

Measurement of cross-sections of yttrium (n,xn) threshold reactions by means of gamma spectroscopy

Petr Chudoba^{1,2}, S. Kilim³, V. Wagner¹, J. Vrzalova^{1,4}, O. Svoboda¹, M. Majerle¹, M. Stefanik¹, M. Suchopar^{1,4}, A. Kugler¹, M. Bielewicz³, E. Strugalska-Gola³, M. Szuta³, D. Hervas¹, T. Herman¹, B. Geier¹

¹ Nuclear Physics Institute of the ASCR, v. v. i., Rez 130, 250 68 Rez, Czech Republic

² Charles University in Prague, Faculty of Mathematics and Physics, Ke Karlovu 3, 121 16 Praha 2, Czech Republic

³ National Centre for Nuclear Research, Otwock-Swierk 05-400, Poland

⁴ Faculty of Nuclear Sciences and Physical Engineering, Czech Technical University in Prague, Brehova 7, 11519 Praha, Czech Republic

Abstract

Neutron activation and gamma spectrometry are usable also for the determination of cross-sections of different neutron reactions. We have studied the cross-sections of yttrium (n,xn) threshold reactions using quasi-monoenergetic neutron source based on the reaction on ⁷Li target at Nuclear Physics Institute of ASCR in Rez. Yttrium (n,xn) threshold reactions are suitable candidates for fast neutron field measurement by activation detectors. Fast neutron field monitoring is necessary already today at a wide range of accelerator facilities and will gain on importance in future fast reactors of generation IV, accelerator transmutation systems or fusion reactors. The knowledge of the cross-sections is crucial for such purpose. Unfortunately, the cross-section is sufficiently known only for ⁸⁹Y(n,2n)⁸⁸Y reaction. For higher orders of reactions there are almost no experimental data. Special attention was paid to the ⁸⁹Y(n,3n)⁸⁷Y reaction. The cross-sections of both ⁸⁹Y(n,2n)⁸⁸Y and ⁸⁹Y(n,3n)⁸⁷Y reactions were analyzed.

1 Introduction and motivation

For future advanced reactor systems same as for fusion reactors and advanced spallation sources of neutrons, the suitable activation neutron detectors will be necessary. During the experiments of the international collaboration 'Energy and Transmutation of RadioActive Waste' it was shown that yttrium is good candidate for monitoring of neutron fields with activation samples, mainly because of its (n,xn) threshold reactions, whose products are easily identifiable and with good half-life for γ -spectrometry. Unfortunately the knowledge of cross-sections of these reactions is insufficient. So we decided to measure cross-section of yttrium (n,xn) reactions at different neutron energies. Quasi-monoenergetic (QM) neutron source at Nuclear Physics Institute (NPI) of the Academy of Sciences of the Czech Republic in Rez [1] was used as the neutron source. The source is based on ⁷Li(p,n)⁷Be reaction. This series of measurements is continuation of previous measurements [2] made by our group using NPI source and quasi-monoenergetic neutron source at The Svedberg Laboratory in Uppsala, Sweden [3].

2 Samples and measurements

The measurements were done using six different proton energies. Four irradiations were part of ERINDA project and two next irradiations were on base of CANAM. Positions of QM peak were 17.4, 24.5, 24.8, 27.9, 28.7 and 33.5 MeV. Two of them were almost the same, so there is a good opportunity to check the consistency of evaluation procedure and systematic uncertainty sources. Two types of samples were used: 'YN' samples which were made of solid yttrium foil with dimensions 25 × 25 × 0.64 mm³, with

weight ~ 1.8 g and 'YO' samples which had form of pills made of compressed yttrium powder with dimensions $\varnothing 9 \times 1.5$ mm³, with weight $\sim 0.6 - 0.8$ g. The samples were fixed on a paper attached to an aluminum holder which was mounted behind the neutron source. Gold samples were irradiated together with the yttrium samples. Both samples were on the same position on the holder. The yttrium sample was placed on top of the gold sample. The gold samples with much better known cross-sections of neutron reactions were used as the experimental condition monitors. Figures 1 (a) and (b) shows the arrangement of samples and neutron source. Both pictures were made using VISED [4] program. For each irradiated sample, the neutron spectrum was calculated by means of MCNPX [5] simulation.

