E1-2007-7

ABOUT THE FIRST EXPERIMENT ON INVESTIGATION OF ^{129}I, ^{237}Np, ^{238}Pu AND ^{239}Pu TRANSMUTATION AT THE NUCLOTRON 2.52 GeV DEUTERON BEAM IN NEUTRON FIELD GENERATED IN U/Pb-ASSEMBLY «ENERGY PLUS TRANSMUTATION»

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Кривопустов М. И. и др. E1-2007-7
О первом эксперименте по исследованию трансмутации ^{129}I, ^{237}Np, ^{238}Pu и ^{239}Pu на дейтронном пучке нуклотрона в поле нейтронов, генерируемых в U/Pb-сборке установки «Энергия плюс трансмутация» при энергии 2,52 ГэВ

Приведены результаты первого эксперимента по исследованию трансмутации высокотоксичных радиоактивных отходов атомных реакторов на дейтронном пучке нуклотрона ОИЯИ при энергии 2,52 ГэВ и электроядерной установке «Энергия плюс трансмутация». В поле нейтронов, генерируемых в свинцовой мишени и размножающихся в бланкете из естественного урана, экспонировались активационные пороговые детекторы (от Al до Bi), твердотельные трековые детекторы, образцы ^{129}I, ^{237}Np, ^{238}Pu и ^{239}Pu для изучения их трансмутации и ядерные эмульсии. Трансмутация (выжигание) долгоживущих отходов (радиоэкологический аспект) изучалась методом гамма-спектрометрии по определению выходов нуклидов, образующихся в реакциях с нейтронами. С помощью активационных пороговых детекторов и ^{3}He-счетчиков нейтронов исследовано энергетическое распределение нейтронов в объеме и на поверхности уранового бланкета. Проведено сравнение экспериментальных результатов и данных компьютерного моделирования по программе MCNPX.

Работа выполнена в Лаборатории высоких энергий им. В. И. Векслера и А. М. Балдина и в Лаборатории ядерных проблем им. В. П. Джелепова ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 2007

Krivopustov M. I. et al. E1-2007-7
About the First Experiment on Investigation of ^{129}I, ^{237}Np, ^{238}Pu and ^{239}Pu Transmutation at the Nuclotron 2.52 GeV Deuteron Beam in Neutron Field Generated in U/Pb-Assembly «Energy Plus Transmutation»

Preliminary results of the first experiment with deuteron beam with energy 2.52 GeV at the electronuclear setup which consists of Pb-target (diameter 8.4 cm, length 45.6 cm) and $^{\mathrm{nat}}$U-blanket (206.4 kg), transmutation samples of ^{129}I, ^{237}Np, ^{238}Pu and ^{239}Pu (radioecological aspect) are described. Hermetically sealed samples in notable amounts are gathered in atomic reactors and setups of industries which use nuclear materials and nuclear technologies were irradiated in the field of neutrons produced in the Pb-target and propagated in the $^{\mathrm{nat}}$U-blanket. Estimates of transmutations were obtained as result of measurements of gamma activities of the samples. The information about the space and energy distribution of neutrons in the volume of the lead target and the uranium blanket was obtained with help of sets of activation threshold detectors (Al, Co, Y, I, Au, Bi and others), solid-state nuclear track detectors, ^{3}He neutron detectors and nuclear emulsion. Comparison of the experimental data with the results of simulation with the MCNPX program was performed.

The investigation has been performed at the Veksler and Baldin Laboratory of High Energies and at the Dzhelepov Laboratory of Nuclear Problems, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna, 2007

This work is dedicated to the cherished memory of Prof. Vladimir Pavlovich Perelygin

INTRODUCTION

The investigations described in this paper were performed within the framework of the scientific programme called *«Investigations of physical aspect of electronuclear energy generation and atomic reactors radioactive waste transmutation using high energy beams of the Synchrophasotron/Nuclotron JINR (Dubna)»* — programme «Energy plus Transmutation». It has been introduced into the nuclear science community by Krivopustov et al. [1–4]. The scientific description of the project, including main ideas, history, former experiment description and results, uranium fission calorimeter description, experimental methodology used for neutron and proton field properties' investigation (activation and Solid State Nuclear Track Detectors (SSNTD), nuclear emulsions, ^{3}He detectors, etc.), can be found in publications of the «Energy plus Transmutation» collaboration [3–8] and in the overview of JINR research by Baldin et al. and Malakhov et al. (see Ref. [9]).

During 1999–2004 various experiments were made with the «Energy plus Transmutation» assembly employing proton beams with kinetic energies in the range from 0.7 to 2.0 GeV. The experiments were focused on general aspects of energy generation by future Accelerator Driven Systems (ADS), such as neutron generation and multiplication, neutron spectra determination (division into thermal, resonance, fast and high-energy groups), generation of secondary isotopes inside the Pb-target and U-blanket, energy generation and deposition, neutron induced transmutation of long-lived minor-actinides (^{237}Np and ^{241}Am), fission products (^{129}I), and plutonium isotopes (^{238}Pu and ^{239}Pu) [3–17]. These investigations appear to be very important for the development of ADS usable for future nuclear energy production and nuclear fuel cycle safety. This technology has recently attracted considerable attention [18–23].

Usage of the deuteron beam was motivated by the possibility of comparison of the data of neutron generation in our setup with results of Tolstov et al. [24], who used a Pb-slot of $50 \times 50 \times 80$ cm^3, and Vasillkov et al. [25], who used cylinders with diameters from 16 to 20 cm and lengths from 60 to 76 cm. Data can also be compared with our previous proton results and with computer simulations which are in progress now. An interesting result for comparison with protons is the average energy cost for one neutron, i. e., the energy which is needed for production of one neutron in ADS. This value depends on setup, beam type, beam energy, and other variables.

The investigations of transmutation (incineration) of radioactive waste at the high energy accelerator which were started in 1996 in our experiments at the Synchrophasotron proton beams (LHE, JINR) [10] were continued in the present work. This paper describes the first experiment investigating the transmutation of the radioactive nuclides ^{129}I, ^{237}Np, ^{238}Pu and ^{239}Pu with a deuteron beam [13, 15, 17]. The experiment used the setup «Energy plus Transmutation» [1–4] and the superconducting accelerator Nuclotron of the Veksler and Baldin Laboratory of High Energies at JINR (A. D. Kovalenko Ref. [9]).

1. EXPERIMENTAL SETUP

The general scheme of «Energy plus Transmutation» setup [2–4], which was built in 1998–1999 for spent fuels isotopes transmutation investigations is shown in Figs. 1 and 2. The detailed technical design was carried out by the All-Russian Institute of Nuclear Energy Machine Building (VNIIAEM) in Moscow and manufacturing of the steel structure was performed at the mechanical workshop of the Laboratory of High Energies JINR.

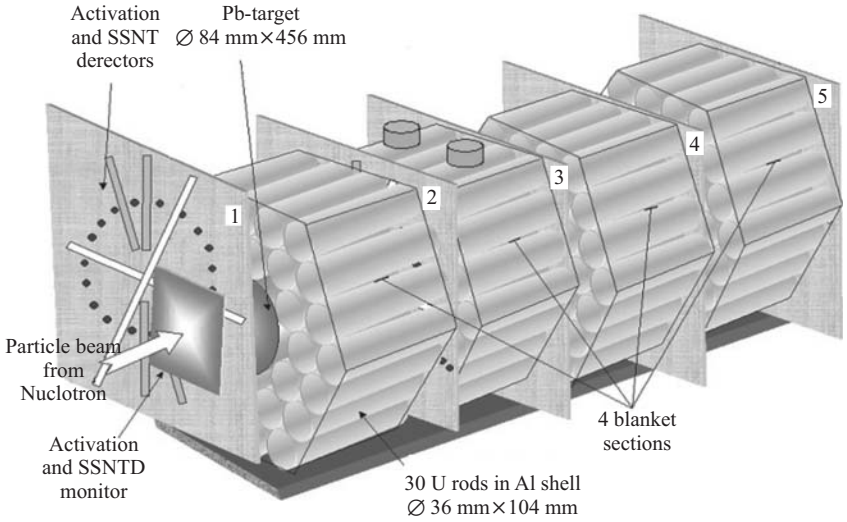

Fig. 1. Scheme of the four-section «Energy plus Transmutation» setup with a massive lead target and uranium subcritical blanket [1–4]. The placement of transmutation samples is presented at the surface of the second section of U-blanket

The «Energy plus Transmutation» setup consists of the following systems:

- Lead target divided into four sections (diameter of 8.4 cm and total length of 45.6 cm; weight of 28.6 kg).

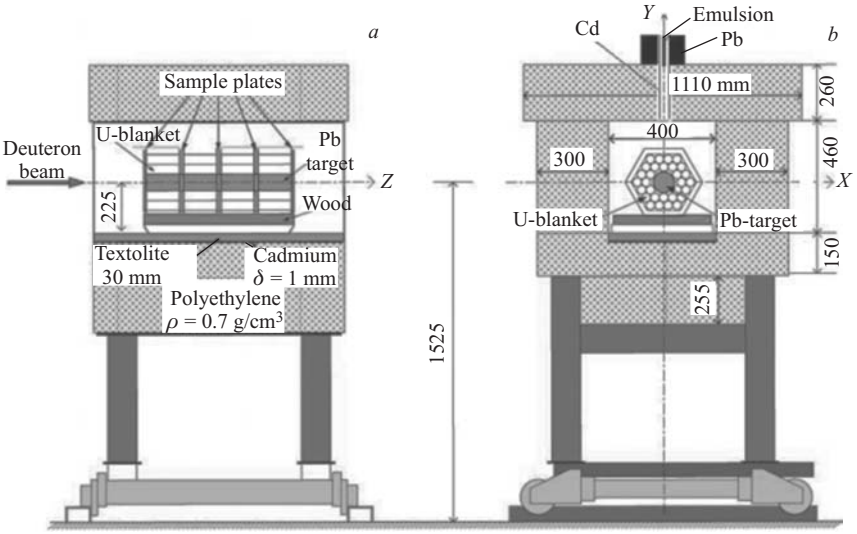

Fig. 2. The U/Pb-assembly of «Energy plus Transmutation» setup [1–5] is inside of a massive shielding and placed onto a mobile platform, which can be moved into and out of the beam line. The left side (a — YZ cross section) of this figure gives a cut through the setup along the beam line, the right side (b — XY cross section) shows a cut through the assembly perpendicular to the beam line in the position. In the holes, on the upside shielding and between the first and second sections of the U/Pb-assembly nuclear emulsion spectrometers are mounted

- Uranium blanket divided into four sections; each section consists of 30 fuel rods of natural uranium inside an aluminium cover (34 mm diameter, 104 mm length, weight of 1.72 kg). Each section contains 51.6 kg of uranium, so the whole blanket contains 206.4 kg of natural uranium.

