Preparation of ^{242}Cm from Neutron-irradiated ^{241}Am

Masanobu SAKANOUE and Mitsuo NAKATANI

Radiochemical Laboratory, Department of Chemistry,

Faculty of Science, Kanazawa University,

Kanazawa

Received June 14, 1971

The target of ^{241}Am irradiated with thermal neutron flux was subjected to the cation ion-exchange resin separation to separate ^{242}Cm , fission products and target ^{241}Am . The optimum conditions for ion-exchange chromatography was examined with cation exchange papers and by paper electrophoresis. The automatic recording device was applied to ascertain α emitters in the effluent. The radiochemically pure ^{242}Cm was obtained easily by this study.

1. Introduction

As shown in Fig. 1, ^{242}Cm can be prepared by thermal neutron irradiation of ^{241}Am . The fission cross-section of the simultaneously formed nuclides ^{242}Am and $^{242\text{m}}\text{Am}$ are as large as ~ 3000 and 6390 barn respectively, i.e., fission products are present after the irradiation. Hence, to obtain pure ^{242}Cm , chemical separation from these fission products and the target nuclide ^{241}Am is necessary. Because of the physical and chemical similarity of Am and Cm, they could be effectively separated by ion-exchange method only^{(1)~(7)}. In these experiments, α -hydroxyisobutyric acid (α -HIBA) was used as eluant. Optimum condition for the separation was found out with electrophoresis and ion-exchange paper. The elution curve of the α -emitters ^{241}Am and ^{242}Cm was automatically recorded with a surface barrier silicon semiconductor detector.

The details of the ^{242}Cm preparation are given below.

2. Apparatuses and Reagents

2.1 Measurement of the radioactivity

Gamma-rays was detected with a NaI (TI) well type scintillation counter, and the spectra were analyzed by a 200 channel pulse height analyzer, Model EDS-34208-1 made by Toshiba Electric Co.

Fig. 1 The production of ^{242}Cm by irradiation of ^{241}Am with thermal neutron.

The α -spectra were measured with a surface barrier silicon semiconductor, Model M-8813, connected with a pre-amplifier, Model RU-104, and a linear-amplifier, Model RU-108, by Toshiba Electric Co.

2.2 Apparatus for the chemical separation

The apparatus for the chemical separation is illustrated in Fig. 2. "A" indicates the reservoir for the eluant, "B" the ion-exchange column, "C" the glass-fiber, "D" the resin bed, "E" the condenser, "F" the device for

heating trichloroethylene and circulating the vapor to maintain the column temperature at 87°C .

2.3 Automatic recording of the elution curves of α -emitter

In the same figure (Fig. 2), "G"~"M" show an automatic recorder for elution curves of α -emitting nuclides. If the α -emitting effluent from the column flows on a mylar (G) (0.89 mg/cm^2), the α -rays pass through it and are recorded immediately by a surface barrier silicon semiconductor detector (H) connected with a preamplifier (I), linear-amplifier (K), linear-rate-meter (L) and recorder (M). The silicon detector was sealed in a box, because it was sensitive against light. Only α -emitters being recorded, this detector does not respond to β and γ rays.

A=eluant reservoir; B=column;
C=glass fiber; D=resin bed;
E=condenser; F=trichloroethylene;
G=mylar; H=silicon semiconductor detector; I=preamplifier; L=rate meter; M=recorder

Fig. 2 The apparatus for ion-exchange chromatography and automatic recorder for elution curves of α -emitter nuclide.

2.4 Preparation of the α -hydroxyisobutyric acid (α -HIBA) solutions

A weighed portion of extra pure α -HIBA (made by Wako Chemical Co.) was diluted to 1 M solution and stored in a polyethylene

bottle (Solution A). Solution B was obtained by passing ammonia gas through solution A so as to adjust the pH value to 7~8.

The solutions A and B were diluted to a concentration of 0.5M.

