BAW-3714-16
UC-25
April 1970

GEL-ADDITION PROCESS CHEMICAL STUDIES

- Quarterly Progress Report No. 14 -

November 1969 - January 1970

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AEC Contract No. AT(30-1)-3714
B&W Contract No. 680-3100

Prepared for
THE U. S. ATOMIC ENERGY COMMISSION
by

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Key Words: Sol Gel, $(U,Pu)O_2$, Fast Reactor Fuels

ABSTRACT

The development of a sol-gel process and related processes for preparing $(U,Pu)O_2$ fast reactor fuels has continued at the Babcock & Wilcox Nuclear Development Center. Progress for the 14th quarter, ending January 31, 1970, is reported. The program is sponsored by the U. S. Atomic Energy Commission under Contract No. AT(30-1)-3714.

About 6 kg of $^{235}UO_2$ sol and 550 g of PuO_2 sol were prepared by precipitation-peptization techniques. The unit-operation-scale spheroidizing system was operated without the organic regeneration system. About 250 g of coarse and 150 g of fine $^{235}UO_2$-20% PuO_2 microspheres were prepared.

The $^{235}UO_2$-20% PuO_2 pellets and shards required for the initial EBR-II irradiation test were nearly completed. Because of a design change in the EBR-II pins, additional UO_2 blanket and insulator pellets were prepared.

Four capsules in a series designed to show the differences in restructuring kinetics of pellets and vibratory compacted spheres and shards were irradiated in the BAWTR. The exposure time varied from 3 to 81 days at 6 to 20 kW/ft.

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1. INTRODUCTION

This is the 14th 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(30-1)-3714. This report covers the period from November 1, 1969, to January 31, 1970.

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 FY-1970 the prime objective is the preparation of fuel and the fabrication of 18 unencapsulated fuel pins for irradiation in the EBR-II as part of 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 in FY-1970.

During this report period the preparation of $^{235}UO_2$ sol and PuO_2 sol was completed and about 400 grams of $^{235}UO_2$-20% PuO_2 microspheres were formed. In addition the $^{235}UO_2$-20% PuO_2 pellets and shards for the initial EBR-II irradiation test were nearly completed. Four capsules were irradiated at 6 to 20 kW/ft for 3 to 81 days in the BAWTR, and another capsule was assembled for irradiation at a later date. Characterization of the fuel being irradiated in the BAWTR during FY-1970 is nearly complete. Progress in all technical areas of the program is reviewed in the following sections.

2. RESULTS AND CONCLUSIONS

2.1. Fuel Preparation

About 6 kg of $^{235}UO_2$ sol and 550 g of PuO_2 sol were prepared by precipitation-peptization techniques. The $^{235}UO_2$ sol was somewhat lighter in color than was previous sol and had a shelf-life of less than a month. Difficulty in thermally denitrating the high-nitrate PuO_2 sols was overcome by treating the initial plutonium feed solution with H_2O_2.

The unit-operation-scale spheroidizing system was operated successfully without the organic regeneration system. Additional operating experience is required to bring the regeneration system on line. About 250 g of coarse and 150 g of fine $^{235}UO_2$-20% PuO_2 microspheres were prepared.

2.2. Fuel Fabrication

The $^{235}UO_2$-20% PuO_2 pellets and shards required for the initial EBR-II irradiation test were nearly completed. Only a small quantity of coarse shards remains to be prepared. Because of a design change in the EBR-II pins, another lot of UO_2 blanket and insulator pellets were fabricated. The density of these pellets averaged 95.1 $\pm$ 1.6% of theoretical.

2.3. Irradiation Testing

Four capsules were irradiated in the BAWTR for from 3 to 81 days at 6 to 20 kW/ft. One of these (GA-9) was scheduled for an 81-day irradiation at a linear heat rate of 6 kW/ft. However, it was removed after five days because of the erratic behavior of thermocouples, which is unexplained.

The stainless steel tubing for the EBR-II pins has been received and ultrasonically inspected. The welding chamber has been modified to handle the 40-inch EBR-II pins, and the welding and xenon-tagging procedures are nearly complete.

2.4. Analytical Support

No difficulty has been encountered in complying with the quality assurance plan written for the FY-1970 fuel order. Characterization of the fuel being irradiated in the BAWTR during FY-1970 is nearly complete.

3. FUEL PREPARATION
(R. V. Carlson)

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 are scheduled for insertion in July 1970 as part of a subassembly to be shared with ORNL. 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.

