GEL-ADDITION PROCESS CHEMICAL STUDIES
— Quarterly Progress Report No. 13 —

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ABSTRACT

The development of a sol-gel process and related processes for preparing (U,Pu)O₂ fast reactor fuels has continued in Babcock & Wilcox's laboratories. Progress for the thirteenth quarter, ending October 31, 1969, is reported. The program is sponsored by the U. S. Atomic Energy Commission under Contract No. AT(30-1)-3714.

About 4.3 kg of ceramic-grade powder containing 20% PuO₂ in fully enriched UO₂ were prepared by continuous coprecipitation for irradiation in the EBR-II. The construction and cold checkout of a unit-operation-scale system for forming (U,Pu)O₂ sol-gel microspheres were completed.

All pellet, vipac, and sphere-pac fuel pins required for the FY-1970 irradiation program in the BAWTR were fabricated and are awaiting end-cap welding. The pellets containing 20% PuO₂ in fully enriched UO₂ were fabricated as required for the initial EBR-II irradiation test.

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1. INTRODUCTION

This is the thirteenth progress report on a continuing program of experimental work on sol-gel and related methods for preparing mixed uranium-plutonium oxide fuels for fast reactor application. The program is sponsored by the U. S. Atomic Energy Commission under Contract No. AT(30)-3714. This report covers the period from August 1 to October 31, 1969.

The work program for this contract establishes several technical tasks, including fuel preparation and fabrication, reactor testing in both thermal and fast neutron fields, and analytical support. For fiscal year 1970, the prime objective is the preparation of fuel and the fabrication of fuel pins for irradiation in the EBR-II of 18 unencapsulated pins in a 37-pin subassembly to be shared with ORNL. This fuel will be fully enriched UO_2 -20% PuO_2 in the form of sol-gel microspheres, shards (crushed pellets), and pellets. In addition, eight capsules containing UO_2 -20% PuO_2 in the same three fuel forms are scheduled for irradiation in the BAWTR at linear heat ratings of 6 to 20 kW/ft for 1 to 81 days. By the end of the fiscal year, 15 of 17 capsules in a series designed to show the differences in restructuring kinetics of pellets and vibratory compacted spheres and shards will have been irradiated in the BAWTR. Five of these capsules are to be examined in the B&W hot cell facility.

During this report period, 4.3 kg of ceramic-grade coprecipitated powder were prepared for the EBR-II irradiations. Construction and cold checkout of a unit-operation-scale system for spheroidizing UO_2 - PuO_2 sols were completed. All fuel pins required for the FY-1970 BAWTR irradiation program were fabricated. Progress in all technical areas of the program is reviewed in the following sections.

2. RESULTS AND CONCLUSIONS

2.1. Fuel Preparation

About 4.3 kg of fully enriched UO_2 -20% PuO_2 powder were prepared by continuous coprecipitation of ammonium diuranate-plutonium hydroxide from a uranium-plutonium nitrate solution. Of the fifteen 315-g batches run, one was rejected on the basis of a $\text{Pu}/(\text{Pu}+\text{U})$ ratio outside the specifications of $20.0 \pm 0.5\%$. Owing to holdup in the large equipment, the average yield was only about 86%. This completes the powder requirements for the EBR-II irradiation tests currently scheduled under this contract.

About 400 g of plutonium were converted into stable PuO_2 sols using the ORNL precipitation-peptization flow sheet. An unexplained difficulty in reducing the NO_3^-/Pu mole ratio to less than 0.2 was encountered in three batches.

About 8.32 kg of depleted uranium and 700 g of fully enriched uranium were converted into stable UO_2 sols using the ORNL formate flow sheet. The depleted UO_2 sol was used in the cold checkout of the unit-operation-scale spheroidizing system, which is now operational. The fully enriched UO_2 sol and the PuO_2 sol will be used in the preparation of 1300 g of UO_2 -20% PuO_2 sol-gel microspheres for the EBR-II irradiation tests.

2.2. Fuel Fabrication

Twenty-four BAWTR specimens were fabricated-six pins containing pellets, six containing shards, and 12 containing sol-gel microspheres. This completes the fuel pin fabrication requirements for the FY-1970 BAWTR irradiation program under this contract.

All the UO_2 blanket pellets required for the EBR-II pins were fabricated. In addition, the fully enriched UO_2 -20% PuO_2 pellets for the

EBR-II pellet and shard pins were pressed. The binder will be burned out, and the pellets will be fired during the next report period.

Ceramography of the EBR-II blanket pellets and the BAWTR fuel pellets showed a small percentage of an apparent metallic phase in the microstructure. In addition, the alpha autoradiographs indicated that the (U,Pu)O₂ pellets contain regions in which the Pu/(Pu+U) ratio is as high as 25%. However, the material is considered to be acceptable for these irradiation tests.

2.3. Irradiation Testing

The assembly of additional capsules for irradiation in the BAWTR was continued. The first of eight capsules scheduled for irradiation in FY-1970 will be inserted into the BAWTR in time for the December 1969 startup for an 81-day irradiation at 6 kW/ft. In addition, the request for "approval-in-principle" for the irradiation of an unencapsulated bundle (to be shared with ORNL) in the EBR-II was submitted to the USAEC.

2.4. Analytical Support

A formal quality assurance plan for the fuel being prepared in FY-1970 has been written and submitted to the USAEC. Characterization of the fuel fabricated for the FY-1970 BAWTR irradiation program and the UO₂ blanket pellets for the EBR-II tests has continued. The impurity levels were generally low, and the plutonium content and oxygen-to-metal ratio were acceptable.

3. FUEL PREPARATION (R. V. Carlson)

3.1. Introduction

The gel-addition irradiation program includes the exposure of spherical particles, angular particles, and pellets to the fast reactor conditions obtainable in the EBR-II. Eighteen unencapsulated pins in a subassembly to be shared with ORNL are scheduled for insertion in July 1970. The FY-1970 fuel order for the EBR-II irradiations includes 1300 g of sol-gel microspheres and 4100 g of coprecipitated powder containing 20% PuO_2 in UO_2 enriched to 93% ^{235}U .

