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Department of Commerce
Washington 25, D. C.

ANL-5095

ARGONNE NATIONAL LABORATORY
P. O. Box 299
Lemont, Illinois

December 23, 1953

DETERMINATION OF THE CONVERSION RATIO
OF THE EXPERIMENTAL BREEDER REACTOR
BY RADIOCHEMICAL METHODS

"Production and Consumption Patterns in the EBR"

by M. Levenson

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DETERMINATION OF THE CONVERSION RATIO OF THE EXPERIMENTAL BREEDER REACTOR
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SUMMARY

The Experimental Breeder Reactor, as first loaded and operated, was liberally sampled after 485,000 KWH of operation. Based on radiochemical assays the EBR produced 1.00 ± 0.04 atoms of plutonium for each atom of uranium 235 consumed. For each 100 atoms of uranium 235 consumed in the core (fission plus capture consumption), 110 net atoms of plutonium (total produced minus amount fissioned) were produced. However, during this period 10 atoms of uranium 235 were consumed in the natural uranium blanket, so that the net conversion ratio on an atomic basis is 1.00. The conversion ratio determined by physical measurements as reported in ANL-5222 is between 0.99 and 1.02. An analysis of the neutron losses peculiar to the first loading and design of this reactor confirms the fact that an appreciably higher conversion ratio can be achieved with better design.

Of the total uranium 235 consumed, 79.8 per cent was due to fission in the core and 7.7 per cent to fission in the blankets. The other 12.5 per cent of the uranium 235 consumed was lost by parasitic capture to form uranium 236. Of this 11.1 per cent occurred in the core and 1.4 per cent in the blankets. The "fast bonus" (fission of uranium 238) amounted to 13 atoms of uranium 238 fissioned per 100 atoms of uranium 235 consumed.

The plutonium was essentially all produced in the blankets, only 0.4 per cent of it being produced in the core. The outer blanket production represented 63.9 per cent of the net production and the inner blanket production was 35.7 per cent of the net total. All plutonium production figures are net values, i.e., the amount of plutonium actually present at the time of sampling. No credit is taken for plutonium previously formed and subsequently consumed in the reactor nor is a charge made for the plutonium fissioned.

Values of α , $\sigma_{\text{capture}}/\sigma_{\text{fission}}$, were determined for uranium 235 and plutonium. They were somewhat higher than originally anticipated (ANL-4284). The value of α for uranium 235 in the core varied from 0.11 to 0.17. The plutonium α varied from a value of 0.07 at the inner edge of the inner blanket to a value of 0.55 at the outer edge of the outer blanket.

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INTRODUCTION

The usual analysis and evaluation of a reactor is a very complex problem necessitating many accurate determinations of the neutron flux and neutron spectrum in the various parts of the reactor. From these data and from known cross-sections, the number of events of interest (fissions, captures, etc.) can be computed. The detailed analysis of the Experimental Breeder Reactor (ANL-4356) posed such a problem. As a check on the physical methods of evaluation of the reactor, a program of evaluation by chemical methods was undertaken. It was decided to determine the plutonium production patterns and the uranium consumption patterns by chemical assay. This report describes the analytical methods used and the results thereof.

1. SCOPE OF THE PROGRAM

1.1 Plutonium Production and Quality

The information desired from this part of the program was (1) a knowledge of the total amount of plutonium produced for conversion ratio calculations, (2) a knowledge of the distribution pattern of the plutonium produced to aid in establishing the best loading geometry, and (3) a knowledge of the plutonium quality (ratio plutonium 238/plutonium 240/plutonium 239) to determine the blanket cycle time and the specific activity of the plutonium.

Samples from all parts of the reactor were removed for plutonium assays. Samples of plutonium were isolated, decontaminated and analyzed for the isotopic contents of plutonium 238, plutonium 239 and plutonium 240. The amount of plutonium consumed in the present EBR loading is negligible, but capture data are necessary to evaluate the proposed plutonium fuel loading. Values of the plutonium alpha, $\sigma_{\text{capture}}/\sigma_{\text{fission}}$, were calculated from the plutonium 240 content and the estimated plutonium burnup data.

1.2 Uranium Fission Patterns

The determination of the fission patterns was a substantial problem. The burnup at the time of sampling was too small to be detected by direct uranium analysis or by an isotopic shift in the uranium isotopes. Therefore, fission product analyses had to be utilized. A calibration experiment (Appendix I) was performed to relate the amount of cesium 137 activity produced with the number of fissions of uranium. Samples of the reactor core and blankets were dissolved and analyzed for cesium 137, and the total number of fissions that had occurred in each type of material was calculated. In the enriched core essentially all this fission was due to uranium 235 and it was so attributed. However, in the blankets between 50 and 80 per cent of the total fissions were of uranium 238. This ratio was determined by using the results of a second calibration experiment (Appendix II). The fission yield curve (mass of the nuclide versus per cent abundance) varies with the mass of the isotope being fissioned, large changes occurring in the light element peak and in the valley. Ruthenium 106 was selected as an isotope whose yield was very mass sensitive. Foils of highly enriched and highly depleted uranium were irradiated and the ratios of ruthenium 106 counts to cesium 137 counts were determined for uranium 235 fission and for uranium 238 fission. Since the yield of cesium is essentially constant, the magnitude of the ratio of ruthenium to cesium activities in an unknown sample is a measure of the fraction of the uranium fissions that is due to uranium 235.

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1.3 Uranium Capture Patterns

Uranium 235 is also consumed by parasitic capture yielding uranium 236. Samples of the enriched fuel were decontaminated and analyzed for uranium 236 by mass spectroscopy; samples of this material were also analyzed for cesium to determine the amount of uranium 235 fissioned. Alpha, the ratio of the cross section for capture to the cross section for fission, was thus determined.

2. SAMPLING

The first samples for breeding gain determination were removed from the reactor after it had been operating for approximately 485,000 KWH. Figure 1 is a schematic diagram of the reactor. The samples consisted of seven outer blanket bricks (one complete stack), eight inner blanket rods (two complete radii), seventeen fuel rods from the core (two complete radii), seventeen upper blanket and seventeen lower blanket rods (two complete radii), and two samples from the base slab. Whenever two radii were sampled, one radius was through a flat of the hexagonal core and the other radius was through a vertex. The actual results indicate no difference between such radii. The various reactor components that were used as samples were treated in the following ways:

2.1 Outer Blanket

Each brick was cut into three pieces, each piece being dissolved and sampled separately. These samples represented the inner 24 per cent by weight, the center 41 per cent by weight and the outer 35 per cent by weight of each brick. In addition, holes were drilled in each brick and the chips were dissolved for spot check samples. All of the dissolved metal from the top six bricks was blended and sampled as a check on the integration and the individual analyses.

2.2 Inner Blanket

An inner blanket rod consists of five uranium pellets, each four inches long. The bottom section of the center pellet was dissolved separately as its vertical position approximated the nuclear center of the reactor. One rod was dissolved pellet by pellet to determine vertical variations and the rest of the rods were treated as single samples, the five pellets of each rod being dissolved together.

