Production of Curium by the Neutron Irradiation of Americium-241

著者	SUZUKI Shin, SATO Akiko, HARA Mitsuo, MITSUGASHIRA Toshiaki, KAWASUJI Isamu, KIKUCHI Hideo, FUKASAWA Tetsuo
journal or publication title	Science reports of the Research Institutes, Tohoku University. Ser. A, Physics, chemistry and metallurgy
volume	28
number	1
page range	73-79
year	1979-12-05
URL	http://hdl.handle.net/10097/28088

Production of Curium by the Neutron Irradiation of Americium-241*

Shin SUZUKI, Akiko SATÔ, Mitsuo HARA, Toshiaki MITSUGASHIRA,
Isamu KAWASUJI, Hideo KIKUCHI** and Tetsuo FUKASAWA

The Research Institute for Iron, Steel and Other Metals

(Received August 30, 1979)

Synopsis

^{241}Am was irradiated by the Japan Material Testing Reactor. The group separation of transuranium elements from fission products and cladding materials were carried out, and then Np, Pu, Am and Cm were isolated by using the ion exchange method. The isotopic ratios of Cm and Am were determined by the α- and γ-ray spectrometry.

I. Introduction

We have a plan to irradiate ^{241}Am and ^{243}Am in the Japan Material Testing Reactor (JMTR) and to investigate the chemical properties of Cm, Bk and Cf which are obtained from the irradiated Am target. This work is one of the preliminary works to establish the separation method of Cm from a heavily irradiated Am target and to obtain the knowledge on the future production of Bk and Cf.

When ^{241}Am was irradiated in JMTR for more several hundreds days, even a few milligrams of the irradiated sample would have the radioactivity of Ci order after the suitable cooling. The removal of most fission products and cladding materials must be performed by the remote operation, therefore this separation procedure needs to be simple and easy.

Moreover, considering the group separation of transuranium elements from the fission products such as rare earth elements and the mutual separation of transuranium elements, the ion exchange method was taken for the chemical separation, because the technique has been relatively established in detail.

The isotopic ratios of Cm and Am were determined by the α- and γ-ray spectrometry.

II. Experimental

1. Reagents and irradiated ^{241}Am.

All the chemical reagents used in this work were of analytical grade. LiCl was produced from Li_2CO_3 and HCl, and recrystallized twice. The cation exchange resin Dowex 50W×12 (100–200 mesh) and the anion exchange resin Dowex 1×8 (200–400 mesh) were used.

---

* The **1701th** report of the Research Institute for Iron, Steel and Other Metals.

** Present address: The Miyagi Prefecture Office, Sendai.

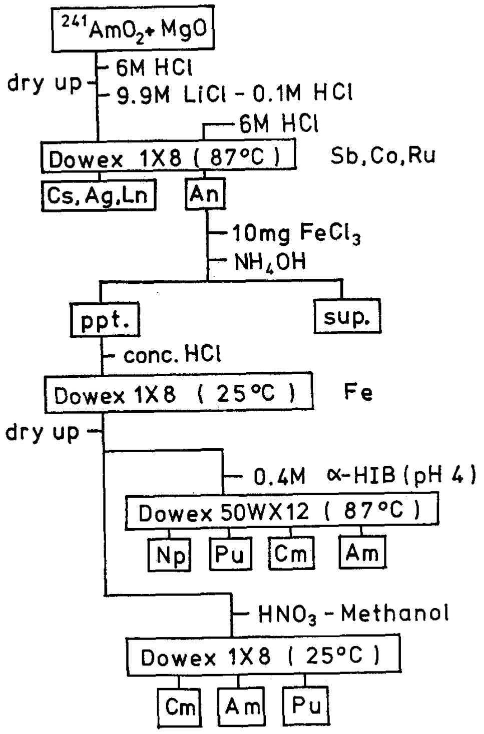

Fig. 1. Separation scheme.

$^{241}AmO_2$ was irradiated in the reflection area of JMTR, intermittently for 384 days from July 9, 1971 to July 22, 1972$^{(1)}$. The authors got a part of it from Japan Atomic Energy Research Institute and used in this work.

