SPATIAL DISTRIBUTION OF ^{nat}U(n, f), ^{238}U(n, g) REACTION RATES IN SPALLATION NEUTRON FIELDS PRODUCED BY DEUTERONS AND ^{12}C IONS ON THE MASSIVE URANIUM TARGET

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Abstract

The results of the experiments carried out within the framework of "Energy and Transmutation of RAW" at JINR NUCLOTRON accelerator are presented. The target assembly QUINTA consisting of 512 kg natural uranium was irradiated by deuteron and carbon beams with energies 1, 2, 4 and 8 GeV (deuterons), 24 and 48 GeV (carbon). Spatial distribution and total number of capture reaction and fission reaction rates was obtained using the activation technique. The integral number of fissions reactions in the volume of uranium target remains approximately constant within our statistical errors for 1, 2, 4 and 8 GeV deuteron beams and for 24 and 48 GeV carbon beams (per one primary particle and per 1 GeV of beam energy). For the integral number of capture reactions with deuteron beams we have seen maximum at 2 GeV. Some of the obtained experimental data was analyzed using the MCNPX transport code. For spatial distribution of reaction rates in case of 4 and 8 GeV deuteron beams we have seen a discrepancy between the experimental and calculated values in backward direction.

INTRODUCTION

Starting from 2010 studies of spallation neutron fields, which are generated by relativistic particle beams in the 500 kg uranium target of sub-critical assembly QUINTA [1], were carried out within the framework of international collaboration "Energy and Transmutation RAW" at NUCLOTRON accelerator of the Joint Institute for Nuclear Research, Dubna, Russia. In those experiments the capture, fission and transmutation reaction in the neutron field composed of spallation and fission neutrons were investigated [2 -4].

One of the main collaboration objectives is to study of dependence of neutron generation in the uranium target on primary beam energy and accelerated particles type. In this paper we describe the spatial distribution of ^{238}U(n, γ) and ^{nat}U(n, f) reaction rates obtained with deuteron and ^{12}C ion beams at various energies. As the result of the experiments, the total numbers of above reactions in the volume of uranium target were estimated.

EXPERIMENTAL PROCEDURE

The deeply (k_{eff} < 0.3) sub-critical assembly QUINTA (Figure 1) consists of 5 hexagonal sections filled with natural metallic uranium cylindrical rods (diameter 3.6 cm, length 10.4 cm, weight 1.72 kg) and surrounded by 10 cm thick lead bricks. Each section is 114 mm long (140 mm inscribed circle radius) and separated by a 17 mm air gap, where samples mounted onto detector plates. The total mass of natural uranium in fife section is 512 kg. For more technical detail, see reference [1].

The uranium target was irradiated with 1, 2, 4 and 8 GeV deuteron and with 24 and 48 GeV carbon beams from the NUCLOTRON accelerator of the JINR.

The total number of deuterons and ^{12}C ions to hit the target was determined from the activation of Al and Cu foils in the $^{27}Al(a, x)^{24}Na$ and $^{nat}Cu(a, x)^{24}Na$ reaction.

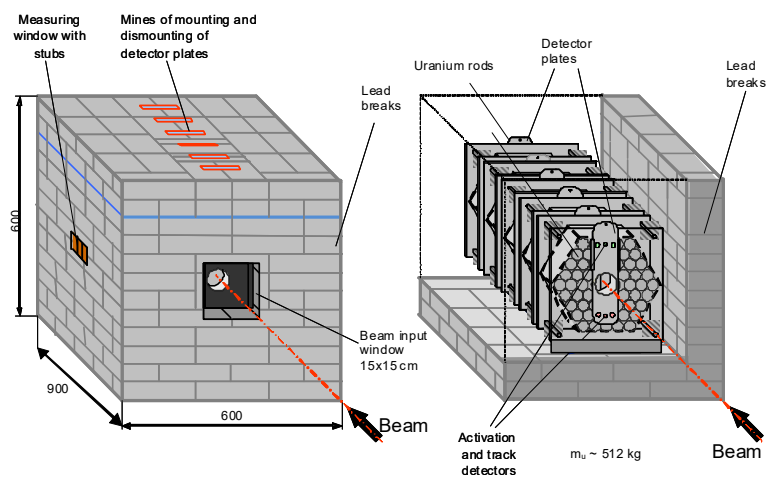

Figure 1: Assembly QUINTA.

