^{232}Th(n,γ)^{233}Th Thermal Reaction Cross-Section Measurement

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Abstract. The ^{232}Th(n,γ)^{233}Th thermal neutron-capture reaction cross section was measured using targets of $\sim$ 1.5 mg of high-purity metallic thorium irradiated in the IPEN IEA-R1m 5 MW pool research reactor. The ^{197}Au(n,γ)^{198}Au reaction was used to monitor the thermal and epithermal neutron fluxes in the irradiation position, which was found using the Westcott formalism. The residual gamma-ray activity was followed with an HPGe detector. The detector efficiency curve was fitted by the least-squares method applying covariance analysis to all uncertainties involved. The experimental result is $\sigma_0 = 7.20 \pm 0.20$ b, in agreement with previous published values.

INTRODUCTION

The Th-U fuel cycle needs accurate experimental cross-section data in order to obtain precise reactor-parameter predictions. The IAEA Consultants Meeting on Assessment of Nuclear Data Needs for Thorium and other Advanced Cycles [1] specified the needed precision for the different reactions of interest.

A previous measurement of a ^{232}Th(n,γ)^{233}Th thermal neutron reaction cross section was published by Jones [2], and evaluations or calculations on other neutron energy ranges were reported [3, 4] taking into account the data needed to develop accelerator-driven technologies. An IAEA Coordinated Research Program on Evaluated Nuclear Data for the Th-U fuel cycle is in progress. The first co-ordination meeting summary presents data on this reaction [5].

This paper reports the measurement of the thermal neutron absorption cross section for the ^{232}Th(n,γ)^{233}Th reaction in a reactor core irradiation, where the neutron flux was determined from Au monitor activation, and the cross section was evaluated from residual radioactivity determination with HPGe detectors. The adopted procedures were used before to measure the thermal neutron absorption cross sections for ^{137}Cs, ^{241}Am, and ^{57}Co [6, 7, 8].

EXPERIMENTAL METHOD

The target consisted of $\sim 1.5 \times 10^{-3}$ g natural isotopic composition metallic thorium (20 μg uncertainty, 100% ^{232}Th), obtained from a foil 0.0508 mm thick, 99,8503% purity (Reactor Experiments Inc., USA). The target nuclide ^{232}Th, 1.4×10^{10} y half-life [9], decays by β^- particle emission. The neutron activation product nucleus ^{233}Th, 22 min half-life [9], decays by β^- to ^{233}Pa, 27 d half-life [10], which in turn decays to ^{233}U by β^- emission. The gamma rays following this last decay, with energies equal to 300, 312, and 341 keV, and intensities 6.62%, 38.6% and 4.47% photons per decay [9], respectively, allowed the measurement of the reaction yield. Figure 1 shows the reaction and decay scheme of the nuclei involved in this experiment.

The irradiations were performed at the IPEN 5 MW pool-type research reactor operating at 2 MW. The irradiation sample consisted of a thorium target and Au-Al foils, irradiated in pairs with and without a 0.5-mm-thick Cd shield, wrapped in thin commercial aluminum foils to avoid loss of mass and external contamination, besides keeping the foils in the desired relative positions when irradiating. The sample was placed inside a polyethylene rabbit, transported to the reactor core by means of a pneumatic tube, and irradiated during 20 min. The time