2. Chemical separation.

Fig. 1 shows the flow sheet of the isolation procedure of Cm. Ln represents the lanthanide elements, namely Eu, Ce and so on, and An the actinide elements, Np, Pu, Am and Cm. The separation of fission products from transuranium elements were done by the remote control of ion exchange method in the 6th hot cell of the hot laboratory of the Ôarai Laboratory for Irradiation Experiment of the Research Institute for Iron, Steel and Other Metals.

After the dissolution of the irradiated sample, transuranium elements were separated from most of fission products and cladding materials by the anion exchange procedure with 9.9M LiCl–0.1M HCl solution and then eluted with 6M HCl solution at 87°C. The size of the column was 80 mm in length, 8 mm in diameter and the flow rate was 0.25–0.35 ml/cm^2/min.

The transuranium elements in the HCl solution containing a small amount of LiCl were precipitated with $Fe(OH)_3$, the precipitates were dissolved with conc.HCl solution and the transuranium elements and Fe were separated by means of the anion exchange method. Sequently, transuranium elements were separated mutually by the following two methods.

---

(1) K. Ueno, K. Watanabe, C. Sagawa and T. Ishimori, J. Nucl. Sci. Technol., **12** (1975), 356.

Method 1; The cation exchange separation with α-hydroxyisobutyrate (α-HIB) was performed. The HCl solution of transurnaium elements was dried up and several drops of 0.4M α-HIB (pH 4) were added. This solution was placed and adsorbed on the top of the resin which was maintained at 87°C by the steam of trichloroethylene and eluted slowly. Suitable aliquots of the effluent were taken into the polyethylene tubes. Each fraction was analysed radiochemically and the purity of Cm and Am was determined. The size of the column of the cation exchange resin was 117 mm in length, 4 mm in diameter and the flow rate was 0.3–0.6 ml/cm^2/min.

Method 2; The anion exchange separation with HNO_3-methanol solution was performed. The column size of the anion exchange resin was 70 mm in length, 4 mm in diameter and the flow rate was 0.25–0.35 ml/cm^2/min. One milliliter of 1.0M HNO_3 –90% methanol solution of transuranium elements, in which ^{152}Eu and ^{137}Cs were added designedly, was adsorbed on the resin and eluted with the solution of 0.5M HNO_3-methanol. The concentration of methanol was decreased from 90% to 0% in sequence according to the elution of elements.

3. Determination of the isotopic ratios of Cm and Am.

After the chemical separation, isotopic ratios of Cm and Am were determined by analysing their α- and γ-ray spectra obtained by the use of Si- and Ge(Li)-detector, respectively. ^{242m}Am was quantified by analysing the growth curve of ^{238}Np which was the α-decay product of ^{242m}Am. The measured value of isotopic ratios was compared with the value calculated along with the irradiation history of JMTR in order to consider the irradiation condition of ^{241}Am.

III. Results and discussion

1. Isolation of Cm.

The α- and γ-ray spectra of the irradiated sample showed that it contained the nuclides of ^{238}Pu, $^{241-243}$Am and $^{242-244}$Cm as the transuranium elements, ^{134,137}Cs, ^{144}Ce, ^{106}Ru and ^{125}Sb as the fission products, and ^{110m}Ag and ^{60}Co as the cladding materials. By the anion exchange procedure with LiCl solution, Cs, Ce, Sb, Co and most of Ag and Ru were separated from transuranium elements. Ag and Ru were also removed by the cation and anion exchange methods with HCl solution. In the case of cation exchange procedure with α-HIB, the distribution ratios of transuranium elements depend greatly on the pH value of α-HIB. The most appropriate pH value was found to be 4.0. Figure 2 shows the elution curve of transuranium elements, Np, Pu, Am and Cm, with 0.4M α-HIB of pH 3.98. Although Cm cannot be perfectly separated from Am, the separation factor of Cm/Am was found to be about 1.4 and similar value has been reported by others(2).

The result of the anion exchange separation with HNO_3-methanol solution is shown in Fig. 3. Two large peaks are attributed to Cm and Am. Cs was not adsorbed and eluted at the first peak, Eu eluted before Cm, and, Ce and Pu after Am.

