Radiochemical Determination of Neutron Capture Cross Sections of $^{241}$Am

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(Received October 21, 1996)

The thermal neutron capture cross sections and the neutron capture resonance integrals of $^{241}$Am leading to the production of the isomer $^{242m}$Am and the ground-state $^{242g}$Am were measured radiochemically by the Cd-ratio technique with neutron flux monitors of Co/Al and Au/Al alloy. Highly-purified $^{241}$Am targets were irradiated in an aluminum capsule by using JMTR. The neutron fluxes and their epithermal neutron fractions were determined by measuring γ-rays of $^{60}$Co and $^{198}$Au. The yields of $^{242m}$Am and $^{242g}$Am were decided by analyzing growth and decay curves of the α-ray activity ratios $^{242}$Cm/$^{241}$Am. The resultant thermal neutron capture cross sections are $85.7 \pm 6.3$ b and $768 \pm 58$ b for $^{242m}$Am and $^{242g}$Am, and the resonance integrals $114 \pm 7$ b and $1,694 \pm 146$ b, respectively. The differences between the present results and the evaluated values by Mughabghab are $38$–$59\%$. The isomeric ratios, $g/(m+g)$, of $0.90 \pm 0.09$ for thermal neutrons and $0.94 \pm 0.11$ for epithermal neutrons are, however, almost consistent with evaluated values.

KEYWORDS: neutron capture cross sections, americium 241, thermal neutrons, resonance integrals, epithermal neutrons, resonance integral, americium 242m, americium 242g, curium 242, isomeric ratio, chemical separation, alpha spectrometry, evaluations

I. INTRODUCTION

Minor actinides are produced from successive neutron capture reactions starting from $^{238}$U in nuclear fuel and accumulated together with fission products in a high burnup reactor. The minor actinides cause severe problems in waste management. One of the important reactions in the buildup of the minor actinides is the neutron capture of $^{241}$Am. Both isomer-state $^{242m}$Am (48.63 keV) and ground-state $^{242g}$Am are produced by the $^{241}$Am$(n,\gamma)$ reaction. Isomeric $^{242m}$Am is very significant because the isomer has a very large capture cross section at thermal energies$^1$ and consequently can be transmuted into higher americium isotopes. The decay heat from the relatively short-lived isotope of $^{242}$Cm, which is the daughter isotope of $^{242}$Am, gives a consequential problem in spent-fuel handling. However, there is still discrepancy among the data on the absorption cross sections of $^{241}$Am leading to the production of $^{242m}$Am and $^{242g}$Am$^2$. The accurate data with accuracy of 5% on the capture cross section of $^{241}$Am have been required as registered in the World Request List for Nuclear Data WRENDA 91/92$^3$.

In this study, highly-purified $^{241}$Am targets were irradiated in Japan Material Testing Reactor (JMTR). The thermal neutron capture cross sections, the resonance integrals of $^{241}$Am and the isomeric ratios have been radiochemically measured. The obtained data are compared with evaluated and previously measured values.

II. EXPERIMENTAL

1. Principle for Determination of the Capture Cross Sections and the Resonance Integrals

For the purpose of measuring the capture cross section for 2,200 m/s neutrons and the resonance integral, this experiment was carried out by the Cd-ratio method$^4$: a Cd-covered target and another without Cd-cover are irradiated together with neutron flux monitors. In the convention by Westcott et al.$^5$, the effective cross section for Maxwellian neutrons, $\bar{\sigma}$, is defined by

$$R = n_{v0} \bar{\sigma},$$

where $R$ is the reaction rate per atom in the case of the irradiation without Cd-cover, $n$ the neutron density including both thermal and epithermal neutrons, and $v_0=2,200$ m/s. The quantity $n_{v0}$ plays the role of the “flux”. The cross section $\bar{\sigma}$ is also given by

$$\bar{\sigma} = \sigma_0 \left[ g G_{th} + r \sqrt{\frac{T}{T_0}} s_0 G_{epi} \right],$$

where $\sigma_0$ is the reaction cross section for 2,200 m/s neutrons, and $g$ the measure of the cross section deviation from the 1/$u$ law in the thermal energy region. The $r(T/T_0)^{1/2}$ gives the fraction of epithermal neutrons in the neutron spectrum, and $s_0$ is defined by

$$s_0 = \frac{2}{\sqrt{\pi}} \frac{I_0'}{\sigma_0},$$

where $I_0'$ is the resonance integral after subtracting the 1/$u$ component. The $G_{th}$ and $G_{epi}$ denote self-shielding coefficients for thermal and epithermal neutrons, respec-
tively. The values of \( g \) and \( G_{th} \) are almost unity in this experiment\(^6\). From Eqs. (1) and (2), the next equation is deduced,

\[
\frac{R}{\sigma_0} = n\nu_0 + n\nu_0 T \sqrt{\frac{T}{I_0}} s_0 G_{epi} = \phi_1 + \phi_2 s_0 G_{epi}. \tag{4}
\]

In the case of the irradiation with Cd-cover, similar equations are derived as follows:

\[
\frac{R'}{\sigma_0} = \phi_1' + \phi_2' s_0 G_{epi}. \tag{5}
\]

From a Maxwellian distribution of the neutron field in JMTR\(^7\), the epithermal neutron distribution can be cut off at 0.5 eV (a Cd cut-off energy) as assumed in Refs. (5) and (8).

