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Activation Cross Section Measurement of the (n,2n) Reaction on ^{203}Tl at 16.4 MeV, 18.9 MeV and Theoretical Calculations via the EMPIRE code

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Abstract The aim of the present work is to study the cross section of the (n,2n) and (n,3n) reactions on ^{203}Tl , by irradiating a natural TlCl pellet target with monoenergetic neutron beams at 16.4 and 18.9 MeV. The cross section measurements were carried out using the activation method, with respect to the $^{197}\text{Au}(n,2n)^{196}\text{Au}$ and $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reference reactions. The monoenergetic neutron beams were generated in the 5.5 MV Tandem accelerator of NCSR “Demokritos”, using the $^3\text{H}(d,n)^4\text{He}$ reaction. Monte Carlo simulations have been performed to take into account the γ -ray self-absorption results as well as the estimation of the neutron flux through the reference foils. Theoretical calculations with the EMPIRE code have also been performed, using the same parameterization implemented in the theoretical study of Ir and Au nuclei in an attempt to find a suitable model for the description of all the experimental results in this mass region.

Keywords thallium, cross section, neutron activation analysis, Monte Carlo, EMPIRE code

INTRODUCTION

The significance of studying neutron-induced reactions lies in their importance to basic research and their relevance for practical applications in medicine, nuclear technology and industry [1-3]. Thallium is widely used in electronics, pharmaceuticals, nuclear medicine, infrared detectors and fiber optics. Nevertheless, little available information exists in literature for neutron-induced reactions on Tl isotopes. Especially in the case of the $^{203}\text{Tl}(n,2n)^{202}\text{Tl}$ reaction, many discrepancies among the existing experimental data exist, mainly in the energy region above 12 MeV [4,5], as can be seen in Fig. 1.

EXPERIMENTAL DETAILS

Reaction

The impingement of the highly energetic neutron beams on the ^{203}Tl isotope produced the highly excited compound nucleus $^{204}\text{Tl}^*$, while among the possible exit channels, the (n,2n) channel opens just above 8 MeV and the (n,3n) channel well above 15 MeV. The experimental spectra of the thallium foils after activation are shown in Fig. 2 for the 16.4 and 18.9 MeV along with their background spectra. The characteristic γ -line peak of the $^{203}\text{Tl}(n,2n)^{202}\text{Tl}$ reaction lies at 439.5 keV, while the one of $^{203}\text{Tl}(n,3n)^{201}\text{Tl}$ reaction at 167.5 keV, with half-lives of 12.31 d and 3.04 d for the two residual nuclei respectively, as shown in Table 1.

Table 1. Characteristics of the studied channels

Reaction	E_γ (keV)	I_γ (%)	Half Life (d)
$^{203}\text{Tl}(n,2n)^{202}\text{Tl}$	439.51	91.5	12.31
$^{203}\text{Tl}(n,3n)^{201}\text{Tl}$	167.47	10	3.04

