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Measurement of (n,xnγ) Reactions of Interest for the New Nuclear Reactors

J. C. Thiry,[1,*] C. Borcea,[2] P. Dessagne,[1] J. C. Drohé,[3] E. Jericha,[4] H. Karam,[1] M. Kerveno,[1] A. J. Koning,[5] A. Negret,[2] A. Pavlik,[6] A. Plompen,[3] C. Rouki,[3] G. Rudolf[1] and M. Stanoiu[2,3]

[1] *Université de Strasbourg, IPHC, 23 rue du Loess 67037 Strasbourg, France and CNRS, UMR7178, 67037 Strasbourg, France*

[2] *Horia Hulubei National Institute of Physics and Nuclear Engineering, Str. Atomistilor 407, 077125 Bucharest - Magurele, Romania*

[3] *Institute for Reference Materials and Measurements, Retieseweg 111, B-2440 Geel, Belgium*

[4] *Technische Universität Wien, Atominstitut der Österreichischen Universitäten, Stadionallee 2, 1020 Wien, Austria*

[5] *Nuclear Research and Consultancy Group, Westerduinweg 3, NL-1755 ZG Petten, The Netherlands*

[6] *Universtität Wien, Fakultät für Physik, Währinger Str. 17, 1090 Wien, Austria*

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The design of Generation IV nuclear reactors and the research of new fuel cycles require knowledge of the cross sections of different nuclear reactions. Our research is focused on cross section measurements of (n,xnγ) reactions occurring in these new reactors. The aim is to measure unknown cross sections and to reduce the uncertainty on present data relative to reactions and isotopes present in transmutation or regeneration processes. The current work consists of studying ^{232}Th(n,n'γ) and ^{235}U(n,xnγ) reactions in the fast neutron energy domain (up to 20 MeV). The experiments are performed at the GEel LINear Accelerator (GELINA) which delivers a pulsed, white neutron beam at IRMM, Belgium. The time characteristics of the beam enable us to measure neutron energies with the time of flight (TOF) technique. The neutron induced reactions (in this case inelastic scattering and (n,2n) reactions) are identified by online prompt γ spectroscopy with an experimental setup including 4 HPGe detectors. A double layered fission chamber is used to monitor the incident neutron flux. The experimental setup and analysis methods will be presented and a comparison between the obtained cross sections and the TALYS predictions will be discussed. This work is a first step in the preparation of the measurement of ^{233}U(n,xnγ) reactions, which are completely unknown at this stage although of very high importance in the ^{232}Th regeneration process.

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I. INTRODUCTION

Precise knowledge of (n,xnγ) reactions is a key issue in present day's reactor development studies. The new Generation IV nuclear reactors explore new energy domains, and imply reaction rates unknown at this stage.

On one side, (n,xn) reactions are crucial for the design of new reactors. They are an important energy loss mechanism which has to be taken into account in the calculations of new reactors as they lead to neutron multiplication and production of radioactive isotopes.

On the other side, (n,xnγ) reactions allow to validate theoretical codes, such as TALYS [1], and to verify the level density models. These are fundamental for the prediction of γ ray productions. Theoretical knowledge of (n,xn) reactions also implies a good knowledge of fission parameters as a strong competition exists between neutron emission and fission in the studied nuclei.

The presented work is performed using the (n,xnγ) technique, for which a high precision experimental setup was designed. It has already been used to measure (n,xnγ) reactions on isotopes such as ^{235}U, ^{232}Th, and ^{nat}W.

II. EXPERIMENTAL SETUP

This section treats the applied measurement techniques as well as the experimental setup, shown in Fig. 1.

1. The (n,xnγ) technique

A sample enriched in ^{A}X isotopes is irradiated by a neutron beam, inducing (n,xn) reactions. This leads to production of $^{A-(x-1)}$X isotopes in excited states. Decay of these isotopes leads to emission of characteristic γ rays, witnessing a prior reaction. They yield the cross section of isotope production in a given excited state.

*E-mail: jean-claude.thiry@ires.in2p3.fr

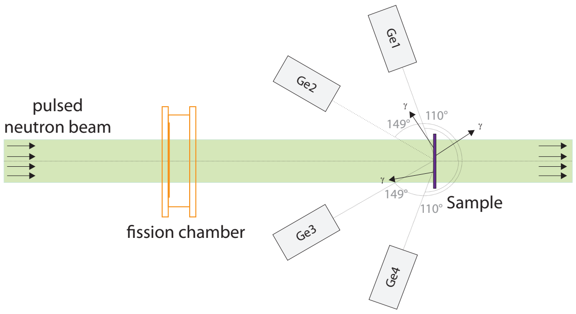

Fig. 1. (Color online) Experimental setup used at GELINA, FP16/30 m.

