NEUTRON–PHYSICAL CHARACTERISTICS OF THE SUBCRITICAL SETUP WITH NATURAL URANIUM BLANKET BOMBARDED BY 4 GeV DEUTERONS

M. Artiushenko, Yu. Petrusenko, V. Sotnikov, V. Voronko, NSC KIPT, Kharkov, Ukraine
A. Patapenka, A. Safronava, I. Zhuk, JIPNR-Sosny NASB, Minsk, Belarus

Abstract

The operation of the nuclear reactor in a subcritical mode is one of the options of safe power engineering. In this case, the reactor must be irradiated by a neutron flux. The neutrons can be produced by using intense beams of relativistic protons or heavier nuclei. In view of this, recent attention of scientists has been focused on both the basic research and the development of specific projects of installations that produce energy. This paper describes experiments that are a part of the research program "Investigations of physical aspect of electronuclear energy generation and atomic reactors radioactive waste transmutation using high energy beams of JINR Nuclotron (Dubna)" ("Energy plus Transmutation" Project). An extended U/Pb-assembly was irradiated with a beam of deuterons having an energy of 4 GeV. Information about the space and energy distributions of neutrons in the volume of the assembly was obtained by means of activation and track detectors. The results are compared with the data on irradiation of U/Pb subcritical assembly by deuterons with energies of 1.6 and 2.52 GeV. The results of numerical calculations performed using the program FLUKA2008 are given.

INTRODUCTION

Transmutations of long-lived actinides and fission products from nuclear waste, plutonium from nuclear weapons, or thorium (as an energy source) have been investigated with increasing interest in the last two decades. Various concepts of transmutation also involve the use of Accelerator Driven Systems (ADS) [1] based on a subcritical nuclear reactor driven by an external spallation neutron source. The spallation reaction is a process, in which a light projectile (*p, n*, light nucleus) with E_n = 0.1 ÷ 1 GeV interacts with a heavy nucleus (e.g., W, Pb) and causes emission of a large number of hadrons. Spallation has two stages: intranuclear cascade (INC) (including pre-equilibrium emission) and deexcitation (evaporation or fission). In the case of a thick target, high-energy particles (mainly neutrons) emitted from the nucleus in the course of intranuclear cascade can induce further spallation reactions and generate internuclear cascade. For some target materials, spallation neutrons with E_n = 10 MeV can increase neutron production by (n, xn) reactions. Mainly, the incident particle induces the production of a great number of neutrons with wide energy spectra, which can be used for transmutation of relevant nuclei.

"Energy plus Transmutation" (E+T) is an international project [2] intended to investigate spallation reactions, neutron production and transport, and the transmutation of fission products and higher actinides by spallation neutrons. A massive lead target surrounded by a subcritical, natural uranium blanket is the center of the experimental assembly. It was irradiated with extracted proton beams of energies 0.7, 1.0, 1.5, 2.0 GeV and deuteron beams of energies 1.6, 2.52, 4 GeV for transmutation studies of radioactive samples of ^{129}I, ^{237}Np, ^{238}Pu, ^{241}Am and ^{239}Pu (see [3, 4]). A variety of different experimental techniques were used, such as classical radiochemical measurements, activation detectors, solid state nuclear track detectors (SSNTD), nuclear emulsions and uranium fission calorimeters. Other research groups [5, 6] have also studied the ADS. Tolstov et al. [7, 8] and Vassil'kov et al. [9] performed irradiation of extended lead targets with protons, deuterons and heavier ions.

EXPERIMENTAL SETUP AND METHODS

The E+T international collaboration has designed a setup for the purpose of transmutation studies in high-energy neutron fields. The setup consists of a cylindrical lead target (diameter 84 mm, length 456 mm) and a surrounding subcritical uranium blanket (206.4 kg of natural uranium). A detailed description of (E+T) assembly can be found in [2-4]. The setup was irradiated for 18 hours with deuterons of kinetic energy 4 GeV at the Nuclotron accelerator LPHE (JINR, Dubna). The integral beam intensity on the target was monitored with aluminium foils, employing the nuclear reaction ^{27}Al(d,αp)^{24}Na, for which the cross section at the corresponding bombarding energy was found in [10] to be 14.63±1.13 mb. The total number of deuterons was determined to be $(1.99\pm0.15)\cdot10^{13}$.

