Determination of neutron capture cross sections of ^{232}Th at 14.1 MeV and 14.8 MeV using the neutron activation method *

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Abstract: The ^{232}Th(n, γ)^{233}Th neutron capture reaction cross sections were measured at average neutron energies of 14.1 MeV and 14.8 MeV using the activation method. The neutron flux was determined using the monitor reaction ^{27}Al(n,α)^{24}Na. The induced gamma-ray activities were measured using a low background gamma ray spectrometer equipped with a high resolution HPGe detector. The experimentally determined cross sections were compared with the data in the literature, and the evaluated data of ENDF/B-VII.1, JENDL-4.0u+, and CENDL-3.1. The excitation functions of the ^{232}Th(n,γ)^{233}Th reaction were also calculated theoretically using the TALYS1.6 computer code.

Keywords: ^{232}Th(n,γ)^{233}Th reaction, cross section, neutron activation method

PACS: 25.40.-h, 28.20.Np **DOI:** 10.1088/1674-1137/41/4/044002

1 Introduction

The database of cross sections for the reactions induced by neutrons around 14 MeV plays a key role for the applied physics and the design of nuclear reactors. Specifically, the neutron capture cross section of ^{232}Th is an important parameter for the design of any nuclear reactor based on the Th-U fuel cycle[1]. In this cycle, ^{233}U is the fissile isotope and is formed from ^{232}Th by neutron capture followed by two β-decays.

$$^{232}\mathrm{Th} + \mathrm{n} \longrightarrow {}^{233}\mathrm{Th} \xrightarrow{\beta^-(22.3\,\mathrm{min})} {}^{233}\mathrm{Pa} \xrightarrow{\beta^-(26.975\mathrm{d})} {}^{233}\mathrm{U}$$

Moreover, the production of the ^{233}U depends on the ^{232}Th(n,γ) reaction cross section. This is currently required to be known within an accuracy of 1%–2% in order to be used safely in simulated techniques for predicting the dynamical behavior of complex arrangements in fast reactor or thorium molten salt reactors (TMSR) [2].

For the ^{232}Th(n, γ)^{233}Th reaction, many researchers have measured the cross section over a wide range of neutron energies from thermal to 17.28 MeV [3]. Around 14 MeV, only two data points for the ^{232}Th(n,γ) reaction cross-section are available, at 14.5 MeV and 14.8 MeV using the activation technique, and there is relatively large disagreement and uncertainty between these results. Meanwhile, the discrepancies between the evaluated data files (ENDF/B-VII.1 [4], JENDL-4.0u+ [5], CENDL-3.1 [6]) reach 40% at the neutron energy of 14.1 MeV. Hence, the neutron capture reaction cross section of ^{232}Th need further precise measurements to strengthen the reliability of the databases.

In this paper, we present the results of the ^{232}Th(n, γ)^{233}Th reaction cross sections at neutron energies of 14.1 and 14.8 MeV using the activation method and off-line γ-ray spectrometric technique. The measured results are discussed and compared with the previous experimental data and evaluated data, as well as the calculated results from TALYS1.6 code.

2 Experimental method

Two targets (thorium dioxide powder of 99.7% purity) and monitors (aluminum foil of 99.99% purity) were made into circular disks with a diameter of 20 mm for irradiation. The ThO_2 samples were 1.05 mm and 1.07 mm thick respectively, and the Al samples were 0.06 mm thick. A stack of Al-Th-Al was made from these two samples and mounted at 0 and 90° angles respectively, relative to the deuteron beam direction and centered about the tritium–titanium (T-Ti) target at distances of 35 mm.

The irradiations were carried out using the CPNG-600 neutron generator at China Institute of Atomic En-

Received 3 August 2016, Revised 22 October 2016
* Supported by Chinese TMSR Strategic Pioneer Science and Technology Project-The Th-U Fuel Physics Term (XDA02010100) and National Natural Science Foundation of China (11205076, 21327801)
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ergy (CIAE) and lasted approximately 5 hours. The neutrons had a yield of about $\sim 1.5\times10^{10}$n/(4π·s) and were produced by the T(d,n)^{4}He reaction. The ion beam current was up to 300 μA with effective deuteron energy of 300 keV. A solid tritium-titanium (T-Ti) target was used in the generator, with a thickness of 1.0 mg/cm^2.

The neutron energies in these positions were calculated by the Q equation [7] and compared with the method of cross-section ratios for ^{90}Zr(n,2n)$^{89m+g}$Zr and ^{93}Nb(n,2n)^{92m}Nb reactions [8,9] before irradiation. The determined neutron energies were 14.1±0.2 MeV and 14.8±0.2 MeV, respectively.

During irradiation, the neutron flux was determined using the monitor reaction ^{27}Al(n, α)^{24}Na, and the variation of the neutron yield was monitored by accompanying α-particles so that the corrections could be made for the fluctuation of neutron flux. The schematic view of the accompanying α-particle monitor was the same as shown in Ref. [10]. The Au–Si surface barrier detector used at 135°, accompanying the α-particle tube, was at a distance of 110 cm from the target.

