oxidation of UO_2 and UO_2 Plus Fissia Sintered Pellets", NAA-SR-4677, 1960.
 - (v) P. E. Blackburn, J. Weissbart, and E. A. Gulbranson, "Oxidation of UO_2 ", Westinghouse Laboratories Report 100FF 942-R2.

Table 3.9

Effect of Oxidation-Reduction Process Parameters
on Measured Weight Changes for UO₂-20% PuO₂

Sintered at 1600°C for 1 Hour in N₂-6% H₂
 Oxidized at 800°C for 1 Hour in O₂
 Reduced at 700°C for 1 Hour in N₂-6% H₂
 (Theoretical Weight Gain in O₂ = 3.15%)

<u>Oxidation Procedure</u>	<u>Weight Gain on Oxidation %</u>	<u>Weight Loss on Reduction %</u>	<u>Calculated O/M</u>
In - cool; Out - hot	2.55	2.45	1.983
In - hot; Out - hot	2.18	1.75	1.929
In - hot; Out - cool	2.30	2.20	1.983
In - cool; Out - cool	3.47	3.27	1.968

nonstoichiometric, with an oxygen-to-metal ratio of 2.04, which could explain the somewhat high value for the activation energy.

A limited number of sintered pellets of the compositions UO_2 -5 w/o PuO_2 , UO_2 -12.5 w/o PuO_2 , UO_2 -20 w/o PuO_2 , and UO_2 -35 w/o PuO_2 were oxidized in the temperature range of 400 to 500°C. A summary of data is presented in Table 3.10. Diffusion equations derived from these data are tabulated in Table 3.11. For the UO_2 -12.5 w/o PuO_2 composition (Pu20), pellets were prepared from powder that was hammermilled as well as from non-milled powder. The surface area of the starting hammermilled powder was 6.1 M^2/gm ; the surface area of the final oxidized powder was 1.22 M^2/gm . The oxygen diffusion coefficient was calculated using a particle radius of 2.6×10^{-5} cm as derived from this latter surface area determination. The apparent activation energy for this composition is quite low, and the data are being reanalyzed, although this low energy could be explained by the presence of UO_3 in the oxidized material. X-ray analysis indicates that UO_3 may be present in addition to a considerable amount of U_3O_8 . For the UO_2 -20 w/o PuO_2 composition (CP20), x-ray analysis of the powder formed by oxidation at 480°C for 11 hours indicates the presence of a disproportionate amount of PuO_2 which is probably due to incomplete conversion since the resulting powder was extremely coarse. For the UO_2 -35 w/o PuO_2 composition (Pu19), the gross nonstoichiometry obscured the initial reaction to U_4O_9 .

A typical series of oxidation curves are shown in Figure 3.4 which shows the oxygen pickup as a function of time for the oxidation of the UO_2 -5 w/o PuO_2 (Pu14A) sintered pellets. The rate of diffusion of oxygen through a barrier layer of U_3O_7 or U_4O_9 is shown for the reaction of 407°C prior to the time when the secondary reaction $\text{U}_4\text{O}_9 \rightarrow \text{U}_3\text{O}_8$ becomes predominant. This secondary reaction is a complex mechanism of nucleus formation and growth of U_3O_8 with the behavior being described by a sigmoidal rate curve. The sigmoidal rate is better shown for the oxidation reaction at the higher temperatures of 500 and 502°C. On these curves are shown an initial period of accelerating growth, a period of linear growth until completion is nearly reached and, finally, an extended period during which the reaction slows down. Subsequent x-ray analysis of the pellet oxidized at 502°C showed that the oxidation of the single phase material had gone completely to U_3O_8 and PuO_2 with U_4O_9 , UO_3 , or any single phase $(\text{UPu})\text{O}_2$, being completely absent or present to less than 5%, the limit of detection. The linear portions of these oxygen pickup curves were fitted with "a least squares fit" to determine the rate of growth of the U_3O_8 phase. The data from the initial reaction where the diffusion process is predominant was used to calculate the diffusion coefficient of oxygen.

