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PURIFICATION OF KILOCURIE QUANTITIES OF
PROMETHIUM-147 BY ION EXCHANGE

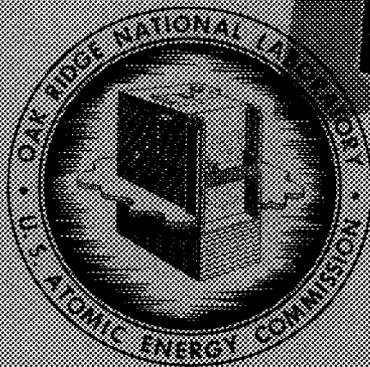
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ISOTOPES DIVISION

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R. S. Pressly
C. L. Ottfinger
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R. S. Pressly P. B. Orr
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ABSTRACT

Twenty thousand curies of Pm^{147} in a mixture of rare-earth fission products was processed by ion exchange on a cation resin, using ammonium citrate solution as the eluant, until the desired purity of Pm^{147} was obtained. Promethium was present to the extent of about 1.6% in the starting mixture and was increased to 60% in the final product.

Procedures, typical elution curves, methods of analysis, losses, material balances, and a description of the equipment are given for the process.

Multikilocurie quantities can be produced with similar equipment, with little difficulty contributed by radiation or radiation damage when feed material of the appropriate concentration is available.

INTRODUCTION

One of the major efforts in the Isotopes Division of Oak Ridge National Laboratory is the separation of fission products from reactor wastes. Although the long-lived fission products have been available in curie quantities for several years, the Fission Products Pilot Plant (F3P) provides the first relatively large-scale production of these isotopes. Kilocurie amounts of Cs^{137} , Ru^{106} , Sr^{90} , Ce^{144} , and Pm^{147} , as well as grams of Tc^{99} , can now be processed annually.¹

Increasing interest in Pm^{147} has developed because this isotope has a combination of properties which makes it attractive for the preparation of large radiation sources. It has a reasonably long half life (2.6 years); it decays by low-energy beta emission only, thus minimizing radiation hazards; and its compounds can be fabricated into sources with relative ease. Some of the potential uses for large quantities are as follows:

1. self-luminescent devices (the low specific ionization of the beta particles results in less damage to sources),
2. direct beta radiation sources (large sources can be used for nonpenetrating radiation treatment of surfaces for sterilization),
3. secondary x-ray sources,
4. devices for the conversion of radiation energy into electrical energy.

¹E. Lamb, H. E. Seagren, and E. E. Beauchamp, *Proc. U.N. Intern. Conf. Peaceful Uses Atomic Energy, 2nd Geneva, 1958* 20, 38 (1958).

Historical

Element 61, promethium, evidently does not exist in nature.² The first characterization of this element was done by Marinsky, Glendenin, and Coryell.³ Using ion exchange techniques to investigate rare-earth fission products, these workers identified Pm^{147} as the daughter of Nd^{147} and, by studies of order of elution from cation resin, proved it to be an isotope of element 61. Weighable quantities were produced later as more enriched rare-earth fractions became available.⁴ Fourteen isotopes of promethium have been identified, but most of these have such short half lives and/or low fission yields that they can be neglected during the processing of Pm^{147} . However, the 42-day isotope Pm^{148} contributes gamma activity to relatively short-decayed promethium fractions and necessitates using long-decayed wastes for the production of promethium for use as a pure beta emitter.

The development of ion exchange processes has provided an excellent method for the separation of rare earths. Other techniques such as fractional crystallization, complex formation, and

²G. E. Boyd, *J. Chem. Educ.* 36, 3 (1959).

³J. A. Marinsky, L. E. Glendenin, and C. D. Coryell, *J. Am. Chem. Soc.* 69, 2781 (1947).

⁴G. W. Parker and P. M. Lantz, *The Separation of Milligram Quantities of Element 61 From Fission [Products]*, AE CD-2160 (June 18, 1948).

fractional precipitation are tedious and seldom very effective. Solvent extraction has interesting possibilities but has not been used extensively for rare-earth separations. Ion exchange can be readily adapted to the processing of radioactive solutions because the equipment needed is relatively simple, easily shielded, and can be adapted for remote-control operation. From tracer levels to amounts of several curies, ion exchange has been used successfully to separate radioactive rare earths.

Separation of Pm¹⁴⁷ at ORNL

Separation of Pm¹⁴⁷ for sale by Oak Ridge National Laboratory was begun in 1950. An ion exchange method for the separation of fission products yielded a fraction enriched in rare earths, and this fraction was further processed by ion exchange to partially purify promethium.⁵ Later, a rare-earth fraction was separated from aged waste solutions of fission products by precipitation as oxalates; ion exchange was used to separate promethium from the other rare earths.⁶

As the demand for this isotope increased, production increased from 11 mc in 1950 to several curies per year by 1957 (see Table 1).

