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LOSSES ASSOCIATED WITH THE INTERIM
PURIFICATION PROCESSING OF NEPTUNIUM☐

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May 19, 1959

V. R. Cooper, Manager
Research and Engineering
Chemical Processing Department

LOSSES ASSOCIATED WITH THE INTERIM PURIFICATION
PROCESSING OF NEPTUNIUM

1.0 INTRODUCTION

The interim program for the production of neptunium oxide at HAPO applies the following processing steps: isolation of neptunium from the Purex process streams, using Purex flow sheets specially adapted for this purpose; purification of the neptunium nitrate by an ion exchange process carried out in one of the Redox laboratory (222-S) multi-curie cells; and precipitation of neptunium oxalate and conversion of the oxalate to oxide in laboratory-type equipment. The process, being still in the developmental stages, is as yet subject to extreme fluctuations, both in conditions and results. For example, the feed processed in the April purification runs contained large quantities of other salts, a factor which affected resin column performance. Since one of the deleterious effects was a marked increase in waste loss, this history of the interim program, with a discussion of the factors which affect waste losses, has been prepared.

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2.0 SUMMARY

Neptunium purification experience, with the experimental equipment and processes being used for the interim program, has shown that with good quality feed the ion exchange waste losses can be held to less than two per cent. In the April runs, however, the feed contained enough thorium (material from UCN) or uranium (material from Purex) that the normal operating procedures permitted the loss of up to 21 per cent of the feed neptunium to the resin column waste effluents. Determination of the extent of the loss to waste was delayed by the presence of appreciable amounts of plutonium and the dependence upon total alpha counting for verification that the system was behaving normally. In consequence of a) the lack of a quick, specific neptunium determination procedure, b) the lack of adequate waste storage capacity, and c) the desire to meet a tight shipping schedule, the waste from the first two of the three runs, containing 69 grams of neptunium, was discarded to the Redox waste tanks. The waste from the third run of the series, containing 93 grams, was held and re-worked, with a four gram loss to waste. A B-PID of -35 grams resulted for this series of runs.

The general conclusions are drawn that with the experimental-type facility now in use, tight control over waste losses is not compatible with a tight processing schedule. Plans to increase processing and waste storage capacity and to arrange for rapid, specific neptunium determinations on selected waste samples will alleviate the problem. However, it should be recognized that for the rest of the interim program, it will be necessary to lengthen (perhaps double) the total purification cycle, if good control of waste losses is to be maintained.

3.0 GENERAL

3.1 Interim Program Requirements

The interim purification and oxide-production program was established to meet two needs: to take care of production requirements until a permanent processing facility could be planned and built; and to provide means for the development and verification of processing flowsheets. Selection of the location and personnel for the program was based upon consideration of the above needs and the following problems: a) the equipment, although simple, was needed quickly and had to be installed in available space; b) the purification process requires shielded and properly enclosed storage, feed, make-up, and waste tanks and processing equipment; c) the processing operations are necessarily carried out on an intermittent, campaign-type schedule, because of the intermittent (bi-monthly) availability of the feed

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and the pressure to turn out product as quickly as possible; and d) rather wide variations in feed composition and quality are to be expected, giving even more emphasis to the developmental nature of the operations.

3.2 Process, Equipment, and Operations

The first purification run, in the fall of 1958, was made by Chemical Research personnel, in temporary equipment in a 222-S multi-curie cell which had been used previously for research work on solvent extraction processes. When this proved successful and the program was put on a more formal basis, administration was turned over to CPD's Process Chemistry Operation. The ion exchange facility, consisting of a minimum of new equipment in addition to the tanks, pumps, valves, lines, etc. which had previously been part of the solvent extraction unit, was revamped to increase its capacity and make it easier to operate. These changes, of course, while making the installation better adapted to its use on an interim basis, did not convert it to a true production facility.