- Beam monitoring and adjustment of activation system and solid-state nuclear track detectors, proportional ionization chambers and Polaroid films.

- ^{3}He detector system with automatic movement along the setup. The present experiment was the first one where ^{3}He detectors [26] were used in electronuclear investigations. These detectors were used to determine the spatial and energetic distributions of neutron flux density.

- Set of radioactive samples for transmutation studies. This set contains ^{129}I, ^{237}Np, ^{238}Pu and ^{239}Pu nuclides, each hermetically packed inside an aluminium container (see Figs. 3 and 4).

- Backplate for fixation of radioactive samples and other foil-based detectors to the top of the second section of the blanket (see Figs. 2 and 3).

- Five polyethylene plates for fixation of activation threshold and SSNT detectors.

- Five spectrometers based on nuclear emulsion for neutron registration by proton recoil. The results of the first usage of nuclear emulsion technique for investigation of fast electronuclear neutron spectra are presented in our work [5].

- Shielding box filled with granulated polyethylene with boron carbide, and with a cadmium cover; the outside box is made from wood. The box has dimensions of $100 \times 106 \times 111$ cm^3 and weighs 950 kg. It can be moved to the irradiation place at focus F3N of the Nuclotron experimental complex using a special rail system (see Fig. 2).

More detailed information of each part of the setup is given in our publications [1–5].

The system of activation threshold detectors, nuclear emulsion, SSNTD, ^{3}He neutron detectors, and thermometric techniques are in general called uranium fission calorimeter [3].

2. TRANSMUTATION SAMPLES

The transmutation samples (^{129}I, ^{237}Np, ^{238}Pu and ^{239}Pu) were placed on the top of the second section of the uranium blanket (see Figs. 1 and 3) fixed on the

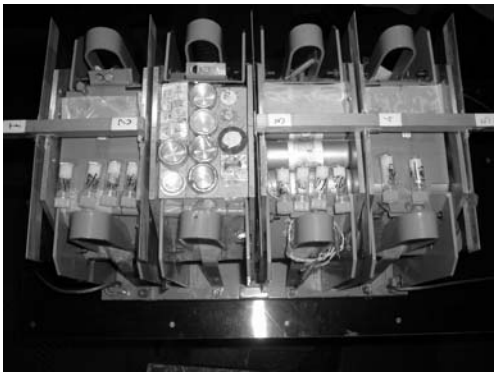

Fig. 3. The four U-blanket sections (top view). In this image the gaps between the blanket section and detector plates (Number 1, 2, 3, 4 and 5) can be seen. On top of the blanket on the surface sections activation detectors and radioactive waste samples of ^{129}I, ^{237}Np, ^{238}Pu and ^{239}Pu for transmutation studies are placed

Table 1. Basic properties of radioactive samples for transmutation studies

Sample	Decay type	Half-life, y	Weight, g	Isotopes, %
^{129}I	β^-	1.57×10^7	0.591 0.121	85 ^{129}I 15 ^{127}I
^{237}Np	α	2.14×10^6	1.085	~ 100 ^{237}Np
^{238}Pu	α	87.7	0.0477	72.92 ^{238}Pu 16.75 ^{239}Pu 2.87 ^{240}Pu 0.35 ^{241}Pu 0.11 ^{242}Pu rest is unknown
^{239}Pu	α	2.41×10^4	0.455	$\sim 100\%$ ^{239}Pu

special paper backplate ($104 \times 140 \times 1$ mm^3). In each experiment only one sample of each isotope was used plus one sample with ^{127}I, which is irradiated to subtract the effect of this nuclide in the ^{129}I sample, which contains 15% of ^{127}I. Also the ^{238}Pu sample is not pure and it contains other plutonium isotopes, mainly ^{239}Pu (16.75%). Radioactive materials are enclosed in an aluminium holder (special duralumin alloy) with diameter of 34 mm (Fig. 4). Some properties of the samples used in 2.52 GeV deuteron irradiation are given in Table 1.

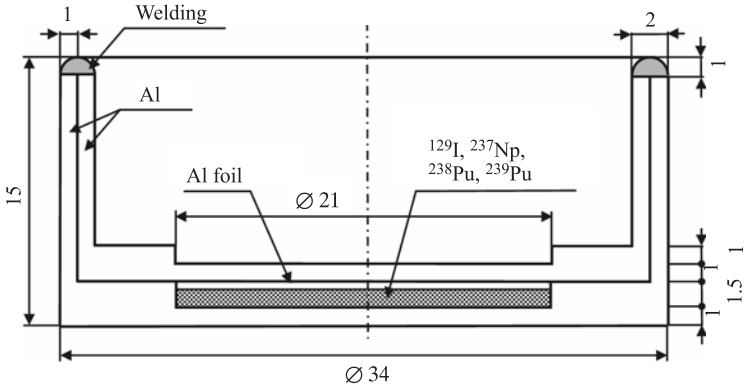

Fig. 4. Weld-sealed aluminium container for ^{129}I, ^{237}Np, ^{238}Pu and ^{239}Pu radioactive samples which were used for this transmutation experiment (dimensions are in mm)

Radioactive samples were manufactured by collaboration of three Russian nuclear research institutes — the Leipunski Institute of Physics and Power En-

gineering in Obninsk, the Bochvar All-Russian Institute of Anorganic Materials in Moscow (VNIINM), and the «Mayak» Plant in Ozersk (Chelyabinsk region). Samples are periodically tested for hermetical properties, especially before and after irradiation by testing for alpha activity on the surface.

3. ACTIVATION AND SSNT DETECTORS, NUCLEAR EMULSIONS AND SAMPLES OF VARIOUS TECHNOLOGICAL MATERIALS

In order to determine the neutron field at the places where transmutation samples were located, activation threshold detectors (of Al, Ti, V, Mn, Fe, Co, Ni, Cu, Y, Nb, In, Dy, Lu, W, Au and Bi) were placed on the second section of the U-blanket (see Figs. 1 and 3).

Neutron distributions were studied along the surface of the U-blanket and on the top of the shielding using SSNT detectors. CR39 foils acting as particle detector were placed parallel to the target axis. One part of the detector was in contact with a neutron converter (Kodak LR115 type 2B, containing $Li_2B_4O_7$). This converter provides information about neutron fluence, detecting the alpha-particle tracks on CR39 emitted via ^{10}B$(n,\ \alpha)^7$Li and ^{6}Li$(n,\ \alpha)^3$H reactions. Another part of CR39 was in contact with the converter and was covered on both sides with 1 mm Cd foils detecting likewise the epithermal neutrons.

The thermal neutron component (up to about 1 eV) was calculated by subtracting the measured track density of the Cd-covered from the Cd-uncovered region of the CR39 detector. Fast neutrons were determined by proton recoil tracks in the detector itself (neutron elastic scattering on hydrogen of the detector) [26]. The neutron energy region detected by proton recoils is between 0.3–3 MeV due to limitations in the proton registration efficiency [27]. SSNTDs were also used as fission detectors. The fissioning target of approximately 100 μg/cm^2 was evaporated on Makrofol E-detector. Targets of ^{235}U and ^{232}Th were used in order to study the thermal (up to about 1 eV) and fast neutron (above 2 MeV) distributions over the U-blanket [26].

SSNTDs were used for beam monitoring (see Sec. 6 of the present work), investigation of the high-energy neutron field ($E > 30$ MeV) between U-blanket sections, determination of fission abundance and energy output in the blanket, share of thermal, epithermal and fast neutrons in the neutron field.

For investigation of spectra of fast neutrons leaving the U/Pb-assembly in our experiment there were used high sensitive thick nuclear emulsions G-5BR (baseless, relativistic) with dimensions of $100 \times 25 \times 1.2$ mm^3. As is demonstrated in Fig. 2 these emulsion spectrometers were placed on upper surface of the shielding of the U/Pb-assembly [5].

Investigations of technical properties of materials used in nuclear facilities, like hafnium, zirconium and epoxy which are of a high importance for accelerators, atomic reactors, physical setup and coupled engineering, have also begun.

4. ^{3}He NEUTRON COUNTERS

The basic characteristics of ^{3}He proportional counter used are summarized in Table 2. The measurement system, presented in Fig. 5, consists of a high-voltage power supply, a preamplifier suitable for proportional counters (Canberra, model 2006), an amplifier (Tennelec model TC205), and a computer based multichannel analyzer (Tennelec PCA-III). The ^{3}He counter was manufactured by LND INC (New York, USA).

Table 2. Basic characteristics of the ^{3}He counter

Detector	Pressure, atm	Gas contens, %	Cathode: material, thickness	Anode: material, diameter	Effective length, cm	Effective diameter, cm
^{3}He	6	^{3}He 64.7 Kr 33.3 CO_2 2.0	Stainless steel 304, 0.089 cm	Tungsten, 0.025 mm	15	5

The system was calibrated using the Tandem, Van de Graaff accelerator facility at the Institute of Nuclear Physics, NCSR Demokritos (Athens, Greece) [28]. The detector was irradiated with mono-energetic neutrons in the energy range of 230 keV $\leqslant E_n \leqslant$ 7.7 MeV, produced via ^{7}Li$(n, p)^7$Be and ^{2}H$(d, n)^3$He reactions. Due to the high pressure and its large dimensions the ^{3}He counter could be used effectively for measuring neutron energies up to about 7 MeV.

A linear response with incident neutron energy was observed for neutron energies up to this energy, both for the full energy peak and the recoil peak. The energy resolution varied from 11% for thermal neutrons down to 4% for higher energies.

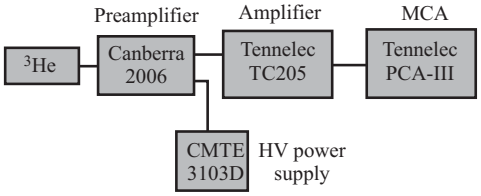

Fig. 5. Neutron counting system (see details in the text)

The disadvantage of ^{3}He counters when they are used in high intensity neutron fields is their relatively high dead time of several tens of μs. In order to avoid space charge effects or even paralyzing of the detector, the maximum count rate must be kept well below 10^4 s^{-1}. In this irradiation the motorized stage, which was specifically designed for holding and moving the counter during

the experiment, was positioned to the maximum possible distance about 4.7 m from the center of U-blanket (see Fig. 6). The cylindrical wall of the counter was covered with 1.2 mm Cd to minimize the contribution of scattered thermal neutrons, coming mainly from the concrete walls. Preliminary ^{3}He counter results are given in Subsec. 7.3. of the present work.