⁵P. O. Schallert, *Production Separations of Fission Product Groups for the Radioisotope Program*, ORNL-1144 (July 28, 1952).

⁶R. S. Pressly, *Preparation of Fission Rare Earth Isotopes*, ORNL-2252 (May 7, 1957).

Table 1. Annual Production of Pm¹⁴⁷ at ORNL

Year	Millicuries Produced
1950	11
1951	295
1952	1,937
1953	0
1954	6,247
1955	19,250
1956	53,405
1957	15,178
1958	828,869

In 1958 a process was developed to provide Pm¹⁴⁷ in larger quantities.⁷ This process differed from previous ones in that solvent extraction was used to separate the rare earths from other fission products and inactive materials in waste solutions. Tributyl phosphate was used to remove rare earths and yttrium from aluminum nitrate wastes obtained from the Idaho Chemical Processing Plant. After the stripping from the tributyl phosphate, the cerium was removed by iodate precipitation and the promethium purified by ion exchange. Approximately 830 curies of Pm¹⁴⁷ was prepared by this method; the process was discontinued with the startup of the F3P.

At the F3P, waste solutions are processed to yield a fraction containing yttrium and the light rare earths from cerium to europium. Removal of most of the cerium and yttrium by solvent extraction provided a rare-earth feed material for ion exchange separation.

Objective of the Project

The project described in this report was designed to demonstrate the use of large-scale ion exchange equipment to separate kilocurie quantities of Pm¹⁴⁷ from the other rare-earth fission products and to prepare the promethium into a form suitable for source fabrication. Specifications for the product were as follows:

1. activity concentration of greater than 450 curies of Pm¹⁴⁷ per gram of total rare-earth oxide,
2. gamma contamination of less than 3 μ C per curie of Pm¹⁴⁷.

PROCEDURE

Equipment

The ion exchange columns for the purification of Pm¹⁴⁷ were installed in a lead-and-concrete-shielded cell.

The following is a list and description of the equipment used to purify multicurie quantities.

Transfer Vessel for Crude Promethium. - A lead-shielded stainless steel tank with a capacity of about 45 liters and a weight of about 6 tons was used for feed transfer. Two Snaptite fittings were used for ease in filling and emptying. One

⁷C. L. Ottinger and R. S. Pressly, ORNL, unpublished work.

fitting was connected to a short leg and the other to a long leg inside the carrier cavity.

The carrier was filled with a rare-earth nitrate solution at the F3P and transferred to Building 3028 for promethium separation. To remove the solution from the carrier and transfer it to columns 1 and 2, air pressure of about 12 psi was used. A stainless steel solenoid valve, actuated by the liquid-level instrument on column 1 or 2, controlled the flow from the transfer vessel.

Column Feed Tanks: FT-1, -2, -3, and -4. – Four polyethylene tanks with a capacity of about 195 liters each were located on the third floor of the building. Liquids from the tanks flowed through rotameters before entering the columns. A stainless steel solenoid valve, actuated by a liquid-level instrument on each column, controlled the flow from the feed tanks to the columns.

Ion Exchange Columns 1 and 2. – These were stainless steel roughing columns 3 in. in diameter by 8 ft long, each with a capacity of 10 liters of resin.

The free space above the resin in these columns contained a liquid-level gage equipped with limit switches to control the maximum and minimum level above the resin. The limit switches activated solenoid valves in the feed tank lines, the line from the crude promethium transfer vessel, or the line from collection vessel CP-1, depending upon the source of the solution feeding the columns.

Discharge lines from these columns were made of polyethylene tubing to eliminate corrosion and the resulting pickup of foreign cations by the resin, which would be produced by the action of citric acid on stainless steel. Each discharge line penetrated an ionization chamber used to monitor the flow of activity from the column. Unfortunately, due to the weak activity of Pm^{147} and also to the interference due to the strong Eu^{154} gamma radiation, promethium was only slightly detected. The main function of the ion chamber was to detect the breakthrough and peak of Eu^{154} , in order to determine sampling frequency. Samplers were located on the discharge lines from the columns.

Ion Exchange Column 3. – Column 3 was also 3 in. in diameter by 8 ft long and was lined with tantalum to reduce the release of foreign cations by corrosion. Optional column heating was available by means of Calrod units on the outside

surface. Temperature control was maintained by thermocouples and related instruments. The eluant was preheated by means of a steam heater before being fed to the column, and the eluate was cooled, upon discharge from the column, by a water-cooled condenser.

