PROCEEDINGS OF SCIENCE

# Measurements of fast neutron spectrum in QUINTA assembly irradiated with 2,4 and 8 GeV deuterons

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Collaboration **E&T - RAW**

Experimental values of high energy neutron flux by application Yttrium-89 foils in QUINTA assembly, using 2, 4 and 8 GeV deuteron beams (Fig. 1) from the JINR Nuclotron (2012 E+T – RAW experiments) are presented. Evaluation of average high energy neutron fluxes for three energy ranges (11,5-20.8, 20,8-32.7, 32,7-100 MeV) using the determined three isotopes production of $^{88}$Y $^{87}$Y and $^{86}$Y [1] are performed. The (n,xn) reaction rates of yttrium samples located inside the assembly were determined through gamma spectrometry. We stared to check values of cross section data for Y-89 neutron reactions (n,xn) experimentally.

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## Introduction

Investigation of physical aspects of radioactive waste transmutation, using hadron relativistic beams (deuteron beams in the energy range to 8 GeV) was undertaken within the project „Quinta“ in the Joint Institute for Nuclear Research (JINR), Dubna, Russia. The QUINTA uranium target is a deeply sub-critical active core consisting of 512 kg of natural uranium rods arranged hexagonally.

To study the fast neutron energy spectrum we have used Yttrium foils (Y-89) which have the following advantages: one stable isotope - $^{89}$Y, easy way to make good shape samples, several resulting isotopes ($^{88}$Y, $^{87}$Y, $^{86}$Y, $^{85}$Y and $^{84}$Y) with long enough half life time – longer than 12 hours, obtained in the (n,xn) reactions for several threshold reactions (11.5, 20.8, 32.7, 42.1 and 54.4 MeV). In order to evaluate the fast neutron energy spectrum the Y-89, was irradiated with 2 GeV, 4 GeV and 8 GeV deuteron beam. The neutron field was determined by twelve Y-89 foils placed in specified positions (given by the radial and axial distance) inside the natural uranium blanket target facility. After irradiation by deuterons, the gamma activity of Y-89 detectors were measured using HPGe spectrometer. Taking into account necessary corrections we have determined the isotope production per one gram of sample and per one beam deuteron – B parameter. Having already determined isotope production per one gram of sample and per one beam deuteron at specified positions of the Quinta setup for the three isotopes Y-88, Y-87 and Y-86 we can evaluate three average high energy neutron fluxes in each Yttrium foils (Y-89) location for certain energy ranges.

## 1. Experiments

The Assembly Quinta “ready to use”, located at the Joint Institute for Nuclear Research (JINR), has been available for the E&T RAW collaboration since the end of the year 2009. The “QUINTA” assembly (Fig.1) has five sections of a hexagonal shape. Each section consists of 61 fuel rods of natural uranium inside an aluminum cover (36 mm diameter, 124 mm length, weight of 1.64 kg). Each section contains 100 kg of uranium, so the whole target contains around 500 kg. The first section have a hole in its center, a beam window. Target was surrounded by a massive lead shielding of total weight 1780 kg. The front of the lead castle has a square window 150 x 150 mm. The $^{89}$Y activation detectors were placed on the detector plates in front of, between the five sections, and on the rear of the setup in two radial distances (4 and 8 cm) from the beam axis (Fig.2).

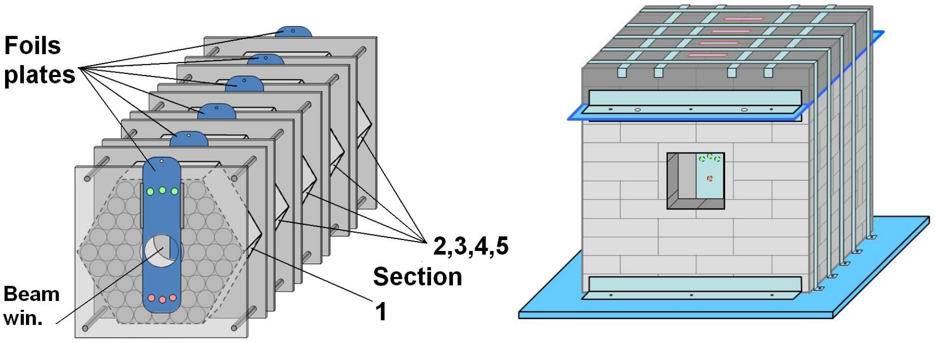

Fig. 1. Schema of The QUINTA setup. Left: the uranium target. Blue means aluminum holder for detectors. Right: the lead shielding enfolding the target.