After irradiation and cooling, the γ-activities of the irradiated ThO_2 and Al samples were measured off-line by a coaxial GMX60 HPGe detector (ORTEC, made in USA) with a relative efficiency of 68% and an energy resolution of 1.82 keV FWHM at 1.33 MeV. The efficiency was previously calibrated with a standard ^{152}Eu source. The measurements were repeated several times to follow the decay of the product radionuclides and make sure the dead time was less than 5%. A typical γ-ray spectrum from the irradiated ThO_2 sample is given in Fig. 1, where the γ-rays of interest have been marked.

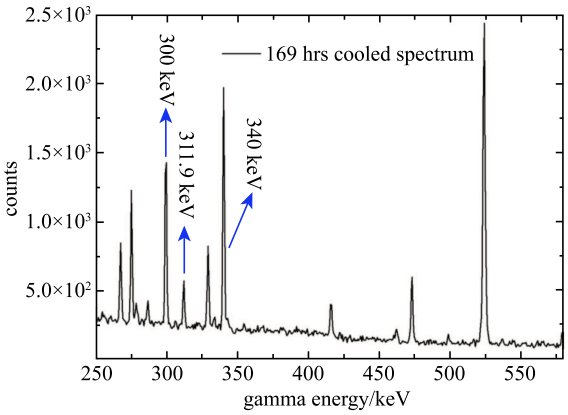

Fig. 1. (color online) Gamma ray spectrum of irradiated ThO_2 showing the γ-ray energy of ^{233}Pa.

3 Determination of ^{232}Th (n,γ)^{233}Th reaction cross-sections

The decay parameters of the product radioisotopes used in this work were taken from Refs. [11, 12] and are summarized in Table 1.

As shown in the Table 1, the half-life of the isotope ^{233}Th is 21.83 minutes, which decays to ^{233}Pa ($T_{1/2}$=26.975 d) within 3 hours. In view of this, the ^{232}Th(n,γ)^{233}Th cross-section can be calculate from the observed photo-peak activity of ^{233}Pa within the long cooled γ spectrum. In the calculation, the photo-peak activity of the 311.904 keV γ-line was selected instead of 300.129 keV and 340.476 keV. This is to avoid the interference of the 300.1 keV γ-line of ^{212}Pb, which is the

Table 1. Reactions and associated decay data of activation products.

reaction	abundance of target isotope (%)	half-life of product $T_{1/2}$	γ-ray energy E_γ/keV	γ-ray intensity I_γ(%)
^{232}Th (n,γ)^{233}Th	100	21.83 m	86.48	2.76
$\rightarrow$ ^{233}Pa		26.975 d	300.129	6.63
			311.904	38.5
			340.476	4.45
^{27}Al(n, α)^{24}Na	100	14.951 h	1368.63	100

α decay product from ^{232}Th, and reduce the statistical error of 340.476 keV because of its low γ-ray intensity.

The following activation formula was used to determine the measured ^{232}Th(n,γ)^{233}Th cross sections, as proposed by X.Z. Kong et al. [13]:

$$\sigma_{\rm x}=\frac{[\eta\varepsilon I_\gamma mKSD]_{\rm m}[\lambda FCA]_{\rm x}}{[\eta\varepsilon I_\gamma mKSD]_{\rm x}[\lambda FCA]_{\rm m}}\sigma_{\rm m}, \quad (1)$$

where the footnotes m and x represent the terms of the monitor reaction and the measured reaction, respectively. $\sigma_{\rm m}$ is the cross section of the monitor reaction, ε is the full-energy peak efficiency of the measured characteristic γ-ray, I_γ is the γ-ray intensity, η is the abundance of the target nuclide, m is the mass of the sample, $S=1-{\rm e}^{-\lambda T}$ is the growth factor of the residual nuclide, λ is the decay constant, and T is the total irradiation time. $D={\rm e}^{-\lambda t_1}-{\rm e}^{-\lambda t_2}$ is the counting collection factor, t_1 and t_2 are time intervals from the end of the irradiation to the start of counting and end of counting, respectively, A is the atomic weight, C is the measured full-energy peak area and F is the total correction factor of the activity:

$$F=F_{\rm s}\times F_{\rm c}\times F_{\rm g} \quad (2)$$

where F_s, F_c and F_g are the correction factors for the self-absorption of the sample at a given γ-energy, the coincidence sum effect of cascade γ-rays in the investigated nuclide and in the counting geometry, respectively.

K is the neutron fluency fluctuation factor,

$$K = \left[\sum_{i}^{L} \Phi_i (1-\mathrm{e}^{-\lambda \Delta t_i})\mathrm{e}^{-\lambda T_i}\right] / \Phi S \qquad (3)$$

In this calculation, we divided the total irradiation time into L parts. L is the number of time intervals into which the irradiation time is divided, ΔT_i is the duration of the i^{th} time interval, T_i is the time interval from the end of the i^{th} interval to the end of irradiation, Φ_i is the neutron flux averaged over the sample during ΔT_i, and Φ is the neutron flux averaged over the sample during the total irradiation time T.