The values of \( \phi_1, \phi_2, \phi_1', \) and \( \phi_2' \) can be obtained from the irradiation of flux monitors (two sets of Co and Au wires in this study) put near the target.

From Eqs. (5) and (6),

\[
s_0 G_{epi} = \frac{\phi_1 - \phi_1'(R/R')}{\phi_2 - \phi_2'(R/R')} \tag{7}
\]

and then

\[
\sigma_0 = \frac{R}{\phi_1 + \phi_2 s_0 G_{epi}} \tag{8}
\]

are obtained, where the quantity \( \sigma_0 \) is the value to be measured first in this study.

Because the thicknesses of the \( ^{241}\text{Am} \) targets prepared in this work are thin enough (1.5 and 3.8 mg/cm\(^2\)) to reasonably assume the \( G_{epi} \) to be unity, the quantity \( I_0 \) can be obtained using Eq. (3). The \( 1/\nu \) contribution to the resonance integral for a Cd cut-off energy of 0.5 eV is given by 0.45\( \sigma_0 \), so

\[
I_0 = I_0' + 0.45\sigma_0, \tag{9}
\]

where \( I_0 \) is the resonance integral to be also determined\(^8\).

2. Radiochemical Procedures

Before target preparation, the americium samples were purified chromatographically from other actinides (U, Np, Pu, and Cm), which disturb the cross section measurement of \( ^{241}\text{Am} \), by an anion exchange method\(^9\).

Figure 1 shows the scheme of the procedure for the ion-exchange separation. The sample was dissolved in a mixture of 0.1 ml of conc. nitric acid and 1 ml of ethyl alcohol. Americium was isolated by eluting from a 4 mm diam. x 40 mm column of MCI GEL CA06Y anion exchange resin with a mixture of 0.5 M nitric acid and 80% of methyl alcohol at room temperature. The ion-exchange separation procedure was carried out twice in order to perfectly isolate the Am from Cm. Chemical recovery of the americium after the ion exchange was more than 99.9%. The purity of the Am was checked by \( \alpha \)- and \( \gamma \)-ray measurements; no \( \alpha \)-rays from \( ^{243}\text{Am} \) and other actinides and no \( \gamma \)-ray from \( ^{239}\text{Np} \) (daugh-

![Fig. 1 Procedure for the chemical separation of Am by an anion-exchange method](image)

The \( ^{241}\text{Am} \) was electrodeposited\(^{10}\) in an active area of 5 mm in diameter on aluminum disks of 12 mm in diam. and 0.5 mm in thickness. The amounts of \( ^{241}\text{Am} \) deposited were determined by a \( \alpha \)-ray spectrometry; the atom number of the target without Cd-cover was \((7.166 \pm 0.018) \times 10^{14} \) (36 kBq) and that with Cd-cover \((1.847 \pm 0.004) \times 10^{15} \) (94 kBq). Weighed 0.504%-Co/Al and 0.061%-Au/Al alloy wires were used for monitoring the neutron fluxes at the target positions.

Gamma-ray activities from \( ^{60}\text{Co} \) and \( ^{198}\text{Au} \) of neutron flux monitors were measured with a coaxial HPGe detector and a multi-channel analyzer. The detection efficiency was calibrated with a mixed standard source (Laboratoire de Mérologie des Rayonnements Ionisants). Absolute activities of the monitors were determined by measuring the \( \gamma \) rays of 1,173 and 1,333 keV for \( ^{60}\text{Co} \) and 412, 676, and 1,088 keV for \( ^{198}\text{Au} \).

Alpha-rays emitted from the \( ^{241}\text{Am} \) targets before and after the irradiation were measured with a silicon surface barrier detector and a multi-channel analyzer. Figure 2 gives \( \alpha \)-ray spectra of the target measured in this study. The \( \alpha \)-ray activity ratios of \( ^{242}\text{Cm} / ^{241}\text{Am} \) in the irradiated targets were computed from the spectra by correcting for the background counting and the tailing of the peaks by a GP method\(^{11}\), which is based on the geometric progression for the far tail of the spectrum.

In order to measure the activity of \( ^{242}\text{Cm} \) growing from the \( ^{242m}\text{Am} \) (for the determination of the amount of \( ^{242m}\text{Am} \), mentioned later in detail), the same chemical procedures of ion exchange and electrodeposition were...
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3. Irradiation

Two sets of the $^{241}\text{Am}$ target with the Co- and Au-flux monitor wires were irradiated in JMTR for 4 days by using a hydraulic type capsule shown in Fig. 3. One set was covered with an Al container lined with Cd and another with an unlined container, in order to measure the cross sections of thermal neutron and the resonance integrals. The JMTR is a high flux reactor with a rated power of 50 MW. The maximum neutron flux is about $4 \times 10^{14} n/cm^2 \cdot s$, and the typical thermal and fast neutron flux distributions and the detailed neutron spectra in the core were given in the literature\(^{7}\).

