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SEPARATION OF AMERICIUM AND PROMETHIUM

R. S. Pressly

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SEPARATION OF AMERICIUM AND PROMETHIUM

R. S Pressly

SUMMARY

Promethium-147 is separated with the long-lived rare-earth fraction in the process of obtaining cesium-137 from radioactive waste material. Because of the fission yield of 2.6% and the half life of 2.6 years, there is contained in two-year decayed solution approximately equal activities of promethium-147 and cesium-137.

Ion exchange methods of separating the individual rare earths separate promethium and americium together. These two activities are difficult to separate from each other. Ion exchange methods separate small quantities effectively. With larger quantities, the precipitation method described in this report has been used and has proved to be more efficient than ion exchange. By this method promethium and americium are coprecipitated as fluorides. This fluoride precipitate is dissolved in boric acid and HNO_3 solution. The resultant solution is made 3.0 *M* in hydrofluosilicic acid (H_2SiF_6), which, upon heating and slow digestion, decomposes into two volatile gases, HF and SiF_4. Promethium fluoride precipitates from the solution, and americium remains complexed in solution.

INTRODUCTION

Promethium, because of the energy of the beta particle (0.226 Mev), is useful for making phosphors and also for radiographic work where the characteristic x ray is produced by beta bombardment of such metals as tantalum and platinum with no bremsstrahlung greater than the energy of the beta.

Americium-241 must be separated from promethium-147, since the alpha particle destroys the phosphor and reduces its life. The gammas associated with the decay of americium will make an x-ray source less useful. For these two uses and other uses, it is necessary to have a source high in specific activity and purity.

Inactive neodymium and samarium are present in the rare-earth group. Because of the high cross section for neutron absorption of the samarium produced in fission, most of it is removed. There remains neodymium from which promethium must be separated.

Solutions received as enriched long-lived rare-earth solutions contain such rare-earth activities as cerium-144, europium-152, -154, -155, promethium-147, and samarium-151. Cerium-144 is removed by the classical method[1] of oxidizing Ce^{3+} to Ce^{4+} and precipitating as ceric iodate. The remaining trivalent rare-earth activities are now promethium-147, europium-152, -154, -155, and samarium-151. By a heated ion exchange process,[2] promethium is separated from these gross activities. It is now contaminated with americium-241, which separates in the ion exchange process with promethium.

Some of the ion exchange methods which have been used to separate promethium-147 from americium-241 are as follows:

1. Americium is removed from a cation column on which both americium and promethium had been adsorbed by eluting[3] with 12–13 *N* HCl solution.
2. From a cationic resin bed, promethium is complexed and removed with 2.0 *M* H_2SiF_6 solution.[4]

The method described in this report has been used to separate promethium from americium in solutions containing approximately 50–60 curies of promethium. With the availability of greater quantities (75,000–125,000 curies per year), this method of purification will be very useful.

PROCEDURE – SEPARATION OF PROMETHIUM-147 FROM AMERICIUM

1. To approximately 100 ml of 8.0 *N* HNO_3 solution of Pm^{147} (~0.5 curies/ml), H_2SiF_6 solution is added until the mixture is 3.0 to 3.3 *M* H_2SiF_6.
2. The solution is heated slowly to boiling and boiled slowly for approximately 20 min. The H_2SiF_6 decomposes to give hydrofluoric acid and silicon fluoride. Both are volatile and the excess of each

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[1] P. H. M.-P. Brinton and C. James, *J. Am. Chem. Soc.* 41, 1980 (1919).

[2] R. S. Pressly and A. F. Rupp, *Purification of Fission-Product Rare Earths by Ion Exchange*, ORNL-1313 (Jan. 26, 1953).

[3] S. G. Thompson *et al.*, *J. Am. Chem. Soc.* 76, 6229 (1954).

[4] A. F. Rupp, *Radioisotope Production and Process Development Annual Report for 1955*, ORNL-2064, p 7.

is removed. Americium is more strongly complexed by SiF_6 than is Pm^{147}. The Pm^{147} precipitates as PmF_3.

3. The granular precipitate settles rapidly and the supernatant liquid is centrifuged to remove the precipitate. The precipitate is washed with 50 ml of H_2O and the water rinse is added to the supernatant liquid.

4. Fifty milliliters of saturated boric acid solution is added to the centrifuge tube to slurry out the precipitate into the beakers containing the bulk of the precipitate. The slurry is agitated and heated to approximately 80°C.

5. Fifty milliliters of 16 *M* HNO_3 is added, and the precipitate is heated to boiling; the precipitate is dissolved and a small amount of SiO_2 is precipitated.

6. The SiO_2 is centrifuged out and the solution is saved. The volume is reduced to ~100 ml by boiling. The solution is now 8.0 *N* HNO_3 and H_3BO_3 (0.44 *M*).

7. Steps 1 through 6 are considered a cycle. This procedure is repeated through approximately eight cycles to decontaminate to the desired factor of ~200 alpha counts per millicurie.

8. To precipitate PmF_3 and AmF_3, HF is added to the H_2SiF_6 supernatant until the solution is 3.0 *N*. After three cycles approximately 15% of the americium is left with the Pm^{147} product and 85% of the americium is in the H_2SiF_6 supernatant waste.

Tables 1 and 2 and Fig. 1 show the data for this procedure.

DISCUSSION

The Chemistry Division of the University of Chicago purified americium by adding Ce(III) to a 3.0 *M* H_2SiF_6 solution containing americium and fission rare earths and thus precipitating cerium and other rare earths as fluorides or fluosilicates.[5]

[5] *Chemistry Division Summary Report for November 1945*, University of Chicago, CS-3359 (Dec. 15, 1945).