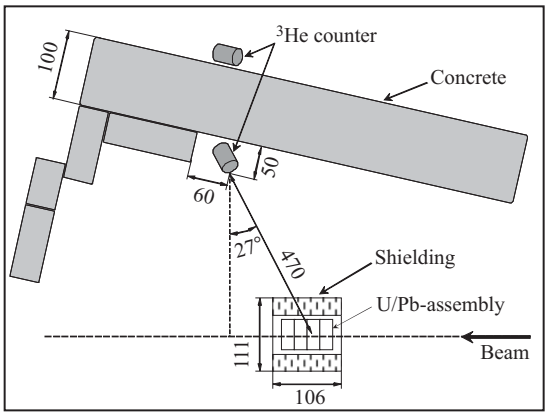

Fig. 6. Arrangement of the counter in respect to the beam direction and U/Pb-assembly of the setup «Energy plus Transmutation» [1–4] (distances are in cm)

5. GAMMA-SPECTRA MEASUREMENT

Analyses of activation threshold detectors, Al and Cu beam monitors and ^{129}I, ^{237}Np, ^{238}Pu and ^{239}Pu transmutation samples started with the gamma-spectra measurement on HPGe detectors provided by Dzhelepov Laboratory of Nuclear Problems of JINR. Description of main parameters of these detectors are given in Table 3. Different geometrical positions as well as various filters composed of Cu, Cd and Pb were used depending on samples activities. Spectra measurements started a few hours after the end of the irradiation and lasted for up to two weeks. The HPGe-detector systems were calibrated using a well-defined ^{57}Co, ^{60}Co, ^{88}Y, ^{137}Cs, ^{152}Eu, ^{154}Eu and ^{228}Th (and daughters products) and ^{133}Ba sources which have several gamma lines ranging from 80 up to 2600 keV.

The gamma spectra were analyzed and the net peak areas were calculated using the DEIMOS program [29]. All necessary corrections for coincidences and background contributions were done. About five hundred gamma-ray spectra were measured and analyzed.

Table 3. Parameters of HPGe gamma-spectrometers

HPGe detector	CANBERRA GR1819	ORTEC GMX-23200	ORTEC GMX-20190-P
Relative efficiency, %	18.9	27.7	28.3
Resolution, keV (at 1332 keV)	1.78	1.86	1.80
Amplifier	ORTEC 973	CANBERRA 2024	CANBERRA 2026
ADC	ORTEC 921 SPECTR. MASTER	ORTEC 919 SPECTR. MASTER	ORTEC 919 SPECTR. MASTER

6. BEAM MONITORING

The beam intensity profile of irradiation of U/Pb-assembly with deuteron beam of the Nuclotron is illustrated in Fig. 7. The irradiation lasted for 7 h 59 min. The total number of deuterons determined by a proportional ionization chamber suffered from a significant systematic error. Therefore, the beam geometry and the total beam fluence were measured separately, each of them by two independent beam monitors.

6.1. Aluminium and Copper Monitors. The total beam intensity was determined by the activation analysis method using foils of Al and Cu. In the process of irradiation, this stable isotope was transmuted by ^{27}Al(d, $3p2n$) reactions into radioactive ^{24}Na. The yield of β radioactive nucleus of ^{24}Na was determined with the help of γ-spectrometry. There is only one available value in the literature of the cross section of the ^{27}Al(d, $3p2n$)^{24}Na reaction [30]:

$$\sigma_{2.33\,\mathrm{GeV}}(^{27}\mathrm{Al}(d,3p2n)^{24}\mathrm{Na}) = (15.25 \pm 1.50)\ \mathrm{mb}. \qquad (1)$$

However, as the excitation function is almost constant in this energy region, we have neglected the difference between the cross sections at 2.33 and 2.52 GeV and we have used the tabulated value (1) also for our calculations.

There were two sets of Al foils activation placed 100 cm in front of the lead target, a distance which is sufficient to suppress the influence of the neutrons evaporated from the target, which could cause the competitive reaction ^{27}Al(n, α)^{24}Na.

The first set was a 10 $\times$ 10 cm^2 Al foil with a thickness of 250 μm**.** Using the cross-section data from (1), the value of the integral deuteron flux determined from this Al foil is $I_d = 6.42(17) \cdot 10^{12}$. The uncertainty of the integral deuteron

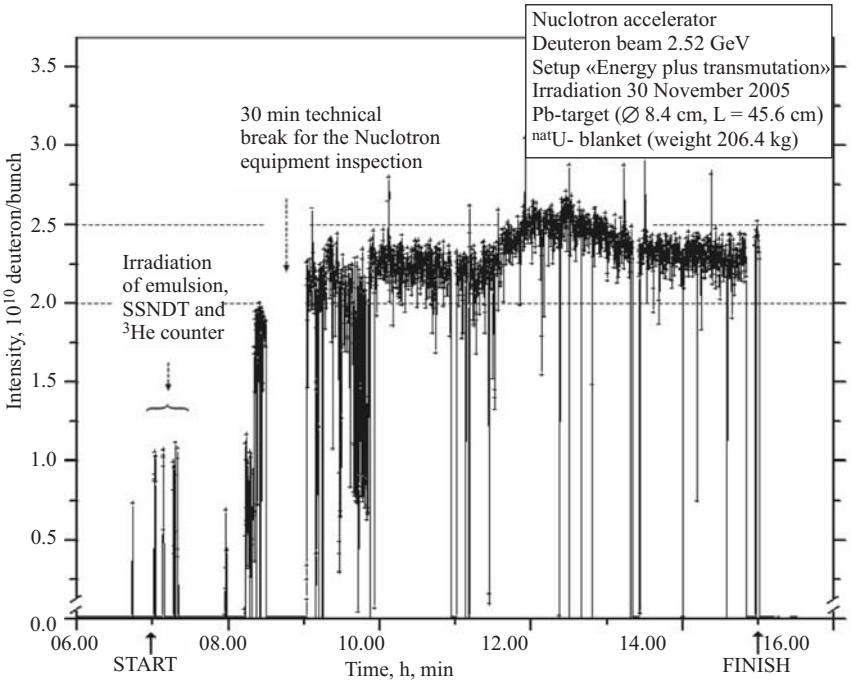

Fig. 7. The intensity profile of the irradiation of the Pb-target with U-blanket at 2.52 GeV deuteron beam, delivered by the Nuclotron [9]

flux given above includes only statistical uncertainties. It is difficult to estimate the inaccuracy of the determination of the corresponding cross section because the only one experimental value is available. The given experimental uncertainty of the known cross section contributes at least $\pm 10\%$.

The second set was a circular ^{27}Al foil with a thickness of 30 μm cuts into an inner circle with diameter of 21 mm and four concentric rings with external diameters of 80, 90, 120 and 160 mm (Table 4). The integral deuteron flux determined from this Al foil is $I_d = 6.50(21) \cdot 10^{12}$ (also statistical uncertainty only). Finally, the weighted average of the integral deuteron flux was determined as $I_d = 6.45(13) \cdot 10^{12}$, where only the statistical uncertainty of the cross-section determination was taken into account.

The beam position was first monitored before the beginning of the irradiation. One bunch of deuterons was sent onto Polaroid films (placed in front of the target) in order to check the shape, location, and alignment of the beam. After the check that the beam

- is centred on the forehead of the target,
- goes approximately in a parallel way with the target axis,

Table 4. Measured activities of single parts of the circular ^{27}Al foil and calculated beam intensities

Inner diameter, cm	Outer diameter, cm	^{24}Na activity, Bq	Statistical uncertainty, %	Beam intensity, 10^9 deuterons/cm^2
0.0	2.1	253	3	7.43
2.1	8.0	274	3	0.60
8.0	9.0	5	20	0.04
9.0	12.0	8	13	0.02
12.0	16.0	11	9	0.01

- has an elliptical shape close to a circle with half-axes of not more than 1 cm length,

two sets of monitors were used to measure the beam profile during the whole period of the target irradiation.

One 6 $\times$ 6 cm^2 Cu foil with a thickness of 100 μm was placed closely in front of the target. After the irradiation, this foil was cut into nine pieces and each of these 2×2 cm^2 foils was measured and analyzed separately. Many products of $^{\rm nat}$Cu(d, X) reactions were measured and the following isotopes were identified: ^{24}Na, ^{43}K, ^{48,44m}Sc, ^{48}V, ^{48}Cr, ^{52}Mn, ^{58,56,55}Co, ^{57}Ni and ^{61}Cu. As there are no available experimental cross sections for $(d + {\rm Cu})$ reactions, only the yields in different foils were compared. We found that the deuteron beam at the entrance to the target was shifted a bit down ($\sim$1 mm) and notably to the right ($\sim$1 cm).

6.2. SSNTD Monitors. The second set of detectors for beam monitoring were SSNTD. The determination of the beam profile using this technique includes the measurement of distributions of fission rates induced in natural lead. Two subsets (lines of 19 lead samples each) were placed just in front of the target in perpendicular directions: from left to right side and from bottom to top. The experimental data were fitted by a Gaussian distribution. The parameters obtained from the fitting were the shift of the beam centre: 0.3(2) cm down, 1.5(2) cm to the right (uncertainties estimated from the inaccuracy of samples location); and the elliptical shape of the beam with the major half-axis in vertical direction ${\rm FWHM_{VER}} = 1.63(20)$ cm and the secondary half-axis in horizontal direction ${\rm FWHM_{HOR}} = 1.56(20)$ cm. These results correspond very well with the results of the first method of the beam measurement. Nevertheless, thanks to its very narrow shape, about 97(3)% of the beam hits the target, as shown in Table 4.

7. PRELIMINARY RESULTS

This Section gives some preliminary results of measurements with the activation threshold detectors from ^{27}Al, ^{87}Y and ^{197}Au (the detectors made of 16 different isotopes and enumerated in Sec. 2 of this paper were used in our experiment), natural U foils, from SSNT detectors and ^{3}He neutron counters. The transmutation (incineration) yield results of radioactive nuclear waste isotopes are also presented. Detailed description of different detectors, methods used in the experiment and analysis of results together with computer simulation will be presented in future publication of the collaboration «Energy plus Transmutation».

