E1-2017-91

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DETERMINING THE FAST NEUTRON FLUX DENSITY AND TRANSMUTATION LEVEL MEASUREMENTS IN THE **ADS** BY THE USE OF A THRESHOLD NUCLEAR REACTION

Submitted to "Particles and Nuclei, Letters"

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Определение плотности потока быстрых нейтронов и уровня трансмутации в измерениях в ADS системе с использованием пороговых ядерных реакций

Целью проекта было определение плотности потока быстрых нейтронов на основе данных, полученных в эксперименте «Quinta» (коллаборация E + T RAW), проведенном 4 декабря 2015 г. в ЛФВЭ ОИЯИ. Экспериментальная сборка на основе природного урана в алюминиевом корпусе облучалась протонным пучком с энергией 660 МэВ фазотрона ЛЯП ОИЯИ. Для исследования энергетического и пространственного распределения нейтронов использовались ядерные пороговые реакции типа $(n,\ xn)$. В этой статье описывается сборка Quinta, экспериментальные результаты, процедуры калибровки и расчеты средних нейтронных потоков на основе иттриевой мишени (Y-89). В будущем результаты и выводы из эксперимента, подобного этому, могут быть полезными при разработке подкритических систем (ADS) или других быстрых реакторов 4-го поколения. Работа проведена на летней практике студентов в ОИЯИ в 2016 г.

Работа выполнена в Лаборатории физики высоких энергий им. В. И. Векслера и А. М. Балдина ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 2017

Hanusek T. et al. E1-2017-91

Determining the Fast Neutron Flux Density and Transmutation Level Measurements in the ADS by the Use of a Threshold Nuclear Reaction

The aim of the project was determining the fast neutron flux density by using data from the Quinta experiment (E + T RAW collaboration), which took place on 4 December 2015 at the Veksler and Baldin Laboratory of High Energy Physics (VBLHEP), the Joint Institute for Nuclear Research (JINR). The experimental assembly based on natural uranium and an aluminum cover was irradiated by a 660 MeV proton beam from the Phasotron, DLNP, JINR. To gain the knowledge about the neutron flux inside the experimental assembly, nuclear threshold reactions of $(n,\ xn)$ type were used. This paper describes the Quinta assembly, experimental results, calibration procedure and average high energy neutron calculation based on yttrium (Y-89) isotopes production. In the future, results and conclusions from an experiment like this could be useful to design accelerator-driven subcritical systems (ADS) or other 4th generation fast reactors. The paper is based on the 2016 Student Summer Practice work at JINR.

The investigation has been performed at the Veksler and Baldin Laboratory of High Energy Physics, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna, 2017

INTRODUCTION

The main goal of the E + T RAW collaboration is determining the fast neutron flux density inside the Quinta experimental assembly. The Quinta based on natural uranium in aluminium cover was irradiated by a 660 MeV proton beam from a cyclotron. To gain the knowledge about the neutron flux inside the experimental assembly, nuclear threshold reactions of $(n,\ xn)$ type were used. This paper describes the Quinta experimental assembly, calibration and normalization procedure, and average high energy neutron calculation based on yttrium (^{89}Y) isotopes production. The paper is based on the 2016 Student Summer Practice work at JINR. Experimental data came from the experiment which took place on 4 December 2015 at the Veksler and Baldin Laboratory of High Energy Physics (VBLHEP), the Joint Institute for Nuclear Research (JINR) at Dubna on the cyclotron Phasotron.

1. EXPERIMENT DESCRIPTION

The Quinta experimental assembly consists of about 512 kg of natural uranium [1]. It is divided into five sections, 114 mm long and separated by a 17 mm air gap. Each section contains uranium cylindrical rods in the aluminium cover, 36 mm in diameter, 104 mm in length and 1.72 kg in mass. Except the first one, each section holds 61 rods. The first section holds only 51 rods because of the window in the centre of the section for a proton beam. The hole is 80 mm in diameter and it works as reduction of the escaping neutrons (including backward scattering and emission). The front and the back of each section are covered by aluminium plates with the dimensions of $350 \times 350 \times 5$ mm. The 17 mm air gap between the Quinta sections allows the placement of samples mounted onto special aluminium plates. There were six plates — four gaps between assembly sections and two positioned in front of and near the assembly (Figs. 1 and 2). The Quinta assembly is surrounded by a lead cover which was made from lead bricks. The cover is 100 mm thick on all six sides and its total weight is 1780 kg. The aims of the cover are radiation shielding for γ rays and neutron reflecting. In the front wall there is a square window 150×150 mm in order to facilitate for the beam getting inside the assembly (Fig. 3).