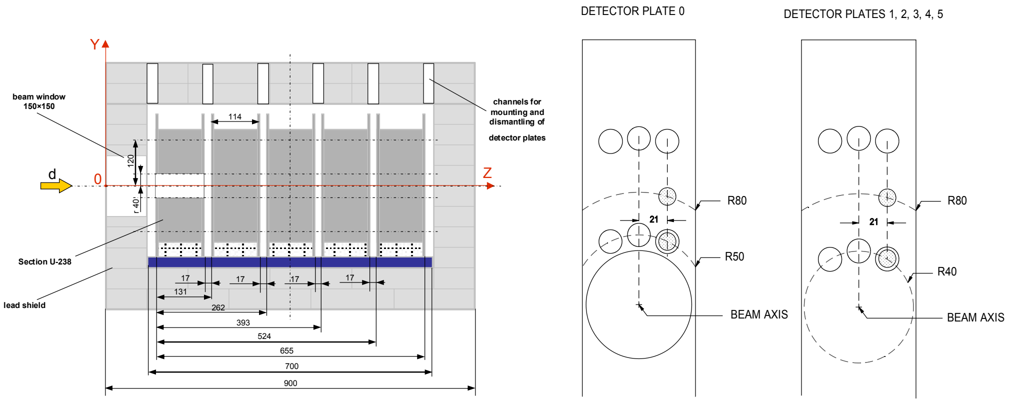

Fig. 2. Dimensions of Quinta assembly and location of yttrium foils . On the left there is a longitudinal view of the assembly, on the right there are yttrium foils locations on the detector plates.

The Quinta target was irradiated with a pulsed deuteron beam of energies 2, 4 and 8 GeV in December 2012. Total number of deuterons of the three irradiations are equal to 3.02(30)x10$^{13,}$, 2.73(27)x10$^{13}$ and 0.91(9)x10$^{13}$ during the time of irradiation equal to 6.27h, 9.35h and 16.7h, respectively. The total number of deuterons to hit the Quinta target was determined by the activation of aluminium via beam induced $^{27}$Al(d,3p2n)$^{24}$Na reactions [2].

To measure the number of $^{89}$Y (n,xn) reactions in $^{89}$Y detectors during irradiation of the assembly, we measured gamma spectra from Y-89 foils on germanium detectors. Measurements began ~1.5 h after irradiation had stopped, continuing for up to 6 days afterwards. A program DEIMOS was applied for the spectra analysis [3]. Using this program one performed the gamma line energy calibrations, and determined line absolute intensity and half width of the line (FWHM - Full Width at Half Maximum) and respective errors. Next, the results spectra analysis were corrected for detector efficiency, cascade effect, function for thickness and shape of detectors, and cooling time [4]. We calibrate all the results to B parameters by the below calibration formula.

$$\boxed{B = N_1 \cdot \frac{1}{m \cdot I} \cdot \frac{\Delta S(G) \cdot \Delta D(E)}{\frac{N_{abs}}{100} \cdot \varepsilon_p(E) \cdot COI(E,G)} \cdot \frac{(\lambda \cdot t_{ira})}{[1 - \exp(-\lambda \cdot t_{ira})]} \cdot \exp(\lambda \cdot t_+) \cdot \frac{\frac{t_{real}}{t_{live}}}{[1 - \exp(-\lambda \cdot t_{real})]}}$$

where

| | |
|---|---|
| B | number of nuclei per gram of a sample material and per one primary proton |
| $N_1$ | peak (line) area |
| $N_{abs}$ | the absolute intensity of given line in percent [%] |
| $\varepsilon_p(E)$ | detector efficiency function of energy (polynomial) |
| COI(E,G) | cascade effect coefficient function of energy and geometry |
| ΔS(G), ΔS(G) | calibrations function for thickness and shape of foils |
| I | total number of primary protons |
| $t_{1/2}$ | half life time |
| $t_{ira}$ | elapsed time of irradiation |
| $t_+$ | elapsed time from the end of irradiation to the beginning of measurement |
| $t_{real}$ | elapsed time of the measurement |

$t_{live}$ “live” time of measurement
m mass of the sample (foils) in grams

Finally, we can show each isotope productions in useful 3D graphs. Production rate inside the Quinta assembly [5] was estimated. An example of the isotope production Y-88 presented in Figure 3, show that the Y-89 isotopes production have maximum of axial distribution in the second position of about 13 cm along the natural uranium spallation target.