2.3 Fuel Rods

The center fuel rod of the reactor consisted of four small pellets totaling seven and a half inches in length. The two center pellets were taken as one sample and the two end pellets as another sample. One of the outermost fuel rods consisting of six pellets (ten inches long over-all) was split into three samples -- top two pellets, center two pellets and bottom two pellets. The other fifteen fuel rods were considered as single samples, each rod being dissolved, sampled and processed as a separate unit.

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FIGURE 1
REACTOR COMPONENTS

PLAN

ELEVATION

Core — Enriched Uranium (93.5 % U²³⁵)
All Blankets — Natural Uranium

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FIGURE 2

EBR CONVERSION RATIO DATA
(at 485,000 KWH)

<u>Reactor Component</u>	<u>U^{235} Consumed by Fission (gms)</u>	<u>U^{235} Consumed by Capture (gms)</u>	<u>Pu Produced (gms)</u>
Core (enriched fuel)	17.84	2.48	0.10
Outer Blanket Bricks	0.89	0.16	12.48
Inner Blanket	0.70	0.13	6.85
Upper and Lower Blankets	0.02	0.00	1.23
Base Slab	0.05	0.01	1.05
Control Rods	0.07	0.01	0.95
Total	19.57	2.79	22.66

The conversion ratio of the first loading is therefore

$$\frac{22.66}{19.57 + 2.79} \times \frac{235}{239} = 1.0$$

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2.4 Upper and Lower Blankets

Two upper and two lower blanket rods, one from the center and one from the edge were sampled pellet by pellet to determine the vertical distributions. For the other fifteen holes, the top and the bottom rods at each radius were dissolved together and treated as a single sample.

2.5 Base Slab

Two samples were taken from the base slab. These samples consisted of a canned slug originally placed in a sample hole in the base. For aid in extrapolating the results of these samples across the base, the bottom brick was considered to be an extension of the base slab (see Figure 1).

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3. RESULTS

3.1 Plutonium Production Patterns

The plutonium concentrations were determined by assay as outlined in Appendix III. These results are plotted in Figures 3 through 11 and are listed in Tables 1 through 4.

Figure 3 (data in Table 1) is a plot of the plutonium concentration versus radius for the inner blanket. The upper curve shows the concentration gradient near the center of the blanket (the bottom slices of the center slugs), and the lower curve indicates the average value for each rod. Figure 4 (data in Table 4) shows the variation in the plutonium concentration along an inner blanket rod from top to bottom.

Figures 5 and 6 (data in Table 2) give the plutonium distribution in the outer blanket bricks. Figure 5 gives the radial variation in each of the seven bricks sampled and Figure 6 shows the variation from the top to the bottom of the reactor.

Figure 7 is a curve of the plutonium concentration existing at 485,000 KWH in a fully inserted natural uranium control rod. Since the control rods are inserted from the bottom, the distribution of plutonium in the half inserted control rods is approximately that of the lower half of the curve.

Figure 8 (data in Table 3) is a plot of the upper and lower blanket plutonium production. Because of the small size of these blankets, and of the small amount of material involved, the two blankets (upper and lower) have been treated as one. The plot shows the average concentration existing at each radius (obtained by dissolving the two rods from that radius at the same time). Figure 9 (data in Table 4) shows the vertical variation along two sets of such rods, one rod from the center of the blanket and the other rod from the outer edge of the blanket.

The plutonium production pattern in the base is shown in Figure 10. As only two samples were available from the slab, the data from the bottom blanket brick was used to help define the slope of the curve.

Figure 11 is a composite plot showing the plutonium concentrations at various horizontal distances from the center line of the reactor.

3.2 Uranium Fission Patterns

The total amount of material fissioned was determined by cesium analyses as described in Appendix I. The per cent of the total fissions that were due to the fission of uranium 235 was determined by ruthenium and cesium analyses as outlined in Appendix II. These results are tabulated in Tables 5, 6, and 7 and graphically depicted in Figures 12 through 19.

The amount of enriched uranium consumed by fission in the reactor core is shown in Figure 12 (Table 5). There is no difference between the two radii sampled, as can be seen from this graph.

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FIGURE 3

PLUTONIUM DISTRIBUTION IN
THE EBR INNER BLANKET.
(at 485,000 KWH)

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Table 1

PLUTONIUM PRODUCTION IN THE INNER BLANKET

(at 485,000 KWH)

<u>Radius (cm)</u>	<u>Uranium Weight (kgs)</u>	<u>Plutonium Concentration (ppm) From Fig. 3</u>	<u>Total Plutonium (grams)</u>	<u>Plutonium Concentration ppm by Analysis</u>	
				<u>Reactor Center</u>	<u>Rod Average</u>
11.07	45.0	17.1	0.769	24.6	17.11
11.64	45.0	16.5	0.742		
12.70	22.5	15.4	0.346		15.50
13.20	22.5	14.9	0.335	20.7	14.87
13.44	45.0	14.6	0.659		
14.14	45.0	13.9	0.626		
15.24	22.5	12.8	0.288		12.81
15.45	45.0	12.6	0.567	17.2	12.55
15.86	45.0	12.2	0.549		
16.66	45.0	11.5	0.518		
17.60	22.5	10.9	0.245	14.6	10.72
17.78	67.5	10.8	0.729		10.98
18.32	<u>45.0</u>	10.6	<u>0.477</u>		
TOTAL	517.5 kgs		6.850 gms		

Average plutonium concentration = 13.24 ppm.

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FIGURE 4

PLUTONIUM DISTRIBUTION IN
AN EBR INNER BLANKET ROD.
(at 485,000 KWH)

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Table 2

PLUTONIUM PRODUCTION IN THE OUTER BLANKET BRICKS

(at 485,000 KWH)

<u>Plutonium Concentration in PPM of Uranium</u>				
<u>Position</u>	<u>Average for the Brick</u>	<u>Inner 24 Per Cent by Weight</u>	<u>Center 41 Per Cent by Weight</u>	<u>Outer 35 Per Cent by Weight</u>
7 (top)	3.74	5.591	3.510	2.733
6	5.04	8.186	4.773	3.233
5	5.58	9.009	5.063	3.760
4	4.70	7.492	4.368	3.170
3	3.54	5.371	3.211	2.602
2	2.56	3.65	2.296	2.084
1 (bottom)	1.68	2.022	1.475	1.686
AVERAGE	3.834	5.903	3.528	2.752

Total plutonium production in the bricks is therefore:

$$(\text{concentration}) \times (\text{number}) \times (\text{weight/brick}) = \frac{3.834 \times 84 \times 38.73 \times 10^3}{10^6} = 12.48 \text{ grams.}$$

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FIGURE 5

PLUTONIUM DISTRIBUTION IN THE EBR
OUTER BLANKET BRICKS.
(at 485,000 KWH)

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FIGURE 6

PLUTONIUM DISTRIBUTION IN THE
EBR OUTER BLANKET BRICKS.
(at 485,000 KWH)

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FIGURE 7

PLUTONIUM DISTRIBUTION IN A FULLY
INSERTED EBR CONTROL ROD.
(at 485,000 KWH)

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FIGURE 8

PLUTONIUM DISTRIBUTION IN THE EBR
UPPER AND LOWER BLANKETS.