In the experiment, the uranium target was irradiated using the proton beam from the Dubna Phasotron. The beam energy was 660 MeV and finally, after 5 h of irradiation, about 10^{15} primary particles were collected. The samples were made from natural yttrium with a purity of 99.9 %, whose isotope Y-89 has 100 % abundance. Preparing the samples to the experiment consisted in weighing and putting them in appropriate positions on earlier prepared aluminium plates. There were 12 samples in different positions in relation to the beam axis and forehead

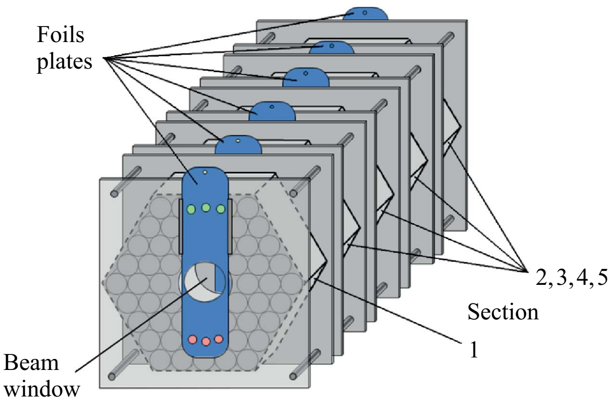

Fig. 1. Scheme of the uranium core of the Quinta set [1,2]

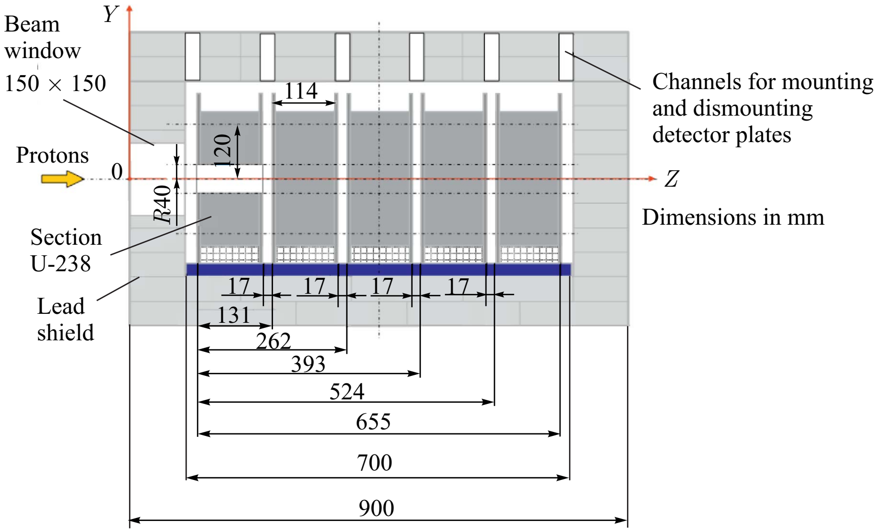

Fig. 2. View of the experimental system along with the lead shield along the beam axis. The dimensions of the set are shown [1,2]

of the set (Fig. 4). On the third plate from the forehead (plate number 2) of the set there were additional sensors to measure fluctuations of the temperature during the experiment. An additional sample was put outside the lead cover to check the level of radiation only, and this sample was not included in the calculations of the average neutron flux density.

