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CALCULATIONS ON CLOSE-COUPLED PROCESSING FOR Pu-238 RECOVERY

By

E. A. Coppinger

PROCESS ENGINEERING DEVELOPMENT OPERATION
CHEMICAL DEVELOPMENT
CHEMICAL LABORATORY
HANFORD LABORATORIES

July 10, 1963

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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CALCULATIONS ON CLOSE-COUPLED PROCESSING FOR Pu-238 RECOVERY

Irradiation of Np-237 in Hanford reactors and recovery of the Pu-238 product in a close-coupled separations plant is currently of interest. Such a concept has the potential of increasing Pu-238 production rates. The results of initial calculations on the subject are presented herein to aid further study and evaluation. Much of the information is presented in terms of the aqueous target system proposed in earlier work⁽¹⁾ (i.e., irradiation and processing of an aqueous neptunium solution). However, most of the information can be converted for evaluation of a solid target system.

Irradiation Yields

Nuclide build-up during irradiation of neptunium-237 in a Hanford reactor is shown in Figure 1. Of major significance is the rapid build-up of short-lived highly radioactive neptunium-238. The build-up curves also show the advantage of short irradiation times in regard to product purity and plutonium-238 yields. As irradiation time increases, plutonium-238 both fissions and captures neutrons to form fission products and higher plutonium isotopes thereby reducing product purity and decreasing the yield per gram of Np-237 consumed. However, a shorter irradiation time means more frequent chemical processing and any savings on neptunium consumption in the reactor could be reduced or eliminated by more frequent neptunium process losses. It should also be noted that the Np-238 content should be added to the Pu-238 content given by the Pu-238 build-up curve to get total Pu-238 production after adequate cooling.

⁽¹⁾ A. M. Platt, "Palmolive Alternate", HW-57720 RD, October 22, 1958

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Power During Irradiation

During irradiation of the Np-237, the power steadily increases due to fission of Np-238, Pu-238, and Pu-239. Lesser amounts of power are also produced from the beta decay of Np-238 and the alpha decay of Pu-238. Assuming an initial target solution concentration of 1 M Np-237, the increase in power during irradiation is shown in Table I. Since the power is directly proportional to the neptunium content, the values in Table I can be readily converted to other neptunium concentrations or to a solid system.

TABLE I
NEPTUNIUM SOLUTION POWER IN REACTOR

1 M Np-237 Solution Irradiated at 2.8×10^{13} neutrons/sec/cm²

<u>Irradiation Time, Days</u>	<u>Power, Watts/Liter</u>			<u>Total</u>
	<u>Fission</u>	<u>Np-238 Decay</u>	<u>Pu-238 Decay</u>	
1	450	139	-	589
5	1290	398	0.2	1688
10	1570	472	0.5	2042
20	1720	485	1.3	2206
50	2320	478	3.4	2801
100	4080	464	6.7	4551
140	6000	453	9.1	6463

The values given in Table I are based on the nuclide build-up in Figure 1 and assumed fission cross sections of 1600, 18, and 1370 barns for Np-238, Pu-238, and Pu-239, respectively.

Power After Discharge

After discharge of the neptunium solution from the reactor, there is still considerable power due to decay of fission products, Np-238, and Pu-238. Table II shows the total decay power in an irradiated solution that was charged to the reactor at a 1 M neptunium concentration. The values in Table II include

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the gamma power associated with the decay. Total radioactive decay power in the neptunium solution and the first cycle Purex feed stream (HAF) are also compared in Table II. This comparison shows the neptunium solution, one hour after reactor discharge, is about 600 times "hotter" than HAF. When irradiated for 10 and 100 days and cooled for 10 days, the neptunium solution is, respectively, about 22 and 36 times "hotter" than HAF. This suggests that a short cooling time on the order of 10 days will be required for aqueous close-coupled reprocessing.

Since a portion of the gamma radiation escapes from the solution, it is, perhaps, more meaningful to compare the alpha and beta decay power of the neptunium solution with the beta decay power of HAF. This comparison is shown in Table III.