7.1. About Transmutation (Incineration) of ^{129}I, ^{237}Np, ^{238}Pu and ^{239}Pu. The transmutation of long-lived radioactive waste nuclides from nuclear reactors and industrial setups which use nuclear technologies and nuclear materials into stable or short-lived radioactive nuclides is the main purpose of the programme «Energy plus Transmutation» [1–4].

The experimental values of transmutation rates or velocity of the reaction $R(A_{\rm res})$ and yield of residual nuclei $B(A_{\rm res})$ are calculated by the following equations:

$$R(A_{\rm res}) = \frac{N(A_{\rm res})}{n_s \cdot I_D}, \quad B(A_{\rm res}) = \frac{N(A_{\rm res})}{m_s \cdot I_D}, \quad R(A_{\rm res}) = B(A_{\rm res})\frac{A_s}{N_{\rm Avo}}, \quad (2)$$

where $N(A_{\rm res})$ — the number of nuclei of isotope $A_{\rm res}$ produced in a sample; n_s and m_s — the number of atoms in the activation detector and its mass in grams; I_D — deuteron fluence in the irradiation; $N_{\rm Avo}$ — the number of Avogadro; A_s — the atomic number of a sample.

Transmutation yields $B(A_{\rm res})$ are ideal benchmark data to test theoretical models that describe the complex processes of interactions of the primary beam particles, the production of spallation neutrons, their secondary reactions in the $^{\rm nat}$U/Pb-assembly, particle transport properties and finally the interaction with the sample nuclide. The correct description of all processes is an essential premise for credible results from the necessary calculations that must be made in order to design a real transmutation setup on the technical or even industrial scale. The ^{129}I, ^{237}Np, ^{238}Pu and ^{239}Pu nuclides are of particular importance as they are long-lived radioactive waste that could be transmuted with neutrons into stable or short-lived isotopes (see the following reactions and Tables 5 and 6):

$$\begin{aligned}
&^{129}\mathrm{I}(n,\gamma)^{130}\mathrm{I}^* \xrightarrow{\beta^-;T_{1/2}=12.36\ \mathrm{h}} {}^{130}\mathrm{Xe}\ (\mathrm{stable}),\\
&^{129}\mathrm{I}(n,nx)\ \text{and}\ {}^{129}\mathrm{I}(n,xnyp) \rightarrow \text{stable and radioactive isotopes},\\
&^{237}\mathrm{Np}(n,f) \rightarrow \text{fission products},\\
&^{237}\mathrm{Np}(n,\gamma)^{238}\mathrm{Np} \xrightarrow{\beta^-;T_{1/2}=2.2\ \mathrm{d}} {}^{238}\mathrm{Pu}^*(T_{1/2}=87.74\ \mathrm{y}),\\
&^{238,239}\mathrm{Pu}(n,f) \rightarrow \text{fission products}.
\end{aligned} \quad (3)$$

Taking into account that samples of ^{237}Np, ^{238}Pu and ^{239}Pu are irradiated with fast neutrons with energies up to hundreds of MeV fission and also spallation reactions are possible.

Radioactive samples that are exemplified nuclear waste isotopes being serious from the safety point of view were irradiated with secondary neutrons generated by the spallation reactions of 2.52 GeV deuterons on the lead target and also by neutrons produced inside the subcritical uranium blanket. The radioactive samples of ^{129}I and ^{237}Np (see Table 1 and Figs. 1 and 3) and stable ^{127}I were irradiated on the top surface of the second section of uranium blanket. Transmutation rates of the isotopes — i. e., the yield of product nuclei — were investigated by gamma-spectrometric methods. We obtained data on the absolute reaction rates as an $R(A_{\rm res})$-parameter, which is the number of product nuclei produced per atom of the sample, per one incident deuteron or proton.

The irradiation of ^{127}I was used for subtraction of a 15% contamination in the ^{129}I sample. The results are given in Table 5. The data from interaction of 2.0 GeV protons with the same targets under the same experimental conditions are also presented [12]. Delay between the end of irradiation and the first γ-spectra measurement (i. e. the cooling time) was 5 h for protons and 11 h for deuterons. Short-lived nuclei such as ^{128}I ($T_{1/2} = 28$ min) therefore could not be detected. It is seen from Table 5, that the results from deuterons of 2.52 GeV (the present work) and protons of 2.0 GeV [12] show remarkably similar yields. This means that the product cross section is mainly determined by the full energy of the input particles — deuterons or protons.

However, for ^{237}Np as sample the yield of product nuclei from 2.52 GeV deuterons is systematically higher, almost the factor of 1.3, than in case of 2.0 GeV protons.

Two plutonium isotopes (^{238}Pu and ^{239}Pu) were also irradiated on top of the second section of the U/Pb-assembly (see Figs.1 and 3), close to the ^{129}I and ^{237}Np samples. In Table 6 product yields are given.

As mentioned above, measurements of γ-spectra were started about 11 hours after the end of irradiation. Due to such long cooling time, it is impossible to see product isotopes with short life-times which were actually observed in experiments with protons. This is especially obvious in ^{238}Pu sample, in which only two nuclei products were observed.

It should be mentioned, that besides our experiments on investigation of transmutation of radioactive waste atomic reactors (^{129}I, ^{237}Np, ^{238}Pu, ^{239}Pu and ^{241}Am) see, for example, our results presented in the publications [5, 10–17] about study of transmutation fission products of ^{99}Tc and ^{129}I at beams of three different accelerators and setups also mentioned in the works [23, 31].

7.2. Studies of Neutron Distributions Using Activation Detectors. We studied the spatial and energetic distributions of neutrons produced at different places of the setup. The neutrons induced γ-emitting products in activation detector

Table 5. Preliminary results about product nuclei observed in ^{127}I, ^{129}I and ^{237}Np samples; $R(A_{\rm res})$ yields resulting from irradiations with proton and deuteron beams

Residual nuclei	$T_{1/2}$	Deuterons 2.52 GeV	Protons 2.0 GeV [12]
^{127}I sample, $R(A_{\rm res}) \cdot 10^{29}$			
^{111}In	16.78 d	0.50(7)	0.38(10)
^{119}Te	16.03 h	1.15(18)	1.31(27)
^{119m}Te	4.70 d	1.15(26)	1.03(12)
^{121}I	2.12 h	3.87(100)	3.13(23)
^{122}Sb	2.72 d	1.24(15)	–
^{123}I	13.3 h	11.6(14)	13.0(10)
^{124}I	4.18 d	18.3(11)	19.0(10)
^{125}Xe	16.9 h	20.1(86)	–
^{126}I	13.11 d	70.4(3)	81(4) 10
^{129}I sample, $R(A_{\rm res}) \cdot 10^{29}$			
^{121}Te	16.78 d	4.93(94)	–
^{124}I	4.18 d	4.38(125)	4.0(5)
^{126}I	13.11 d	10.8(25)	22.5(44)
^{130}I	12.36 h	816(40)	809(33)
^{237}Np sample, $R(A_{\rm res}) \cdot 10^{26}$			
^{97}Zr	17.0 h	0.188(29)	0.159(8)
^{99}Mo	2.75 d	1.64(47)	–
^{132}Te	3.26 d	0.217(32)	0.147(11)
^{133}I	20.8 h	0.265(75)	0.182(28)
^{238}Np	2.12 d	17.0(8)	13.3(3)

foils through (n, xn), (n, α) and (n, γ) reactions. Different thresholds of these reactions allow probing the energy spectrum of neutrons. Foils of three elements (Al, Y and Au) were placed at different positions of the setup and also inside the lead target. The activation detectors of the first set were placed at the distances of 0; 11.8; 24.0; 36.2 and 48.4 cm from the front of the Pb-target and at the distances from 0.0 to 13.5 cm from the Pb-target axis (see Fig. 8).

Table 6. Preliminary results about product nuclei observed in ^{238}Pu and ^{239}Pu samples; $R(A_{\rm res})$ and $B(A_{\rm res})$ yields resulting from the irradiation with a deuteron beam

Product nuclei	$T_{1/2}$	$B(\Delta B)\cdot 10^5$	$R(\Delta R)\cdot 10^{27}$
^{238}Pu sample			
^{97}Zr	16.9 h	4.51(11)	15.6(4)
^{135}Xe	9.14 h	8.0(9)	29(4)
^{239}Pu sample			
^{103}Ru	3.93 d	5.0(4)	19.8(17)
^{128}Sb	9.01 h	0.18(5)	0.72(22)
^{132}Te	3.2 d	4.3(4)	16.9(17)
^{133}I	20.8 h	6.8(7)	27(3)
^{135}I	6.57 h	4.6(8)	18(3)
^{135}Xe	9.14 h	3.0(8)	12(3)
^{140}Ba	12.75 d	5.2(6)	20.4(23)
^{143}Ce	33.04 h	3.6(4)	14.3(15)
^{91}Sr	9.63 h	2.6(4)	10.3(17)
^{97}Zr	16.9 h	5.3(4)	20.9(17)

After the irradiation the activated foils were transported to the gamma-spectroscopy laboratory to investigate activities with the HPGe detectors for the determination of decay rates of produced radioactive nuclides. Activities at the end of the bombardment were converted into production rates $B(A_{\rm res})$ of these nuclei (see Eq. (2)). The production rate $B(A_{\rm res})$ is very sensitive to the energy threshold of the reaction and hence also to the neutron spectrum in the foil position. Gamma-radiation of each foil was measured twice at different times after irradiation to distinguish between isotopes with different half-life.

The results from the analysis of several gamma-ray line intensities from two spectra were used to calculate the experimental production rate $B(A_{\rm res})$. Weighted averages over the number of spectra were determined for each individual isotope and foil. Resulting values were used for further analysis.

Radial distributions of production rates $B(A_{\rm res})$ for several isotopes produced in Au and Al activation detectors are shown in Figs. 9–11 as examples.

