Measurements of the Effective Cross Section of the Th-233(n, γ)Th-234 Reaction Using the KUR Core

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Abstract

Approximately 100 mg of solid thorium (Ⅳ) nitrate tetrahydrate ($Th(NO_3)_4 \cdot 4H_2O$; thorium nitrate) was irradiated together with neutron fluence monitors of 0.1143%Au-Al and 0.483%Co-Al alloy foil, in the Kyoto University Reactor (KUR). The Th was chemically purified after irradiation and measured using high-purity germanium detectors (HPGes) including a low-energy photon spectrometer (LEPS), in order to obtain the gamma ray spectra of Th-234. On the other hand, the thermal neutron flux and epithermal index, i.e., the strength of the epithermal component relative to the density of neutrons including both thermal and epithermal neutrons, were determined by the Westcott convention using the multiple foil activation method in which the Au(n, γ)Au-198 and Co(n, γ)Co-60(g+m) reactions were used. The effective cross sections were determined for various epithermal indexes.

1. Introduction

In the thorium cycle, although the experimental value of the Th-233(n, γ)Th-234 reaction cross section is closely linked with the evaluation of the generation of U-233, only two reports on obtaining experimental values, including an unpublished paper, exist, as far as the author knows [1, 2]. Moreover, no one has ever used HPGe in the determination of the cross section. Therefore, in order to verify the widely used value of the cross section and to improve its accuracy, measurements of the effective cross section have been carried out.

2. Experimental

The experiments were performed three times using irradiation facilities of Hyd (Hydraulic conveyor: at the center of the core) and Slant (Slant exposure tube: outside the reflectors). In the case of using the Slant, experiments were repeated twice: Slant 1st and Slant 2nd. In this paper, unless otherwise noted, we describe the case of the experiment using Hyd, where the neutron flux is the most intense. Configurations and characteristics of the facilities have been presented in Ref. 3. Figure 1 shows the placement of the irradiation samples. Irradiation was applied at 5000 kW for 5 h. In order to wait for Th-233 and Th-231, produced by the reactions of Th-232(n, γ) and Th-232(n,2n), respectively, to decay out, chemical purification was performed more than 13 days after the irradiation. The flow diagram and details of the purification procedure are shown in Fig. 2 and in previous papers [4, 5, 6]. The final precipitate was cemented with glue; this was the Th sample. The recovery rate of Th-232 was roughly 50%.

The activity of Th-234 was determined from the area of the peak at 92.57 keV in the γ-ray spectra, which is a doublet of 92.35 and 92.78 keV with intensities (2.73±0.05) and (2.69±0.05)%, respectively [7] (see Fig. 3). The activity of Th-234 immediately after irradiation was (101,200 ± 2,500) Bq. Activities of Au-198 and Co-60 were also determined using the calibrated HPGes.

3. Determination of the effective cross section in the case of double neutron capture

The activity of Th-234 immediately after irradiation ($\lambda_3 N_3$) is determined by solving the differential equations (1) and (2), which represent the processes of the production and destruction of Th-233 and Th-234, respectively [8].

$$\frac{dN_2}{dt} = nv_0\hat{\sigma}_{12}N_1 - \lambda_2 N_2 \quad (1)$$

$$\frac{dN_3}{dt} = nv_0\hat{\sigma}_{23}N_2 - \lambda_3 N_3 \quad (2)$$

Here, N_1, N_2 and N_3 are the numbers of nuclides of Th-232, Th-233 and Th-234, respectively. $\hat{\sigma}_{12}$ and $\hat{\sigma}_{23}$ are the effective cross sections for the reactions of Th-232(n, γ)Th-233 and Th-233(n, γ)Th-234, respectively. λ_2 and λ_3 are the decay constants of Th-233 and Th-234, respectively. n is the density of neutrons including both thermal and epithermal neutrons. v_0 is 2200 m/s, and t is irradiation time. The solution is formulated as

$$\lambda_3 N_3 = \frac{\hat{\sigma}_{12}\hat{\sigma}_{23}(nv_0)^2 N_1}{\lambda_2(\lambda_2 - \lambda_3)}\left[\lambda_2(1 - e^{-\lambda_3 t}) - \lambda_3(1 - e^{-\lambda_2 t})\right] , \quad (3)$$

where t is 5 h. Therefore, $\hat{\sigma}_{23}$ can be calculated when the following is known: $\lambda_3 N_3$, $\hat{\sigma}_{12}$, amount of recovered Th-232 after the chemical purification (N_1) and Westcott conventionality thermal neutron flux (nv_0).

