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Determination of the ratios of the average ^{239}Pu and ^{237}Np fission cross-sections to the average ^{235}U fission cross-section in QUINTA neutron field

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The ratios of the average ^{239}Pu and ^{237}Np fission cross-sections to the average ^{235}U fission cross-section were determined experimentally. Experiments with U-assembly QUINTA were carried out using the accelerator complex "Nuclotron" of the Veksler and Baldin Laboratory of High Energy Physics (VBHEP) of the Joint Institute for Nuclear Research (JINR) (Dubna, Russia). Experiments were conducted in the frame of the project "E&T RAW". It is based on so called Relativistic Nuclear Technology (RNT) proposed recently [1] by one of the institutions (CPTP "Atomenergomash", Moscow) participating in "E&T RAW" collaboration. The assembly was irradiated by deuterons and carbon ions with energies 2 and 4 GeV/A. Comparison between experimental results and MCNPX calculations are presented. Accumulation and burning of plutonium, neptunium and others isotopes influences the neutron balance of the assembly that attracts an interest in such measurements.

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1. Introduction

The problem of effective disposal of spent nuclear fuel has become a key in discussing the future of the global energy industry in recent years. The world powers have begun to consider seriously the use of accelerator driven systems (Accelerator Driven Systems - ADS in international terms) as an alternative and promising method to solve this problem.

Analysis of the various areas of nuclear power shows limitations of the capabilities of traditional reactors and classic ADS in addressing global energy challenges. In the fission neutron spectrum, the threshold minor actinide burning is ineffective because of their high threshold ($\approx$ 1 MeV). Transmutation of long-lived radioactive waste from the spent fuel is closed very bad due to multistep reactions that lead to the emergence of new long-lived radioactive isotopes. Thus it becomes clear that the only real prospect of radical solutions to the problems of modern nuclear power is the use of more hard than fission's, the neutron spectrum [2], [3].

For practical realization of this path principally new scheme of electronuclear method, based on the relativistic nuclear technologies (RNT) has been worked out.

RNT are the basis of a fundamentally new scheme electronuclear method for energy production and transmutation of radioactive waste. This scheme is aimed at creation extremely hard neutron spectrum inside of the multiplying system. It is expected that such a spectrum would permit to "burn" for energy production natural (depleted) uranium or thorium, and simultaneously utilize the long-lived components of spent nuclear fuel of nuclear power plants.

Due to the features of material composition of the experimental assembly and assembly neutron field it is necessary to determine the ratios of the average ^{239}Pu and ^{237}Np fission cross-sections to the average ^{235}U fission cross-section to determine the long-lived radioactive isotopes transmutation cross sections.

2. Target assembly description

The target assembly QUINTA (Fig. 1) consists of five identical sections (Fig. 2) of hexagonal aluminum containers with an inscribed diameter of 284 mm, each of which is placed for 61 cylindrical uranium block.

Blocks of 36 mm diameter and a length of 104 mm made of metallic natural uranium and placed in sealed aluminum housing (thickness 1.1 mm). Unit weight is 1.72 kg and the total mass of uranium in one section is 104.92 kg. The front section has the cylindrical input beam channel diameter 8 cm. The total mass of uranium in the target assembly is about 500 kg. Connected to each other sections are fixed on aluminum plate and placed on a mobile carriage allows for precise adjustment of the axis of the target relative to the direction of the incident beam.

The lead blanket with thickness of 10 cm with the input beam window (150 $\times$ 150 mm) surrounds QUINTA.

The axis of the setup was aligned with beam axis with the help of the adjustable stand under the whole setup. The alignment of the beam center with the center of the setup was achieved by examining polaroid films placed in front of the target and exposed to a couple of incident particles pulses prior to the installation of the sample plates and the start of the main irradiation.

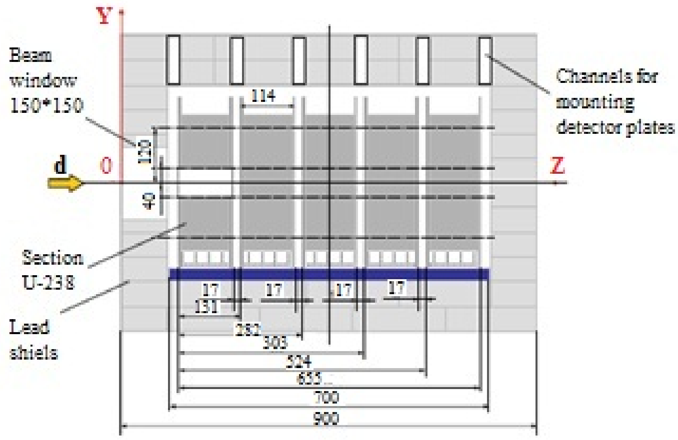

Figure 1: The target assembly QUINTA equipped by lead blanket..