Table 3

PLUTONIUM PRODUCTION IN THE UPPER AND LOWER BLANKET RODS (at 485,000 KWH)

<u>Radius (cm)</u>	<u>Weight of Uranium (gms)</u>	<u>Plutonium Concentration by Assay (ppm)</u>	<u>Plutonium Concentration (ppm) From Figure 8</u>	<u>Total Plutonium</u>
0	406.9	16.56	16.56	.00674
1.255	2439.8	16.25 16.71	16.45	.04013
2.174	2364.6	16.48	16.38	.03873
2.510	2441.8	16.23	16.33	.03987
3.320	4885.0	16.16	16.28	.07953
3.764	2443.9	16.28	16.23	.03966
4.346	4885.4		16.03	.07831
4.524	2443.9		15.95	.03898
5.019	2443.2		15.80	.03860
5.469	4885.2	15.60	15.60	.07621
5.750	4886.4		15.50	.07574
6.274	2442.0		15.30	.03736
6.520	2444.0	15.27	15.22	.03720
6.640	4782.7		15.18	.07260
6.986	4883.0		15.10	.07373
7.529	2443.8		14.80	.03617
7.633	4885.5	14.79	14.70	.07182
7.836	4648.6		14.25	.06624
8.228	4177.2		13.25	.05577
8.693	2088.6	12.50	12.58	.02627
8.783	6265.8	12.41	12.45	.07801
9.048	4177.2		12.20	.05096
9.473	4177.2	11.97	12.00	.05013
10.038	<u>2088.6</u>		11.70	<u>.02444</u>

84,030 gms

1.233 gms

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Table 4

PLUTONIUM DISTRIBUTION WITHIN BLANKET RODS

(Plutonium concentration in ppm at 485,000 KWH)

<u>Slug Location</u>	<u>Inner(1) Blanket Rod at 17.6 cm. Radius</u>	<u>Upper(2) Blanket Rod at 0 cm. Radius</u>	<u>Lower(3) Blanket Rod at 0 cm. Radius</u>	<u>Upper(3) Blanket Rod at 9.47 cm. Radius</u>	<u>Lower(4) Blanket Rod at 9.47 cm. Radius</u>
Top	5.98	8.86	22.66	7.90	
Next to Top	10.11				
Center	(				
Next to Bottom	(13.97	14.16			14.01
Bottom	9.57	22.89	14.74	13.93	

(1) A five-slug rod -- the center and next to bottom slug were dissolved together and treated as a single sample.

(2) This rod contained three slugs.

(3) These rods contained only two slugs.

(4) This rod consisted of a single slug.

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FIGURE 9

THE PLUTONIUM DISTRIBUTION IN TWO
UPPER AND LOWER BLANKET RODS.
(at 485,000 KWH)

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FIGURE 10

PLUTONIUM DISTRIBUTION IN
THE EBR BASE SLAB.
(at 485,000 KWH)

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FIGURE II

PLUTONIUM CONCENTRATION VERSUS RADIUS
AT THE EBR CENTER LINE.
(at 485,000 KWH)

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Table 5

URANIUM FISSION DATA FOR THE EBR CORE

(at 485,000 KWH)

Radius (cm)	Weight (gms) of Uranium 235	Per Cent of Fuel Fissioned (by analysis)	Per Cent of Fuel Fissioned (Fig. 12)	Amount Uranium 235 Fissioned (gms)	$\alpha \left(\frac{\sigma_{\text{capture}}}{\sigma_{\text{fission}}} \right)$ (Fig. 20)	Amount Uranium 235 Consumed by Capture (gms)
0	221.6	0.0414	0.0414	0.0918	0.109	0.0100
1.26	1326.4	0.0415 0.0412	0.0414	0.5498	0.111	0.0610
2.17	1323.0	0.0406	0.0410	0.5424	0.113	0.0612
2.51	1332.4	0.0413	0.0408	0.5435	0.114	0.0619
3.32	2054.5	0.0396	0.0396	1.0505	0.116	0.1225
3.76	1531.2	0.0396	0.0393	0.5251	0.117	0.0612
4.35	1534.6	0.0390	0.0385	0.5140	0.119	0.0612
4.52	2650.0	--	0.0383	1.0172	0.120	0.1220
5.02	1329.0	0.0375	0.0375	0.4984	0.123	0.0613
5.47	2659.5	0.0368	0.0366	0.7787	0.126	0.1234
5.75	2059.0	--	0.0363	0.9056	0.128	0.1236
6.27	1325.5	0.0354	0.0353	0.4061	0.133	0.0622
6.52	1531.2	0.0345	0.0348	0.4632	0.135	0.0625
6.64	2664.3	--	0.0346	0.9217	0.136	0.1254
6.99	2666.8	--	0.0340	0.9068	0.140	0.1270
7.53	1531.2	0.0332	0.0328	0.4366	0.146	0.0638
7.63	2005.5	0.0325	0.0326	0.8688	0.147	0.1277
7.84	5120.6	--	0.0322	1.0050	0.150	0.1508
8.23	3993.1	--	0.0312	1.2458	0.155	0.1931
8.69	2008.3	0.0274	0.0274	0.5502	0.161	0.0666
8.78	6023.1	0.0271	0.0271	1.6322	0.162	0.2644
9.05	4022.6	--	0.0265	1.0661	0.166	0.1770
9.47	3574.7	0.0256	0.0256	0.9152	0.172	0.1574
10.04	--	--	0.0248	--	0.18	--
<hr/>						
Sub-Total	53,558.73 gms			17.7607 gms		2.4692 gms
Partial loading corrections (fuel rods added after startup)						
9.47	344.9		0.00815	0.0273	0.172	0.0047
10.04	663.2		0.00789	0.0523	0.18	0.0094
<hr/>						
TOTAL	-----			17.84 gms		2.48 gms

Total uranium 235 consumed in the core = 20.3 gms.

FIGURE 12
FRACTION OF FUEL FISSIONED IN
EXPERIMENTAL BREEDER REACTOR CORE.
(at 485,000 KWH)

Figure 13 (data in Table 6) shows the fission patterns in the inner blanket rods. The upper curve is the total material consumption due to fission and the bottom curve is the uranium 235 consumption due to fission. All points on both curves are the average values for an entire blanket rod. The variation of burnup and fission ratios along a single rod is shown in Figures 14 and 14B.

The total amount of material consumed by fission in the outer blanket bricks is shown in Figure 15 (Table 7). Figure 16 gives the per cent of the total fissions that are due to uranium 235 and Figure 17 gives the distribution of uranium 235 fission in the bricks.

Only two samples were available from the base slab. As in the plutonium assay, a sample from the bottom brick was used to help define the fission curves shown in Figure 18 (Table 7). The upper curve is for total fissions, and the lower curve is for fissions due to uranium 235.

The fissions in the upper and lower blankets are plotted in Figure 19. The high percentage of uranium 238 fissions may be due to the streaming of fast neutrons up and down in the core.

All fissions other than those of uranium 235 were attributed to uranium 238, giving a fast fission bonus of 13 per cent.

3.3 Uranium Capture Data

The alpha (σ capture/ σ fission) measurements made on enriched core samples are plotted in Figure 20 (data in Table 8). The alpha value for thermal neutrons (0.183) was used in blanket calculations. The alpha values of uranium 238 are shown in Figure 20A.