3. –1 Westcott conventionality thermal neutron flux determined using multiple foil activation method [9, 10]

This method of using the Au(n, γ)Au-198 and Co(n, γ)Co-60(g+m) reactions has been reported in detail in Ref. 3, therefore, in this section, the explanation is restricted to the essential points and the results.

nv_0 and epithermal neutron flux $nv_0 r\sqrt{T/T_0}$ are, respectively, equivalent to the intercept and slope of the linear equation

$$\frac{R}{\sigma_0 g G_{th}} = nv_0 + nv_0\, r\sqrt{T/T_0}\,\frac{s_0 G_{epi}}{g G_{th}}, \quad (4)$$

where R is the reaction rate of the Au(n, γ) or Co(n, γ) reaction. G_{th} and G_{epi} are self-shielding factors within the foil for thermal and epithermal neutrons, respectively, and they are approximately unity, because Au and Co can be considered to be infinite dilution. $r\sqrt{T/T_0}$ is the epithermal index, T is neutron temperature and T_0 is 293.6 K. g and s_0 are measures of the departure of the cross-section law from the 1/v form in the thermal and epithermal regions, respectively. The former factor is the Westcott g-factor, which has been tabulated by Westcott [11] and Gryntakis and Kim [12]. If the cross section obeys the 1/v law, g = 1 and s_0 = 0. Furthermore, s_0 is defined by [13]

$$s_0 = \frac{1}{\sigma_0}\sqrt{\frac{4}{\pi}}\; I_0', \quad (5)$$

where I'_0 is the reduced resonance integral which is obtained by subtracting the 1/v-term from resonance integral I_0, which is given by $I'_0 = I_0 - 0.45\,\sigma_0$ when the cadmium cutoff energy E_{Cd} is 0.5 eV.

The same Eq. (4) with a common intercept and slope will be formed for the Au(n, γ)Au-198 and Co(n, γ)Co-60(g+m) reactions, if Au and Co are irradiated at the same time and at the same position. The nuclear data and parameters used are listed in Table 1. Figure 4 shows the plots of Eq. (4) and the results are shown in Table 2.

3. - 2 Effective cross section of the Th-232(n, γ)Th-233 reaction

The effective cross section is given by

$$\hat{\sigma}_{12} = \sigma_0\left(g + r\sqrt{T/T_0}\, s_0\right). \quad (6)$$

Accordingly, using the evaluated cross section and resonance integral of JENDL -3.2 [15], viz., σ_0 = 7.4 b and I_0 = 84.4 b, s_0 is defined to be 12.4. Therefore, $\hat{\sigma}_{12}$ can be calculated using $r\sqrt{T/T_0}$, which is never determined without experiment, as listed in Table 2, when g is assumed to be unity.

3. –3 Recovered amount of Th-232

The thorium contents in the Th samples were quantitatively analyzed using the activation method. Th samples for which the measurements of Th-234 and Pb-212 activities have been completed, were irradiated together with Th foil of known weights and 10 pieces of 5 mg Au foil in order to determine neutron fluences at the positions of the Th samples and Th foil, using the heavy-water neutron irradiation facility (D2O facility [16]), where uniform neutron flux is available to a certain extent. The targets were attached on a 15×7 cm piece of corrugated cardboard and then fastened onto the Bi layer of the D2O facility; irradiation was performed at 100 kW for 26 h. Relative neutron flux, i.e., relative reaction rates of the Au(n, γ)Au-198 reaction, on the cardboard is shown in Fig. 5. In the figure, -3, 0 and 3 cm on the abscissa are the positions of the Th samples, and -6 and 6 cm are those of the Th foil. The irregularity of the neutron flux was taken into account in the correction of induced activities of Pa-233 produced by the beta decay after the Th-232(n, γ) Th-233 reaction. Recovered amounts of Th-232 were 23.4, 19.7 and 43.7 mg in the cases of Hyd, Slant 1st and Slant 2nd, respectively.

4. Results and discussion

Results of the experiments are listed in Table 2. Johnston et al. [1] determined the effective cross section to be (1470 ± 100) b and the thermal neutron cross section to be 1450 b, and Hyde et al. [2] determined the effective cross section to be 1350 b (quoted from Ref. 1). The effective cross section varies mainly according to the epithermal index and effective cross section of the Th-232(n, γ)Th-233 reaction. Therefore, although, the comparison of the effective cross sections cannot be made unconditionally, our results are slightly smaller than those obtained by Johnston et al. [1] and Hyde et al. [2]. More irradiation experiments using Slant and Pn-2 (pneumatic tube No. 2) are being carried out.