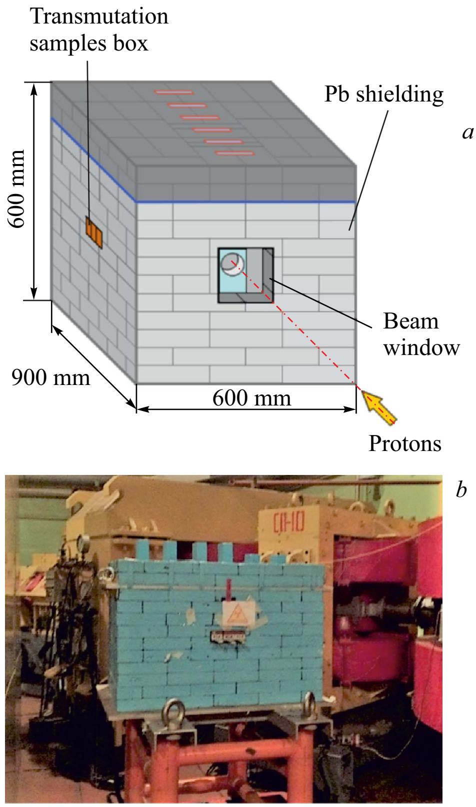

Fig. 3. Quinta experimental assembly in the lead cover: *a*) scheme of the Quinta assembly surrounded by the lead bunker [2]; *b*) the Quinta assembly surrounded by the lead bunker right before the experiment in 2015 (photography of the authors)

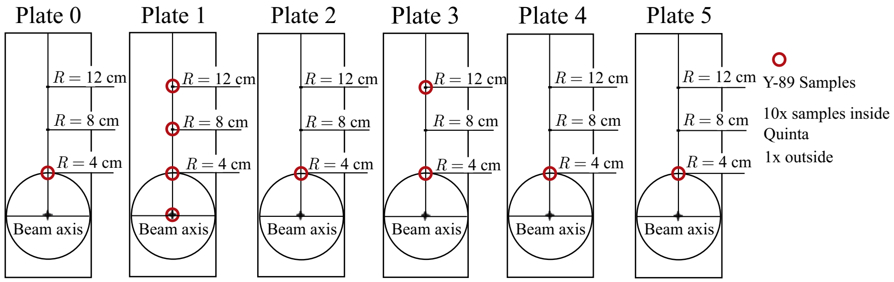

Fig. 4. The placement of the Y-89 samples on the aluminum plates on 4 December 2015. The beam was going from left to right

Samples from the experiment used to calculate the average neutron flux density

Sample number	Mass of sample, g	Plate at exp. set	Radius posi-tion, cm	Measu-rement position number at HPGe detector	Date and time of start of measurement	t_+, s	t_{live}, s	t_{real}, s
73	0.573	0	4	p3	2015.12.04 23:05:45	6645	557.1	655.6
58	0.655	1	4	p8	2015.12.04 22:23:56	4136	556.8	575.2
56	0.722	2	4	p5	2015.12.07 06:45:01	207001	3582.4	3605.0
74	0.933	3	4	p8	2015.12.04 23:22:53	7673	893.5	914.0
75	0.79	4	4	p5	2015.12.04 23:40:51	8751	747.1	779.0
77	0.773	5	4	p3	2015.12.05 00:11:07	10567	1135.0	1184.7
57	0.712	1	0	p8	2015.12.04 22:09:00	3240	534.7	704.0
58	0.655	1	4	p8	2015.12.04 22:23:56	4136	556.8	575.2
59	0.692	1	8	p3	2015.12.04 22:37:26	4946	573.2	604.0
60	0.567	1	12	p3	2015.12.04 22:49:51	5691	770.0	785.4
76	0.851	3	12	p3	2015.12.04 23:55:27	9627	777.6	800.4
43	0.746	5	8	p3	2015.12.05 00:32:49	11869	1650.6	1679.5

Note. t_+ — time between the end of the experiment and the start of the measurement; t_{real} — time of the sample being in the gamma detector; t_{live} — time of the measurement with the dead time correction ($t_{\text{live}} = t_{\text{real}} - t_{\text{dead}}$).

After the proton irradiation for 21900 s, the samples were taken out to the measurement set. The measurement set consisted of an HPGe detector with a liquid nitrogen bottle and a lead cover. Inside the cover, there were eight possible measurement positions at different length from the front of the HPGe detector. The measurements for all samples lasted for many hours, from a few minutes to above 11 h each. There were two sets of each sample measurements: the short one (minutes) just after finishing of the irradiation and the long one (hours) after the first set. The period between the end of the irradiation and start of the first measurement was 54 min (table).