3.4 Plutonium Capture Data

The alpha measurements made on isolated plutonium samples are shown in Figure 21 and Table 9. Because of the extremely small amount of plutonium 240 present and the low burnup, these alpha values are only first approximations. It is expected that better values will be obtained at the next sampling.

3.5 Evaluation of Results

The estimated error of the conversion ratio determined by chemical methods is ± 4 per cent calculated as follows: The estimated error in the plutonium production value is ± 2 per cent*; the estimated error in the uranium fissions in the core is ± 2 per cent*; the estimated error in the parasitic capture losses is ± 10 per cent; and the estimated error in the uranium 235 fissions in the blankets is ± 5 per cent. The estimated error in the total uranium consumption is (consumption due to core fissions) (error in fission determination) $\pm$ (consumption due to blanket fissions) (error in determination) $\pm$ (consumption due to capture) (error in determination) $\approx (0.80)(2) \pm (0.08)(5) \pm (0.12)(10) \approx 3$ per cent. The estimated error in the conversion ratio is therefore $\approx$

$$\sqrt{(\% \text{ error in plutonium production})^2 + (\% \text{ error in uranium 235 consumption})^2} \approx$$

$$\sqrt{(2)^2 + (3)^2} \approx \pm 4 \text{ per cent.}$$

* Discussed in detail in ANL-4720.

Table 6

URANIUM FISSION DATA FOR THE EBR INNER BLANKET RODS

(at 485,000 KWH)

<u>Radius (cm)</u>	<u>Uranium Weight (kgs)</u>	<u>Per Cent of Total Atoms Fissioned (From Figs. 13 and 14)</u>	<u>Total Burnup (gms)</u>	<u>Per Cent of Fissions Due to Uranium 235</u>	<u>Uranium 235 Fissioned (gms)</u>	<u>Ruthenium/Cesium Reference Ratio (by Analysis)</u>
11.07	45.0	9.05×10^{-4}	0.407	23	0.094	
11.64	45.0	8.25×10^{-4}	0.371	24	0.090	
12.70	22.5	6.85×10^{-4}	0.154	26	0.040	6.95
13.20	22.5	6.15×10^{-4}	0.138	27	0.038	
13.44	45.0	5.90×10^{-4}	0.265	28	0.073	
14.14	45.0	5.05×10^{-4}	0.227	30	0.067	
15.24	22.5	4.00×10^{-4}	0.090	32	0.029	6.53
15.45	45.0	3.85×10^{-4}	0.173	32	0.055	
15.86	45.0	3.57×10^{-4}	0.161	32	0.052	
16.66	45.0	3.15×10^{-4}	0.142	32	0.045	
17.60	22.5	2.80×10^{-4}	0.063	32	0.020	
17.78	67.5	2.67×10^{-4}	0.180	32	0.058	6.55
18.32	45.0	2.52×10^{-4}	0.113	32	0.036	
TOTAL	517.5 Kgs		2.48 Gms		0.70 Gms	

FIGURE 13
URANIUM FISSION PATTERN
IN THE EBR INNER BLANKET.
(at 485,000 KWH)

RECORDED

FIGURE 14

FISSION DISTRIBUTIONS IN AN EBR
INNER BLANKET ROD (AT 485,000 KWH)
(ROD LOCATED AT OUTER EDGE OF
INNER BLANKET)

RECORDED

FIGURE 14-B

THE RATIO OF U^{235} FISSIONS TO
TOTAL FISSIONS ALONG AN
EBR INNER BLANKET ROD.
(at 485,000 KWH)

REF ID: A65788

Table 7

URANIUM FISSION DATA FOR THE EBR OUTER BLANKET BRICKS AND BASE SLAB

(at 485,000 KWH)

<u>Brick Position</u>	<u>Brick Fraction</u>	<u>Ruthenium/Cesium Ratio (by Analysis)</u>	<u>Per Cent of Total Atoms That Have Fissioned (from Fig. 15)</u>	<u>Per Cent of Total Fissions Due to Uranium 235</u>
1 bottom	Center 41% (Wt.)	4.79	1.81×10^{-5}	56
2	Outer 35% (Wt.)	2.91	5.47×10^{-5}	83
	Whole Brick	3.60	4.17×10^{-5}	73
3	Inner 24% (Wt.)	6.52	6.94×10^{-5}	33
	Center 41% (Wt.)	5.53	4.16×10^{-5}	47
5	Inner 24% (Wt.)	5.68	16×10^{-5}	44
	Center 41% (Wt.)	—	7.31×10^{-5}	—
	Outer 35% (Wt.)	3.11	8.48×10^{-5}	80
6 next to top	Inner 24% (Wt.)	5.68	15.3×10^{-5}	44
	Center 41% (Wt.)	4.94	6.96×10^{-5}	54
	Outer 35% (Wt.)	3.46	6.22×10^{-5}	75
2 - 7	Composite	5.62	6.74×10^{-5}	45

Slab Samples (Cooling Hole Slugs)

Radius

5.62 inches	5.77	3.74×10^{-5}	43
7.5 inches	5.80	3.18×10^{-5}	43

FIGURE 15

TOTAL FISSION PATTERNS IN
THE EBR OUTER BLANKET BRICKS.
(at 485,000 KWH)

REPRODUCED

33

FIGURE 16

PER CENT OF THE TOTAL FISSIONS IN THE
EBR OUTER BLANKET BRICKS DUE
TO THE FISSION OF URANIUM 235.

34
FIGURE 17URANIUM 235 FISSION PATTERNS
IN THE EBR OUTER BLANKET BRICKS.
(at 485,000 KWH)

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FIGURE 18

URANIUM FISSION PATTERNS
IN THE EBR BASE SLAB.
(at 485,000 KWH)

36

FIGURE 19

FISSION PATTERNS IN THE UPPER AND
LOWER BLANKETS OF THE EXPERIMENTAL
BREEDER REACTOR (AT 485,000 KWH)

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Table 8

URANIUM CAPTURE AND ALPHA DATA

<u>Sample Identity</u>	<u>Amount of Uranium 235 Fissioned in the Sample</u>	<u>Uranium 236/Uranium 235</u>	<u>Alpha 235 (σ_{capture}) (σ_{fission})</u>
Fuel Rod - 0 cm. radius	0.041 %	0.000045 $\pm$ 0.000005	0.11
Fuel Rod - 5.02 cm. radius	0.037 %	0.000046 $\pm$ 0.000006	0.12
Fuel Rod - 9.47 cm. radius	0.026 %	0.000044 $\pm$ 0.000002	0.17
* CP-3 ¹ Foil (Thermal Flux)	0.0171 %	0.000031 $\pm$ 0.000002	0.181
* Uranium 235 - 93.5 per cent enriched as received from Oak Ridge	0.0000 %	0.000004 $\pm$ 0.000002	--
Center 1/2 of a fuel rod (0 radius)	0.0445 %	0.000053 $\pm$ 0.000002	0.12
Center 1/3 of a fuel rod (9.47 cm. radius)	0.0260 %	0.000042 $\pm$ 0.000003	0.16

* The fact that the uranium 236 in the virgin material does not measure exactly zero indicates that the values of alpha given above for the EBR may be slightly high. The fact that alpha for a thermally irradiated sample comes out as it does indicates that the method is giving meaningful results.

