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Measurement of ^{232}Th(n,5nγ) cross sections from 29 to 42 MeV

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Abstract. The excitation function of the reaction ^{232}Th(n, 5nγ)^{228}Th from 29 to 42 MeV has been measured for the first time at the quasi-monoenergetic neutron beam of the UCL cyclotron CYCLONE employing the ^{7}Li(p,n) source reaction. Taking advantage of the good energy resolution of the planar High Purity Germanium (HPGe) detectors, prompt γ-ray spectroscopy was used to detect the γ rays resulting from the decay of excited states of nuclei created by the (n,xn) reactions. The neutron beam was characterized by a combination of time of flight measurements carried out using a liquid scintillation detector and a ^{238}U fission ionization chamber and fluence measurements carried out using a proton recoil telescope. The preliminary results are compared with calculations performed using the TALYS-0.72 code.

1 Introduction

1.1 Motivations

In contrast to the uranium cycle, which was well studied during the development of conventional reactors, the experimental and evaluated data bases required for the innovative thorium cycle are more incomplete. In order to determine the characteristics and performance of this cycle, a correct description of nuclear properties (level densities, fission barriers, etc.) is needed since reliable calculations of nuclear reaction cross sections require this data.

In this context, the excitation function of the reaction ^{232}Th(n, 5n γ)^{228}Th was measured from 29 to 42 MeV for the first time. This work was motivated by the fact that the γ-ray production in (n,xn γ) reactions allows the test of models of the level densities at high excitation energies. These quantities are crucial for the prediction of the exclusive γ-ray production. Cross sections for (n,xn) reactions are also of interest for the determination of fission barriers along the whole ^{232}Th–^{228}Th reaction chain since a strong competition exists between neutron emission and fission. The present measurement will help describe this competition and thus extend the fission barrier database in this region.

1.2 Experimental method

The present work is part of a series of measurements of (n,xn γ) cross sections performed using the in-beam gamma-ray spectroscopy method at different neutron source facilities [1]. This method consists in detecting the γ radiation from the decay of the excited nucleus created by the (n,xn) reaction using High-Purity Germanium (HPGe) detectors. At the GELINA facility of IRMM (Geel, Belgium) [2] which delivers a "white" neutron beam, this technique was combined with time-of-flight (TOF) measurements in order to determine the incident neutron energy. The cross section ^{206,207,208}Pb(n, xn γ) for x = 1 and 2 was measured with a flight path of 200 m [3,4]. At present, measurements of the (n,xn γ) cross sections of uranium isotopes (^{235,238}U) are carried out using a flight path of 30 m.

At the neutron beam facility CYCLONE [5], the cyclotron of the Université Catholique de Louvain (UCL) in Louvain-la-Neuve (Belgium) delivers quasi-monoenergetic neutron beams. There the flight path of 3.28 m is too small to use the TOF technique to discriminate between neutrons from the monoenergetic peak and from the continuum. Thus only spectrum-averaged cross sections can be directly measured and deconvolution procedures have to be applied to derive cross sections for the monoenergetic neutrons from the high-energy peak.

2 Experimental set-up

2.1 Fast neutron beam at the CYCLONE neutron beam facility

The quasi-monoenergetic neutrons beams used for the present experiment were produced by bombarding a ^{nat}Li target with 32, 36.5, 38, 40 and 44 MeV protons. The nominal target thickness was 3 mm. An iron collimator 2 m in length and with an opening angle of 0.24 degrees was employed to produce a collimated neutron beam with neutron emission angles centred

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around 0 degree. The spectral neutron distribution exhibits a prominent high-energy peak resulting from transitions to the ground state and the first excited state ($E_x = 0.43$ MeV) of ^{7}Be and a broad low-energy continuum from ^{7}Li(p,nx) break-up reactions.

The beam characterisation was carried out using a gain-stabilised BC501 liquid scintillation detector, a parallel-plate ^{238}U fission ionisation chamber (UFC) and a recoil proton telescope (RPT). A second ^{238}U transmission ionisation chamber was used as monitor detector. The stability of this counter was checked with a thin NE102 transmission detector and a beam current integrator.

The relative spectral fluence (Φ_E/Φ) was measured using the BC501 detector and the TOF method at beam currents of 20–50 nA. The beam pulse selector available at the CYCLONE facility could only be used for the 36.5 MeV proton beam for a TOF measurement with a reduced repetition rate of 2 MHz and a low frame-overlap threshold of about 2 MeV. At the other beam energies, an experimental response matrix available for this detector was used to disentangle the effect of frame overlap in the TOF distributions and to determine (Φ_E/Φ) above 8 MeV. Figure 1 shows the spectral distribution of all neutron beams used for the present experiment.