Yttrium-89 activation detectors that were already used in former experiments [32] were placed on plastic foils in front of and between the four sections,

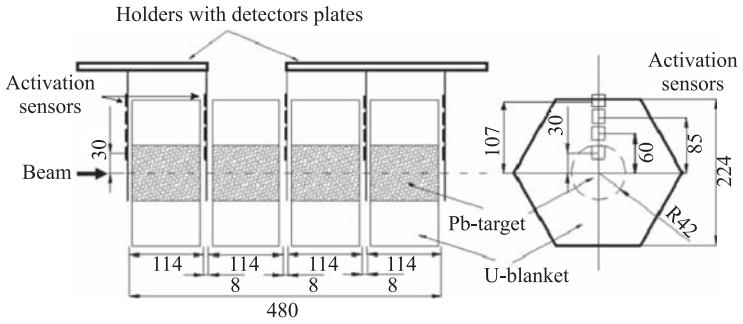

Fig. 8. Typical placement of activation detectors (see also Figs. 2 and 3)

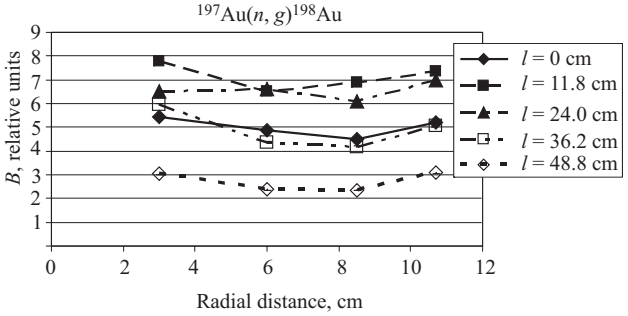

Fig. 9. Radial distributions of production rates $B(A_{\rm res})$ for isotope ^{198}Au produced on Au foils by non-threshold (n, γ) reaction for different distances from the front of the Pb-target. The lines are drawn to guide the eyes

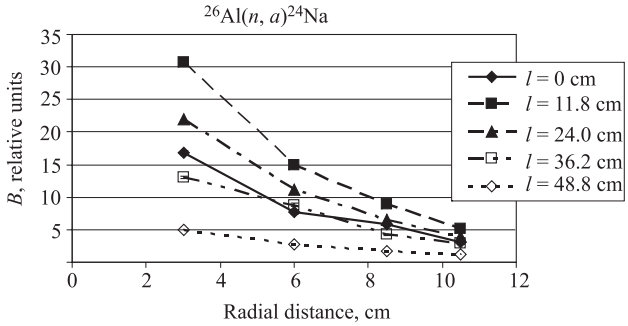

Fig. 10. Radial distributions of production rates $B(A_{\rm res})$ for isotope ^{24}Na produced in Al foils by (n, α) reaction with threshold energy of 5.5 MeV. Distributions are shown for different distances from the front of the Pb-target. The lines are drawn to guide the eyes

as well as on the rear of the U/Pb-assembly in several positions at varying radial distances from assembly's symmetry axis. The preliminary results presented

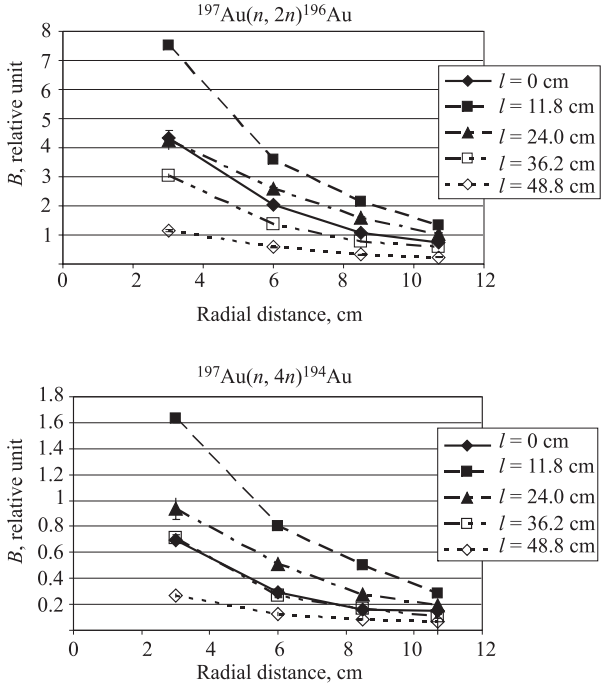

Fig. 11. Radial distributions of production rates $B(A_{\rm res})$ for ^{196}Au (top) and ^{194}Au (bottom) isotopes produced on Au foils by (n, $2n$) and (n, $4n$) reactions with threshold energies of 8.1 and 23.2 MeV, respectively. The lines are drawn to guide the eyes

below give spatial distributions of ^{88}Y, ^{87}Y and ^{86}Y isotopes production from (n, xn) reactions (see Table 7 and Fig. 12).

General features of experimental distributions are as follows:

- there is a clearly defined maximum in yield around the distance $Z = (16 \pm 2)$ cm from the front of the target;
- the $B(A_{\rm res})$ parameter is decreasing with increasing radial distance from the target axis (beam axis position);
- while $B(A_{\rm res})$ parameter distributions for the non-threshold (n, γ) reaction are almost flat, the corresponding distributions of threshold reactions change over magnitude and in case of radial distributions are close to an exponential form.

One can conclude that epithermal and resonance neutron fluxes are almost constant in the Pb-target volume whereas the flux of high energy neutrons strongly changes with radial distance from the target axis. These features are very similar

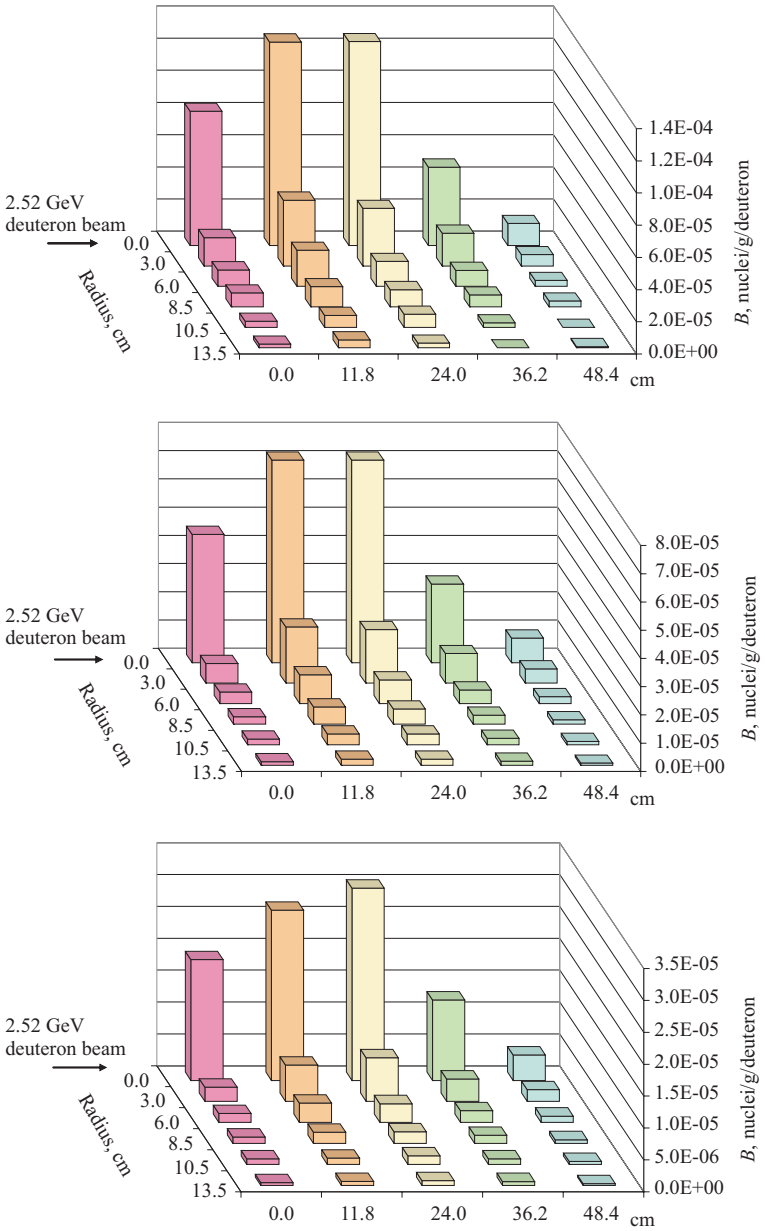

Fig. 12. Distributions of production rates $B(A_{\rm res})$ for isotopes ^{88}Y (top), ^{87}Y (middle) and ^{86}Y (bottom) produced on Y foils by $(n,\, 2n)$, $(n,\, 3n)$, and $(n,\, 4n)$ reactions with threshold energies 11.5, 20.8 and 32.7 MeV, respectively

Table 7. Preliminary results of $B(A_{\rm res})\cdot 10^5$ — parameter for product observed in ^{89}Y detectors, positioned at 0.0; 11.8; 24.0; 36.2 and 48.4 cm in front of the Pb-target (see Figs. 2 and 8)

Residual nuclei, $T_{1/2}$, used γ lines	Radial distance, cm	Axial position, cm				
		0.0	11.8	24.0	36.2	48.4
^{88}Y	0.0	8.34 (6)	12.7 (4)	12.7 (17)	4.874 (12)	1.37 (5)
	3.0	1.77 (11)	4.1 (4)	3.58 (12)	2.04 (13)	0.70 (8)
$T_{1/2} = 106.65$ d,	6.0	1.03 (10)	2.25 (8)	1.56 (3)	0.997 (2)	0.38 (12)
$E\gamma = 898.0$ and	8.5	0.87 (20)	1.26 (5)	1.075 (2)	0.741 (9)	0.373 (5)
1836.0 keV	10.5	0.38 (11)	0.74 (17)	0.82 (3)	0.285 (19)	–
	13.5	0.23 (5)	0.50 (4)	0.30 (10)	–	0.075 (6)
^{87}Y	0.0	4.55 (5)	7.175 (13)	7.18 (4)	2.785 (8)	0.875 (8)
	3.0	0.694 (4)	1.988 (11)	1.899 (21)	1.048 (1)	0.509 (3)
$T_{1/2} = 3.32$ d	6.0	0.415 (10)	1.025 (5)	0.841 (3)	0.483 (5)	0.250 (3)
$E\gamma = 388.5$ and	8.5	0.264 (11)	0.611 (3)	0.540 (13)	0.323 (2)	0.158 (1)
484.8 kev	10.5	0.197 (4)	0.383 (7)	0.379 (9)	0.227 (7)	0.115 (10)
	13.5	0.126 (3)	0.216 (4)	0.211 (2)	0.141 (9)	0.074 (5)
^{86}Y	0.0	1.89 (13)	2.67 (10)	3.02 (15)	1.26 (8)	0.400 (27)
	3.0	0.224 (15)	0.57 (4)	0.69 (3)	0.350 (25)	0.184 (14)
$T_{1/2} = 0.614$ d	6.0	0.141 (17)	0.304 (22)	0.29 (3)	0.185 (17)	0.098 (13)
$E\gamma = 1076.0$ keV	8.5	0.100 (23)	0.174 (16)	0.184 (21)	0.126 (11)	0.057 (8)
	10.5	0.086 (19)	0.099 (10)	0.133 (16)	0.085 (15)	0.046 (10)
	13.5	0.042 (13)	0.060 (8)	0.075 (10)	0.060 (13)	0.026 (6)

to previous experimental results obtained using proton beams [5, 32, 33] and they fit together with systematics of proton experiments.