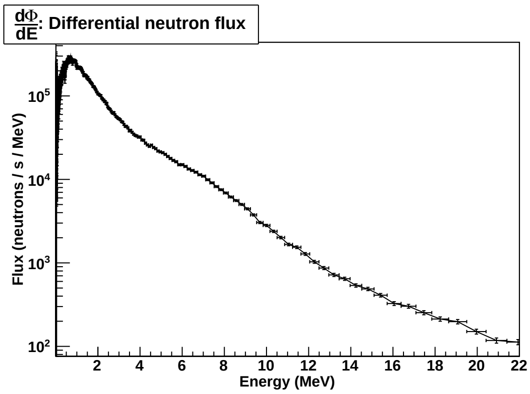

Fig. 2. Differential neutron flux measured at FP16/30 m at GELINA.

2. The TOF technique

The experiment is realized at the GEel LINear Accelerator (GELINA) facility at the Institute for Reference Materials and Measurements (IRMM), Belgium. GELINA produces a white, pulsed neutron beam using the (γ,F) and (γ,xn) reactions on a depleted Uranium target which leads to an incident flux spectrum from a few keV up to several MeV. The pulsed beam enables energy separation of the incident neutrons using a time spectrum, which can be calibrated thanks to the presence of a γ-flash. The experimental setup is located 30 m away from the neutron source. The data acquisition resolution being 10 ns, this flight path is the best compromise between time resolution and flux intensity, allowing a resolution of 1 MeV at a neutron energy of 20 MeV.

3. Data acquisition

The signals arising from the detectors are processed by TNT2[1] cards developed at our institute. Signals are processed online in parallel in two different channels, one determining the event time by applying the Constant Fraction Discriminator (CFD) method and one calculating the γ-ray energy of the incident events using the Jordanov [2] signal treatment method. The events are stored in list mode files, where the energy is encoded on 14 bits and the time resolution is 10 ns.

4. Flux monitoring

Precision of cross section measurements depends very strongly on the uncertainties of the incident neutron flux (Fig. 2), thus it is of upmost importance to have very precise flux data.

The flux is measured using a double layer ^{235}U fission chamber. The deposits, both highly enriched in ^{235}U (>99.5%) are very thin: 324 μg/cm^2 for the vacuum evaporated $^{235}UF_4$ layer and 387 μg/cm^2 for the spray painted $^{235}U_3O_8$ layer. The effective thickness of the fission chamber was chosen between 6 and 7 mm, as this leads to the best ratio of fission fragment energy loss (signal) and radioactivity α particle energy loss (background noise). The measurements show a combined evaporation and fission neutron spectrum, and uncertainties ranging from 2 to 4% at this moment. This should be improved after a detailed study of a high precision measurement recently performed at the Physikalisch-Technische Bundesanstalt (PTB), Braunschweig, Germany.

5. γ detection

The γ-rays, witnessing the decay of created isotopes, are detected using four high purity Germanium (HPGe) counters, made of planar crystals with depths ranging from 2 to 3 cm and surfaces dimensioned between 10 and 28 cm^2. The detectors are optimized for high resolution detection at low energies (resolution of 0.7 keV at 122 keV). They are placed at angles of 110° and 149° which allows the angular dependence to be taken into account. Backward angles were chosen to reduce dead time caused by the observation of events due to γ-flash scattering, arriving up to 50% of the detections.

III. DATA ANALYSIS

1. Differential cross sections

The differential production cross section for a γ transition of interest at a given angle θ_i and energy E_i can be expressed as:

$$\frac{\mathrm{d}\,\sigma}{\mathrm{d}\,\Omega}(\theta_i, E_i) = \frac{1}{4\pi}\,\frac{n_{GE}(\theta_i, E_i)}{n_{FC}(E_i)}\,\frac{\varepsilon_{FC}\sigma_{U,f}(E_i)}{\varepsilon_{GE}(E_i)}\,\frac{\varsigma_{FC}}{\varsigma_{sple}}\,\frac{s_{FC}}{s_{sple}} \quad (1)$$

where n_{GE} and n_{FC} represent the dead time corrected numbers of detections for a given ray in the Ge energy spectrum and for the fission chamber high energy spectrum respectively, ε_{GE} and ε_{FC} the Germanium detector's and the fission chamber's efficiency, $\sigma_{U,f}$ the ^{235}U fission cross section, ς_{FC} and ς_{sple} the areal densities of the Uranium layer in the fission chamber and the sample, s_{FC} and s_{sple} the surfaces of the Uranium layer in the fission chamber and the sample.

