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K REACTOR PU-238 DEMONSTRATION RESULTS
(Interim Report PTA-063)

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(Interim Report PTA-063)

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K REACTOR PU-238 DEMONSTRATION RESULTS

INTRODUCTION

During the first half of FY-1968, a test irradiation¹ of neptunium was performed in KE Reactor. The basic objectives of this test were to (1) show the reactor capability for irradiating neptunium target elements in an elevated flux; (2) demonstrate the charge-discharge of several of the target columns during reactor operation; and (3) establish the Pu-238 production characteristics of the K-type reactor. These objectives were successfully met. This document reports the results and conclusions derived in meeting the third objective - establishing K reactor Pu-238 production characteristics.

SUMMARY AND CONCLUSIONS

The series of major benchmarks which were attained by the irradiation are as follows:

1. Demonstration of the Operational Fueling Facility was carried to a successful conclusion.
2. Enhancement of neutron flux to approximately 150% of normal (9.5×10^{13} neutrons/cm²-sec) was realized.
3. K Reactor Pu-238 production characteristics were established for this specific mode of operation. Of principal significance is the confirmation that the high moderator temperature in the reactor caused a reduction in the burnout-to-formation neutron absorption cross section for Pu-238. The results attained generally show a high degree of analytical precision, and are seen to be favorably consistent with the monoenergetic theoretical model used for prediction.

The collaboration between calculated and analytical results has permitted normalization of the theoretic model and assures reliable prediction of the production parameters for other operating modes.

Graphic representation of the production characteristics is given in the various figures at the end of this document. A report of the conclusions pertaining to the amount of Pu-236 contaminant found in Pu-238 product, not given in this report, will be issued as soon as the analyses are completed.

TEST DESCRIPTION

The neptunium irradiation test block was located on the near side of the central zone of KE Reactor. The zone was a 10 x 10 array which included a 32-tube zone consisting of seven neptunium columns and 25 support enrichment columns. This

1. DUN-2272, "Production Test Authorization 063, Quick Turnaround Neptunium Demonstration," RE Baars, JC Nosler, JP Schmidt, 4-28-67 (Secret)

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zone was then surrounded by a flux-increasing buffer zone. Owing to the high specific powers developed, driver columns were appropriately shortened to maintain tube power limits.

Five target columns were used in the irradiation of 1960 grams of Np-237 in the form of neptunium-aluminum alloy. An additional 680 grams of neptunium were also irradiated in two columns utilizing graphite² as the target element matrix. Fifty aluminum and 16 graphite target elements were charged; the neptunium concentration was approximately 56 grams per foot of column length.

IRRADIATION HISTORY

The first column was charged in May, 1967. The charge and discharge schedule of all neptunium bearing columns is given in Figure 1, Page 5.

DATA COLLECTION

To obtain data at various exposures, columns were irradiated for various time intervals as shown in Figure 1; further variation in nvt was also obtained by analysis of small wafer samples cut from target elements which were in the environment of a high flux for a long duration or low flux for a short duration. Utilization of these two approaches enabled an effective exposure range from 250 Mwd per adjacent fuel column to 56 Mwd per adjacent fuel column.

The 50 aluminum target elements were dissolved and separated utilizing the "C" hot cell facility in Pacific Northwest Laboratories. To acquire data prototypical of a full production mode of operation, dissolver solutions which contained the entire contents of an individual column were processed and sampled separately. After dissolution, the solution was mixed to insure homogeneity and then sampled five times. Three of these redundant samples from each dissolution were submitted to the BNW radiochemistry department for analysis. One sample from two of the dissolutions was delivered to ARHCO analytical laboratories for redundant analysis by a separate laboratory. The raw data obtained in both laboratories is presented in Table 3, Page 14.

Annular wafers approximately 1/8" thick and weighing about five grams were cut from the interior* of various target elements and dissolved for analysis. (Note: reactor location of sampled elements can be found by referring to Table 2, Page 13.)

Measurement of neutron flux and cadmium ratio was accomplished by fitting four neptunium targets and three Tm_2O_3 ² targets with low concentration cobalt flux wires, some of which were enclosed in a cadmium sheath. Post-irradiation analysis of cobalt activation followed by data reduction utilizing program FOILS³ reveals the flux wire results given in Table 5, Page 16.

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*Here interior is used to denote core material greater than 1" from the end of the element.