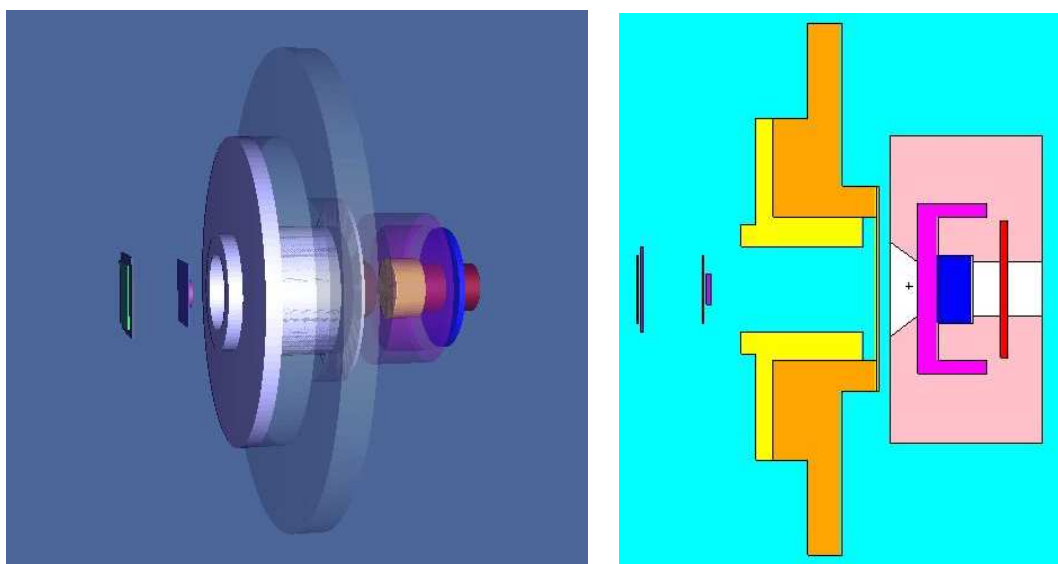


Fig. 1: (a) 3D model of neutron source with samples; (b) visualization of neutron source with samples for MCNPX

2.1 Evaluation of cross-section

The evaluation procedure consists in the calculation of number of produced nuclei N_{yield} for each isotope. Then using this value the cross-section is calculated. The N_{yield} is calculated accordingly to formula

$$N_{yield} = \frac{S_{peak} \cdot C_{abs}(E)}{I_{\gamma} \cdot \varepsilon_p(E) \cdot COI_E \cdot C_{area}} \frac{t_{real}}{t_{live}} \frac{e^{\lambda \cdot t_0}}{1 - e^{-\lambda \cdot t_{real}}} \frac{\lambda \cdot t_{irr}}{1 - e^{-\lambda \cdot t_{irr}}}, \quad (1)$$

where S_{peak} - peak area, $C_{abs}(E)$ - self-absorption correction, I_{γ} - gamma emission probability, $\varepsilon_p(E)$ - detector efficiency, COI_E - true coincidences correction, C_{area} - square emitter correction, t_{real} - real time of measurement, t_{live} - live time of measurement, t_0 - cooling time, t_{irr} - irradiation time, λ - decay constant. The last three fractions represent respectively dead time correction, correction for decay during cooling and measurement and correction for decay during irradiation. The peak area was determined using Canberra's Genie 2000 software. The uncertainties of peak areas were between 0.5% and 3%. The uncertainties brought in by corrections are about 1%, except the detector efficiency which has uncertainty not worst then 3%. Using formula (1) to get the number of produced nuclei it is possible to use formula (2) to calculate the cross-section. This formula has the form

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

where S - foil area, A - molar mass, B_a - beam instability correction, N_n - number of neutrons in peak, N_A - Avogadro's number, m - foil mass. The character of neutron spectra forces to involve one more correction. Since almost half of the produced neutrons are in low energy background tail, it is necessary to involve background subtraction correction.