Fundamentally, the purification facility is comprised of a glass ion exchange column and assorted feed, product, and waste handling equipment. In addition, a second ion exchange column was included to permit recovery of plutonium from any high-plutonium waste streams. Typical operating schemes consist of the following sequence of operations: a) feed make-up; b) passage of the feed through the anion exchange column, for adsorption of the neptunium; c) passage of a reducing solution wash through the column, for removal of any adsorbed plutonium or uranium; d) use of a wash containing fluoride for removal of adsorbed fission products, thorium, etc.; and e) elution of the neptunium from the column with a low-acid solution.

Over-all purification operations are planned and carried out under the direction of a Process Chemistry chemist. Each of the three operating crews used in the 24 hrs./day operation consists of a chemist and an engineering assistant. Operating instructions consist of general rules (regarding limits for discarding waste, etc.) and a specific run plan, developed for each run. Experience thus far has been that the run plan has served as a general guide, but that variations in feed content and behavior have been great enough to force marked departure from the original plans. Changes from the run plans are based upon physical observations of the ion exchange column behavior and upon apparent waste losses.

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3.3 Process Control Measures

For control of the process, reliance has been placed upon the following measures:

- a) Control of the feed make-up process, to assure adsorptability of neptunium in the column feed.
- b) Control of the batch size, to avoid over-loading the column. Experience has shown that with feed which is free of other adsorbing materials, the large column will hold 600-700 grams neptunium. Although the presence of large amounts of such materials as thorium and uranium has been found to decrease the column capacity, it has been assumed that a 400-gram batch of the usual feed can be handled safely.
- c) Analysis of waste stream samples. Because of the complexity of the specific neptunium analysis, waste stream control has depended on total alpha measurements and a knowledge of the process. Thus, although the total alpha count in the feed due to plutonium far exceeds that due to neptunium, the process can be controlled so that very little neptunium should appear in the waste streams (the loading cycle waste and first wash) containing the plutonium. During the wash cycle in which loss of neptunium is probable (the second wash), the total alpha count is a good indication of neptunium content.

The control system has suffered from the lack of sufficient waste storage space to continue operating while holding a waste batch until a specific neptunium determination can be made. It is for this reason that so much reliance has been placed upon total alpha determinations.

3.4 Waste Loss and Material Balance Experience

Product waste loss and material balance experience is summarized in Tables 1 and 2, respectively. As shown in Table 1, the neptunium discarded to waste ranged from negligible amounts to five per cent for runs 1 through 6. The processing operations in April, 1959, (Runs 7-10) resulted in much greater concentrations of neptunium in the ion exchange waste effluents. Because of the lack of adequate waste storage capacity, and because of the pressure on the program, relatively major amounts of neptunium (73 grams, or 3%) were discharged to the Redox waste tanks.

A material balance for runs 7-10 shows a B-PID of -35 grams, or four per cent underaccounted. This may represent errors in measurement of waste losses. The over-all B-PID of about four per cent (Table 2) is well within the limits of present measurement accuracy.

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TABLE 1

NEPTUNIUM IN ION EXCHANGE WASTE

Date	Run No.	Feed Source	Kg Np Rec'd	Gross Salts in Feed (Wt.)	Np in Waste			Disposal of Waste
					Process Step	Gm	%	
12/58	1	Purex	0.12		Total	5.0	5	Discarded
1/59	2	"	0.44	45 g Pu	Total	0.8	0.2	
"	3	"	0.33	16 g Pu	"	5.1	1.5	
	Total - Runs 2 & 3					5.9	0.8	Discarded
3/59	4	Rework	0.35	--	Total	N11	-	
"	5		0.39	--	"	N11	-	
"	6	Purex	0.22	86 g Pu 322 g U	"	0.6		Discarded
4/59	7	LCN	0.11	144 g Th	Loading Wash 2	4 14		
			Total			18	16	Discarded
4/59	8	Purex	0.37	8 g Pu 7 kg U	Loading Wash 1 Wash 2 & 3	25 13 13		20 g held for Purex recovery; eventually all discarded
				Total		51	14	
4/59	9	Purex	0.44	7 g Pu 7 kg U	Loading Wash 1 Wash 2 & 3	15 26 52		
				Total		93	21	Reworked in Run 10
4/59	10	Rework	0.10	11 g Pu 4.7 kg U	Total	4	4	Discarded
				Total - Runs 7 - 10		166	18	73 g (8%) dis- carded; 93 g reworked.