Rare-earth feed for column 3 was transferred from collection vessel CP-1 by means of air pressure.

The discharge line from column 3 passed directly through an ion chamber for measuring the activity being eluted. The ion-chamber reading was used as a guide in sampling the eluate.

Ion Exchange Column 4. – A tantalum-lined column 2 in. in diameter by 8 ft long, with a resin capacity of about 5 liters, was used for final purification. Like column 3, it could be heated and fed only from collection vessel CP-1.

The discharge line was of Saran and polyethylene. The only contact the product had with stainless steel was in the solenoid discharge-control valve. The discharge line passed through an ion chamber for monitoring activity as it was removed from the column. If a good separation was obtained and a large quantity of promethium was on the column, the promethium activity could be recorded on the ion-chamber electrometer. However, this approach was not dependable, and frequent sampling was necessary to determine when the promethium was being eluted. Sampling was done by utilizing the product-removal station, which was designed to accept 1-liter plastic bottles.

Collection Vessel CP-1. – Promethium fractions from columns 1, 2, and 3 were collected in a 200-liter stainless steel tank. This tank was equipped with a liquid-level instrument and a solution addition funnel. Solutions were moved from CP-1 by air pressure to columns 3 and 4 or to waste.

Process

Operation. – A simplified flow diagram for the procedure is presented in Fig. 1.

Feed material for the columns was prepared at the Metal Recovery Plant and at the F3P. The uranium processed was long-irradiated natural uranium which had decayed for approximately two years. Dissolution and extraction of the uranium and plutonium was done at the Metal Recovery Plant, and the fission product solution was transferred to the F3P. There the rare-earth fraction

