TRANSMUTATION MEASUREMENTS LEVEL IN ACCELERATOR-DRIVEN SUBCRITICAL SYSTEMS (ADS)*

M. Bielewicz[a,b], T. Hanusek[c], A. Jaskulak[d]
E. Strugalska-Gola[a], M. Szuta[a], S. Kilim[a], M.J. Peryt[e,b]
S. Tiutiunnikov[b]

[a]National Centre for Nuclear Research, A. Sołtana 7, Otwock-Świerk, Poland
[b]Joint Institute for Nuclear Research, Joliot-Curie 6, Dubna, Russia
[c]Poznan University of Technology, Pl. M. Skłodowskiej-Curie 5, Poznań, Poland
[d]University of Warsaw, Warszawa, Poland
[e]Warsaw University of Technology, Pl. Politechniki 1, Warszawa, Poland

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The aim of the project was the determining the Fast Neutron Flux Density and Transmutation Level measurements in the experimental assembly Quinta based on natural uranium, irradiated by 660 MeV proton beam from cyclotron "Phasotron" at the Laboratory of High Energy Physics, JINR Dubna. A threshold reactions $Y^{89}(n, xn)$ were used. This paper describes the Quinta assembly, experimental results from December 2015, and average high-energy neutron calculation based on Y^{89} isotopes production. The better knowledge about neutron flux density is necessary to constructing the fourth generation nuclear reactors such as fast reactors (FR) and accelerator-driven subcritical systems (ADS).

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1. Experimental assembly

The Quinta experimental assembly based on natural uranium cylindrical rods (diameter 36 mm, length 104 mm and mass 1.72 kg) in aluminum cover. It was irradiated by a 660 MeV proton beam from a Dubna cyclotron "Phasotron". To gain the knowledge about the neutron flux, nuclear threshold reactions of (n, xn) type were used. The Quinta consists of about 512 kg of natural uranium [1]. It is divided into five, separated by 17 mm air gap, sections (114 mm long). Except the first section, each have 61 rods. The first one holds only 51 rods because has the beam window in the center of the

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section. The window (diameter 80 mm) works as reduction of the escaping neutrons (including backward scattering and emission). The front and the back of each section are covered with thick (5 mm) aluminum plates. The air gap between the Quinta sections allows the placement of samples onto additional aluminum plates (Fig. 1). The Quinta assembly is surrounded by a lead cover (thick 100 mm) which was made from lead bricks and its total weight is 1 780 kg. In the front lead wall, there is a square beam window (150 × 150 mm) (Fig. 2). The beam energy was 660 MeV and, finally, after 5 h (21 900 s) of irradiation, about 10^{15} primary particles were collected. The activation samples were made from natural yttrium with a purity of 99.9%, whose isotope Y^{89} has 100% abundance. There were 12 samples (cylinder shape with diameter 10 mm and thickness about 1 mm each) in different positions in relation to the beam axis and forehead of the set (Fig. 3).

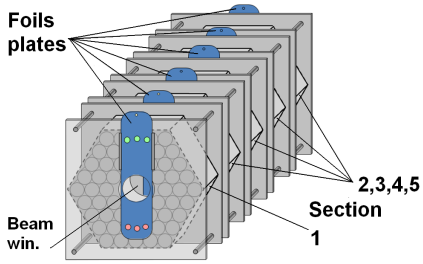

Fig. 1. Scheme of the uranium core of the Quinta set [1].

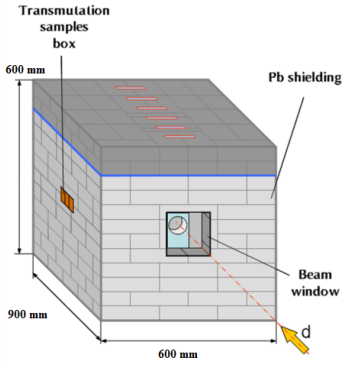

Fig. 2. Scheme of the Quinta assembly surrounded by the lead bunker [2].

After the irradiation, the samples were taken out to the HPGe detector measurement set. 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.