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FIGURE 20
VARIATION OF URANIUM 235 q IN
THE EBR CORE

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FIGURE 20A
VARIATION OF URANIUM 238 α IN
THE EBR BLANKET

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Table 9

PLUTONIUM QUALITY AND ALPHA DATA

<u>Sample Identity</u>	<u>Per Cent Fission of the Plutonium 239 *</u>	<u>Plutonium 240/Plutonium 239</u>	<u>Alpha</u>
Inner Blanket Rod (radius - 11.1 cm.)	0.016	0.000011 $\pm$ 0.000002	0.069
Inner Blanket Rod (radius - 15.45 cm.)	0.009	0.0000100 $\pm$ 0.0000008	0.11
Inner Blanket Rod (radius - 17.6 cm.)	0.007	0.000009 $\pm$ 0.000002	0.13
Inner 24 per cent (wt.) of an outer blanket brick	0.0042	0.000004 $\pm$ 0.0000001	0.10
Center 41 per cent (wt.) of an outer blanket brick (opposite center of reactor)	0.0028	0.000008 $\pm$ 0.000001	0.29
Outer 35 per cent (wt.) of an outer blanket brick (opposite center of reactor)	0.0064	0.000035 $\pm$ 0.000010	0.55
Composite of the top six bricks	0.0044	0.000011 $\pm$ 0.000002	0.25

* Estimated values.

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FIGURE 21

VARIATION OF PLUTONIUM $\alpha \left[\frac{\sigma_{\text{CAPTURE}}}{\sigma_{\text{FISSION}}} \right]$ IN THE
EXPERIMENTAL BREEDER REACTOR
(AT 485,000 KWH)

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Although the reactor is quite symmetrical horizontally, it is quite evident from both the production and consumption curves that it is not symmetrical vertically. The high flux existing at the top of the outer blanket (as compared to the bottom) indicates that an appreciable number of neutrons were streaming out over the "cup".⁽²⁾ The relatively large amount (4.6 per cent of the total) of plutonium produced in the base slab indicates appreciable leakage through the lower blanket. The upper blanket is designed in the same manner as the lower blanket. From these two observations and the actual measured conversion ratio of 1.00, it is apparent that if the upper half of the reactor were symmetrical with the lower half the conversion ratio would have been substantially greater than 1.

4. CALCULATIONS

4.1 Plutonium Production

To determine the total amount of plutonium produced in the inner blanket, the number of rods at each radius was multiplied by the average rod weight to get the total weight of uranium existing at that radius. This weight was then multiplied by the concentration of plutonium existing at that radius as read from Figure 3. These amounts were then totaled (Table 1) giving 6.85 gms. of plutonium produced or 30.2 per cent of the total plutonium produced. Similar treatment of the upper and lower blankets (Table 3) yields a total of 1.23 grams of plutonium or 5.4 per cent of the total.

The amount of plutonium produced in the outer blanket bricks was calculated by multiplying the average brick concentration (3.83 ppm) by the weight of a brick (38.73 kgs.) and the number (84) of bricks, giving a total of 12.48 grams of plutonium or 55.1 per cent of the total plutonium produced. The amount bred in the uranium control rods was calculated in the same manner. Eleven control rods, each weighing 15.52 kgs. were fully inserted during normal operation and the twelfth rod was inserted halfway⁽¹⁾. The average plutonium content of the fully inserted rods was 5.4 ppm and for the partial rod it was 3.6 ppm. The total amount of plutonium in the control rods was 0.95 grams, or 4.2 per cent of the total plutonium produced.

The average concentration of plutonium in the core was 2 ppm. In the 54-kg. core, the production was 0.1 grams of plutonium or 0.4 per cent of the total.

The total amount of plutonium in the base slab is 1.05 grams or 4.6 per cent calculated as follows:

(1) Personal communication from L. J. Koch.

(2) The number of neutrons lost by leakage from the EBR is estimated (ANL-5222) at 0.37 neutrons per fission.

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$$\text{Wt. Pu} = \int_0^R 2\pi RH \rho C_{\text{Pu}} dR \text{ (for solid slab)}$$

where R = radius of the slab

H = thickness of the slab

ρ = density of the slab

C_{Pu} = plutonium concentration

$$= \frac{5.05 - 0.2805 R}{10^6} \text{ (equation of the line from Figure 10)}$$

$$\text{Wt. Pu} = \int_0^{8.855 \text{ inches}} (A - BR)KR dR = 1.149 \text{ grams (for a solid slab)}$$

The safety plug in the center of the slab is thicker than the slab and a correction of 0.068 grams was added (21.4 kgs. of metal containing 3.2 ppm of plutonium). From this was subtracted the loss due to holes as follows:

at 5 5/8-inch radius, 16 1-inch holes at 3.45 ppm

at 7 1/2-inch radius, 18 1-inch holes plus 0.39 kg. of dowel pin holes at 2.95 ppm

at 2 1/2-inch radius, 14 1-inch holes plus 0.7 kg. of dowel pin holes at 4.35 ppm

at the center, 1 1-inch hole at 5.05 ppm.

The total left is 1.07 grams for a slab mass of 316.7 kgs. The actual weight of the slab plus safety plug is 311.4 kgs., the difference (5.3 kgs.) being rounded corners and counterbores. It was assumed that this weight was uniformly distributed, and the 1.07 was multiplied by the ratio of 311.4/316.7 giving 1.05 grams as the total amount of plutonium actually in the slab.

4.2 Uranium Consumption

To determine the uranium 235 consumption in the core, the weight of the rods at each radius was multiplied by the per cent fission at that radius and the results totaled (Table 5). To this total was added the amount of uranium consumed by capture. (The burnup of each radius was multiplied by the alpha of the radius and these products were added.) The amount of fuel (uranium 235) consumed in the core was 17.8 grams by fission and 2.5 grams by capture or an average of 87.8 per cent by fission and 12.2 per cent by capture (yielding uranium 236).

The consumption of uranium 235 in the inner blanket was calculated in a similar manner (Table 6). The weight of the rods at each radius was multiplied by the per cent fission due to uranium 235 at that radius. For all blanket material, the alpha value used was 0.183 (thermal value). The uranium 235

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consumed in the inner blanket was 0.70 grams by fission and 0.13 grams by capture or a total of 4.0 grams per 100 grams of enriched fuel consumed.

The uranium consumption in the six upper layers of outer blanket bricks was calculated from the composite sample of the top six bricks. The consumption in the bottom layer of bricks was calculated from the extended slab data (Figure 18). The amount of uranium 235 consumed by fission was 0.89 grams, the amount consumed by capture was 0.16 grams or a total of 5.1 grams per 100 grams of enriched fuel consumed.

In a similar manner the uranium 235 consumption in the other reactor components of natural uranium was calculated.

Figure 2 shows the summarized consumption and production data together with the conversion ratio calculation.

ACKNOWLEDGEMENTS

The data reported here represent the work and contributions of many people. In particular, the many suggestions of S. Untermyer proved extremely valuable in carrying out this program. D. Engelkemeir, T. Novey and E. Turk performed the experiments necessary to calibrate the fission product methods; C. E. Crouthamel developed the precision plutonium analysis used; A. H. Jaffey performed the alpha pulse analyses used to determine plutonium 238/plutonium 239 ratios, M. G. Inghram and D. Hess did the mass spectrometric work; the many chemical and radiochemical analyses necessary were performed under the direction of D. Krause and E. Turk; the enriched uranium recovery facility was operated by W. B. Seefeldt. Many other members of the Chemical Engineering Division contributed ideas and did much of the supporting work necessary to the project.

