Studies of neutron cross-sections important for spallation experiments using the activation method

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Abstract. A series of experiments devoted to studies of neutron cross-sections by activation method was carried out. The cross-sections of various threshold reactions were studied by means of different quasi-monoenergetic neutron sources with energies from 14 MeV up to 100 MeV. Threshold reactions in various materials are among others used to measure fast neutron fields produced during accelerator driven system studies. For this reason our measurements of neutron cross-sections are crucial. At present, neither experimental nor evaluated data above 30 MeV are available for neutron threshold reactions in Au, I and In published in this proceedings. We studied materials in the form of thin foils and compared our data with the calculations performed using the deterministic code TALYS 1.4.

1. Introduction

Suitable activation detectors are necessary for monitoring of neutron fields in different applications. One example is the study of the international collaboration “Energy and Transmutation of Radioactive Waste” at Joint Institute for Nuclear Research Dubna, Russia. The results of the experiments with activation detectors that were carried out with the spallation setup QUINTA [1] are presented also in these proceedings. This contribution is focused on the experiments carried out at The Svedberg Laboratory (TSL) in Uppsala, Sweden and Nuclear Physics Institute (NPI) in Řež, Czech Republic, especially on the determination of cross-sections of threshold reactions, which were not published in [2-4]. We would like to mention that the values published in [2], denoted as \(^{89}\text{Y}(n,3n)^{87}\text{Y}\) are actually the ground state \(^{87}\text{Y}\) production cross-section (not the total cross-sections).

2. Measurements and data analysis

Proton beam energies from 25 to 97 MeV were used at TSL and proton beam energies from 20 to 38 MeV at NPI. The activation materials Au, Bi, I, In and Ta were in the form of foils with dimensions from 1.5 x 1.5 cm\(^2\) up to 3 x 3 cm\(^2\) and thickness from 50 µm up to 1 mm. The iodine samples were in the form of KIO\(_3\) powder pressed into pellets.

The activation method and \(\gamma\)-ray spectrometry were used for cross-section determination. Measured gamma spectra were analyzed by the DEIMOS code [5]. After identification of the isotope to a gamma peak the yield of activated material was calculated. The cross-sections were determined taking into account the number of atoms in a sample and neutron fluence [2]. Neutron spectra in NPI experiments...
were estimated by interpolating the data from an identical setup [6], neutron spectra in TSL experiments were calculated using the algorithm from [7] for the measured proton energies. The second approach was also applied to determining the neutron spectra in NPI with the help of MCNPX simulation [4]. The presented uncertainties of the neutron energies are actually FWHM of the mono-energetic peaks.

| Table 1. Reactions on tantalum, bismuth, natural indium, iodine and gold. |
|------------------------|------------------------|------------------------|------------------------|------------------------|------------------------|
| Reaction Isotopes      | 181Ta (n,2n) 180Ta     | 209Bi (n,5n) 205Bi     | 209Bi (n,10n) 208Bi    | natIn (n,xn) 113mIn   | natIn (n,xn) 112mIn   |
| Half-life              | 8.152(6) h          | 15.31(4) d            | 36.4(5) m             | 1.6582(6) h           | 20.56(6) m            |
| Eγ [keV]               | 93.326(2)           | 1764.36(4)            | 1026.5(2)             | 391.690(15)           | 156.56(10)            |
| Intensity [%]          | 4.5                  | 32.5(6)               | 100                   | 64.2                  | 13.2(3)               |
| En [MeV] cross-section [barn] |
| 17.5(8)                | ---                  | ---                   | 0.023(3)              | 0.043(7)              | ---                   |
| 30.4(8)                | ---                  | ---                   | 0.159(22)             | ---                   | ---                   |
| 47.0(6)                | 0.139(25)            | 0.81(13)              | ---                   | 0.28(7)               | ---                   |
| 59.0(4)                | 0.059(10)            | 0.30 (5)              | ---                   | 0.019(3)              | ---                   |
| 66.4(4)                | 0.040(7)             | 0.19 (3)              | 0.016(4)              | ---                   | 0.24(4)               |
| 72.8(4)                | 0.042(10)            | 0.151(28)             | 0.0169(29)            | ---                   | 0.174(26)             |
| 89.3(7)                | ---                  | 0.104(18)             | 0.0158(26)            | ---                   | 0.078(12)             |
| 94.0(7)                | ---                  | 0.152(25)             | 0.040(6)              | ---                   | 0.075(25)             |

The high energy neutron source based on proton reactions on a lithium target has the energy spectrum which contains besides the main neutron peak also lower-energy continuum. When determining the neutron fluence we subtracted the contribution of these “background” neutrons (all neutrons below the mono-energetic peak) using the TALYS code. It is estimated as the ratio between the folding of calculated cross-section and neutron spectrum in the peak and the same convolution in the whole spectrum (for more details, see [2]).

