FAST NEUTRON SPECTRUM MEASUREMENT IN THE QUINTA ASSEMBLY OF E+T RAW COLLABORATION

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Abstract

This work is a preliminary step to study of radioactive waste utilization problems (transmutation) of E+T RAW collaboration. The Quinta experimental assembly has about 500 kg of natural uranium rods surrounded by a lead shielding. The Quinta assembly was irradiated with 0.66 GeV proton beam from the Phasotron and with 2, 4 and 8 GeV deuteron beams from the Nuclotron – both are located in the JINR laboratory, Dubna. A method based on yttrium activation detectors was used to obtain neutron flux spectrum. The nuclear (n, xn) threshold reaction rates of ^{89}Y samples located inside the assembly were used to obtain fast neutron energy spectra. The experiments were carried out in December 2012 and 2014. Comparison of the results of both experiments will be presented.

1. Introduction

The main advantage of electronuclear technology, as compared to conventional reactor technologies, is that a subcritical active core plus an external neutron source (accelerator and spallation target) are used. This advantage not only provides intrinsic safety of the system but also makes it possible to obtain high fluxes of high-energy neutrons independent of fission neutrons of the subcritical assembly material. The high-energy neutrons are an ideal tool to induce fission in most trans-uranium isotopes.

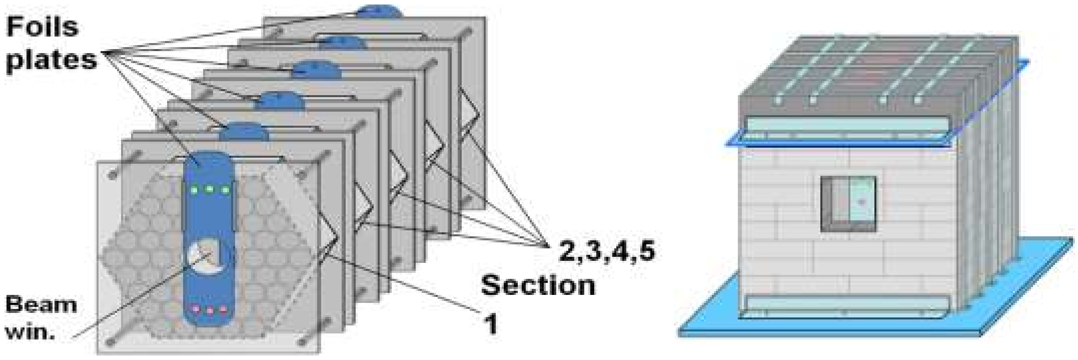

Fig. 1. Scheme of Quinta assembly. On the left there is a view on the uranium target with supporting structures and six metallic plates used for sample placement. On the right there is a view on the lead shielding enfolding the target. [2]

Most activities are concentrated on the classical electronuclear systems – Accelerator Driven Systems (ADS) – based on spallation neutron generation, with a spectrum harder than that of fission neutrons.

Study of deep subcritical electronuclear systems and feasibility of their application for energy production and radioactive waste (RAW) transmutation using relativistic beams from the JINR Nuclotron, which we are involved in, is performed within the E&T RAW collaboration. The long-range goal is the study of the possibilities of such systems with maximally hard neutron spectrum, to carry out transmutation of RAW, and also to gain energy due to burning of AC material.

We have started with experiments which refer to the study of neutron spectra at various points in the volume of the assembly Quinta (Fig. 1.).

2. Experiments

A new "ready to use" assembly Quinta, located at the Joint Institute for Nuclear Research (JINR), Dubna, Russia, has been available for the E&T RAW collaboration since the end of the year 2009. It is an assembly massive uranium target and lead shielding (Fig. 1). The Quinta assembly consists of a total of 512 kg of natural uranium. It is composed of five sections, each being 114 mm long and separated by a 17 mm air gap which allows the placement of samples mounted onto special plates (Figs. 1 and 2). We have 6 plates (measurements positions) because we have 4 gaps between assembly sections and two positions in front of and rear assembly. The sample plates are labeled 0 – 5, starting from the direction of the incident beam (Figs. 2 and 3). The uranium exists as many cylindrical rods, where each rod is 36 mm in diameter, 104 mm in length and 1.72 kg in mass. Excluding the first section, 61 rods are arranged in a hexagonal lattice. The first section contains only 54 rods and the removal of the central 7 rods is to create a beam window. The five sections are mounted onto a single slab of aluminum with thickness 25 mm and surrounded by lead bricks 100 mm thick on all six sides of total weight 1780 kg. This serves as a neutron reflector and to some extent as a biological shielding for γ-rays. The front of the lead bunker has a square window 150 × 150 mm.

