BNWL-SA--1945
CONF-690712--1

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Ion Exchange - A Generic Nuclear Industry
Process for the Recovery and Final Purification
of Am, Cm, Pm, Sr, Pu, Np, Cs, Tc, Rh and Pd

by

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October, 1968

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For presentation at the "International Conference on Ion Exchange in the Process Industries," London, England, July 16-18, 1969.

This work was conducted by Battelle-Northwest for the United States Atomic Energy Commission under Contract AT(45-1)-1830.

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SYNOPSIS

The versatility of a shielded ion-exchange facility operated adjunct to a reactor fuel reprocessing plant is discussed. The feasibility and economic advantage of using such a processing facility for campaign recovery and purification of a large number of valuable elements is pointed out, and several separations flowsheets are briefly reviewed.

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INTRODUCTION

As the nuclear industry, particularly the nuclear power industry, has expanded, an ever increasing number of valuable elements, both stable and radioactive, have become available in gram to kilogram quantities. The potential economic and scientific value of these elements has enticed the chemist to develop processes for their recovery at high purity. Because of the need for shielding, confinement, remote operation of the separations equipment, and other expensive restrictions inherent to the processing of highly radioactive materials, processes and facilities which yield a maximum degree of flexibility with a minimum capital investment are most attractive. Great versatility, simplicity of operation, and high separations efficiency with small amounts of material qualify ion-exchange processing for such consideration.

The versatility of a properly engineered ion-exchange facility installed adjunct to a conventional large-scale chemical separations plant has been demonstrated at the U. S. Atomic Energy Commission's Hanford complex. The purpose of this paper is to describe the salient features of this facility and review briefly both the purification processes which have been carried out in this facility and others which have been demonstrated in other equipment, but which are adaptable to the currently existing facility. It is not the intent of this paper to suggest that this facility be used for the

purposes presented, but that a pilot plant such as this would have potential capability. Individual processes cannot be explored in depth, nor is it the function of this paper to critically review all processes that might have potential application for the purification of these elements.

DISCUSSION

Ion-Exchange Pilot Plant

A small ion-exchange pilot plant has been installed in one cell of the High Level Radiochemistry Facility operated for the United States Atomic Energy Commission by Battelle-Northwest. This facility consists of three manipulator cells shielded with 1.22 m of high density concrete, a small shielded tank farm containing six tanks ranging in capacity from 0.95 m³ up to 68 m³, a ventilation system, cask handling equipment (up to 2.3 x 10⁴ kg) and an assortment of instrumentation and building services. Each cell is lined with polished stainless steel and has floor drain access to a shielded waste tank, a feature which greatly facilitates the decontamination of in-cell equipment.

Nine ion-exchange columns are installed in one 4.57 m wide x 2.13 m deep x 3.35 m high cell. The 2.74 m high columns range from 356 mm I.D. down to 25.4 mm I.D. and each is water jacketed for controlled temperature operation. Five columns, 102 mm I.D. and larger, are stainless steel while the four smaller units are glass. Also in the cell are requisite tanks, valves, and instrumentation. The instrumentation includes three particularly

useful units: an in-cell gamma spectrometer,¹ which can "read" both individual samples and in-line streams, an automated optical spectrometer² for in-line monitoring, and an optical periscope. The sensing units of both spectrometers are in the shielded cell, and the instrumentation is in the operating gallery. The piping arrangement is such that one or more columns can be connected together in series by use of flexible polyethylene jumpers, with the in-line instrumentation sensing units interposed between them or down-flow from the final column. Figure 1 shows a partial view of the metal and glass columns and valve system.