3.1. Preparation of $^{235}UO_2$-20% PuO_2 Powder

During the past 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.[1] This completed the FY-1970 mixed oxide powder order.

3.2. Preparation of $^{235}UO_2$-20% PuO_2 Microspheres

The formation of sufficient $^{235}UO_2$ and PuO_2 sol to produce 1300 g of sol-gel microspheres was completed. The blending and gelling of these sols in the unit-operation-scale spheriodizing system are in progress.

3.2.1. Preparation of UO_2 Sol

About 6 kg of fully enriched uranium were converted into stable UO_2 sols using the ORNL formate flow sheet.[2] The pertinent run data from the sol preparations are summarized in Table 3-1.

The reduction time in each run was substantially less than normally observed in previous runs. This is attributed mainly to having a urea/uranium mole ratio (in the initial feed solution) of 0.8 rather than the flow sheet value of 0.6.[3] In addition, a new stirring rod was used in batches 158 to 173. This permitted more efficient mixing during the reduction step and also seemed to lower the reduction time.

Table 3-1. Data From Preparation of $^{235}UO_2$ Sols

Batch No.	Uranium reduced, g	Input U conc, M	Reduction time, h
145	100.0	0.47	1.7
146	200.0	0.81	<1.0[a]
147	200.0	0.81	2.0
148	200.0	0.81	2.3
149	200.0	0.81	2.5
150	200.0	0.81	3.0
151	300.0	0.89	3.6
152	300.0	0.89	2.0
153	300.0	0.89	1.8
154	302.1	0.89	2.1
155	295.2	0.89	2.9
156	287.2	0.88	2.4
157	294.5	0.88	2.4
158	295.2	0.88	1.0[b]
159	150.0	0.86	1.2[b]
160	255.3	0.89	1.1[b]
161	278.7	0.87	1.1[b]
162	299.4	0.89	1.9[b]
163	291.4	0.88	1.2[b]
168	298.9	0.89	1.3[b]
169	25.3	0.72	0.7[b]
170	300.0	0.89	2.1[b]
171	300.0	0.89	2.0[b]
172	300.0	0.89	1.3[b]
173	300.0	0.89	2.1[b]

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[a] Over-reduction occurred.

[b] New stirring rod, which allowed greater mixing, was used.

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

Batch No.	Stock solution		Pu input, g	4 M NH_4OH, ℓ	No. washes	Final filtrate, pH	NO_3^-/Pu mole ratio to peptize	Peptizing time, h	Baking time, h	NO_3^-/Pu mole ratio	Yield of denitrated powder, g	Pu recovered as sol, %[a]
	Pu, g/ℓ	H^+, M										
33	25.38	1.00	101.0	2.85	4	7.3	1.23	0.50	–[b]	--	--	--
34	25.37	1.00	101.0	2.85	4	7.2	1.23	0.50	–[b]	--	--	--
35[c]	25.59	1.00	55.1	1.54	5	7.2	1.19	0.50	5.0	0.15	46.4	71.6[d]
36[c]	25.37	1.00	101.0	2.85	5	7.7	1.23	0.50	6.0	0.12	114.7	96.5
37[c]	25.32	1.00	119.0	3.40	5	7.9	1.20	0.50	5.8	0.14	127.2	90.9
38[c]	25.32	1.00	119.0	3.40	5	7.9	1.20	0.50	1.8	0.12	133.4	95.3
39[c]	25.32	1.00	119.0	3.40	5	7.8	1.20	0.50	2.0	0.14	130.5	93.2
40[c]	25.32	1.00	119.0	3.40	5	7.9	1.20	0.75	2.0	0.13	131.1	93.6

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

(b) Baking discontinued because NO_2 fumes were not observed and powder would not resuspend in water.

(c) Pu feed solution treated with H_2O_2.

(d) Yield decreased because of large number of samples removed.

The UO_2 sol prepared during the quarter was somewhat lighter in color than is normally observed. In addition the shelf-life was less than one month, whereas a life of up to six months had been previously observed. The reason for this behavior is not clear, but as a result of the short shelf-life, a large quantity of the sol had spontaneously gelled before it could be run through the spheroidizing system.

3.2.2. Preparation of PuO_2 Sol

About 550 g of plutonium were converted into stable PuO_2 sols using the ORNL precipitation-peptization flow sheet.[4] The operating procedure has been reported,[5] and the run data from these batches are summarized in Table 3-2.