Information on the space and energy distributions of neutrons in the volume of the lead target and the uranium blanket was obtained with sets of activation detectors ^{nat}U and solid-state nuclear track detectors (SSNTD). Spatial distributions of the ^{nat}U fission rate in the volume of the target and blanket assembly were obtained using SSNTD techniques. The activation method was used to obtain the spatial distributions of ^{238}U(n, γ), ^{238}U(n,f) reaction rates. The measuring sensors were located on five detector plates (Z = 0, 118, 236, 354, and 472 mm from the end of the target). Therewith, each plate had 6 positions at different distances from the longitudinal symmetry axis of the target R = 0, 30, 60, 85, 110 and 135 mm. The sensors consisted of a radiator (uranium foils: diameter – 8 mm, thickness – 1 mm), which was in close contact with the

SSNTD. Thus, the foils simultaneously served as a radiator for the SSNTD and as an activation detector. Artificial mica was used as a SSNTD. This type of the track detector has high registration efficiency of fission fragments and eliminates the recoil nucleus background level at the exposure in the hard spectrum of neutron fields.

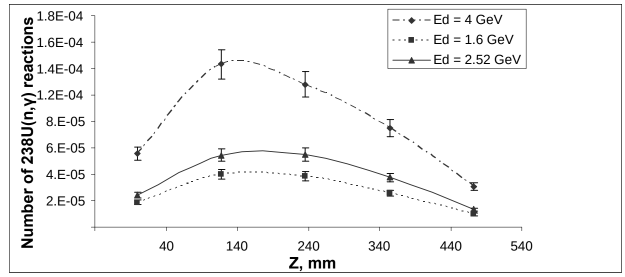
The SSNTD were used to obtain the spatial distribution of ^{nat}U fission reactions in the U/Pb assembly (for more information about working with SSNTD see refs. [11, 12]).

Spatial distribution of the number of ^{238}U neutron radiative capture reactions was measured using uranium foils of natural composition. The number of ^{238}U neutron radiative capture reactions corresponds to the number of ^{239}Pu nuclei, which are formed by the chain of ^{239}U β-decay: $^{238}U(n,\gamma)^{239}U$ *(23,54 min)* $\beta^- \rightarrow {}^{239}Np$ *(2,36 d)* $\beta^- \rightarrow {}^{239}Pu$. The γ-spectra of irradiated foils were measured using two high-purity germanium detectors (HPG). The first detector (with Be window) was used to determine the number of radiative capture reactions (by measuring the yield of 74.66 keV γ-line, which accompanied the ^{239}U decay) immediately after the irradiation. After a four-hour exposure (more than ten ^{239}U decay periods), the second detector (measuring the yield of the 277.6 keV γ-line, which accompanied the ^{239}Np decay) was used. The number of radiative capture reactions was well determined by two detectors; the obtained data were in close agreement within the experimental error. In addition, in the spectra measured by the second detector a great number of γ-lines, which corresponded to the decay of radioactive fission fragments in the range of mass numbers A = 88 ... 146 (^{88}Kr, ^{91}Sr, ^{97}Zr, ^{105}Ru, ^{131}I, ^{132}Te, ^{133}I, ^{135}I, ^{135}Xe, ^{143}Ce, ^{146}Ce, etc.) were identified. From the measured γ-line intensities the total number of nuclei of the nuclides accumulated for the whole irradiation session at different spatial positions of the uranium-lead assembly, was determined. Using the measured data for the fission products, whose yields per fission by neutrons are close in a wide energy range, we can determine the number of ^{238}U fission reactions. The number of nuclear fissions was determined by averaging the results for the following fragments: ^{97}Zr (5.7%), ^{131}I (3.7%), ^{133}I (6.2%), and ^{143}Ce (4.9%).

RESULTS AND DISCUSSION

The spatial distribution of neutron capture reactions ^{238}U (n,γ) for the U/Pb assembly at deuteron energies of 1.6, 2.52, and 4 GeV is shown in Fig. 1.