3.2. Preparation of Microspheres Containing 20% PuO_2 in Fully Enriched UO_2

The formation of sufficient $^{235}\text{UO}_2$ and PuO_2 sol to produce 1300 g of sol-gel microspheres is in progress. The sols will be blended and gelled in the unit-operation-scale spheriodizing system during the next quarter.

3.2.1. Preparation of UO_2 Sol

About 8.32 kg of depleted uranium and 700 g of fully enriched uranium were converted into stable UO_2 sols using the ORNL formate flow sheet.¹ The pertinent run data from the sol preparations are summarized in Table 3-1.

The depleted UO_2 sols were prepared for the cold testing of both a geometry-controlled ^{235}U mixing system and a unit-operation-scale spheroidizing system. During these runs, an apparent relationship between the urea/uranium mole ratio (in the initial feed solution) and the U(VI) reduction time was observed. In batches 115 - 120, the urea/uranium mole ratio was inadvertently adjusted to 0.40 instead of to the flow sheet value of 0.60. With the uranium concentration in excess of 1 M, quantitative reduction of U(VI) in a reasonable length of

time was not possible. The reduction of batches 115 - 117 was discontinued after the temperature of the reaction vessel had been near room temperature for at least 1 hour.

Since the net reaction taking place during reduction is exothermic, the reaction vessel is always warm during a normal reduction. Therefore, the lack of heat generation was assumed to indicate that the desired reaction was not taking place. Difficulty previously observed in reducing U(VI) was overcome by diluting the uranium to a concentration of 0.83 M and decreasing the total quantity of uranium per batch.² This technique was successful for batches 118 - 120. In batch 121 the urea/uranium mole ratio was adjusted to 0.60, and the reduction time was decreased substantially. The average reduction time was about 4.6 hours for batches 123 - 133 and about 9.0 hours for batches 134 - 144. Since different operators prepared each series of runs, the difference in reduction time is assumed to be related to a change in technique, such as stirring speed. However, it should be noted that quantitative reduction in these batches was obtained in a reasonable length of time as compared to that for batches 115 and 116.

The fully enriched UO_2 sols were prepared as feed material for the formation of (U,Pu) O_2 microspheres. Since a low urea/uranium mole ratio apparently results in extremely long reduction times, the urea/uranium mole ratio was adjusted to the unusually high value of about 0.80 to ensure that an adequate amount of urea was present. In addition, the quantity of uranium per batch was reduced. The data for batches 145 - 149 in Table 3-1 show that complete reduction was achieved within a short period of time. The only processing problem encountered during these five runs was the over-reduction of batch 146 in less than 1 hour. The reason for this extremely short reduction time is not known. Additional fully enriched UO_2 sol will be prepared next quarter in larger batch sizes.

3.2.2. Preparation of PuO_2 Sol

About 400 g of plutonium were converted into stable PuO_2 sols using the ORNL precipitation-peptization flow sheet.³ The operating procedure has been reported,⁴ and the run data from these batches are summarized in Table 3-2.

Table 3-1. Data From Preparation of UO_2 Sols

Batch No.	^{235}U enrichment, wt %	U reduced, g	Input U conc, M	Urea/U mole ratio	Reduction time, h
115	<0.71	367.5	1.03	0.40	>18.0 ^(a)
116	<0.71	367.5	1.03	0.40	>13.0 ^(a)
117	<0.71	367.5	1.03	0.40	>6.0 ^(a)
118	<0.71	52.5	0.35	0.40	3.75
119	<0.71	122.4	0.79	0.40	5.5
120	<0.71	244.8	1.12	0.40	6.75
121	<0.71	183.8	0.92	0.60	<3.75 ^(b)
122	<0.71	186.0	0.92	0.59	1.67
123	<0.71	367.5	1.06	0.60	2.5
124	<0.71	367.5	1.06	0.60	3.75
125	<0.71	367.5	1.06	0.60	4.25
126	<0.71	367.5	1.06	0.60	1.75
127	<0.71	367.5	1.06	0.60	6.75
128	<0.71	367.5	1.06	0.60	5.75
129	<0.71	367.5	1.06	0.60	4.5
130	<0.71	367.5	1.06	0.60	5.75
131	<0.71	367.5	1.06	0.60	5.5
132	<0.71	367.5	1.06	0.60	5.75
133	<0.71	367.5	1.06	0.60	4.67
134	<0.71	367.5	1.06	0.60	10.0
135	<0.71	367.5	1.06	0.65	9.75
136	<0.71	367.5	1.06	0.65	8.17
137	<0.71	367.5	1.06	0.65	11.5
138	<0.71	367.5	1.06	0.65	8.0
139	<0.71	367.5	1.06	0.65	7.5
140	<0.71	367.5	1.06	0.65	11.0
141	<0.71	367.5	1.06	0.65	8.0
142	<0.71	367.5	1.06	0.65	7.75
143	<0.71	367.5	1.06	0.65	<8.0 ^(b)
144	<0.71	367.5	1.06	0.65	9.0
145	92.68	100.0	0.47	0.81	1.67
146	92.68	200.0	0.81	0.79	<1.0 ^(b)
147	92.68	200.0	0.81	0.79	2.0
148	92.68	200.0	0.81	0.79	2.33
149	92.68	200.0	0.81	0.79	2.5

(a) U(VI) reduction was not quantitative.

(b) Over-reduction occurred.