Purification of Fission Product Strontium

The cation-exchange purification of fission product Sr was demonstrated in 1961 utilizing a much lower capacity, less versatile, six-column, pilot plant installed in the manipulator cell described previously.³ In seven production runs, 75,000 Ci* of ⁹⁰Sr were isolated and purified. The isotopic purity of the product was 56% ⁹⁰Sr and the average chemical purity was 95% Sr. The ⁹⁵Zr-Nb contamination of the product was less than 5×10^{-5} Ci/Ci ⁹⁰Sr, and the ¹⁴⁴Ce-Pr contamination was less than 1×10^{-5} Ci/Ci ⁹⁰Sr. As a result of process improvement during the campaign, the final production run yielded 16,500 Ci ⁹⁰Sr in an 8×10^{-3} m³ product solution containing less than one Ci total radiochemical impurities. The chemical purity exceeded 98% Sr.

*Ci is the adopted abbreviation for curie.

Figure 1
Shielded Ion-Exchange Facility

Processing consisted of four steps: feed preparation, feed absorption, elution and product removal. The approximate composition of the feed stock was:

$$\text{Sr} = 0.32 \text{ kg/m}^3 (2.6 \times 10^4 \text{ Ci/m}^3)$$

$$\text{Ca} = 0.1 \text{ kg/m}^3$$

$$\text{Ba} = 0.05 \text{ kg/m}^3$$

$$\text{Fe} = 8.0 \text{ kg/m}^3$$

$$\text{Pb} = 5.6 \text{ kg/m}^3$$

$$\text{Na} = 10.0 \text{ kg/m}^3$$

$$^{95}\text{Zr-Nb} = 7 \times 10^4 \text{ Ci/m}^3$$

$$^{144}\text{Ce-Pr} = 7.5 \times 10^5 \text{ Ci/m}^3$$

To prevent the absorption of Ce, Fe, Pb and a large fraction of the Zr-Nb during the feed absorption step, N-hydroxyethylethylenediaminetriacetic acid (HEDTA) was added and the feed solution pH was adjusted to pH 4 with NH_4OH . Under these conditions, the stated impurities are strongly complexed by HEDTA and are not absorbed by the resin. Following a water wash, the absorbed band was eluted less than half a band length with a 0.015 molar solution of ethylenediaminetetraacetic acid (EDTA), buffered to pH 8.5 with NH_4OH , to affect a separation of Ca, Sr and Ba into individual bands. Three 102 mm I.D. glass columns filled to a depth of 2.5 m with Dowex 50W, X-12 (20-50 mesh)* resin were used for feed absorption and three 50.4 mm I.D. glass columns filled to the same depth with Dowex 50W, X-12 (50-100 mesh) resin were used for the elution cycle. Since Ca^{+2} precedes Sr^{+2} in this elution sequence,

*A product of Dow Chemical Co.

Ca^{+2} was used as the restraining ion band on the first two elution columns and Y^{+3} on the final elution column. The function of a restraining ion is described elsewhere.⁴⁻⁵ By proper valve manipulation, the leading Ca, a small amount of Sr and the trailing Ba were routed to waste and only the pure Sr was eluted onto the final column behind the Y^{+3} restraining ion. The Y^{+3} barrier was then preferentially removed with an HEDTA solution buffered to pH 4 and the purified Sr was removed with HNO_3 or $\text{HNO}_3 \cdot \text{NH}_4\text{NO}_3$. The photograph in Figure 2, taken through a 1.22 m thick lead glass window and utilizing only the light emitted by the ^{90}Sr decay, shows the product removal step.

In the intervening years, much has been learned about this type of process. Utilization of technology that will be disclosed in the following section and only three of the nine ion-exchange columns in the now-existing facility would permit purification of up to 400,000 Ci of ^{90}Sr per run. The cycle time would not exceed 72 hours and the product purity would be much enhanced over that previously obtained.