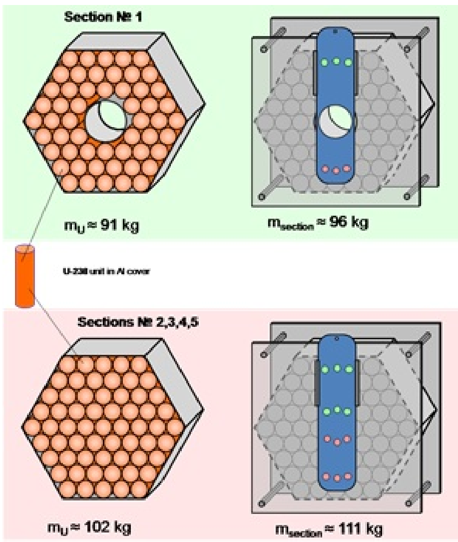

Figure 2: The sections of QUINTA.

2.1 Experimental details

QUINTA was irradiated by deuterons and carbon ions with energies 2 and 4 GeV/A. The beams were produced by superconducting, strong focusing synchrotron Nuclotron.

The extracted pulsed incident particles beam of the NUCLOTRON hits the target from the exit window located on it in 3.32 m. The spatial and time profile, as well as an intensity of each incident particles pulse were monitored using calibrated and position sensitive ionization chambers in coincidence with two scintillation telescopes. The beam position on the target and the integral flux were controlled by the profilometer and activation monitors from aluminum foil and solid state nuclear track detectors placed before the target.

The main objectives of the experiments were:

- Determination of the ratios of the average ^{239}Pu and ^{237}Np fission cross-sections to the average ^{235}U fission cross-section.

- Comparison between experimental results and calculations.

3. Experimental technique

The method of the SSNTD was chosen for the determination of the ratios of the average ^{239}Pu and ^{237}Np fission cross-sections to the average ^{235}U fission cross-section.

This technique was developed by I. Zhuk and A. Malikhin. It was applied for fission reactions rates measurements in reactor systems [4], [5].

SSNTD-sensors consist of two parts: of the heavy metal that interacts with incident particles via nuclear fission (irradiator) and of the material in which fission fragments leave tracks (track detector). The detectors material is artificial mica (Fluorophlogopite) (Fig. 3). Sensors were placed on backplate of left side on lead blanket of QUINTA. The procedure for SSNTD-sensors calibration is described in [6].

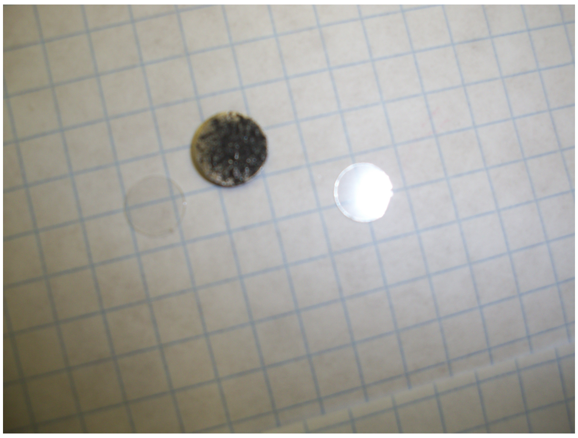

Figure 3: The sensor.

The procedure of the determination of the ratios of the average fissionable nuclides cross-sections to the average ^{235}U fission cross-sections $\frac{\overline{\sigma}^i_f}{\overline{\sigma}^{25}_f}$ is based on correlation between the track density on a track detector N_i irradiated in contact with target (it consist of i - fissionable nuclide), and energy neutron flux density of investigated neutron field φ_E:

$$N^i = A^i_k\, \mu^i\, \varepsilon^i\, t\, d^i\, \rho^i \int_0^\infty \sigma^i_k(E)\varphi(E)\, dE \tag{3.1}$$

where A^i_k -number of charged particles produced in k -reaction of i -nuclide; μ^i -the fraction of charged particles reaching the detector; ε -detection efficiency of the charged particle track detectors; t -duration of sensors the exposure, sec; d^i -the radiator thickness, cm; ρ^i -nuclear density of i -nuclide in the radiator, nuclei/cm^3; $\sigma^i_k(E)$ -microscopic cross-section of k -reaction on i -nuclide.

Figure 4: The placement of sample.

