

Measurements of cross-sections of (n,xn) threshold reactions in various materials

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Abstract. Experimental cross-section data for (n,xn) reactions with x higher than four and neutron energies over ~30 MeV are very scarce. We performed seven successful (n,xn) cross-section measurements in two campaigns exploiting the quasi-monoenergetic neutron source at The Svedberg Laboratory in Uppsala, Sweden. Neutron energies from the ⁷Li(p,n)⁷Be based source were in the region 22 to 94 MeV. We carried out additional five irradiations with neutron energies from 17 up to 34 MeV using the quasi-monoenergetic neutron source of the Nuclear Physics Institute in Řež. We have irradiated Al, Au, Bi, I, In, Ta and Y materials in the form of thin foils. We observed good agreement with the few existing experimental data about corresponding cross-sections in EXFOR database and also with the calculations performed in deterministic code TALYS.

1 Introduction

As members of international collaboration “Energy and Transmutation of Radioactive Waste” we routinely use (n,xn) threshold reactions in various materials to measure high energy neutron flux from spallation reactions [1]. The usually used materials for activation detectors are Al, Au, Bi, I, In, Ta and Y. However the detailed measurements of the neutron reactions cross-sections in the energy range of interest in above mentioned transmutation studies are still missing. Therefore we lunched corresponding experiments using neutron energies 17, 22, 30 and 35 MeV from the quasi-monoenergetic neutron source in Řež and neutron energies 22, 47 and 94 MeV in Uppsala. The last experiment was carried out in February 2010 in Uppsala using neutron energies 59, 66, 72 and 89 MeV. Evaluated libraries offer (n,xn) cross-sections for higher x and energies up to hundred of MeV. Deterministic code TALYS can calculate cross-sections up to incident neutron energy 200 MeV. Evaluated libraries and TALYS describe well the shape of the cross-section, but they differ in absolute value.

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2 Cyclotrons at TSL Uppsala and at NPI Řež

A high energy neutron source with well known monoenergetic spectrum is needed for cross-section studies. This source must be in addition highly intensive for experiments done by the means of activation method. Unfortunately, the only available sources to use are quasi-monoenergetic. We use neutron source based on ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction, see “Fig.1.”. Approximately a half of the intensity was in the peak with FWHM = 1 MeV (corresponds to the ground state and first excited state at 0.43 MeV at ${}^7\text{Be}$) and half of the intensity was in the continuum in lower energies (corresponds to higher excited state, multiple-particle emission etc.)

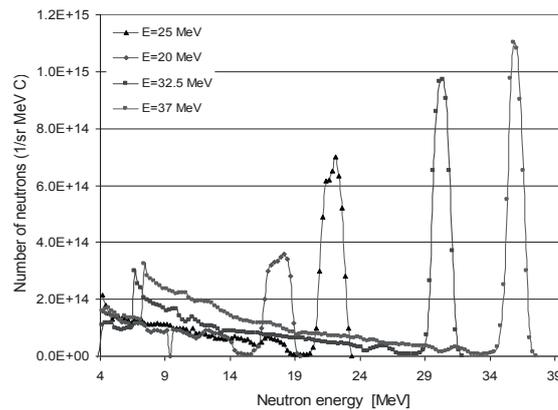


Fig. 1. Neutron spectra for different proton energies (NPI ASCR Řež).

The cyclotron at TSL Uppsala has the energy range from 20 MeV to 180 MeV for protons and the maximum neutron flux density $10^5 \text{ cm}^{-2}\text{s}^{-1}$ [2]. The remaining part of proton beam behind the lithium target was deflected by a magnet and guided onto a graphite beam dump. The neutron beam was formed by a 100 cm long iron collimator (50 cm in diameter). Samples were placed 373 cm from the lithium target. Time of irradiation was about 8 hours. The uncertainty of the neutron spectra determination as well as the uncertainty of the beam intensity determination was about 10 %.

Energy range of neutron source at NPI of ASCR at Řež is between 10 and 37 MeV for protons and the maximum neutron flux density is $10^8 \text{ cm}^{-2}\text{s}^{-1}$ [3]. High energy protons from the cyclotron were lead to lithium target with carbon backing to stop protons. The thickness of the neutron source was 2 mm. Samples were located from 11 to 16 cm from the lithium target. Time of irradiation was about 20 hours. The uncertainty of the neutron spectra determination was 10 %, the uncertainty of the beam intensity determination was in this case 5 %.

