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Measurement of neutron-induced activation cross-sections using spallation source at JINR and neutronic validation of the Dubna code

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Abstract. A beam of 1 GeV proton coming from Dubna Nuclotron colliding with a lead target surrounded by 6 cm paraffin produces spallation neutrons. A Th-foil was kept on lead target (neutron spallation source) in a direct stream of neutrons for activation and other samples of ^{197}Au, ^{209}Bi, ^{59}Co, ^{115}In and ^{181}Ta were irradiated by moderated beam of neutrons passing through 6 cm paraffin moderator. The gamma spectra of irradiated samples were analyzed using gamma spectrometry and DEIMOS software to measure the neutron cross-section. For this purpose neutron fluence at the positions of samples is also estimated using PREPRO software. The results of cross-sections for reactions ^{232}Th(n,γ), ^{232}Th$(n,2n)$, ^{197}Au(n,γ), ^{197}Au(n,α), ^{197}Au(n,xn), ^{59}Co(n,α), ^{59}Co(n,xn), ^{181}Ta(n,γ) and ^{181}Ta(n,xn) are given in this paper. Neutronics validation of the Dubna Cascade Code is also done using cross-section data by other experiments.

Keywords. Spallation neutrons; PREPRO; DEIMOS; activation cross-section; (n,xn) reactions; Dubna Cascade Code.

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1. Introduction

At an energy less than 20 MeV, experimental neutron cross-sections are available for so many years and at higher energies such cross-sections are available very rarely [1]. For conventional reactors, (n,xn) reactions play a role up to $x=2$ and in the case of accelerator driven sub-critical systems, because of high-energy neutrons the (n,xn) reactions play a very important role due to neutron multiplication as well as competition of (n,f) and (n,xn) reactions [2]. Measurements of the high-energy

Table 1. Geometrical details of Co- and Ta-activation detectors used in Gamma-2 experiment.

Element	^{181}Ta	^{59}Co
Cross-sectional area	2.64 cm^2	1.23 cm^2
Thickness (mm)	0.431	3.5
Weight (g)	1.896	3.831
Density (g/cm^3)	16.66	8.9
Atoms/cm^2	2.38×10^{21}	3.16×10^{22}
Waiting period, t_2	4.57 h	3.79 h

cross-sections for a variety of materials are required not only from the point of nuclear studies but also for the selection of both structure and fuel material of the ADS. In the absence of mono-energetic high-energy neutron sources the available quasi-energetic [1] or even the wide spectrum spallation source may provide useful information for the preliminary design and modeling of ADS like it was used in the early stage of design and modeling of the reactors.

In our earlier experiment Gamma-2, a solid lead (Pb) target set-up was irradiated to 1 GeV proton beam from the Nuclotron (detailed description of the basic parameters of the Nuclotron can be found elsewhere [3]). Details of the complete set-up and preliminary data analysis are given in refs [4–6]. Fluences of the spallation and the moderated neutrons in some positions of the target had been measured using the activation detectors and these results have been reported in an earlier work [7] which were also found in good agreement with the Monte-Carlo calculations performed by Barashenkov *et al* [8] using Dubna Cascade Code. In ref. [7] one can see the details of the activated detectors, Au, Bi, and Th as well as the methodology of analysis for the estimation of 'one group cross-section'. Further details of the Co- and Ta-activation detectors are given in table 1.

2. Experimental results

2.1 *Measurement of the neutronic fluence*

The neutron fluence was measured at selected positions of (a) inner cylindrical surface of the lead spallation target, (b) outer cylindrical surface of the paraffin moderator and (c) the circular downstream end (outside the paraffin moderator). Results of measurement of the overall fluence in the first two cases (a) and (b) have been published in an earlier work [7] and the fluence in case (c) is estimated as follows. A sample of tantalum of size $1.6 \times 1.65 \times 0.0431$ cm^3 was activated by the moderated neutrons escaped from the circular downstream end of the paraffin under the experiment Gamma-2 in the year 2002 and its data of gamma spectrometry are analyzed recently to validate the code in downstream direction. The energy distribution of the moderated neutrons passing through the Ta-sample simulated from the Dubna Cascade Code is given in figure 1.