TABLE 1. SEPARATION OF PROMETHIUM AND AMERICIUM

Cycle	Sample	Americium (Total Alphas)	Alpha (%)	Total Promethium (mc)	Pm^{147} (%)	β/α Ratio	Remarks
	Pm-0	3.03×10^9	100	59,700	100	3.2×10^4	Starting solution
1	Pm-0-4	4.1×10^8	14	258	0.4	1.7×10^3	H_2SiF_6 supernatant
	Pm-0-5	2.8×10^9	92.0	Sample not good		5.0×10^4	Dissolved PmF_3
2	Pm-0-6	1.7×10^9	56.0	2,552.0	4.3	1.7×10^3	H_2SiF_6 supernatant
	Pm-0-7	2.1×10^9	69.0	1,581.0	2.6	4.2×10^3	Am recovered from Pm-0-4 and Pm-0-6
3	Pm-0-8	6.5×10^8	20.0	2,100.0	3.5	5.6×10^3	H_2SiF_6 supernatant
4	Pm-0-9	1.5×10^7	0.5	823.0	1.4	9.0×10^4	H_2SiF_6 supernatant
	Pm-0-10	4.8×10^8	16.1	Analysis not good			Dissolved PmF_3
5	Pm-0-11	1.6×10^8	5.2	1,799.0	3.0	2.0×10^4	H_2SiF_6 supernatant
6	Pm-0-12	1.3×10^8	4.3	5,161.0	8.6	6.0×10^4	H_2SiF_6 supernatant
7	Pm-0-13	4.6×10^7	0.2	4,111.0	6.8	1.5×10^5	H_2SiF_6 supernatant
	Pm-0-14	2.1×10^7	0.06	35,290.0	59.1	2.85×10^6	Dissolved PmF_3
8	Pm-0-15	1.5×10^7	0.05	3,800.0	6.4	4.3×10^5	H_2SiF_6 supernatant
	Pm-0-16	7.7×10^6	0.003	32,900.0	55.0	7.4×10^6	Dissolved PmF_3
	Pm-0-17	3.1×10^8	10.3	11,600.0	19.4	7.0×10^4	Recovery from all H_2SiF_6, cycles 3–8

TABLE 2. SUMMARY OF RUN Pm-0

Sample	Remarks	Pm^{147} (curies)	Pm^{147} (%)	Alpha Counts Per Millicurie
Pm-0	Starting solution	59.7	100	50,000
Pm $(NO_3)_3$-23	Product	22.52	37.7	383
Pm $(NO_3)_3$-24	Product	5.9	9.88	277
Pm-0-7	Recovered Am fraction	1.58	2.6	1.4×10^6
Pm-0-20	Second recovery of Pm from waste H_2SiF_6	0.772	1.3	
Pm-0-21	Discarded in H_2SiF_6 solution	3.788	6.3	
Pm-0-17	Recovered from H_2SiF_6 solution	11.6	19.4	
	Total	46.16	77	
	Loss unaccounted for in samples, SiO_2, etc.	13.5	22	

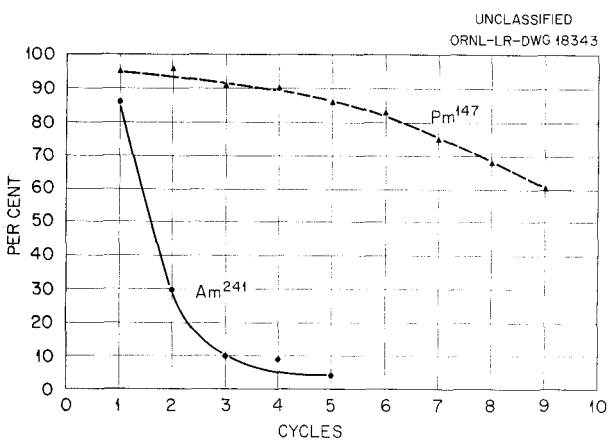

Fig. 1. Per Cent of Am^{241} and Pm^{147} Remaining in PmF_3 After Each Cycle.

Their problem was to obtain americium separated from fission rare earths and plutonium. The beta/alpha ratio of their final product was 100/1.

The problem discussed in this report was the obtainment of the product Pm^{147} separated from Am^{241}. The beta/alpha ratio of the starting solutions was 3.2×10^4 and the calculated purity of the final product was to be a beta/alpha ratio of approximately 1.2×10^7 based on 52% geometry of the alpha counter.

Upon first investigation, it was found that the distribution coefficient of Am^{241} was larger than that of Pm^{147}, as determined with Nalcite HCR (100–200 mesh) in 2.0 *M* H_2SiF_6. An ion exchange method was developed to separate Am^{241} and Pm^{147} in a time of three weeks of continuous operation. With the advent of larger amounts of Pm^{147} from the Cs^{137} purification process, the ion exchange process became less useful and the precipitation method described in this report was developed and used on a solution containing 50 to 60 curies of Pm^{147}.

Small quantities of Pm^{147} can be effectively separated from americium by adding Ce(III) to the promethium, carrying the Pm^{147} through the process, and then removing the cerium by oxidizing to Ce^{4+} and precipitating ceric iodate.

There are several fission chains which produce inactive neodymium. This neodymium should be separated from promethium quite completely by ion exchange before the H_2SiF_6 precipitation process.

Losses into the H_2SiF_6 waste after recovery precipitation of promethium as fluorides should not be greater than 6%. Other losses are caused by adsorption of the precipitate on equipment or into the silicon dioxide. These losses will be minimized by careful operation.

The separation factor of beta activity from alpha for each cycle is calculated to be approximately 3.7.

This method can be effectively used to separate large quantities of inactive rare earths from either americium or plutonium.