Table 3-2. Summary of Batch Preparation of Pu(IV) Sols

Batch No.	Stock solution		Pu input, g	4M NH_4OH , g	No. washes	Final filtrate, pH	NO_3^-/Pu mole ratio to peptize	Peptizing time, h	Baking time, h	Final NO_3^-/Pu mole ratio	Yield of denitrated powder, g	Pu recovered as sol, % ^(a)
	Pu, g/l	H^+ , M										
25	25.46	1.00	101.7	2.86	5	7.3	1.17	2.75	3.5	0.14	110.3	92.2
26	25.46	1.00	113.0	2.86	4	7.3	1.06	2.5	2.0	0.13	123.6	93.0
27	25.46	1.00	85.3	2.86	4	7.0	1.20	2.75	2.0	0.14	95.3	94.9
28	25.57	1.00	114.2	2.86	3	6.8	1.23	3.0	2.3	0.79 ^(b)	--	--
29	25.57	1.00	101.0	2.86	3	7.2	1.20	2.0	1.0	0.81 ^(b)	--	--
30	26.18	1.00	117.4	2.86	3	7.2	1.20	2.25	4.0	0.19	≈40 ^(c)	≈30
31	25.58	1.00	113.5	3.20	-- ^(d)	--	--	--	--	--	--	--
32	25.56	1.00	102.0	2.86	4	6.5	1.21	0.50	2.3	0.11	104.5	87.1

(a) Assuming denitrated powder is 85 wt % Pu.

(b) Further reduction of NO_3^-/Pu mole ratio was not possible.

(c) Only 1/3 of powder was satisfactory.

(d) Pu(IV) polymer apparently peptized before initial filtration.

No problems were encountered in preparing batches 25 - 27. However, a problem similar to one encountered previously at B&W occurred with batches 28 and 29.⁵ The processing proceeded normally during these two runs up to the thermal denitration step. In each case, nitrate decomposed normally during the baking operation until the NO_3^-/Pu mole ratio was about 0.8. However, when the powder was placed back on the hot plate after measuring the nitrate content, no additional NO_2 fumes were observed. It was then discovered that samples of the black powder removed for nitrate determination would not re-suspend in water. Failure to resuspend is the usual result of overbaking, i. e., reducing the NO_3^-/Pu mole ratio to less than about 0.1. However, in every case previously observed at B&W, overbaked powder was green rather than black. Therefore, since no NO_2 fumes were observed after the NO_3^-/Pu mole ratio was determined to be about 0.8, and since the powder was black, it is felt that this powder was not overbaked.

In batch 30, processing again proceeded normally up to the thermal denitration step. At this point, the powder was divided into three approximately equal lots. In the first lot, nitrate decomposed until the NO_3^-/Pu mole ratio was 0.37. During additional baking, no nitrate fumes were observed, and the powder would not resuspend in water. The second lot was overbaked—the powder was green and would not resuspend. The third lot was baked until the NO_3^-/Pu mole ratio was 0.19, and the powder did resuspend in water to form a stable sol.

The reason for failure to reduce the NO_3^-/Pu mole ratio of the powder to less than 0.2 and to resuspend the high-nitrate powder in water is not clear. One possible explanation could be related to an excessive temperature during baking. In batches 28 - 30 the powder was baked in a stainless steel beaker placed directly on a hot plate rather than in a sand bath. This procedure has been used previously to reduce the baking time with no difficulty. However, in these runs the temperature could have been higher than in the previous runs.

Batch 31 was abandoned before the hydrous Pu(IV) polymer was washed. The precipitation was carried out as usual, but the filtration of the slurry was interrupted, and the slurry was allowed to

stand about 16 hours before the filtration was continued. During this period, the precipitate spontaneously peptized to a translucent green dispersion, which readily passed through a Whatman No. 42 filter paper. This phenomenon has not been seen before at B&W even though filtration was interrupted in several previous runs. The PuO_2 sol preparation procedure has been modified to eliminate any delay in carrying out the filtration.

The processing of batch 32 was normal. The first filtration was completed immediately after the precipitation, and a sand bath was used for temperature control during thermal denitration. The digested polymer was peptized by nitric acid in a shorter length of time than it normally is, but this apparently had no undesirable effect on the powder. The yield of stable sol was lower than the normal yield because a large number of samples were taken during the baking in order to monitor the NO_3^-/Pu mole ratio very closely.

3.3. Preparation of Fully Enriched UO_2 -20% PuO_2 Powder by Coprecipitation

A total of 4100 g of powder containing 20% PuO_2 in fully enriched UO_2 is required for the EBR-II irradiation tests under the gel-addition contract. During this quarter about 4.3 kg of mixed oxide powder were prepared by continuous coprecipitation of ammonium diuranate-plutonium hydroxide from a uranium-plutonium nitrate solution. The flow sheet used is summarized in Figure 3-1.

Although the capacity of the continuous coprecipitation equipment is about 2 kg/day, the fissile material mass limit of the glove box restricted the throughput to 315 g (on a metal basis) per day for these runs. The $\text{Pu}(\text{NO}_3)_4$ and $\text{UO}_2(\text{NO}_3)_2$ feed solutions were mixed on a weight basis from previously analyzed stock solutions. The compositions of these stock solutions are shown in Table 3-3. The pH was held at about 9.5 during precipitation to ensure homogeneity of the plutonium and uranium in the powder.^{6,7}

To dry the wet filter cake and then calcine the powder, it is necessary to transfer the material twice between laboratory units established for maintaining criticality control. Therefore, to increase the accuracy of these fissile material transfers, an empirical value for the weight losses during drying and calcination was sought by making a

Figure 3-1. Flow Sheet for Preparation of UO_2 -20% PuO_2 by Continuous Coprecipitation Process

number of weighings. These data are summarized in Table 3-4. The weight loss during drying averaged about 71%. During calcination, the weight loss averaged about 30%; however, the data show large variations from the average for a given batch. This indicates that a 12-hour drying at 125 C is not sufficient to obtain a reproducible product.

The plutonium content of each batch of calcined powder was measured before being blended into larger lots. The specification calls for a Pu/(Pu+U) ratio of $20.0 \pm 0.5\%$. The data in Table 3-4 show that all batches were within these limits except for 8-B-1, which was therefore rejected.