In batches 33 and 34 it was not possible to thermally denitrate the dried powder. This problem was reported previously[3] but has not been explained. It was thought that the problem might be related to a high Pu(VI) content in the initial feed solution. Therefore, the feed solution in batch 35 was treated with H_2O_2 to destroy any Pu(VI) that might be present. This treatment involved heating the feed solution (1.0M in free acid) to 70 C, adding 25 ml of 30% H_2O_2 over a period of five minutes while stirring the solution rapidly, and cooling to room temperature. Samples of the stock solution taken before and after the H_2O_2 treatment were analyzed for Pu(VI) by controlled potential coulometry. The Pu(VI) content in both samples was below the limit of detection (≈1%). Nevertheless, no difficulty was encountered with the thermal denitration step in batch 35, and a stable PuO_2 sol was formed. Therefore, the initial plutonium feed solution in batches 36 to 40 was also treated with H_2O_2, and each of these runs was successful.

3.2.3. Microsphere Formation

The initial effort to prepare $^{235}UO_2$-20% PuO_2 microspheres in the unit-operation-scale spheroidizing system was unsuccessful because of an inability to find the proper surfactant conditions. No difficulty was observed in the laboratory-scale system, which does not contain a still for organic regeneration. Therefore, the large system was operated with the organic bypassing the still.

The following procedure was used:

> A total of 32 ℓ of 2-EH containing 0.5 vol % water and the required surfactant concentrations was placed in the system at the start of each day's run. Then 4 ℓ of wet 2-EH were removed and 4 ℓ of dry 2-EH were added to the system every 30 minutes during operation. All rejected organic was pumped from the box to waste drums.

Using this method, satisfactory microspheres can be formed, and it will be possible to have the FY-1970 fuel order prepared on schedule. Later, additional runs will be made with the system to determine the proper surfactant addition rates while recirculating the 2-EH through the organic regeneration system.

About 250 g of acceptable coarse product and 150 g of fines have been prepared. The remainder of the fuel order will be completed during the next quarter.

4. FUEL FABRICATION
(J. M. Kerr)

4.1. Oxide Characterization (A. H. Bremser)

Sorbed gas release values were obtained for the ORNL microspheres to be irradiated in the BAWTR and for two of the three firings of UO_2 blanket pellets to be used in the EBR-II irradiation specimens. These results are shown in Table 4-1.

Table 4-1. Sorbed Gas Release Values

Type of material	Gas release, cc/g	
	Run 1	Run 2
ORNL microspheres[a]	0.016	0.016
EBR-II blanket pellets, firing 1	0.004	0.005
EBR-II blanket pellets, firing 2	0.004	0.006

[a] Analysis performed on a mixture of 73% coarse and 27% fine microspheres in order to simulate the fuel composition in a pin.

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

The $^{235}UO_2$-20% PuO_2 pellets required for the EBR-II pellet and shard pins were pressed previously.[6] During this report period, they were fired and separated into two lots—one to be used in pellet pins and one to be crushed into shards. The results of statistical analyses of the physical characteristics of these pellets are shown in Table 4-2.

The density of each pellet in the lot to be used in pellet pins was measured to ensure that each was within the specification of 93 $\pm$ 2% of

theoretical. These pellets will be ground, washed, dried, and loaded into the EBR-II irradiation specimens during the next report period.

The other lot of pellets was crushed to produce the shards for the vipac pins, but the quantity of coarse particles prepared was insufficient for the entire fuel order. Therefore, additional $^{235}UO_2$-20% PuO_2 pellets will be produced and crushed during the next quarter.

It was previously reported that all the UO_2 blanket pellets for the EBR-II pins were fabricated and stacked.[6] However, because of a pin design change, additional UO_2 blanket and insulator pellets were required. These were fabricated with an average density of 95.1 $\pm$ 1.6% of theoretical. All the UO_2 pellets will be assembled into 7-inch stacks with an average stack smeared density between 91 and 95% of theoretical.

Table 4-2. Statistical Analysis of Pellet Lots for EBR-II Pins

	Lot for pellet pins	Lot for vipac pins
No. of pellets sampled	30	30
Mean density, % theor	93.02	93.78
Standard deviation	0.60	1.35
95% confidence interval	91.85-94.19	91.13-96.43
Mean diameter, in.	0.211	0.210
Standard deviation	0.0015	0.0016
95% confidence interval	0.208-0.214	0.207-0.213
Mean length, in.	0.247	0.248
Standard deviation	0.005	0.007
95% confidence interval	0.237-0.257	0.234-0.262
Mean weight, g	1.4529	1.4484
Standard deviation	0.0424	0.0461
95% confidence interval	1.3697-1.5361	1.3581-1.5387

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

The gel-addition irradiation task, as described previously,[7] 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.

