EXPERIMENTAL DETERMINATION OF HEAVY NUCLEI FISSION CROSS-SECTIONS UNDER RELATIVISTIC DEUTERONS IRRADIATION ON THE ACCELERATOR COMPLEX “NUCLOTRON” FOR PURPOSES OF TRANSMUTATION AND ENERGY AMPLIFICATION

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Abstract

Experimental studies of neutron spectra of three different subcritical assemblies driven by an accelerator (Accelerator Driven Systems – ADS) for investigation of the possibility of transmutation and energy amplification have been carried out. The assemblies were constructed in the framework of the international project “Energy and Transmutation of Radioactive Wastes” and the experiments with them are running in the Veksler and Baldin Laboratory of High Energy Physics of the Joint Institute for Nuclear Research (Dubna, Russia) at the accelerator complex “Nuclotron”. In this paper the results of measurements of ^{239}Pu(n,f), ^{235}U(n,f), ^{238}U(n,f) and ^{238}U(n,γ) reactions cross-sections and reactions rates using solid state nuclear track detectors and activation gamma-spectroscopy are presented. A comparison of the experimental results with FLUKA calculations is given. The obtained experimental values characterize the neutron spectra in the experimental points and allow the efficiency of the ADS technology for the systems with similar parameters to be evaluated.

INTRODUCTION

One of the greatest obstacles facing nuclear energy is how to properly handle the highly radioactive waste which is generated during irradiation in reactors. In the past years more and more studies have been carried out on advanced waste management strategy. An innovative concept of a hybrid system for transmutation of long-lived radioisotopes, i.e. the combination of a subcritical nuclear reactor with a high energy particle accelerator, has been suggested. Such systems can generate electricity converting transuranium waste [1].

The perspective ADS facility is supposed to consist of the neutron generating heavy metal (Pb, Bi, ^{nat}U) target surrounded by blanket of fissile material. The main element of nuclear fuel for ADS is ^{238}U in composition of natural or enriched uranium.

To develop such facility it is necessary to get neutron cross-sections data, neutron and γ-quantum multiplicity data in a wide range of energy from 20 MeV to 2 GeV. However there is lack of data and experimental results in this range of energy [2].

This paper presents the results of experiments which were carried out on different ADS driven by the superconducting strong focusing accelerator “Nuclotron”. Cross-sections and rates of ^{239}Pu(n,f), ^{235}U(n,f), ^{238}U(n,f), ^{238}U(n,γ) reactions were obtained using solid state nuclear track detectors and activation gamma-spectroscopy.

A comparison of the experimental results with FLUKA calculations is given.

EXPERIMENTAL INSTALLATIONS

Within the “E&T - RAW” collaboration a number of target systems of different geometries and composition were developed and irradiated. In this paper the results of investigation of three of them are presented.

- ***Lead/graphite-assembly “Gamma-MD”*** is a lead neutron generating target with 8 cm in diameter and 60 cm long surrounded by graphite. “Gamma-MD” is illustrated in Fig. 1.

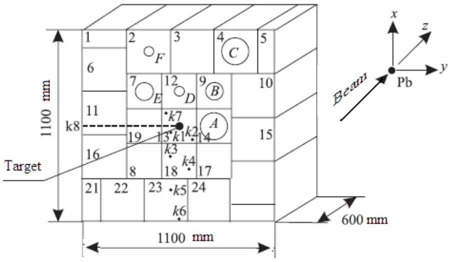

Figure 1: Working area of “Gamma-MD” setup.

- ***Uranium/Lead-assembly “Energy plus Transmutation”*** (“E+T”) is illustrated in Fig. 2.

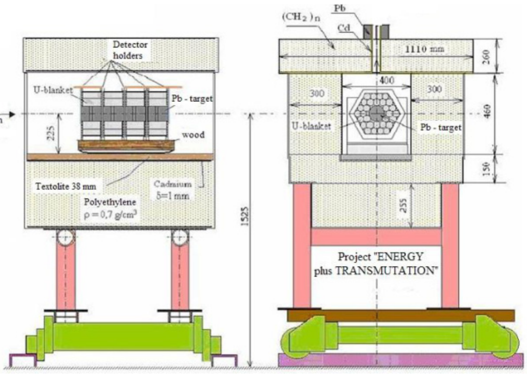

Figure 2: Cross-section and front view of “E+T” setup.

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"E+T" setup consists of a cylindrical lead target (diameter 84 mm, total length 480 mm) and a surrounding subcritical uranium blanket (206.4 kg of natural uranium). Target and blanket are divided into four sections. Between the sections there are 8 mm gaps for samples and detectors.