The average yield for all batches except 8-B-1 was about 86%. The principal source of material loss was holdup in the large equipment. After each run, the entire system was flushed with dilute nitric acid, and the resultant solution was stored for scrap recovery. In running large batches through this equipment, the quantity of material holdup is about the same, but the percentage loss is reduced.

The 14 batches of calcined powder meeting the plutonium content specification were blended into two lots. The first contained 2142.0 g of powder and was ball-milled for 12 hours. This lot was then used to prepare pellets and shards for the unencapsulated EBR-II bundle to be shared with ORNL (see section 4.2). The second lot contained 2155.2 g of powder and was ball-milled for 8 hours. This lot will remain in storage until the encapsulated EBR-II subassembly is fabricated (see section 5.4).

3.4. Pilot Plant Equipment (R. N. Gurley)

A unit-operation-scale spheroidizing system was designed late in FY-1968,⁸ and construction was nearly complete early in FY-1969.⁹ However, owing to funding restrictions, it was placed in a standby status before the cold checkout was completed. During this quarter, about 15# of depleted UO_2 sol were run through the system to complete the cold checkout. The FY-1970 sol-gel microsphere fuel order for the EBR-II irradiations will be produced with this system during the next quarter.

Table 3-3. Composition of Stock Solutions
for Coprecipitation

<u>Plutonium Nitrate</u>	
Plutonium, mg/g soln	156.4 ± 0.08
Free acid, M	5.2
²³⁸ Pu, wt %	0.047
²³⁹ Pu, wt %	90.497
²⁴⁰ Pu, wt %	8.344
²⁴¹ Pu, wt %	1.033
²⁴² Pu, wt %	0.079
<u>Uranyl Nitrate</u>	
Uranium, mg/g soln	227.5 ± 0.5
²³⁴ U, wt %	0.99 ± 0.01
²³⁵ U, wt %	92.68 ± 0.05
²³⁶ U, wt %	0.38 ± 0.01
²³⁸ U, wt %	5.95 ± 0.03

Table 3-4. Data From Preparation of Coprecipitated Powder

Batch No.	Weight of wet cake, g	Weight of dried cake, g	Weight of calcined product, g	Weight loss during drying, g	Weight loss during calcination, %	Pu/(Pu+U), %	Yield, %
8-A-91	1286	380	313	70.5	17.6	20.16±0.08	87.5
8-A-96	NM(a)	412	305	--	26.0	20.17±0.04	85.3
8-A-97	1425	380	302	73.3	20.5	20.05±0.09	84.5
8-A-98	1417	400	300	71.8	24.8	20.20±0.08	84.0
8-A-99	1463	447	300	69.4	32.9	20.09±0.05	83.8
8-B-1	1333	430	258	67.7	40.0	23.00±0.08	72.2
8-B-2	1605	431	302	73.1	29.9	20.20±0.24	84.5
8-B-3	1471	407	308	72.3	24.3	20.12±0.06	86.1
8-B-4	1450	417	303	71.2	27.3	20.14±0.04	84.7
8-B-5	1815	514	309	71.7	30.8	20.24±0.05	86.4
8-B-6	1476	415	301	71.9	27.5	20.14±0.15	84.0
8-B-7	1614	454	295	71.9	35.0	20.33±0.03	82.5
8-B-9	NM(a)	499	313	--	37.3	20.36±0.06	87.5
8-B-10	NM(a)	499	302	--	39.5	20.32±0.08	84.3
8-B-13	NM(a)	545	345	--	36.7	20.29±0.03	96.5

(a) Not measured.

The unit-operation-scale spheroidizing system incorporates many improvements over the laboratory-scale system previously used. These improvements include:

1. A $\text{UO}_2\text{-PuO}_2$ sol throughput capacity of at least 500 g of uranium plus plutonium per 8-hour shift. This throughput is obtained with a single two-fluid nozzle. The sol injection needle is stainless steel and contains four axial holes. The holes in a given needle are identical in size and range from 12 to 24 microns in diameter in the needles available for this work. A sol feed rate of 8 to 10 cc/minute is easily obtainable.

2. A simplified and versatile sol injection control system. The sol feed rate and drop size are controlled by nitrogen gas pressure rather than by a pump.

3. A mixed-sol feed system that should overcome any tendency for the UO_2 and PuO_2 to segregate. The UO_2 and PuO_2 sols are each introduced initially into the primary sol mixing tank. After blending, the mixed sol is transferred to the spheroidizing column feed tank, which is equipped with a stirrer. The mixed sol, therefore, is essentially stirred continuously from the time it is introduced into the system until it is injected into the column.

4. A capability for recovery of the 2-ethyl-1-hexanol (2-EH). A side-stream of wet 2-EH (containing about 1% water) is withdrawn from the main 2-EH recirculation loop and sent to an electrically heated still. Water containing trace amounts of 2-EH is distilled and sent to waste, and the dry 2-EH (containing about 0.5% water) is returned to the main 2-EH recirculation loop.

5. Ethanol and aqueous ammonia washing of the spheres, as well as heated inert gas drying of the washed spheres.

6. Complete control of atmosphere during processing. An inert atmosphere is provided in all tanks and lines to minimize UO_2 sol oxidation. The vessels from which UO_2 sol is introduced to the system, as well as the vessels in which $(\text{U,Pu})\text{O}_2$ gel microspheres are removed from the system, also contain an inert atmosphere.