2. DUN-2272 SUP 3, "Supplement to PTA-063, Quick Turnaround Neptunium Demonstration," JC Nosler, 8-21-67 (Secret)
3. HW-81367, "FOILS: A Program for Computing Neutron Exposures from Foil-Activation Data," WC Morgan, April, 1964 (Unclassified)

Figure 1

NEPTUNIUM IRRADIATION TIME SCHEDULE

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*Authorized by DUN-2272 SUP 1, "Supplement 1 to PTA 063, Quick Turnaround Neptunium Demonstration," JC Nosler, 5-26-67 (Secret)

**F.P.D. = Full Power Days

DATA ANALYSIS

Np-237 burnup, Pu-238 buildup, plutonium isotopic, and the efficiency of conversion are the four basic parameters on which models have been constructed for effective representation of production. The first step in the calculation of the above mentioned parameters is the measurement of the initial amount of neptunium associated with each analysis. This can be calculated from the raw data as:

$$Np \text{ (initial)} = Np \text{ (final)} + Pu \text{ (total)} + C \text{ (fission products)}$$

where: the fission products were determined from the cesium concentration assuming the fission yield of cesium-137 and its precursors is .0657. From this,

$$C = 5.98712 \times 10^{-21} \text{ grams neptunium destroyed/atom of cesium-137.}$$

The appropriate production parameters are then related to the measured isotopic concentration as follows:

$$1-\beta = \frac{\text{neptunium destroyed}}{\text{neptunium charged}} = \frac{Pu \text{ (total)} + F.P. \text{ (cesium)}}{Np \text{ (final)} + Pu \text{ (total)} + F.P. \text{ (cesium)}}$$

$$\alpha = \frac{Pu^{238} \text{ produced}}{\text{Neptunium charged}} = \frac{Pu^{238}}{Np \text{ (final)} + Pu \text{ (total)} + F.P. \text{ (cesium)}}$$

$$\epsilon = \frac{\alpha}{1-\beta} = \text{conversion efficiency} = \frac{Pu^{238}}{Pu \text{ (total)} + F.P. \text{ (cesium)}}$$

$$Q = Pu^{238} \text{ isotopic abundance}$$

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Production parameters from raw data obtained from wafer samples and dissolver solutions are given in Tables 6 and 7. Graphics representation of the rate of neptunium burnup is presented in Figure 2. The ordinate in this graph is a measure of full power operation for specific columns. The abscissa is $1-\beta$ as defined above. Figures 3, 4, and 5 are graphs showing the correlation between α , ϵ , and Q with neptunium burnup. In these graphs, $1-\beta$ is the abscissa. The solid curves on the graphs referenced above are theoretical predictions based on data obtained from program PUPA.⁴ The major parametric factors used were a flux of 9.5×10^{13} neutrons/cm²-sec and a Pu^{238} to Np^{237} cross-section ratio of 1.8 ($\sigma_{Pu^{238}} = 460$, $\sigma_{Np^{237}} = 256.7$, and $\sigma_{Np^{238}} = 1600$).

All dissolver solution data is presented within this document; however, about 15% of the wafer sample results are not included because of inconsistencies in the results which were not checked by re-analysis. Such action was justified by detected errors in analytical procedures and errors in 10% of the measurements

4. DUN-2911, "PUPA - a Code for the Calculation of Pu-238 Production," K. J. Matty, 8-27-67 (Unclassified)

subsequently checked by redundant measurements. Total conversion data based on average production parameters from each process channel is presented in Figure 1. Note that graphite diluted target element results are approximate. Since column average parameters are not yet available on these elements, a front-to-rear flux distribution was used in conjunction with one wafer sample to approximate column averages.

The possible sources of errors in the analytical determinations of data which affect the precision are those of volume measurement, sample evaporation, non-representative sampling, and inaccuracy in analytical procedure. Measurements were taken in various ways; the most accurate of all data is the isotopic analysis which employs mass spectroscopy techniques. Four or five significant figures are given in this analysis at a confidence level of 95%. Measurements taken of the cesium-137, Pu-238, and Np-237 solution concentrations were assigned $\pm 2\%$ precision with a 95% confidence level. The consistency of the data presented in this document and agreement with predicted relationships indicates, however, precision within the 1/2 to 1% range.