Purification of Promethium

Among the heat producing radioisotopes, ^{147}Pm has unique value because no intense primary gamma radiation is emitted and the soft beta radiation yields a maximum bremsstrahlung energy of only 0.225 MeV. Fission product Pm contains minor amounts of ^{146}Pm and ^{148}Pm , both of which emit hard gamma radiation. However, the 42-day ^{148}Pm is removed from consideration by aging prior to final processing and ^{146}Pm contaminates ^{147}Pm to the extent of only 0.2 to 0.3 ppm by activity in current Hanford waste. The contamination level is slightly higher in power reactor waste and is a function of reactor design and operation. Thus,

Figure 2
Elution of Sr^{90} From Ion-Exchange Column

to yield a product of value, the Pm separations process must remove Eu to levels less than 10^{-7} Ci $^{154}\text{Eu}/\text{Ci } ^{147}\text{Pm}$ and other radionuclides to even lower levels. To reach this level the impurity decontamination factors* must exceed 10^6 .

The Pm purification effort at Hanford has typified the value of an ion-exchange pilot plant coupled to a conventional separations plant. As with the Sr program, the feed material for the Pm ion-exchange purification was recovered from Purex high-activity waste by a series of precipitation, solvent extraction and thermal concentration steps performed in large complex facilities by personnel of the Atlantic Richfield Hanford Company. The solution contained all of the rare earth fission products, Am, small amounts of Fe, Cr, Al, Pb, Na and traces of other fission products in dilute HNO_3 .

The initial Pm purification process utilized the much publicized ethylenediaminetetraacetic acid (EDTA) flowsheet developed at the Ames Laboratory,⁵ but with two modifications, Zn^{+2} was used in place of Cu^{+2} as a restraining ion and the temperature was increased to 60°C , thus improving the kinetics of the system and permitting a 3-fold increase in elution rate. Cu^{+2} cannot be used because of partial reduction to the metal in the presence of the eluting agent and a high radiation field.⁶ As a result of process experience and laboratory research, the eluting agent was changed first to diethylenetriamine-pentaacetic acid (DTPA) and then to nitrilotriacetic acid (NTA).⁷⁻⁹ As these two eluting agents are efficient for other separations, the operating conditions found optimum are given in Table I.

*Decontamination factor or D.F., defined as:
$$\frac{(\text{conc. impurity}/\text{conc. Pm})_{\text{feed}}}{(\text{conc. impurity}/\text{conc. Pm})_{\text{product}}}$$

Table I
Eluting Conditions Used

	<u>DTPA</u>	<u>NTA</u>
Eluant Concentration	20 kg/m ³	20 kg/m ³
Eluant pH	6.5	6.4
Buffering Reagent	NH ₄ OH	NH ₄ OH
Elution Flow Rate	5.8 x 10 ⁻⁴ ms ⁻¹	13.3 x 10 ⁻⁴ ms ⁻¹
Column Operating Temperature	60-65°C	60-65°C
Restraining Ion	Zn ⁺²	Zn ⁺²
Resin	Dowex 50W, X-8 (50-100 mesh)	

The initial EDTA separation run required 21 days of continuous operation and yielded 62,000 Ci of pure product. With the NTA flowsheet now in use and full utilization of the processing equipment, over 300,000 Ci of pure Pm are obtained in only 3 days. The product contains less than 10⁻⁹ Ci of radiochemical impurities per Ci of ¹⁴⁷Pm and the chemical purity exceeds 99%, the only detectable impurities being Nd and Sm. Higher chemical purity is obtainable but because of the decay of ¹⁴⁷Pm to stable ¹⁴⁷Sm at a rate of 2.2% per month, higher purity is of little significant value. In excess of three million curies of Pm have been purified in the past three years.

Purification of Americium and Curium

Using flowsheets similar to those previously described, the elution sequences of Am⁺³ and Cm⁺³ were determined using several amino polyacetic acid eluting agents. The DTPA and NTA results proved to be the most interesting and are shown in Table II.

Table II
Ion-Exchange Elution Sequence

With DTPA

With NTA

In the DTPA system, Cm and Am elute close to each other and on the trailing edge the Er-Ho-Dy triad which does not separate. Gd and Eu elute in order but with little separation between them. With NTA, the sequences of Y and Am are altered. The two flowsheets thus complement each other. Since very little Dy could be expected in any fission product mixture, DTPA would yield a relatively clean Cm-Am fraction with good separation from Gd and Eu. NTA could then be used in a second cycle to separate Cm and Am from each other.