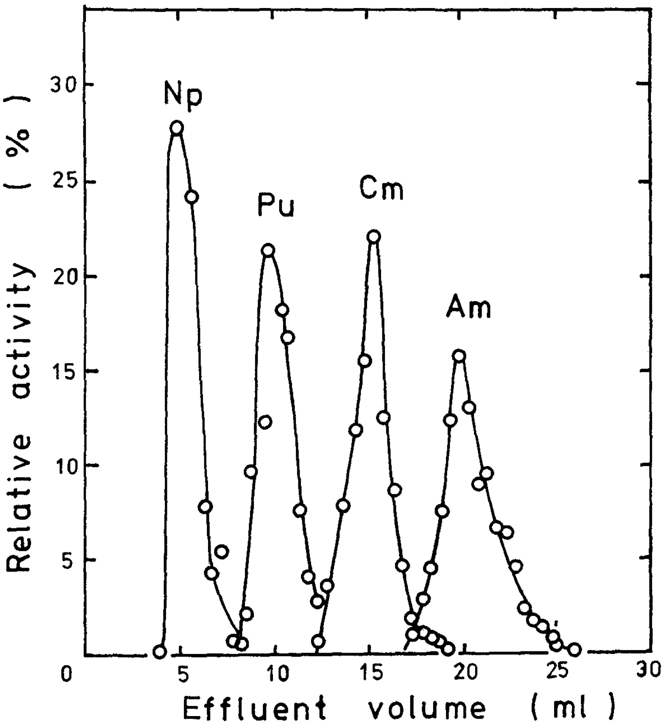

Fig. 2. Elution curves for Np, Pu, Am and Cm.
Eluent: 0.4M α-HIB pH 3.98. Column: 117 mml×4 mmφ. Resin: Dowex 50W×12 (100–200 mesh). Flow rate: 0.3–0.6 ml/cm²/min (87°C).

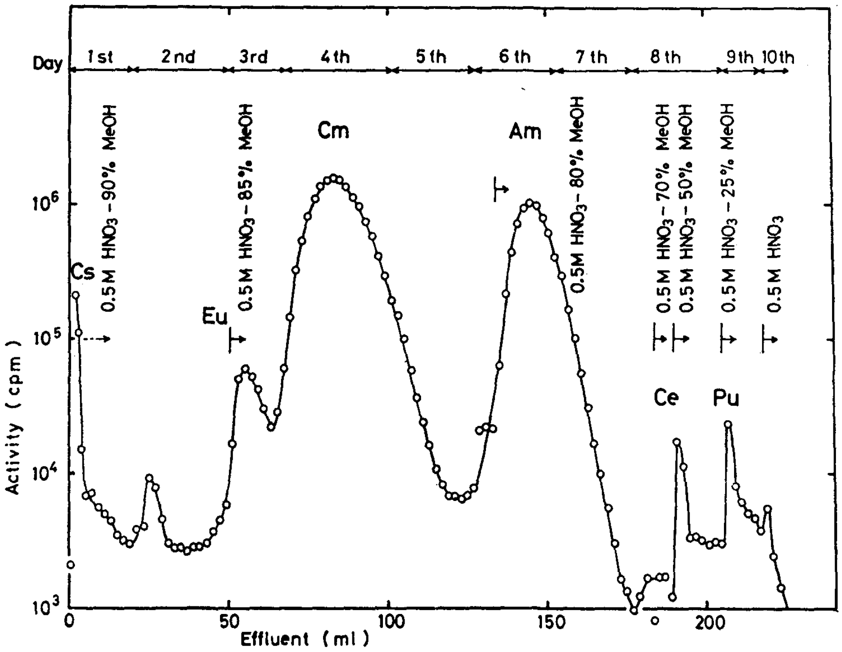

Fig. 3. Elution curve of HNO_3-methanol system.
Column: 70 mml×4 mmφ. Resin: Dowex 1×8 (200–400 mesh). Flow rate: 0.25–0.35 ml/cm²/min (25°C).