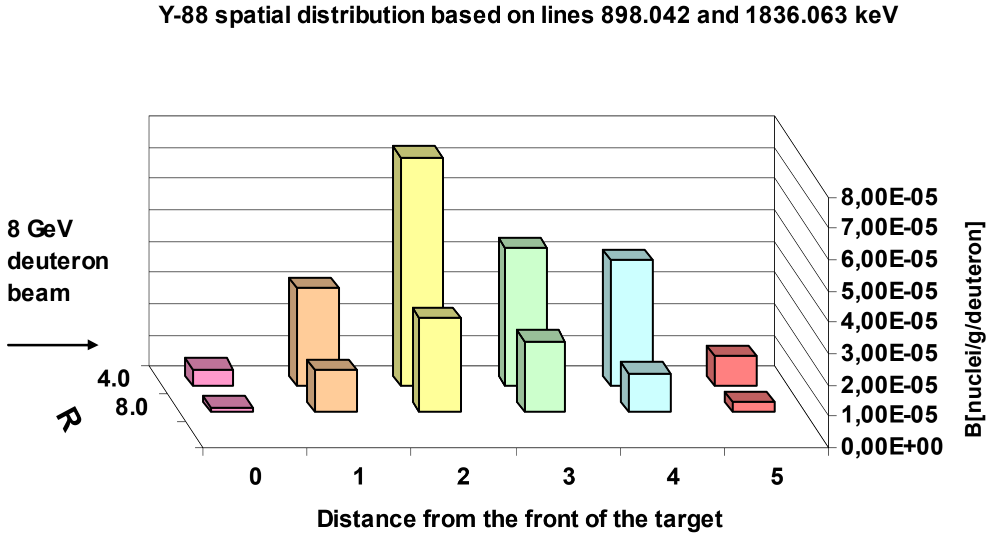

Fig. 3. Spatial distribution (radial & axial) of Y88 production for the deuteron beam 8GeV.

## 2. Evaluation of average high energy neutron flux in the Yttrium-89 detectors location inside the Quinta assembly.

Having already determined isotope production per one gram of sample and per one beam deuteron at specified positions of the Quinta setup for the three isotopes Y-88, Y-87 and Y-86 we can evaluate three average high energy neutron fluxes in each Yttrium-89 detectors location for certain energy ranges. The energy ranges is roughly determined by the microscopic cross section in function of energy for the (n,xn) reactions of the three isotopes $^{88}$Y, $^{87}$Y and $^{86}$Y. The following three threshold energy 11.5, 20.8 and 32.7MeV for the reactions $^{89}$Y(n, 2n)$^{88}$Y, $^{89}$Y (n, 3n)$^{87}$Y and $^{89}$Y(n,4n) $^{86}$Y appoint the first two energy ranges (11.5 - 20.8 MeV) and (20.8 - 32.7MeV) of the neutron fluxes $\overline{\phi_1}$ and $\overline{\phi_2}$. The third energy range begins at the energy 32.7MeV and ends when the microscopic cross section is comparatively low (100 MeV) with the maximum cross section of $^{89}$Y(n,4n) $^{86}$Y reaction where is evaluated the neutron flux $\overline{\phi_3}$.

To calculate the high energy neutron field we need to know the microscopic cross section for the (n,xn) $^{89}$Y reactions. We used TALYS code [6], [7] for cross section calculation because the experimental nuclear data libraries are poor for Yttrium isotope [8]. In general, the number of yttrium isotopes (Y) in the yttrium 89 detector of volume V in the given energy range can be expressed by the formula:

$$Y = V\,\bar{\varphi}\,N\,\bar{\sigma}\,t\ ,$$

where:

$\overline{\varphi}$ - average neutron flux [n/cm$^2$·s]
N – number of $^{89}$Y isotopes in volume unit [cm$^{-3}$]
$\overline{\sigma}$ - average microscopic cross section for the reaction (n,xn) in the energy range (E$_1$–E$_2$) [barns]