4. Analysis of the Decay and Growth Curves of the Activity Ratio $^{242}\text{Cm}/^{241}\text{Am}$

After the neutron capture reaction of $^{241}\text{Am}$, the $^{242}\text{Cm}$ nuclide is formed through the decays of the produced nuclides, i.e. $^{242\nu}\text{Am}$ and $^{242}\text{Am}$. In this study, the growth and decay curves on the $\alpha$-ray activity ratios of $^{242}\text{Cm}/^{241}\text{Am}$ have been precisely observed by the $\alpha$-spectrometric method. By analyzing the curves in detail, the atom number ratios of $^{242}\text{Cm}/^{241}\text{Am}$ and $^{242\nu}\text{Am}/^{241}\text{Am}$ can be computed as follows:

1. Determination of the Atom Number Ratios of $^{242}\text{Cm}/^{241}\text{Am}$ from the Decay Curves

We define the numbers of atoms as $N_0$, $N_1$, $N_2$, $N_3$ and $N_4$ for $^{241}\text{Am}$, $^{242\nu}\text{Am}$, $^{242}\text{Cm}$, $^{242}\text{Cm}$ and $^{238}\text{Pu}$, respectively. We assume that only the $^{241}\text{Am}$ target and the decay product $^{242}\text{Cm}$ from the $^{242}\text{Am}$ which had been directly produced by the reaction existed at EOI, because the contribution of the $^{242}\text{Cm}$ supplied from $^{242\nu}\text{Am}$ can be neglected as mentioned later. The amount of the $^{238}\text{Pu}$ accumulated from the $\alpha$-decay of $^{242}\text{Cm}$ during the four-day irradiation is negligibly small (the activity of $^{238}\text{Pu}$ was estimated to be <0.01% of that of $^{242}\text{Cm}$). Therefore, the activity ratio, $A_{mes}$, of
$^{242}\text{Cm}/(^{241}\text{Am}+^{238}\text{Pu})$ measured in this study, where $\alpha$-rays with almost the same energy of 5.5 MeV are released both from the $^{241}\text{Am}$ target and from the decay product $^{238}\text{Pu}$ of $^{242}\text{Cm}$, is expressed by

$$A_{meas} = \frac{dN_3}{dt} = \frac{dN_0}{dt} + \frac{dN_4}{dt}$$

$$= \frac{\lambda_3 N_3}{\lambda_0 N_0 + \lambda_4 N_4}$$

$$= \frac{\lambda_3 N_3^0 e^{-\lambda_3 t}}{\left[ \frac{N_0^0}{N_0} e^{-\lambda_0 t} + \frac{1}{\lambda_4 - \lambda_3} e^{-\lambda_4 t} \right]}$$

$$+ \frac{\lambda_4 N_4^0 e^{-\lambda_4 t}}{\left[ \frac{N_0^0}{N_0} e^{-\lambda_0 t} + \frac{1}{\lambda_3 - \lambda_4} e^{-\lambda_3 t} \right]},$$

where $N^0_0$ means the number of atoms at EOI. By least squares fitting of Eq. (13) to the decay curves measured in this study (see Fig. 5), the atom number ratios of $^{242}\text{Cm}/^{241}\text{Am}$ at EOI, $N^0_0/N^0_0$, can be determined. Since the $^{242m}\text{Am}$ that was directly produced by the $^{241}\text{Am}(n, \gamma)^{242m}\text{Am}$ reaction existed at EOI and decayed completely to $^{242}\text{Cm}$ at the activity measurement, the value of $N^0_0/N^0_0$ to be obtained in the decay analysis is actually $(N^0_0 + N^0_2)/N^0_0$ under this assumption. Therefore, the net atom number of $N^0_3$ must be obtained by correction for the contribution of $^{242m}\text{Am}$ with the calculated value of $N^0_2$ as mentioned later.

(2) Determination of the Atom Number Ratios of $^{242m}\text{Am}/^{241}\text{Am}$ from the Growth Curves

When the atom numbers after the chemical separation of the irradiated targets are indicated as $N_0c, N_{1c}, N_{2c}, N_{3c}$ and $N_{4c}$ for $^{241}\text{Am}, ^{242m}\text{Am}, ^{242}\text{gAm}, ^{242}\text{Cm}$ and $^{238}\text{Pu}$, respectively, those are expressed by

$$N_0c = N_0^0 e^{-\lambda_0 t},$$

$$N_{1c} = N_1^0 e^{-\lambda_1 t},$$

$$N_{2c} = k_1 \lambda_1 N_{1c}^0 \left( \frac{1}{\lambda_2 - \lambda_1} e^{-\lambda_2 t} + \frac{1}{\lambda_1 - \lambda_2} e^{-\lambda_1 t} \right)$$

$$+ N_2^0 e^{-\lambda_2 t},$$

$$N_{3c} = k_1 \lambda_1 \lambda_2 N_{1c}^0 \left( \frac{1}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} e^{-\lambda_3 t} + \frac{1}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} e^{-\lambda_1 t} \right)$$

$$+ \frac{1}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} e^{-\lambda_2 t}$$

$$+ \frac{1}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} e^{-\lambda_3 t} \right)$$

$$+ k_2 \lambda_2 N_{2c}^0 \left( \frac{1}{\lambda_3 - \lambda_2} e^{-\lambda_3 t} + \frac{1}{\lambda_2 - \lambda_3} e^{-\lambda_2 t} \right),$$