3 Evaluation of cross-section measurements

For the (n,xn) cross-section measurement we have used neutron activation and gamma spectra evaluation. In all irradiations were studied Au, Al, Bi, In and Ta materials. The studied materials were in the form of thin foils. Thickness of the foils was ranging from 0.05 mm up to 1 mm, weights of the foils varied from 0.2 up to 7 grams depending on the foil type and beam energy. Foils were wrapped in two layers of paper, so the nuclide transport between the foils was minimized. Iodine samples were in the form of solid KIO_4 tablet. Transport from the irradiation hall to the spectrometer took approximately two minutes in Uppsala, ten minutes in Řež.

3.1 DEIMOS32

The DEIMOS32 code [4] was used to evaluate the gamma spectrum, i.e. to determine the area and also the uncertainty of the area for a specific γ peak. Every selected peak is fitted with a Gaussian curve and the fit procedure is based on a non-linear least squares method, see “Fig.2.”.

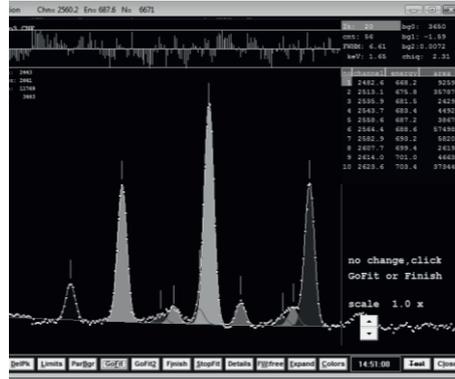


Fig. 2. Graphical interface of the DEIMOS32 code

3.2 The total yield and calculation of cross-section

The total yield of observed radioactive nuclei per one gram of activated material and one neutron was calculated with consideration of the spectroscopic corrections by “Eq.1.”:

$$N_{yield} = \frac{S_p \cdot C_{abs}(E)}{I_\gamma \cdot \varepsilon_p(E) \cdot COI \cdot C_{area}} \cdot \frac{t_{real}}{t_{live}} \cdot \frac{1}{m_{foil}} \cdot \frac{e^{(\lambda \cdot t_0)}}{1 - e^{(-\lambda \cdot t_{real})}} \cdot \frac{\lambda \cdot t_{irr}}{1 - e^{(-\lambda \cdot t_{irr})}} \quad (1)$$

where S_p is the peak area, C_{abs} – self-absorption correction, I_γ – gamma line intensity, $\varepsilon_p(E)$ – detector efficiency, COI – correction for real coincidences, C_{area} – square-emitter correction, t_{real}/t_{live} – dead time correction, m_{foil} – mass of foil and the last two fractions represent decay during cooling and measurement and decay during irradiation.

The following “Eq.2.” was used for determination of reaction cross-section σ .

$$\sigma = \frac{N_{yield} \cdot S \cdot A}{N_n \cdot N_A} \quad (2)$$

N_{yield} – yield of studied isotope, S – area of the foil, A – molar weight, N_n – number of neutrons in the peak, N_A – Avogadro number.

4 Background subtraction

Neutron energy spectrum of quasi-monoenergetic source used contains besides the main neutron peak also lower continuum stretching up to thermal energies “Fig.3.”. This spectrum is different at every irradiation facility. Because of the large amount of low energy neutrons, production of the isotope by these “background” neutrons was not negligible for most of the isotopes studied. This was solved by the subtraction of their contribution. Only reactions with the threshold few MeV lower than the neutron peak could be used to direct cross-section evaluation. Background contribution was determined by folding of the neutron source spectrum and calculated cross-sections. The cross-sections were calculated by means of the deterministic code TALYS 1.0 [5]. The background

subtraction procedure is a potential source of unknown systematic uncertainty. It is insensitive to the absolute value of the cross-section, but a modification in the cross-section shape or in the neutron spectrum shape can change it. TALYS enables five basic settings of nuclear level densities, which can influence the course of the cross-section.