It can be seen that the curve maxima of axial distributions of the number of captures for the U/Pb assembly are located at distances of about Z = 100 - 130 mm. This suggests that, generally, the process of neutron generation is completed. Then all the curves fall off almost linearly to the rear end of the target. In the radial direction, the rate of the neutron capture reaction ^{238}U (n,γ) decreases with the increasing distance from the axis of the U/Pb assembly. The increased contribution of the neutrons resulting from fission of the uranium blanket is seen at the periphery.



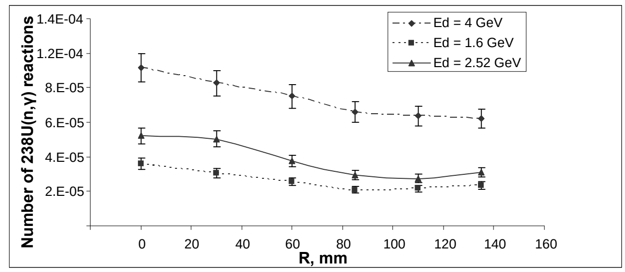


Figure 1. Spatial distributions of neutron capture reactions ^{238}U(n,γ) at deuteron energies of 1.6, 2.52, and 4 GeV. The data are given for one ^{238}U gram and one deuteron incident on the target: top picture shows axial distributions (R = 60, along the axis of the blanket), bottom picture gives radial distributions (Z = 118).

It can be seen that the curve maxima of axial distributions of the number of captures for the U/Pb assembly are located at distances of about Z = 100 - 130 mm. This suggests that, generally, the process of neutron generation is completed. Then all the curves fall off almost linearly to the rear end of the target. In the radial direction, the rate of the neutron capture reaction ^{238}U (n,γ) decreases with the increasing distance from the axis of the U/Pb assembly. The increased contribution of the neutrons resulting from fission of the uranium blanket is seen at the periphery.

The spatial distributions of fission reactions ^{238}U(n,f) measured at deuteron energies of 1.6, 2.52, 4 GeV and the distribution of ^{238}U(n,f) for 4 GeV calculated with the computer numerical code FLUKA2008 are shown in Fig. 2.

It should be noted that the ^{238}U fission is a threshold process, therefore the distribution of the number of fissions displays the distribution of neutrons of energy E > 1 MeV. This is evidenced by the fact that in the case of fission reactions at the periphery of the assembly, the contribution from neutrons produced by fission of the uranium blanket is not observed. One can see that a quicker reduction in the number of fissions in R-distributions in comparison with Z-distributions indicates that the neutron spectrum in the longitudinal direction is harder. Good agreement between the measured and calculated data (Fig. 2) indicates that the model used correctly describes the transport of particles in the blanket and target.

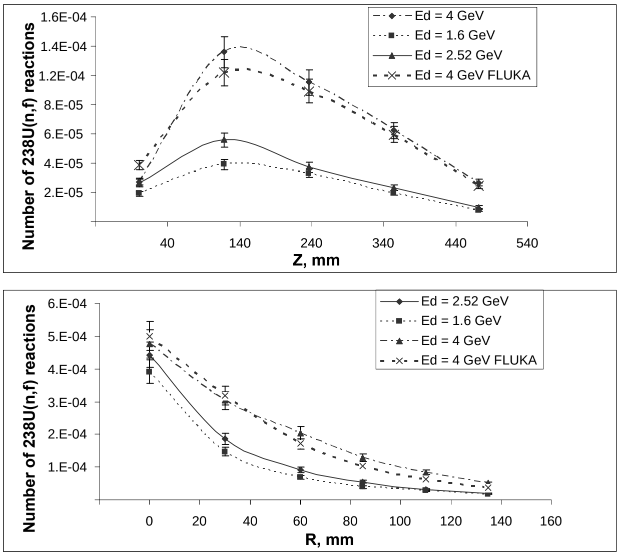


Figure 2. Spatial distributions of fission reactions ^{238}U (n,f) at deuteron energies of 1.6, 2.52, 4 GeV. Data are given per ^{238}U gram and one deuteron incident on the target: top picture shows axial distributions (R = 85, along the axis of the blanket), bottom picture gives radial distributions (Z =118).