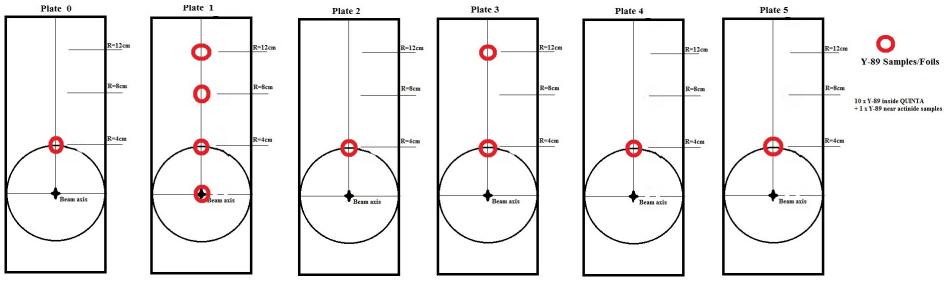

Fig. 3. The placement of the Y^{89} samples on the aluminum plates on December 4, 2015. The beam was going from left to right.

2. Measurements results

All samples had to be under a qualitative and quantitative analysis. For the energy calibration characteristic of the HPGe detector, gamma radiation lines 190.79 keV (from Y^{86}) and 1836.063 keV (from Y^{88}) were used. For the efficiency calibration of the HPGe detector, a set of radionuclide calibration standards was used (Co-60, Ba-133, Cs-137, Eu-152, Bi-207, Na-22, Cs-134 and Th-228). The standards gave us absolute radiation intensity on October 1, 2013 with accuracy of 5–7%. The standard set was measured in the same conditions and positions as the yttrium samples before. After that, an approximating function was created, which is different for each measurement position inside of HPGe detector. For the gamma lines identification, analysis and energy calibration, the DEIMOS program [2] was used. 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 possible results comparison from many different experiments, there is a special formula for changing the absolute results to the relative value. The results have to be converted to one normalized B reaction rate $Y^{89}(n, xn)$, per 1 gram of the sample and per 1 nuclei from the accelerator. Parameter B is given by the formula [3, 4]

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

where

— B — amount of nuclei per 1 gram of the sample and per 1 nuclei 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 nucleus 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 about 1 mm and of the small-circle shape diameter 10 mm). These two corrections were much smaller (less than 1%) than the rest of them [4]. The three last parts of B parameter in the formula 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. 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. It is called dead time [4]. The biggest contribution to the total error is the statistical error from the DEIMOS program (counts of the peak) and the error of the total quantity of protons from the accelerator (minimum error is about 15%). To minimize the error from the peak area, only the stronger lines were used. The rest of the error is mostly about 1–3%. After all necessary corrections and normalization, the spatial distributions of reaction rates $\text{Y}^{89}(n, xn)$ in the experimental assembly were obtained. Examples are presented in Figs. 4 and 5. One of the irradiated samples in radial position 4 cm and axial position 2 (26.2 cm on axial position) was not considered in the results. This sample was measured by other group 57 h after the end of the irradiation which can be the reason of much bigger errors.

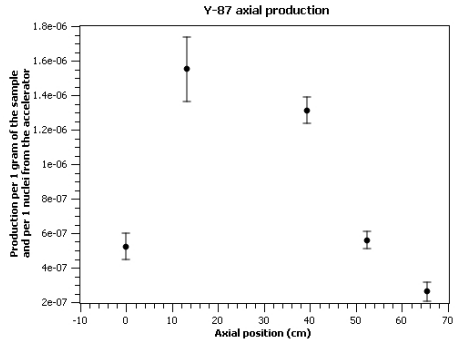

Fig. 4. Axial reaction rate of Y^{87} in the Quinta experimental assembly, at radial position 4 cm.

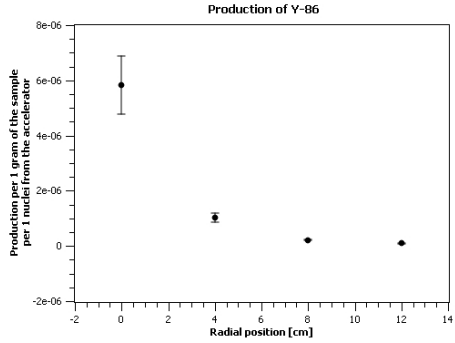

Fig. 5. Radial reaction rate of Y^{86} in the Quinta experimental assembly at axial position 13.1 cm.