Finally, we can write the following three algebraic equations:

$$B^{88}\,C = \bar{\varphi}_1 \bar{\sigma}_{11} + \bar{\varphi}_2 \bar{\sigma}_{12} + \bar{\varphi}_3 \bar{\sigma}_{13}$$
$$B^{87}\,C = 0 + \bar{\varphi}_2 \bar{\sigma}_{22} + \bar{\varphi}_3 \bar{\sigma}_{23}$$
$$B^{86}\,C = 0 + 0 + \bar{\varphi}_3 \bar{\sigma}_{33}$$

where
B$^{88}$, B$^{87}$, B$^{86}$ - measured isotopes of $^{88}$Y, $^{87}$Y and $^{86}$Y respectively per one gram of $^{89}$Y activation foils and per one beam deuteron,
$\overline{\sigma_{11}}\,. - .\overline{\sigma_{33}}$ - average microscopic cross section of the measured isotopes for the reaction (n, xn) in the three chosen energy ranges,
$\overline{\phi_1}, \cdot\overline{\phi_i}\cdot, \overline{\phi_3}$ - unknown average neutron fluxes in the three chosen energy ranges,
and C are a physic constants C=(G$^{89}$ S) / (A t) where
G$^{89}$ - gram-atom of $^{89}$Y,
A - Avogadro's number,
t - deuteron beam irradiation time,
S - total number of deuterons.

The solution of the above three equations let us to evaluate the average neutron fluxes in the three energy ranges expressed in [n/cm$^2$·s] [5]. Example of calculation results we can see in Figure 4a, 4b and 4c. Those three figures coming from experiment with 8GeV beam. In Table.1 are presented (as an example) relevant numerical values of average neutron flux for three energy ranges for deuteron beam 8 GeV. Those data were used to create Figures 4a, 4b, 4c.

## 3. Neutron energy intervals based on $^{89}$Y (n,xn) reaction cross section.

To calculate the high energy neutron field we need to know the microscopic cross section for the $^{89}$Y-(n,xn) reactions. Some cross section data for neutron induced reactions were retrieved from the literature [8] and some cross sections were calculated using the TALYS code [6, 7]. No neutron induced reaction cross section for reaction $^{89}$Y(n,4n) were found in the EXFOR data base or open literature. Only the data for the cross section of $^{89}$Y(n, 2n) and several points of $^{89}$Y(n, 3n)$^{87}$Y reactions we could find [10, 11].

TALYS is a software for the simulation of nuclear reactions. Many state-of-the-art nuclear models are included to cover all main reaction mechanisms encountered in light particle-induced nuclear reactions. TALYS provides a complete description of all reaction channels and observables, and is user-friendly. TALYS is a versatile tool to analyse basic microscopic experiments and to generate nuclear data for applications. It calculates total and partial cross-sections, energy spectra, angular distributions, double-differential spectra, residual production cross sections, and recoils. It has been tested with experimental data with very good results.

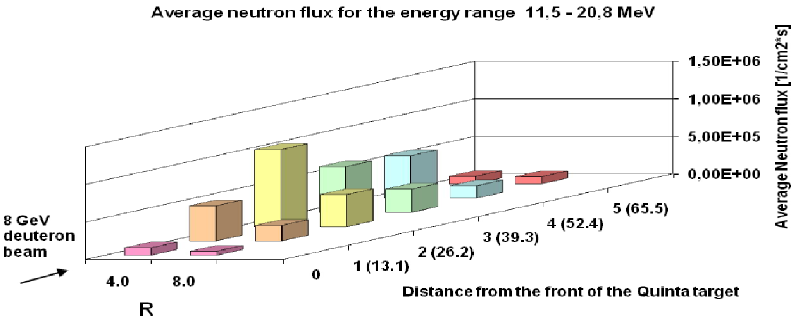

Fig. 4a. Spatial average neutron flux distribution in the Quinta assembly for the neutron energy range (11.5 – 20.8) MeV for the deuteron beam of 8.0 GeV. Distance are in [cm].

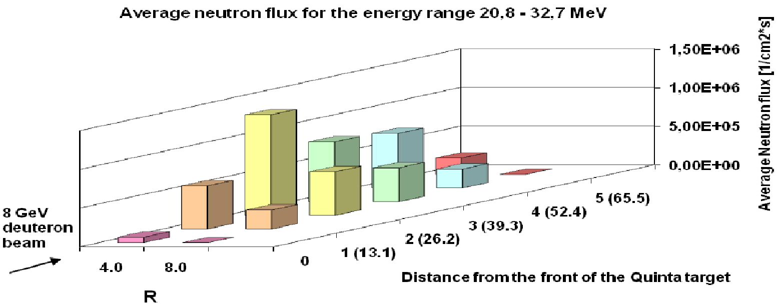

Fig. 4b. Spatial average neutron flux distribution in the Quinta assembly for the neutron energy range (20,8 – 32.7) MeV for the deuteron beam of 8.0 GeV. Distance are in [cm].

