Measurement of 232Th(n,2n)231Th reaction cross-sections at neutron energies of 14.1 MeV and 14.8 MeV using neutron activation method (Postprint)

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Abstract

In this study, the activation cross-sections were measured for 232Th(n,2n)231Th reactions at neutron energies of 14.1 and 14.8 MeV, which were produced by a neutron generator through a T(d,n)4He reaction. Induced gamma-ray activities were measured using a low background gamma ray spectrometer equipped with a high resolution HPGe detector. In the cross-section calculations, corrections were made regarding the effects of gamma-ray attenuation, dead-time, fluctuation of the neutron flux, and low energy neutrons. The measured cross-sections were compared with the literature data, evaluation data (ENDF-B/VII.1, JENDL-4.0 and CENDL-3.1), and the results of the model calculation (TALYS1.6).

Full Text

Preamble

Measurement of 232Th(n,2n)231Th Reaction Cross-Sections at Neutron Energies of 14.1 MeV and 14.8 MeV Using Neutron Activation Method

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Abstract: In this study, activation cross-sections were measured for the 232Th(n,2n)231Th reaction at neutron energies of 14.1 and 14.8 MeV, produced by a neutron generator through the T(d,n)4He reaction. Induced gamma-ray activities were measured using a low-background gamma-ray spectrometer equipped with a high-resolution HPGe detector. In the cross-section calculations, corrections were made for gamma-ray attenuation, dead-time, neutron flux fluctuation, and low-energy neutrons. The measured cross-sections were compared with literature data, evaluated data (ENDF-B/VII.1, JENDL-4.0, and CENDL-3.1), and model calculation results (TALYS1.6).

Keywords: 232Th(n,2n)231Th reaction, Cross-section, Neutron activation method
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Introduction

Accurate knowledge of neutron-induced reaction cross-sections around 14 MeV is essential for the design of fusion reactors, molten salt reactors, hybrid subcritical systems, accelerator-driven subcritical systems (ADSs), and nuclear transmutation applications, as well as for validating and improving nuclear reaction models. Since thorium-added MOX-fueled subcritical hybrid reactors are intended for energy production, the cross-section of the Th(n,2n) reaction plays a key role in the Th-U nuclear fuel cycle, particularly for neutron balance calculations due to its relatively high (n,2n) reaction cross-section. Despite its importance, measurements of the 232Th(n,2n)231Th reaction cross-section by various laboratories show relatively large disagreements and uncertainties. Furthermore, discrepancies among evaluated data files (ENDF/B-VII.1, JEFF-3.2, JENDL-4.0, CENDL-3.1) can reach 30–50% in the 13–15 MeV energy range. Currently, the reaction cross-section is required within an accuracy of 1–2% for safe use in simulation techniques predicting the dynamic behavior of complex arrangements in thorium molten salt reactors (TMSR). Therefore, further precise and accurate measurements are necessary to strengthen database reliability. In the present work, 232Th(n,2n)231Th reaction cross-sections were measured at neutron energies of 14.1 and 14.8 MeV using the activation technique. The measured results are discussed and compared with literature data, evaluated database values, and results from TALYS 1.6 model calculations.

Experimental Procedures

The experiment was conducted using the CPNG-600 neutron generator at the China Institute of Atomic Energy (CIAE). Neutrons with a yield of approximately 1.5×10^{10} n/4πs were produced via the T(d,n)4He reaction. The ion beam current reached up to 300 μA with an effective deuteron energy of 300 keV. A solid tritium-titanium (T-Ti) target with a thickness of 1.0 mg/cm^2 was used in the generator. During irradiation, neutron yield variation was monitored by accompanying α-particles to enable corrections for neutron flux fluctuations. The schematic of the accompanying α-particle monitor was identical to that shown in Ref. [3]. The Au-Si surface barrier detector in the 135° accompanying α-particle tube was positioned 110 cm from the target.

Thorium dioxide powder of 99.7% purity (from China North Nuclear Fuel Corporation Limited) was pressed into circular thin samples with a diameter of 20 mm. Two ThO_2 targets with thicknesses of 1.05 mm and 1.07 mm were used, each placed between two natural aluminum foils of the same diameter. All Al foils had purities better than 99.99% and a thickness of 0.06 mm. The samples were positioned at 0° and 90° angles relative to the deuteron beam direction, centered about the T-Ti target at a distance of approximately 3.5 cm. Neutron energies at these positions were calculated using the Q-equation [4] and verified through cross-section ratio methods for the 90Zr(n,2n)89m+gZr and 93Nb(n,2n)92mNb reactions [5, 6] before irradiation. The determined neutron energies were (14.1 ± 0.2) MeV and (14.8 ± 0.2) MeV, respectively.