This flowsheet was tested in December, 1967.¹⁰ Atlantic Richfield Hanford Company personnel processed 13 1/2 tons of Shippingport Power Reactor blanket rods and recovered the lanthanide-actinide elements which included approximately 140 grams Pm, 50 grams of ^{144}Cm and 1,000 grams of a mixture of ^{241}Am and ^{243}Am . Following transfer by shielded cask to the ion-exchange facility, this material was processed through the two-cycle

flowsheet. Because of the quantity of material, two DTPA runs were required to separate Cm and Am from the rare earth. One NTA run was then used to successfully separate the Cm from the Am. Figures 3 and 4 show the DTPA and NTA product elution curves as obtained with the in-cell gamma spectrometer. Because of the small amount of Cm relative to Am, the binary mixing zone is relatively large. This could be corrected by using a smaller diameter final column or feed containing a higher ratio of Cm to Am. The ^{147}Pm contained in this feed was recovered from the DTPA cycles.

The large amount of ^{244}Cm provided a meaningful test for ion-exchange processing. Gas generation and radiolytic destruction of the resin and eluting agent proved to be problems that were not overwhelming. With proper process improvement, separations involving several hundred grams of ^{244}Cm are reasonable to contemplate.

Recovery and Purification of Cesium

At the Hanford Plant, high activity wastes from the chemical separations plants are concentrated, neutralized, and stored in large underground tanks. Most of the fission products and inert components then precipitate and settle to the bottom of the tanks, but Na and Cs remain in solution. A rather large amount of research has been conducted and various procedures have been proposed for the recovery of Cs from such alkaline waste¹¹⁻¹³ as well as neutral or acidic waste,¹⁴⁻¹⁵. A number of ion-exchange materials exhibit high selectivity for cesium. These include the mineral exchanger clinoptilolite, synthetic alumino silicates such as Decalso*, the synthetic zeolites, and several synthetic phosphate exchangers.

*Manufactured by Permutit Co.

Figure 3

Figure 4

In general, these proposed recovery flowsheets are simple and specify pumping untreated waste supernate through a bed of the exchanger or a series of beds until activity breakthrough is achieved. The loaded bed is washed with pure water and then the cesium is removed. Utilizing shielded casks filled with Decalso, and transported by rail, over 2×10^6 Ci of Cs have been recovered from the alkaline supernate held in Hanford waste storage tanks, transported to another AEC installation, eluted from the resin casks and processed into radiation sources.

Recovery of Technetium, Rhodium and Palladium

C. A. Rohrmann¹⁶ and more recently Deonigi, McKee and Haffner¹⁷ have drawn attention to the impressive quantities of these three and many other immensely valuable isotopes that will be available for recovery in a very few years as by-products of the power reactor reprocessing industry.

⁹⁹Tc is absorbed to high capacity by strong base anion-exchange resins (such as Rohm & Haas Co. IRA-401), even in the presence of strong concentrations of sulfate, carbonate, nitrate and nitrite ions. Recovery and purification has been demonstrated on both a pilot plant and full production basis using a fixed bed operation.¹⁸ The processing steps include absorption of Tc from the alkaline feed solution, successive wash steps with 1 molar NaOH, then water and finally a 0.25 molar HNO₃ combined wash and nitrate ion conversion, then elution of the product from the resin bed with 6 molar HNO₃. Work with Rh and Pd is not as advanced, but the use of multiple cycles coupled with selective elution has yielded these elements in high purity. Over a kilogram of Tc and gram amounts of the others have been recovered and purified.