Table 1 Nuclear data and parameters used in the determination of the neutron fluxes

Reactions	σ_0 (b) [a]	I_0 (b) [a]	g (at 40°C)	G_{th}	G_{epi}
Au(n, γ) Au-198	98.65±0.09	1550±28	1.005 [b]	0.999	0.996
Co(n, γ) Co-60(g+m)	37.18±0.06	74±2	1	1	1

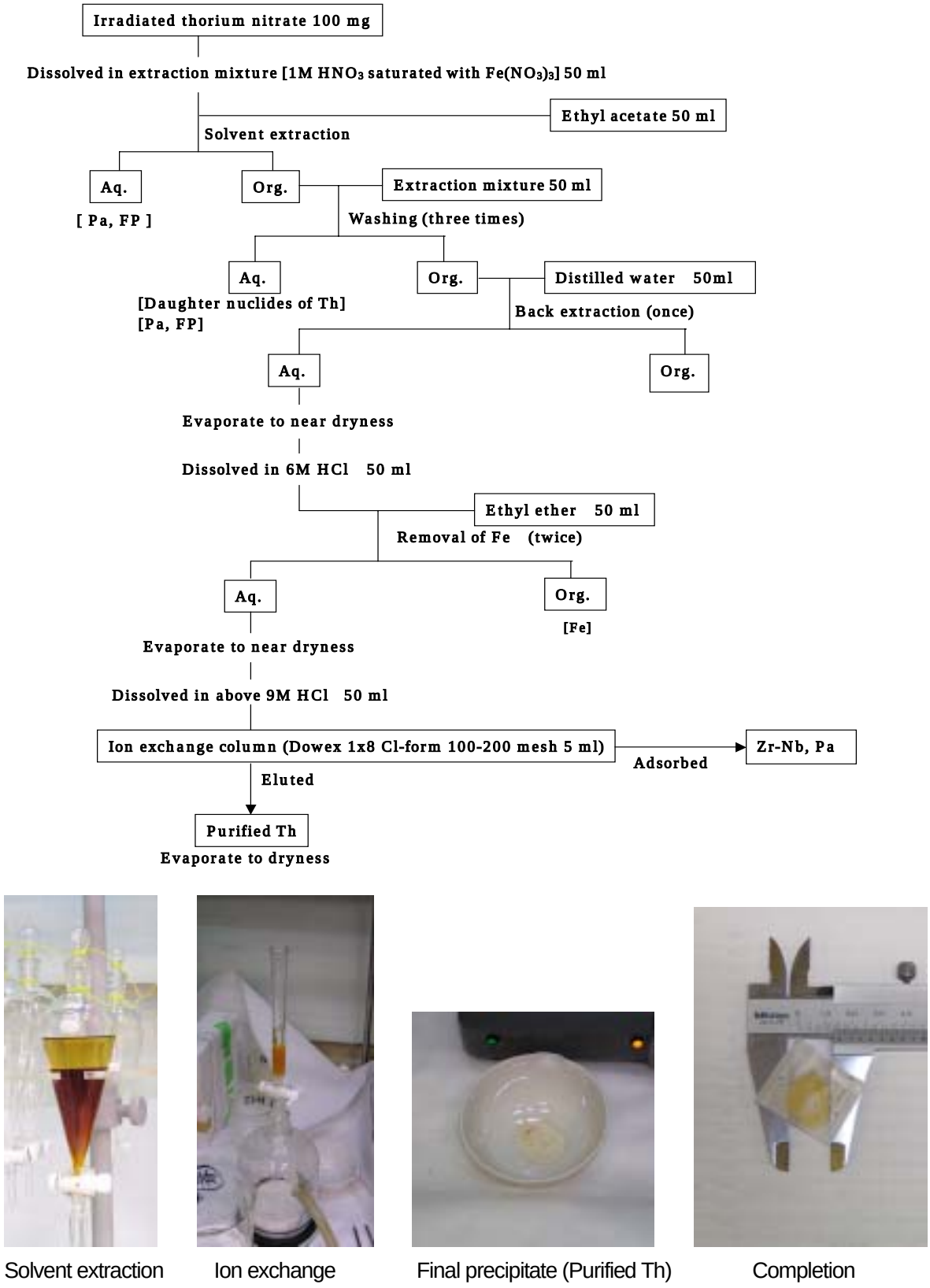
a) Ref. [14]. b) Ref. [12]