The comparison between experimental and simulated production rates of many threshold reactions makes it possible to test the accuracy of the description of neutron production over a wide neutron energy interval. It is also possible to restore the neutron energy spectrum using the cross sections dependence on neutron energy, provided that a sufficient number of production reactions have been determined.

7.3. Methodical Tests of Neutron Measurements Using ^{3}He Counters. At the beginning of the experiment (see Fig. 7), dedicated to the irradiations of emulsions and track detectors were made and several neutron spectra were simultaneously collected. In all of these spectra a distortion of the thermal peak (the reaction ^{3}He(n, p)^{3}H has a Q-value of +764 keV) due to space charge effects is observed, despite the relatively small intensity of the beam. As an example, the

spectra collected during the Polaroid exposure (1 pulse) and during the irradiation of emulsions (6 pulses) are presented in Fig. 7. The count rate during these measurements was calculated to be in the range from 14 to 17 kcps. To prevent detector overload the main counter was placed behind the concrete shield for the rest of the experiment (Fig. 6).

The spectrum collected there during this irradiation is also presented in Fig. 13. The mean count rate during this measurement was about 20 kcps. The second peak at about 1.5 MeV presented in all spectra is a coincidence peak of two thermal neutrons.

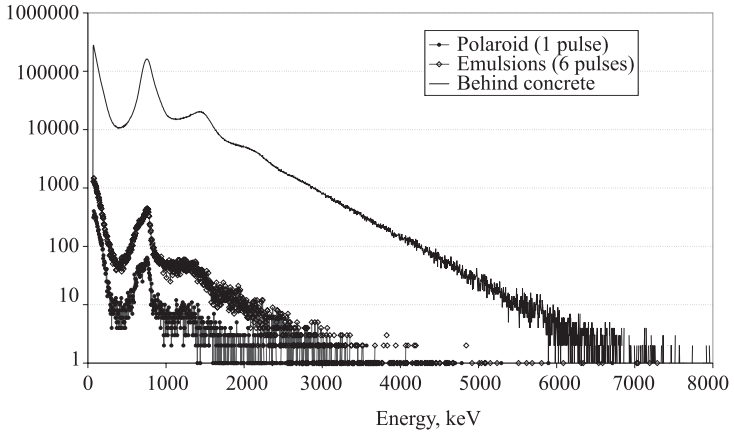

Fig. 13. The spectra collected during the irradiation of the «Energy plus Transmutation» setup with a deuteron beam of 2.52 GeV (Fig. 7). The «bottom» and the «middle» spectra were accumulated by the detector, placed in front of the concrete wall, during 1 and 6 impulses of the deuteron beam, respectively. The «top» spectrum shows the neutron spectrum, collected with the detector, placed behind the concrete wall (see Fig. 6), during 7 h of irradiation of the target

According to the above observations the count rate in all collected spectra exceeded the limit of this system for neutron spectroscopy. Useful information about neutron spectra of «Energy plus Transmutation» setup can be obtained only with lower deuteron intensity and pulses of longer duration.

7.4. Measurements of Neutron Characteristics Using SSNTD. The SSNT detectors mentioned in Sec. 3 of the present work were used in the same way as in the series of previous experiments at the «Energy plus Transmutation» setup at the Synchrophasotron and Nuclotron proton beams [4–8, 13, 16].

7.4.1. The fast neutron distribution along the U-blanket. The neutron flux on the U-blanket surface reflects neutron production along the Pb-target from spallation reactions by primary and secondary particles and also neutron production in the uranium blanket by secondary particles, keeping in mind that the beam diameter is smaller than the target diameter.

According to the experimental data, thermal neutrons were not detected because no difference between the tracks density on the Cd-covered and the Cd-uncovered regions of CR39 with $Li_2B_4O_7$ converter was observed. The same conclusion was verified by the supplementary measurements of thermal neutrons using ^{235}U fission detectors [34]. The track density of the Cd-covered region of CR39 with $Li_2B_4O_7$ converter corresponds to epithermal neutrons (up to 10 keV) fluence which presented to be one order of magnitude less than fast neutrons fluence (between 0.3–3 MeV) detected by proton recoil on the CR39 detector itself. Thus, it is obvious that the U/Pb-assembly produces mostly fast neutrons.

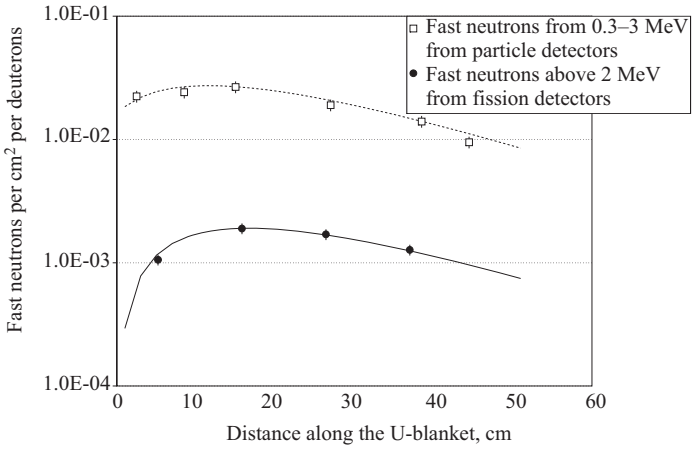

Fig. 14. The fast neutron distribution along the U-blanket measured by particles and fission detectors

Fission detectors with ^{232}Th converter were also used in order to measure fast neutron fluxes above 2 MeV. The fast neutron distributions along the U-blanket measured by both methods are in good agreement as is seen in Fig. 14. In addition, approximately the same values of the neutron fluxes have been calculated using the high-energy transport code DCM-DEM (Dubna). The fast neutron production increases up to about 15 cm from the beam entrance and then decreases along the target as presented in Fig. 15. The same intensity distribution and approximately the same intensities of fast neutrons have been determined in an irradiation of the same spallation source by a 1.5 GeV proton beam [35].

The fluence of thermal and fast neutrons escaping the shielding was also measured with CR39 detectors. The intensity of fast neutrons escaping the polyethylene was two orders of magnitude less than on the U-blanket surface (Fig. 15).

While less than a half of these neutrons are in the thermal energy range about $2 \cdot 10^5$ neutrons per cm^2 per deuterons. Therefore the polyethylene shielding proves to be an efficient moderator thermalizing a large amount of the fast neutrons produced by the spallation neutron source.

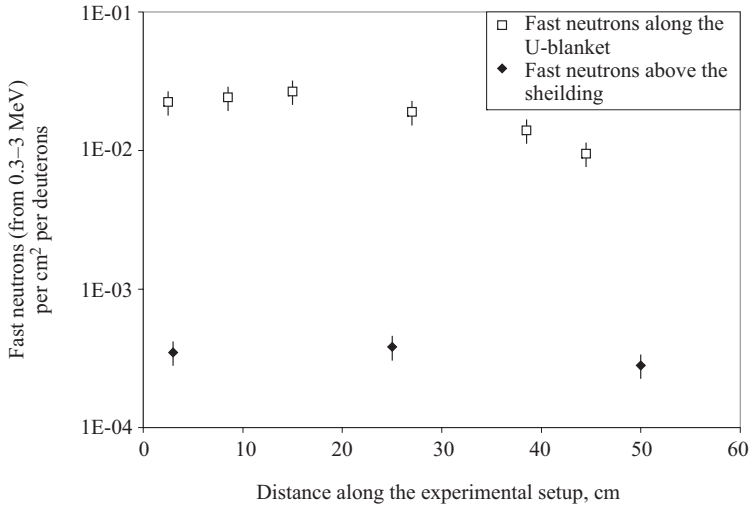

Fig. 15. Neutron distribution escaping the polyethylene shielding and comparison with neutron distribution along the U-blanket surface

7.4.2. The spatial and energy distribution of neutrons. This part includes measured results of fission rate distributions of $^{\mathrm{nat}}$Pb and ^{238}U, capture reactions of neutrons by ^{238}U, and also the spectral index $\overline{\sigma}_{\mathrm{capt}}^{238\mathrm{U}} / \overline{\sigma}_{\mathrm{f}}^{238\mathrm{U}}$ at various radial positions at the U/Pb-assembly. Moreover, comparison of the experimental data with results of calculations using modern mathematical programs of simulation on the basis of Monte Carlo method are presented. One of the primary aims was the development of a technique of measurement of the spectral index. This technique allows determination of the spatial distribution of ^{239}Pu-accumulation in the $^{\mathrm{nat}}$U-blanket as a result of neutron capture reactions by ^{238}U. The spectral index is defined as the ratio of the number capture reactions to the number of fissions in the U-target material. Results of measurements and calculations of axial and radial distributions of ^{238}U (n, γ) neutron capture reactions are given in Fig. 16.

It can be seen from Fig. 16, a that results of measurements and calculation of reaction rates of neutron capture of ^{238}U in the center blanket differ by 30%. In radial direction (Fig. 16, b) the cross section of ^{238}U (n, γ) neutron capture reactions decreases with distance from the Pb-target axis. It should be noted that experimental results for distance of 85–135 mm from the axis of assembly are of special interest for comparison with the calculations by various codes. The significant difference that the experimental reaction rate exceeds calculation on average by the factor of 1.5 is explained by the influence of the neutrons slowed down and reflected by the biological shield.

