COMPARISON OF NEUTRON-PHYSICAL CHARACTERISTICS OF URANIUM TARGET OF ASSEMBLY "QUINTA" IRRADIATED BY RELATIVISTIC DEUTERONS AND ^{12}C NUCLEI

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At the present time disposal of spent nuclear fuel and fuel supply problem are two main reasons preventing wide distribution of nuclear power. One of the ways to solve this problem is using Nuclear Relativistic Technologies aimed at forming of maximum hard neutron spectrum in natural or depleted massive uranium targets irradiated by high energy (2…10 GeV) beams of relativistic particles. This paper describes the neutron generation in massive natural uranium target (assembly "QUINTA", $m_U \sim 500$ kg) irradiated by beams of relativistic deuterons and ^{12}C ions with energies of 2 and 4 AGeV at the accelerator Nuclotron (JINR, Dubna). The reactions ^{nat}U(n,f), ^{238}U(n,γ), and ^{59}Co(n,x) were investigated using activation technique. Comparison of obtained experimental results in dependence on energy of incident beam and type of particles was carried out.

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INTRODUCTION

The increased in the past two decades interest in the study of subcritical accelerator-driven systems (ADS) for the purpose of using them to solve applied problems led to theoretical and experimental studies in this field in the world's leading nuclear centers. At the present time the powerful multipurpose accelerator centers such as SNS, USA (~1.3 MW power) [1], J-PARC, Japan (~1 MW) [2], the PSI, Switzerland (~0.75 MW) [3] have been operated. They have different departments in structure that study materials, transmutation of radioactive waste, medical radioisotopes production, etc. These centers use proton accelerators with energies of about 1 GeV (or less) for neutron generation. Meanwhile, a number of experiments have studied neutron production in thick targets (JINR, LANL, KEK, and ITEP) and found that more energy accelerators should be used for more effective neutron generation. In particular, the group of V. Yurevich (JINR, Dubna) performed the cycle of experimental studies [4, 5] and analysis of available experimental data of neutron energy spectra that produced in thick targets. It was shown that for thick targets secondary nuclear interactions in the target give additional contribution to neutron emission. Neutron multiplication with simultaneous weakening of the charged particle release makes thick target, especially as neutron sources. The ratio of total energy expended on the neutron formation to particle beam energy shows weak growth with beam energy and does not depend on type of particle primary beam. The effect of average neutron energy increasing per unit of beam energy with increasing of beam energy has also been noted. That could be used for further neutron multiplication when using a quasi-infinite target. In this context, a more efficient use of beam energy for neutron production will significantly increase the value of optimum beam particle energy.

During the last 5 years at JINR in the framework of collaboration "Energy and Transmutation of RAW" the nuclear physical characteristics of neutron fields generated in massive uranium target irradiated by deuterons with energy 1…8 GeV have been studied [6]. One of the main collaboration objectives is to study of dependence of neutron generation in the uranium target on primary beam energy. This paper describes experiments on the irradiation of uranium assembly, surrounded by the lead blanket (assembly "QUINTA") by deuteron and ^{12}C nuclei beams with energies 2 and 4 AGeV at the "Nuclotron" accelerator of JINR.

The purposes of this work are:

1. To perform the monitoring of deuteron and ^{12}C nuclei beams.
2. Using the activation technique to obtain the spatial distribution of density of radiative capture reactions ^{238}U(n,γ), fission reactions ^{nat}U(n,f), ^{59}Co(n,x) reactions, spectral indices in the volume of uranium target, as well as the total number of capture and fission reactions.
3. To compare the experimental results depending on beam energy and type of accelerated particles.

1. EXPERIMENT AND METHODS

The experimental setup "QUINTA" [6, 7] consists of uranium target (^{nat}U m = 512 kg), surrounded by lead blanket with thickness of 10 cm. Uranium target contains five sections. Before irradiation detector plates with sets of activation and track detectors were placed in the gaps between sections, as well at the front and rear end of the target. At the entrance of beam in the assembly the lead shielding has beam input window with size of 150×150 mm. Beside this first section also has input window with diameter of 80 mm. Natural uranium activation detectors with thickness of 1 mm and diameter of 8 mm were used in the experiment to obtain spatial distribution of ^{nat}U fission reactions and neutron capture reactions on ^{238}U. Activation detectors were placed on six detector plates (Z = 0, 123, 254, 385, 516, 647 mm). Each plate except the first one contained 5 detectors at following distances from center R = 0, -40, -80, -120, +80 mm.

