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ADVANTAGES OF PALMOLIVE ALTERNATE

by

E. A. Coppinger and E. T. Merrill

Programming Operation
HANFORD LABORATORIES OPERATION

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND. WASHINGTON

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INTRODUCTION

It has been proposed* that Pu-238 be produced by irradiating ^{neptunium}~~plutonium~~ solution in one or more loops in a reactor and then recovering the ~~Pu-238~~ in a close-coupled separations plant. Such a scheme could replace the more conventional scheme of solid element fabrication, irradiation, and reprocessing for plutonium and neptunium recovery. This document presents the advantages of such a scheme from the standpoint of product purity and Pu-238 production.

SUMMARY

Four Pu-238 production schemes in a Hanford reactor are compared here. Scheme 1 and 2, which keep 50 and 75% of the available neptunium inventory in the reactor, respectively, represent the probable range of solid element systems. Scheme 3 represents the proposed liquid system* which allows a short Np-238 cooling time and keeps 50 per cent of the available inventory in the reactor. Scheme 4 also represents a liquid system but differs in that the total available inventory is kept in the reactor during operating periods. Assuming neptunium solution irradiation begins in 1962 with a 24 kg inventory and 0.75 kg of neptunium is added to the inventory each month, Pu-238 production rates and cumulative production after January 1, 1962, are compared below for the four schemes.

Date	Production Rate, g Pu-238/mo.				Cumulative Production, Kg Pu-238			
	Scheme				Scheme			
	1	2	3	4	1	2	3	4
Jan. 1, 1962	149	192	161	317	-	-	-	-
Jan. 1, 1963	185	237	203	370	2	2.8	2.2	4.1
Jan. 1, 1964	219	276	241	414	4.4	5.9	4.8	8.8
Jan. 1, 1965	248	311	275	451	7.2	9.4	7.9	14.0
Jan. 1, 1967	298	369	334	506	13.8	17.6	15.2	25.5
Jan. 1, 1969	337	414	382	544	21.5	27.0	23.8	38.1

The liquid systems offer the advantage of increased product purity by decreasing Pu-238 dilution with higher plutonium isotopes. The product from the four schemes has the following Pu-238 content:

Scheme	Pu-238 Content Wt. %
1	95.2
2	86.6
3	99.2
4	98.7

*HW-57720-RD, Palmolive Alternate, A. M. Platt, October 22, 1958.

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DISCUSSION

Production Schemes

Four schemes for the production of Pu-238 in a Hanford reactor are compared in this document. Major variables used for each scheme are shown below:

	Scheme			
	1	2	3	4
Reactor Residence Time, days	120	365	20	24-29
Irradiation Time, days	96	292	16	24
Time Outside Reactor, days	120	120	20	1-6
Cycle, days	240	485	40	30
Production Rate, g Pu-238/Mo/Kg Inventory	6.2	8.0	6.7	13.2
Burn-up Rate, g Np-237/Mo/Kg Inventory	6.9	9.3	6.9	13.8
Processing Loss, g Np-237/cycle/Kg Inventory	20	20	2	2
Disappearance Rate, g Np-237/Mo/Kg Inventory	9.4	10.5	8.4	15.8
Pu-238 Recovery	100%	100%	100%	100%

Scheme 1 and 2 represent the probable range of production methods using solid neptunium fuels which involve irradiation, cooling, processing for Np-237 and Pu-238 recovery, and refabrication of solid fuel elements. For the chosen cycle time 50% and 75% of the neptunium inventory is in the reactor for scheme 1 and 2, respectively. Actual conditions will depend on the Pu-238 demand and desired product purity.

Scheme 3 represents Pu-238 production by the proposed scheme of irradiating neptunium solution in reactor tubes and recovering the Pu-238 in a close-coupled separations plant. In scheme 3 only 50% of the inventory is in the reactor. Solution is continuously or semi-continuously removed from the reactor, cooled (for Np-238 decay), processed in the close-coupled separations plant, and returned to the reactor.

In scheme 4 the entire inventory, as neptunium solution, is in the reactor for the entire operating period. During a maintenance or charge-discharge shut-down, the solution is removed and processed through the close-coupled separations plant where Pu-238 is recovered and fission and degradation products are removed. After processing, the solution is returned to the reactor.

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Production and burn-up rates were obtained from Figure 1 which shows irradiation yields in a Hanford reactor for a standard Westcott flux of 2.6×10^{13} neutrons per sec. per sq. cm.

Neptunium processing losses are higher for schemes 1 and 2 because of the larger number of processing steps. In schemes 1 and 2 the irradiated neptunium (as a solid element) is subjected to dissolution, two or more cycles of separations processing, calcination, and element fabrication before its return to the reactor. In schemes 3 and 4 the irradiated neptunium solution is subjected to one cycle of ion exchange or solvent extraction before its return to the reactor.