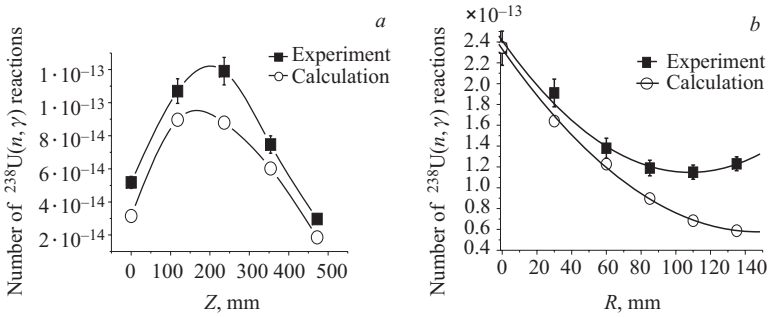

Fig. 16. *a*) Longitudinal distribution along the uranium blanket (at radial distance $R = 85$ mm from the symmetry axis of the assembly); *b*) Radial distribution (at longitudinal distance $Z = 118$ mm from the target front) of number of neutron capture reactions ^{238}U (n, γ). The data are normalized to one atom of ^{238}U and one incident deuteron

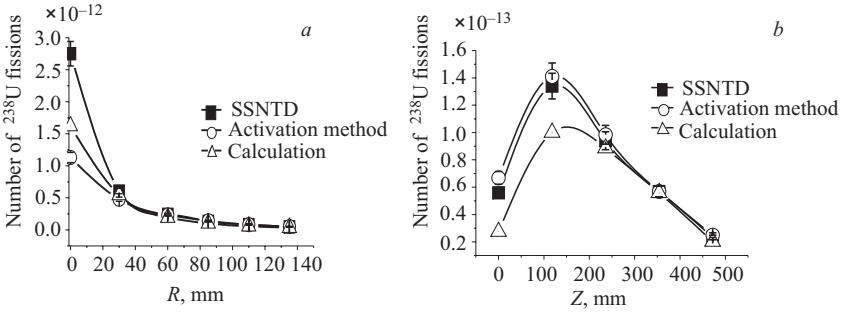

Fig. 17. *a*) Radial (at longitudinal distance $Z = 118$ mm) and *b*) axial (at radial distance $R = 85$ mm from the target symmetry axis) distributions of number of fission reactions of ^{238}U. The data are normalized to one nucleus of ^{238}U and one incident deuteron

Knowing the yield of fission products [36], it is possible to determine the fission density distribution of ^{238}U. It is interesting to compare results of the measurements of two independent experimental methods among themselves and also with calculated results.

It is obvious from Fig. 17, *a* that radial distributions of numbers of fission reactions of ^{238}U determined by two experimental methods coincide within the limit of experimental uncertainties at distances larger 30 mm from the Pb-target axis. Calculation also describes fission process in the blanket material (natural uranium) and on the periphery of the assembly very well.

Results of experiment and calculated values of a spectral index coincide within limits of experimental uncertainties (Fig. 18). A difference (1.5 times) is observed at the periphery of the assembly. This also shows the underestimation in calculations of the influence of neutrons moderated in and reflected by the biological shielding.

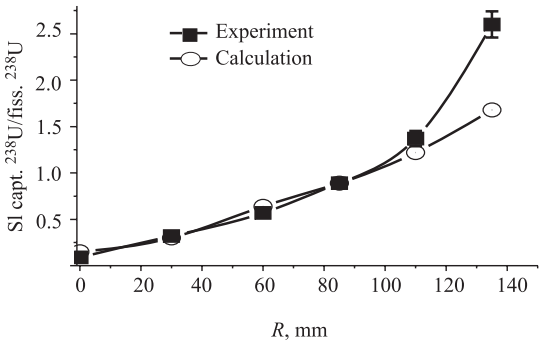

Fig. 18. Radial distribution of the spectral index (longitudinal distance $Z = 118$ mm from the Pb-target front). Lines connecting the data points are drawn to guide the eyes

It is obvious from Fig. 18 that the number of fissions of ^{238}U in a material blanket exceeds the number of neutron captures by the factor of three at the border between the lead target and the blanket ($R = 32$ mm). Neutron capture then begins to prevail with increasing radial distance, probably because neutrons are moderated by collisions in the blanket material.

The procedure of combining the track counting and gamma-spectrometry techniques for the determination of spectral indices is a new development. It involves receiving of information from the same sample by SSNTD methods, i. e., counting the fission tracks of ^{238}U, and by γ-ray spectrometry methods, i. e., counting a γ line from the nuclide ^{239}Np at 277.6 keV.

$$^{238}\mathrm{U}(n,\gamma)^{239}\mathrm{U}^{*} \xrightarrow{\beta^{-};T_{1/2}=23.4\ \mathrm{min}} {}^{239}\mathrm{Np}^{*} \xrightarrow{\beta^{-};T_{1/2}=2.36\ \mathrm{d}} {}^{239}\mathrm{Pu}^{*} \xrightarrow{\beta^{-};T_{1/2}=2.41.10^{4}\mathrm{y}} . \quad (4)$$

The novel methods allows one to measure the spectral index with an uncertainty of no more than $\pm 15\%$. The new technique allows quantitative determination of ^{239}Pu accumulation in the $^{\mathrm{nat}}$U-blanket. Experimental value of total mass of ^{239}Pu accumulated in the blanket is $1.6(2) \cdot 10^{-8}$ g, which compares well with the calculated value of $1.43 \cdot 10^{-8}$ g using the MCNPX code [37].

CONCLUSIONS

We performed the first experiment on investigations of transmutation (incineration) of radioactive waste atomic reactors (^{129}I, ^{237}Np, ^{238}Pu and ^{239}Pu) using deuteron beam with 2.52 GeV energy. Neutron spatial distributions were studied using activation threshold detectors.

Neutron distributions along the spallation source containing the Pb-target surrounded by U-blanket, were measured with SSNTDs. Comparison between measurements with particle detectors and fission detectors were performed. The spallation neutron source proved to provide high fluxes of fast neutrons very effectively, while the epithermal neutron flux is one order of magnitude less intense than flux of fast neutrons and thermal neutrons are negligible. The fast neutron distribution along the uranium blanket surface shows an increase in intensity up to about 15 cm after beam impact and then a decrease along the target.

For the first time in investigations at accelerator driven systems (ADC), methodical measurements with ^{3}He counters were done in order to determine the neutron spectra.

Natural uranium foils were used to determine the (n, γ)-reaction rates along the target length and radius. Spectral indices (capture to fission ratios) were also determined. Experimental results were compared with MCNPX simulations. Experimental and simulated values of spectral indices agree well. The total experimental value of the ^{239}Pu production in the whole $^{\rm nat}$U-blanket was estimated as $1.6(2) \cdot 10^{-8}$ g, the calculated value using the MCNPX code is $1.43 \cdot 10^{-8}$ g.

The measured experimental data complement our systematics obtained earlier using proton beams with energies between 0.7 and 2.0 GeV [5, 12–14, 16].

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REFERENCES

1. *M. I. Krivopustov, D. Chultem.* Experiments on Electronuclear Technology and Transmutation of Nuclear Waste Using Synchrophasotron Beams. JINR News. Dubna, 1998. V. 3. P. 27–30.

2. *M. I. Krivopustov, D. Chultem, Ts. Tumendelger et al.* Modeling of the Electronuclear Method of Energy Production and Study of Radioactive Waste Transmutation Using a Proton Beam of the JINR Synchrophasotron/Nuclotron. Research Program of the Laboratory of High Energies JINR /Ed. A. M. Baldin. JINR, 99–266. Dubna, 1999. P. 135–139;

   Book of Abstracts of the 3rd International Conference on Accelerator Driven Transmutation Technologies and Applications, June 7–11, 1999, Praha, Czech Republic. P. 55.

3. *M. I. Krivopustov, D. Chultem, Ts. Tumendelger et al.* On the First Experiment on Calorimetry of the Uranium Blanket of the Model U/Pb-Assembly of Electronuclear Installation «Energy plus Transmutation» from the JINR Synchrophasotron at Proton Energy 1.5 GeV. JINR Preprint P1-2000-168. Dubna, 2000; Kerntechnik. 2003. V. 68. P. 48–55.

4. *M. I. Krivopustov, D. Chultem, Tc. Tumendelger et al.* Experimental Studies of Electronuclear Method of Energy Production and Transmutation of Radioactive Wastes Using Relativistic Beams from JINR Synchrophasotron/Nuclotron // Proc. of the XV International Seminar on High Energy Physics Problems «Relativistic Nuclear Physics and Quantum Chromodynamics», Dubna, September, 2000. Dubna, 2001. V. 2. P. 3–21.

5. *M. I. Krivopustov, J. Adam, Yu. A. Batusov et al.* Investigation of Neutron Spectra and Transmutation of ^{129}I, ^{237}Np and Other Nuclides with 1.5 GeV Protons from the Nuclotron Using the Electronuclear «Energy plus Transmutation» Setup. JINR Preprint E1-2004-79. Dubna, 2004. P. 1–33; Nucl. Instrum. Meth. Phys. Res. A (submitted).

6. *M. Zamani, M. Manolopoulou, M. Fragopolou et al.* Neutron Generation in the New Transmutation Setup «Energy plus Transmutation» in Dubna // Nucl. Instrum. Meth. Phys. Res. A. 2003. V. 508. P. 454–459;

   S. Stoulos, M. Zamani, M. Manolopoulou et al. Application of Activation Method in Experimental Transmutation Installations in Dubna // Appl. Rad. Isot. 2003. V. 58. P. 169–175.

7. *I. V. Zhuk, M. K. Kievets, M. I. Krivopustov et al.* Investigation of Space and Energy Distribution of Neutrons in the Lead Target and the Uranium Blanket of the Electronuclear Installation «Energy plus Transmutation» During Irradiation with Proton at 1.5 GeV. JINR Communication P1-2002-84. Dubna, 2002; Vesti NAN Belarus. Ser. Phys. Math. Minsk, 2003. V. 12. P. 31–35;

   B. A. Marcynkevich, A. M. Khilmanovich, M. I. Krivopustov et al. Unfolding of Fast Neutron Spectra in the Wide Energy Range (up to 200 MeV) in a Heterogeneous Subcritical Assembly of an Electronuclear System «Energy plus Transmutation». JINR Preprint P1-2002-65. Dubna, 2002; Vesti NAN Belarus, Ser. Phys. Math. Minsk, 2004. V. 1. P. 90–95.

8. *S. R. Hashemi-Nezhad, R. Brandt, M. I. Krivopustov et al.* Experimental Studies and Monte-Carlo Computer Simulation of Space Distribution of Neutrons Generated in the Electronuclear System // Rad. Meas. 2003. V. 36. P. 295–300;

   S. R. Hashemi-Nezhad, I. V. Zhuk, M. I. Krivopustov et al. Studies of the Spatial Distribution and Nuclear Waste Transmutation Capabilities of the Spallation Neutrons at the JINR (Dubna) «Energy plus Transmutation» Setup // Proc. of the 15th Pacific Basin Nuclear Conference, 15PBNC, October 15–20, 2006, Sydney, Australia (in press).