2.2 Background subtraction method

The quasi-monoenergetic neutron source based on ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction has the energy spectrum with contributions from monoenergetic peak and continuum at lower energies. Fig. 2 shows an example of the neutron spectrum with the cross-sections for (n,2n) and (n,3n) reactions on yttrium. The neutron background is negligible only in case, when the threshold energy is just under the energy of the peak, for other cases subtraction procedure [6] was involved. This procedure is based on the ratio between the folding of calculated cross-section $\sigma(E)$ and neutron spectrum $N(E)$ in the peak energy interval and the same convolution in the whole spectrum interval. Since the neutron spectrum is binned, the integral operators are replaced by sum operators. The background subtraction correction factor is defined accordingly to formula (3). Using this coefficient it is possible to correct the number of produced nuclei for the ones produced by background neutrons accordingly to (4).

$$C_{bgr} = \frac{\int_{Peak} \sigma(E) \cdot N(E) dE}{\int_{Spectrum} \sigma(E) \cdot N(E) dE} \longrightarrow \frac{\sum_{i \in Peak} \sigma_i \cdot N_i}{\sum_i \sigma_i \cdot N_i} \quad (3)$$

$$N_{yield} \longrightarrow N_{yield,peak} = N_{yield} \cdot C_{bgr} \quad (4)$$

Cross-sections are calculated using TALYS 1.4 [7]. The background subtraction procedure is independent on the absolute value of cross-section. It is dependent only on the shape of the cross-section. The advantage of it will be seen in discussion of results. If the shape wouldn't be accurate, then the values for higher energies would diverge from it. Direction of divergence would be opposite than the difference between the correct shape and the used one. In order to validate simulated neutron flux by MCNPX, the gold samples were irradiated together with yttrium samples. Difference between simulated and calculated neutron fluency is not exceeding 2 %. Two uncertainties are brought in the final cross-section by the background subtraction procedure: 1) uncertainty of the used cross-sections below QM peak, 2) uncertainty of the used neutron spectra. We estimate the sum of these two uncertainties to be around 10%.

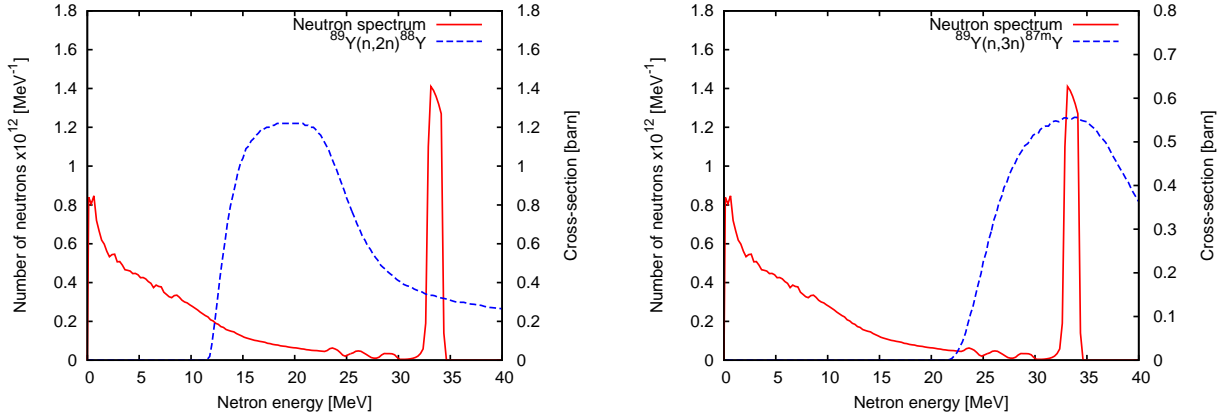


Fig. 2: Neutron spectrum and cross-section for (a) (n,2n) reaction; (b) (n,3n) reaction

3 Cross-section results

Currently the experimental cross-section data of yttrium (n,xn) reactions for higher are almost nonexistent. Only for (n,2n) reaction there are enough experimental points in the EXFOR database. For (n,3n) and higher order reactions, there are almost no experimental data. The data in this work are still preliminary, but they are in good agreement with the current experimental data in EXFOR. The cross-sections of the reaction ${}^{89}\text{Y}(n,3n){}^{87m}\text{Y}$ are available only in [2] for neutron energies from 59.0 to 89.3 MeV, in

bachelor thesis [8] for neutron energy 32 MeV and in this contribution for neutron energies from 24.5 to 33.5 MeV. The results together with data from [8] marked as 'Rez 2011', TALYS calculations and EXFOR data are shown in Figs. 3 and 4.