The overall fluence of these neutrons is estimated using the following two reactions:

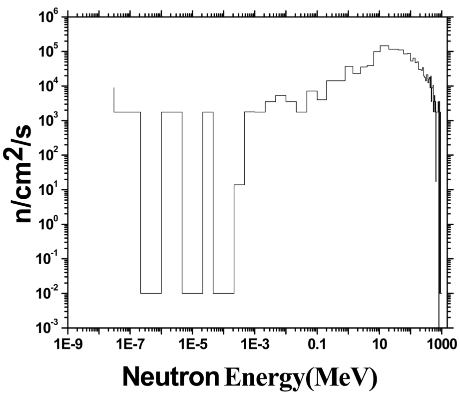

Figure 1. Fluence passing through the Ta-sample placed at the circular downstream face (outer side of paraffin) at 20° to the line joining the two paraffin dees.

(i) For the ^{181}Ta$(n,\gamma)^{182}$Ta reaction, the product nucleus ^{182}Ta emits a prominent γ-ray with energy $E_\gamma = 67.75$ keV. The one group cross-section for this reaction for the simulated energy distribution from the software PREPRO [10] using the ENDF-VI.1 library comes out to be 1.13 b. This in turn gives an average neutron fluence of $1.24 \pm 0.29 \times 10^6$ (n/cm^2/s) on the Ta-detector position.

(ii) Similarly, in the case of ^{181}Ta$(n,2n)^{180}$Ta reaction, the product nucleus ^{180}Ta emits a prominent gamma with energy $E\gamma = 93.3$ keV. The one group cross-section for this reaction from PREPRO comes out to be 0.612 b which in turn gives a neutron fluence of $1.37 \pm 0.06 \times 10^6$ (n/cm^2/s) which verifies the earlier fluence. The average value of the two fluences in position of the Ta-detector comes out to be $1.36 \pm 0.06 \times 10^6$ (n/cm^2/s) which is close to that given by the Dubna simulation from the Dubna Cascade Code, i.e. 1.43×10^6 (n/cm^2/s). Simulation errors depend on the number of histories and are very small. This has been given in table 2 along with that were obtained in other radial directions in earlier publications.

2.2 *Spectrum-averaged cross-sections of different reactions*

Measured neutron fluences (n/cm^2/s) in selected positions of (a) inner cylindrical surface of lead spallation target, (b) outer cylindrical surface of the paraffin moderator and (c) at the circular downstream end outside the paraffin moderator along with the values from the Dubna Cascade are given in table 2 and the fluence values which are given in table 2 have been used for the estimation of spectrum-averaged cross-sections of (i) ^{232}Th(n,γ), (ii) ^{232}Th$(n,2n)$ reactions by using fluence $1.496 \pm 0.087 \times 10^7$ (n/cm^2/s) corresponding to the downstream end of the cylindrical surface of NSS in between lead and paraffin, (iii) ^{197}Au(n,γ), (iv) ^{197}Au(n,α), (v) ^{197}Au$(n,6n+p)$, (vi) ^{197}Au(n,xn) for $x = 2,4,6,7$ and 8, (vii) ^{59}Co(n,xn)

Table 2.

Detector and its position	Fluence (experimental) (n/cm^2/s)	Fluence (Cascade) (n/cm^2/s)
(a) Th at the inner cylindrical surface	$1.496 \pm 0.087 \times 10^7$	1.46×10^7
(b) Au, Co, Bi at the outer cylindrical surface of paraffin	$0.657 \pm 0.016 \times 10^7$	0.64×10^7
(c) ^{181}Ta at the circular downstream end of paraffin	$1.37 \pm 0.06 \times 10^6$	1.43×10^6