4. FUEL FABRICATION (J. M. Kerr)

4.1. Oxide Characterization (A. H. Bremser)

The first lot of coprecipitated powder containing fully enriched UO_2 -20% PuO_2 was received (see section 3.3). This material will be used in preparing pellets and shards for the unencapsulated EBR-II fuel pins. Because of the nonreproducible sintering characteristics of different lots of ceramic-grade powders, a sample of powder is routinely pressed into pellets and fired to ensure that the final density requirements can be achieved using the tested pellet-pressing and firing conditions. The heating cycle for the sinterability test on the EBR-II powder was as follows: Under N_2 -6% H_2 atmosphere, heat to 1000 C at 330 C/h, hold at 1000 C for 1 h, heat to 1650 C at 330 C/h, and hold at 1650 C for 2 h. The results from this test (Table 4-1) indicate that the test cycle is adequate to meet the density specification of $93 \pm 2\%$ theoretical.

A large amount of a metallic phase was previously observed in coprecipitated mixed oxide containing depleted UO_2 -20% PuO_2 .¹⁰ It was found that the metallic inclusions were being introduced by flaking of the metallic retort liner in the powder calcining furnace, and that most of it could be removed by passing the powder through a magnetic separator several times. Additional pellets were prepared from this powder for BAWTR irradiation (see section 4.2). The presence of the remaining metallic inclusions can be seen in the microstructure shown in Figure 4-1. In addition to the BAWTR pellets, the EBR-II pellets prepared for the sinterability test were examined ceramographically. The microstructure in Figure 4-2 shows that this material also contains a small amount of an apparent metallic phase. However, the results of both the ceramographic examination and the trace metallic impurity determination (Table 6-2) indicate that the amount of metallic phase is insufficient to warrant an attempt at either positive identification or separation.

Table 4-1. Sinterability Test Data for
EBR-II Fuel Pellets

	<u>Green pellets</u>	<u>Fired pellets</u>
Number of pellets sampled	10	14
Mean density, % theor.	48.27 ± 1.29	94.30 ± 0.69
Mean diameter, in.	0.2720 ± 0.00001	0.2119 ± 0.0017
Mean length, in.	0.3071 ± 0.0017	0.2436 ± 0.0021
Mean weight, g	1.549 ± 0.046	1.457 ± 0.030
Mean linear shrinkage, %	--	20.64 ± 0.25

Figure 4-1. Microstructure of Depleted
 UO_2 -20% PuO_2 Pellet

Unetched

Bright Field

100x

Figure 4-2. Microstructure of Fully Enriched
 UO_2 -20% PuO_2 Pellet

Figure 4-3. Alpha Autoradiograph of Depleted
 UO_2 -20% PuO_2 Pellet

Figure 4-3 is an alpha autoradiograph of a depleted UO_2 -20% PuO_2 pellet. The bright white spots in the figure are artifacts of the processing. However, the different shades of gray are an indication of inhomogeneity in the plutonium concentration. A comparison with standards indicates that the lighter areas may contain a $\text{Pu}/(\text{Pu}+\text{U})$ ratio as high as 25%. Kodak high resolution film (type SO-343), currently used for autoradiography at B&W, has increased the resolution over that of the earlier techniques.¹¹ Therefore, the presence of inhomogeneity, such as shown in Figure 4-3, would not have been detected in previous work. The source of this apparent inhomogeneity is not known, but the fuel is judged to be acceptable for the current irradiation tests.

Ceramography of the pellets from two firing batches of UO_2 blanket pellets for the EBR-II pins showed the presence of a small amount of an apparent metallic phase (see Figures 4-4 and 4-5). These flecks were also observed in the as-received UO_2 powder. The levels of the trace metallic impurities measured (see Table 6-2) were well within the fuel specifications. Therefore, these pellets were considered acceptable.

4.2. Fabrication of BAWTR Specimens (L. J. Ferrell)

All pellet, vipac, and sphere-pac specimens for the FY-1970 BAWTR irradiations have been fabricated and placed in storage until the irradiation capsules are assembled. A topical report on the fabrication and characterization of the FY-1969 BAWTR irradiation specimens was issued.¹²

The flow sheet for preparing pellets is similar to the one discussed previously.¹³ The depleted UO_2 -20% PuO_2 coprecipitated powder was prepressed at 40,000 psi, granulated through a 20-mesh screen, and final-pressed at 40,000 psi. The green pellets were then fired at 1650 C for 2 hours in a flowing N_2 -6% H_2 atmosphere. The BAWTR pellet pin loading data are shown in Table 4-2, and the pin fabrication data are given in Table 4-3.

The sphere-pac pins were loaded according to the flow sheet described earlier.¹³ Ten of the pins contained microspheres prepared at B&W, and the remaining two contained ORNL microspheres. All of the material was 20% PuO_2 in UO_2 enriched to 20% ^{235}U . The fabrication data for the BAWTR sphere-pac pins are shown in Table 4-4.

The flow sheet described previously was followed in loading the vipac pins.¹³ All of the feed material was coprecipitated powder containing depleted UO_2 -20% PuO_2 . The fabrication data for the BAWTR vipac pins are shown in Table 4-5.

4.3. Fabrication of EBR-II Specimens (L. J. Ferrell)

The pellets required for the EBR-II pellet and shard pins were fabricated with an automatic mechanical press. Binder was added to the coprecipitated powder, and the powder was dried, prepressed at 40,000 psi, granulated, and final-pressed at 40,000 psi. The binder will be burned out and the pellets will be fired during the next report period.

The UO_2 blanket pellets required for the EBR-II pins were fabricated in two batches and stacked into 5-inch lengths. The oxide, purchased from Eldorado Nuclear Limited, was prepressed at 40,000 psi, granulated through a 20-mesh screen, and final-pressed at 40,000 psi. The first batch was fired at 1450 C in a flowing N_2 -6% H_2 atmosphere, and the second batch was fired at 1500 C. The average density was $91.96 \pm 0.61\%$ theoretical for the pellets from the first firing and $93.12 \pm 1.24\%$ theoretical for those from the second. The data from the 5-inch pellet stacks are shown in table 4-6.