The Quinta target was irradiated with 2, 4 and 8 GeV pulsed deuteron beams from the Nuclotron and with 0.66 GeV proton beam from the Phasotron – both are located in the JINR laboratory, Dubna. Total number of deuterons of the three deuteron irradiations are equal to $3.02(30) \cdot 10^{13}$, $2.73(27) \cdot 10^{13}$, $0.91(9) \cdot 10^{13}$ and over 10^{15} protons from Phasotron. During the time of irradiation equal to 6.27 h, 9.35 h and 16.7 h, and 5.5 h respectively.

All ^{89}Y activation foils (purity > 99.99%) were placed in the Quinta target on the detector plates in front of, between the five sections, and on the rear of the target in two positions at varying radial distances (4 and 8 cm) (Figs. 2 and 3) from the beam axis for each irradiations. Foils had form of pills made of compressed yttrium powder with dimensions diameter 9 × 1.5 mm^3, with weight ~ 0.6 – 0.8 g.

After irradiation, the samples were removed and transported away to be analyzed with gamma spectrometry, using HPGe n-type coaxial detector. Measurements began ~0.5 – 1 h after irradiation had stopped and continuing few days. All spectra were analyzed using the DEIMOS [5] program. Finally we calibrate all the results to B parameters by the below calibration formula (1). Spatial distributions of ^{88}Y, ^{87}Y, ^{86}Y and ^{85}Y isotope production for the deuteron beam of 2.0, 4.0 and 8.0 (December 2012) GeV and proton beam 660 MeV (December 2014) in the Quinta assembly were measured. The error of spatial distributions of ^{88}Y, ^{87}Y, ^{86}Y, ^{85}Y isotopes production are from 7E-08 to 1E-06 (error range 10 – 20%) it deepens of the point.

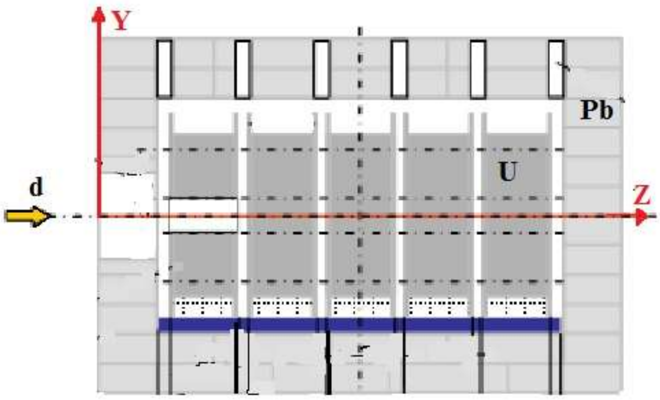

Fig. 2. Longitudinal view of the Quinta assembly. Measurements plates (Fig.3) are located in the air gaps between the Uranium “U” sections [2].

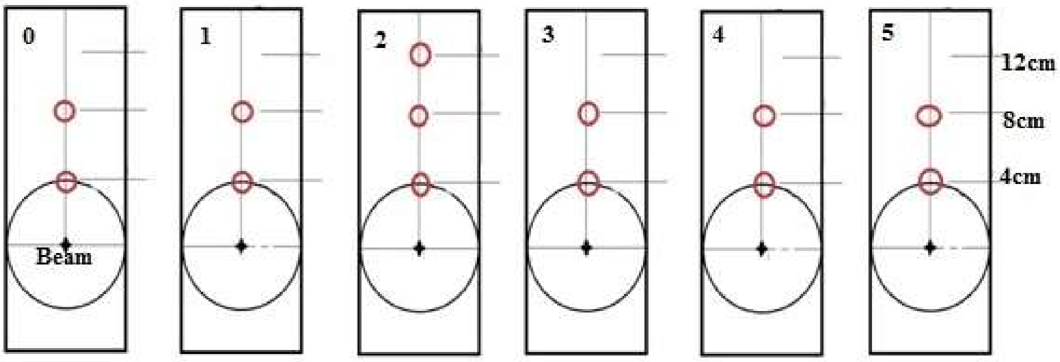

Fig. 3. Location of ^{89}Y foils during the proton irradiation (red circles). Measurement plates distance from the front of assembly 0=0 cm, 1=13.1 cm, 2=26.2 cm, 3=39.3 cm, 4=52.4 cm and 5=65.5 cm. [2]