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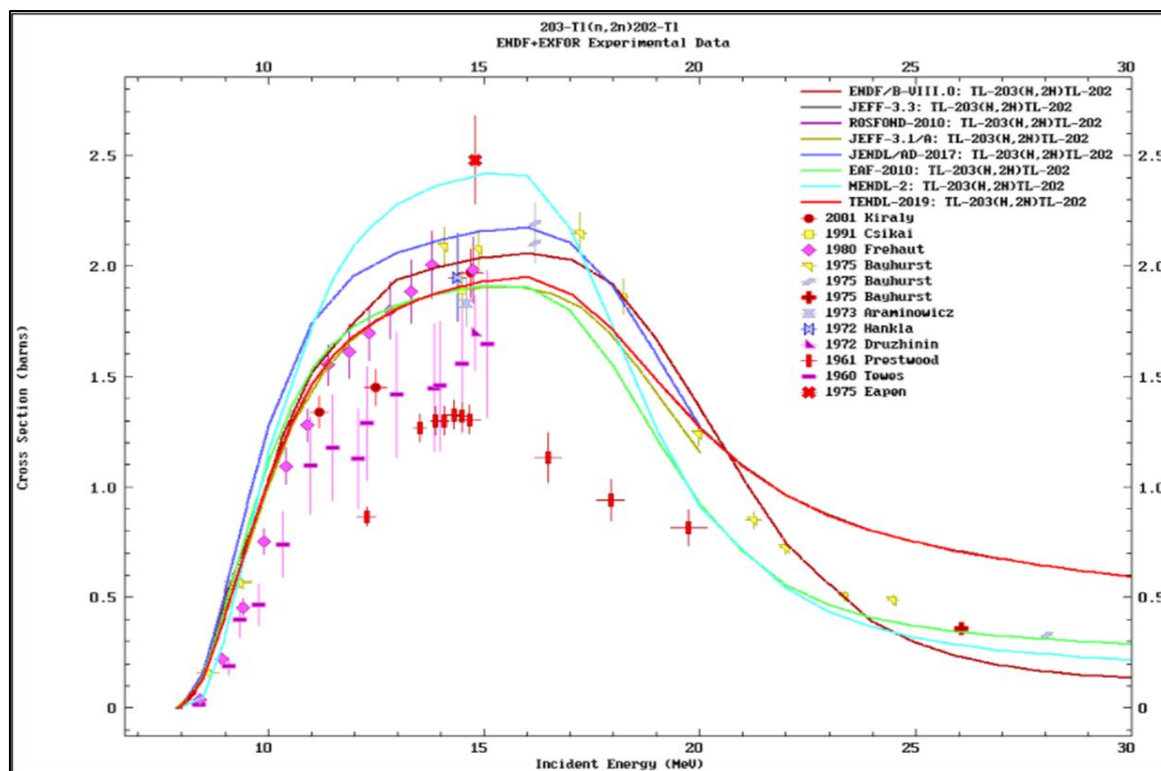


Figure 1. Existing experimental data of the $^{203}\text{Tl}(n,2n)^{202}\text{Tl}$ reaction

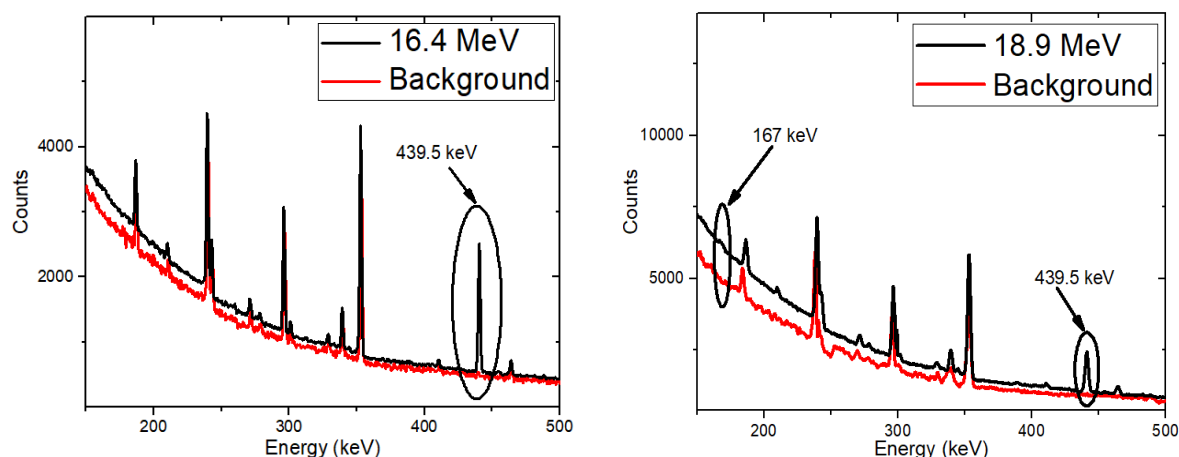


Figure 2. Derived spectra of the irradiated thallium foils at 16.4 and 18.9 MeV, compared to the time normalized background spectrum. At 18.9 MeV the appearance of the 167.5 keV characteristic transition for the $(n,3n)$ reaction is obvious