2. Angle integration

The quantity of interest is the total reaction cross section, which requires integration of Eq. (1). One can show

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that the differential cross section can be expressed as a finite sum over even degree Legendre polynomials [3]:

$$\frac{\mathrm{d}\,\sigma}{\mathrm{d}\,\Omega}(\theta) = \frac{\sigma_{tot}}{4\pi} \cdot \sum_{i=0}^{\infty} \alpha_i \, \mathrm{P}_i(\cos\theta) \tag{2}$$

where σ_{tot} is the total angle integrated cross section, and the α_i are coefficients depending on the angular momentum of the initial and final state J_i, J_f, and the transition multipolarity L [4]. As the highest order Legendre polynomial in the decay distribution has order $\leq 2L$ and $\leq 2J_i$, this infinite summation can be limited to M terms, where M = $\min\{2L,2J_i\}$.

Usually the sum can be limited to even Legendre polynomials up to the order of 6 as the contribution of higher-order polynomials is small. Under this assumption the integrated cross section can be obtained in very good approximation from measurements at only two angles where the value of the fourth-order Legendre polynomial P_4 is zero according to:

$$\sigma_{tot} \approx 4\pi \left[w_1^* \frac{\mathrm{d}\,\sigma}{\mathrm{d}\,\Omega}(\theta_1^*) + w_2^* \frac{\mathrm{d}\,\sigma}{\mathrm{d}\,\Omega}(\theta_2^*) \right] \tag{3}$$

with θ_1^* = (30.6° or 149.4°), θ_2^* = (70.1° or 109.9°), w_1^* = 0.3479, and w_2^* = 0.6521 for the zeroes of P_4 [3].

IV. RESULTS

In this paper the (preliminary) results for two different measurement sets are presented. The measurements on ^{235}U were part of the thesis work of H. C. Karam [5].

1. The ^{235}U isotope

In this section we present a preliminary data set of measurements of the $\frac{5}{2}^+ \rightarrow \frac{7}{2}^-$ transition, observed through emission of a 129.3 keV γ-ray. This γ-ray transition due to an inelastic scattering reaction on ^{235}U has never been measured before. Averaged cross-sections were derived from the time-of-flight spectra for neutron energy bins of suitable sizes. The integral cross sections were summed according to Eq. (3) using the results obtained from both angles (110° and 149°). These data were corrected for internal conversion resulting in *transition cross sections* between different levels. The total beam time of this experiment was 1059 hours.

Figure 3 shows that the measured data points are located below the model calculation results obtained with the TALYS code for neutron energies below 3 MeV, and above the calculation results for energies higher than 3 MeV. The used TALYS code was optimized for the fission cross section of the isotope of interest and its descendants [6]. We can observe the same order of magnitude for the measured data, as well as a fairly good agreement in the shape of the curves.

For the ^{235}U(n,2n)^{234}U reaction channel the 152.7 keV γ-ray transition has been observed. It results from a $6^+ \rightarrow 4^+$ transition. The obtained results are shown in Fig. 4. For the observed ^{235}U(n,2n)^{234}U reaction, a good agreement of the shape, but a factor 2 in amplitude are noticed between our measurements and the TALYS simulations.

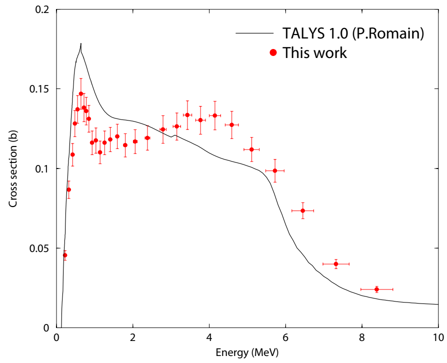

Fig. 3. (Color online) Transition cross section (γ-ray production cross section corrected for internal conversion) for the 129.3 keV transition of ^{235}U.

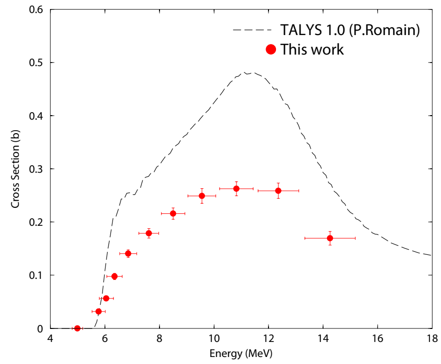

Fig. 4. (Color online) Transition cross section for the 152.7 keV transition of ^{234}U.

Due to the observed differences, the data will be subject to further investigations as well from experimental as theoretical point of view.