These solutions were mixed in order to be adjusted to the desired pH value. The α -HIBA solution was stored in form of a 1M solution because solutions of lower concentrations get spoiled soon.

2.5 Ion-exchange resin and column

Small amounts of the commercially available spherical cation exchange resin, Dowex 50 (100~200 mesh, H-form) were gained in an agate mortar and poured into a fine glass tube filled with water. The difference in the sedimentation rate was utilized for grading. A resin of an appropriate mesh number was filled into the column and washed three times with alternately 0.5M HCl, distilled water and a 0.5M NH_4Cl solution, and finally with ammonia solution of pH=3.3 so as to give the ammonium form.

The inner radius of the column was 5 mm and the resin was filled to a height of 45mm. The temperature of the column was kept at 87°C with the vapor of trichloroethylene, the flow rate being adjusted to 0.03 ml/min.

3. Experimental

3.1 Thermal neutron irradiation

A solution of $14\text{ }\mu\text{g}$ of ^{241}Am was filled into a quartz tube, and heated to dryness. After sealing, the sample was irradiated in the center of the Kyoto University Reactor for 90 hours.

The thermal neutron flux was $\sim 10^{13}\text{ n/cm}^2\cdot\text{sec}$.

3.2 Examination of the pH value of the eluant

The distribution coefficients and also the separation factors of trivalent lanthanide and actinide ions depend on the pH value of the organic acid used as eluant. Hence prior to the ion-exchange of Am and Cm, the electrophoretic mobilities of ^{147}Pm (carrier free) and ^{152}Eu (high specific activity) were deter-

mined at various pH values of the background electrolyte α -HIBA, the details of this method being reported in other paper⁸⁾. According to the result of this experiment, it was found that in the range between pH=3.0 and 5.0, the lower the pH is, the larger are the separation factors of trivalent lanthanides. This fact may hold true for ion-exchange experiments, for actinides too.

Furthermore, the difference in the distribution of Am in α -HIBA solutions of various pH was examined with a cation exchange paper. A cation exchange paper (Amberlite SA-2) ($1 \times 1 \text{ cm}^2$) was immersed in a 0.1 N HCl solution containing ^{241}Am , and stored for at least ten minutes. After taking it out, it was dried at room temperature and immersed in α -HIBA solutions of various pH values. The elution degree of ^{241}Am was examined in terms of the period of storage in the α -HIBA solutions, the pH, and the temperature of these solutions.

Fig. 3 shows that the distribution coefficient of ^{241}Am on the cation exchange resin varies little with different times and temperatures but largely with different pHs of the α -HIBA solutions. The elution degree for ^{241}Am from the cation exchange resin is very small in the low pH range (pH<3.3). It increased largely between pH=3.1 and 3.9, and above pH=4.1 Am is almost completely

Fig. 3 The elution of ^{241}Am from cation-exchange paper (Amberlite SA-2) by 0.5M α -hydroxyisobutyric acid.

eluted. This together with the paper electrophoretic results show that the optimum pH for the separation of lanthanides and actinides is pH=3.3~3.7.

3.3 Separation of ^{242}Cm from ^{241}Am and fission products

The irradiated ^{241}Am was allowed to stand for about 10 days until the short-living fission products had decayed. Then the sample was dissolved in 1 ml of 0.5M α -HIBA (pH=2.0), and loaded on the top of resin bed. The column had been kept at 87°C for a sufficiently long period and stirred to remove any air bubbles in the resin.

^{95}Zr - ^{95}Nb were eluted first with an α -HIBA solution of pH=3.3.

After ^{95}Zr - ^{95}Nb had been removed completely, ^{91}Sr was eluted with an α -HIBA solution of pH=3.5. Under these conditions Am and Cm moved slightly from the top of the resin bed. When ^{91}Sr had passed through, Am and Cm were eluted by an α -HIBA of pH=3.7.

The effluent was collected in polyethylene tubes, and each fraction was counted with a NaI (TI) scintillation counter.