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TABLE 2
MATERIAL BALANCE^(a) - NEPTUNIUM PURIFICATION
AND OXIDE PREPARATION

Run Series	Date	Receipts (Kg)		Product Transfers (Kg)		Waste Discards			Run-End Inv. (Kg)		Cumulative B-PID	
		Run	Cumulative	Run	Cumulative	Run		Cum.	Book	Phys.	Kg	%
						Kg	%					
1	12/58	0.12	0.12	0.08	0.08	0.01	1	.01	0.03	0.04	+0.01	+8
2,3	1/59	0.77	0.89	0	0.08	0.01	1	.02	0.79	0.78	-0.02	-2
4-6	3/59	0.21 ^(b)	1.10	0.91	0.99	Nil		.02	0.09	--		
7-10	4/59	0.92	2.02	0.74	1.73	0.07	8	.09	0.20	0.11	-0.09	-4

(a) Receipt values are receiver measurements. Product transfer values are as measured as neptunium oxide, nitrate, etc. transferred out of the facility.

(b) A total of 0.96 kg was processed. This included 0.74 kg product from the previous run, which was reprocessed.

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4.0 CONDITIONS CONTRIBUTING TO HIGH WASTE LOSSES

The unusually high waste losses experienced in runs 7-10 may be ascribed to a combination of circumstances:

- a) Production and finished element shipping schedules have emphasized the need for speed in the purification operation. Incentive for keeping the over-all purification schedule short has come from the need to borrow personnel from other groups for the period of the run. It has also come from considerations of the increased radiation hazard in handling the oxide with increased length of the period since ion exchange treatment.
- b) The facility does not have adequate waste storage capacity. The result is that, in order to keep the processing going, it is often necessary to discard waste solutions on the basis of total alpha measurements, before a specific analysis for neptunium can be run. In prior runs, with relatively clean feeds, this procedure has been satisfactory.
- c) All the feed for these runs had large quantities of other salts which tended to reduce the amount of neptunium the resin could hold, and to spread out the neptunium band. In consequence, neptunium was lost to the waste effluents during all phases of the run (see Table 1). The loss was obscured at the time by the high plutonium alpha content of the solutions, and in consequence the decision was made that, although the losses appeared to be higher than usual, they would be accepted to avoid stopping the processing operation in order to re-work the waste.

5.0 CONCLUSIONS

Consideration of the factors described above has led to the following conclusions:

- a. The purification processing schedule which we have attempted to follow is not compatible with the routine achievement of good yields in an experimental system. Waste losses in runs 7-10 would have been lower had more time been taken.
- b. The facility does not have enough waste storage capacity.
- c. In spite of endeavors to plan ahead, run plans are usually invalidated by variation in the feed stream composition from batch to batch.

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- d. The intermittent nature of the operation, with the consequent requirements for borrowed personnel, unusual laboratory service in great quantity, etc., makes it difficult to arrive at a balance with other, competing functions which must also be carried on.

6.0 RECOMMENDATIONS

- a. Plans for installation of a larger column, to give added column capacity and consequently added protection against losses, should be expedited. Plans for increasing the waste storage capacity should also be expedited.
- b. Efforts should be made to adapt analytical methods now in use to provide a quick, specific neptunium determination. (Forthcoming installation of an alpha-energy analyzer in the Redox laboratory will do much to solve this problem.)
- c. Decisions as to waste discards should be made only after direct determination of the neptunium content. Discard of unusual quantities (more than two per cent of the feed) should be reviewed and approved by the Manager, Process Chemistry, or his authorized delegate.

K. M. Harmon

Manager, Process Chemistry
Research and Engineering
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