Experimental setup

The experiments were conducted at the 5.5 MV tandem accelerator of N.C.S.R. “DEMOKRITOS”. The neutron beam that irradiated the natural TlCl pellet, in each irradiation (see Table 2), was produced via the $^3\text{H}(d,n)^4\text{He}$ reaction, using a Ti-T solid target. For the determination of the neutron flux, the reference samples that were used were Al and Au foils of the same diameter (13 mm diameter) as the thallium target. The target and reference foil assembly were placed at approximately 1.5-2 cm from the tritium target, where the neutron beam is practically isotropic and monoenergetic. A BF_3 detector was placed at ~ 3 m from the tritium flange for the monitoring of the neutron beam fluctuations. The induced radiation of the samples was measured using High Purity Germanium (HPGe) detectors, with 80% and 100% relative efficiency depending on each irradiation,

which were properly shielded with lead blocks. The irradiated samples were placed at a distance of 10 cm from the entrance window of the HPGe detectors. The absolute efficiency of the detectors was deduced with the use of a calibrated ^{152}Eu point source of (217 ± 3) kBq activity, placed at the same distance as the samples.

Table 2. Characteristics of the two performed irradiations

En (MeV)	Irradiation time (h)	Measurement time (d)	Deuteron Energy (MeV)	Neutron fluence (n/cm ²)
16.4	~26	~5	2.25	$(1.75 \pm 0.09) \cdot 10^{10}$
18.9	~28	~8	2.64	$(1.37 \pm 0.08) \cdot 10^{10}$

CROSS SECTION MEASUREMENT

The cross section σ of the $^{203}\text{Tl}(n,2n)^{202}\text{Tl}$ and $^{203}\text{Tl}(n,3n)^{201}\text{Tl}$ reactions was measured with respect to the $^{197}\text{Au}(n,2n)^{196}\text{Au}$ and $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reference reactions (see Fig. 3), implementing the activation method, and was calculated by the formula:

$$\sigma = \frac{N_p}{N_t} \frac{1}{\Phi} \quad (1)$$

N_p : the number of unstable nuclei produced by the neutron irradiation

N_t : the number of target nuclei irradiated

Φ : neutron flux calculated with Monte Carlo simulations applying the MCNP code [6] along with the experimental results from the reference reactions.

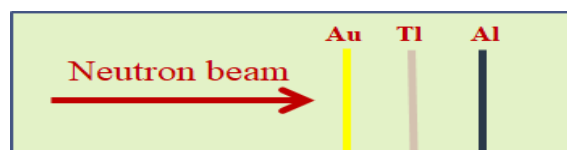


Figure 3. Schematic configuration of the targets

The de-excitation of the produced nuclei from the end of the irradiation until the end of the activity measurement, as well as the balance between production and de-excitation of nuclei during the irradiation, were taken into account for the calculation of the number of nuclei produced by the neutron irradiation.

RESULTS AND DISCUSSION

The preliminary experimental cross section results are presented in Table 3 and Fig. 4, along with the existing experimental data in literature [5], as well as the ENDF-VIII, JENDL-5 and TENDL-2019 evaluations [4] of the $^{203}\text{Tl}(n,2n)^{202}\text{Tl}$ and $^{203}\text{Tl}(n,3n)^{201}\text{Tl}$ reactions.

Table 3. Experimental Cross Section Results

En (MeV)	σ (barn)	
	$^{203}\text{Tl}(n,2n)^{202}\text{Tl}$	$^{203}\text{Tl}(n,3n)^{202}\text{Tl}$
16.4	1.87 ± 0.11	—
18.9	1.49 ± 0.09	0.39 ± 0.06