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To determine the spatial neutron flux distribution and the reactions caused by them in the volume of the uranium target various types of the detectors have been used. These detectors are placed on 6 removable detector plates. For ^{238}U(n, γ) and ^{nat}U(n, f) reaction rate measurements the activation detectors from natural metallic uranium (diameter 8 mm, thickness 1 mm, mass ~ 0.9 g) were used. In each irradiation 29 activation detectors were used. These detectors were mounted onto six removable detector plates (fig.1) at 0 cm, 4 cm, 8 cm and 12 cm (R) from target axis (Z).

The fission and capture reaction rates of ^{nat}U were determined via gamma spectroscopy of the irradiated uranium foils using HPGe detectors.

The number of neutron radiation capture reactions was determined by the yield of γ-line with 277.6 keV accompanying ^{239}Np decay:

$$^{238}U(n,\gamma)^{239}U\ (\beta^-\ 23.5\ min) \rightarrow {}^{239}Np\ (\beta^-\ 2,4\ day) \rightarrow {}^{239}Pu$$

Fissions reaction rates were determined by yield of gamma-lines 743.4 keV, 364.5 keV, 529.9 keV (87%), and 293.3 keV which accompany decay of fission products ^{97}Zr, ^{131}I, ^{133}I and ^{143}Ce respectively. Cumulative yields (CY) of these mass of products not greatly changed in a wide range of neutron energies. We used the following values of CY: ^{97}Zr -5.5%, ^{131}I -3.6%, ^{133}I - 6.3%, ^{143}Ce -4.3% (average evaluated data for fission-spectrum and 14 MeV). Fission rates obtained for ^{97}Zr, ^{131}I, ^{133}I and ^{143}Ce are averaged.

The overall uncertainties of the reaction rates obtained through gamma spectroscopy method are 12 - 15%, mainly due to errors in cross-section of monitor reactions.

MONTE CARLO SIMULATIONS

The irradiation of the Quinta assembly was simulated using MCNPX 2.7 code [5]. The INCL4-ABLA physics model and LAQGSM code were used. The neutron and charge particle spectrum at the location of irradiated samples was calculated and then the convolution with cross-sections from ENDF/B-VII evaluation performed. In the simulation the detailed geometric model of the target QUINTA and experimental data about beam shape and position on the target were employed.

RESULTS AND COMPARISON WITH MCNPX PREDICTION

Fig. 2 shows the experimental axial distribution of ^{nat}U fission rates for deuteron and ^{12}C ion primary beams with various energy. Value of reaction rates normalized per one gram of ^{nat}U, per one incident particle and per 1 GeV particle energy. For this normalization, the distributions of reaction rates of one type of primary particles is close together for all beam energies. But one can see that with increasing beam energy, a relative decrease in the number of reactions at the first half of the target is observed in the axial distributions of uranium fission events, and a slight increase in these quantities is observed at the same time at the second half of the target along the direction of the primary beam.

Axial distributions of capture reaction rates are similar to axial distributions of fission reaction rates.

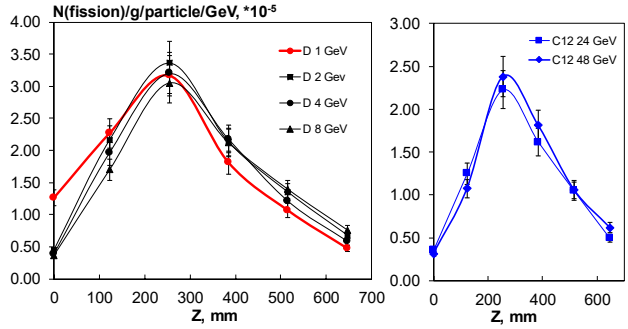

Figure 2: Axial distributions of ^{nat}U(n, f) reaction rates average over radial distribution at each detector plate.