(17)

where $N^0_0$ means the number of atoms as of the chemical separation (180 days after EOI) and $N^0_2=0$ because of the Am purification. The amount of $^{238}\text{Pu}$ produced from $^{242}\text{Cm}$, $N_{4c}$, is negligibly small as compared with that of $^{241}\text{Am}$ (target) in this growth analysis. The values $k_1$ and $k_2$ are the correction factors for branching ratios of $\Gamma$ of $^{242m}\text{Am}$ (0.995) and $\beta$-decay of $^{242}\text{gAm}$ (0.827), respectively. Therefore, the growth of the activity ratio of $^{242}\text{Cm}/^{241}\text{Am}$ can be expressed by

$$\frac{dN_{3c}}{dt} = \frac{dN_{0c}}{dt} = \frac{\lambda_3 N_{3c}}{\lambda_0 N_{0c}}$$

$$= \frac{k_1 \lambda_1 \lambda_2 \lambda_3}{\lambda_0(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} \cdot N_0^0 e^{-(\lambda_2 - \lambda_1)t}$$

$$+ \frac{k_1 \lambda_1 \lambda_2 \lambda_3}{\lambda_0(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} \cdot N_{1c}^0 e^{-(\lambda_3 - \lambda_2)t}$$

$$+ \frac{k_2 \lambda_2 \lambda_3}{\lambda_0(\lambda_3 - \lambda_2)} \cdot N_{2c}^0 e^{-(\lambda_3 - \lambda_2)t}.$$

By least squares fitting of this formula to the growth curve (see Fig. 5), the atom number ratio of $^{242m}\text{Am}/^{241}\text{Am}$ at the chemical separation, $N_{1c}/N_{0c}$, can be determined, where the $N_{2c}/N_{0c}$ is the atom number ratio of the decay product $^{242}\text{gAm}$ of $^{242m}\text{Am}$ to the $^{241}\text{Am}$ target at the separation and is a useless value in this study.

III. RESULTS AND DISCUSSION

1. Neutron Flux

The reaction rates, $R$ and $R'$, of $^{59}\text{Co}$ and $^{197}\text{Au}$ (neutron flux monitors) were obtained from the absolute activities measured by the $\gamma$-ray spectrometry. The values of $R_0/\sigma_0$ and $R'/\sigma_0$ in Eqs. (5) and (6) were computed by using the data of $\sigma_0(0^+)8$ listed in Table 1. The obtained values of $R_0/\sigma_0$ and $R'/\sigma_0$ were plotted as a function of the parameter $S_0 G_{\text{epi}}$ as shown in Fig. 4, where the other parameters used here are also given in Table 1. By least squares fitting of Eq. (5) or (6) to the data (see Fig. 4), the neutron fluxes $p_1, p_2, p'_1$ and $p'_2$ were determined as shown in Table 2. The fitting errors are given in the table. The ratios of $p'_1$ to $p_1$ and $p'_2$ to $p_2$ are 0.0026 ± 0.0002 and 1.1 ± 0.1, respectively.

2. Atom Number Ratios of $^{242}\text{Cm}/^{241}\text{Am}$ and $^{242m}\text{Am}/^{241}\text{Am}$ at EOI

Figure 5 shows the decay and growth curves of the activity ratios of $^{242}\text{Cm}/^{241}\text{Am}$ measured both for the irradiated $^{241}\text{Am}$ targets and for the radiometric sources.
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prepared chemically from the irradiated targets. Statistical errors of the activity ratios measured in this work were 0.2-1.1%, because the high-resolution sources for a-ray spectrometry could be prepared by the electrodeposition\(^{(10)}\). The atom number ratios of $^{242}$Cm/$^{241}$Am (actually, $(^{242}$Cm+$^{242g}$Am)/$^{241}$Am) and $^{242m}$Am/$^{241}$Am at EOI, obtained by the least squares fitting and decay correction, are shown in Table 3, where the errors of the ratios result from the fitting. Using these values, the cross sections of the reactions of $^{241}$Am(n, g)$^{242m}$Am and $^{241}$Am(n, g)$^{242g}$Am were determined by an iteration calculation as described below.