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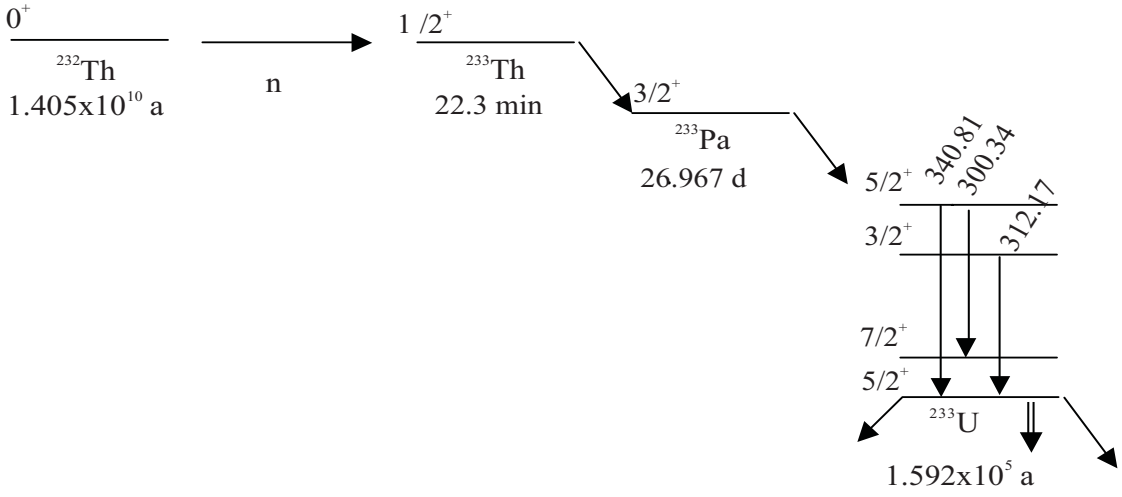

FIGURE 1. ^{232}Th neutron capture reaction and decay scheme, showing the main gamma-ray transitions of the decay chain [9].

interval between the end of the irradiation and the beginning of the residual radioactivity measurement was approximately one day for the Au flux monitor foils and three days for the thorium samples.

The thermal and epithermal average neutron fluxes, about 6×10^{11} and 2×10^{11} neutrons·cm^2/s, respectively, were monitored through the ^{197}Au(n, γ)^{198}Au reaction using 0.1% gold in Au-Al alloy foils and obtained through the Westcott formalism [11], as described in Maidana et al. [8]. The accuracy in the Au content in the alloy was previously checked by simultaneous irradiation with pure gold foils.

ACTIVITY DETERMINATION

The gamma-ray measurements were performed with a HPGe detector (35% ORTEC), shielded by a 10-cm-thick lead wall and previously calibrated with an standard ^{152}Eu source [12]. The ^{198}Au and ^{233}Pa residual activities were counted in a reproducible geometry at 250 mm from the detector end cap and calculated considering corrections like decay waiting time and gamma-ray self-absorption (calculated for the 312-keV gamma ray). Figure 2 shows the gamma-ray spectrum of one of the ^{232}Th irradiated samples. All the measurements were performed in live time counting methodology with pile-up rejection. The decay scheme parameters used in the activity determination appear in Table 1.

THERMAL CROSS SECTION DETERMINATION

The thermal cross section, σ_0, was calculated using the expression

$$\sigma_0 = \frac{A'}{N_0 \phi_{th} \sqrt{\frac{4T}{\pi T_0}}}$$

where:

A' = the thermal neutron induced ^{233}Th activity;
N_0 = the number of atoms of ^{232}Th sample;
ϕ_{th} = the thermal neutron flux determined with the Au-Al monitors and
T_0, T = absolute reference (20^{o}C) and moderator neutron temperatures, respectively.

RESULTS AND DISCUSSION

The final result for the ^{232}Th(n,γ)^{233}Th thermal neutron reaction cross section is the average of the values obtained in two irradiations, and it is presented in Table 2 along with the other experimental data found in the available literature. The analysis of the gamma-ray spectra did not show any other activity besides that following neutron capture by ^{232}Th, which was expected, because the samples were irradiated in a position far from the reactor core.

The 2.8% relative uncertainty obtained in this work is due to two kinds of errors. On one hand are errors that will be averaged out when evaluating independent data, from which the most important in this work was the correction for the activation by epithermal neutrons, 1.9%

TABLE 1. Decay parameters used in the activity determination [9, 10, 13]. The numbers in parentheses are the standard deviations in units of the last digit.