Table 2 Results

Irradiation facilities:	Hyd	Slant (1st exp.)	Slant (2nd exp.)
nv_0 (n/s/cm^2)	(1.08±0.03)×10^{14}	(1.07±0.03)×10^{13}	(9.90±0.03)×10^{12}
$r\sqrt{T/T_0}$	0.0322 ± 0.0023	0.0129 ± 0.0009	0.0118 ± 0.0008
$\hat{\sigma}_{12}$ (b)	10.34 ± 0.21	8.59 ± 0.17	8.48 ± 0.17
$\hat{\sigma}_{23}$ (b)	1340 ± 90	1330 ± 90	1270 ± 90

Acknowledgement

We acknowledge Dr. S. Yamada, Associate Prof. (ret.) for his comprehensive assistance on the use of the LEPS, and Dr. Y. Sakurai for his suggestions and assistance on the use of the D2O facility.



Solvent extraction

Ion exchange

Final precipitate (Purified Th)

Completion

Fig. 2 Flow diagram of the chemical purification and the photographs. It begins with the dissolution of the irradiated thorium nitrate into 1M nitric acid saturated with $Fe(NO_3)_3$ more than 13 days after irradiation.

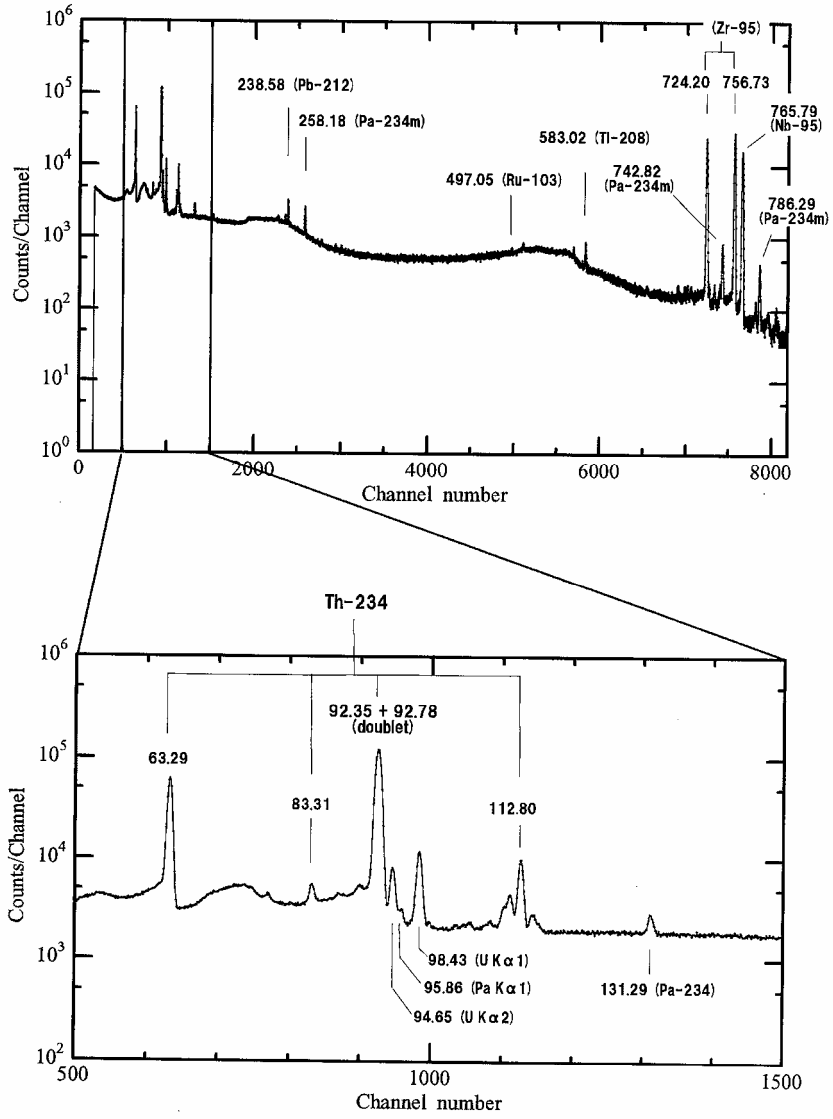


Fig. 3
Gamma-ray spectrum of the Th sample in the energy range from 20 to 820 keV (upper spectrum), obtained using a low-background high-resolution HPGe with a relative efficiency of 50%, at a S (Source) to D (Detector) distance