Corrections were also made for γ-ray self-absorption in the sample, for γ-ray coincidence summing effects, for fluctuation of the neutron flux during the irradiation and for sample geometry. The uncertainties in our results mainly include the counting statistics (10%–15%), detector efficiency (~3%), neutron energy and fluency uncertainties (~2.5%), monitor reaction cross section (~1.5%), weight of samples (<0.1%), self-absorption of γ-ray (~1%), coincidence summing effect of cascade γ-rays (~1%), sample geometry (~1%), and the irradiation, cooling and measuring times (<0.8%), etc. The overall uncertainty was calculated using the quadratic method.

4 Results and discussion

The cross sections of the ^{232}Th(n,γ)^{233}Th reaction measured in the present work and the values around neutron energy 14 MeV given in the literature are summarized in Table 2 together with the cross section of the monitor reaction ^{27}Al(n,α)^{24}Na. In the calculations, the cross sections for the ^{27}Al(n,α)^{24}Na monitor reaction were obtained by interpolating the evaluated values from the literature [12].

Table 2. Summary of the cross sections.

reaction	cross-sections (in mb) at various neutron energies		
	E_n = 14.1±0.2 MeV	E_n=14.5 MeV	E_n=14.8±0.2 MeV
^{232}Th(n,γ)^{233}Th	2.320±0.378		2.201±0.328
		5.2±0.624 [20]	1.569±0.257 [19]
^{27}Al(n,α)^{24}Na	121.6±0.6		111.9±0.5

It can be seen from Table 2 that our measured results are in good agreement with the data from Ref. [19] but much lower than in Ref. [20]. For comparison, the ^{232}Th(n,γ)^{233}Th reaction cross sections from the present work and similar experimental data from Refs. [21–27] are plotted in Fig. 2 together with the evaluated data [4–6].

Theoretically, the excitation function of the ^{232}Th(n,γ)^{233}Th reaction cross sections at different neutron energies from 1 MeV to 20 MeV was calculated using the computer code TALYS, version 1.6. [14, 15]. The TALYS-1.6 code system is able to analyze and predict nuclear reactions based on physics models and parameterizations. It can calculate nuclear reactions involving neutrons, photons, protons, deuterons, tritons, ^{3}He and alpha-particles, in the 1 keV–200 MeV energy range and for target nuclides of mass 12 and heavier. For the ^{232}Th target, the optical model parameters for neutrons were obtained by a local potential proposed by Koning and Delaroche [16]. Similarly, the compound nucleus contribution was calculated by the Hauser– Feshbach model [17]. The two- component exciton model developed by Kalbach [18] was used for calculating the pre-equilibrium contribution. The default values were used for parameters concerning nuclear masses, ground-state deformations, discrete levels, decay schemes, and strength functions. Meanwhile, all the possible outgoing channels for a given neutron energy were considered, including inelastic and fission channels.

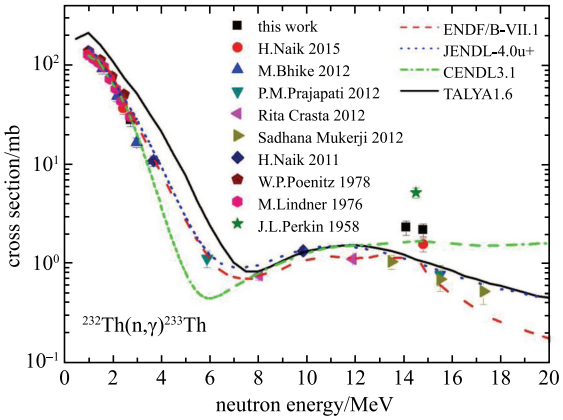

Fig. 2. (color online) Plot of the experimental, evaluated and theoretical calculated ^{232}Th(n,γ)^{233}Th reaction cross sections as a function of the neutron energy from 1 MeV to 20 MeV.

As shown in Fig. 2, the ^{232}Th(n,γ)^{233}Th reaction cross sections show a sharp decreasing trend from 1 MeV

to 7.5 MeV and then increasing up to the neutron energy of 12 MeV. This dip is mainly due to the opening of the (n,2n) and (n,nf) reaction channels . It can also be seen from Fig. 2 that the experimental cross sections of ^{232}Th within the neutron energy range 14.1–14.8 MeV are significantly higher than the calculated results from TALYS and evaluated data from ENDF/B-VII.1, JENDL-4.0u+, but not the evaluated data of CENDL-3.1.

5 Conclusions

We have measured the activation neutron capture cross sections of ^{232}Th at neutron energies of 14.1 and 14.8 MeV using the off-line gamma ray spectrometric technique. During the work we used the latest nuclear data for decay characteristics of the product radionuclide. The excitation function of the ^{232}Th(n,γ)^{233}Th reaction was also calculated using computer code TALYS 1.6 and compared with the recent evaluated data of ENDF/B-VII.1, JENDL-4.0u+ and CENDL-3.1. Our results are useful for verifying the accuracy of nuclear models used in the calculation of cross sections and also be useful for the design, evaluation and construction of nuclear reactors based on the ^{232}Th-^{233}U fuel cycle.

We would like to thank the Intense Neutron Generator group at China Institute of Atomic Energy for performing the irradiations.

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