- ***Massive uranium/Lead shielding assembly "Quinta"*** consists of five identical sections of hexagonal aluminium containers with an inner diameter of 284 mm, each of which is filled with 61 cylindrical metallic natural uranium blocks of 36 mm diameter and a length of 104 mm aluminium cover. The front section has the cylindrical input beam channel of 8 cm in diameter. The total mass of uranium in the target assembly is about 500 kg. Between the sections there are 6 experimental plates for detectors and samples. "Quinta" setup is illustrated in Fig. 3.

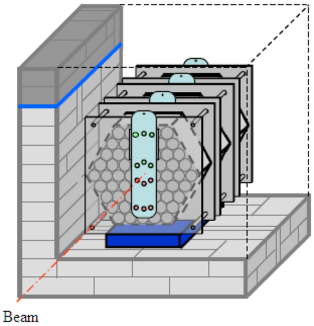

Figure 3: Schema of "Quinta" setup.

"Gamma-MD" and "E+T" setups were irradiated by deuterons with energy of 2.3 GeV, "Quinta" setup – with energy of 2 GeV.

EXPERIMENTAL TECHNIQUES

To determine reactions cross-sections and reactions rates we used solid state nuclear track detectors (SSNTD) and activation gamma-spectroscopy.

SSNTD technique is based on correlation of tracks density on irradiated detectors and flux density of investigated neutron field. After irradiation of track detector in contact with fission foil (source of fission fragments) the damaged zone which is called a latent track is formed on the track detector surface. Latent tracks are visible in optical microscope after chemical etching.

The track density at external surface of a sensor (track detector plus fissionable nuclei) is given by the following equation [3]:

$$\rho = n \cdot \mu \cdot \varepsilon \cdot d \cdot N_v \cdot t \int_0^\infty \sigma_f(E)\varphi(E)dE \qquad (1)$$

where N_v is number of fissionable nuclei per unit volume of the foil, $\sigma_f(E)$ and $\varphi(E)$ are the energy dependent fission cross-section and projectile flux respectively, t is the irradiation time, n is number of the fragments emitted per fission, d is the thickness of the foil, ε is an efficiency factor which include the critical angle effect as well as the limitations imposed by the minimum detectable track size and track observation conditions, μ accounts for different foil thicknesses.

Equation for fission cross section determination is derived from Eq. 1:

$$\sigma = \rho / P \cdot w \qquad (2)$$

where P is the number of primaries, $w = n \cdot \mu \cdot \varepsilon \cdot d \cdot N_v$ is calibration factor.

The procedure of calculation and experiments on calibration are well described in the paper [4].

The number of primaries (or total intensity) is determined by activation gamma-spectroscopy using activation Al foils via ^{27}Al(d,3p2n) ^{24}Na reaction.

We used artificial mica (Fluoroflogopite) as a track detector for fission fragments registration and heavy metals (^{239}Pu, ^{235}U, ^{238}U) as the fissionable nuclei. After the exposure the detectors are etched in 7 % hydrofluoric acid (HF) at 60°C. The duration of the etching time is decided on the basis of the track population in a given sample, in order to minimize the overlapping of the track openings.

EXPERIMENTAL RESULTS

The cross-sections for calculation of ^{239}Pu(n,f) reaction rate were taken from ENDF-B database and FLUKA [5,6] library for energy range from 0 to 20 MeV and from EXFOR database for energies above 30 MeV.

To compare the results the experimental samples were located in the same positions in "E+T" and "Quinta" setups (point 1: z = 12 cm, x=0 cm, y = 8,5 cm; point 2: z = 12 cm, x=0 cm, y = 12 cm), in "Gamma-MD" setup – in position z= 16 cm, x=y=9,2 cm.

For simulation of the process of interaction of projectiles with the assemblies' material the FLUKA code was used.

Table 1 presents the experimental and simulation results in the investigated point 1 and point 2 respectively.

Table 1: ^{239}Pu(n,f) Cross-Section and Fission Rates

Setup	**Deuteron energy, GeV**	**Neutron flux, 1/cm^2·d**	**Average cross-sections, barn**	**^{239}Pu(n,f) fission rates**	
				Calculation	**Experiment**
Point 1					
Quinta	2	0,2198	1,6129± 0,0083	0,3546± 0,0018	0,34± 0,04
E+T	2,3	0,1154	4,9430± 0,0845	0,5702± 0,0097	0,55± 0,07
Gamma-MD	2,3	0,1140	125,16± 0,0354	14,272± 0,0040	13,0± 1,6
Point 2					
Quinta	2	0,1153	1,6121± 0,0089	0,1859± 0,0010	0,19± 0,02
E+T	2,3	0,0534	19,4068± 0,0563	1,0369± 0,0030	1,14± 0,14

It is clear that due to neutron generation in the uranium of “Quinta” setup the neutron flux is higher here.