Figure 4-4. Microstructure of UO_2 Blanket Pellet for EBR-II Pins, Batch 1

Unetched

200×

Figure 4-5. Microstructure of UO_2 Blanket Pellet for EBR-II Pins, Batch 2

Unetched

200×

Table 4-2. Loading Data for BAWTR Pellet Pins

Pin No.	C-110	C-114	C-10	C-19	C-112	C-109
End cap	--	--	--	--	--	--
ZrO ₂ foam	--	--	--	--	--	--
UO ₂ insulator	--	--	--	--	--	--
Pellet, in. dia/% TD	0.206/93.45	0.207/91.41	0.207/91.96	0.207/91.73	0.208/91.79	0.207/91.80
Pellet, in. dia/% TD	0.208/93.56	0.208/91.38	0.207/91.82	0.208/91.53	0.208/91.42	0.206/95.02
Pellet, in. dia/% TD	0.207/93.00	0.206/94.27	0.208/92.88	0.206/93.12	0.209/91.61	0.205/94.62
Pellet, in. dia/% TD	0.208/91.30	0.208/92.45	0.207/94.84	0.206/95.19	0.207/94.60	0.207/94.53
Pellet, in. dia/% TD	0.207/94.50	0.206/93.78	0.206/94.12	0.207/94.10	0.207/94.56	0.208/91.93
Pellet, in. dia/% TD	0.208/91.46	0.207/94.47	0.208/92.84	0.206/94.01	0.207/92.06	0.208/91.95
Pellet, in. dia/% TD	0.207/92.67	0.208/91.75	0.208/92.02	0.207/93.58	0.208/91.83	0.208/92.58
Pellet, in. dia/% TD	0.206/94.06	0.208/94.32	0.207/93.17	0.205/94.92	0.208/91.75	0.208/91.83
Pellet, in. dia/% TD	0.206/95.26	0.207/93.97	0.208/93.53	0.207/94.87	--	0.206/93.93
UO ₂ insulator	--	--	--	--	--	--
End cap	--	--	--	--	--	--
Fuel weight, g	11.3896	11.3888	11.3738	11.3603	11.4969	11.4969

Table 4-3. Fabrication Data for BAWTR Pellet Pins

<u>Pin No.</u>	<u>Fuel weight, g</u>	<u>Fuel length, in.</u>	<u>Insulator weight, g</u>	<u>Insulator length, in.</u>	<u>ZrO₂ filler length, in.</u>
C-110	11.3896	2.007	2.5524	0.5518	1.1912
C-114	11.3888	2.002	2.6318	0.5546	1.1934
C-10	11.3738	2.004	2.5902	0.5527	1.1933
C-19	11.3603	2.002	2.8306	0.5411	1.2069
C-112	10.8544	1.914	2.8156	0.5667	1.2693
C-109	11.4969	2.025	2.8470	0.5688	1.1562

Note: The following conditions apply for all pins:

Pin length, in. 4.500
 Smeared density, % TD 82.5 ± 1.0
 Fuel form Pellet
 Insulator form Depleted UO₂ pellets

Table 4-4. Fabrication Data for BAWTR Sphere-Pac Pins

<u>Pin No.</u>	<u>Fuel length, in.</u>	<u>Smeared density, % theor</u>	<u>Insulator length, in.</u>	<u>ZrO₂ filler length, in.</u>	<u>²³⁵U content, wt %</u>
C-98	3.922	82.89	0.508	5.320	0.258
C-132	3.970	81.88	0.500	5.280	0.258
C-146	3.970	81.88	0.506	5.274	0.258
C-141	3.981	81.66	0.516	5.253	0.258
C-140	3.966	81.97	0.518	5.266	0.258
C-123	3.903	83.29	0.512	5.335	0.258
C-134	3.968	81.93	0.505	5.277	19.68
C-138	3.981	81.66	0.503	5.266	19.68
C-125	3.956	82.17	0.503	5.291	19.68
C-46	3.947	82.36	0.511	5.292	19.68
C-122(a)	3.916	83.15	0.500	5.334	20.14
C-52(a)	3.941	82.63	0.499	5.310	20.14

Note: The following conditions apply for all pins:

Pin length, in. 10.500
 Fuel weight, g 22.00
 Fuel form Spheres

(a) Contain ORNL microspheres.

Table 4-5. Fabrication Data for BAWTR Vipac Pins

Pin No.	Fuel length, in.	Smear density, % theor	Insulator length, in.	ZrO ₂ filler length, in.
C-84	3.886	84.20	0.506	5.358
C-53	3.966	82.50	0.502	5.282
C-137	4.000	81.80	0.502	5.248
C-126	3.982	82.17	0.503	5.265
C-129	4.001	81.78	0.504	5.245
C-128	4.001	81.78	0.500	5.249

Note: The following conditions apply for all pins:

Pin length, in.	10.500
Fuel weight, g	22.144
Fuel form	Shards
²³⁵ U content, wt %	0.258

Table 4-6. Stack Data for EBR-II Blanket

Stack No.	Length, in.	Weight, g	Stack No.	Length, in.	Weight, g
EBR-IN-1	5.062	30.89	EBR-IN-15	5.040	31.03
EBR-IN-2	5.053	30.96	EBR-IN-16	5.064	30.98
EBR-IN-3	5.049	30.94	EBR-IN-17	5.055	30.92
EBR-IN-4	5.062	30.89	EBR-IN-18	5.052	30.99
EBR-IN-5	5.057	31.02	EBR-IN-19	5.062	30.94
EBR-IN-6	5.049	30.97	EBR-IN-20	5.052	30.86
EBR-IN-7	5.053	31.00	EBR-IN-21	5.045	30.99
EBR-IN-8	5.044	30.95	EBR-IN-22	5.062	30.94
EBR-IN-9	5.054	30.92	EBR-IN-23	5.032	30.82
EBR-IN-10	5.038	30.90	EBR-IN-24	5.059	30.88
EBR-IN-11	5.038	31.00	EBR-IN-25	5.050	31.02
EBR-IN-12	5.045	30.93	EBR-IN-26	5.054	30.97
EBR-IN-13	5.048	30.96	EBR-IN-27	5.059	31.07
EBR-IN-14	5.041	31.03	EBR-IN-28	5.060	30.87

Average length, in. 5.051 ± 0.009
Average weight, g 30.95 ± 0.06

5. IRRADIATION TESTING (C. J. Baroch & E. N. Harbinson)

5.1. Introduction

The gel-addition irradiation task, as described previously,¹⁴ is divided into three subtasks:

1. Time Required for In-Pile Restructuring.
2. Sorbed Gas Behavior.
3. EBR-II Irradiation.

The scope and the progress for each of these subtasks are described in this section.