of 10 cm. Accumulated for 52537 s. Waiting time after the chemical purification was 16 d. Numerical values are the energies of photons in keV. Lower spectrum is enlargement of the energy range from 50 to 150 keV. Activity of Th-234 was determined from the peak area of 92.57 keV (average of the two γ-ray energies of 92.35 and 92.78 keV). There is no appreciable interference caused by unwanted nuclides in the analysis of the 92.57 keV peak.

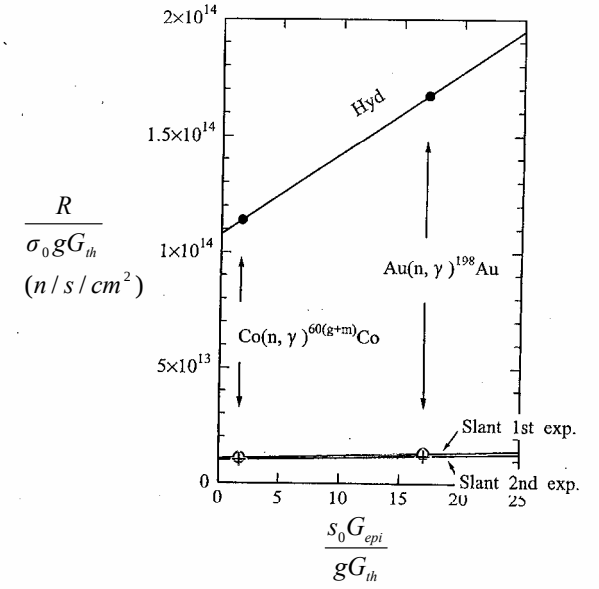


Fig. 4
Westcott conventionality thermal neutron fluxes nv_0 (intercepts) and epithermal neutron fluxes $nv_0 r\sqrt{T/T_0}$ (slopes) at the irradiation positions of thorium nitrate, determined from Eq. (4). The results are shown in Table 2.

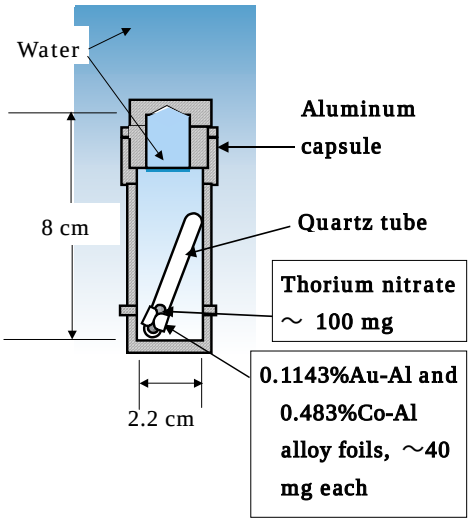


Fig. 1 Irradiation samples in an aluminum capsule of Hyd.

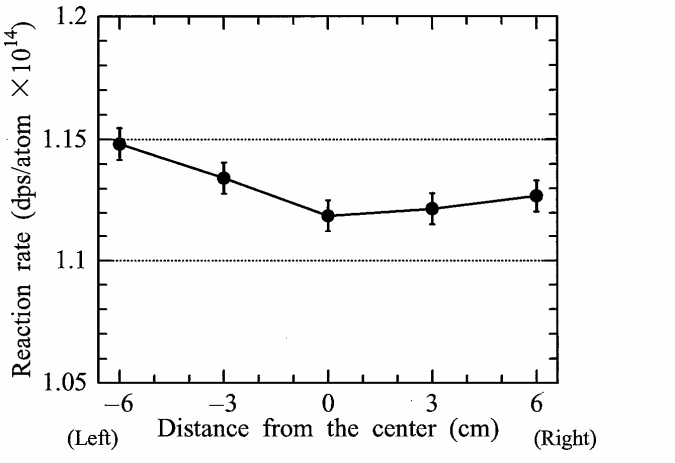


Fig. 5 Distribution of the reaction rates of the Au(n. γ)Au-198 reaction on the corrugated cardboard, which corresponds with that of neutron fluxes at the Th samples and Th foil positions.

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