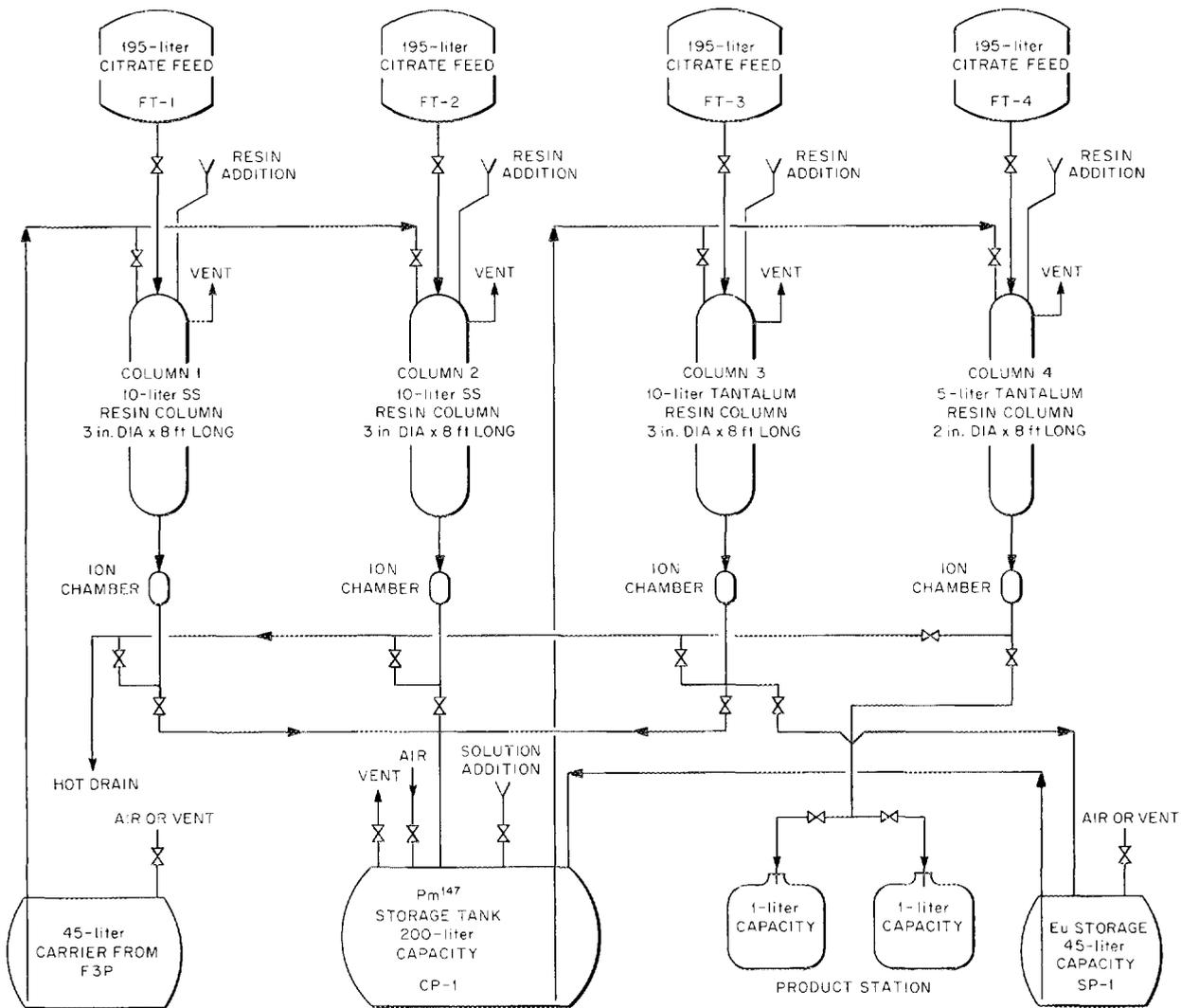


Fig. 1. Flowsheet for Pm^{147} Purification by Ion Exchange.

was separated from the other fission products. A batchwise solvent extraction procedure was used to remove approximately 98% of the cerium and approximately 92% of the yttrium from the rare-earth fraction.⁸ This removal greatly reduced the processing time, cost, and radiation problems in the ion exchange operation.

⁸E. E. Ketchen and T. A. Butler, ORNL, *Solvent Extraction Separation of Cerium and Yttrium from Other Rare-Earth Fission Products* (to be published).

After this initial separation at the F3P, the rare-earth feed was transferred to the ion exchange equipment by means of a 6-ton, 45-liter shielded carrier. Analysis of a typical batch of feed is given in Table 2.

Acid normality of the feed was not allowed to exceed 0.4, since excess acid will spread the rare-earth band on the columns and result in less-efficient separation.

The carrier was placed in a stainless steel tank and connected to a line to columns 1 and 2

Table 2. Typical Feed Analysis

Volume	40 liters
Acidity	0.3 N
Approximate activities, mc/ml	
Ce ¹⁴⁴	3.7
Y ⁹¹	3.0
Eu ¹⁵⁴	0.2
Pm ¹⁴⁷	90.0
Relative rare-earth content, g	
Promethium	1.0
Yttrium	1.6
Lanthanum	9.1
Cerium	0.4
Praseodymium	9.6
Neodymium	28.1
Samarium	8.4
Europium	3.4
Total rare earths/Pm ¹⁴⁷	60.6

by means of a flexible line and quick-opening fittings. Another flexible line was connected to the carrier for use as an air or vent line. A cover was placed on top of the tank after the quick-action fittings were secured to assure protection against leakage when the carrier was pressurized.

Air pressure at 10 to 12 psi was used to force the solution from the carrier directly to columns 1 and 2, one column at a time. These columns contained 10 liters of coarse Dowex 50W, 12% cross-linked, resin in the hydrogen form, prepared from twice-washed 50- to 100-mesh resin. No more than one-third of the resin was loaded with cations from the feed in order to assure enough length of hydrogen-form resin for separation. Loading was done at a rate of about 15 liters/hr, which was usually as fast as the resin would permit the solution to flow. The loaded resin was washed with about 30 liters of distilled water to remove excess acid from the column.

Elution was done with 0.2 M citric acid adjusted to a pH of 3.0 with NH₄OH. Flow was adjusted to one column volume (10 liters) per hour. Both columns 1 and 2 were eluted at the same time.

The eluate was sampled three times per 8-hr shift and analyzed for promethium. When the

electrometer measuring the column discharge showed an increase in activity, sampling frequency was stepped up to once an hour.

Yttrium-91 and mixed europium activities, in that order, were eluted first. These elements were eluted to waste until analysis showed promethium to the extent of about 25 mc/ml, at which time collection was started. Beginning the collection at this point minimized the loss of promethium and reduced the amount of yttrium and europium collected. Collection was stopped when the promethium peak was passed and the activity dropped to about 25 mc/ml. About half of the neodymium and rare earths of lower atomic number was discarded by stopping the promethium collection at this point, eluting the column, and sending the eluate to the waste tank.

Neodymium and other cations remaining on columns 1 and 2 were stripped off with 60 to 70 liters of 0.2 M citric acid at a pH of 5.0 and then washed with approximately 30 liters of distilled water to remove excess ammonium citrate. To prepare the columns for re-use and to be sure that all activities were off the columns, the resin was converted to the hydrogen form with 60 to 80 liters of 3 N HNO₃. Excess acid was removed with 30 to 40 liters of distilled water.