^{239}Np ($T_{1/2}$=2.35 d) which is the daughter of ^{243}Am eluted while ^{243}Am remained in the resin and its behavior was not clearly known.

In comparison with the cation exchange method with α-HIB, the anion exchange method with HNO_3-methanol is superior to the cation exchange method because of the large separation factor of Cm from Am, which is reported to be about 3 in Ref. 3. The anion exchange method has also the merits because the separation can be carried out at room temperature and the salt-free sample for the α-ray measurement can be prepared easily. But on the other hand, the latter is inferior to the former with respect to the small flow rate of eluent, and then it takes quite a long time for the separation. The α-ray spectra of purified Am and Cm are shown in Figs. 4 and 5, respectively.

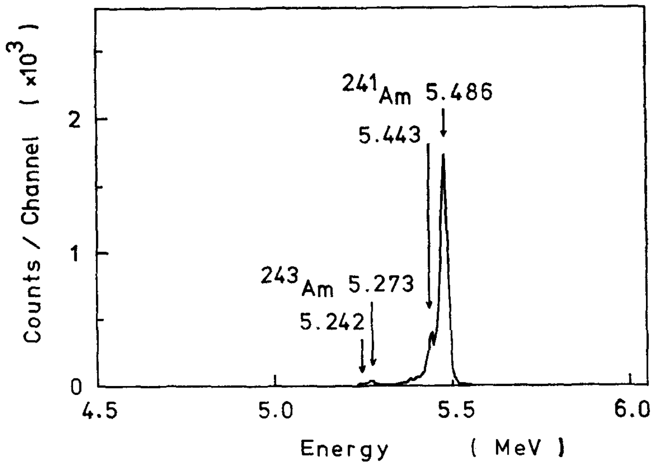

Fig. 4. α-ray spectrum of Am fraction.

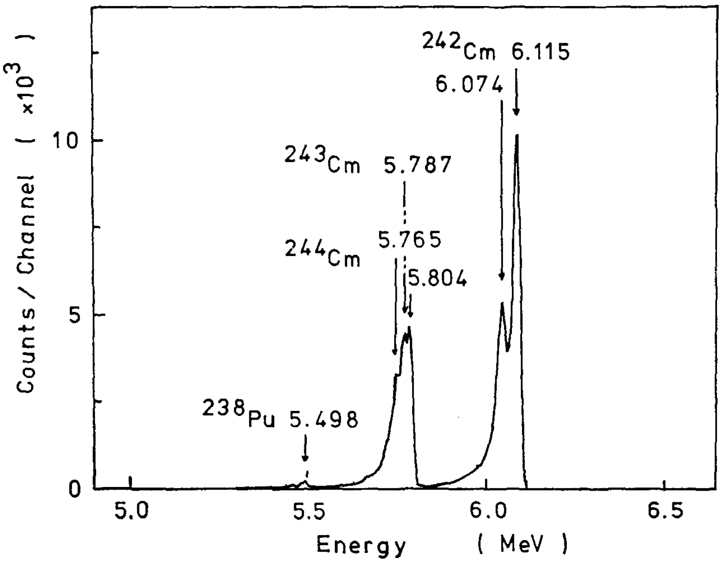

Fig. 5. α-ray spectrum of Cm fraction.

---

(2) G.R. Choppin, B.G. Harvey and S.G. Thompson, J. Inorg. Nucl. Chem., **2** (1956), 66.

(3) L.I. Guseva, I.A. Lebedev, B.F. Myasoedov and G.S. Tikhomirova, Sov. Radiochem., **17** (1975), 324.

2. Determination of the isotopic ratios of Cm and Am(4).

The isotopic ratios of Cm and Am were determined by the α- and γ-ray spectrometry and the results are shown in Table 1. The yields of the nuclides of Cm and Am at the time of mutual separation were computed by a computer program(5). For the calculation the thermal neutron flux (ϕ) and the fraction of the total density in the epithermal distribution (f) were chosen as adjustable variables. The values of $\phi=(2.15\pm0.20)\times10^{14}$n/cm²/sec and f=0.084±0.026 were found to represent the isotopic ratios measured within the error of 2σ as is shown in Fig. 6.