Fission product power for Tables II and III was approximated by using the fission power from Table I and the Untermeyer-Weill's empirical constants⁽²⁾ which give a ratio of fission product power to reactor power for U-235 fission. The fission product power does not take into account any fission products in the recycle neptunium stream.

Neptunium-238 beta and gamma power is based on the decay scheme³ summarized in Table IV.

(2) H. Etherington, Nuclear Engineering Handbook, 1st Ed., New York: McGraw-Hill, 1958, p. 1-39.

(3) R. G. Albridge and J. M. Hollander, "The Decay of Np-238", Nuclear Physics, Vol. 21, No. 3, December, 1960, p. 438.

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TABLE II

TOTAL DECAY POWER IN NEPTUNIUM SOLUTION

Basis: 1 M Np Solution Irradiated at 2.8×10^{13} neutrons/sec/cm²

Cooling Time	Total Decay Power, Watts/Liter									
	10 Day Irradiation					100 Day Irradiation				
	F.P.'s	Np-238 Decay	Pu-238 Decay	Total	Factor >(1) HAF	F.P.'s	Np-238 Decay	Pu-238 Decay	Total	Factor >(1) HAF
1 Hr.	16.5	466	0.55	483	568	52	456	6.75	515	606
12 Hr.	6	400	0.58	406	478	22	392	6.78	421	496
1 Day	4	339	0.61	343	404	18	332	6.81	357	420
5 Day	1.4	90.7	0.72	92.8	109	9.4	89	6.93	105	124
10 Day	0.8	17.4	0.76	18.9	22	6.8	17.1	6.95	30.8	36
20 Day	0.45	0.65	0.77	1.87	2.2	4.7	0.63	6.97	12.3	14

(1) Assumes total beta plus gamma decay power of 0.85 watts/liter of Purex HAF.

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TABLE III

ALPHA AND BETA POWER IN NEPTUNIUM SOLUTION

Basis: 1 M Np Solution Irradiated at 2.8×10^{13} neutrons/sec/cm²

Cooling Time	Alpha and Beta Power, Watts/Liter									
	10 Day Irradiation					100 Day Irradiation				
	F.P.'s ⁽¹⁾	Np-238 Decay	Pu-238 Decay	Total	Factor > ⁽²⁾ HAF	F.P.'s ⁽¹⁾	Np-238 Decay	Pu-238 Decay	Total	Factor > ⁽²⁾ HAF
1 Hr.	8.2	148	0.55	156.7	373	26	145	6.75	177.7	423
12 Hr.	3	127	0.58	130.6	311	11	125	6.78	142.8	340
1 Day	2	108	0.61	110.6	263	9	106	6.81	121.8	290
5 Day	0.7	29	0.72	30.4	72	4.7	28	6.93	39.6	94
10 Day	0.4	5.5	0.76	6.7	16	3.4	5.4	6.95	15.7	37
20 Day	0.22	0.21	0.77	1.2	2.8	2.3	0.2	6.97	9.5	22

(1) Assumes beta power of fission products is 50 percent of total.

(2) Assumes beta power of 0.42 watts/liter of Purex HAF.

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TABLE IV
Np-238 DECAY

<u>Beta</u>		<u>Gamma</u>	
<u>Mev</u>	<u>Abundance, %</u>	<u>Mev</u>	<u>Abundance, %</u>
0.219	8.4	0.044	77.4
0.26	38	0.102	4.3
1.14	3	0.885	1.2
1.246	45	0.925	3.1
?	5.6	0.986	20
		1.027	5.3
		1.03	17

Using this decay scheme, the specific power has been calculated to be 1144 watts per gram. Of this, there are 364 beta watts per gram and 780 gamma watts per gram. The large amount of gamma energy indicates that, with short cooling, recycle neptunium solution requires heavy shielding regardless of the fission product decontamination factor. In the solid system, short cooling results in remote fabrication and remote charging of the recycle neptunium. (However, some degree of shielding may be required for recycle neptunium even after long cooling periods because of the regrowth of neptunium daughters.)

The decay power from Pu-238 is essentially from alpha particles and the specific power has been computed to be 0.54 watts per gram.