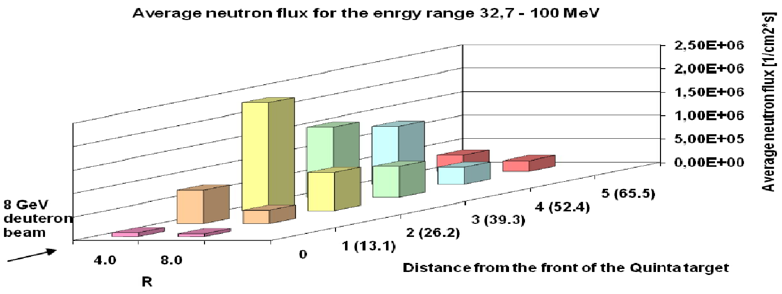

Fig. 4c. Spatial average neutron flux distribution in the Quinta assembly for the neutron energy range (32,7 – 100) MeV for the deuteron beam of 8.0 GeV. Distance are in [cm].

Tab. 1 Numerical values of average neutron flux for three energy ranges for deuteron beam 8 GeV inside QUINTA assembly in december 2012.

| **Beam (8.0 GeV) Energy range [MeV]** | **Radius [cm]** | **Average neutron flux from Y89 detectors [1/cm2•s]*(10E+05)** Axial position [cm] | | | | | |
|---|---|---|---|---|---|---|---|
| | | **0** | **1 (13.1)** | **2 (26.2)** | **3 (39.3)** | **4 (52.4)** | **5 (65.5)** |
| Neutron flux1<br>11,5- 20,8 | 4.0 | 0.98± 0.3 | 4.7 ±2.6 | 10.3 ±5.7 | 6.1 ±3.5 | 5.7 ±1.8 | 1.0 ±0.8 |
| | 8.0 | 0.53 ±0.5 | 2.1 ±1.1 | 4.3 ±1.6 | 3.1 ±1.2 | 1.6 ±0.5 | 0.96 ±0.7 |
| Neutron flux 2<br>20,8-32,7 | 4.0 | 0.71 ±0.05 | 5.6 ±0.3 | 13,0 ±1.1 | 7.7 ±0.5 | 7.1 ±0.8 | 2.1 ±0.2 |
| | 8.0 | 0.12 ±0.04 | 2.5 ±0.2 | 5.6 ±0.4 | 4.4 ±0.3 | 2.5 ±0.2 | 0.4 ±0.5 |
| Neutron flux 3<br>32,7-100 | 4.0 | 0.86 ±0.06 | 7.1 ±0.4 | 22,9 ±0.1 | 15.0 ±0.7 | 12.4 ±0.8 | 3.4 ±0.2 |
| | 8.0 | 0.62 ±0.05 | 2.9 ±0.2 | 8.2 ±0.5 | 6.7 ±0.4 | 3.7 ±0.2 | 2.2 ±0.2 |