5.1. Time Required for In-Pile Restructuring

The effect of heat rate and time on the restructuring characteristics of sol-gel PuO_2-UO_2 spheres and on angular particles and pellets prepared from coprecipitated PuO_2-UO_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.

Four capsules (GA-6, GA-9, GA-10, and GA-12) were irradiated during the quarter. Table 5-1 shows the conditions under which these capsules were irradiated, as well as the data from the previously irradiated capsules. Capsule GA-10 contains a pellet fuel pin and two sphere-pac fuel pins. One of these sphere-pac pins was made from spherical sol-gel particles prepared by ORNL and the other from spherical sol-gel particles prepared by B&W. All other capsules contain fuel pins made from spherical particles prepared by B&W and from angular particles and pellets prepared from coprecipitated powder.

Table 5-1. Operating Conditions for BAWTR Capsules

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-12	81[c]	6	4.8 - 6.0	1040	1015 - 1025
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 - 1045
GA-8[a]	3	20	17.0 - 19.8	1150	1050 - 1140
GA-10	3	20	20.0 - 22.0	1150	1120 - 1160
GA-6	9	20	17.0 - 20.0	1150	1060 - 1120
GA-9[b]	5	6	6.2 - 7.3	1040	1140 - 1225

[a] Capsules GA-8, GA-13, GA-14, and GA-15 were irradiated simultaneously.

[b] Capsule GA-9 was to be irradiated for 81 days, but it was removed after 5 days of operation because of indicated abnormal temperatures.

[c] Irradiation in progress.

Capsule GA-9 was scheduled for an 81-day irradiation in the BAWTR at a linear heat rate of 6 kW/ft. During the first five days of operation, the thermocouple in the NaK annulus adjacent to the top fuel pin indicated a temperature of 1140 F, while the thermocouple in the NaK annulus adjacent to the bottom fuel pin indicated a temperature of 1225 F. On the fifth day of irradiation the temperature indicated by the top thermocouple rose from 1140 to 1270 F, while the bottom thermocouple reading remained at 1225 F. Initially, this behavior was attributed to a thermocouple failure. However, approximately 10 hours later the bottom thermocouple reading changed from 1225 F to above 1500 F within a period of 10 to 15 minutes. At this point the capsule was removed from the reactor until this erratic behavior could be understood.

After removing the capsule, the resistance of both thermocouples was checked, and their temperature readings were compared with the pool temperature. Each test indicated that both thermocouples were functioning properly. In addition, neutron radiographs did not reveal any abnormal appearance.

Additional tests will be performed on this capsule to determine the reason for the erratic behavior. However, there are no plans to reinsert it into the BAWTR. Instead, capsule GA-12, which was scheduled for a 27-day irradiation at 6 kW/ft, was substituted for GA-9 and is operating satisfactorily. A new capsule (GA-19) will be fabricated and irradiated under the conditions originally scheduled for GA-12.

Capsule GA-16, fabricated during this quarter, is scheduled for irradiation in April for 1 day at 20 kW/ft. Like GA-10 it contains one fuel pin with ORNL spheres, one pin with B&W spheres, and one pin with pellets.

5.2. 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.

5.3. EBR-II Irradiations

The primary objective of the EBR-II irradiations is to obtain data on the operational characteristics of particulate fuels irradiated in a fast reactor. The effect of heat rate and burnup on the overall performance of sol-gel spherical particles and of angular particles and pellets prepared from coprecipitated UO_2-20% PuO_2 powder will be compared and evaluated. This subtask includes the irradiation of 30 fuel pins in a 19-pin encapsulated subassembly in row 7 of the EBR-II, and the

irradiation of 28 unencapsulated pins in a 37-pin subassembly to be shared with ORNL. The heat rating for the encapsulated and unencapsulated fuel pins will be approximately 8 to 10 kW/ft and 14 to 16 kW/ft, respectively.

The B&W fuel pins proposed for the unencapsulated subassembly are described in Table 5-2. A summary sheet for both the B&W and the ORNL fuel pins to be irradiated in this subassembly is shown in Table 5-3. All three fuel types will consist of UO_2-20% PuO_2, the UO_2 being fully enriched in ^{235}U, and all fuel pins will have a smeared density of 82 $\pm$ 2%. The pellet density will be 93 $\pm$ 2% of theoretical, and the smeared density will be adjusted by varying the radial gap. The fuel column length will be 13.5 inches with a total pin length of 40 inches. The fuel pins will be clad with 0.250-in. OD × 0.016-in. wall Type-316 stainless steel. A modified Mark-J subassembly will be used, so that peak cladding temperatures of 1100 to 1200 F will be attained.