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Reactor Physics

TABLE I

PLUTONIUM-238 PRODUCTION DATA FROM COLUMN DISSOLUTION SAMPLES

<u>Column No.</u>	<u>Mwd/Adj. Column</u>	<u>Full Power Days</u>	<u>Np Charged g.</u>	<u>Np Converted</u>	<u>Pu-238 Produced</u>	<u>Pu-238/ Total Pu</u>	<u>Type and Number Elements</u>
2857	146.2	90.0	308.7	50.47	40.75	88.49	8 aluminum alloy
3157	172.8	104.5	342.0	62.31	49.01	87.12	9 aluminum alloy
2955	186.0	115.1	343.0	63.91	50.66	87.43	9 aluminum alloy
2759	225.4	139.0	454.2	105.92	79.13	84.95	12 impregnated in graphite
2960	250.0	154.5	481.3	122.73	89.14	84.37	12 impregnated in graphite
2857*	104.5	64.5	316.4	~31.3	~27.5	~92.64	8 impregnated in graphite
3157*	56.7	35.0	364.0	~28.09	~26.2	~93.44	8 compacted with graphite

*Estimated from wafer samples

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FIGURE 2

PERCENT OF ORIGINAL NP-237 CONSUMED VS. FULL POWER DAYS

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FIGURE 3

PU-238 PRODUCED (α) VS. NP-237 CONSUMED ($1-\beta$)

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FIGURE 4

IN-REACTOR CONVERSION EFFICIENCY VS. NEPTUNIUM CONSUMED

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FIGURE 5

PU-238 ISOTOPIC ABUNDANCE VS. NEPTUNIUM CONSUMED

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TABLE II

NEPTUNIUM LOADING CONFIGURATION

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<u>Column No.</u>	<u>2857</u>	<u>3157</u>	<u>2955</u>	<u>2759</u>	<u>2960</u>
	4-2	3-14	3-23	3-10	1-1
	4-7*	3-4*	4-9	3-2*	1-2
	3-22	3-25	4-6	3-3	1-3*
<u>Piece No.</u>	3-1*	4-8	4-5	3-5*	1-4
	1-10*	3-24*	4-4*	3-28	2-2*
	2-1	4-3	3-21	3-26*	2-3
	3-11	3-13	3-19	3-18	2-4*
	4-1	3-20	3-16*	3-17*	2-5
		3-12	3-15	3-6	1-6
				3-7	1-7
				3-8	1-8
				3-9	1-9*

Upstream

Downstream

*Denotes those target elements which were sampled

TABLE IIIDATA DERIVED FROM DISSOLVER SOLUTION ANALYSIS

<u>Analytical</u>	<u>From Process Tube</u>	<u>Cs¹³⁷ Atom/l x 10¹⁹</u>	<u>Pu²³⁸ g/l</u>	<u>Np²³⁷ g/l</u>	<u>Plutonium Isotopic</u>				
					<u>238</u>	<u>239</u>	<u>240</u>	<u>241</u>	<u>242</u>
5565	2857	.439	.250	1.58	88.47	9.32	1.83	.335	.0388
5566	2857	.449	.247	1.55	88.53	9.28	1.82	.328	.0374
5567	2857	.448	.250	1.61	88.51	9.30	1.82	.331	.0391
5568	3157	.625	.317	1.80	87.11	10.17	2.22	.440	.056
5569	3157	.631	.278	1.64	87.12	10.17	2.21	.437	.055
5570	3157	.619	.317	1.76	87.13	10.17	2.21	.436	.055
5573	2955	.639	.326	1.79	87.43	9.93	2.15	.427	.054
5574	2955	.639	.329	1.81	87.47	9.90	2.14	.430	.054
5575	2955	.638	.325	1.80	87.44	9.94	2.14	.430	.056
5718	2759	1.52	.562	2.47	84.95	11.02	3.18	.729	.118
5719	2759	1.52	.564	2.50	84.92	11.05	3.19	.726	.117
5720	2759	1.52	.569	2.49	84.94	11.02	3.19	.730	.118
5756	2960	1.53	.539	2.13	84.37	11.29	3.39	.808	.139
5757	2960	1.52	.540	2.19	84.41	11.26	3.39	.803	.138
5758	2960	1.53	.534	2.18	84.40	11.26	3.40	.802	.137
ARHCO	2960		.530	2.20					

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TABLE IVDATA DERIVED FROM WAFER SAMPLES