The radioactivity of each activated product was determined via low-background γ-ray spectroscopy using a coaxial GMX60 HPGe detector (ORTEC, USA) with a relative efficiency of 68% and an energy resolution of 1.82 keV FWHM at 1.33 MeV. The distance between the sample and detector was 5 cm. Efficiency calibration was performed using point-like calibrated gamma-ray sources. The decay characteristics of product radioisotopes and natural abundances of target isotopes are summarized in Table 1 [7, 8]. Count rates were corrected for background neutron contributions, sample self-absorption, and cascade γ-ray pile-up and coincidence losses. Uncertainties primarily included counting statistics (1-2%), detection efficiency (2%), sample mass (<0.1%), neutron energy and fluence uncertainties (2.5%), γ-ray self-absorption (0.5%), and irradiation, cooling, and measuring times (<0.8%).

Experiment Data Deduction and Nuclear Model Calculation

Cross-sections of the 27Al(n,α)24Na reaction were selected as monitors for measuring the 232Th(n,2n)231Th reaction cross-section. The measured cross-sections, σ , were calculated using the activation formula [9]:

$$\frac{[\eta\varepsilon I_\gamma mKSD]_m[\lambda FCA]_x}{[\eta\varepsilon I_\gamma mKSD]_x[\lambda FCA]_m}$$

where σ represents the cross-section;   is the abundance of the target nuclide;   is the full-energy peak efficiency of the measured characteristic γ-ray; Iγ is the γ-ray intensity; m is the sample mass; K is the neutron fluence fluctuation factor; S = 1 - e^(-λT) is the growth factor of the residual nuclide, λ is the decay constant, and T is the total irradiation time; D = e^(-λt_1) - e^(-λt_2)

is the counting collection factor, where t_1 and t_2 are time intervals from the end of irradiation to the start and end of counting, respectively; F is the total correction factor for activity; C is the measured full-energy peak area; and A is the atomic weight. The subscripts m and x represent terms for the monitor reaction and measured reaction, respectively.

In the equation, Fs, Fc, and Fg are correction factors for sample self-absorption at a given γ-energy, coincidence sum effects of cascade γ-rays in the investigated nuclide, and counting geometry, respectively. The total irradiation time was divided into L parts, where L is the number of time intervals, ΔTi is the duration of the ith interval, Ti is the time from the end of the ith interval to the end of irradiation, Φi is the neutron flux averaged over the sample during ΔTi, and Φ is the neutron flux averaged over the sample during the total irradiation time T.

The measured 232Th(n,2n)231Th cross-sections are presented in Table 2 together with the monitor reaction 27Al(n,α)24Na cross-sections [10]. In this work, the excitation function for 232Th(n,2n)231Th reaction cross-sections from threshold to 20 MeV was calculated theoretically using the TALYS 1.6 computer code [11, 12]. The TALYS-1.6 code system analyzes and predicts nuclear reactions based on physics models and parameterizations. It calculates nuclear reactions involving neutrons, photons, protons, deuterons, tritons, ^{3}He, and α-particles in the 1 keV–200 MeV energy range for target nuclides with mass numbers of 12 and heavier. For the 232Th target, default optical model parameters for neutrons and all possible outgoing channels for a given neutron energy were considered, including inelastic and fission channels [13].

Results and Discussion

Cross-sections of the 232Th(n,2n)231Th reaction at neutron energies of 14.1 and 14.8 MeV were obtained relative to the 27Al(n,α)24Na reaction. In calculations, the monitor reaction cross-sections were obtained by interpolating evaluated literature values. The measured cross-sections and monitor reaction values are summarized in Table 2. Our values were measured with approximately 9.5% uncertainty.

Figure 1 [Figure 1: see original paper] shows the 232Th(n,2n)231Th reaction cross-section along with literature results [14–29]. The experimental cross-sections exhibit a sharp increase from threshold to 8.0 MeV neutron energy, then remain constant up to 13.6 MeV. Above 13.6 MeV, cross-sections decrease, primarily due to opening of (n,3n) and (n,2nf) reaction channels.

Based on the D-T neutron generator capabilities, experimental measurements were performed in the 13–15 MeV range. Accordingly, measured cross-sections from this work and literature data from EXFOR [30] are plotted in Figure 2 [Figure 2: see original paper] for the 12.5–15.8 MeV range, along with evaluated data [31–33] and TALYS1.6 theoretical values. Around 14 MeV, our results agree well with those of Reyhancan et al. [17], Karius et al. [23], and Butler et al. [25]

within experimental uncertainties. Figure 2 shows that ENDF-B/VII.1 and CENDL-3.1 evaluations agree well with our values, while TALYS1.6 calculations overpredict the experimental 232Th(n,2n)231Th cross-section, except for data from Filatenkov et al. [20].

[Figure 1: see original paper]

[Figure 2: see original paper]

Conclusion

In this work, activation cross-sections for the 232Th(n,2n)231Th reaction were obtained at neutron energies of 14.1 MeV and 14.8 MeV with considerably improved accuracy. Our measurements generally agree with recent literature data, though some discrepancies exist among literature values, possibly attributable to variations in neutron flux measurement methods, sample characteristics, or nuclear parameters used. The results were compared with previously reported cross-sections and TALYS-1.6 nuclear model calculations. These new results are valuable for verifying the accuracy of nuclear models used in cross-section calculations and for practical applications.

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