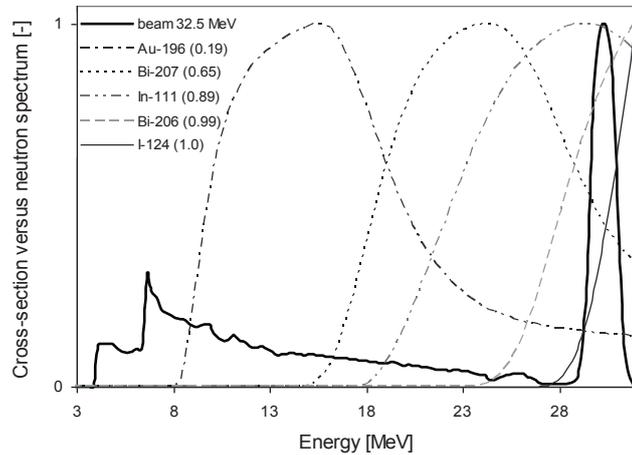


Fig. 3. Example of different influence of background (values in the parentheses are ratios between production in the peak and total production).

5 Cross-section results

Cross-section measurements at NPI and TSL cover a wide range of energies. A lot of new experimental data were measured. Good agreement with TALYS 1.0 and known experimental data from EXFOR was observed for most of the isotopes. For energies higher than 40 MeV and reactions of the order higher than (n,4n) no data are available in EXFOR (except bismuth) [6], so it is possible to say, that our data are unique. Some examples of our measurements are in “Fig.4.”- “Fig.8.”.

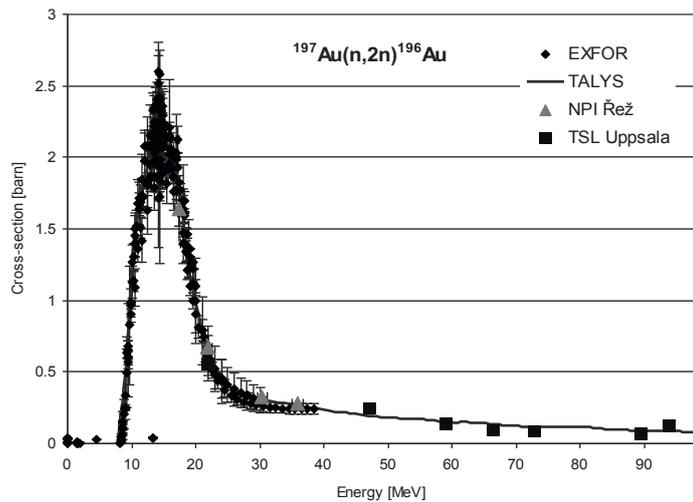


Fig. 4. Comparison of experiment data from NPI and TSL with TALYS and EXFOR, $^{197}\text{Au}(n,2n)^{196}\text{Au}$ reaction.

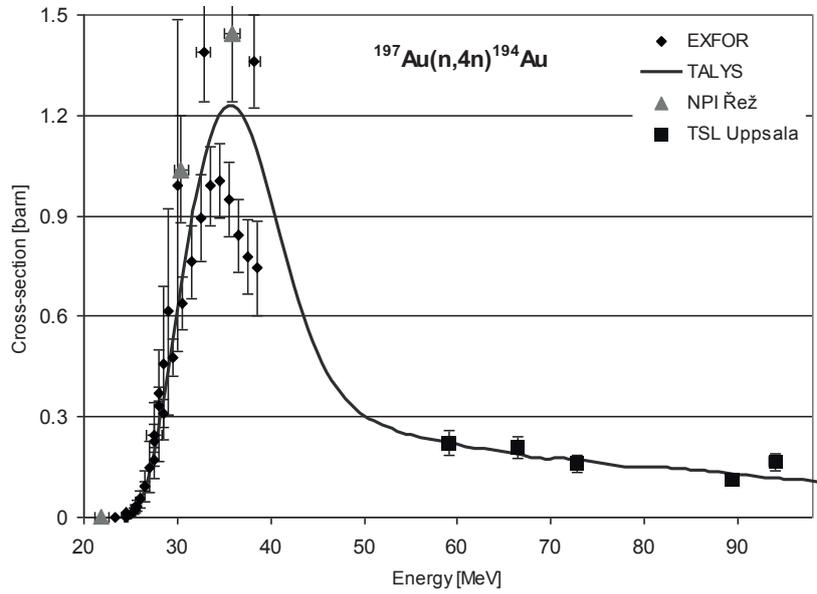


Fig. 5. Comparison of experiment data from NPI and TSL with TALYS and EXFOR, $^{197}\text{Au}(n,4n)^{194}\text{Au}$ reaction.