5.2. Time Required for In-Pile Restructuring

The effect of heat rate and time on the restructuring characteristics of sol-gel UO_2 - PuO_2 spheres and on angular particles and pellets prepared from coprecipitated UO_2 - PuO_2 powder will be evaluated. A total of 17 capsules will be irradiated in the BAWTR. The time required for restructuring at linear heat rates of about 6, 10, 15, and 20 kW/ft will be determined. The program will include a comparison of the irradiation behavior of spherical sol-gel particles prepared by ORNL and B&W.

No capsules were irradiated during this quarter, but the assembly of additional capsules was continued. The first of eight capsules scheduled for irradiation during FY-1970 will be inserted into the BAWTR in time for the December 1969 startup for an 81-day irradiation at 6 kW/ft. Seven capsules are ready for examination in the B&W hot cell facility. Table 5-1 shows the conditions under which these capsules were irradiated. The hot cell examinations of these capsules will begin when the B&W alpha-beta-gamma hot cell facility is completed in January 1970.

Table 5-1. BAWTR Capsule Irradiation Conditions

Capsule No.	Exposure time, d	Linear heat rate, kW/ft		NaK temperature, F	
		Design	Actual	Design	Actual
GA-15(a)	3	6	5.0-6.5	1040	1140
GA-14(a)	3	10	7.5-10.3	1080	950-1110
GA-11	1	15	15.5-17.0	1100	1040-1060
GA-13(a)	3	15	15.2-16.1	1100	1140-1170
GA-2	9	15	15.3-16.7	1100	1080
GA-4	1	20	17.2-19.6	1150	965-1095
GA-8(a)	3	20	17.0-19.8	1150	1050-1140

(a) Capsules GA-8, GA-13, GA-14, and GA-15 were irradiated simultaneously.

5.3. Sorbed Gas Behavior

The pressure created by the sorbed gas released from particulate fuel during irradiation will be determined. If practicable, each capsule will contain two pins in which the gas pressure is continuously monitored. One pin will contain spherical sol-gel particles and the other will contain angular coprecipitated particles. The gas-release characteristics of spherical sol-gel particles prepared by ORNL and B&W will be determined. By varying the linear heat rate, it should be possible to determine the amount of gas released as a function of average fuel temperature. The fuel pins will be irradiated at design linear heat rates of 6, 10, 15, and 20 kW/ft. No work has been performed on this subtask because of funding limitations.

5.4. EBR-II Irradiations

The irradiations in EBR-II will provide performance data on particulate fuels under the fast reactor conditions obtainable in EBR-II. The effect of heat rate and burnup on the overall performance of pellets, spherical particles, and angular particles will be evaluated. The smeared density for all three materials will be $82 \pm 2\%$. This subtask includes the irradiation of one 19-pin encapsulated subassembly in row

7 of the EBR-II and the irradiation of 18 unencapsulated pins in a 37-pin subassembly to be shared with ORNL. The pins for the unencapsulated bundle are described in Table 5-2. During this report period, the request for "approval-in-principle" for irradiation of the unencapsulated bundle was submitted to the USAEC.

Table 5-2. Number of B&W Fuel Pins for Unencapsulated Bundle

Burnup, GWd/T	Type of fuel(a)		
	Angular particles	Spherical particles	Pellets
25	2	3	2
50	2	2	2
75	2	3	2
100	3	3	2

(a) Some positions within the bundle will be used twice; for example, pins removed at an exposure of 75 GWd/T will be replaced with pins that will be irradiated to an exposure of 25 GWd/T.

6. ANALYTICAL SUPPORT (D. H. Schmitt)

6.1. Quality Assurance (D. H. Schmitt, R. V. Carlson, J. M. Kerr)

A formal quality assurance plan for the fuel being prepared in FY-1970 was written and submitted to the USAEC. The procedures in MIL-STD-414¹⁵ are followed to ensure representative sampling of pellet lots, and the procedures given by Jones¹⁶ are followed to ensure representative sampling of sphere and shard lots. The acceptance criteria are based on the finished fuel specifications. The basic quality assurance program is summarized in the following paragraphs.

6.1.1. Preparation of Coprecipitated (U,Pu)O₂ Powder

The Pu(NO₃)₄ and UO₂(NO₃)₂ feed solutions are each received as a single blended lot and analyzed on a weight basis. The input solutions for each coprecipitation run are then measured by weight. A "grab" sample is removed from each batch of powder and analyzed for Pu/(Pu+U). To be accepted, a batch must have an average Pu/(Pu+U) content between 19.5 and 20.5% (see Table 3-4). Two lots of coprecipitated powder are each formed by blending seven calcined (U,Pu)O₂ batches.