Table 3.10
Sintered Pellet Oxidation Data

<u>Sample</u>	<u>Composition</u>	<u>Pellet Density % Theoretical</u>	<u>Initial Oxygen-to-Metal Ratio of Uranium Fraction</u>	<u>Reaction Temperature °C</u>	<u>UO₂ → U₄O₉ Diffusion Coefficient cm²/sec x 10¹⁶</u>	<u>U₄O₉ → U₃O₈ Oxidation Rate mg/min/gm</u>
Pu14A	UO ₂ -5 w/o PuO ₂	96	1.98	500	.190	.444
Pu14A	UO ₂ -5 w/o PuO ₂	96	1.98	502	.172	.437
Pu14A	UO ₂ -5 w/o PuO ₂	96	1.98	407	.073	.0176
Pu20HM	UO ₂ -12.5 w/o PuO ₂	95	2.07	472	3.7	.161
Pu20HM	UO ₂ -12.5 w/o PuO ₂	95	2.07	432	1.4	.055
Pu20HM	UO ₂ -12.5 w/o PuO ₂	95	2.07	451	2.2	.057
Pu20NM	UO ₂ -12.5 w/o PuO ₂	95	1.98	500	132.0	.320
Pu20NM	UO ₂ -12.5 w/o PuO ₂	96	1.98	460	72.0	.177
Pu20NM	UO ₂ -12.5 w/o PuO ₂	96	1.98	481	105.0	.212
Pu20NM	UO ₂ -12.5 w/o PuO ₂	96	1.98	441	44.0	.194
CP20	UO ₂ -20 w/o PuO ₂	94	2.01	420	5.92	.0372
CP20	UO ₂ -20 w/o PuO ₂	95	2.01	480	.95	.377
CP20	UO ₂ -20 w/o PuO ₂	94	2.01	400	.49	.0334
Pu19	UO ₂ -35 w/o PuO ₂	89	2.14	478	-	.174
Pu19	UO ₂ -35 w/o PuO ₂	89	2.14	498	-	.244

Table 3.11

Diffusion Equations

<u>Pellet Composition</u>	<u>Diffusion Equation</u>
UO ₂ .04	$D = 2.12 \times 10^{-3} \exp \left[- (25,380 \pm 1,900)/RT \right]$
UO ₂ -5 w/o PuO ₂	$D = 3.29 \times 10^{-5} \exp \left[- (38,900 \pm 3,900)/RT \right]$
UO ₂ -12.5 w/o PuO ₂	$D = D_0 \exp \left[- (16,700)/RT \right]$
UO ₂ -12.5 w/o PuO ₂	$D = D_0 \exp \left[- (20,620)/RT \right]$
UO ₂ -20 w/o PuO ₂	$D = 2.39 \times 10^{-6} \exp \left[- (33,170 \pm 3,000)/RT \right]$
<u>Powder Composition</u>	<u>Diffusion Equation</u>
UO ₂ -5 w/o PuO ₂	$D = 1.20 \times 10^{-6} \exp \left[- (20,800)/RT \right]$

OXIDATION PICKUP WITH TIME FOR $\text{UO}_2\text{-5}\%$ PuO_2 PELLETS

FIG. 3.4

The analysis of the oxidation kinetics of UO_2 -5 w/o PuO_2 powder (Pu14B) is continuing. Seventeen oxidation runs have been made on this powder in the temperature range of 148 to 403°C. The results to date are given in Table 3.12, and the resultant diffusion equation is given in Table 3.11. An Arrhenius plot of the diffusion coefficient of oxygen in UO_2 -5 w/o PuO_2 is shown in Figure 3.5. The oxidation of a low PuO_2 composition material, UO_2 -0.5 w/o PuO_2 , is also being studied. This powder has been re-reduced and has good stoichiometry and a low surface area. The preliminary results are also presented in Table 3.11.

Physical Property Measurements
(E. K. Halteman, R. Gerrish, J. Roth)

The high temperature x-ray diffractometer was committed to plutonium during this reporting period and has been used mainly for compound identification purposes on a service basis. The limiting conditions for analysis presently are a maximum temperature of about 1500°C and a maximum pressure of 15 microns with a nitrogen flush atmosphere. The platinum stage of the high temperature furnace has been replaced with a tungsten stage after it was discovered that reaction had occurred between ThO_2 and UO_2 and platinum. A possible explanation for this reaction is a U-O dissociation at high temperatures with possible alloying of free uranium.

The conversion from the platinum to the tungsten stage necessitated the adoption of an internal standard to be used for temperature indication since there is no thermocouple attached to the tungsten stage. Thoria was selected to serve this purpose since the coefficient of thermal expansion is well defined in the literature using x-ray techniques. Also, there is little or no reaction between thoria and UO_2 or PuO_2 under the operating conditions. It is anticipated that the temperature determined by this method will be good to $\pm 20^\circ\text{C}$, which will result in an error of about 3% in the operable temperature range to 1500°C.