Iodine Production

Instantaneous production rates for 8.05-day I-131, 21-hr I-133, and 6.7-hr I-135 have been approximated for specific days during the irradiation period. Results are shown in Table V. These rates are based on assumed effective fission yields of 3.7, 6.5, and 6.6 percent for I-131, I-133, and I-135, respectively. No allowance is included for decay during the equipment holdup time. Additional activity from other shorter-lived iodine isotopes will also be present.

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TABLE V

IODINE PRODUCTION RATES

Specific Day	Production Rate, Curies/Day/Kg Np-237		
	<u>I-131</u>	<u>I-133</u>	<u>I-135</u>
10	17	283	905
50	26	425	1360
100	46	742	2480

For means of comparison, one ton of uranium, irradiated to 600 MWD/T at a specific power of 10 MW/T, contains 8.4 curies of I-131 after 120 days of cooling. The Purex Plant would handle 218 curies per day at an instantaneous processing rate of 26 tons of uranium per day. If 90 percent of an assumed 100 kilogram inventory of Np-237 is in the reactor as an aqueous solution and all of the I-131 is evolved, the evolution rate is 1530, 2340, and 4140 curies per day for irradiation day 10, 50, and 100, respectively. Evolution rates for the other iodine isotopes are relative to production rates in Table V.

Radiolytic Gas Production

The water and nitric acid in an aqueous solution of neptunium nitrate and nitric acid will be decomposed by the high-energy radiation present during irradiation. Water is decomposed to hydrogen, hydrogen peroxide, and oxygen. Nitric acid will probably be decomposed to hydrogen and oxides of nitrogen. In an operating homogeneous reactor, over 96 percent of the hydrogen gas produced in solution is due to the fission recoil particles⁽⁴⁾. Reference (4) also lists $G_{(H_2)}$ values of about unity for reactor-irradiated 1M uranyl nitrate solution. Possible gas evolution rates for a 1M neptunium solution are shown in Table VI. These rates were calculated assuming a $G_{(H_2)}$ value of unity and absorption of 90 percent of the fission power shown in Table I.

(4) J. A. Lane, et. al., Fluid Fuel Reactors, 1st Ed., Addison-Wesley

TABLE VI

GAS PRODUCTION DURING IRRADIATION

<u>Irradiation Time, Days</u>	<u>Gas Rate*, Liters at STP/Min/Liter of Solution</u>
1	0.1
10	0.4
100	1.0

*Hydrogen and equivalent amount of oxygen plus hydrogen peroxide.

The Homogeneous Reactor Test at ORNL produced 10 cfm of gas at STP when operating at full power of 1000 KW_(t) without recombination catalysts⁽⁵⁾. This gas rate can be nearly reproduced with the assumptions used to calculate the values of Table VI. Copper salts are employed as catalysts for recombination of hydrogen and oxygen in homogeneous reactors. They may be applicable here.

Aqueous System Production Rates

Plutonium-238 production rates depend on the interaction of many factors. Some of major importance are available inventory of Np-237, percent of available inventory in the reactor, irradiation yields, cycle time, and virgin Np-237 recovery rate. Production rates and operating variables for several schemes employing an aqueous neptunium solution are shown in Table VII to illustrate the effect of various factors. In all schemes, the initial solution concentration is assumed to be 1 M Np-237 (about 0.75 atom percent Np-237) and 1 M nitric acid. The Np-237 inventory is assumed to be 100 kilograms. At a 1 M concentration, a 100 kilogram inventory represents a solution volume of 422 liters or 111.5 gallons.

(5) C. F. Bonilla, Nuclear Engineering, New York, McGraw-Hill, 1957, p. 750.