2. CALIBRATION AND ANALYSIS

The samples had to be subjected to a qualitative and quantitative analysis. The energy and efficiency calibration of the HPGe detector were needed. For the energy calibration characteristic, gamma radiation lines from yttrium isotopes were used.

The energy range of the used HPGe detector was from 40 keV (channel number 104) to 3143 keV (channel number 8191). All used lines in later calculation were from 380.79 to 1920.72 keV. Considering this, the authors decided to choose two lines from the beginning and end of the range — lines 190.79 keV (from Y-86) and 1836.063 keV (from Y-88) to prepare the HPGe detector energy calibration. The efficiency of the HPGe detector can be changed in a long time period because of the temperature or level of the electronic noise, and results on the same detector can be completely different within a few days or even less. For the efficiency calibration of the HPGe detector, a set of radionuclide calibration standards was used. Each used source is supplied with a certificate of calibration, which contains the total radioactivity level in a specific moment in the past. The standards set was measured in the same conditions and positions as the yttrium samples before. To create the HPGe detector efficiency curve, Co-60, Ba-133, Cs-137, Eu-152, Bi-207, Na-22, Cs-134 and Th-228 standard samples were used. After that an approximating function was created, which is different for each measurement position (inside the HPGe detector). For position number 5, the approximating function is given by the formula

$$\text{Efficiency} = \exp\left(-0.1282\left(\ln\left(\frac{Eg}{1000}\right)\right)^5 - 0.4278\left(\ln\left(\frac{Eg}{1000}\right)\right)^4 - 0.1863\left(\ln\left(\frac{Eg}{1000}\right)\right)^3 + 0.3512\left(\ln\left(\frac{Eg}{1000}\right)\right)^2 - 0.8407\left(\ln\left(\frac{Eg}{1000}\right)\right) - 6.5952\right). \quad (1)$$

For the gamma lines identification, analysis and energy calibration, the DEIMOS program [3] was used, which allowed detailed analysis. In DEIMOS, the user can receive specified parameters of every single line, such as number of counts, full width at half maximum (FWHM), energy of line, statistic error and more. The program uses iteration process to fit the best Gaussian function and consider the radiation level of the background. To make comparison of many different experiments, there is a special formula for changing the absolute results to the relative value that can be compared. The results have to be converted to one normalized B parameter, which is a number of produced specific isotopes per 1 g of the sample and per 1 proton from the accelerator. Parameter B is given by the formula [4, 5]

$$B = N_1 \frac{1}{m \cdot I} \frac{\Delta S(G) \cdot \Delta D(E)}{\frac{N_{\text{abs}}}{100} \cdot \varepsilon_p(E) \cdot \text{COI}(E, G)} \frac{\lambda \cdot t_{\text{ira}}}{1 - \exp(-\lambda \cdot t_{\text{ira}})} \times \\ \times \exp(\lambda \cdot t_+) \frac{t_{\text{real}}/t_{\text{live}}}{1 - \exp(-\lambda \cdot t_{\text{real}})}, \quad (2)$$

where

- B — number of nuclei per 1 g of the sample and per 1 nucleus from the accelerator;
- N_1 — area of the peak (number of counts);
- N_{abs} — absolute intensity of the line in percentage;
- $E_p(E)$ — detector efficiency in the energy function;
- $\text{COI}(E, G)$ — cascade effects in the energy and geometry function;
- I — absolute number of nuclei in the beam from the accelerator;
- m — mass of the sample;
- $\Delta S(G)$ — sample area correction in the geometry function;
- $\Delta D(E)$ — self-absorption correction in the energy function;
- Λ — decay constant;
- $t_{1/2}$ — half-time period;
- t_{ira} — irradiation time;
- t_+ — time between the end of the experiment and the start of the measurement;
- t_{real} — time of the sample being in the gamma detector;
- t_{live} — time of the measurement with the dead time correction.

Self-absorption sample correction $\Delta D(E)$ and size of sample correction (no point sample) $\Delta S(G)$ were very small (the used samples were quite thin and of the small-circle shape). These two corrections were much smaller than the rest of them [6]. The three last parts of B parameter in formula (2) are connected with time. The first takes into account the time of the irradiation and the fact that after the end of the irradiation some of the isotopes created at the beginning decayed.