Radionuclide	Half-life (d)	Gamma ray Energy (keV)	Photons per Decay (%)
^{152}Eu	4933(11)	121.7824(4)	28.37(13)
		244.6989(10)	7.53(4)
		344.2811(19)	26.57(11)
		411.126(4)	2.238(10)
		443.965(4)	3.125(14)
		778.903(6)	12.97(6)
		964.055(4)	14.63(6)
		1085.842(4)	10.13(5)
		1112.087(6)	13.54(6)
		1408.022(4)	20.85(9)
^{198}Au	2.6943(8)	411.8044(11)	99.448(8)
^{233}Pa	26.975(13)	300.34(2)	6.62(13)
		312.17(2)	38.6(4)
		340.81(3)	4.47(4)

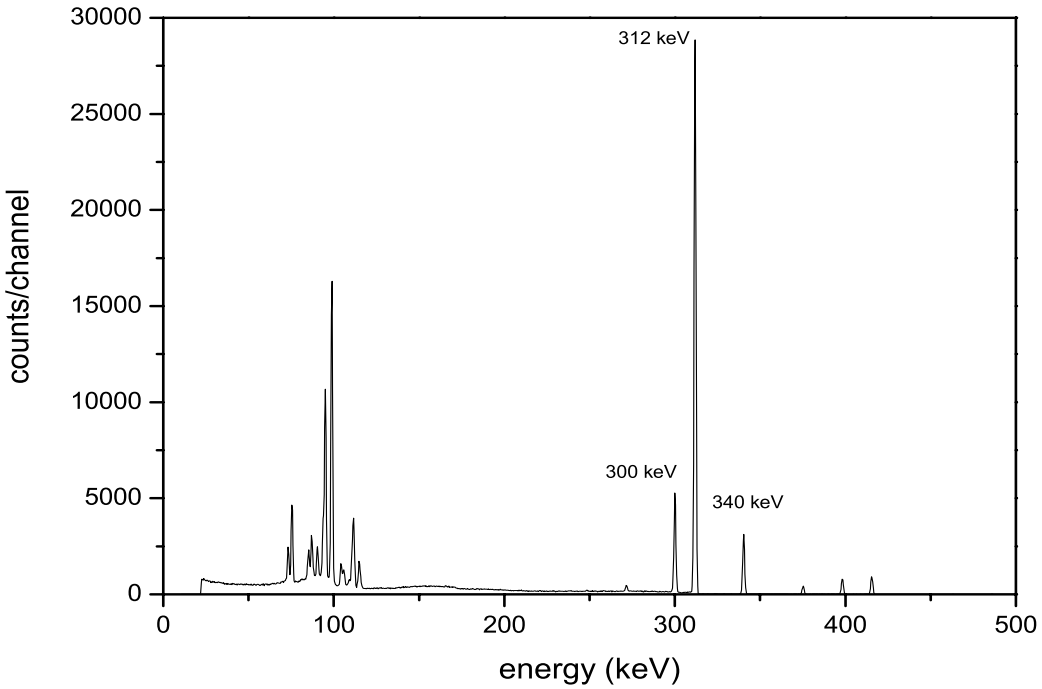

FIGURE 2. One-hour counting time gamma-ray spectrum of the sample acquired four days after irradiation. The 300-, 312-, and 340-keV ^{233}U gamma-ray transitions are marked. The energy dispersion is 0.2 keV/channel.

TABLE 2. Experimental thermal neutron absorption cross section, σ_0.

Reaction	σ_0 (b)	
	this work	Jones [2]
^{232}Th(n,γ)^{233}Th	7.20±0.20	7.33±0.17

of the cross section, and other smaller contributions, like time measurements and counting statistics. On the other hand are the errors that affect all results, from which the main contribution came from the emission probability of the 312-keV gamma ray, 1.0% relative standard deviation, followed by the relative precision in the Au cross section used as the monitor, 0.3%. The uncertainty due to the efficiency calibration is almost negligible because the final result depends only on the relative efficiency of the 312 keV from ^{233}Pa and 411 keV from ^{198}Au decay gamma rays.

The U-Th nuclear fuel cycle requires the knowledge of the ^{232}Th neutron-capture cross section with 1% relative standard deviation [1], which is not achievable by a single experiment by any procedure. For each methodology, there is a minimum precision bound for the obtained cross section, which is given by the errors that affect all results obtained by similar experiments. We have seen that by the methodology described here, the minimum precision bound is given by the ^{233}Pa 312-keV gamma-ray emission probability; the ^{198}Au cross section is already known with better precision than that required in the ^{232}Th cross section. Therefore, a better knowledge of the ^{233}Pa decay scheme is required to improve on the precision of the final result by this methodology, besides repeating the experiment many times.

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