Table 3.12
Coprecipitated Powder Oxidation Data

<u>Sample</u>	<u>Composition</u>	<u>Surface Area</u> <u>M²/gm</u>	<u>Initial</u> <u>Oxygen-to-Metal Ratio</u> <u>of Uranium Fraction</u>	<u>Reaction</u> <u>Temperature</u> <u>°C</u>	<u>Diffusion</u> <u>Coefficient</u> <u>cm²/sec x 10¹⁴</u>
Pu14B	UO ₂ -5 w/o PuO ₂	4.66	2.16	213	1.13
				184	.28
				189	.26
				189	.24
				170	.13
				169	.081
				174	.093
Pu41	UO ₂ -0.5 w/o PuO ₂	0.7	2.004	185	1.55
				219	4.09

OXYGEN DIFFUSION IN UO₂-5^w% PuO₂ POWDER

FIG. 3.5

FUEL ELEMENT FABRICATION AND EVALUATION

Task 4.00

I. D. Thomas

Fabrication of High Burnup Test Specimens
(R. M. Horgos, M. D. Houston)

The high burnup fuel samples are now being fabricated in accordance with the design criteria discussed under Task 5.00. As described in previous sections, all fuel powders have been processed, and sintering trials have been run to determine the required pressing and sintering conditions.

RADIATION TESTING AND EVALUATION

Task 5.00

R. M. Horgos

Short-Term Irradiation Tests (R. M. Horgos)

The fourteen capsules have been at the reactor site awaiting insertion since the last report period.⁽ⁱ⁾ Technical difficulties with the reactor necessitated low power operation. This lower power consequently resulted in a considerably lower thermal neutron flux in the trail cable facility than required for our irradiations. As a result, the three capsules which require irradiation during the start-of-cycle peak flux period have not yet been inserted.

High Burnup Irradiation Tests (R. M. Horgos)

Detailed design of the high burnup fuel elements and test capsules has been completed.

To meet the program objectives described previously,⁽ⁱⁱ⁾ fuel elements will be fabricated using a variety of potential fast breeder fuels and thermal converter fuels. The fuels will be prepared by a number of methods to assess the behavioral effects of exaggerated differences in processing variables; especially, the effects of inhomogeneities that might arise due to poor mechanical blending of component powders and the effects of the presence of reduced phases that might be present initially or that could develop during irradiation due to oxygen pickup by fission products. All fuel elements will be fabricated by the pellet-in-tube route. The fuel element composition and fuel preparation routes are tabulated in Table 5.1.

(i) NUMEC P-105, Progress Report, "Development of Plutonium-Bearing Fuel Materials", page 34.

(ii) NUMEC P-105, Progress Report, "Development of Plutonium-Bearing Fuel Materials", page 35.

Table 5.1

Fuels for High Burnup Irradiation Tests

<u>Capsule Number</u>	<u>Element Number</u>	<u>Fuel Composition</u>	<u>Fuel Preparation Route</u>
NU-15	N-15-1	UO ₂ -5 w/o PuO ₂ (7.5 w/o enriched UO ₂)	Coprecipitated solid solution UO ₂ -PuO ₂ .
	N-15-2	UO ₂ -5 w/o PuO ₂ (7.5 w/o enriched UO ₂)	Mechanically blended oxides using near-stoichiometric PuO ₂ powder.
	N-15-3	UO ₂ -5 w/o PuO ₂ (7.5 w/o enriched UO ₂)	Mechanically blended oxides using PuO ₂ powder containing approximately 20% Pu ₂ O ₃ .
	N-15-4	UO ₂ -5 w/o PuO ₂ (7.5 w/o enriched UO ₂)	High density PuO ₂ particulates blended with UO ₂ powder.
	N-15-5	UO ₂ -5 w/o PuO ₂ (7.5 w/o enriched UO ₂)	High density PuO ₂ particulates blended with coprecipitated UO ₂ -PuO ₂ .
NU-16	N-16-1 through N-16-5	UO ₂ -5 w/o PuO ₂ (7.5 w/o enriched UO ₂)	Duplicates of Capsule 15 elements.
NU-17	N-17-1	UO ₂ -20 w/o PuO ₂ (Natural UO ₂)	Coprecipitated solid solution UO ₂ -PuO ₂ .
	N-17-2	UO ₂ -20 w/o PuO ₂ (Natural UO ₂)	Mechanically blended oxides using near-stoichiometric PuO ₂ powder.
	N-17-3	UO ₂ -20 w/o PuO ₂ (Natural UO ₂)	Mechanically blended oxides using PuO ₂ powder containing approximately 20% PuO ₂ .
	N-17-4	UO ₂ -20 w/o PuO ₂ (Natural UO ₂)	UO ₂ -20 w/o PuO ₂ coprecipitated powder formed into dense particulates, mechanically packed and infiltrated with tungsten.
	N-17-5	UO ₂ -20 w/o PuO ₂ (Natural UO ₂)	UO ₂ -20 w/o PuO ₂ coprecipitated powder formed into low density particulates, mechanically packed and infiltrated with tungsten.
NU-18	N-18-1 through N-18-5	UO ₂ -20 w/o PuO ₂ (Natural UO ₂)	Replicate of Capsule 17 elements.
NU-19	N-19-1 through N-19-5	UO ₂ -20 w/o PuO ₂ (Natural UO ₂)	Replicate of Capsule 17 elements.