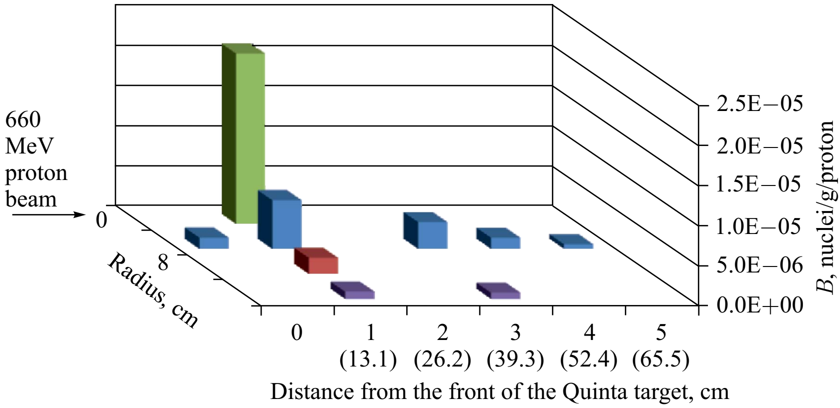

Fig. 5. Production of yttrium-88 in the Quinta experimental assembly

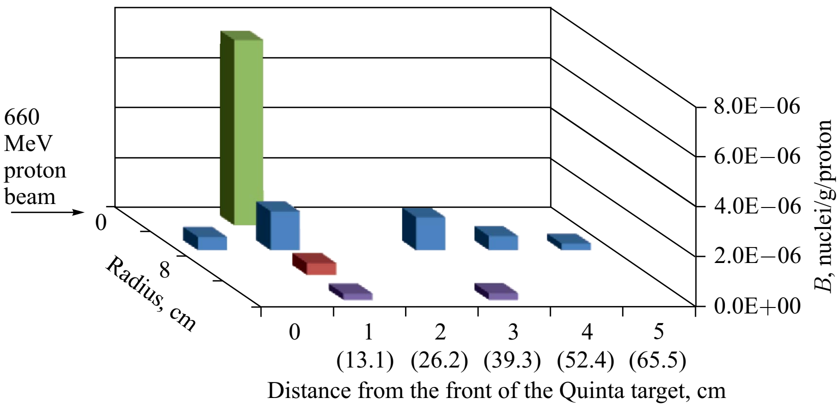

Fig. 6. Production of yttrium-87 in the Quinta experimental assembly

The second part (the fourth in the equation) is the correction because of the period between the end of the irradiation and the start of the measurements of each sample. The fifth part is the correction because of the time of the measurements and the relation between the "real" time and the "live" time. The real time is the physical time of the measurement. Every detector has time when it is insensitive to any gamma rays because of clearing buffer — it is called dead time [6]. The dead time and the live time in sum give the real time. The biggest contribution to the total error is the error from the DEIMOS program (counts of the peak) and the error of the total quantity of protons from the accelerator. To minimize the error from the peak area only the stronger lines were used, because errors can be about 1–20%. The error from the quantity of the protons was assumed to

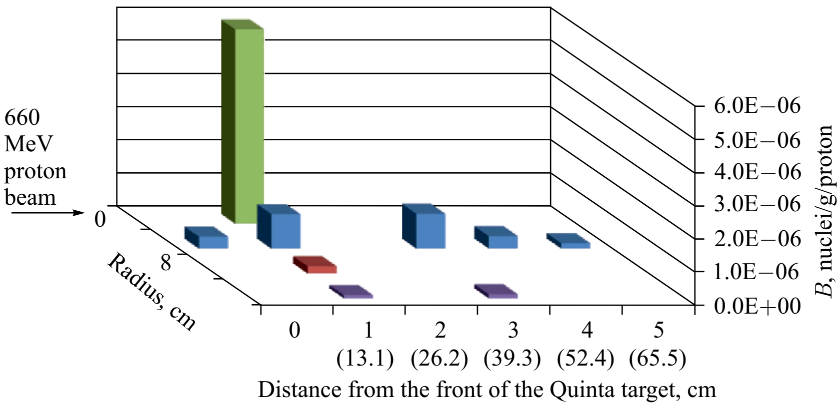

Fig. 7. Production of yttrium-86 in the Quinta experimental assembly

be about 15%. The rest of the error is mostly about 1–3%. After all necessary correction and normalization, the spatial distributions of parameter B (production of yttrium: 88, 87 and 86) in the experimental assembly were obtained, which are presented in Figs. 5–7. One of the irradiated samples in radial position 4 cm and axial position 2 was not considered in the results. This sample was measured by other scientists 57 h after the end of the irradiation which can be the reason of much bigger errors of that.