The nuclides were identified by γ -ray spectrometry. From each fraction, 1~10 drops were put on a glass plate, heated to dryness, and the α -spectra were measured with a surface barrier silicon detector. The activity ratio of each fraction containing both Am and Cm was determined, giving detailed information about the separation. For fractions with small activity, a condensing method was applied as follows. A cation exchange paper was immersed into the fractions after the pHs had been adjusted to about 2. After taking the paper out, it was washed with 0.1 N HCl and the nuclides were eluted from it with as small an amount of 8 N HCl as possible. The effluent was evaporated to dryness on a glass plate, and the α -spectra recorded.

Fig. 4 shows the elution curve. In the fraction containing ^{241}Am and ^{242}Cm , fission products were not detected. For the fraction containing both ^{241}Am and ^{242}Cm , the separa-

Fig. 4 The elution curve for irradiated ^{241}Am with 0.5M α -hydroxyisobutyric acid (pH=3.5 and 3.7) at 87°C.

Fig. 5 Elution curve for ^{241}Am and ^{242}Cm with 0.5M α -hydroxyisobutyric acid (pH=3.5) at 87°C.

tion was again carried out in the same way with an α -HIBA of pH=3.5 as eluant. The result is shown in Fig. 5. Fig. 6(a) shows the elution curve of ^{242}Cm in this separation recorded with automatic recording device of elution curve mentioned above.

4. Conclusion

Separation conditions for ion-exchange chromatography can be examined easily by paper electrophoresis and with a cation exchange paper.

The automatic recorder for elution curves can only be used for α -emitter with high-

Fig. 6 (a) The elution curve of ^{242}Cm recorded by automatic recording device with surface barrier silicon semiconductor detector.

(b) The elution curve by γ countings.

activity. For α -emitter with low-activity, it is not sensitive enough because only small part of the α -particles are recorded, a great part is lost due to unavoidable absorbance, i. e. water, mylar, air, etc.

Fig. 7 and Fig. 8 show the α -spectra of neutron irradiated ^{241}Am and the separated ^{242}Cm , respectively. The separation of ^{241}Am and ^{242}Cm was complete, and the γ -spectra show that even a trace amount of fission

Fig. 7 The α -spectrum for neutron irradiated ^{241}Am . Fig. 8 The α -spectrum for prepared ^{242}Cm .

products was not present.

References

- 1) W. Cohn, et al.: Metallurgical Project, CC-2827 (June 1945)
- 2) D.H. Harris, and E.R. Tompkins: *J. Am. Chem. Soc.*, **69**, 2777 (1947)
- 3) F.H. Spedding, et al.: *J. Am. Chem. Soc.*, **69**, 2800 (1947)
- 4) K. Street, Jr., S.G. Thompson, and G. T. Seaberg: *J. Am. Chem. Soc.*, **72**, 4832 (1950)
- 5) G.R. Choppin, B.G. Harvey, S.G. Thompson, and G.T. Seaberg: *J. Am. Chem. Soc.*, **76**, 6229 (1954)
- 6) G.R. Choppin, B.G. Harvey, and S.G. Thompson: *J. Inorg. Nucl. Chem.*, **2**, 66 (1956)
- 7) G. Höhle, H.J. Born, and W. Weinländer: *Radiochim. Acta*, **10**, 85 (1968)
- 8) M. Sakanoue and M. Nakatani: *Bull. Chem. Soc. Japan*, to be submitted.

要 旨

^{242}Cm の中性子照射 ^{241}Am よりの調製

阪上正信, 中谷光雄
金沢大学理学部

中性子照射をうけた ^{241}Am ターゲットから陽イオン交換樹脂による分離法を適用して, ^{242}Cm , 核分裂生成物を分離した。

陽イオン交換分離の溶離最適条件を陽イオン交換紙およびろ紙電気泳動法で検討する一方, 溶離 α 放射体の自動記録のための装置をくふうして用いた。これらにより放射化学的純度のよい ^{242}Cm を得た。