Approval-in-principle has been received for the EBR-II irradiation of the unencapsulated subassembly to be shared with ORNL. The stainless steel tubing has been received and ultrasonically inspected to a defect level of 1 mil × 5 mils × 30 mils. The welding chamber has been modified to accept the 40-in. EBR-II pins for welding and xenon tagging, and the qualification of a welder is in progress. In addition, the welding and xenon-tagging procedures are nearly complete. During the next report period all 28 pins for the unencapsulated bundle are scheduled for fabrication.

5.4. Status of Hot Cell Modification (W. S. Thomasson)

B&W is modifying its hot cell facility to accommodate the post-irradiation examination of PuO_2-UO_2 fuel pins irradiated in the BAWTR and EBR-II under the Gel-Addition Contract. The new alpha-containment box has been constructed, and Cell I has been prepared for the insertion of this box. Equipment checkout is proceeding normally.

The following 10 capsules are available for post-irradiation examination: GA-2, 4, 6, 8, 9, 10, 11, 13, 14, 15. All 10 are scheduled to be placed in the alpha-containment box in March for initiation of the hot-cell examination.

Table 5-2. Proposed Program for 18 Positions Allotted to B&W in 37-Pin Unencapsulated EBR-II Subassembly

Position	Material	Bundle burnup, MWd/t × 10^{-3}				
		0	25	50	75	100
1	Vipac	Pin 1				Pin 38
2	Vipac	Pin 2				Pin 39
3	Sphere-pac	Pin 3				Pin 40
4	Sphere-pac	Pin 4				Pin 41
5	Sphere-pac	Pin 5				Pin 42
6	Pellets	Pin 6				Pin 43
7	Pellets	Pin 7				Pin 44
8	Vipac	Pin 8			Pin 45	
9	Sphere-pac	Pin 9			Pin 46	
10	Pellet	Pin 10			Pin 47	
11	Vipac	Pin 11				
12	Vipac	Pin 12				
13	Vipac	Pin 13				
14	Sphere-pac	Pin 14				
15	Sphere-pac	Pin 15				
16	Sphere-pac	Pin 16				
17	Pellet	Pin 17				
18	Pellet	Pin 18				

	Pins Removed, MWd/t × 10^{-3}				
Type of fuel	25	50	75	100	Total number
Vipac	2	2	2	3	9
Sphere-pac	3	2	3	3	11
Pellet	2	2	2	2	8

Table 5-3. Summary of B&W and ORNL Programs

Type of fuel	Pins removed, MWd/t × 10^{-3}				
	25	50	75	100	Total
Sphere-pac					
ORNL	2	10	2	5	19
B&W	3	2	3	3	11
Vipac					
B&W	2	2	2	3	9
Pellets					
ORNL	0	6	0	4	10
B&W	2	2	2	2	8

6. ANALYTICAL SUPPORT
(D. H. Schmitt)

6.1. Characterization of BAWTR Fuel
(R. H. Dodd and R. J. Boden)

A complete characterization of all fuel prepared for the BAWTR irradiation program (see section 5.2) is in progress. The results for the fuel being irradiated in FY-1970 are shown in Tables 6-1 and 6-2. The characterization of the fuel previously irradiated will be completed during the next quarter.

6.2. Quality Assurance Plan Compliance

6.2.1. Acceptance of Coprecipitated Powder
(R. V. Carlson)

A grab sample was removed from each batch of coprecipitated powder and analyzed in quadruplicate for Pu/(Pu+U). The results are shown in Table 6-3. To be accepted, a batch must have an average Pu/(Pu+U) ratio of 20.0 $\pm$ 0.5%. Using this criterion, batch 8-B-1 was rejected and the remaining 14 batches were accepted.