3. AVERAGE FAST NEUTRON FLUX DENSITY

To obtain the average flux neutron density, inside the experimental Quinta assembly, first B^y (B parameter) has to be calculated. The average flux density ($\overline{\Phi}$) is given by the formula [5]

$$\overline{\Phi} = \frac{B^y \cdot S \cdot G}{\overline{\sigma} \cdot A \cdot t}, \tag{3}$$

where

- B^y — production of yttrium isotope;
- S — total number of protons from accelerator;
- G — atomic mass;
- $\overline{\sigma}$ — average cross section for the reaction $(n,\, xn)$ in a particular energy range;
- A — Avogadro constant;
- t — time of irradiation.

To simplify the equations, it was decided to divide the spectrum of the neutron energy into three ranges:

- 11.5–20.8 MeV;
- 20.8–32.7 MeV;
- 32.7–100 MeV.

The levels of 11.5, 20.8, and 32.7 MeV are the threshold energies for the reactions $(n, 2n)$, $(n, 3n)$, and $(n, 4n)$, respectively. 100 MeV was taken to make sure almost 100% reactions were included and the value was chosen arbitrarily. Set of equations to solve is

$$\begin{cases} B^{88} \cdot C = \Phi_1 \cdot \sigma_{11} + \Phi_2 \cdot \sigma_{12} + \Phi_3 \cdot \sigma_{13} \\ B^{87} \cdot C = 0 + \Phi_2 \cdot \sigma_{22} + \Phi_3 \cdot \sigma_{23} \\ B^{86} \cdot C = 0 + 0 + \Phi_3 \cdot \sigma_{33} \end{cases} \quad (4)$$

where

- B^y — production of yttrium isotope;
- C — constant, $C = \dfrac{S \cdot G^{89}}{A \cdot t}$;
- Φ_n — average fast neutron flux density;
- σ_n — average cross section for the reaction in the given energy range.

Using this set of equations and specific B parameter value, the obtained fast neutron flux density is as shown in Figs. 8–10. For this calculation, made by TALYS CODE [7], the cross section value was used for the reason that the EXFOR experimental data base for yttrium (n, xn) reaction was very poor [8,9].

The spatial distribution of the neutron flux density is given in three energy ranges.

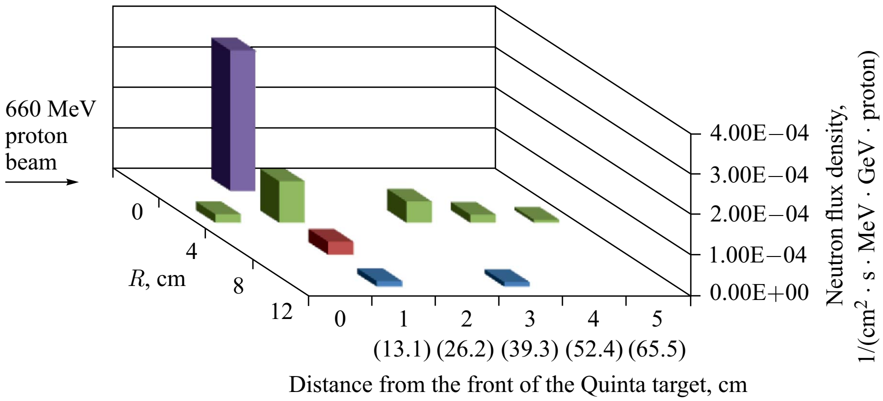

Fig. 8. Average neutron flux density inside the Quinta assembly for the energy range from 11.5 to 20.8 MeV