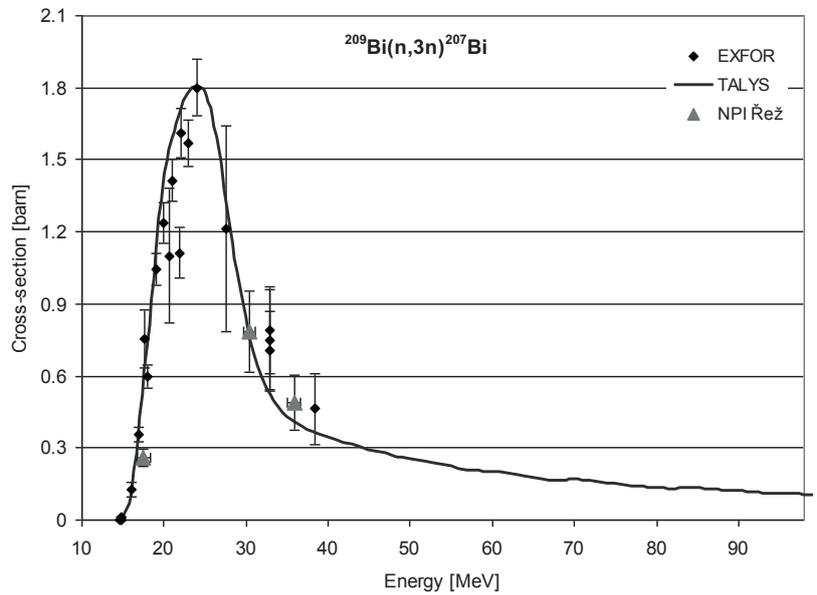


Fig. 6. Comparison of experiment data from NPI and TSL with TALYS and EXFOR, $^{209}\text{Bi}(n,3n)^{207}\text{Bi}$ reaction.

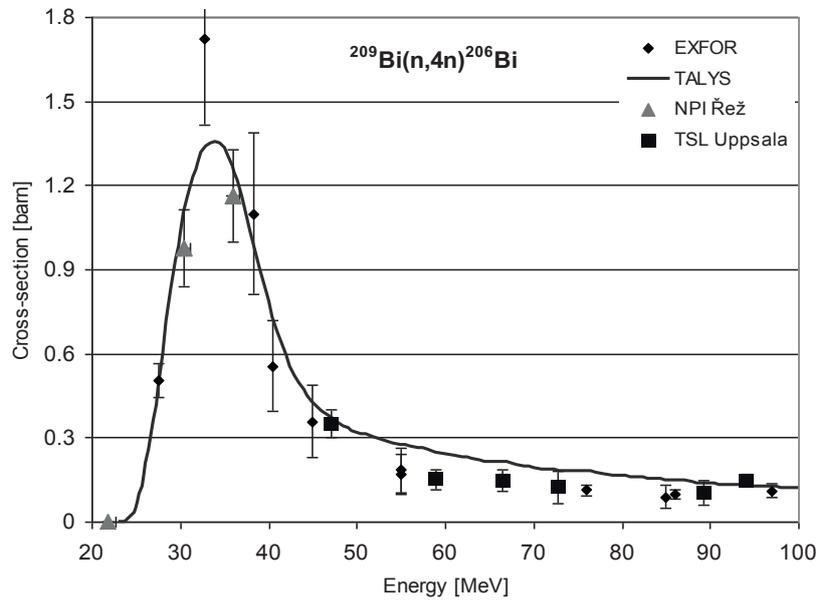


Fig.7. Comparison of experiment data from NPI and TSL with TALYS and EXFOR, $^{209}\text{Bi}(n,4n)^{206}\text{Bi}$ reaction.