The average plutonium cross-section is minimal in the uranium setup due to hard spectrum and maximal in lead/graphite setup because of thermal neutron spectrum.

In the “E+T” setup average ^{239}Pu cross-section increase higher than four times in radial direction that is explained by reflection from the biological shielding.

The calculation is converged with the experimental result in the range of ±20%. The main error of experimental result is projectile particle flux determination error.

Neutron spectra of investigated setups in point 1 and point 2 are shown on Fig. 4.

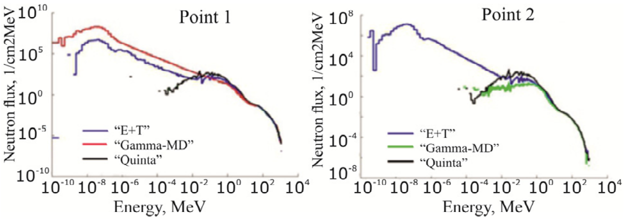

Figure 4: Neutron spectra in point 1 and point 2.

In the range of energy up to 2 MeV in the “E+T” and “Quinta” spectra dominate the fission neutrons. For these two setups there are no neutrons with energy bellow 10 eV in their spectra due to absence of moderation. While the spectrum of “Gamma-MD” setup includes considerable part of the thermal and epithermal neutrons.

In Fig. 5 the ^{239}Pu(n,f), ^{235}U(n,f), ^{238}U(n,f) and ^{238}U(n,γ) reactions cross-sections are presented.

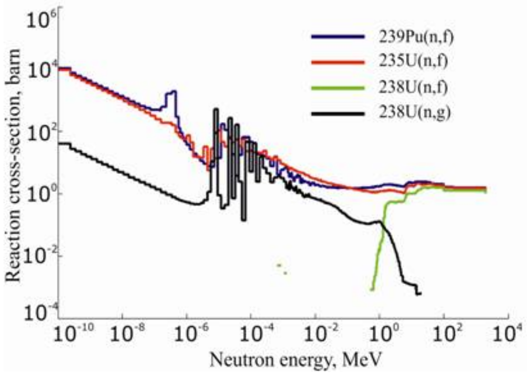

Figure 5: Reaction cross-sections.

It is noticeable that the cross-sections of ^{239}Pu(n,f), ^{235}U(n,f) are practically the same. The difference in the epithermal region is explained by plutonium resonance at this energy.

Fission of ^{238}U is a threshold reaction with the threshold of 1 MeV and fission rate of this reaction defines the fast neutron distribution in the setup volume.

^{238}U(n,f) and ^{238}U(n,γ) reactions characterize the main processes in the subcritical facilities: fission and radiation capture.

The rates of ^{239}Pu accumulation in “Quinta” and “E+T” setups are shown in Fig. 6.

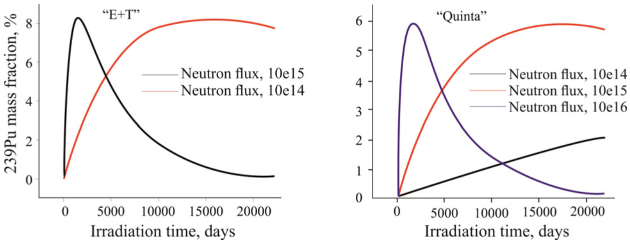

Figure 6: ^{239}Pu accumulation.

It is shown that mass fraction maximal value of plutonium accumulation depends on cross-section and does not depend on neutron flux.

^{239}Pu is accumulated non-uniformly in the blanket volume even in case of constant spectrum inside. In the target area the maximum can be reached quite quickly that can results in formation of areas with different subcriticality. This aspect should be taking in account in ADS development.

CONCLUSION

Reactions cross-sections and reactions rates of ^{239}Pu(n,f), ^{235}U(n,f), ^{238}U(n,f) and ^{238}U(n,γ) reactions for different subcritical assemblies were determined using solid state nuclear track detectors and activation gamma-spectroscopy. Comparison was made between the results of the measurements and results of Monte Carlo simulations using FLUKA code. The obtained experimental values characterize the neutron spectra in the experimental points and allow the efficiency of the ADS technology for the systems with similar parameters to be evaluated.

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