6.2.2. Acceptance of $^{235}UO_2$ and PuO_2 Sol Blends
(R. V. Carlson)

Since a lot of $^{235}UO_2$ sol and a lot of PuO_2 sol were each prepared by blending several individual sol batches together, a method to ensure the homogeneity of each blended lot was necessary. The following method, based on the natural gamma activity of uranium and plutonium, was chosen:

For the $^{235}UO_2$ sol, eight samples were taken from a flowing stream of sol, a 10-ml aliquot of each sample was transferred to a small polyethylene screw-cap bottle, the weight of UO_2 sol in each bottle was determined, and finally each bottle was sealed in a polyethylene bag. For the PuO_2 sol, three 1-ml samples were withdrawn from each container (one sample each from the top, middle, and bottom), each sample was placed in a small polyethylene screw-cap bottle, the weight of PuO_2 in each bottle was determined, and each bottle was sealed in two separate polyethylene bags. The gross gamma activity of each sample was measured five times for one minute with a 3 in × 3 in. NaI(Tℓ) crystal. The samples were counted in a random order from a stationary platform 7 in. above the detector for the $^{235}UO_2$ sol and 25 in. for the PuO_2 sol. For each lot of sol to be considered homogeneous, the average measured gamma activity of each sample had to be within 0.5% of the average for all samples. The data in Tables 6-4 and 6-5 show that both sol-lots did meet the homogeneity criterion.

It should be noted that the gamma-counting technique used for proving lot homogeneity is relatively simple and rapid, and its precision is comparable to that of other techniques which are tedious and time consuming. The precision of the technique is limited mainly by the statistical nature of radioactive decay processes and the variation of the sample position relative to the detector. The errors in the average c/min/g shown in Tables 6-4 and 6-5 were calculated on the basis of the deviation of each individual count from the average for each sample and therefore include all random errors. The error due to counting statistics is equal to the square root of the total counts accumulated[8] and averaged about ±29 c/min/g (0.25%) for the $^{235}UO_2$ samples and ±440 c/min/g (0.19%) for the PuO_2 samples. These could be reduced by accumulating a larger number of counts. The statistical difference between the total error and the counting error (about 0.3% for the ^{235}U samples and 0.9% for the PuO_2 samples) is primarily caused by the non-reproducibility of the sample position for each count. Although sample placement at essentially the same distance from the crystal for each count presented no problem, maintaining a constant "sample thickness" was difficult because of the presence of the plastic bags (for contamination control), which tended to tilt the sample bottle. Because of

self-absorption of the low-energy gamma rays from these samples, the count rate is extremely sensitive to slight changes in "sample thickness." The data show that this problem was greater for the PuO_2 samples, which were enclosed in two separate bags. If gamma-counting of ^{235}U and Pu samples was done routinely, a technique could be devised that would essentially eliminate the sample positioning error.

The results of this work show not only that the two sol lots were homogeneous, but also that the natural radioactivity of plutonium and uranium can be used advantageously in performing rapid and precise analyses.

6.2.3. Acceptance of EBR-II Fuel Pellets (L. J. Ferrell)

The density of all fired pellets was measured. To be accepted, the pellet density must be 93 $\pm$ 2% of theoretical and the pellet diameter must be 0.206 $\pm$ 0.001 mil. A histogram of the density distribution for the accepted pellets is given in Figure 6-1.

Table 6-1. BAWTR Fuel Characterization

Analysis	$(U,Pu)O_2$ pellets[a]	$(U,Pu)O_2$ shards[a]	$(U,Pu)O_2$ microspheres[a] (420-590μ)	ORNL $(U,Pu)O_2$ microspheres[a] 420-590μ	ORNL $(U,Pu)O_2$ microspheres[a] <25μ
Pu/(Pu+U), wt %	19.93±0.66	19.93±0.66	19.00±0.11	20.74±0.08	20.01±0.15
^{235}U content, wt %	<0.7[b]	<0.7[b]	19.68±0.02	20.16[c]	20.08[c]
O/M, atom ratio	1.975±0.006	1.977±0.007	1.974±0.003	1.994±0.001	1.997±0.003
Moisture, ppm oxide	4.6±0.7	19.8±6.1	11.6±5.4	7.0±1.2	67±15
Sorbed gas, cc/g	0.013	0.094	0.053	0.016[d]	0.016[d]
Density, % theor	91.3 - 95.3	--	96.2±0.9	97.1±0.1	--
Nitrogen, ppm metal	21±11	14±5	12±4	12±3	18±2
Carbon, ppm metal	64±13	35±16	12±2	10±3	21±5
Chloride, ppm metal	5.0±2.7	5.5±2.2	6.0±2.2	5.3±0.7	9.7±1.3
Fluoride, ppm metal	5.1±1.1	4.5±0.6	0.5±0.2	0.7±0.3	0.9±0.8

[a] Errors are for 95% confidence limits on the mean.

[b] ^{235}U content was not determined but should be less than normal.

[c] ^{235}U content determined by ORNL.