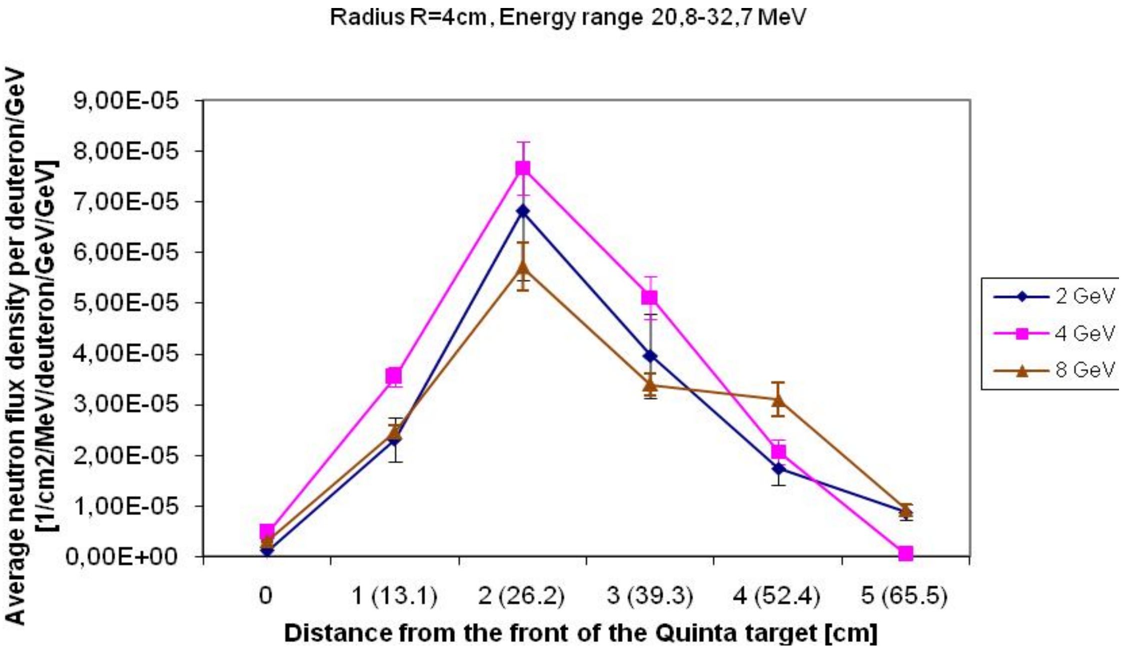

Fig. 5. Average neutron flux density per deuteron and its energy in function of Quinta target axis at R=4 cm for three deuteron energies (2, 4, 8 GeV) in the neutron energy range 20.8-32.7 MeV

The TALYS code was used to generate microscopic cross sections for several $^{89}$Y(n, xn) reactions. Comparison of experimental microscopic cross sections of the reaction $^{89}$Y(n, 2n)$^{88}$Y and the reaction $^{89}$Y(n, 3n)$^{87}$Y with the generated microscopic cross sections for the two reactions shows very good agreement. This gives us confidence in the calculated cross sections by TALYS code of the $^{89}$Y (n, 3n)$^{87}$Y and $^{89}$Y(n, 4n)$^{86}$Y reactions which we have checked experimentally by selection anyway. That is why we have performed measurement of neutron reaction cross-sections for selected energies of these reactions by means of quasi mono-energetic neutron sources at Nuclear Physics Institute (NPI) in Řež, Czech Republic (2012) and at The Svedberg Laboratory (TSL) in Uppsala, Sweden (2011).

The measurements were done using six different proton energies. Positions of quasi-mono-energetic neutron peak were 17.4, 24.5, 24.8, 27.9, 28.7 and 33.5 MeV. The samples were fixed on an aluminum holder which was mounted behind the neutron source. Gold samples were irradiated together with the yttrium samples. The gold samples with much better known cross-sections of neutron reactions were used as the experimental condition monitors For each irradiated sample, the neutron spectrum was calculated by means of MCNPX.

In addition to the experimental cross section data retrieved from the EXFOR data base for (n,2n) reaction and several points for (n,3n) reaction we have collected data for 32 MeV from and for 59.0 to 89.3 MeV from [12, 13] together with our measurement for 17.4, 24.5, 24.8, 27.9, 28.7 and 33.5 MeV neutron energies. The data shows good agreement with the TALYS and other experiments results from EXFOR database. More detailed description of our results is presented in P. Chudoba et al. contribution in these proceedings (POS-54 2015).

## 4. Conclusions

The general feature of the experimental spatial distribution of 88Y, 87Y, 86Y and 85Y isotopes production is that the maximum yield is at about 13 cm from the front of the natural uranium spallation target and that the yield is decreasing with increasing radial distance from the target axis.

Shape of neutron flux density per deuteron in the Quinta assembly produced by the neutrons generated in the assembly irradiated by the relativistic deuteron beam of 2 GeV, 4 GeV and 8 GeV energies in general is the same.

The main contribution to uncertainties in the experimental results are due to peak area calculation and statistical error coming from DEIMOS program. Moreover there are uncertainties in the measurements involving the total number of the primary deuterons in each experiment. We estimate that the overall uncertainties of the experimental data to be in the range 15%-20%.

Measurements of neutron reaction cross-sections for selected energies of these reactions by using of quasi mono-energetic neutron sources are very important and should be continues in the future. We plan next experiments in Uppsala laboratory in January 2015.

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