The average cross-section of k -reaction on i -nuclide is equal to

$$\overline{\sigma}_k^i = \frac{\int\limits_0^\infty \sigma_k^i(E)\varphi(E)\,dE}{\int\limits_0^\infty \varphi(E)\,dE} \tag{3.2}$$

Then for $\frac{\overline{\sigma}_f^i}{\overline{\sigma}_f^{25}}$:

$$\frac{\overline{\sigma}_f^i}{\overline{\sigma}_f^{25}} = \frac{\int\limits_0^\infty \sigma_f^i(E)\varphi(E)\,dE}{\int\limits_0^\infty \sigma_f^{25}(E)\varphi(E)\,dE} \tag{3.3}$$

Absolute and relative methods are used for the determination of the ratios of the average cross-sections. Equations (3.1) and (3.3) are used for absolute methods. But absolute methods have an additional uncertainties from determination of ε, ρ, d uncertainties.

In relative methods ε, ρ, d are determined from calibration measurements in standart neutron field.

So equation for the ratio of average fissionable nuclides cross-sections is

$$\frac{\overline{\sigma}_f^i}{\overline{\sigma}_f^{25}} = \frac{Nm^i}{Nm^{25}}\frac{Ns^i}{Ns^{25}}\frac{(\sigma_f^i)_s}{(\sigma_f^{25})_s} \tag{3.4}$$

where m and s are the results from investigated and standart neutron fields.

We denoted $b = \frac{Nm^i}{Nm^{25}}$ and $a = \frac{Ns^i}{Ns^{25}}$. Then we obtain following ratio:

$$\frac{\overline{\sigma}_f^i}{\overline{\sigma}_f^{25}} = \frac{b}{a}\frac{(\sigma_f^i)_s}{(\sigma_f^{25})_s} \tag{3.5}$$

Next we need doing necessary corrections. More information about corrections in [4].

For calculation of the ratio of the average ^{239}Pu fission cross-section to the average ^{235}U fission cross-section the nextformula is used:

$$\frac{\overline{\sigma}_f^{49}}{\overline{\sigma}_f^{25}} = k\frac{b}{a}\left(\frac{\sigma_{fo}^{49}}{\sigma_{fo}^{25}}\frac{g_{fo}^{49}}{g_{fo}^{25}} + \sum_i \frac{\sigma_{f0}^{i}}{\sigma_{f0}^{25}}\frac{g_{fo}^{i}}{g_{fo}^{25}}\frac{\chi_{49o}^{i}}{\chi_{49o}^{49}}\right)\left(1+\sum_{imp}\frac{\overline{\sigma}_f^{imp}}{\overline{\sigma}_f^{25}}\frac{\chi_{25}^{imp}}{\chi_{25}^{25}}\right)_m e^{-\lambda_{49}(t_o - t_m)} -$$

$$-\sum_i \frac{\overline{\sigma}_f^{i}}{\overline{\sigma}_f^{25}}\frac{\chi_i^{9m}}{\chi_9^{9m}} \tag{3.6}$$

where b and a -ratios of the track density from plutonium target to the track density from uranium target at the irradiation by investigated beam and standard neutron field respectively; i -impurities in the target; σ_{fo}^{i} -^{239}Pu fission cross-sectionby neutrons with υ=2200 m/s; g_{fo}^{49} -Westcott factor for ^{239}Pu at T= 293 K; χ_{49}^{i} -the fractions of i -nuclide at the investigated target; m-index and o-index means that we consider the nuclide content in the target at the exposure time at the investigated beam and standatr neutrons field respectively; λ_{49} - ^{239}Pu decay constant; t_o and t_m -time between making the target and the target irradiation in the investigated beam and standart neutrons field respectively; k -correction that includes self-shielding in the targets.

For calculation of the ratio of the average ^{237}Np and ^{232}Th fission cross-section to the average ^{235}U fission cross-section the next formula is used:

$$\frac{\overline{\sigma}_f^{i}}{\overline{\sigma}_f^{25}} = \frac{b}{a}\frac{(\sigma_f^{i})_s}{(\sigma_f^{25})_s}\frac{\left(1+\frac{\overline{\sigma}_f^{24}}{\overline{\sigma}_f^{25}}\frac{\chi_{25}^{24}}{\chi_{25}^{25}}+\frac{\overline{\sigma}_f^{28}}{\overline{\sigma}_f^{25}}\frac{\chi_{25}^{28}}{\chi_{25}^{25}}\right)_m}{\left(1+\frac{\sigma_f^{24}}{\sigma_f^{25}}\frac{\chi_{25}^{24}}{\chi_{25}^{25}}+\frac{\sigma_f^{28}}{\sigma_f^{25}}\frac{\chi_{25}^{28}}{\chi_{25}^{25}}\right)_m} \tag{3.7}$$

where i -we refer to ^{237}Np or ^{232}Th; other see for (3.6) and (3.8).