Gamma spectra of irradiated samples were measured with HPGe detector. Energy and efficiency calibration of the detectors was performed using a set of conventional radioactive sources (^{54}Mn, ^{57}Co, ^{60}Co, ^{88}Y, ^{133}Ba, ^{137}Cs, ^{139}Ce, ^{152}Eu, ^{228}Th).

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

$$^{238}U(n,\gamma)^{239}U\ \beta^- \rightarrow {}^{239}Np\ \beta^- \rightarrow {}^{239}Pu.$$

The number of fissions was determined by yield of gamma-lines 743.36, 364.49, 529.9, and 293.3 keV of fission fragments ^{97}Zr, ^{131}I, ^{133}I and ^{143}Ce respectively. Cumulative yields (CY) of these fragments remain approximately constant in a wide range of neutron energies from the fission-spectrum neutrons to neutrons with energy 22 MeV [12]. We used the next values of CY: ^{97}Zr – 5.7%, ^{131}I – 3.6%, ^{133}I – 6.3%, ^{143}Ce – 4.3%.

Additionally ^{59}Co activation detectors ~ 3 mm thick and ~ 15 mm diameter were used to study neutron spectrum characteristics. Six ^{59}Co detectors, that were placed one on each plate at the distance R = +40 mm from center of the target, were used in each irradiation. It should be noted that products of threshold reactions $^{59}Co(n,x)$ with E_{th} from ~1 to ~100 MeV were observed in the measured γ-spectra of cobalt detectors. Obtained spatial distributions of reaction density were used to analyze the neutron spectrum produced in the uranium target of assembly "QUINTA".

Monitoring of deuteron and ^{12}C nuclei beams was carried out by activation of aluminum and copper foils in the reactions ^{27}Al(d,x)^{24}Na, ^{27}Al(^{12}C,x)^{24}Na, ^{64}Cu(d,x)^{24}Na and ^{64}Cu(^{12}C,x)^{24}Na. Cross sections of these reactions for given beam energy were chosen by averaging and interpolation of known experimental values [8 - 10]. Techniques of beam monitoring has described in detail in paper [8]. Obtained total intensities of incident particles and used cross sections are shown in Table 1.

Table 1

Cross sections of the reactions ^{27}Al(d,x)^{24}Na, ^{27}Al(^{12}C,x)^{24}Na, ^{64}Cu(d,x)^{24}Na, ^{64}Cu(^{12}C,x)^{24}Na, and total intensities of incident particles

Isotope	Energy, AGeV	CS (Al), mb	CS (Cu), mb	Total intensity
d	2	14.6	6.0	$2.2 \cdot 10^{13}$
	4	14.0	6.3	$6.1 \cdot 10^{12}$
^{12}C	2	19.4	9.5	$2.1 \cdot 10^{11}$
	4	19.0	9.5	$6.2 \cdot 10^{10}$

2. RESULTS AND DISCUSSION

Fig. 1 shows radial density distributions of ^{238}U(n,γ) and ^{nat}U(n,f) reaction rates, as well radial dependences of $\bar{\sigma}^{238U}_{capt} / \bar{\sigma}^{238U}_{f}$ spectral index for three detector plates (Z = 0, 254, 647 mm) obtained in the experiments with ^{12}C and deuteron beams of 2 AGeV energy. Reaction rates are given per one accelerated particle, one gram of ^{nat}U, and 1 GeV of beam energy.