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

where B is a number of nuclei per 1 gram of a sample material and per 1 primary proton;

N_1 – a peak area (line) – number of counts;

N_{abs} – the absolute intensity of given line in percent [%];

$\varepsilon_p(E)$ – a detector efficiency function of energy (polynomial);

$COI(E,G)$ – a cascade effect coefficient function of energy and geometry;

$\Delta D(G)$ – calibrations function for self-absorption inside of samples (foils)

$\Delta S(E)$ – calibrations function for shape of samples(foils) - (non point);

I – a total number of primary protons;

$t_{1/2}$ – a half life time [s];

t_{ira} – an elapsed time of irradiation [s];

t_+ – an elapsed time from the end of irradiation to the beginning of measurement [s];

t_{real} – an elapsed time of the measurement [s];
t_{live} – a “live” time of measurement [s];
m – a mass of the sample (foils) in grams [g].

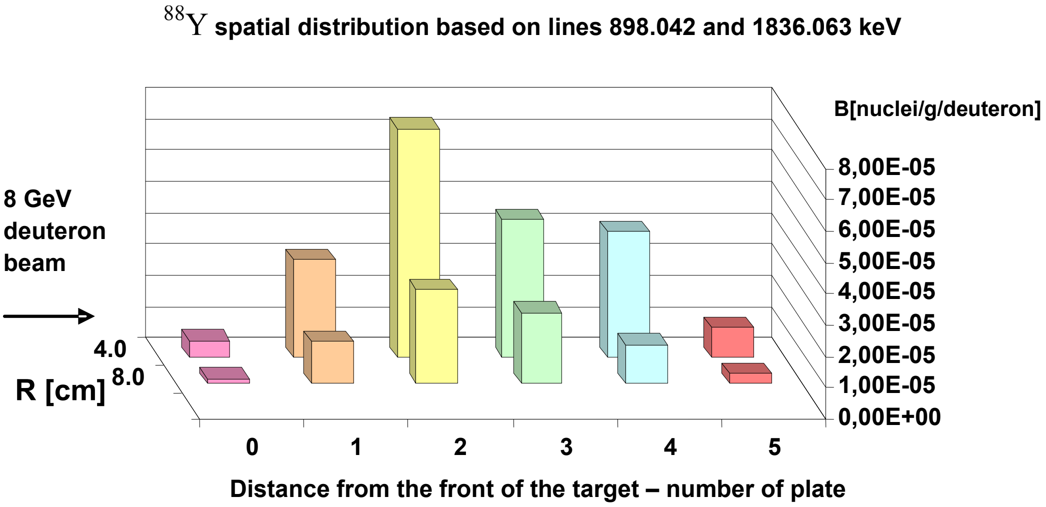

Fig. 4. Spatial distribution (radial & axial) of ^{88}Y production (*B* parameter) for the deuteron beam 8 GeV. On X axis we have number of measurements plates (distance in [cm] 1=13.1, 2=26.2, 3=39.3, 4=52.4 and 5=65.5). Y axis – radial distance in [cm].

All our results are presented in 3D graphs form. An example of the isotope production ^{88}Y presented in Fig. 4 show that the Spatial distribution (radial & axial) of Y88 production. The maximum yield of production is at about 26 cm from the front of the Quinta assembly (about 13 cm from ^{238}U spallation target) and that the yield is decreasing with increasing radial distance from the target axis.

3. Evaluation of average high energy neutron flux in the ^{89}Y foils location inside the U/Pb assembly

Having determined isotope production per one gram of sample and per one beam deuteron at specified positions of the Quinta setup for the three isotopes ^{88}Y, ^{87}Y and ^{86}Y, we can evaluate three average high energy neutron fluxes in each ^{89}Y foils location for certain energy ranges. The energy ranges are 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 32.7 MeV 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 32.7 MeV and ends at 100 MeV. Microscopic cross section for energy 100 MeV for reaction ^{89}Y(n, 4n)^{86}Y is very low.