Due to the high starting ratio of total rare earths to promethium, in the feed, little separation of the promethium was done on columns 1 and 2. However, a reduction of the total rare earths by a factor of 2.5 was accomplished. The following is a list of the approximate amounts of the elements remaining per gram of promethium, after this first ion exchange separation:

Element	Weight (g)
Europium	1.7
Samarium	8.4
Promethium	1.0
Neodymium	14.0
Total	25.1

This represented a ratio of 24.1 g of rare earths per gram of promethium.

The promethium fraction collected from columns 1 and 2 was acidified with 1.5 ml of 16

N HNO₃ per liter of eluate to break the rare-earth complex. This adjusted the solution to a pH of about 1 to permit readsorption on the next column. Transfer of the acidified promethium fraction to column 3 was accomplished by applying air pressure (about 14 psi) to the receiving vessel for columns 1 and 2. Column 3 contained 10 liters of the hydrogen-form Dowex 50W, 12% cross-linked, prepared in the same way as that used in columns 1 and 2. The flow rate during adsorption was not allowed to exceed two column volumes (20 liters) an hour. When all the promethium fraction was loaded on the column, it was washed with 30 to 40 liters of distilled water to remove all traces of acid.

Elution of column 3 was done with 0.2 M citrate at a pH of 3.0, with a flow rate of one column volume (10 liters) per hour. Before the column-3 electrometer indicated a rise in activity, sampling was done about three times per 8-hr shift. After the activity began to rise, sampling was done once an hour. The activity was allowed to go to waste until the promethium activity in the samples increased to approximately 25 mc/ml, at which time promethium collection was started. After the concentration of the promethium had reached a maximum and then declined to about 20 mc/ml, collection was stopped. The quantity of the other rare earths in the promethium fraction was reduced 90% by column 3. The ratio of the other rare earths (total) to promethium at this point was about 2.6 to 1.

The pH of the eluant was adjusted to 5.0, and the remaining rare earths on column 3 were stripped to waste. The column was then washed with 30 to 40 liters of water to remove the citrate. Since the ammonium citrate at pH 5.0 may not have removed all the cations, the column was stripped with about 70 liters of 3 N HNO₃ and then washed with 30 to 40 liters of distilled water.

The column-4 procedure for the further ion exchange purification of the rare-earth fraction eluted from column 3 was almost identical with that used for column 3. The fraction was acidified with nitric acid and transferred to column 4 by using air pressure at 12 to 14 psi. Column 4 has a capacity of about 5 liters of Dowex 50W, 12% cross-linked, resin prepared in the same manner as that used in columns 1, 2, and 3. Loading was done at a rate of about 5 liters/hr, after which the loaded column was washed with 15 to 20 liters

of water to remove all traces of acid. Elution was done at a rate of not more than 5 liters/hr, and the eluate was sampled three times per 8-hr shift until the ion-chamber electrometer on the column discharge indicated an activity rise. The electrometer required close observation, since very little hard beta or gamma activity was present on the column to cause a large deflection. When a rise in activity occurred, sampling was done at a minimum rate of once an hour. After the promethium activity in the samples rose to 5 mc/ml, collection was started and was continued until the peak was past and the promethium content dropped to 1 mc/ml. Collection was done in 1-liter polyethylene bottles, and each bottle was analyzed for Y⁹¹, Pm¹⁴⁷, Eu¹⁵⁴, and total rare earths.

At the peak of the promethium elution, the analysis showed as much as 650 curies (689 mg) of Pm¹⁴⁷ per gram of total rare earths. According to the analysis of each fraction, those fractions which met the curies-per-gram specifications were combined as products. Those fractions which were not within specifications were reserved for reprocessing.

Elution Curves. -- A convenient method for describing the performance of an ion exchange column is the elution curve. By plotting concentration vs volume of eluate through the column, a curve results which is characteristic of the performance of a column under a certain set of conditions. Such factors as flow rate, resin size, and operating temperature influence the elution curve; these must be kept constant in order to obtain a definitive curve for operating purposes.

For elutions of rare earths with 0.2 M citrate, the elution curve is bell-shaped, with each rare earth forming a band which is dilute at the front and rear boundaries and more concentrated in the middle. As the elution progresses, these bands move down the column and begin to separate. The degree of separation, that is, the amount of overlap between bands, can be determined by plotting elution curves for all components on the same graph.

In the following figures, several typical elution curves are presented. Activity values are determined by the assay of frequent samples taken from the column outlets. The term "column volume" is here defined as the apparent volume of wet resin in the column after washing and settling. Only promethium and europium, the

components which can be determined by counting techniques, are used in these representative curves.