Based on spatial distributions of neutron capture reactions and fissions (both for experimental values and simulated using MCNPX 2.7) the total number of ^{239}Pu production and the total number of ^{nat}U fissions were obtained in the volume of uranium target (see Fig. 3 and Fig. 4). Estimation of these values was carried out in the approximation of the cylindrical target with a radius R = 140 mm (inscribed circle radius of uranium sections). The radial distance between the activation detectors and the primary beam axis calculated taking into account beam position at the front end of the target.

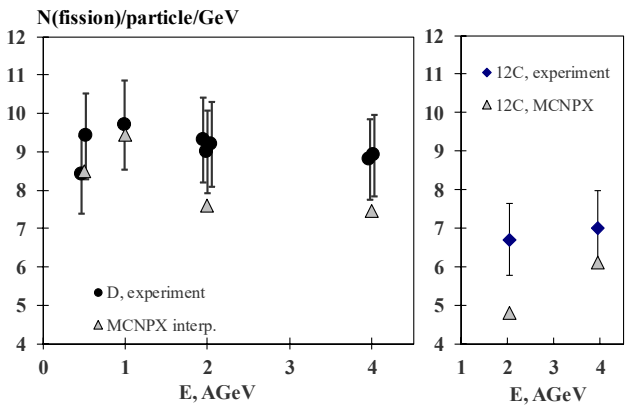

Figure 3: Integral numbers of ^{nat}U(n, f) reactions in the volume of uranium target as a function of beam energy.

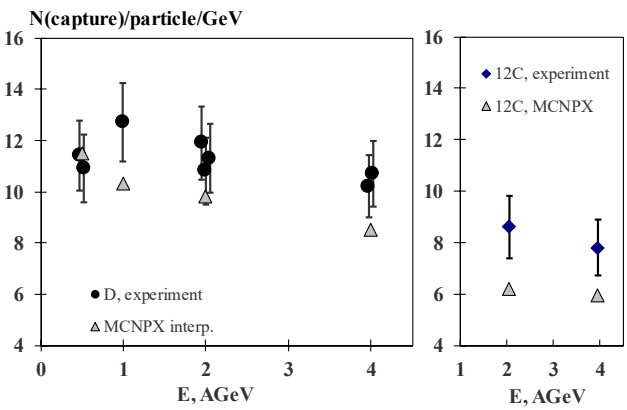

Figure 4: Integral numbers of ^{238}U(n, γ) reactions in the volume of uranium target as a function of beam energy.

The integral number of fissions reactions ($N_{fission}$) in the volume of uranium target of QUINTA, that was obtained by means of gamma spectroscopy, remains approximately constant within our 14% of the experimental errors for 1, 2, 4 and 8 GeV deuteron (per a deuteron and per 1 GeV energy of the beam or per unit of primary beam power). The integral number of fission reactions for carbon beams is smaller than for deuteron beams by 20% - 25%.

For the integral number of capture reactions ($N_{capture}$) we have seen maximum at 2 GeV (1 GeV per nucleon) for deuteron beam. With a further increase in the beam energy, the $N_{capture}$ values decrease.

Note that in the case of a deuteron beam, the $N_{fission}(E)$ dependence may be similar to the capture curve. One can see feebly marked maximum at 2 GeV and an insignificant decrease with increasing energy. However, experimental errors do not lead to a final conclusion.

The ratio of total number of capture to total number of fission decrease from 1.3 at beam energy 1 GeV/nucleon to 1.2 at beam energy 4 GeV/nucleon for deuteron beams, and from 1.3 at beam energy 2 GeV/nucleon to 1.1 at beam energy 4 GeV/nucleon for carbon beams.

The $N_{capture}/N_{fission}$ ratio is independent of beam intensity monitoring and corresponds to average over the target $^{238}U(n, \gamma)/^{nat}U(n, f)$ spectral index. A decrease of these indices shows a hardening of the neutron spectrum inside the uranium target with increasing of primary beam energy.

MCNPX code simulation of integral number of fissions and capture reactions is in agreement with the experimental data in the range of 20%. In case of carbon beam we have 35% deviation between experimental and simulated data.

Another discrepancy between the experiment and the Monte Carlo calculation is shown in Fig. 5.