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 are in the EXFOR data base [6] (only for reaction ^{89}Y(n, 2n) and several points for ^{89}Y(n, 3n)^{87}Y). That is why our cross sections were calculated using the TALYS code [7].

When we consider three isotopes then we have to solve three equations [8]. Solution of the three equations let us to evaluate the average neutron fluxes in the three energy ranges expressed in [n/cm^2·s] (2, 3, 4):

$$\overline{\phi_1} = \frac{C}{\overline{\sigma_{11}}}[B^{88} - B^{87}\frac{\overline{\sigma_{12}}}{\overline{\sigma_{22}}} + B^{86}(\frac{\overline{\sigma_{23}}\,\overline{\sigma_{12}}}{\overline{\sigma_{33}}\,\overline{\sigma_{22}}} - \frac{\overline{\sigma_{13}}}{\overline{\sigma_{33}}})], \tag{2}$$

$$\overline{\phi_2} = \frac{C}{\overline{\sigma_{22}}}[B^{87} - B^{86}\frac{\overline{\sigma_{23}}}{\overline{\sigma_{33}}}], \tag{3}$$

$$\overline{\phi_3} = \frac{C}{\overline{\sigma_{33}}}B^{86}, \tag{4}$$

where B^{88}, B^{87}, B^{86} are measured isotopes (B parameters) of ^{88}Y, ^{87}Y and ^{86}Y respectively per one gram of detector and per one beam deuteron (protons);

$$C = \frac{S \cdot {}^{89}G}{A \cdot t};$$

S is a total number of deuterons (protons);
^{89}G – the gram-atom of ^{89}Y [g];
A – the Avogadro’s number;
t – a deuteron (proton) irradiation time [s];
$\overline{\sigma_{11}} \div \overline{\sigma_{33}}$ – average microscopic cross sections of the measured isotopes for the reaction (n, xn) in the three chosen energy ranges;
$\overline{\phi_1}, \overline{\phi_2}, \overline{\phi_3}$ are unknown average neutron fluxes in the three chosen energy ranges.