Table 1. Isotopic ratios of Cm and Am (in June 1976)

Nuclide	Isotopic ratio	
	Measured	Calculated*
Am-241	1	1
242m	0.0116±0.0017	0.0112
243	0.245 ±0.026	0.240
Cm-242	1	1
243	16.7 ±2.5	16.5
244	17.6 ±2.7	19.65

*: $\phi=2.1\times10^{14}$ n/cm²/sec and f=0.09.

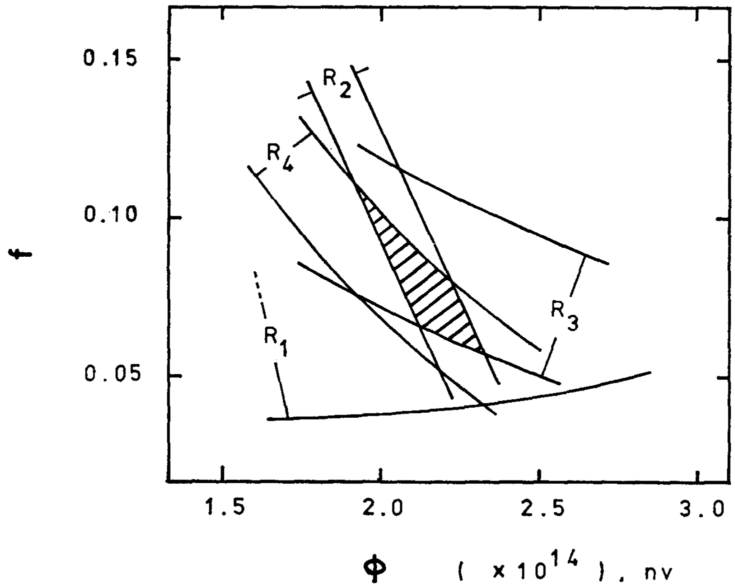

Fig. 6. Region of ϕ and f in JMTR which satisfies the measured isotopic ratios of Cm and Am within the error of 2σ.
R_1: Am-242m/Am-241. R_2: Am-243/Am-241. R_3: Cm-243/Cm-242.
R_4: Cm-244/Cm-242.

---

(4) I. Kawasuji, T. Fukasawa and S. Suzuki, Radiochem. Radioanal. Lett., **40** (1979), 215.

(5) M. Hara, T. Mitsugashira, A. Satô and S. Suzuki, Sci. Rep. RITU, **A28** (1979), 41.

The isotopic ratios which were calculated using the values of $\phi=2.1\times10^{14}$n/cm^2/sec and f=0.09 are shown in Table 1.

The effective cross-sections of Cm and Am in JMTR were calculated by the method of C.H. Westcott(6). The values calculated at f=0.09 are shown in Table 2. These values are useful for the planning of the production of transcurium elements by JMTR.

Table 2. Effective cross-sections

Nuclide	Reaction	Corss-section, barns		
		σ_{eff}	σ_0	Σ'
Am-241	(n, γ)	900 ±100	670	2100
	(n, γ)*	100 ± 20	70	300
	(n, f)	5.43± 1.01	3.15	21
242m	(n, γ)	2260 ±120	2000	2400
	(n, f)	7590 ±530	6400	10950
243	(n, γ)	315 ±109	78	2250
	(n, f)	0.07	0.07	0
Cm-242	(n, γ)	36 ± 7	20	150
	(n, f)	5	5	0
243	(n, γ)	225	225	0
	(n, f)	600	600	0
244	(n, γ)	80 ± 29	14	608
	(n, f)	1.2	1.2	0

*: ^{241}Am(n, γ) ^{242m}Am. σ_{eff}: Effective cross-section.
σ_0: Thermal neutron cross-section.
Σ': Resonance integral.

Acknowledgement

We wish to thank Dr. K. Ueno of Japan Atomic Energy Research Institute for providing the irradiated ^{241}Am. And we are thankful to the members of the Ôarai Laboratory for Irradiation Experiment of the Research Institute for Iron, Steel and Other Metals for the helpful support to our experiment.

---

(6) C.H. Westcott, CRRP-960 (1960).