Figures 2 and 3 show the elution of europium and promethium from 3- and 2-in.-dia columns respectively. The columns are operated continuously at a flow rate of one column volume per hour. Thus for a 3-in.-dia column containing 10 liters of resin, the elution time required to reach the peak for promethium is 40 to 45 hr, and to elute the promethium completely requires about 60 hr.

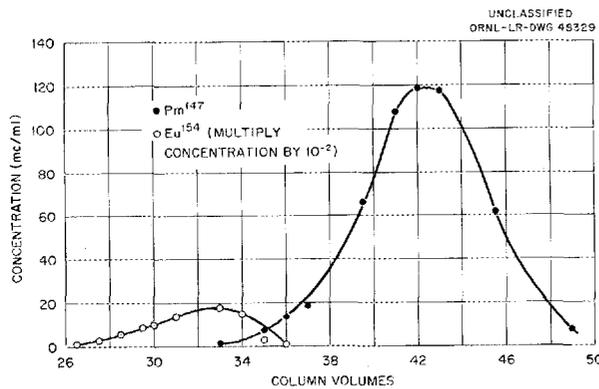


Fig. 2. Typical Elution Curve for a 3-in.-dia Column. Column volume, 8 liters; Dowex 50W, 12% cross-linked resin, 50 to 100 mesh.

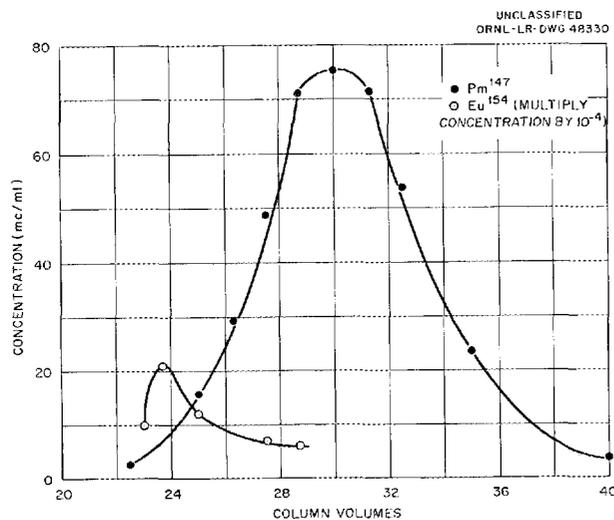


Fig. 3. Typical Elution Curve for a 2-in.-dia Column. Column volume, 4 liters; Dowex 50W, 12% cross-linked resin, 50 to 100 mesh.

The reproducibility of the elution curve is illustrated in Fig. 4. Four runs, containing varying amounts of promethium, are plotted together. Conditions for all these runs were the same except for the amount of activity and the volume of resin in the column; the latter varied from 8 to 10 liters.

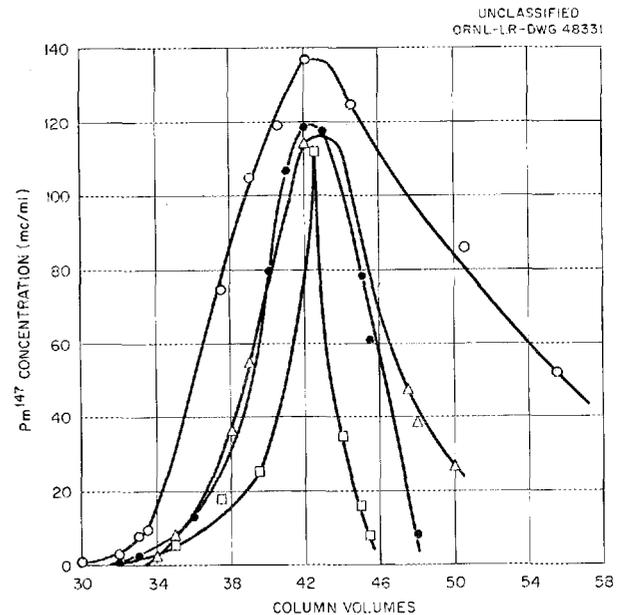


Fig. 4. Elution Curves for Pm^{147} Obtained by Use of 3-in.-dia Column. Volume of resin, 8 to 10 liters.

One condition which has a pronounced effect upon the operation of an ion exchange column is temperature. In Fig. 5, elution curves are presented for two runs under identical conditions except for the operating temperature. Heating of the column accelerates the approach to equilibrium; thus the separation is improved.