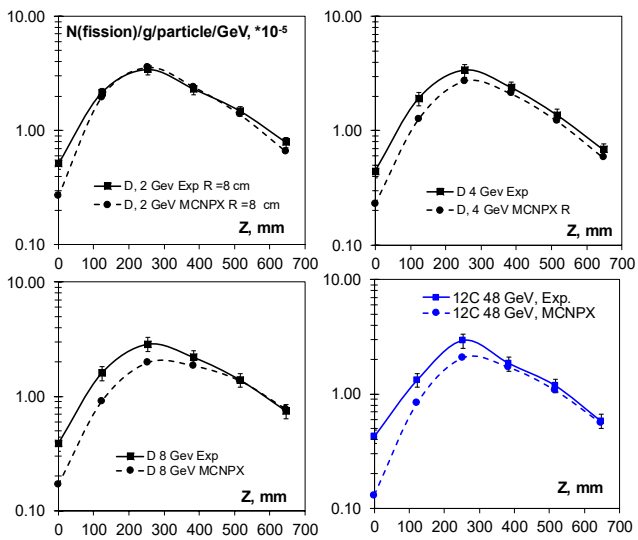

Figure 5: Experimental and Monte Carlo calculation Axial distributions of ^{nat}U fission rates for samples at radial distances of R = 8 cm.

Fig. 5 shows the axial distribution of ^{nat}U fission rates for 6 uranium samples at R = 8 cm. As can be seen in the case of the 2 GeV deuteron beam there is a good agreement between the calculation and experiment. But with increasing beam energy there is an increasing discrepancy between the experimental and calculated data only for the first three detector plates (Z = 0, 123 and 254 mm). Such a discrepancy in backward direction from beam path in target is also observed for samples at R = 4 cm and 12 cm.

CONCLUSIONS

Measurement of the spatial distributions and fission and capture reaction rates allows to estimate such integral characteristics of the QUINTA uranium target as the total number of fissions in the target, the total number of produced plutonium nuclear and the ^{238}U (n, γ)/^{nat}U (n, f) average spectral index.

It is shown that, within 14% of the experimental errors, the total number of fissions in the target normalized per unit of primary beam power is practically independent of the beam energy, and the total produced plutonium nuclei number in the target decreases with beam energy for both deuteron and carbon beams. As a consequence, the average spectral index also decreases, showing a hardening of the neutron spectrum inside the target with energy of the incident particles increasing.

The integral number of fission and capture reactions for carbon beams is smaller than for deuteron beams by 20% - 25% at the same beam power and for beam energy 2 - 4 GeV/nucleon.

We have seen the large discrepancy between the experimental and calculated values of reaction rates in backword direction from beam path in target (first and second target section) for deuteron and carbon beams with 2 and 4 GeV/nucleon.

REFERENCES

[1] W. Furman, J. Adam, A. Baldin, *et. al.*, "Recent results of the study of ADS with 500 kg natural uranium target assembly QUINTA irradiated by deuterons with energies from 1 to 8 GeV at JINR NUCLOTRON", in *Proceeding of Science (Baldin ISHEPP XXI)* 086, Dubna, Russia, Sept. 2012.

[2] N.L. Asquith, S.R. Hashemi-Nezhad, S. Tyutyunnikov, et al., "Study of sub-actinide (^{209}Bi, ^{nat}Pb, ^{197}Au) fission in a sub-critical natural uranium assembly irradiated with 1 and 4 GeV deuterons", *Annals of Nuclear Energy*, vol. 63, pp. 742–750, 2014.

[3] L. Zavorka, J. Adam, V.V. Sotnikov, et al., "Neutron-induced transmutation reactions in ^{237}Np, ^{238}Pu, and ^{239}Pu at the massive natural uranium spallation target", *Nucl. Instr & Meth in Phys. Research, B.,* no. 349, pp.31-38, 2015.

[4] L. Zavorka, J. Adam, M. Artiushenko, et. al., "Validation of Monte Carlo simulation of neutron production in a spallation experiment", *Annals of Nuclear Energy*, vol. 80, pp.178-187, 2015.

[5] D.B. Pelowitz, *MCNPX User's Manual, Version 2.7.0,* Los Alamos National Laboratory, USA, Apr. 2011, 46 LA-CP-11-00438.