6.1.2. Preparation of Sol-Gel (U,Pu)O₂ Microspheres

The fully enriched UO₂ sol and PuO₂ sol input are each blended into a single lot and analyzed on a weight basis. Proof of lot homogeneity is obtained by requiring the measured gamma activity of each of eight consecutive samples to be within 0.5% of the average of all eight samples. The input sols for each sphere-forming run are then measured by weight. Each 315-g batch of gel microspheres is fired separately. Samples are selected randomly from each of four batches of coarse and four batches of fine microspheres and analyzed

for Pu/(Pu+U) and carbon content. If the average Pu/(Pu+U) ratio for each sampled batch is within $\pm 0.5\%$ (absolute) of the average for all batches sampled, and the average carbon content of each batch sampled is within ± 10 ppm (metal basis) of the average for all batches sampled, then all the material is accepted. If any batch sampled does not meet the Pu/(Pu+U) acceptance criterion, then all remaining batches are sampled and analyzed. Any batch in which the average Pu/(Pu+U) ratio is outside $20.0 \pm 0.5\%$ is rejected. The carbon content is measured to determine large differences between firings. If any sampled batch does not meet the carbon acceptance criterion, then the O/M ratio of each batch is measured. Any batch in which the average O/M atom ratio is outside the range of 1.97 to 2.00 is rejected. All acceptable batches of microspheres are blended into two lots—one containing the coarse fraction and one containing the fine fraction. Eight 3- to 4-g random samples (a total of at least 30 g) are taken from each lot for final characterization.

6.1.3. Fabrication of (U,Pu)O₂ Pellets and Shards for BAWTR Irradiation

All the pellets are pressed, using powder from a blended lot of depleted UO₂-20% PuO₂, and fired in two batches. The density of all fired pellets is measured. For a pellet to be accepted, its density must be in the normal operating range on the standard pellet-diameter-vs-pellet-density curves.¹³ Rejected pellets are centerless-ground, re-fired, or transferred to scrap recovery. Pellets for the fabrication of the BAWTR pellet pins are selected from one firing batch. The remaining pellets from this batch, as well as all pellets from the other batch, are crushed into shards and blended into one lot. At least 15 pellets from the pellet lot and 30 g of material from the shard lot are randomly selected for final characterization.

6.1.4. Fabrication of (U,Pu)O₂ Pellets and Shards for EBR-II Irradiation

All of the pellets are pressed, using powder from a blended lot of fully enriched UO₂-20% PuO₂, and fired in two batches. For a pellet to be accepted, its density must be $93 \pm 2\%$ of theoretical. Each batch of pellets is handled in the same manner as described in

section 6.1.3. At least 20 pellets from the final pellet lot and 30 g of material from the shard lot are randomly selected for final characterization.

6.2. Final Characterization of BAWTR Fuel

The final characterization of the depleted UO_2 -20% PuO_2 pellets and shards to be used in the FY-1970 BAWTR irradiation tests is nearly complete. In addition, the UO_2 blanket pellets for the EBR-II irradiation tests and the 20% PuO_2 in UO_2 enriched to 20% ^{235}U microspheres for the BAWTR tests are nearly characterized. These results are summarized in Tables 6-1 and 6-2. The aluminum content in the BAWTR pellets and shards is high, probably because of the use of low-density alumina balls during the ball milling of this coprecipitated powder. The silicon content of the microspheres is also high, but the source of this impurity is not known. However, it is felt that these impurity levels are acceptable for these BAWTR irradiation tests.

Table 6-1. Summary of Fuel Analyses

Analysis	BAWTR pellets ^(a)	BAWTR shards ^(a)	BAWTR microspheres ^(a)	EBR-II UO ₂ blanket pellets	
				Batch 1(a)	Batch 2(a)
Pu/(Pu+U), wt %	19.93±0.66	19.93±0.66	19.00±0.11	--	--
²³⁵ U content, wt %	<0.7(b)	<0.7(b)	19.68±0.02	<0.7(b)	<0.7(b)
O/M, atom ratio	1.975±0.006	1.977±0.007	1.974±0.003	2.007±0.000	2.007±0.000
Moisture, ppm oxide	4.6±0.7	19.8±6.1	11.6±5.4	10.3±2.2	3.2±0.7
Sorbed gas, cc/g	0.013	0.094	0.053	NA ^(c)	NA ^(c)
Density, % theor	91.3 - 95.3	NA ^(c)	96.2±0.9	NA ^(c)	NA ^(c)
Nitrogen, ppm metal	21±11	NA ^(c)	12.5±3.8	24±12	24±4
Carbon, ppm metal	64±13	35±16	11.8±2.4	14±11	12±9
Chloride, ppm metal	5.0±2.7	5.5±2.2	6.0±2.2	2.6±0.9	6.3±2.0
Fluoride, ppm metal	5.1±1.1	4.5±0.6	0.5±0.2	0.5±0.3	0.7±0.4

(a) Errors are for 95% confidence limits on the mean.

(b) ²³⁵U content was not determined but it should be less than normal.

(c) Not available at this time.

Table 6-2. Trace Metallic Impurities in Fuel^(a)

<u>Element</u>	<u>BAWTR pellets</u>	<u>BAWTR shards</u>	<u>BAWTR micro-spheres</u>	<u>EBR-II UO₂ blanket pellets</u>	
				<u>Batch 1</u>	<u>Batch 2</u>
Aluminum	>500	>500	305	<15	<15
Beryllium	<0.5	<0.5	<0.5	--	--
Bismuth	<1	<1	--	<1	<1
Boron	<0.5	<0.5	1.7	0.5	0.4
Cadmium	<0.5	<0.5	<1	<0.3	<0.3
Calcium	9	<11	--	55	40
Chromium	27	<23	<20	100	47
Copper	170	86	46	4	3
Iron	39	36	620	130	150
Lead	2	6	<1	<2	<2
Lithium	--	--	--	<2	<2
Magnesium	48	21	<23	<3	<3
Manganese	3	5	3	<6	<6
Molybdenum	<34	<34	<25	21	16
Nickel	<11	<11	<20	<10	<10
Silicon	28	7	>250	15	16
Silver	<0.5	<0.5	<1	<0.25	<0.25
Sodium	45	<4	24	15	19
Tin	10	2	3	22	15
Titanium	<5	<11	<15	--	--
Vanadium	<1	<1	<1	<3	<3
Zinc	<10	<10	<20	<50	<50

(a) Values given are in ppm of metal and the average of four determinations.

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