Product Purity

Schemes 3 and 4 produce a purer Pu-238 product than schemes 1 or 2. The shorter irradiation times permitted by the liquid systems decrease the dilution of Pu-238 product with Pu-239, Pu-240, and Pu-241. The build-up of Pu-238 and higher plutonium isotopes are shown in Figure 1. Product compositions for the four schemes are shown below:

<u>Isotope</u>	<u>Weight Per cent</u>			
	<u>Scheme 1</u>	<u>Scheme 2</u>	<u>Scheme 3</u>	<u>Scheme 4</u>
Pu-238	95.2	86.6	99.2	98.7
Pu-239	4.6	12.2	0.8	1.3
Pu-240 - 241	0.2	1.2	<0.005	<0.01

Inventory Build-up

For a given production scheme plutonium-238 production at any given time depends on the neptunium inventory which has built up. Inventory build-up depends on the rate of neptunium addition to the inventory, neptunium burn-up rate, and neptunium processing losses. In order to compare production capabilities of the four schemes, it is estimated that neptunium solution irradiation would begin January 1, 1962. The rate of neptunium addition to the inventory is assumed to be 0.75 kg/mo or roughly the recovery rate at the Purex plant. The inventory, as of January 1, 1962, is assumed to be 24 kilograms or roughly the build-up if scheme 1 or 2 is begun in early 1959, at the above addition rate to the inventory. The inventory build-up from January 1, 1962, has been calculated and is shown in Figure 2 for the four schemes for the period 1962 to 1969.

Production Rate

Plutonium-238 production rates (g/mo) at any specific time, have been calculated from the inventory build-up curves and production rates per kilogram of inventory given earlier. They are shown in Figure 3 for the four schemes for the period 1962 to 1969.

The curves in Figure 3 vividly show the increase in production rates that is immediately available from scheme 4 when irradiation of neptunium solution begins. Scheme 4 provides about a 60 to 100% increase in instantaneous production rate over scheme 1 and about a 30 to 60% increase over scheme 2.

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Scheme 3 provides production rates between those of schemes 1 and 2. Thus, any method developed for neptunium solution irradiation and close-coupled separations processing should keep most or all of the inventory in the reactor during operating periods.

Figure 3 also illustrates another feature of the liquid systems. Should a certain production rate be desired, a liquid system may provide such rates sooner than solid element systems. For example, the production rate of scheme 1 (solid elements) is 337 g Pu-238/mo. in 1969. Scheme 4 reaches this rate in about May of 1962 - a difference of almost 7 years.

Cumulative Production

Integration of the production rate curves in Figure 3 gives the total amount of Pu-238 produced at any time since January 1, 1962. Such curves of cumulative production are shown in Figure 4 for the four schemes for the period 1962 to 1969.

The curves show how cumulative production increases after neptunium solution irradiation is begun in 1962. They also show that after startup in 1962, any cumulative Pu-238 production goal can be met quicker with scheme 4. For example, Figure 4 shows that 21.5 kg of Pu-238 will be produced by scheme 1 during the period 1962 to 1969. With the total neptunium inventory in the reactor as solution, 21.5 kg would be produced by May, 1966. This further illustrates the desirability of developing a solution irradiation scheme which keeps most or all of the inventory in the reactor during operating periods.

Possible advantages of some liquid systems indicate the desirability of studying the feasibility of a general purpose liquid loop in a reactor for research and development purposes.

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Programming

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Np-237

FIGURE 1

IRRADIATION YIELDS

GRAMS / KG Np-237 CHARGED

100

10

1.0

0

DAYS

50 100 150 200 250 300 350

BD-7600-027 (3-58) ARC-DE RICHARD, WASH.

IRRADIATED AT STANDARD WESTCOTT $\phi = 2.6 \times 10^{13}$

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Pu-238

Pu-239

Pu-240 + 241

FIGURE 2

INVENTORY BUILDUP

Scheme 1 - Solid elements - 50% inventory in Pile

Scheme 2 - Solid elements - 75% inventory in Pile

Scheme 3 - Liquid System - 50% inventory in Pile

Scheme 4 - Liquid System - 100% inventory in Pile

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FIGURE 3

PRODUCTION RATE

- Scheme 1 - Solid elements - 50% inventory in reactor
- Scheme 2 - Solid elements - 75% inventory in reactor
- Scheme 3 - Liquid System - 50% inventory in reactor
- Scheme 4 - Liquid System - 100% inventory in reactor

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FIGURE 4

CUMULATIVE PRODUCTION

Scheme 1 - Solid elements - 50% inventory in reactor

Scheme 2 - Solid elements - 75% inventory in reactor

Scheme 3 - Liquid System - 50% inventory in reactor

Scheme 4 - Liquid System - 100% inventory in reactor

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