Spectral indices or ratios of the average (effective) capture/fission cross sections correspond to the ratio of the neutron number in different energy groups of the neutron spectrum. Thus, they carry information about the spectral composition of the neutron flux in the nuclear installation. A comparison between the measured data and numerical calculations can reveal the source of errors when designing nuclear-physics installations. The experimental values of spectral indices are the most accurate and convenient to use for comparison with the simulation data obtained with the FLUKA program. They do not contain the error associated with the uncertainty of the primary particle fluence bombarding the target in ADS-devices.

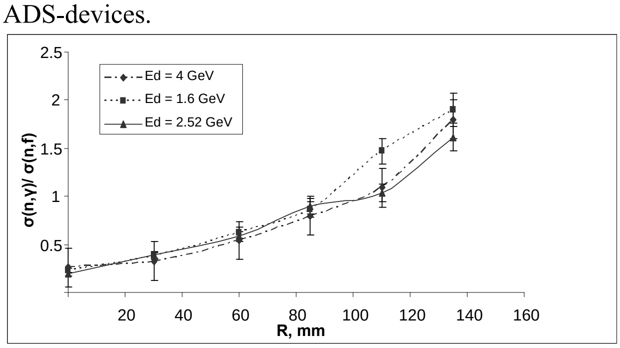


Figure 3. Spatial distributions of spectral indices for U/Pb assembly (E + T) at deuteron energies of 1.6, 2.52, 4 GeV (Z = 118).

The method of solid-state track detectors and the activation technique were used to determine the rates of (n, f) and (n, γ) reactions. Figure 3 shows the spatial distribution of spectral indices for the U/Pb assembly at deuteron energies of 1.6, 2.52, 4 GeV.

Based on the spatial distributions of (n,γ) and (n,f) reactions of ^{238}U in the blanket of assembly (total 15 points), the total number of ^{239}Pu and total number of fission reactions accumulated for all time of irradiation can also be determined. The results are shown in the table.

Table 1: Total number of ^{239}Pu nuclei and total number of ^{238}U (n,f) fission reactions at deuteron energies of 1.6, 2.52, 4 GeV. Data are given for one deuteron incident on the target and GeV^{-1}.

E, GeV	**(n,γ) reaction, experiment**	**(n,f) reaction, experiment**	**(n,f) reaction, FLUKA**
1.60	3.3	3.1	3.1
2.52	3.1	2.8	3.0
4.00	3.9	4.3	3.6

Good agreement between the calculation and experiment is seen from the table. The results given for energy 4 GeV are exceed the results for energies 1.6 and 2.52 GeV.

REFERENCES

[1] C. Rubbia et al., ENEA, ISBN 88-8286-008-6, p.128 (2001).
[2] M. I. Krivopustov, D. Chultem, J. Adam et al., Kerntechnik 68 (2003) 48 – 54.
[3] I. V. Zhuk, M. I. Krivopustov, W. Westmeier et al., Radiation Measurements 31 (1999) 515 – 520.
[4] M. I. Krivopustov, A. V. Pavliouk, A. D. Kovalenko et al., Journal of Radioanalytical and Nuclear Chemistry 279 (2009) 567 – 584.
[5] C. D. Bowman, E. D. Arthur, P. W. Lisowski et al., Nucl. Instr. Meth. Phys. Res A320 (1992) 336 – 345.
[6] A. Abánades, J. Aleixandre, S. Andriamonje et al., Nucl. Instr. Meth. Phys. Res. A478 (2002) 577 – 588.
[7] V. A. Voronko, V. M. D'yachenko, V. Ya. Kostin, K. D. Tolstov et al., Atomic Energy, Springer New York 66 (1989) 252 – 254.
[8] V. M. D'yachenko, V. A. Voronko, V Ya. Kostin, K. D. Tolstov et al., Atomic Energy, Springer New York 67 (1989) 783 – 785.
[9] R. G. Vassil‘kov, V. I. Gol‘danskii, V. V. Orlov et al., Atomic Energy, Springer New York 79 (1995) 257 – 264.
[10] W. Westmeier Private communication - 2010.
[11] S. R. Hashemi-Nezhad, I. V. Zhuk, A. S. Potapenko, et al., Radiation Measurements 43 (2008) 204 – 209.
[12] S. R. Hashemi-Nezhad, I. V. Zhuk, M. Kievets, et al., Nuclear Instruments and Methods in Physics Research Section A 591 (2008) 517 – 529.