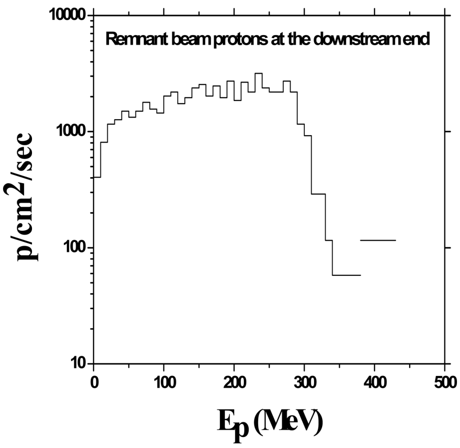

Figure 2. Energy spectrum of remnant protons.

for $x = 2, 4$ and 5, (viii) ^{59}Co(n, α) reactions by using fluence $0.657 \pm 0.016 \times 10^7$ (n/cm^2/s) corresponding to the outer cylindrical surface of paraffin and similarly (ix) ^{181}Ta(n, γ), (x) ^{181}Ta(n, pn), (xi) ^{181}Ta(n, xn) for $x = 2$, 4 and 5 reactions by using the value of neutron fluence $1.3647 \pm 0.0597 \times 10^6$ (n/cm^2/s) corresponding to the circular downstream end of paraffin.

Additional spectrum averaged cross-sections of ^{232}Th, ^{197}Au and that of ^{59}Co and ^{181}Ta of the present work using the corresponding n-fluences are listed in table 3. The values given with the * are after correcting for the background reactions which may give the same gamma in this experimental environment where remaining protons are also in existence. Their spectrum deduced from the code is given in figure 2.

3. Discussion and conclusions

1. In the case of the Ta-sample which is positioned at the downstream end there are definite chances that the remnant beam protons also initiate a reaction in Ta which has same residual nucleus as was available in a (n, xn) reaction. For example, ^{180}Ta nucleus obtained in the case of the ^{181}Ta$(n, 2n)^{180}$Ta reaction may

Table 3. Spectrum averaged cross-sections.

Reaction	E_γ (keV)	σ
^{232}Th$(n,\gamma)^{233}$Th ^{233}Th => ^{233}Pa	311.9	72.18 ± 0.36 mb
^{232}Th$(n,2n)^{231}$Th	84.22	0.262±0.09 b
^{197}Au$(n,\gamma)^{198}$Au	411.80	26.9±0.67 b
^{197}Au$(n,\alpha)^{194}$Ir	328.40	7.72 ± 0.05 mb
^{197}Au$(n,p+6n)^{191}$Pt	538.86	241±3.7 mb
^{197}Au$(n,2n)^{196}$Au	355.69	167.9±6.9 mb
^{197}Au$(n,4n)^{194}$Au	328.40	59.29±2.7 mb
^{197}Au$(n,6n)^{192}$Au	316.50	33.67±1.1 mb
^{197}Au$(n,7n)^{191}$Au	586.45	29.63±2.0 mb
^{197}Au$(n,8n)^{190}$Au	295.8	08.83±0.14 mb
^{59}Co$(n,\alpha)^{56}$Mn	846.77	3.46±0.16 mb
^{59}Co$(n,2n)^{58}$Co	810.81	98.0±2.69 mb
^{59}Co$(n,4n)^{56}$Co	846.77	1.70±0.12 mb
^{59}Co$(n,5n)^{55}$Co	931.5	0.33±0.06 mb
^{181}Ta$(n,\gamma)^{182}$Ta	67.75	1.03±0.05 b
^{181}Ta$(n,2n)^{180}$Ta	93.3	0.619±0.03 b (0.507 b)*
^{181}Ta$(n,4n)^{178\mathrm{m1}}$Ta	426.38	0.304±0.013 b (0.252 b)*
^{181}Ta$(n,5n)^{177}$Ta	112.95	0.130±0.006 b (0.077 b)*
^{181}Ta$(n,pn)^{180\mathrm{m}}$Hf	332.28	0.053±0.004 b