9. *A. M. Baldin, A. I. Malakhov, A. N. Sissakian.* Problems of Relativistic Nuclear Physics and Multiple Particle Production. JINR Communication P1-2001-106. Dubna, 2001. P. 55–56; Phys. Part. and Nucl. 2001. V. 32, Suppl. 1. P. s4–s30;

   A. I. Malakhov, A. N. Sissakian, A. S. Sorin, S. Vokal. Relativistic Nuclear Physics at the Joint Institute for Nuclear Research. JINR Preprint P1-2006-93. Dubna, 2006. P. 60–61;

   A. D. Kovalenko, A. M. Baldin, A. I. Malakhov et al. Status of the Nuclotron // Proc. of the EPAC-94, June 1994, London, 1995. V. 1. P. 161–164; Cryogenic System of the Nuclotron — A New Superconducting Synchrotron // Adv. Cryogenic Eng. 1994. V. 39. P. 501–508; Main Results and Development Plans // J. At. En. 2002. V. 93. P. 479–485.

10. *M. I. Krivopustov, R. Brandt, M. Heck et al.* First Experiments on Transmutation Studies of ^{129}I and ^{237}Np Using Relativistic Protons of 3.7 GeV. JINR Preprint E1-97-59. Dubna, 1997; Radioanal. Nucl. Chem. 1997. V. 222. P. 267–271.

11. *R. Brandt, M. Heck, M. I. Krivopustov et al.* Transmutation of Radioactive Waste with the Help of Relativistic Heavy Ions. JINR Preprint E1-97-349. Dubna, 1997; Kerntechnik. 1998. V. 63. P. 167–172.

12. *J. Adam, K. Katovsky, A. R. Balabekian et al.* Transmutation of ^{129}I, ^{237}Np, ^{239}Pu and ^{241}Am Using Neutrons Produced in Pb-target with ^{nat}U-blanket System «Energy plus Transmutation» Bombarded by 2.0 GeV Relativistic Protons // Proc. of the International Conference on Nuclear Data for Science and Technology, September 26 – October 1, 2004, Santa Fe, New Mexico, USA. N. Y., 2005. V. 2. P. 1560–1563.

13. *M. I. Krivopustov et al.* Investigation of Neutron Transmutation of Radioactive Waste of Atomic Reactors (^{129}I, ^{237}Np, ^{238}Pu, ^{239}Pu and ^{241}Am) at the Setup «Energy plus Transmutation» with Proton and Deuteron Beams with Energies from 0.7 to 2.5 GeV of the JINR Synchrophasotron / Nuclotron (Dubna) // Proc. XIV International Seminar on Interaction of Neutrons with Nuclei, May 25–30, 2006, Dubna, Russia (in press).

14. *J. Adam, K. Katovsky, A. R. Balabekian et al.* Transmutation of ^{129}I, ^{237}Np, ^{238}Pu, ^{239}Pu and ^{241}Am Using Neutron Produced in Pb-target with ^{nat}U-blanket System «Energy plus Transmutation» by Relativistic Protons // Abstracts of 56th International Conference on Problems of Nuclear Spectroscopy and Structure of Atomic Nucleus, «Nucleus-2006», September 4–8, 2006, Sarov, Russia (in press).

15. *J. Adam, K. Katovsky, M. I. Krivopustov et al.* Interaction of Radioactive Nuclei ^{129}I, ^{237}Np, ^{238}Pu and ^{239}Pu and Stable ^{127}I with the Secondary Neutrons from the Pb-target with ^{nat}U-blanket System «Energy plus Transmutation» Irradiated by 2.52 GeV Deuterons // Abstracts of the 56th International Conference on Problems of Nuclear Spectroscopy and Structure of Atomic Nucleus, «Nucleus-2006», September 4–8, 2006, Sarov, Russia (in press).

16. *M. I. Krivopustov et al.* Investigation of the Neutron Field of Pb-target with ^{nat}U-Blanket and Radioactive Waste Transmutation of ^{129}I, ^{237}Np, ^{238}Pu, ^{239}Pu and ^{241}Am on Proton Beam with Energies from 0.7 to 2.0 GeV at Synchrophasotron // Book of Abstracts of the XVIII Intern. Baldin Seminar on High Energy Physics Problems «Relativistic Nuclear Physics and Quantum Chromodynamics», September 25–30, 2006, Dubna, Russia. JINR E1,2-2006-129. Dubna, 2006 (in press).

17. *M. I. Krivopustov et al.* About First Experiment at Deuteron Beam with Energy 2.52 GeV on Investigation of Transmutation of ^{129}I, ^{237}Np, ^{238}Pu and ^{238}Pu in the Field of Neutron Generated in Pb-target with ^{nat}U-Blanket // Book of Abstracts of the XVIII Intern. Baldin Seminar on High Energy Physics Problems. «Relativistic Nuclear Physics and Quantum Chromodynamics», September 25–30, 2006, Dubna, Russia. JINR E1,2-2006. Dubna, 2006 (in press).

18. *C. D. Bowman., E. D. Arthur, P. W. Lisowski et al.* Nuclear Energy Generation and Waste Transmutation Using an Accelerator-Driven Intense Thermal Neutron Source // Nucl. Instrum. Meth. Phys. Res. A. 1992. V. 320. P. 336–367.

19. *C. Rubbia, J. A. Rubio, S. Buano et al.* Conceptual Design of a Fast Neutron Operated High Power Energy Amplifier. Report CERN-AT-95-44. Geneva, 1995.

20. *H. Nifenecker, S. David, J. M. Loiseaux et al.* Hybrid Nuclear Reactors // Prog. Part. Nucl. Phys. 1999. V. 43. P. 683–827.

21. *C. Rubbia, J. Aleixandre, S. Andriamonje et al.* A European Roadmap for Developing Accelerator Driven Systems (ADS) for Nuclear Waste Incineration. ISBN 88-8286-008-6, ENEA, 2001; www.fzk.de/adsroadmap.html

22. *A. Abanades, J. Aleixandre, S. Andriamonje et al.* Results from the TARC Experiment: Spallation Neutron Phenomenology in Lead and Neutron-Driven Nuclear Transmutation by Adiabatic Resonance Crossing // Nucl. Instrum. Meth. Phys. Res. A. 2002. V. 478. P. 577–730; NIM. A. 2001. V. 463. P. 586–592.

23. *R. Brandt, M. I. Krivopustov, W. Westmeier et al.* Accelerator Driven Systems for Transmutation and Energy Production Challenges and Dangers // Kerntechnik. 2004. V. 69. P. 37–50.

24. *K. D. Tolstov, V. A. Voronko, V. Ya. Migalenya et al.* Generation Neutrons in Thick Lead Target // Atomic Energy. 1990. V. 68. P. 449–454; Energy Generation Neutrons Light Nuclei in Lead Slot. JINR Rapid Communication, 2(53), Dubna, 1992. P. 17–25.

25. *R. G. Vassil'kov, V. I. Gol'danskii, V. V. Orlov.* Electronuclear Method Generation Neutrons // Atomic Energy. 1970. V. 29. P. 151–187; About Electronuclear Briding // J. Succ. Phys. Sci. 1983. V. 139. P. 435–478; Neutron Yeild from Massive Lead Target Irradiation Relativistic Nucleus // At. Energy. 1995. V. 79. P. 257–264.

26. *S. Stoulos, M. Zamani, M. Manolopoulou et al.* Neutron Measurements by Passive Methods at the Dubna Transmutation Assemblies // Nucl. Instrum. Meth. Phys. Res. A. 2004. V. 519. P. 651–658.

27. *J. R. Harvey, R. J. Tanner, W. G. Arbert et al.* The Contribution of Eurados and Cendos to Etch Track Neutron Dosimetry: The Current Status in Europe // Rad. Protection. Dosimetry. 1998. V. 77. P. 267–304.

28. *M. Manolopoulou, M. Fragopoulou, M. Zamani et al.* Studies on the Response of ^{3}He and ^{4}He Proportional Counters to Monoenergetic Fast Neutrons // Nucl. Instrum. Meth. Phys. Res. A. 2006. V. 562. P. 371–379.

29. *J. Frána.* Program DEIMOS 32 for Gamma-Ray Spectra Evaluation // J. Rad. Nucl. Chem. 2003. V. 257. P. 583–587.

30. *J. Banaigs, J. Berger, J. Duplo et al.* // Nucl. Instrum. Meth. A. 1971. V. 95. P. 307.

31. *Yu. E. Titarenko, O. V. Shvedov, V. F. Batyaev et al.* Nuclei Product Yields in ^{99}Tc Transmutation under 0.1–2.6 GeV Proton Bombardment // Proc. of the Intern. Conf. on Subcritical Accelelerator Driven Systems, October 11–15, 1999, Moscow, Russia. Moscow, 1999. P. 204–211;

    S. E. Chigrinov, I. V. Rachno, A. I. Kievitskaya et al. Monte-Carlo Calculation of Iodine-129 and Neptunium-237 Transmutation with Relativistic Protons // Proc. of the Intern. Conf. on Accelerator Driven Transmutation Technologies and Applications, June 7–11, 1999, Praha, Czech Republic.

32. *M. Bielevich, M. Szuta, E. Strugalska-Gola et al.* On the Experiment of Neutron Spectrum Investigation on U/Pb-Assembly Using 0.7 GeV Proton Beam from the Nuclotron (Dubna) // Proc. of XVII International Baldin Seminar on High Energy Physics Problems «Relativistic Nuclear Physics and Quantum Chromodynamics», September 27 – October 2, 2004, Dubna, Russia. Dubna, 2005. V. II. P. 125–132.

33. *F. Krízek, V. Wagner, J. Adam et al.* The Study of Spallation Reactions, Neutron Production and Transport in a Thick Lead Target and a Uranium Blanket During 1.5 GeV Proton Irradiation // Czech. Journ. Phys. 2006. V. 56. P. 243–252.

34. *M. Zamani, M. Manolopoulou, M. Fragopolou et al.* A Spallation Neutron Source Based on Pb-Target Surrounded by U-Blanket. Radiation Measurement // Proc. of the 23rd Intern. Conf. SSNTD, September 11–15, 2006, Beijing, China (in press).

35. *M. Zamani, M. Manolopoulou, M. Fragopolou et al.* Spallation Neutron Production at the New Dubna Transmutation Assembly // Nucl. Instrum. Meth. Phys. Res. A. 2003. V. 508. P. 454–459.

36. *V. M. Gorbachev, Yu. S. Zamyatin, A. A. Lbov*. Interaction Ray with Heavy Element and Fission Nuclei. M.: Atomizdat, 1978.

37. *L. W. Hendricks, G. W. McKinney, L. S. Waters et al.* MCNPX, VERSION 2.5.e. Report No. LA-UR-04-0569, Los Alamos National Laboratory, USA, 2004.

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