The normalisation of the relative spectral fluence (Φ_E/Φ) was determined relative to the ^{238}U(n,f) cross section [6] by TOF measurements using the UFC at distances of 5.45 m and 9.79 m from the Li target and at beam currents of about 5 μA. The fluence (Φ_0/M) of neutrons in high-energy peak per monitor count M measured at the larger distance was compared to the peak fluence Φ_0 measured using the RPT at a distance of 5.64 m. The average ratio of the peak fluence measured with the RPT and the UFC was (0.925 ± 0.019) which is consistent with the ratio of neutron fluences measured earlier using the same instruments [7].

At a distance of 5.45 m, the full beam was covered by the ^{238}U layers. Thus the total number (N_0/M) of neutrons in the high-energy peak per monitor count could be determined as required for the analysis of the measurements of the photon emission from the ^{232}Th sample which was also larger than the beam size.

In addition, these measurements were used to correct the relative spectral fluence measured with the BC501 detector for slight increases in the ratio of the low-energy continuum relative to the high-energy peak observed when the beam current was increased to the maximum value. At most, these corrections amounted to 7%.

2.2 Gamma detection

The thorium sample (6 × 6 cm^2, 12.6 g) was placed 3.28 m downstream the neutron source and oriented at 40° with respect to the beam direction to minimize the absorption effects. For the gamma detection, two High Purity Germanium planar detectors placed at 110° and 150° were used. At these backward angles the background from neutrons and gamma is lower. Figure 2 shows the neutron beam facility and the gamma detection set-up.

With this set-up is was possible to identify three gamma transitions between the lowest levels of the ground-state rotational band of ^{228}Th formed by the ^{232}Th(n, 5n) reaction: $4^+ \to 2^+(E_\gamma = 129.065\,\text{keV}), 6^+ \to 4^+(E_\gamma = 191.349\,\text{keV})$ and $8^+ \to 6^+(E_\gamma = 244.3\,\text{keV})$. Unfortunately the $4^+ \to 2^+$ transition and, to a lesser extent, the $6^+ \to 4^+$ transition are also fed by the decay of the ^{228}Ac from the ^{232}Th radioactive decay chain. Therefore background measurements without beam were carried out to subtract this contribution.

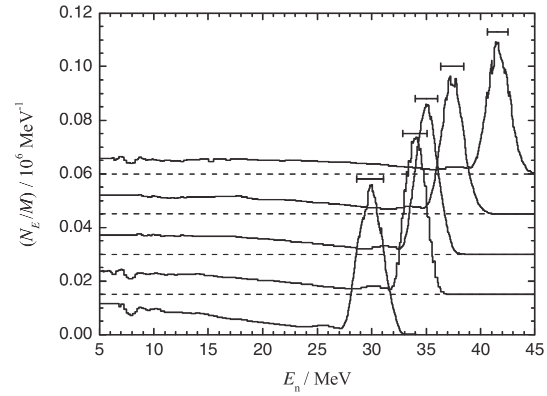

Fig. 1. Spectral number of neutrons N_E per monitor count M for the neutron beams produced by 32, 36.5, 38, 40 and 44 MeV protons incident on a ^{nat}Li target with a nominal thickness of 3 mm. The shape of the high-energy peak reflects the time resolution of the BC501 detector and the pulse duration of the proton beam. The intrinsic width as calculated from the nominal target thickness is shown by the horizontal error bars.

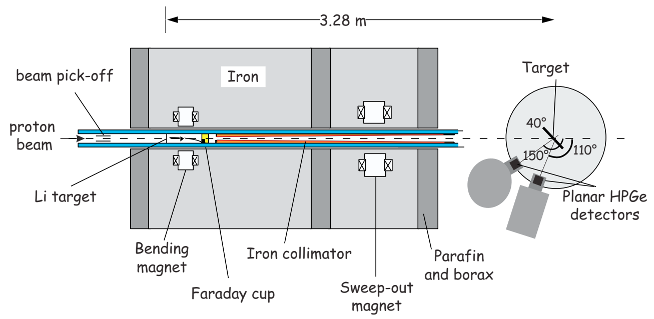

Fig. 2. Schematic view of the neutron beam facility and the γ-detection system used for the ^{232}Th(n, 5nγ) cross section measurements.