Methods of Analysis

Promethium-147. — Promethium-147 was difficult to identify in the presence of other activities because of its low-energy beta decay. An end-window G-M counter was standardized for total disintegrations by using purified Pm^{147} , Y^{91} , Eu^{154} , and other activities which were present. A small aliquot of the solution was evaporated on a watch glass, covered with cellophane, and counted. A 60-mg aluminum absorber was then placed over the mounted sample, which was

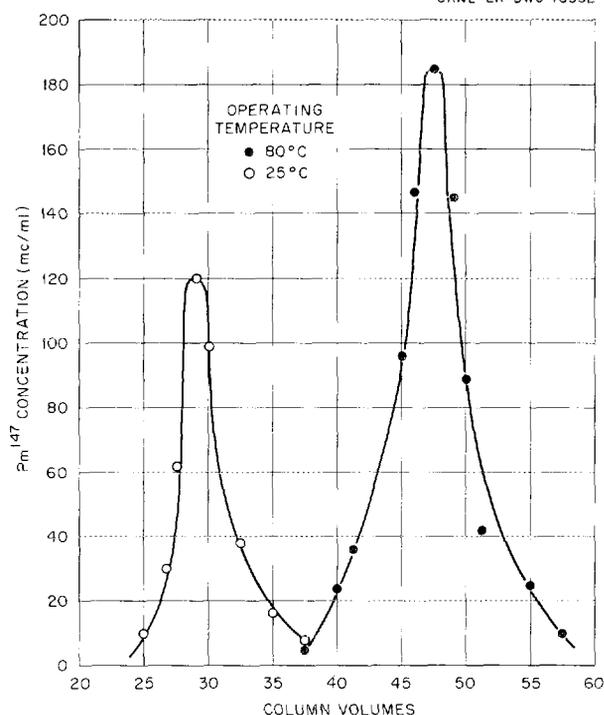


Fig. 5. Effect of Temperature on Elution of Pm¹⁴⁷. Column volume, 4 liters; Dowex 50W, 12% cross-linked resin, 50 to 100 mesh; column diameter, 2 in.; flow rate, 1 column volume per hour.

re-counted. This difference in counts was calculated as Pm¹⁴⁷, although the presence of other rare-earth activities contributed to the calculated amount of Pm¹⁴⁷.

As the Pm¹⁴⁷ was separated into more concentrated and purer fractions, the analyses became more accurate and could be made by beta proportional or liquid scintillation counting.

Yttrium-91. — Calculation of the Y⁹¹ content was based on the beta activity that was not absorbed by the 60-mg aluminum absorber when the G-M counter and the mount for determining Pm¹⁴⁷ were used. This method was not very accurate; however, it was useful when combined with gamma scintillation analysis to determine the increase or decrease of Y⁹¹ during column operations.

Europium-154. — The presence of Eu¹⁵⁴ was determined by using a single-channel gamma spectrometer. When the presence of Y⁹¹ was shown by beta counting, the 970-keV gamma

peak was measured for Eu¹⁵⁴. If Y⁹¹ was not present, the 1.25-MeV gamma peak was measured to determine the Eu¹⁵⁴.

Inactive Rare Earths. — Inactive rare earths were present in quantities dependent upon the irradiation time in the reactor and the decay after removal from the reactor. As these rare earths were separated from Pm¹⁴⁷ by ion exchange, it was necessary to be able to analyze quickly in order to determine the performance of the columns. When the results of the analyses of total rare earths contained in the eluate were associated with the ion-chamber readings and radiochemical analyses, a fairly accurate interpretation could be made as to the quantities of inactive neodymium, europium, and samarium which accompanied or were separated from Pm¹⁴⁷ as the solution proceeded through the columns.

The total rare earths in the samples were precipitated from the 0.2 M citrate solution as oxalates. The precipitate was dissolved in nitric acid which contained hydrogen peroxide, and this solution was heated in the presence of platinum black to remove excess hydrogen peroxide. The solution was then adjusted with sodium hydroxide to a pH of 4.5 to 6.0 and then buffered with acetate. A drop of mercuric Versenate was added, and the rare earths were titrated potentiometrically with 0.05 M sodium Versenate solution. One electrode was a mercury-plated gold wire, and the other was calomel. The potential between the mercury electrode and the mercuric ion in solution was measured. When the endpoint was reached, the potential changed sharply due to a decrease in the concentration of the mercuric ion. By comparison with a standard rare-earth solution, a value for total rare earth was obtained.⁹

Promethium-148. — Promethium-148 is produced by the (*n*, γ) reaction on Pm¹⁴⁷. This isotope has two isomers, one of 5.2-day half life and the other of 42-day half life; the decay scheme of neither of these has as yet been definitely established. Studies have been made by Eldridge¹⁰ and by Bhattacharjee, Sahai, and Baba¹¹ which

⁹H. Kubota, *Anal. Chem. Ann. Prog. Rep. Dec. 31, 1959*, ORNL-2866, p 11.