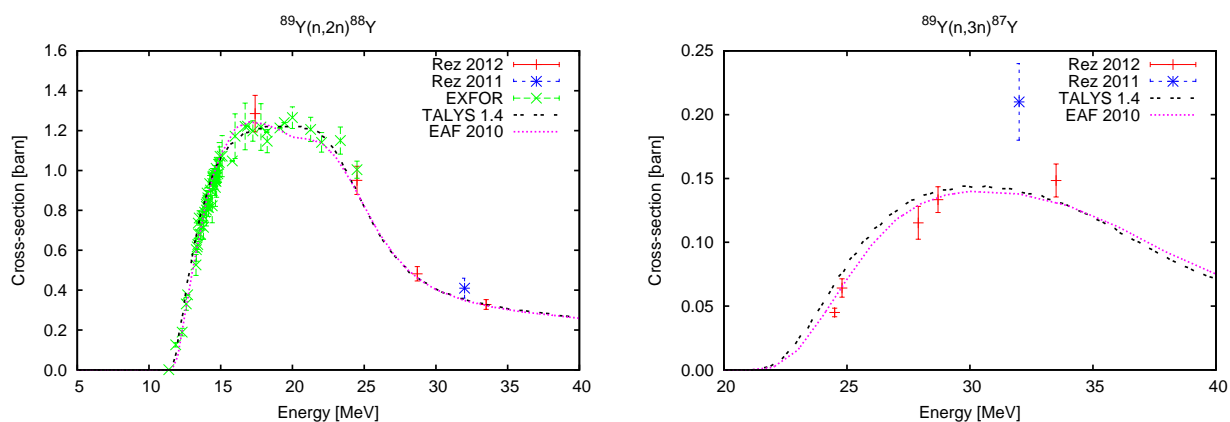


Fig. 3: Cross-sections for (a) $^{89}\text{Y}(n,2n)^{88}\text{Y}$ reaction; (b) $^{89}\text{Y}(n,3n)^{87}\text{Y}$ reaction

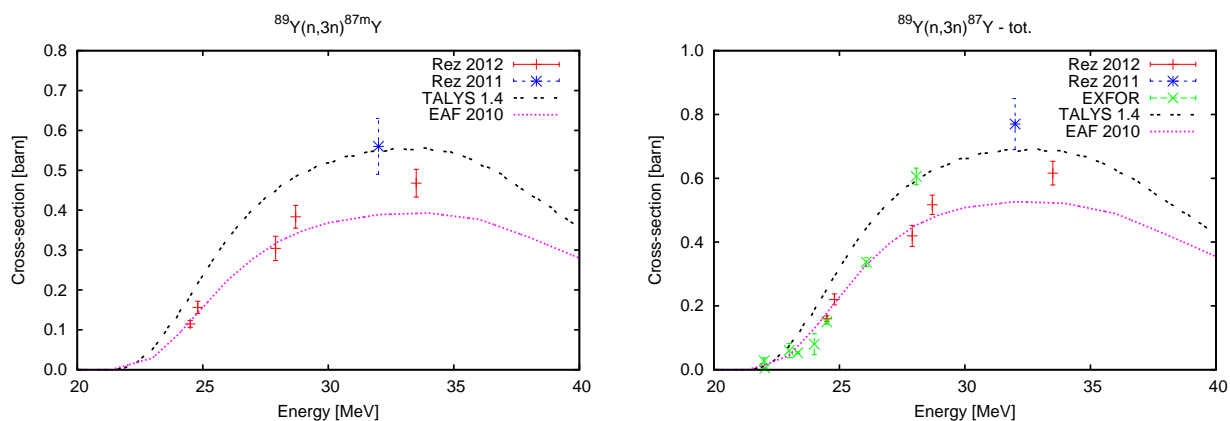


Fig. 4: Cross-sections for (a) $^{89}\text{Y}(n,3n)^{87m}\text{Y}$ reaction; (b) $^{89}\text{Y}(n,3n)^{87}\text{Y}$ reaction - total production

