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RECEIVED BY DTIE MAY 22 1970

B PLANT RECOVERY OF CESIUM FROM PUREX SUPERNATANT

H. L. Brandt

March 23, 1970

Atlantic Richfield Hanford Company

Richland, Washington 99352

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B PLANT RECOVERY OF CESIUM FROM PUREX SUPERNATANTINTRODUCTION

A column of zeolite in B Plant is used to remove cesium by ion exchange from the supernatant liquid in Purex stored alkaline waste (PSN) and Purex sludge supernatant (PSS). The same column is also used to remove cesium which has been recovered from Purex current acid wastes (CAW) by phosphotungstate (PTA) precipitation. The CAW, however, has a much smaller loading volume and is processed on a campaign basis between PSN and PSS runs. The cesium, after removal by the zeolite, is eluted as aqueous ammonium carbonate. The carbonate is concentrated, transformed to cesium nitrate, and stored.

Several improvements in B Plant ion exchange have been made since plant startup. The purpose of this report is to provide an updated flowsheet, which shows the effect of these changes, for PSN processing.

PROCESS DESCRIPTIONDiscussion

A high degree of decontamination of cesium from sodium is obtained during the loading step as the PSN is pumped through the B Plant ion exchange column. The column contains a fixed bed of zeolite, an alumino-silicate crystal, which is used as a cation exchanger. This exchanger has a much higher selectivity for cesium than for sodium as these two cations displace ammonium ion from the bed which was put there by the ammonium carbonate eluent during the previous exchange cycle. Further sodium removal from the exchanger bed is achieved with a stream of dilute ammonium carbonate in a subsequent scrub step. Finally, the cesium is removed as a relatively pure product from the zeolite during the elution step with strong ammonium carbonate. The ammonium carbonate containing the cesium is distilled for reuse while the product cesium solution is simultaneously concentrated for subsequent nitration and storage.

The plant equipment has a demonstrated capability to recover greater than 95 percent of the cesium from PSN. The purity of the product cesium has been consistently better than the established standard of less than ten moles of sodium per mole of total cesium. Recently, the purity has improved significantly with several of the latest ion exchange runs having a product with the sodium-to-cesium molar ratio as low as two.

The individual steps and equipment details are described below. A flow diagram including a material balance and time cycle are shown in Figure 1. Figure 2 shows the process solutions required for ion exchange makeup.

Loading Step

PSN feed for the ion exchange is pumped to the cesium supernatant receiver (TK-17-2) from TK-105-C (500,000-gallon capacity) in C Farm. From tank 17-2 the feed is pumped to the IXF pump tank (TK-18-3) and from there to the top of T-18-2, a steel cylinder 6 feet in diameter by 12 feet in height, for downflow loading. The column contains a fixed bed of approximately 11,000 pounds (1850 gallons) of a synthetic zeolite, AW-500*. Its ultimate capacity is over 15,000 pounds (2500 gallons) of zeolite.

The working exchange capacity of the column for the PSN and PSS processed in recent months is about 0.5 MCi. Thus, about 83,000 gallons of a "typical" feed with a cesium concentration of 6 Ci/gal can be loaded with less than 5 percent instantaneous breakthrough. The capacity of the zeolite is inversely related to the sodium-to-cesium molar ratio. This value for capacity is therefore conservative since the exchanger has to accommodate feed with the highest sodium-to-cesium ratio. No significant difference between PSN and PSS has been noted in the sodium-to-cesium ratios. The ratios for both PSN and PSS range between 5000 and 15,000.

The CAW cesium has a sodium-to-cesium ratio of less than 100 and, consequently, loads cesium onto the exchanger with no detectable loss. The favorable sodium-to-cesium ratio allows the cesium to be taken up early in the zeolite bed so that considerably more CAW cesium can be exchanged onto the bed with CAW cesium than from PSN.

The waste stream from the PSN is monitored for cesium by a gamma energy analyzer (RR-18-2-3). The waste is routed to the IXW receiver (tank 18-1). Demineralized water follows the loading step to remove residual feed and to rinse the zeolite particles. At this phase, the sodium-to-cesium ratio in the zeolite lattice ranges from about 50-100 to 1.

Sodium Removal Step

A large number of the sodium ions which exchange with the ammonium ions in the zeolite during the loading step are removed by a 0.2M

*Trademark of the Linde Division, Union Carbide

ammonium carbonate-ammonium hydroxide scrub solution. A 20,000-gallon volume of scrub is used downflow without removing a significant quantity of cesium. In this step, the sodium-to-cesium ratio is reduced to less than ten on the zeolite.

Cesium Elution Step

The cesium is removed from the exchanger by 13,000 gallons of an upflowing $3\text{M } (\text{NH}_4)_2\text{CO}_3 - 2\text{M } \text{NH}_4\text{OH}$ solution at 50 C. The large concentration of NH_4^+ overcomes the greater affinity that Cs^+ has for the cationic exchange site in the zeolite by mass action and reconverts the zeolite to the ammonium form, thus preparing it for the next cycle. The dilute product solution is routed to the IXP receiver (tank 19-1).

Cesium Carbonate Concentration Step

The IXP receiver serves also as the feed tank for the IXP concentrator (E-20-2). The dilute cesium product is concentrated by a factor of about fifty. Up to three IX runs can be cumulatively processed in E-20-2. The overheads from E-20-2 are condensed in the E-20-3 condenser and collected in the cesium eluent storage tank (tank 21-1). The non-condensable gases from E-20-3 are vented to an ammonium carbonate absorber (T-21-2) on top of TK-21-1. Liquid is recycled from TK-21-1 to the top of the absorber to recover any NH_3 and CO_2 before these gases are vented to the NH_3 scrubber (T-22-2) on top of the condensate receiver tank (tank 22-1). To make up for gas losses incurred in the process and through the absorber, NH_3 and CO_2 are sparged as required into the liquid in TK-21-1 from their respective headers.

Nitration of Concentrated Carbonate

After the cesium carbonate has been concentrated in E-20-2, the carbonate solution is retained for transformation to the nitrate. To accomplish this, 2M to 3M HNO_3 is pumped through the upper spray nozzle at 6 to 9 gpm from TK-20-2 by way of Nozzle 47D onto the boiling carbonate solution held at a nominally constant liquid level in the concentrator. An excess of nitric acid is added so that not only is the conversion to nitrate complete, but also the solution has a H^+ of 0.05M or greater. The chemical reaction generates CO_2 and H_2O but at a rate well below the capacity of the off-gas system to handle the gases. After the acid addition has been completed, the liquid volume is further concentrated to about 180 gallons. The nitrate solution is then transferred by jet to TK-20-1. After the nitrate has been removed from the concentrator, a water calibration of that vessel is made to determine the volume of residual solids.

FIGURE 1

CESIUM RECOVERY FROM PUREX ALKALINE SUPERNATANT
BY LINDE AW-500 ZEOLITE ION EXCHANGE

<u>PROCESS STREAM</u>	<u>MAKEUP TANK</u>	<u>COMPOSITION</u>
Bed Makeup	H-317	Linde AW-500 and Water
Sodium Eluent	MNB-173 or TK-21-1	0.2M $(\text{NH}_4)_2\text{CO}_3$; 0.1M NH_4OH
Cesium Eluent	TK-21-1	3M $(\text{NH}_4)_2\text{CO}_3$; 2M NH_4OH

FIGURE 2
PROCESS SOLUTIONS