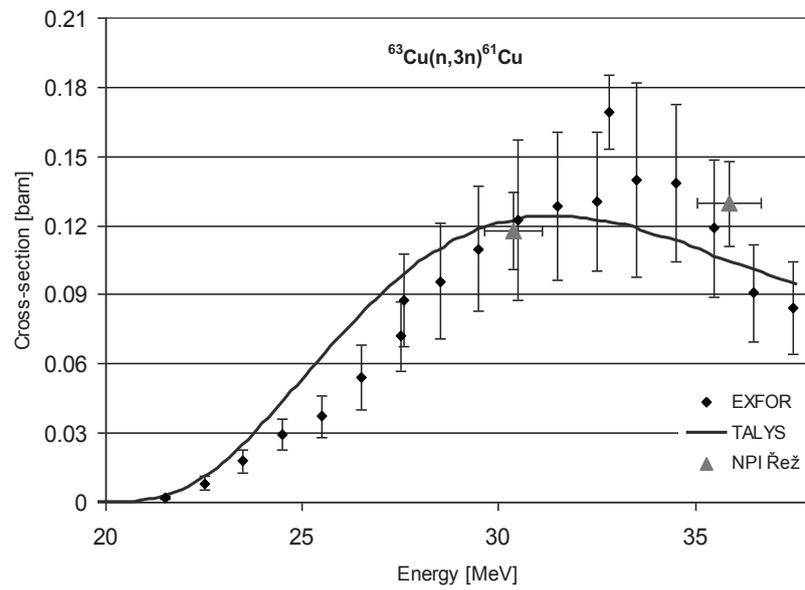


Fig.8. Comparison of experiment data from NPI and TSL with TALYS and EXFOR, $^{63}\text{Cu}(n,3n)^{61}\text{Cu}$ reaction.

Summary

Cross-sections of neutron threshold reactions in Al, Au, Bi, I, In, and Ta were studied in the energy region from 17 to 94 MeV by the means of activation analysis and gamma spectroscopy. We used quasi-monoenergetic neutron sources based on ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction. The results are in good agreement with the cross-sections published in EXFOR. You can find more details in [8-11]. Previous measurements in NPI and TSL were published at scientific workshop EFNUDAT – Slow and Resonance neutrons in 2009 in Budapest [8] and at International Conference on Nuclear Data for Science and Technology in 2010 in South Korea [9]. This article presents final cross-sections data, which was preliminarily published at scientific workshop NEMEA-6 on Nuclear Measurements, Evaluations and Applications in 2010 in Poland [10].

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References

1. A. Krása et al., “Neutron production in a Pb/U-setup irradiated with 0.7-2.5 GeV protons and deuterons”, *Nucl. Instr. and Meth. in Nucl. Res.*, A615, No.1, 70-77 (2010)
2. A. V. Prokofiev et al., “The TSL Neutron Beam Facility”, *Rad. Prot. Dos.*, 126, 18-22 (2007)
3. P. Bém et al., “The NPI cyclotron-based fast neutron facility”, *Proceedings of the International Conference on Nuclear Data for Science and Technology – ND2007*, 555-558 (2007)
4. J. Frána, “Program DEIMOS32 for Gamma-Ray Spectra Evaluation”, *J. Rad. Nucl. Chem.*, V.257, No.3, 583-587 (2003)
5. A. J. Koning et al., “TALYS-1.0.”, *Proceedings of the International Conference on Nuclear Data for Science and Technology–ND 2007*, 211-214 (2007)
6. Experimental Nuclear Reaction Data (EXFOR/CSISRS) [<http://www.nndc.bnl.gov/exfor>], 7.10.2011
7. European Facilities for Nuclear Data Measurements [www.efnudat.eu]
8. O. Svoboda et al., *Proceedings of the 2nd EFNUDAT scientific workshop on Neutron Measurements, Theory and Applications, EFNUDAT – Slow and Resonance Neutrons*, Budapest, Hungary, 155-161 (2009)
9. O. Svoboda et al., *Proceedings of the International Conference on Nuclear Data for Science and Technology–ND2010*, Jeju, South Korea, 1709-1712 (2010)
10. J. Vrzalová et al., *Proceedings of the NEMEA-6 scientific workshop on Nuclear Measurements, Evaluations and Applications*, Krakow, Poland, 259-265 (2010)
11. O. Svoboda, *Experimental Study of Neutron Production and Transport for ADTT*, Dissertation Thesis, Czech Technical University, Faculty of Nuclear Sciences and Physical Engineering, Prague 2011, [http://ojs.ujf.cas.cz/~wagner/transmutace/diplomky/PHD_Svoboda.pdf]