A drawing of the test element is shown in Figure 5.1. The fuel pellets will be centerless ground and clad in Type 347 stainless steel. In addition to the 1.5 inch fuel volume, the capsule contains a 1.0 inch length void volume to accommodate fission gases. A relatively small, 0.200 inch, fuel diameter was chosen to obtain high burnup in a reasonable time period.

A drawing of the general capsule arrangement is shown in Figure 5.2. The capsule is made of thick-walled Zircaloy to allow accurate positioning of four pairs of thermocouples. With these thermocouples, the temperature gradient will be continuously monitored to provide heat flux and burnup data and thereby to provide the information necessary for capsule positioning at the maximum safe flux levels in the GETR RAFT facility. Such positioning at the maximum flux levels will insure maximum burnup in the shortest time span.

Preliminary technical discussions have been held with reactor personnel to insure subsequent approval of the fuel element and capsule design with a minimum of alteration.

Figure 5.2

REACTOR PHYSICS AND ENGINEERING PARAMETRIC STUDIES

Task 8.00

J. D. Clement

High Burnup Irradiation Tests

During this period, fuel compositions, flux requirements, heating rates and specimen temperatures were determined for the high burnup irradiation tests. The results are presented in Table 8.1. With the freedom to move samples during reactor operation as provided by the RAFT facility, it will be possible to maintain maximum heat generation and maximum centerline temperature throughout the irradiation period.

In Table 8.2 are shown the anticipated burnup rates, the irradiation units required and the time span necessary to achieve the desired burnup levels.

Table 8.1

High Burnup Irradiation Capsule Characteristics

Capsules NU-15 and 16
(UO₂-5 w/o PuO₂)

	<u>Centerline Temperature</u>	<u>Average Unperturbed Flux</u>	<u>Average Heating Rate</u>	<u>Heat Flux - BTU/hr/ft²</u>	
				<u>Out of Fuel</u>	<u>Out of Capsule</u>
(Maximum)	2700°C	5.52 x 10 ¹³ nv	1744 watts/cc	702,000	102,000
(Minimum)	2550°C	5.22 x 10 ¹³ nv	1650 watts/cc	664,000	97,000
(Design)	2650°C	5.43 x 10 ¹³ nv	1716 watts/cc	691,000	101,000

Capsules NU-17, 18, 19
(UO₂-20 w/o PuO₂)

	<u>Centerline Temperature</u>	<u>Average Unperturbed Flux</u>	<u>Average Heating Rate</u>	<u>Heat Flux - BTU/hr/ft²</u>	
				<u>Out of Fuel</u>	<u>Out of Capsule</u>
(Maximum)	2700°C	4.29 x 10 ¹³ nv	1744 watts/cc	702,000	102,000
(Minimum)	2550°C	4.05 x 10 ¹³ nv	1650 watts/cc	664,000	97,000
(Design)	2650°C	4.21 x 10 ¹³ nv	1716 watts/cc	691,000	101,000

Table 8.2

Burnup Rate and Irradiation Schedule

<u>Capsule Number</u>	<u>Planned Irradiation Period (MWD/Tonne)</u>	<u>Average Burnup Rate (MWD/Tonne per Reactor Cycle)</u>	<u>Number of Reactor Cycles</u>	<u>Irradiation Units (10^{20} nvt/in²)</u>	<u>Scheduled Insertion Date</u>	<u>Scheduled Discharge Date</u>
NU-15	10,000	3150	3	207	4/64	7/64
NU-16	20,000	3150	6	414	4/64	10/64
NU-17	20,000	3050	6	321	6/64	12/64
NU-18	50,000	3050	15	802	6/64	9/65
NU-19	100,000	3050	30	1605	6/64	12/66