¹⁰J. S. Eldridge, ORNL, private communication, Aug. 1, 1959.

¹¹S. K. Bhattacharjee, B. Sahai, and C. V. K. Baba, *Nuclear Phys.* 12, 356 (1959).

permit calculation of the amount of Pm^{148} . The disintegrations (630-kev gamma rays) of the 42-day isomer are calculated as representing 80% of the total disintegrations of the isotope.

This analysis was made only on very pure Pm^{147} in which the europium content was less than 10 μC per curie of Pm^{147} .

DISCUSSION

Promethium-147 was received in relatively small quantities, as compared with the capacity of the columns. The product was partially separated from the other rare earths and stored until the ion exchange beds could be loaded for more efficient separations. Fractions of the eluate containing a large ratio of inactive rare earths to Pm^{147} were reprocessed rather than discarded. This increased the process time, and the loss by decay was appreciable. With the development of more accurate methods of analyses and better methods of operation, the purification of large quantities of fission-produced rare earths can be made more efficiently with the existing equipment, and the process can be expanded to purify multi-kilocurie amounts of Pm^{147} routinely.

Less discernible advantages of the ion exchange method for purification include the following: (1) the volumes of eluate containing the fission products were easily decreased by altering their pH's and adsorbing the fission products prior to re-eluting them with a new, smaller volume of eluant; and (2) products of corrosion and radiation were removed continuously.

The problem of radiation damage to the ion exchange resin and to the citrate elutriant was not studied quantitatively. Flow rate through each resin column was checked after each regeneration to detect any effect due to changes in the resin. The resin was replaced in column 3 after having been exposed to approximately 50,000 curies of Pm^{147} for the time (60 hr) of adsorption and elution. This represents approximately 1.29 kw hr of beta radiation. In order to reduce the possibility of contamination from previously absorbed activities, the resin in the final purification column (No. 4) was changed after each 20,000-curie run. Possible radiation damage was not considered to be the primary reason for replacing the resin.

The citrate eluate was damaged by radiation, as evidenced by a color change during the time of storage. No precipitates formed which interfered

with the recovery of Pm^{147} either by adsorption on resin or by precipitation methods.

The solution received for purification contained the fission-produced rare earths from which most of the Ce^{144} and Y^{91} had been removed. The Pm^{147} was purified to the extent that less than 2 μC of Eu^{154} was present in each curie of Pm^{147} and more than 425 curies of Pm^{147} per gram of rare-earth oxide.

A large portion of the separation was accomplished by adsorbing on and eluting from four ion exchange columns. The first column should be considered a feed-preparation column, as the rare earths were processed under nonideal conditions to prepare a citrate solution for a second column. From this first column, approximately 90% of the Pm^{147} was recovered; it contained a portion of the Eu, Y, Sm, and Nd. Such radioelements as Cs^{137} , Ru^{106} , and Ce^{144} were almost completely removed.

The second column was designed such that the peak of Pm^{147} desorbed could be readsorbed on the same column to prepare for the final-separation column a feed solution which contained less than 50 μC of Eu^{154} per curie of Pm^{147} and less than 3 g of rare earths per gram of Pm^{147} . The second column also received solutions from the final ion exchange separation which were not of product specification.

If separation factors based on the ratio of column volumes to the respective activity peaks (Figs. 2 and 3) are computed, the separation factors for Pm^{147} from Eu^{154} are 1.29 and 1.27. These compare favorably with the theoretical separation factor of 1.45 which is based on the ratio of distribution factors as determined by equilibration studies.¹²

Table 3 shows the material balance for the process.

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¹²F. C. Nachod and J. Schubert (eds.), *Ion Exchange Technology*, p 420, Academic Press, New York, 1956.

Table 3. Material Balance for Purification of Pm¹⁴⁷ by Ion Exchange

	Curies	Per Cent
Pm ¹⁴⁷ received	35,401	
Pm ¹⁴⁷ decayed	1,770	
Pm ¹⁴⁷ processed (amount received minus amount decayed)	33,631	100
Losses, known	1,804	5.4
Losses, unknown	993	3.0
Sent back for reprocessing	5,504	16.4
Products		
Purity of less than 2 μ c Eu ¹⁵⁴ per curie of Pm ¹⁴⁷ and more than 425 curies of Pm ¹⁴⁷ per gram of rare- earth oxides	18,200	54.1
Purity of less than 40 μ c of Eu ¹⁵⁴ per curie of Pm ¹⁴⁷ and more than 425 curies of Pm ¹⁴⁷ per gram of rare- earth oxides	1,750	5.2
With Sm ¹⁵¹	750	2.2
With neodymium	200	0.6
Partially separated	4,400	13.1

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