3. Cross Sections of $^{241}$Am(n, $\gamma$)$^{242m}$Am and $^{241}$Am(n, $\gamma$)$^{242g}$Am

During the irradiation, the atom number of the target $^{241}$Am before irradiation, $N_0$, and $N_1$, $N_2$, and $N_3$, are the atom numbers of $^{242m}$Am, $^{242g}$Am and $^{242}$Cm during irradiation at time $t$, respectively; $\sigma_0$, $\sigma_{0m}$, and $\sigma_{0g}$ are the absorption cross section of $^{241}$Am, the capture cross section of $^{241}$Am leading to $^{242m}$Am, and the capture cross section of $^{241}$Am leading to $^{242g}$Am, respectively. The time-derivative of atom number is as follows; in this calculation, the sum of the two capture cross sections, i.e. $\sigma_{242mAm} + \sigma_{242gAm}$, is supposed to be the absorption cross section of $^{241}$Am because the fission cross section of $^{241}$Am (for example, about 3 b in the JENDL evaluation\(^{(1)}\)) is smaller than the absorption cross section (about 600 b).

\[
\frac{dN_1(t)}{dt} = N_0^{init} \phi_0 (1 - \sigma_0^{abs} \phi t) - N_1(t) (\lambda_1 + \sigma_1 \phi)
\]

\[(19)\]

Table 1 Nuclear data used for the neutron-flux monitoring and the cross-section determination\(^{(6)(8)}\)

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>$\sigma_0$ (b)</th>
<th>$I_0$ (b)</th>
<th>$s_0$</th>
<th>$G_{epi}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{55}$Co</td>
<td>37.18</td>
<td>74</td>
<td>1.83</td>
<td>0.995</td>
</tr>
<tr>
<td>$^{197}$Au</td>
<td>98.65</td>
<td>1,550</td>
<td>17.02</td>
<td>0.993</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>Decay (branching %)</th>
<th>$\sigma^{abs}$ (b)</th>
<th>$\sigma^{abs}'$ (b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{241}$Am</td>
<td>432.2$\pm$0.2 yr</td>
<td>$\alpha$ (100)</td>
<td>603$\pm$24</td>
<td>1,319$\pm$53</td>
</tr>
<tr>
<td>$^{242m}$Am</td>
<td>141$\pm$2 yr</td>
<td>$\beta$ (99.541$\pm$0.012)</td>
<td>7,660$\pm$766</td>
<td>1,810$\pm$181</td>
</tr>
<tr>
<td>$^{242g}$Am</td>
<td>16.02$\pm$0.02 h</td>
<td>$\beta$ (82.7$\pm$0.3)</td>
<td>7,600$\pm$760</td>
<td>1,650$\pm$165</td>
</tr>
<tr>
<td>$^{242}$Cm</td>
<td>162.8$\pm$0.2 d</td>
<td>$\alpha$ (100)</td>
<td>21$\pm$6</td>
<td>128$\pm$38</td>
</tr>
<tr>
<td>$^{238}$Pu</td>
<td>87.7$\pm$0.3 yr</td>
<td>$\alpha$ (100)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$s_0$: Cross section for 2,200 m/s neutrons

$I_0$: Resonance integral

$s_0 = \frac{2}{\left(\frac{I_0}{\sigma_0}\right)}$, where $I_0$ is the resonance integral after subtracting the 1/v component

$G_{epi}$: Self-shielding coefficient for epithermal neutrons

$\sigma^{abs}$: Absorption cross section for 2,200 m/s neutrons

$\sigma^{abs}'$: Absorption cross section for epithermal neutrons

Table 2 Neutron flux determined by the flux monitors

<table>
<thead>
<tr>
<th>Sample</th>
<th>$\phi_1$ or $\phi_1'$ (n/cm$^2$s)</th>
<th>$\phi_2$ or $\phi_2'$ (n/cm$^2$s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No Cd-cover</td>
<td>$(1.06 \pm 0.05) \times 10^{14}$</td>
<td>$(5.99 \pm 0.34) \times 10^{12}$</td>
</tr>
<tr>
<td>With Cd-cover</td>
<td>$(2.78 \pm 0.11) \times 10^{11}$</td>
<td>$(6.50 \pm 0.09) \times 10^{12}$</td>
</tr>
</tbody>
</table>

$R/s_0 = \phi_1 + \phi_2 G_{epi}$, $\phi_1 = n v_0$, $\phi_2 = n v_0 \sigma \sqrt{T/T_0}$

Fig. 4 Plot of $R/s_0$ against $G_{epi}$ for neutron flux monitors irradiated with and without Cd-cover

Statistical errors of the $R/s_0$ values are depicted in the figure.
respectively, where the amount of $^{242m}$Am supplied from the IT decay of $^{242m}$Am is negligibly small in this irradiation because of the large difference of the half-lives of these nuclides. The time-derivative of atom number of $^{242}$Cm is expressed by

$$\frac{dN_2(t)}{dt} = N_0^{init} \phi \sigma_0(1 - \sigma_0^{bs} \phi t) - N_2(t)(\lambda_2 + \sigma_2 \phi),$$

and

$$\frac{dN_3(t)}{dt} = k_3N_2(t)\lambda_2 - N_3(t)(\lambda_3 + \sigma_3 \phi).$$

The solutions of these differential equations are respectively

$$N_1(t) = N_0^{init} \sigma_0 \left( \frac{\phi}{A_1} + \phi^2 \sigma_0^{abs} \left( \frac{1}{A_1^2} - \frac{t}{A_1} \right) \right) + C_1 e^{-\lambda_1 t},$$

$$N_2(t) = N_0^{init} \sigma_0 \left( \frac{\phi}{A_2} + \phi^2 \sigma_0^{abs} \left( \frac{1}{A_2^2} - \frac{t}{A_2} \right) \right) + C_2 e^{-\lambda_2 t},$$

and

$$N_3(t) = k_3N_2(t)\lambda_2 \sigma_0 \left[ \frac{\phi}{A_1 A_3} + \phi^2 \sigma_0^{abs} \left( \frac{1}{A_2 A_3^2} + \frac{t}{A_2 A_3} \right) \right] + C_3 e^{-\lambda_3 t} + C_4 e^{-\lambda_4 t},$$