The data shows good agreement both with the TALYS and EAF 2010 [9] cross-sections. For $^{89}\text{Y}(n,3n)^{87m}\text{Y}$ reaction there is a shift in absolute value, but the shape agrees very well with the TALYS calculations. In case of the EAF 2010 library and reaction $^{89}\text{Y}(n,3n)^{87m}\text{Y}$, there is possible to see difference even in shape. Fortunately, except of the highest energy, all the measured neutron energies were so close to the threshold, that the effect of the background subtraction was less or equal than 1 %. Due to this fact, it is possible to expect accurate description of the cross-section behavior in the measured region of energies. Difference between background subtraction using TALYS results and EAF 2010 library are negligible in our accuracy, so we kept to use TALYS, due to the better agreement with the shape in case of $^{89}\text{Y}(n,3n)^{87m}\text{Y}$ reaction.

4 Conclusion

Using the NPI quasi-monoenergetic $^7\text{Li}(p,n)^7\text{Be}$ neutron source, six irradiation of yttrium samples with neutron energies 17.4, 24.5, 24.8, 27.9, 28.7 and 33.5 MeV were made. Obtained cross-sections are in this contribution. The agreement of obtained $^{89}\text{Y}(n,2n)^{88}\text{Y}$ cross-sections and the cross-sections from EXFOR shows good applicability of the discussed method of background subtraction. The systematic shift between the data 'Rez 2012' and 'Rez 2011' is most probably due to different method of obtaining

the neutron spectra. The difference will be subject of further analysis. The data are still preliminary, but there should not be significant changes in the results.

Acknowledgements

On this place, we would like to thank the personnel of fast neutron generators at the NPI and the NPI cyclotron staff for excellent beam quality and service. This experiment was supported by ERINDA and CANAM programs.

References

- [1] P. Bem, V. Burjan, J. Dobes, U. Fischer, M. Gotz, M. Honusek, V. Kroha, J. Novak, S. Simakov, E. Simeckova, The npi cyclotron-based fast neutron facility, in: International Conference on Nuclear Data for Science and Technology 2007, 2007.
- [2] J. Vrzalova, O. Svoboda, A. Krasa, A. Kugler, M. Majerle, M. Suchopar, V. Wagner, Studies of (n,xn) cross-sections in al, au, bi, cu, fe, i, in, mg, ni, ta, y, and zn by the activation method, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment vol. 726 (2013) 84–90.
- [3] A. V. Prokofiev, J. Blomgren, O. Bystrom, C. Ekstrom, S. Pomp, U. Tippawan, V. Ziemann, M. Osterlund, The tsl neutron beam facility, Radiation Protection Dosimetry vol. 126 (1-4) (2007-05-13) 18–22.
- [4] R. A. Schwarz, L. L. Carter, Visual editor to create and display mcnp input files, Journal Name: Transactions of the American Nuclear Society.
- [5] G. X-6, Mcnpx 2.3.0 - monte carlo n-particle transport code system for multiparticle and high energy application (2002).
- [6] O. Svoboda, Experimental study of neutron production and transport for adtt, Dissertation thesis, Czech Technical University in Prague (2011), http://ojs.ujf.cas.cz/~wagner/transmutace/diplomky/PHD_Svoboda.pdf.
- [7] A. J. Koning, S. Hilaire, M. C. Duijvestijn, Talys-1.0, in: O. Bersillon, F. Gunsing, E. Bauge, R. Jacqmin, S. Leray (Eds.), Proceedings of the International Conference on Nuclear Data for Science and Technology, April 22 to April 27, 2007, Nice, France, EDP Sciences, 2008, pp. 211–214.
- [8] B. Geier, Experimental determination of neutron cross sections of yttrium by activation method, Bachelor thesis, Graz University of Technology (2011), http://ojs.ujf.cas.cz/~wagner/transmutace/diplomky/BakkArbeit_last.pdf.
- [9] J.-Ch. Sublet, L. W. Packer, J. Kopecky, R. A. Forrest, A. J. Koning, D. A. Rochman, The European Activation File: EAF-2010 neutron-induced cross section library, CCFE Report, 2010, CCFE-R (10) 05.