2. The ^{232}Th isotope

The following section presents the data obtained with an enriched ^{232}Th sample. In this preliminary analysis we studied the transitions $8^+ \rightarrow 6^+$ producing a γ-ray of intensity 223.6 keV (Fig. 5), $6^+ \rightarrow 4^+$ with a 171.2 keV γ-ray (Fig. 6), $4^+ \rightarrow 2^+$ for which E_γ = 112.75 keV (Fig. 7) and $2^+ \rightarrow 0^+$ yielding a 49.37 keV γ-ray (Fig. 8). The data for this measurement has been acquired within 375 hours and presents uncertainties ranging from 4 to 6% for the 223.6, 171.2, and 112.75 keV γ-rays and 7 to 10% for the 49.37 γ-ray.

The data obtained by our measurements seem in good agreement with the TALYS predictions concerning the ascending part and the maximum of the excitation curve. Nevertheless one can clearly observe a difference in the descending part of the cross section yields where the actual data declines earlier and faster than the TALYS predictions. This effect has already been observed in

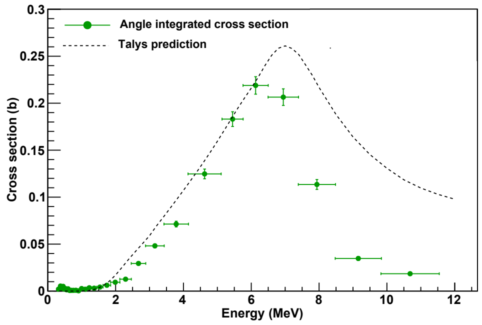

Fig. 5. (Color online) Transition cross section for the 223.6 keV transition of ^{232}Th.

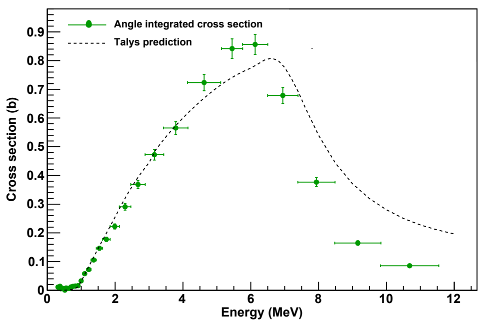

Fig. 6. (Color online) Transition cross section for the 171.2 keV transition of ^{232}Th.

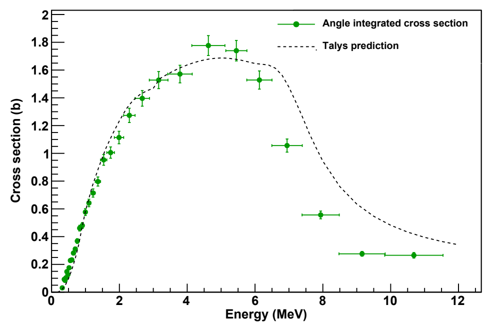

Fig. 7. (Color online) Transition cross section for the 112.75 keV transition of ^{232}Th.

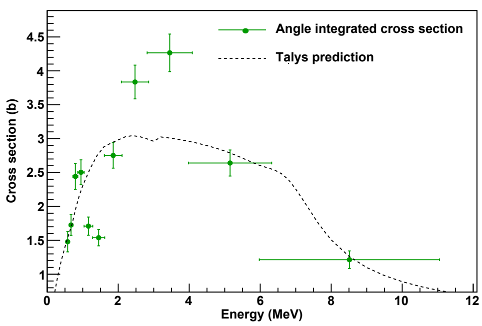

Fig. 8. (Color online) Transition cross section for the 49.37 keV transition of ^{232}Th.

^{232}Th(n,5n)^{228}Th cross section measurements performed with a different experiment at the CYCLONE facility in Louvain-la-Neuve [7]. This can be explained by the level density parameters used in TALYS to obtain the predictions. Our experiments may give a new insight on those densities and be used to improve the predictions for residual nuclei.

The 49.37 γ-ray data has to be considered with caution, as its measurement is affected by high correction factors. The results will need further investigations.

V. CONCLUSIONS

The upcoming projects include finalizing the analysis of the ^{232}Th(n,n'γ)^{232}Th data, and remeasuring the ^{235}U(n,n'γ)^{235}U and ^{235}U(n,2nγ)^{234}U data to improve precision. The presented results will be subject of further investigations from experimental as well as from theoretical point of view.

After a consequent work on the uncertainties of these measurements, we are currently able to measure (n,xnγ) cross sections of a precision ranging from 5 to 7%.

Future measurement campaigns include enriched tungsten samples and ^{238}U. The ultimate goal of developing these measurement techniques is to study (n,xn) reactions on ^{233}U, lacking experimental data, which is of upmost importance for the Thorium cycle. For example, the ^{233}U(n,2n)^{232}U reaction leads in its decay to ^{208}Pb, emitter of a 2.6 MeV γ-ray, which has a major impact on the reactor core temperature.

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