[d] Sorbed gas analyses were performed on a mixture of 73% coarse and 27% fine microspheres in order to simulate the fuel composition in a pin.

Table 6-2. Trace Metallic Impurities in BAWTR Fuel[a]

Element	$(U,Pu)O_2$ pellets	$(U,Pu)O_2$ shards	$(U,Pu)O_2$ microspheres (420-590μ)	ORNL $(U,Pu)O_2$ microspheres[a] 420-590μ	<25μ
Aluminum	>500	>500	305	11	31
Beryllium	<0.5	<0.5	<0.5	<0.25	<0.25
Bismuth	<1	<1	--	<1	<1
Boron	<0.5	<0.5	1.7	<0.4	3
Cadmium	<0.5	<0.5	<1	<0.5	4
Calcium	9	<11	--	21	115
Chromium	27	<23	<20	<9	>100
Copper	170	86	46	<2	20
Iron	39	36	620	26	106
Lead	2	6	<1	<1	<1
Lithium	--	--	--	<0.5	<0.5
Magnesium	48	21	<23	<2.5	6
Manganese	3	5	3	<1	3.5
Molybdenum	<34	<34	<25	100	4
Nickel	<11	<11	<20	<10	58
Silicon	28	7	<250	33	120
Silver	<0.5	<0.5	<1	12	11
Sodium	45	<4	24	<1	<1
Tin	10	2	3	4	42
Titanium	<5	<11	<15	<5	20
Vanadium	<1	<1	<1	<1	<1
Zinc	<10	<10	<20	<50	<50

[a]Values given are in ppm of metal and the average of four determinations.

Table 6-3. Quality Assurance Data for Coprecipitated Powder

Batch No.	Pu/(Pu+U), %
8-A-91	20.16±0.08
8-A-96	20.17±0.04
8-A-97	20.05±0.09
8-A-98	20.20±0.08
8-A-99	20.09±0.05
8-B-1	23.00±0.08
8-B-2	20.20±0.24
8-B-3	20.12±0.06
8-B-4	20.14±0.04
8-B-5	20.24±0.05
8-B-6	20.14±0.15
8-B-7	20.33±0.03
8-B-9	20.36±0.06
8-B-10	20.32±0.08
8-B-13	20.29±0.03

Table 6-4. Quality Assurance Data for $^{235}UO_2$ Sol Lot

Sample No.	Sample wt, g	Count 1, c/min/g	Count 2, c/min/g	Count 3, c/min/g	Count 4, c/min/g	Count 5, c/min/g	Average, c/min/g
1	13.2525	11,445	11,371	11,320	11,468	11,428	11,406 ± 60
2	13.3005	11,392	11,400	11,413	11,475	11,493	11,435 ± 46
3	13.2741	11,319	11,337	11,417	11,327	11,416	11,363 ± 49
4	13.3061	11,384	11,356	11,391	11,305	11,311	11,349 ± 40
5	13.2884	11,405	11,319	11,431	11,374	11,419	11,390 ± 45
6	13.2823	11,380	11,403	11,360	11,369	11,402	11,383 ± 22
7	13.2925	11,287	11,334	11,328	11,350	11,415	11,343 ± 47
8	13.2795	11,382	11,382	11,394	11,422	11,473	11,411 ± 38

Overall average = 11,385 c/min/g.

Acceptable sample average = 11,328 - 11,442 c/min/g.

Order in which samples were counted:

Count 1 — 4, 3, 2, 1, 5, 6, 7, 8

Count 2 — 6, 5, 7, 4, 2, 3, 8, 1

Count 3 — 4, 3, 1, 2, 6, 8, 5, 7

Count 4 — 2, 5, 1, 4, 6, 3, 8, 7

Count 5 — 2, 7, 6, 5, 8, 3, 1, 4

Table 6-5. Quality Assurance Data for PuO_2 Sol Lot

Sample No.	Sample wt, g	Count 1, c/min/g	Count 2, c/min/g	Count 3, c/min/g	Count 4, c/min/g	Count 5, c/min/g	Average, c/min/g
8-B-22	1.2100	232,749	231,823	232,946	235,473	236,886	233,975 ± 2116
8-B-23	1.2104	233,325	233,966	234,987	236,583	233,219	234,416 ± 1401
8-B-24	1.1909	233,344	230,498	237,115	235,611	236,694	234,652 ± 2816
8-B-25	1.2136	232,032	230,690	228,776	236,111	236,454	232,813 ± 3374
8-B-26	1.2260	230,857	232,622	232,822	233,987	232,765	232,609 ± 1119
8-B-27	1.2055	233,795	233,901	229,032	235,677	230,912	232,663 ± 2653

Overall average = 233,521 c/min/g.