Table 3 Measured atom ratios of $^{242}$Cm/$^{241}$Am and $^{242m}$Am/$^{241}$Am at EOI and the calculated results by the iteration method

<table>
<thead>
<tr>
<th>Sample</th>
<th>Atom ratio measured $^{242}$Cm/$^{241}$Am</th>
<th>Atom ratio measured $^{242m}$Am/$^{241}$Am</th>
</tr>
</thead>
<tbody>
<tr>
<td>No Cd-cover</td>
<td>$(2.468 \pm 0.005) \times 10^{-2}$</td>
<td>$(2.866 \pm 0.032) \times 10^{-3}$</td>
</tr>
<tr>
<td>With Cd-cover</td>
<td>$(2.869 \pm 0.011) \times 10^{-3}$</td>
<td>$(5.120 \pm 0.048) \times 10^{-4}$</td>
</tr>
</tbody>
</table>

† The measured atom numbers of $^{241}$Am (target) before the irradiation are $(7.166 \pm 0.018) \times 10^{14}$ and $(1.847 \pm 0.004) \times 10^{15}$ for the sample without Cd-cover and for the sample with Cd-cover, respectively.
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where $A_j$ and $C_j$ are the constants and the integral constants, i.e.,

\begin{align}
A_1 &= \lambda_1 + \sigma_1 \phi, \\
A_2 &= \lambda_2 + \sigma_2 \phi, \\
A_3 &= \lambda_3 + \sigma_3 \phi, \\
C_1 &= -N_0^{init} \sigma_{om} \left( \frac{\phi}{A_1} + \frac{\phi^2 \sigma_{abs}^{ns}}{A_1^2} \right), \\
C_2 &= -N_0^{init} \sigma_{og} \left( \frac{\phi}{A_2} + \frac{\phi^2 \sigma_{abs}^{ns}}{A_2^2} \right), \\
C_3 &= \frac{k_2 N_0^{init} \lambda_2 \sigma_0 (\phi A_2 + \phi^2 \sigma_{abs}^{ns})}{A_2^2 (A_2 - A_3)}, \\
C_4 &= \frac{k_2 N_0^{init} \lambda_2 \sigma_0 (\phi A_3 + \phi^2 \sigma_{abs}^{ns})}{A_3^2 (A_3 - A_2)}. 
\end{align}

Algorithm to obtain the atom numbers of $^{242m}\text{Am}$, $^{242g}\text{Am}$, and $^{242}\text{Cm}$ at EOI by an iteration method is as follows:

(i) Setting an initial value of $\sigma_{abs}^{ns}$ of $^{241}\text{Am}$ temporarily (i.e., $603 \pm 24$ b). The absorption cross sections of $^{242m}\text{Am}$, $^{242g}\text{Am}$ and $^{242}\text{Cm}$ evaluated in JENDL-3.2(1) were used as constant values in this calculation. The nuclear data and their uncertainties are shown in Table 1.

(ii) From the measured atom ratio of $^{242m}\text{Am}/^{241}\text{Am}$, $^{242m}\text{Am}/^{241}\text{Am}_{\text{measured}}$, calculating the atom number $N_1(t_I)$ of $^{242m}\text{Am}$ at $t_I$ by the relation of

\begin{equation}
N_1(t_I) = \left( \frac{^{242m}\text{Am}}{^{241}\text{Am}} \right)_{\text{measured}} N_0^{init} (1 - \sigma_{abs}^{ns} \phi t_I), \tag{32}
\end{equation}

where $t_I$ is the irradiation time (4 d).

(iii) Getting the temporary value of $\sigma_{om}$ by using Eq. (22).

(iv) From the measured atom ratio of $^{242}\text{Cm}/^{241}\text{Am}$, $^{242}\text{Cm}/^{241}\text{Am}_{\text{measured}}$, calculating the atom number $N_3(t_I)$ of $^{242}\text{Cm}$ at time $t_I$ by the relation of

\begin{equation}
N_3(t_I) = \left( \frac{^{242}\text{Cm}}{^{241}\text{Am}} \right)_{\text{measured}} N_0^{init} (1 - \sigma_{abs}^{ns} \phi t_I). \tag{33}
\end{equation}

The obtained $N_3(t_I)$ value contains the component of the decay product $^{242}$Cm of $^{242g}\text{Am}$ that was directly produced by the $^{241}\text{Am}(n, \gamma)^{242g}\text{Am}$ reaction. Such $^{242}\text{Am}$ existed at EOI but decayed out thoroughly at the activity measurement in this study.