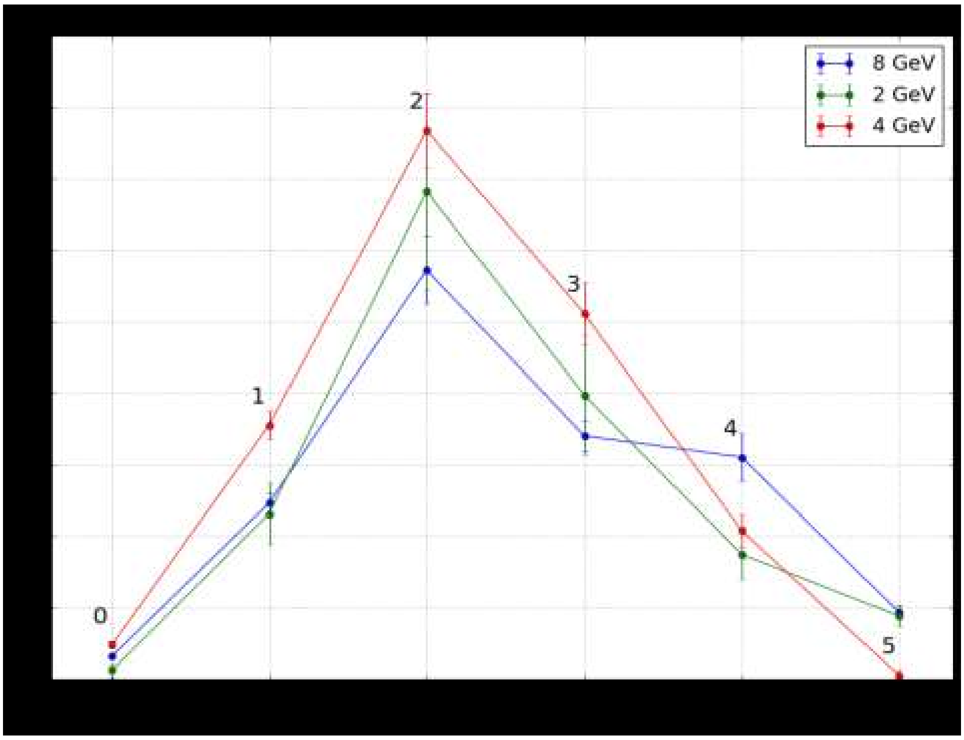

Fig. 5. Average neutron flux density per deuteron and its energy in function of Quinta target axis at R= 4 cm for three deuteron energies (2, 4, 8 GeV) in the neutron energy range (20.8 – 32.7 MeV). Errors range < 20%.

Number of measured isotopes in the detector assigns the number of algebraic equations what in turn assigns the number of unknown neutron fluxes in the chosen neutron energy ranges which are possible to be evaluated. The more of measured isotopes in the detector the more precise evaluation of the high energy neutron spectrum is obtained. Using the microscopic cross sections for the reactions ^{89}Y(n, 2n), (n, 3n) and (n,4n) generated by TALYS code and the experimental data (parameter B) we have evaluated the average high energy neutron flux in the ^{89}Y foils located inside the Quinta assembly for the three energy ranges (11.5 – 20.8 MeV), (20.8 – 32.7MeV) and (32.7 – 100 MeV).

An example of comparison of average neutron flux density per deuteron and per unit energy of deuteron for the three deuteron beams of energies equal to 2, 4 and 8 GeV in function of Quinta target axis at R=4 cm and the energy range 20.8 – 32.7 MeV is presented in Fig. 5. In Fig. 6 we have comparisons for all energies (including 660MeV proton beam - gray curve) in function of Quinta target axis at R= 4 cm and R= 8 cm. The curves of the neutron flux density per deuteron or proton and its energy (0.66, 2, 4 and 8 GeV) overlap in the energy ranges 20.8 – 32.7 MeV and nearly overlap in the energy range 11.5 – 20.8 MeV and 32.7 – 100 MeV for radius 4 cm and 8 cm. It is expected that for the deuteron and proton beam energies from about 1 GeV, the average neutron flux densities per deuteron and per one GeV beam energy should be equal.