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TABLE VIIAQUEOUS SYSTEM PRODUCTION RATES AND VARIABLES

	<u>Scheme 1</u>	<u>Scheme 2</u>	<u>Scheme 3</u>
Reactor Residence Time, days	99	11	99
Irradiation Time, days	79	9	63
Time Outside Reactor, days	11	11	11
Cycle Time, days	110	22	110
Solution Volume Processed, gal/yr	370	1850	370
Average Process Rate, gal/day	1.0	5.1	1.0
Instantaneous Process Rate, gal/ day	1.2 to 11.2	6.3 to 55.7	1.2 to 11.2
Solution Volume In Reactor, gal.	100.3	55.7	100.3
Solution Volume Outside Reactor, gal.	11.2	55.7	11.2
Np-237 Consumption, kg/yr.	16.7	12.3	13.7
Production Rate, kg Pu-238/yr.	15.17	8.96	11.65
Production Rate, watts Pu 238/yr.	8190	4840	6290
Product Purity, w/o Pu-238	96.1	>99.5	96.7

In Scheme 1, 90 percent of the available inventory is kept in the reactor. Also, Scheme 1 assumes that the solution is either irradiated in place without recirculation outside the reactor or the amount of time the neptunium spends outside the reactor in recirculation piping is negligible. An operating efficiency of 80 percent is assumed for the reactor and processing plant. The time outside the reactor consists of 10 days for cooling and one day for processing and reactor solution make-up. The instantaneous processing rate could possibly range from that necessary for continuous operation to that resulting from intermittent processing of the entire out-of-reactor volume in one day after cooling. Neptunium-237 consumption is based on the amount consumed in the reactor plus a 0.2 percent process loss per cycle.

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Scheme 2 is similar to Scheme 1 except that only 50 percent of the inventory is kept in the reactor. The reduced production rate shows the incentive to keep a large percentage of the inventory in the reactor.

Scheme 3 is similar to Scheme 1 in that 90 percent of the inventory is kept in the reactor system. However, in Scheme 3, it is assumed that the volume of the out-of-reactor portion of the recirculation system results in the neptunium being in the reactor only 80 percent of the operating days. The reduced production rate shows the incentive for minimizing the holdup in the out-of-reactor portion of the recirculation system.

The large consumption of Np-237, even with a rather minimum process loss, indicates the importance of the virgin Np-237 production rate in ultimately determining the Pu-238 production capabilities. Also, the large Np-237 consumption relative to Pu-238 production in Scheme 2 shows the effect of process losses resulting from short irradiation times and frequent processing.

Solid System

Solid element systems have not been examined in detail. In general, such systems require a longer time cycle outside the reactor to refabricate fuel elements. Neptunium process losses would be higher due to the additional process steps and a greater Np-237 production rate would be required to sustain or build-up the inventory. However, a solid system is not troubled with the possibility of having a portion of the inventory outside the reactor in a recirculation system.

One possible solid system that has been proposed consists of cans of loosely packed neptunium oxide. Commercially available alumina could be added to get any desired Np-237 dilution. For example, a fuel mix containing four atom percent Np-237 would consist of 37.5 weight percent neptunium

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oxide in alumina (40 to 50 mesh). Such a mix would represent a volume of roughly 136 liters or 36 gallons for a 100 kilogram inventory of Np-237.

Available data⁽⁶⁾ on the effective thermal conductivity of loosely packed alumina (18 to 120 mesh) indicate effective thermal conductivities of about 0.1 and 0.2 Btu/(hr)/(sq. ft.)/(°F/ft) at mean temperatures of 200 and 1000 °F, respectively. Assuming a mean thermal conductivity of 0.15 Btu/(hr)/(sq. ft.)/(°F/ft), Table VIII shows the approximate radial temperature drop across the oxide described in the previous paragraph.

TABLE VIII
TEMPERATURE DROP ACROSS OXIDE MIX

<u>Irradiation Time, Days</u>	<u>Radial Temperature Drop, °F</u>	
	<u>1 in. I.D.</u>	<u>1.4 in. I.D.</u>
1	510	1000
10	1780	3500
100	3960	7780

Temperature drop could be reduced by further dilution of the neptunium oxide with alumina, by filling the void volume with helium, or by reducing the can diameter further.

(6) W. V. DeMier, "Interim Report - Thermal Conductivity Studies on Various Calcined Cold Wastes", HW-62614, March 11, 1960.

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