also be produced in ^{181}Ta$(p,pn)^{180}$Ta reaction. In this situation, Ta(n,xn) cross-sections given in table 3 may be treated as overestimated. In order to estimate the contribution of ^{181}Ta$(p,pn)^{180}$Ta reaction, both the energy distribution of remnant beam protons escaping through the Ta-sample and their reaction cross-sections are simulated by the Dubna Cascade Code. The energy spectrum of the remnant beam protons is given in figure 2. In this way, reaction cross-sections of ^{181}Ta$(p,pn)^{180}$Ta, ^{181}Ta$(p,p3n)^{178}$Ta and ^{181}Ta$(p,p4n)^{177}$Ta come out to be 112.2, 51.75 and 53.1 mb respectively. Thus, the corrected values of cross-sections corresponding to the ^{181}Ta$(n,2n)^{180}$Ta, ^{181}Ta$(n,4n)^{178\mathrm{m1}}$Ta and ^{181}Ta$(n,5n)^{177}$Ta reactions are 507 mb, 252 mb and 77 mb respectively.

On the cylindrical side, the number of protons scattered out of the spallation target vary from 0.2% at the upstream end to 1.3% at the downstream end compared to the scattered neutrons. After loosing energy by ionization process in paraffin the proton energy will reduce drastically. In this way error introduced in (n,xn) reaction cross-section measurement due to the protons will be negligibly small.

2. In all reactions studied above, one must consider possible errors caused by the use of evaluated values of cross-section data by PREPRO in the high-energy range ($E > 20$ MeV) because no experimental data are known in this energy range.

3. PREPRO does not give errors for the 'flat spectrum averaged cross-sections' [7] in an energy group corresponding to the errors associated with the individual experimental data point. This would enhance uncertainty of the measured fluence and hence of cross-section.

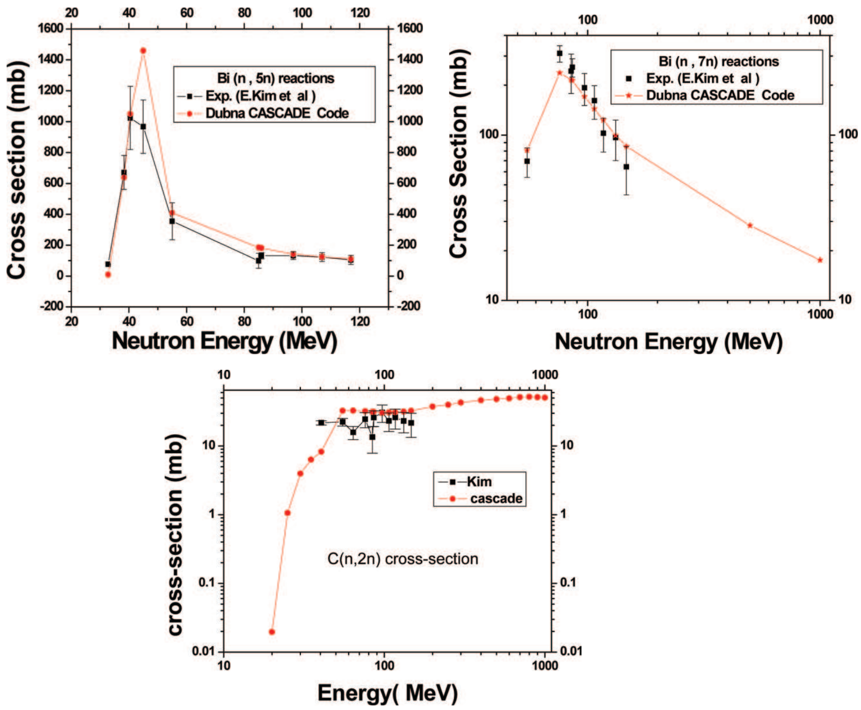

Figure 3. Comparison of the cross-sections from experiments and the Dubna Code.

4. Using the neutron energy profile similar to that used by Kim *et al* [1] we have generated cross-sections from the Dubna Code they are in agreement with that of Kim *et al* [1] (see figure 3). This further validates the Dubna Code with respect to the neutronics.

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