(v) Obtaining the temporary value of $\sigma_{og}$ (i.e., $\sigma_{og}^{ns} - \sigma_{om}$) by using the $\sigma_{om}$ value calculated in (iii).

(vi) Getting the atom number $N_2(t_I)$ of $^{242g}\text{Am}$ by using the $\sigma_{og}$ value and Eq. (23).

(vii) Calculating the net atom number $N_3(t_I)^{\text{net}}$ of $^{242}\text{Cm}$, which does not contain the component of the decay product $^{242}\text{Cm}$ from the directly produced $^{242g}\text{Am}$, by using

\begin{equation}
N_3(t_I)^{\text{net}} = N_3(t_I) - k_2 N_2(t_I). \tag{34}
\end{equation}

(viii) Getting the temporary value of $\sigma_{og}$ by using the $N_3(t_I)^{\text{net}}$ value and Eq. (24).

(ix) Calculating $\sigma_{abs}^{ns}$ by summing $\sigma_{om}$ and $\sigma_{og}$.

(x) Iterating the procedures (ii)–(ix) until the value of $\sigma_{abs}^{ns}$ converges.

(xi) Calculating $N_1(t_I)$ and $N_2(t_I)$ by using the converged value $\sigma_{abs}^{ns}$ which is the actual cross section in the neutron spectrum of JMTR.

The same iteration calculation as above was carried out for the Cd-covered sample by temporarily setting an initial value of $\sigma_{abs}^{ns}$ (i.e., $1,319 \pm 53$ b), then $N_1(t_I)'$ and $N_2(t_I)'$ were also obtained by using the converged value $\sigma_{abs}^{ns}'$.

The final results calculated by the iteration are given in Table 3. On the basis of the obtained values, the nuclear transmutation of $^{241}\text{Am}$ by the neutron-induced reaction can be understood quantitatively; 3.5% of the atom number of the $^{241}\text{Am}$ target without Cd-cover is transmuted to other nuclides after 4-day irradiation in JMTR. The transmuted amount goes to $^{242m}\text{Am}$ (8.0%), $^{242g}\text{Am}$ (19.4%) and $^{242}\text{Cm}$ (63.9%), while the remaining percentage (8.7%) may be attributed to the nuclides of $^{243}\text{Am}$ and $^{243}\text{Am}$ produced by two-neutron capture, other minor actinides as decay products like $^{238}\text{Pu}$, $^{239}\text{Pu}$, etc., and fission products. The radioactivities of $^{243}\text{Am}$ and $^{243}\text{Cm}$ could not be detected in this a-ray spectrometry. On the other hand, in the case of the irradiation of the Cd-covered target, 0.4% of the $^{241}\text{Am}$ target is lost by the irradiation and the transmuted amount changes to $^{242m}\text{Am}$ (5.2%), $^{242g}\text{Am}$ (21.7%) and $^{242}\text{Cm}$ (69.5%). Remaining part of the products (3.6%) may be occupied by $^{243}\text{Am}$, $^{243}\text{Cm}$, other actinides, and fission products.

From the values of $\sigma_{abs}^{ns}$, $\sigma_{abs}^{ns}'$, $N_1(t_I)$, $N_1(t_I)'$, $N_2(t_I)$ and $N_2(t_I)'$ obtained by the iteration calculation, the ratios of $R/R'$ for $^{242m}\text{Am}$ and $^{242g}\text{Am}$ can be calculated by using the relations of

\begin{equation}
R_1 = \sigma_{om} \phi \\
\phantom{R_1} = \frac{N_1(t_I)}{N_0^{init}} \frac{N_0^{init} (1 - \sigma_{abs}^{ns} \phi t_I)}{A_1} \tag{35}
\end{equation}

\begin{equation}
R_1' = \sigma_{om} \phi' \\
\phantom{R_1'} = \frac{N_1(t_I)'}{N_0^{init}'} \frac{N_0^{init} (1 - \sigma_{abs}^{ns} \phi t_I)}{A_1}, \tag{36}
\end{equation}

\begin{equation}
R_2 = \sigma_{og} \phi \\
\phantom{R_2} = \frac{N_2(t_I)}{N_0^{init}} \frac{N_0^{init} (1 - \sigma_{abs}^{ns} \phi t_I)}{A_2}, \tag{37}
\end{equation}

and
\[ R_2' \equiv \sigma_{09} \phi' \]

\[
= \frac{N_2(t_f)^{'} \lambda_0}{A^{\text{thin}}_0 (1 + \phi_2'^{'} \sigma_0^{\text{abs}})} \left( 1 - e^{-A_2 t_f} \right) - \frac{\phi_0'^{'} \sigma_0^{\text{abs}} t_f}{A_2^{'}}, \tag{38}
\]

where \( A_1' = \lambda_1 + \sigma_1' \phi' \) and \( A_2' = \lambda_2 + \sigma_2' \phi' \).