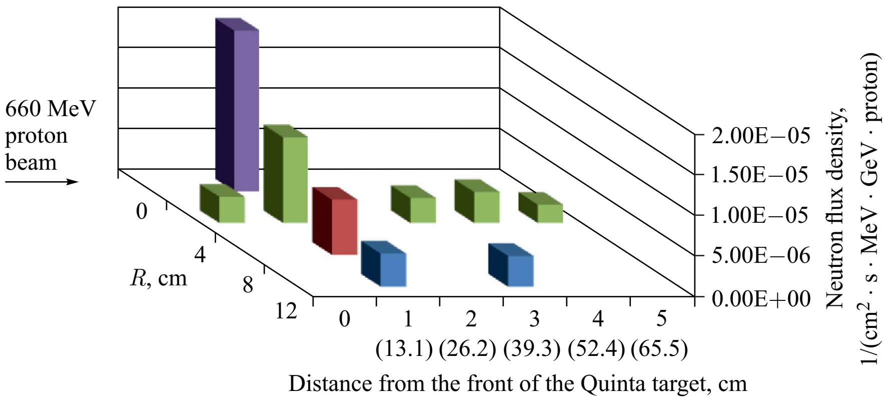

Fig. 9. Average neutron flux density inside the Quinta assembly for the energy range from 20.8 to 32.7 MeV

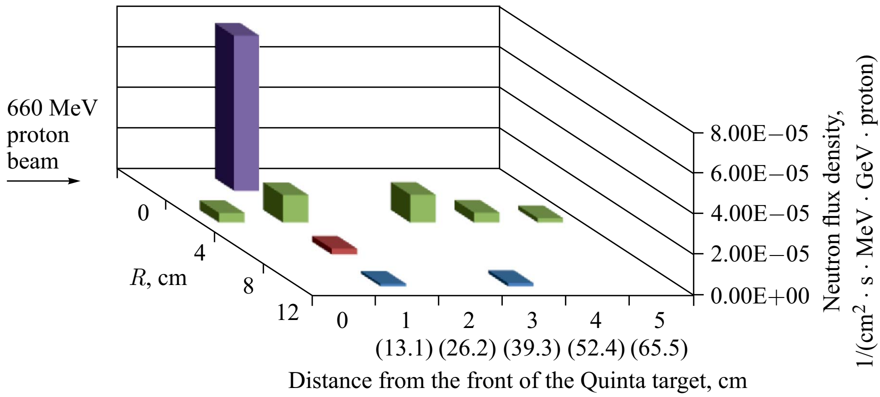

Fig. 10. Average neutron flux density inside the Quinta assembly for the energy range from 32.7 to 100 MeV

CONCLUSIONS

After the experiment, it is possible to perform measurements which give us the isotope level production inside the experimental assembly. Based on the measurements, with a knowledge of nuclear reactions cross section values and parameter equations, we were able to obtain the average neutron flux density inside the Quinta assembly. The qualitatively obtained results are compatible with the expectations from previous experiments [2, 4–6]. Unexpectedly, the results for the range from 20.8 to 32.7 MeV (see Fig. 9) showed some difference as compared to the rest of the ranges. The highest peak in the middle range should be bigger. Moveover, one of the samples, in radial position 4 cm and axial position 2, was not considered in our results. This sample was measured by other scientists 57 h after the end of the irradiation which can be the cause of much bigger, unpredictable errors for this sample. Considering this, it was decided to delete this point from further analysis and figures. The presented method with yttrium samples is effective and quite simple to obtain results. We see that future experimental measurements of the yttrium (n, xn) cross section are necessary to make this method more precise. In the future, we should continue experimental measurement of cross section (n, xn) value [9] and experiment with different type of target (up to now it was lead and uranium target [10]). Additionally, we are planning checking our activation detector material in another type of measurement using positron annihilation spectroscopy [11]. The parameters of the Quinta assembly were very similar to the conditions provided in future ADS reactors. That is why the results should be applied to designing and building future ADS reactors that can transmute long-lived radioisotope from conventional nuclear reactors.

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Received on December 12, 2017.

Редактор *Е. И. Кравченко*

Подписано в печать 05.02.2018.
Формат 60 × 90/16. Бумага офсетная. Печать офсетная.
Усл. печ. л. 0,94. Уч.-изд. л. 1,29. Тираж 255 экз. Заказ № 59328.

Издательский отдел Объединенного института ядерных исследований
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