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APPENDIX I

CESIUM 137 RADIOCHEMICAL METHODS FOR COUNTING URANIUM FISSIONS

The activity of the fission nuclide cesium 137 has been used as a means of determining the number of fissions which have occurred in a uranium specimen upon neutron bombardment. All the techniques used have been standardized and calibrated using samples which have undergone a known number of fissions to give a factor relating the amount of cesium 137 as observed counts per minute to total uranium fissions. Unknown samples are analyzed for cesium 137 using the same standardized techniques, and the observed counts per minute of cesium 137 is converted to fissions by the calibration factor whose units are: counts per minute cesium 137 per fission.

This method offers some advantages over the foil activation techniques usually employed:

- (1) The neutron flux does not have to be known.
- (2) The method eliminates the need for accurate fission cross section values.
- (3) Long irradiations can be carried out with minor corrections necessary for fission nuclide decay during irradiation.
- (4) Samples can be analyzed at any convenient time after irradiation.
- (5) All counting can be done on a relative basis eliminating the difficult task of absolute counting.
- (6) The method gives an integrated value of the fissions with only very rough estimates of time needed for corrections.

The major disadvantage is that the minimum usable irradiations are longer than those usually necessary for foil activations.

A. Experimental

1. Equipment

An all glass filtration apparatus shown in Figure 23 is used for collecting the isolated cesium 137 samples. These samples after drying and weighing are mounted on an aluminum card (2 1/2 x 3 1/4 inches) as shown in Figure 24.

The beta activity of the sample is measured by means of an Amperex 100 C. halogen quenched GM tube mounted in a standard GM tube mount. The counting equipment is shown in Figures 24B and 24C. A special high voltage supply (+ 1 volt per 24 hours at 1500 volts) was used to stabilize the counters; this would not be necessary if one were willing to calibrate the equipment each time it is used. This regulated voltage supply has made it necessary to calibrate the equipment only once a month, and in two years of operation the equipment has been stable in the counting rate of standards (better than ± 1 per cent on uranium metal standards).

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FIGURE 22

THE VARIATION IN FISSION PRODUCT
COUNTING RATIOS WITH DIFFERENT
 $U^{235} - U^{238}$ FISSION RATIOS.

*This ratio is the empirical one obtained by calibration and counting — it is not a fission yield ratio.

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Fig. 23
Filtration Apparatus

Note: Only critical dimension is diameter of chimney. This defines an area of 2.0cm^2 .

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Fig. 24
Sample Mount

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FIGURE 24B
COUNTING EQUIPMENT

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FIGURE 24C

COUNTING PLATE HOLDER

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2. Calibration Procedure

Six samples of enriched uranium metal were irradiated in the fission counting apparatus of Freedman and Steinberg⁽¹⁾. These samples were dissolved and analyzed for cesium 137 activity as described in the next section. This procedure gave a factor relating counts per minute cesium 137 to fissions. The data in Table 10 represents the average values obtained from the six samples using two identical counting arrangements which were matched to count with ± 2 per cent variation on the same sample. All samples were counted on both counters to avoid errors caused by faulty counter performance.

3. Sample Analysis

The sample is dissolved and aliquots are taken for cesium analysis using the perchlorate procedure of Glendenin and Nelson⁽²⁾. The aliquot size is selected to give a sample whose counting rate will be 2000-4000 counts per minute.

The final sample is collected using the filtration apparatus shown in Figure 23. After drying and weighing the sample is mounted as shown in Figure 24. The sample is counted on the second shelf of the GM tube mount.

To be sure that the samples contain no significant amounts of cesium 136 (13-day half-life) all samples are allowed to decay at least 60 days after irradiation.

B. Calculations

1. Calibration Factor

The cesium 137 counting data of the six calibration samples are used to give a calibration factor as follows (see Table 10):

$$K_{Cs^{137}} = \frac{(\text{counts per minute } Cs^{137} \text{ per sample})}{(\text{fissions per sample})}$$

2. Sample Analysis

The cesium 137 activity of a sample is converted to fissions as follows:

$$\text{fissions} = (c/m \text{ } Cs^{137})(\text{decay corrections}) \left(\frac{\text{Volume of sample solution}}{\text{Volume of aliquot}} \right) \left(\frac{1}{K_{Cs^{137}}} \right)$$

-
- (1) Radiochemical Studies: The Fission Products by Coryell and Sugarman, Book 3, Paper 200, "Absolute-Fission-Yield Experiments" by M. S. Freedman and E. P. Steinberg, Figure 200.2.
- (2) Radiochemical Studies: The Fission Products by Coryell and Sugarman, Book 3, Paper 283, "Perchlorate Method for the Determination of Cesium Activity in Fission" by L. E. Glendenin and C. M. Nelson.

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Table 10

Calibration Sample Data

<u>Sample Number</u>	<u>Weight of Sample (gms)(a)</u>	<u>Total Fissions (x 10⁻¹²)</u>	<u>Counts per Minute Cesium 137 Observed</u>	<u>Calibration Factor (Counts per Minute Cesium 137 per 10¹² Fissions)</u>
1	3.7953	20.60	1905	92.5
2	3.8189	20.21	1942	96.1
3	3.7893	22.15	2078	93.8
4	3.7827	20.76	1917	92.3
5	3.8076	19.21	1805	94.0
6	3.8043	12.43	1134	<u>91.2</u>

93.3 $\pm$ 1.8 (Standard
Error)

(a) Isotopic composition of uranium used:

1.1 per cent uranium 234, 93.5 per cent uranium 235,
5.4 per cent uranium 238.

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APPENDIX II

RADIOCHEMICAL METHOD FOR DIFFERENTIATING URANIUM 235 AND URANIUM 238 FISSIONS

The ratio of the two fission nuclides ruthenium 106 and cesium 137 has been calibrated as a measure of the ratio of uranium 235 to uranium 238 fissions. This ratio based on the latest published fission yields varies by a factor of six. The fission yield of cesium 137 is almost constant for both uranium 235 and uranium 238 fission at about 6 per cent, but the fission yield of ruthenium 106 varies making the ruthenium 106/cesium 137 ratio a sensitive indicator of the source of the uranium fissions. The change in the fission yield of ruthenium 106 is largest in going from uranium 235 to uranium 238 fission, but it also varies with neutron energy so the method must be calibrated using uranium 235 and uranium 238 foils and neutrons of approximately the same energy as will irradiate the samples.

A. Experimental1. Equipment

The equipment needed is the same as described in Appendix I, plus two aluminum absorbers, 164 mg./cm.² and 2050 mg./cm.² (for counting the samples).

B. Procedure1. Calibration

Two standard uranium samples are irradiated with neutrons whose energy spectrum is similar to that used on the unknown samples. One sample is enriched uranium (93 per cent uranium 235) and the other depleted uranium (10 to 40 atoms uranium 235 per 10⁴ atoms uranium 238) to provide samples of essentially pure uranium 235 and uranium 238 from which the ruthenium 106/cesium 137 ratio can be established for these two sources of fissions. The results of one such calibration are shown in Figure 22.