The procedure is insensitive to the absolute value of the TALYS [8] cross-section, but sensitive to the shape of cross-section dependency on neutron energy. If we use TALYS models with different nuclear level density, the shapes of cross-section dependency on energy will change. We can calculate with five preset options of level densities; with three phenomenological level density models and two options for microscopic level densities (see figure 2b). Uncertainty caused by the influence of different level density models does not exceed 10 % and was added to the total cross-section uncertainty.

3. Results
The numerical values of measured cross-sections are in table 1 and in figures 1-2 are their graphical representation. In table 1 are also published decay data [9] for the reactions used in this work. New data were obtained in the energy regions where no data existed up to now. The experimental results are compared with model calculations using the TALYS 1.4 code (basic setting). The cross-sections of the reactions 181Ta(n,2n)180Ta, 206Bi(n,5n)208Bi and 206Bi(n,10n)208Bi are also compared with the data from the EXFOR database (for Ta [10]; for Bi [11]). Our data show good agreement with TALYS in cases of In, I and reaction (n,5n) on Bi. In case of the 181Ta(n,2n)180Ta, 197Au(n,7n)191Au and 209Bi(n,10n)208Bi reactions the shapes agree with the TALYS calculation, however, the measured cross-sections are smaller (approximately by 40% for Ta, by 50% for Au and even by 80% for (n,10n)

* These data have been already published in [2].
reaction on Bi), but we can see in figure 3a that our data are in agreement with other experimental data [11].

Figure 1. (a) Cross-section of $^{nat}\text{In}(n,xn)^{112m}\text{In}$ (b) Cross-section of $^{181}\text{Ta}(n,2n)^{180}\text{Ta}$ reaction.

Figure 2. (a) Cross-section of $^{127}\text{I}(n,8n)^{120}\text{I}$ (b) Cross-section of $^{197}\text{Au}(n,7n)^{191}\text{Au}$ reaction (TALYS calculations with five available densities are shown as well).

Figure 3. (a) Cross-section of $^{209}\text{Bi}(n,10n)^{206}\text{Bi}$ (b) Cross-section of $^{nat}\text{In}(n,xn)^{113m}\text{In}$ reaction.

The main contributions to the total uncertainty of cross-sections are uncertainties of Gauss-fit of peaks in DEIMOS (1-10%), detector efficiency (3%) beam intensity and neutron spectra (10% for both sources).
4. Conclusion

We present measurements of neutron-induced reaction cross-sections of the reactions \((n,xn)\) on natural indium, \((n,2n)\) on tantalum, \((n,5n)\) on bismuth, \((n,10n)\) on bismuth, \((n,8n)\) on iodine and \((n,7n)\) on gold from 17 up to 94 MeV. Overall uncertainties in the best cases are near 15%. Our data were compared with data from the EXFOR database and with the TALYS 1.4 code. A good agreement between our data and TALYS was observed for In, I and \((n,5n)\) on Bi, while TALYS predicts significantly higher values than the measured ones for Au, Ta and \((n,10n)\) on Bi. The \((209\text{Bi}(n,5n)205\text{Bi})\) and \((209\text{Bi}(n,10n)200\text{Bi})\) are the only cases where earlier experimental data are available and they agree with each other. No evaluated data libraries contain cross-sections of the presented reactions on Au, In and I in the measured energy regions.

Using our measured cross-sections, it becomes possible to better analyze neutron fluxes in different places of experimental setups consisting of lead, natural uranium, and graphite irradiated by relativistic protons and deuterons to study transmutation of radioactive materials by spallation neutrons at JINR Dubna. Such studies are intended for providing important data serving for the construction of larger-scale Accelerator Driven Systems (ADS).

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