3 Data analysis

Figure 3 shows a portion of the prompt gamma spectrum without background subtraction measured using a 38 MeV proton beam (mean energy of the neutrons in the high-energy peak 35.03 MeV), during an accumulation time of 9 hours. One can see the three interesting gamma transition peaks in ^{228}Th. For the actual data analysis, only runs with the lower background conditions were used. Consequently, the effective accumulation time used for each neutron energy amounts to about 6 hours.

The general angular distribution of the emitted photons can be written as

$$\frac{d\sigma}{d\Omega}(\theta, E_n) = \frac{1}{4\pi}\frac{n_\gamma}{\eta_T \cdot N}, \tag{1}$$

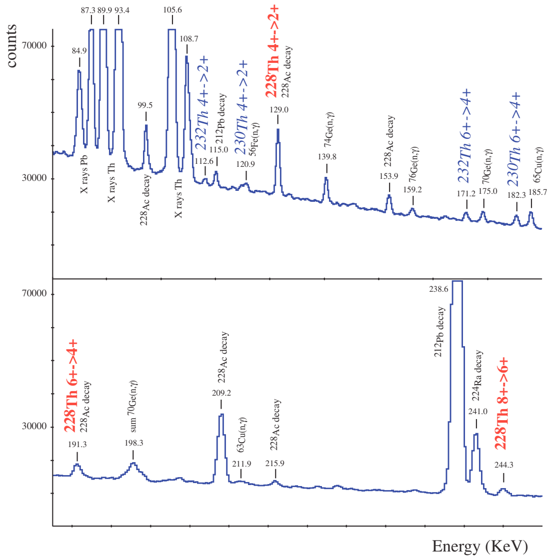

Fig. 3. Portion of the gamma spectrum obtained for $E_p = 38$ MeV (high-energy neutron peak at $E_0 = 35.03$ MeV) and an accumulation time of 9 h. The three interesting γ-rays in ^{228}Th and the peaks corresponding to transitions in ^{232}Th and ^{230}Th are indicated in bold and italic, respectively. The latter γ-rays are produced by the ^{232}Th(n,n') and ^{232}Th(n,3n) reactions.

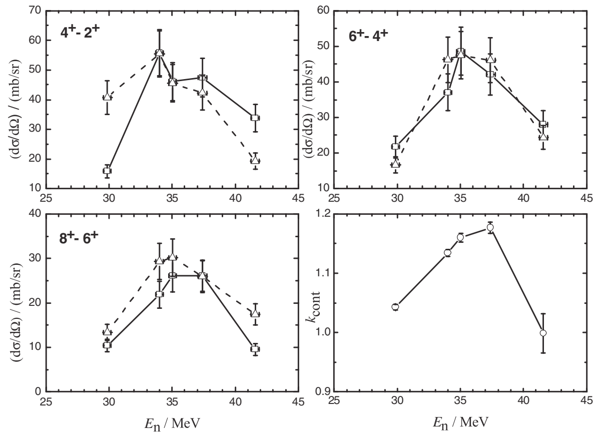

Fig. 4. Excitation functions at the emission angles $\theta_1 = 110°$ (squares, solid lines) and $\theta_2 = 150°$ (triangles, dashed lines) for the three transitions 129 keV (top, left), 191 keV (top, right) and 244 keV (bottom, right). The lower right panel shows the correction factor k_{cont} for the influence of the continuum neutrons.

where n_γ is the rate of photons with emission angle θ and energy E_γ for isotropic emission, η_T is the number of ^{232}Th nuclei in the sample unit area and N the rate of neutrons with energy E_{n} incident neutrons on the thorium sample.

The photon rate n_γ was determined from the integral count rate in the full-energy peak and corrected for ambient background, detection efficiency and dead-time losses. The detection efficiency of the two detectors was calculated using GEANT 3 [8] simulations including the size of the beam spot and the thickness of the thorium sample. The results of the simulations were validated by efficiency measurements performed with a calibrated ^{152}Eu source between the neutron runs. The deviation of the measured and calculated efficiencies was less than a few %.

The multipolarity of the gamma transitions used in the present experiment is pure E2. Thus the total photon emission cross section can be calculated from the differential cross section measured at two selected photon emission angles [9]

$$\sigma(E_{\mathrm{n}}) = \int_{4\pi} \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\,\mathrm{d}\Omega = 2\pi\left(w_1 \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(\theta_1, E_{\mathrm{n}}) + w_2 \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(\theta_2, E_{\mathrm{n}})\right) \tag{2}$$

where $w_1 = 0.695$ and $w_2 = 1.305$ denote the weighting factors for the emission angles $\theta_1 = 30°$, $150°$ and $\theta_2 = 70°$, $110°$ calculated from the Legendre expansion of the E2 angular distribution.