Separation and Purification of Plutonium and Neptunium

Perhaps the classic example of the advantageous use of ion-exchange processing in the nuclear industry is the anion-exchange purification of plutonium in the nitric acid system, a process now employed in more than half a dozen processing plants in the United States and Europe. Durham and Mills¹⁹ found that Pu^{IV} exhibits very high distribution coefficients on quaternary amine anion-exchange resin from 7 molar nitric acid or metal nitrate systems of similar nitrate concentration. An extensive investigation of the equilibria and kinetics of both the absorption and desorption steps in this system with most commercially available resins provided the sound basis for this very simple three-step process.²⁰⁻²²

Pu^{IV} feed solution containing 6 to 8 molar HNO_3 is pumped through the resin at rates as high as $0.013 \text{ kg Pu s}^{-1}\text{m}^{-2}$. The loading progress can be followed by visual observation of the movement, down the column, of the dark green absorption band if glass columns are used or by detectors on the effluent solution if a metal system is used. When the desired loading is achieved, the loaded resin is washed with HNO_3 of the same concentration to remove impurities. Since the common impurities have very low distribution coefficients on the resin, impurity decontamination factors are very high. Removal of the Pu is accomplished by elution with 0.35 molar HNO_3 .

The Pu hexanitrate ion is very large and considerable resin contraction takes place on loading. As a result the process is kinetically limited and careful consideration must be given to the choice of resin and the operating temperature if optimum throughput is required. Under optimum conditions

resin capacities as high as 1.2×10^{-4} kg Pu m^{-3} can be obtained with Dow Chemical Company's Dowex-1, X-4 (50-100 mesh) resin and 0.70×10^{-4} kg Pu m^{-3} with Permutit Company's Permutit SK (20-50 mesh) resin. The product concentration is largely independent of resin loading for these two resins, and concentrations in excess of 4.0×10^{-5} kg Pu m^{-3} can be obtained. In this process which employs HNO_3 as the sole process chemical, uranium decontamination factors of 10^6 and fission product decontamination factors of 5×10^3 have been demonstrated in a single cycle. Fission product decontamination factors of 4×10^7 have been demonstrated in two successive cycles.

J. L. Ryan found that by employing the proper oxidizing agents, the anion-exchange process could be adapted to include Np.²³ When ferrous sulfamate and hydrazine are added to a 6 to 7 molar HNO_3 solution containing Pu and Np, their oxidation states are adjusted to Pu^{III} and Np^{IV}. Pu^{III} is oxidized to Pu^{IV} when this solution is heated and air sparged. Both are then absorbed by the anion-exchange resin when this solution is pumped through the resin column. Employment of an 8 molar HNO_3 co-decontamination wash containing a little KF is effective in removing Th and fission products from the loaded resin. Pu is then reduced to Pu⁺³ and removed from the resin by elution with 5.7 molar HNO_3 containing ferrous sulfamate and hydrazine. The Np is removed with 0.35 molar HNO_3 . The Pu fraction can be processed through a straight HNO_3 -second cycle to remove impurities and yield a concentrated product. This process has been demonstrated on a pilot plant scale for processing ²³⁸Pu.

CONCLUSIONS

The flowsheets used for the final purification of Sr, Pm, Cm and Am are specific examples of restrained-band displacement chromatography. In such systems, i.e., the separation of Pm from Sm and Nd or the separation of Sr from Ca and Ba, resin selectivity is not a significant factor since the separation is an aqueous phase process governed by the relative stability of the complexes formed between the chelating agent and the ions to be separated. The resin provides a fixed matrix with which the uncomplexed ions can exchange and the pre-absorbed restraining ion constrains the leading edge of the absorption band, forcing multiple exchange (multiple application of the separation coefficient) to take place and producing efficient separation. With the variety of chelating agents now available, this process can be used to separate a very broad spectrum of the chemical elements.

The flowsheets described for the other separations represent examples in which the resin exhibits unusually high selectivity for a particular chemical species or in which the aqueous system chosen produces a complex ion for which the resin is highly specific. These separations are usually much less expensive and more direct than displacement chromatography but are applicable to a restricted number of elements.

This paper by no means represents an exhaustive review of all applicable ion-exchange work. Rather, the intent is to show a variety of processes and the broad spectrum of interesting and valuable elements that can be efficiently purified on a campaign basis in a single rather inexpensive facility.

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