<u>Piece No.</u>	<u>From Process Tube</u>	<u>Analytical</u>	<u>Cs¹³⁷ Atoms/l x 10¹⁹</u>	<u>Pu²³⁸ g/l</u>	<u>Np²³⁷ g/l</u>	<u>Plutonium Isotopic</u>				
						<u>238</u>	<u>239</u>	<u>240</u>	<u>241</u>	<u>242</u>
1-3	2960	5275	1.12	.403	1.70	84.09	11.60	3.39	.790	.132
1-9	2960	5306	2.25	.989	4.48	86.34	10.02	2.89	.650	.100
3-1	2857	5086	1.62	.810	5.19	88.22	9.45	1.93	.363	.042
3-4	3157	5089	1.46	.714	4.50	87.68	9.84	2.03	.408	.052
3-5	2759	5272	2.68	.958	4.30	84.50	11.35	3.26	.759	.125
3-16	2955	5088	1.58	.763	4.23	86.07	10.17	2.25	.459	.057
3-17	2759	5273	2.86	1.018	4.28	84.67	11.07	3.34	.790	.140
3-24	3157	5090	1.55	.703	3.87	86.92	10.18	2.35	.490	.063
3-26	2759	5274	2.65	.890	3.65	84.17	11.37	3.50	.820	.140
4-4	2955	5087	1.82	.807	4.52	86.73	10.28	2.41	.505	.070
4-7	2857	5008	1.22	.683	4.42	88.57	9.3	1.78	.322	.033
#7 IMP	2857	5314	.186	.286	2.92	92.64	6.51	.755	.088	.007
#7 COMP	3157	5315	.116	.184	2.45	93.44	5.73	.700	.120	.022

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TABLE V

FLUX WIRE ACTIVATION AND COMPUTATIONAL RESULTS

<u>Target Element No.</u>	<u>Activation</u>	<u>Measurement Time Since Discharge</u>	<u>Irradiation Time</u>	<u>Flux</u>
FWID 3-18	1.92×10^7	86	140	1.07×10^{14}
FWCID 3-17	1.30×10^7	86	140	7.15×10^{13}
FWOD 3-26	2.05×10^7	86	140	1.14×10^{14}
FWCOD 3-28	1.34×10^7	86	140	7.38×10^{13}
3-S	1.80×10^7	0	115	1.17×10^{14}
2-U	1.99×10^7	0	115	1.3×10^{14}
1-S	1.88×10^7	0	115	1.23×10^{14}

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TABLE VI
PRODUCTION PARAMETERS FROM WAFER SAMPLES

<u>Sample No.</u>	<u>From Process Tube</u>	<u>α</u>	<u>1-8</u>	<u>ϵ</u>	<u>Q</u>
1-3	2960	.179	.243	73.81	84.09
1-9	2960	.172	.222	77.27	86.34
3-1	2857	.131	.164	79.80	88.22
3-4	3157	.132	.167	79.25	87.68
3-5	2759	.171	.231	74.03	84.50
3-16	2955	.146	.188	77.78	86.07
3-17	2759	.180	.243	74.14	84.67
3-24	3157	.147	.189	77.94	86.92
3-26	2759	.183	.250	73.20	84.17
4-4	2955	.145	.187	77.60	86.73
4-7	2857	.130	.159	80.92	88.57
#7 IMP	2857	.088	.099	.894	92.64
#7 COMP	3157	.069	.077	.902	93.44

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TABLE VII

PRODUCTION PARAMETERS FROM DISSOLVER SOLUTIONS

<u>Sample No.</u>	<u>From Process Tube</u>	<u>Residence Time</u>	<u>α</u>	<u>1-β</u>	<u>ϵ</u>	<u>Q</u>
5565			.1323	.1636	80.91	88.47
5566	2857	90.0	.1331	.1648	80.72	88.53
5567			.1303	.1610	80.91	88.51
5568			.1440	.1822	79.05	87.11
5569	3157	104.5	.1392	.1788	77.87	87.12
5570			.1467	.1856	79.05	87.13
5573			.1481	.1867	79.32	87.43
5574	2955	115.1	.1479	.1862	79.47	87.47
5575			.1471	.1855	79.27	87.44
ARHCO						
5718			.1744	.2336	74.63	84.95
5719	2759	139.0	.1733	.2320	74.70	84.92
5720			.1750	.2341	74.77	84.94
5756			.1884	.2555	73.73	84.37
5757	2960	154.5	.1849	.2503	73.87	84.41
5758			.1838	.2496	73.66	84.40
ARHCO						

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