2. Analyses

The calibration samples are dissolved and aliquots taken for cesium 137 and ruthenium 106 analyses. The cesium analysis is done as described in Appendix I and the ruthenium analysis is done by the distillation method of Glendenin⁽¹⁾. The final samples are collected using the apparatus shown in Figure 23 and mounted as shown in Figure 24.

The beta counting of the cesium samples is done on the second shelf of the GM tube mount using no added absorber.

The ruthenium samples contain two fission nuclides of importance, ruthenium 106 (one-year half-life) and ruthenium 103 (40-day half-life).

(1) Radiochemical Studies: The Fission Products by Coryell and Sugarman, Book 3, Paper 260, "Improved Determination of Ruthenium Activity in Fission" by L. E. Glendenin.

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To get a count representative of only the ruthenium 106 content of the sample, counts are taken through 164 mg./cm.² and 2050 mg./cm.² (sample on the second shelf and absorber on the first shelf). The difference in these two counting rates is proportional to the ruthenium 106 content of the sample.

C. Calculations

1. Calibration Factors

The counting data of the ruthenium 106 and cesium 137 isolated from the calibration samples are used as follows to give calibration factors:

$$K_{U238} = \frac{(c/m/ml \text{ Ru}^{106})}{(c/m/ml \text{ Cs}^{137})_{U238}} \qquad K_{U235} = \frac{(c/m/ml \text{ Ru}^{106})}{(c/m/ml \text{ Cs}^{137})_{U235}}$$

D. Sample Analysis

The counting data of the ruthenium 106 and cesium 137 isolated from unknown samples are treated as follows:

$$\frac{(c/m/ml \text{ Ru}^{106})}{(c/m/ml \text{ Cs}^{137})_{\text{sample}}} = (X \cdot K_{U238}) + (1 - X) K_{U235}$$

where X = fraction of fission in uranium 238 atoms

$1 - X$ = fraction of fission in uranium 235 atoms.

For breeding gain calculations, values of X were taken from Figure 22.

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APPENDIX III

URANIUM AND PLUTONIUM ANALYTICAL METHODS

A. Uranium Analytical Methods

The cesium, ruthenium and plutonium analyses used to calculate the conversion ratio are all expressed as results per volume of solution. To convert this data to the absolute basis needed, the uranium content of these solutions must be determined with the same accuracy as the other analyses.

The uranium content of the solutions was measured by two independent procedures and the results averaged. The results varied by about one per cent for the two procedures although each method had a precision better than this. The two procedures used were the acetone-thiocyanate precision colorimetric procedure (described in reports ANL-4750 and ANL-4940) and the potentiometric titration procedure described below.

Potentiometric Uranium Procedure

The apparatus used for the titration is shown in Figure 25. The sample to be titrated (approximately 2 mg. uranium) is placed in a 25 mm x 100 mm round bottom test tube and fumed to dryness twice using 3 ml. 5 per cent H_2SO_4 each time (to remove any nitrate ion present in the sample). A stirring bar and 5 ml. 0.5 N H_2SO_4 are added to the test tube, and it is placed in position as shown in Figure 25.

Carbon dioxide gas is passed over the sample during the titration to exclude air. The sample is heated to 80 to 90°C. by means of the nichrome heating coil and 2 to 3 drops of chromous ion are added from the 10-ml. syringe. (The chromous ion is prepared by filling the syringe containing amalgamated zinc with a saturated chrome alum solution thus reducing the chromium to the chromous ion.) When the reduction of uranyl to uranous ion is complete and excess chromous ion is present, the pH meter will indicate an EMF of 1000 millivolts. The solution is then titrated with 0.01 N ferric sulphate from the burette. The solution is stirred by means of a magnetic stirrer. Two end points will be observed, chromous to chromic at 875 to 925 millivolts and uranous to uranyl at 400 millivolts.

To obtain high accuracy and precision on a micro scale, the ferric solution to be used is first standardized against a sample of known uranium content to measure the uranium equivalent of the ferric solution. This result is expressed as milli-moles uranium per ml. ferric sulphate solution. The uranium content of unknown samples is measured by titrating them with the calibrated 0.01 N ferric sulphate and multiplying the ml. of titrant used by its calibration value (milli-moles uranium per ml.).

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FIG. 25

POTENTIOMETRIC URANIUM TITRATION APPARATUS

B. Plutonium Analytical Method

The plutonium concentrations in the EBR solutions were determined by counting the plutonium alpha particles. Before counting, the plutonium was separated from uranium and fission products by extraction with 0.5 M thenoyltrifluoroacetone (TTA) in xylene. The specific activity of pure plutonium 239, 1.36×10^5 disintegrations per minute per microgram, was used in all calculations as mass spectrographic analyses and alpha pulse analyses showed negligible amounts of plutonium 238 or plutonium 240.

1. Plutonium Extraction Procedure

Four individual aliquots of each sample were extracted and four separate samples from the organic phase of each extraction were mounted giving a total of sixteen counting plates per sample. The TTA used was recrystallized twice from cyclohexane before use.

Sample aliquots of 0.750 or 0.900 ml. were used throughout the work and these were diluted to either 5 or 10 ml. in order to achieve a final acidity of 0.5 to 0.6 M just prior to extraction. The amount of hydroxylamine necessary for reduction of the plutonium to the III state and the sodium nitrite necessary for the re-oxidation to the IV valence state were calculated and added to the sample before dilution to the standard volume. The samples were extracted with an equal volume of the TTA-xylene reagent. The exact procedure was as follows:

- a. Pipet aliquot into a 50-ml. Dri-Filmed volumetric flask and add previously calculated quantity of water or nitric acid and calculated volumes of hydroxylamine-hydrochloride solution. Shake flask and contents for 10 minutes.
- b. Add calculated volume of sodium nitrite solution and again shake for 10 minutes.
- c. Add to the flask, using calibrated equipment, a volume of 0.5 M TTA in xylene equal to the volume of aqueous solution. It is necessary that the volume of the TTA-xylene be accurately known for purposes of final calculations.
- d. Shake flask and contents for a period of 20 to 30 minutes.
- e. Allow phases to separate and mount four 0.200 ml. aliquots of the organic phase on polished stainless steel discs. Rinse pipet three times with xylene and add rinses to disc. Evaporate to dryness on special plate dryer. (ANL drawing No. CE-9G3-3429)
- f. When dry carefully flame plates to red heat over a burner.
- g. Submit plates to counting room for counting.

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2. Calibration of Counting Equipment

Two Nuclear Measurements Corporation PC-3 alpha counters were used throughout the work and were calibrated against a low geometry alpha counter. Since the geometry of the low geometry counter was very accurately known, the absolute disintegration rate of a standard plutonium plate could be determined. This plate was then placed in the counting chamber of the PC-3 instrument and an operating plateau versus voltage was determined at a sensitivity of 1 MV. When the plateau was established, the voltage was set at a point only about one-third of the distance along the plateau in order to minimize the interference of beta-gamma background. A series of counts was taken at the established voltage to determine the geometry, and the so determined geometry value was used in all calculations. A secondary standard was then calibrated on the PC-3 instruments and used for all routine checking. Background counts and counts on the secondary standard were taken every four hours (start, middle and end of each shift). A high level cesium 137 plate was placed in the counting chamber daily to check the beta-gamma sensitivity of the counters.