3. Neutron flux density

To obtain the average flux neutron density, inside the experimental Quinta assembly, first B_y ((n, xn) reaction rate) has to be calculated. The average flux density($\bar{\Phi}$) is given by formula from [5]. To simplify the equations, the high-energy neutron spectrum was divided into 3 ranges: 11.5–20.8 MeV, 20.8–32.7 MeV and 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 of that. Finally, the spatial distribution of the neutron flux density is given in three energy ranges. Fast neutron flux density presented in Figs. 6, 7 was obtained using formula from [4] and specific B parameter value. The cross-section value for this calculation was made by TALYS code [5], for the reason that the EXFOR experimental data base for $Y(n, xn)$ reaction was very poor [6, 7].

The qualitatively obtained results are compatible with the expectations from previous experiments [1, 3, 4]. Unexpectedly, the results for the range from 20.8 to 32.7 MeV (Fig. 7) showed some difference as compared to the rest of the ranges. The highest peak in the middle range should be bigger. Moreover, one of the samples, in radial position 4 cm and axial position 2, was not considered in our results. This sample was measured by others with much bigger unpredictable error. Considering this, it was decided to delete this point from further analysis and figures. 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 [7] and an experiment with different type of target (up to now it was lead and uranium target [8]). Additionally, we plan to check the impact of the state of target on the measured here characteristics. For this reason, other techniques as *e.g.* positron annihilation spectroscopy [9] will be applied to describe the state of sample before our experiment.

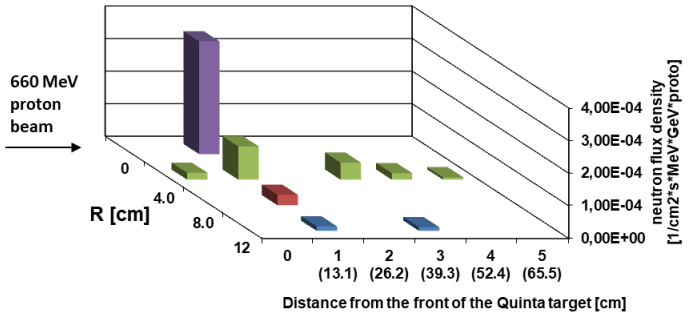

Fig. 6. Average neutron flux density in the Quinta assembly for the energy range of 11.5–20.8 MeV.

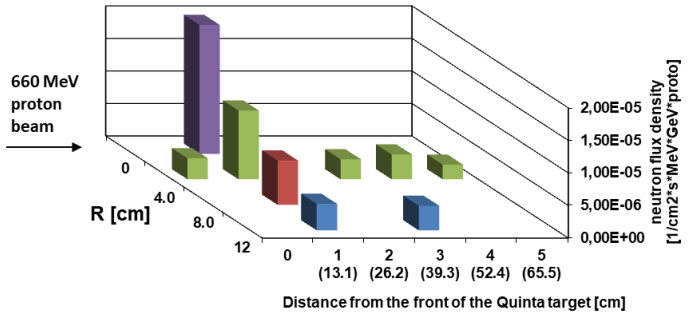

Fig. 7. Average neutron flux density in the Quinta assembly for the energy range of 20.8–32.7 MeV.

4. Conclusions

The presented method with yttrium samples is effective and quite simple to obtain results. After the experiment, it is possible to perform measurements which give us distribution of the reaction rates $Y^{89}(n, xn)$ 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 or ADS and FR. 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 radioisotopes.

REFERENCES

[1] W. Furman *et al.*, *PoS* **(Baldin ISHEPP XXI)**, 086 (2013).

[2] J. Frána, *J. Radioanal. Nucl. Chem.* **257**, 583 (2003).

[3] M. Bielewicz *et al.*, *J. Korean Phys. Soc.* **59**, 2014 (2011).

[4] M. Bielewicz, Spectrometry of Fast (>10 MeV) Neutrons in Lead–Uranium Experimental Setup, Dissertation Thesis, Warsaw: NCBJ, 2015; https://www.ncbj.gov.pl/en/dokumenty/doktorskie

[5] A.J. Koning, S. Hilaire, M.C. Duijvestijn, *AIP Conf. Proc.* **769**, 1154 (2005).

[6] Experimental Nuclear Reaction Data; EXFOR/CSISRS, https://www-nds.iaea.org/exfor/

[7] M. Bielewicz *et al.*, *EPJ Web. Conf.* **146**, 11032 (2017).

[8] M. Bielewicz *et al.*, *Nucl. Data Sheets* **119**, 296 (2014).

[9] P. Horodek *et al.*, *Radiat. Phys. Chem.* **122**, 60 (2016).