The ultimate cross sections of \( ^{241}\text{Am}(n, \gamma)^{242m}\text{Am} \) and \( ^{241}\text{Am}(n, \gamma)^{242g}\text{Am} \) for 2,200 m/s neutrons, \( \sigma_0 \), were obtained from Eqs. (7) and (8) by using the measured values of \( \phi_1, \phi_2, \phi'_1, \phi'_2 \), the reaction rate by thermal neutrons \( (R) \) and that by epithermal neutrons \( (R') \) obtained by the iteration calculation. The resonance integrals, \( I_0 \), were computed from Eqs. (3) and (9). Table 4 shows the cross sections obtained by this method. Considering the propagation of the errors both in the measured atom number ratios (0.2 to 1.1%) and the measured neutron fluxes (1.4 to 5.7%) and in the evaluated half-lives (0.1 to 1.4%) and the evaluated cross sections (4 to 30%) used for the iteration calculation, the overall uncertainties of the cross sections measured in this study are estimated to be 6.2 to 8.6%.

Figure 6 shows the formation process of \( ^{242m}\text{Am}, ^{242g}\text{Am} \) and \( ^{242}\text{Cm} \) in the \( ^{241}\text{Am} \) target irradiated by thermal neutrons, calculated from Eqs. (22)-(24) using the cross sections measured in this study. The formation of \( ^{242m}\text{Am}, ^{242g}\text{Am} \) and \( ^{242}\text{Cm} \) can be quantitatively understood from this figure. In order to quantitatively consider the further transmutation of the products starting from \( ^{241}\text{Am} \), it is essential to obtain the precise nuclear data of cross section, half-life and branching ratio of decay on \( ^{242m}\text{Am}, ^{242g}\text{Am}, ^{242}\text{Cm}, ^{243}\text{Am} \) and \( ^{243}\text{Cm} \).

### Table 4 Cross sections and the isomeric yield ratios of \( ^{242}\text{Am} \) produced by the \( ^{241}\text{Am}(n, \gamma) \) reaction

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Cross section (b)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>( ^{241}\text{Am}(n, \gamma)^{242m}\text{Am} )</td>
<td>( \sigma_0 ) 85.7±6.3</td>
<td>114±7</td>
</tr>
<tr>
<td></td>
<td>( \sigma_0 ) 54±5</td>
<td>195±20</td>
</tr>
<tr>
<td>( ^{241}\text{Am}(n, \gamma)^{242g}\text{Am} )</td>
<td>76±58 1,694±146</td>
<td>Mughabghab(12)</td>
</tr>
<tr>
<td>Total capture</td>
<td>533±13 1,230±100</td>
<td>This work</td>
</tr>
<tr>
<td>( ^{241}\text{Am}(n, \gamma)^{242m}\text{Am} )</td>
<td>854±58 1,808±146</td>
<td>Mughabghab(12)</td>
</tr>
<tr>
<td></td>
<td>587±12 1,425±100</td>
<td>Mughabghab(12)</td>
</tr>
<tr>
<td></td>
<td>600.4 1,305</td>
<td>JENDL-3.2(1)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Neutron energy</th>
<th>Isomeric ratio (g/total)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal</td>
<td>0.90±0.09</td>
<td>This work</td>
</tr>
<tr>
<td></td>
<td>0.91±0.02</td>
<td>Mughabghab(12)</td>
</tr>
<tr>
<td></td>
<td>0.92±0.06(^\dagger)</td>
<td>Wisshak et al.(13)</td>
</tr>
<tr>
<td>Epithermal</td>
<td>0.94±0.11</td>
<td>This work</td>
</tr>
<tr>
<td></td>
<td>0.86±0.09</td>
<td>Mughabghab(12)</td>
</tr>
<tr>
<td></td>
<td>0.65±0.05(^\dagger)</td>
<td>Wisshak et al.(13)</td>
</tr>
</tbody>
</table>

\(^\dagger\) value at 14.75-meV neutrons, \(^\dagger\) at ~30-keV neutrons
The isomeric ratios of the ground state to the total capture cross sections, $g/(m+g)$, are also given in Table 4. The ratio of 0.90 measured at thermal neutron energy agrees excellently with the evaluated value by Mughabghab(12) and the previous data measured by Wisshak et al.(13) However, the isomeric ratio of 0.94 in the epithermal region is not consistent with the value of 0.65 by Wisshak et al. It seems that this discrepancy is due to the difference between our measurement performed with a reactor neutron spectrum and their quasi-monoenergetic experiment with ~30-keV neutrons.

**ACKNOWLEDGMENT**

We would like to appreciate many persons of Japan Atomic Energy Research Institute (JAERI) for their help to make this study possible in many ways, namely, the staff of Mechanical Engineering Division for fabricating the capsule, the JMTR crew for irradiation, the Hot Lab. crew, Mr. S. Motoishi and Mr. K. Kobayashi for cutting the irradiated outer and inner capsules, and Mr. T. Sukegawa and Mr. A. Sato for sample transportation. Thanks are also due to Dr. Y. Nakagawa for comments on the nuclear data of minor actinides and Dr. M. Tanase for encouragement in this study.

**REFERENCES**