The experimental results yield only spectrum-averaged cross sections

$$\bar{\sigma} = \int_{E_{\mathrm{th}}}^{E_{\max}} \sigma(E_{\mathrm{n}}) \left(\frac{N_E}{N_{\mathrm{th}}}\right) \mathrm{d}E_{\mathrm{n}} = k_{\mathrm{cont}}^{-1} \sigma(E_0), \tag{3}$$

where E_0 is the energy of the high-energy peak and (N_E/N_{th}) denotes the spectral neutron rate normalized to the integral rate N_{th} of neutrons with energies above the threshold $E_{\mathrm{th}} = 23.9$ MeV for the ^{232}Th(n, 5nγ) reaction. To determine the correction factor k_{cont} for the effect of the continuum neutrons, a deconvolution procedure has to be used.

For this purpose, it was assumed that the shape of the cross section resembles that predicted by the TALYS code [10] (see below), i.e. that it can be approximated by a Gaussian distribution which is continuously matched to an exponential distribution on the high-energy side. The prior information for this parameterisation was taken from the TALYS result and the posterior distributions of the parameters given the experimental spectrum-averaged cross sections were determined using Bayesian parameter estimation programmed with the WinBUGS software [11, 12] which employs the Markov chain Monte Carlo method to evaluate Bayes' equation. To reduce the number of free parameters, the same Gaussian width and centroid energy as well as the matching energy for the exponential was used for the three transitions. Furthermore, the ratio of the maximum cross sections for the three transitions was set equal to the TALYS result and treated as a constant. Thus the correction factor k_{cont} depends on E_0 only and not on the specific transition. For reasons explained below, the experimental data for the $4^+ \to 2^+$ transition were not included in this analysis.

4 Preliminary results and model calculations

The spectrum-averaged differential photon emission cross sections for the three transitions and the two detection angles are shown in figure 4 together with the correction factor k_{cont}.

Figure 5 shows the corrected experimental integral cross sections $\sigma(E_0)$ together with the TALYS-0.72 calculations performed with default parameters and the results of the deconvolution using the present experimental spectrum-averaged cross sections.

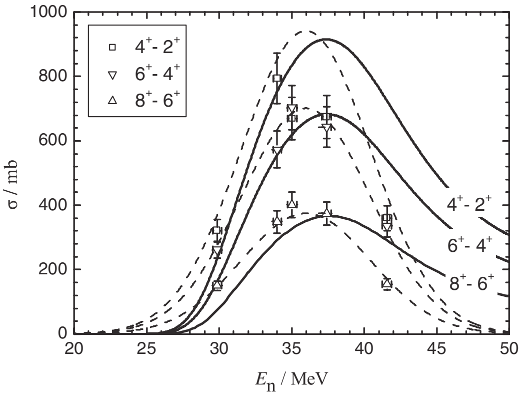

Fig. 5. The total experimental excitation functions of the ^{232}Th(n, 5n γ) reactions obtained in our experiment (symbols) are compared to TALYS-0.72 calculations with default parameters (solid lines). The dashed lines show the cross sections determined using the deconvolution procedure. The horizontal error bars indicate the uncertainty of the mean energy E_0 of the high-energy peak.

The present experimental results exhibit a shift of the low-energy slope of the ^{232}Th(n, 5n γ) cross sections by about -1.3 MeV compared with the TALYS result. Furthermore, the experimental results for $E_0 = 41.6$ MeV indicates a significantly steeper decrease of the cross section on the high-energy side. However, more data in this energy region would be necessary to underpin this conclusion. The $4^+ \rightarrow 2^+$ transition shows a striking behaviour at the three higher neutron energies. This could be caused by deficiencies in subtraction of the ^{228}Ac decay component but further study is required to reveal the reason for the discrepancy. As the TALYS calculations have been made with default parameters, improvements will for example be possible by updating the level density parameters. Moreover, the (n,6n) and (n,7n) channels have to be checked in TALYS since they seem to be underestimated. This could explain the observed differences in the behaviour of the excitation function at high energies between TALYS and experimental data.

5 Conclusions

The ^{232}Th(n, 5nγ) cross sections was measured in the neutron energy range from 29 MeV to 42 MeV for the first time. Preliminary results presented in this contribution are compared to TALYS calculations and show discrepancies in the position of the maximum of the excitation function and in the decrease of the excitation function for higher neutron energies. For the experimental part, more investigations, in particular on the background subtraction procedure, are still needed to arrive at final conclusions. For the TALYS code, improvements of thorium spectroscopic parameters are needed.

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