3. Sample Counting

A sample plate was placed in the counting chamber and the chamber was flushed with P-10 gas (90 per cent argon, 10 per cent methane) for a period of one minute prior to counting. All plates were counted for 20,000 counts or for five minutes, whichever was greater.

4. Calculations

The counting values from the four plates of each extraction were averaged and reported as a single value. The average of the values from the four extractions on each sample was used to calculate the plutonium concentration.

The calculations of the plutonium concentrations were carried out as follows:

$$\frac{(\text{Sample}) (\text{TTA})}{(\text{Aliquot, ml.}) (\text{Aliquot, ml.}) (\text{Geometry of Counter}) (1.36 \times 10^5 \text{ d/m/}\mu\text{gm. Pu}) (\text{mg. U/ml. Solution})}$$

$$= \frac{\mu \text{ gm. Pu}}{\text{mg. U}}$$

or P.P.M. of plutonium in uranium.

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APPENDIX IV

ENRICHED URANIUM RECOVERY FACILITIES

Introduction

When the decision was made to determine the EBR breeding gain by radio-chemical methods, it was also decided to erect facilities at Argonne capable of decontaminating and recovering the enriched uranium used in the analyses. Such facilities were subsequently designed and erected and were ready for operation at the end of the summer of 1951. The operating area of this unit is shown in Figure 26. The equipment installed provided means of dissolving EBR fuel elements, decontaminating the enriched uranium dissolver solution by a solvent extraction process, reducing the product solution volume so that the final concentration was 200 to 300 grams of uranium per liter of solution, removing residual trace fission products from and clarifying of the product solution by passing it through a silica gel column, reducing the volume of the fission product containing aqueous waste stream from the extraction step, and sampling all vessels. The flow sheet for this process is shown in Figure 27.

A. Design Criteria

The original design philosophy called for a continuous process in which all equipment was to be designed for "infinite safety" relative to uranium 235 criticality. A preliminary economic study caused abandonment of this design in favor of a design in which batch size was to be limited to less than one-half of criticality. Other process considerations affecting design were:

- (1) Fuel rods were to be de jacketed from the stainless steel jackets, rinsed in alcohol to effect NaK removal and placed in aluminum cans at the reactor site prior to shipment to the Chemical Engineering Division at Argonne.
- (2) Decontamination was to be sufficient to reduce both beta and gamma activity of the product solution to less than twice that of unirradiated enriched uranium.
- (3) Capacity for three-shift operation to be about one kilogram of uranium per day.

B. Process and Equipment1. Dissolution

Slugs were charged to the dissolver with aluminum shipping cans intact. A single step dissolution was accomplished using boiling nitric acid containing mercuric ion catalyst. In six to eight hours cans and slugs were dissolved completely; no metal heel was allowed to accumulate in the dissolver.

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FIGURE 26

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RECOVERY FACILITY CONTROL AREA

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FLWSHEET FOR ONE-CYCLE UNIT SOLVENT EXTRACTION PLANT.

2. Sampling

The sampling unit used for sampling dissolver solution, and all other process vessels, contained several features not previously incorporated in samplers. A standard circulating liquid system, in which vacuum is furnished by an air jet, is utilized. The novel features of this unit are:

- (1) It is so constructed that a sample is taken while the sample vial is in a shielded carrier. Hence, the operator does not need to transfer the vial from a sampler to a carrier. This eliminates personnel exposure and the danger of dropping the vial.
- (2) The vial is capped before the sample is taken. Again, the operator is subjected to no exposure.
- (3) The sample vial is in a secondary container at all times.
- (4) Provisions are included for returning unused sample portions to the process and for rinsing the vials. Both operations are accomplished without removing the vials from the carrier. This eliminates the disposal problem of unused samples, makes possible the easy recovery of the fissionable material involved, and simplifies vial disposal.
- (5) The sampler is incapable of sampling unless the vial is tightly capped.
- (6) The construction is such that key parts can be replaced easily and rapidly. This sampler is described in greater detail in ANL-4820, pages 50 through 55.

3. Decontamination

A one-cycle solvent extraction process was used to decontaminate the uranium from the dissolver solution. The process utilizing 0.4 molar tributyl phosphate in carbon tetrachloride as the extractant was developed for this recovery at ANL, and is described in ANL-4530.

The primary decontamination equipment consists of two packed columns fabricated from 1-inch stainless steel pipe. The first column consists of a 23-foot extraction section and a 12-foot scrubbing section. The second column consists of a 20-foot stripping section. The columns are packed with 1/4-inch stainless steel raschig rings. Milton-Roy pumps are used for motivating all non-radioactive process streams. The fission product and uranium-containing feed was fed to the process by an ANL-designed "Egyptian water wheel". The advantages of this device are:

- (1) The elimination of check valves with the resulting elimination of rate changes due to improper valve seating;
- (2) Elimination of bellows, seals, packing glands, and other like sources of leakage;
- (3) Elimination of flow rate changes due to possible particulate matter in the feed stream.

This metering device is described in ANL-4872, page 49.

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4. Product Volume Reduction

The product of a solvent extraction run, 30 to 40 liters of aqueous solution, was reduced in volume to about one-half liter in a specially designed batch evaporator. Heat was supplied by a bottom steam jacket only. Side wall temperatures were always less than that of adjacent vapor or liquid. Resulting condensation occurring on these walls prevented the crystallization of uranyl nitrate. A slightly sloping bottom caused the evaporation to be self-quenching, thus eliminating concern over evaporating the solution to dryness. A spray ring on the under side of the cover made thorough rinsing of the unit possible.

5. Product Purification

The concentrated product solution containing residual traces of fission products, primarily zirconium and niobium, and a slight amount of solids, was passed through a three-foot bed of graded silica gel. This both clarified and further decontaminated the product. The silica gel column was backwashed with hot oxalic acid to remove the zirconium and niobium fission products.

6. Aqueous Waste Volume Reduction

The fission products containing aqueous wastes from the solvent extraction step were reduced in volume about threefold in the collection tank. Design was such that in the event of continuous operation, material could be evaporated concurrently with the operation of the extraction columns.

7. Ventilation

All equipment is ventilated and the off gases are scrubbed. The off gas system is actuated by a standard steam jet. The flow of gas is from vessels and other equipment to a central header, through a caustic scrub tower, thence to the actuating steam jet. The jet discharges into a tube and shell condenser. Following the condenser, the non-condensables pass through CWS filters to the atmosphere. Condensate passes through a mixed bed ion exchange column packed to a depth of three feet to remove any residual activity. The resin column effluent is discharged directly to the suspect drain.

The cell air was exhausted via CWS filters to the atmosphere.

C. Product Quality and Recovery

The beta and gamma activities of the product solutions in no case exceeded twice that of unirradiated enriched uranium. The average material balance for the first 20 runs was 100.4 per cent. The average recovery was 100.1 per cent. The average loss for the solvent extraction cycle plus associated steps was 0.006 per cent. The amount of uranium retained by the silica gel column, recoverable by recycling the backwash, averaged 0.1 per cent.

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