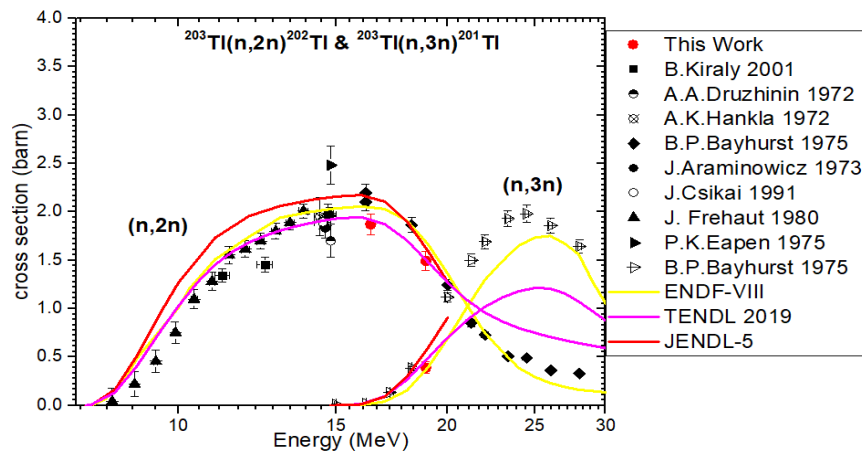


Figure 4. Existing experimental data, this work and ENDF-VIII, JENDL-5 and TENDL-2019 evaluations of the $^{203}\text{Tl}(n,2n)^{202}\text{Tl}$ and $^{203}\text{Tl}(n,3n)^{201}\text{Tl}$ reactions

THEORETICAL CALCULATIONS – EMPIRE CODE

The theoretical calculations in the framework of the Hauser-Feshbach compound nucleus theory using the code EMPIRE (version 3.2.3) [7] and the comparison with the available experimental cross section data were carried out, with the aim of finding a suitable model for the description of the reaction cross sections under study. The best agreement between theoretical calculations with the EMPIRE code and experimental data, especially in the high energy region (see Fig. 5), was achieved with the parameters below:

- Optical Model: Wilmore, 1964 [8]
- Enhanced Generalized Superfluid Model (EGSM) nuclear level densities [9]
- Enabled MSD
- Enabled MSC
- Disabled Direct input

Furthermore, these parameters describe very well the $^{205}\text{Tl}(n,2n)^{204}\text{Tl}$ reaction, thus confirming their suitability, as shown in Fig. 6. The same parameters were used in the case of the investigation of neutron-induced reactions on Ir and Au [10,11] and the fair agreement between theoretical calculations and experimental data provides an indication of how successfully the theoretical models can reproduce the experimental results in this mass region with the same parametrization.

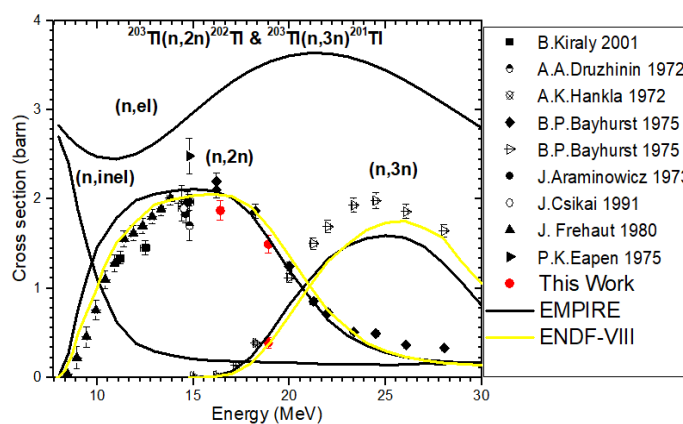


Figure 5. Existing experimental data, this work, ENDF-VIII and theoretical calculations

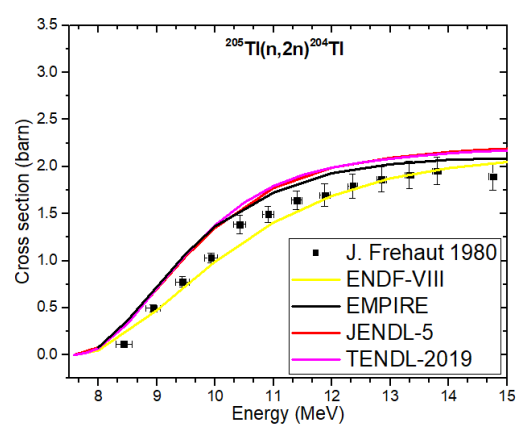


Figure 6. Existing experimental data, ENDF-VIII, JENDL-5, TENDL-2019 and theoretical calculations

However, further theoretical investigation is needed in order to achieve better agreement between theoretical calculations and experimental data, mainly for the $^{203}\text{Tl}(n,2n)$ reaction below 15 MeV and the $^{203}\text{Tl}(n,3n)$ reaction above 20 MeV. Further measurements are also planned in the near future to cover the energy range from 15 up to 20 MeV and also at lower energies, below 12 MeV, where only the data by Frehaut et al. [12] are available.

Acknowledgments

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