Acceptable sample average = 232,353 - 234,689 c/min/g.

Order in which samples were counted:

Count 1 — 22, 23, 26, 27, 25, 24

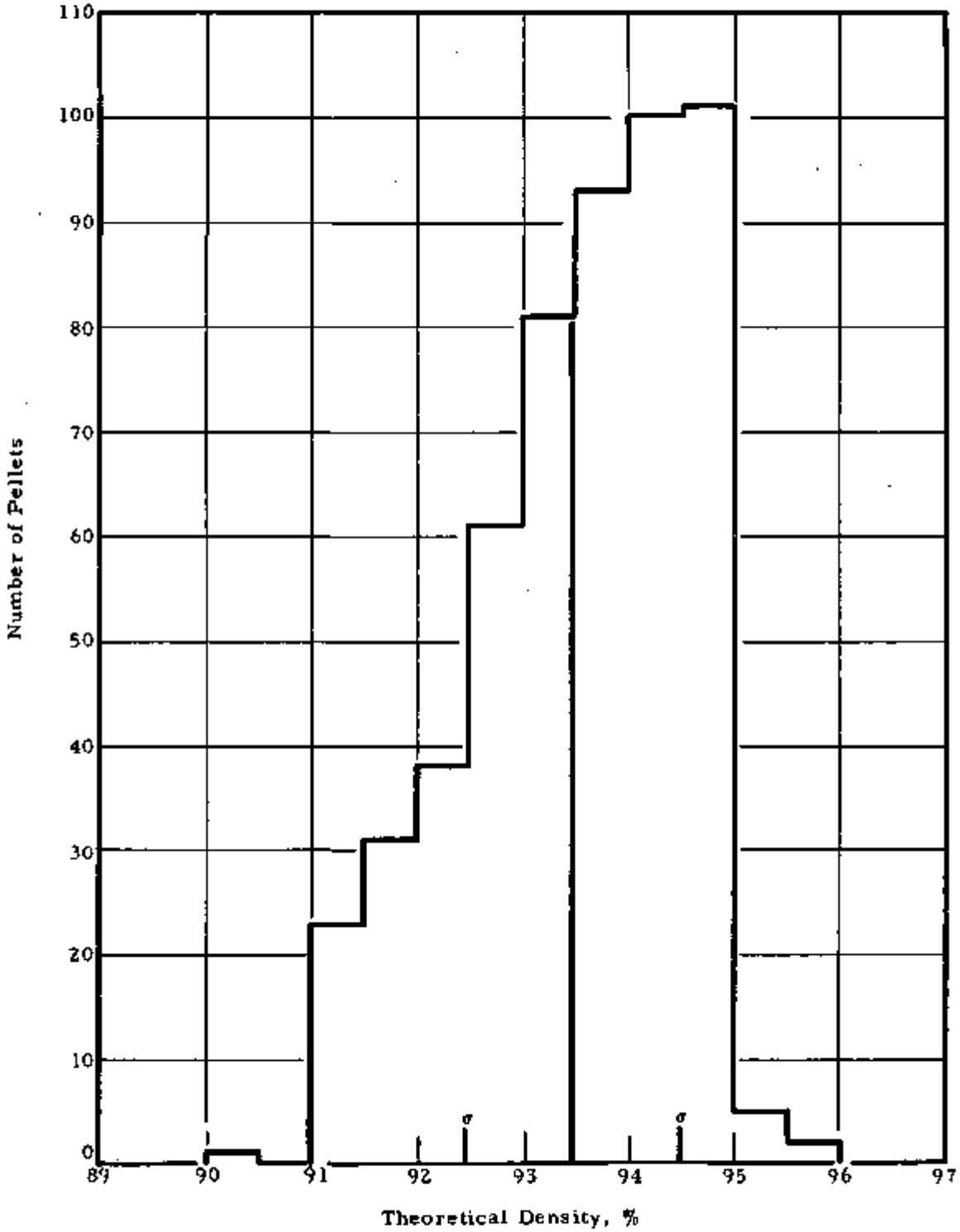
Count 2 — 27, 24, 23, 22, 25, 26

Count 3 — 24, 21, 25, 27, 23, 26

Count 4 — 26, 23, 24, 25, 27, 22

Count 5 — 23, 25, 26, 27, 22, 24

Figure 6-1. Histogram of EBR II Fuel Pellet Density



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