For calculation of the ratio of the average ^{238}U fission cross-section to the average ^{235}U fission cross-section the next formula is used:

$$\frac{\overline{\sigma}_f^{28}}{\overline{\sigma}_f^{25}} = \frac{\left(1+\frac{\chi_i^{24}}{\chi_i^{25}}\frac{\overline{\sigma}_f^{24}}{\overline{\sigma}_f^{25}}\right)_m - k\frac{b}{a}\left(1+\frac{\chi_j^{24}}{\chi_j^{25}}\frac{\overline{\sigma}_f^{24}}{\overline{\sigma}_f^{25}}+\frac{\chi_j^{26}}{\chi_j^{25}}\frac{\overline{\sigma}_f^{26}}{\overline{\sigma}_f^{25}}\right)_m}{k\frac{b}{a}\frac{\chi_j^{28}}{\chi_j^{25}} - \frac{\chi_j^{28}}{\chi_j^{25}}} \tag{3.8}$$

where $\left(\frac{\overline{\sigma}_f^{24}}{\overline{\sigma}_f^{25}}\right)_m$, $\left(\frac{\overline{\sigma}_f^{26}}{\overline{\sigma}_f^{25}}\right)_m$ -the ratio of average ^{234}U and ^{236}U fission cross-sections to the average ^{235}U fission cross-section at the investigated neutron field respectively; χ_j^{κ} and χ_i^{κ} -the fractions of κ -nuclides; k -correction that includes self-shielding in the targets.

4. Results and discussion

This part includes measured results of the ratios of the average ^{239}Pu, ^{237}Np, ^{232}Th and ^{238}U fission cross-sections to the average ^{235}U fission cross-section. Moreover, comparison of the experimental data with results of calculations are presented.

Table 1 presents experimental data from the track detectors, which were irradiated in contact with investigated targets. The values of a taken from [4], [5].

Experimental results for the ratios of the average ^{239}Pu, ^{237}Np, ^{232}Th and ^{238}U fission cross-sections to the average ^{235}U fission cross-section (Table 2) were obtained from the formulas (3.6 — 3.8).

Table 1: Experimental data from the track detectors.

	d, 2 GeV/A		d, 4 GeV/A		^{12}C, 2 GeV/A		^{12}C, 4 GeV/A	
Nuclide	Track density, tr./ cm^2	*b*	Track density, tr./ cm^2	*b*	Track density, tr./ cm^2	*b*	Track density, tr./ cm^2	*b*
6.5 U	2,87·10^6	-	1,4·10^6	-	1,29·10^5	-	6,7·10^4	-
^{nat}U	1,34·10^6	0,47	6,4·10^6	0,46	6,0·10^4	0,46	3,2·10^4	0,47
^{239}Pu	2,14·10^6	0,74	1,06·10^6	0,75	1,0·10^5	0,74	4,5·10^4	0,68
^{237}Np	1,99·10^6	0,69	1,0·10^6	0,71	8,7·10^4	0,67	4,6·10^4	0,69
^{232}Th	3,8·10^5	0,13	1,83·10^5	0,13	1,8·10^4	0,14	8,6·10^3	0,13

Table 2: The ratios of the average ^{239}Pu, ^{237}Np, ^{232}Th and ^{238}U fission cross-sections to the average ^{235}U fission cross-section.

	d, 2 GeV/A		d, 4 GeV/A		^{12}C, 2 GeV/A		^{12}C, 4 GeV/A	
	Exp	Calc	Exp	Calc	Exp	Calc	Exp	Calc
^{238}U/ ^{235}U	0,041 ±0,010	0,040	0,040 ±0,010	0,048	0,041 ±0,010	0,051	0,042 ±0,010	0,055
^{239}Pu/ ^{235}U	1,17 ±0,14	1,17	1,17 ±0,14	1,17	1,16 ±0,14	1,17	1,07 ±0,13	1,18
^{237}Np/ ^{235}U	0,35 ±0,04	0,38	0,35 ±0,04	0,38	0,34 ±0,04	0,39	0,35 ±0,04	0,41
^{232}Th/ ^{235}U	0,0119 ±0,0014	0,0123	0,0116 ±0,0014	0,0123	0,0126 ±0,0015	0,0133	0,0117 ±0,0014	0,0143

The calculations performed with the computer code MCNPX 2.7e with energy neutron data library ENDF70, JANIS 4.0 database, models ISABEL, INCL4/ABLA, CEM2K.

Measurement uncertainties are estimated by the international standard ISO/IEC 17025: 1999.

The experimental data and calculation are in good agreement.

The ratio of average fissionable nuclides cross-sections does not depend on type of incident particles and their energies.

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