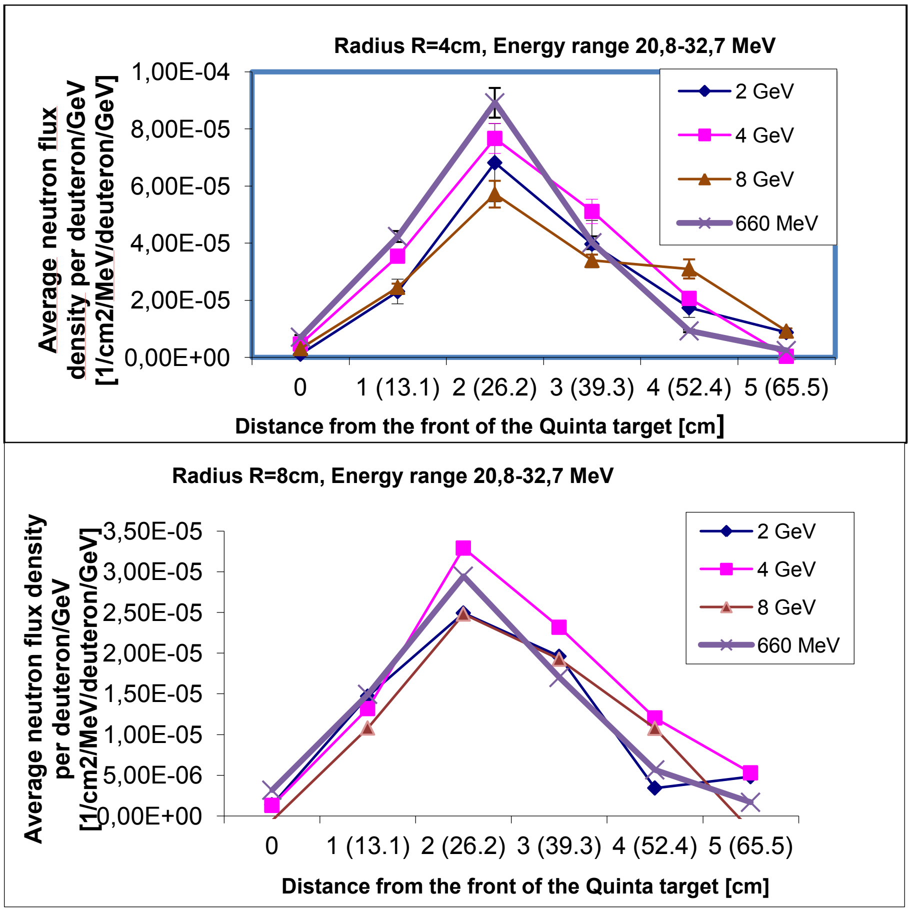

Fig. 6. Average neutron flux density per one deuteron or proton and its energy in function of Quinta target axis at R=4 cm (upper picture) and R=8 cm (down picture) for three deuteron energies (2, 4, 8 GeV) and for proton energy 660 MeV, in the neutron energy range (20.8 – 32.7 MeV). Errors range < 20%.

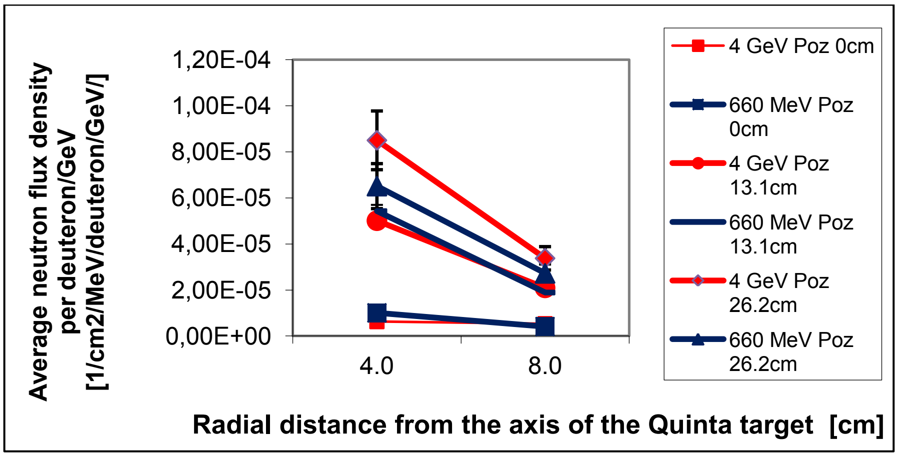

Fig. 7. Average neutron flux density per one deuteron or proton and its energy in function of radial distance from Quinta axis (for three different lengths from the front of the assembly). For deuteron energy beam 4 GeV and for proton energy beam 660 MeV, in the neutron energy range (20.8 – 32.7 MeV). Errors range < 20%.

In Fig. 7 we can see average neutron flux density per one particle in function of radial distance from Quinta axis. Red curve refers to the experiment in Dec 2012 for the deuteron energy 4 GeV and blue curve refers to the experiment in Dec 2014 (Cyclotron) for the proton energy 0.66 GeV The general feature of the experimental average neutron flux density per 1 MeV it is decreasing with increasing radial distance from the target axis. Curve from one longitudinal position are overlap each other.

4. Conclusions

The general feature of the experimental spatial distribution of ^{88}Y, ^{87}Y, ^{86}Y and ^{85}Y isotopes production is that the maximum yield is at about 13 cm from the front of the ^{238}U 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 proton beam 0.66 GeV and deuteron beams 2 GeV, 4 GeV, 8 GeV energies in general is the same.

The main contributions to uncertainties in the experimental results are due to peak area calculation and statistical error coming from DEIMOS program [5]. 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%.

Transmutation yield normalized to 1 GeV beam energy is generally the same for each beam energy from about 0.5 – 1 GeV. Energy range 1 – 1.5 GeV seems to be the optimal energy for ADS installations. No specific difference was observed in transmutation rate comparing proton and deuteron beam.

The presented here research results are of great importance for future use of Accelerator Driven Systems (ADS) for long lived nuclear waste utilization. ADS parameters like what target material to be used, what impinging particle, what particle energy, what geometry should be selected to get the highest transmutation efficiency.

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