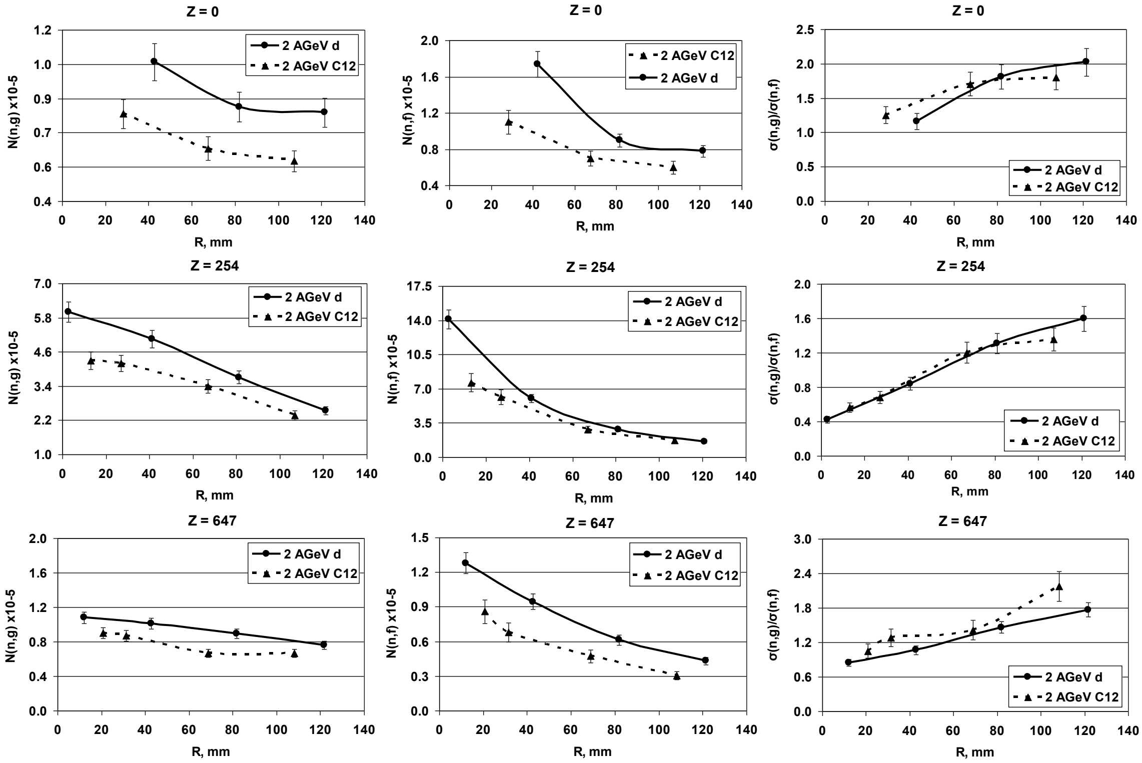

Fig. 1. The radial distributions of density of ^{238}U(n, γ) neutron capture reaction rate (left), ^{nat}U(n, f) fission reaction rate (center) and spectral indices (right) for deuterons and ^{12}C nuclei with energy of 2 AGeV for detector plates Z = 0, 254, 647 mm

This figure shows that yields of radiative capture and fission reactions for detector plate Z = 0 in case of deuterons are about 25% higher in comparison with case of ^{12}C nuclei. For detector plates Z = 254, 647 mm the difference between yields reduces, but in the case of deuterons yields are always larger. The radial dependences of spectral index for all detector plates are about identical for both deuterons and ^{12}C nuclei. The values of spectral index change from ~ 0.5 to ~ 2.

For the 4 AGeV energy incident particles all radial distributions agree within experimental errors with the corresponding distributions shown in Fig. 1 for the 2 AGeV energy incident particles.

This suggests that, firstly, with Z increasing neutron spectrum softening occurs, secondly, the neutron spectra for both types of incident particles and both energies (2 and 4 AGeV) are approximately identical.

Based on obtained spatial distributions of neutron capture reactions on ^{238}U and fissions of ^{nat}U in the uranium target of assembly "QUINTA" the total number of ^{239}Pu production and the total number of ^{nat}U fissions were determined at the volume of uranium. Table 2 shows the experimental (obtained by interpolation of experimental points on uranium volume) integral numbers of ^{239}Pu nuclei and integral number of ^{nat}U fissions for deuterons and ^{12}C nuclei with energies of 2 and 4 AGeV. The results are shown per 1 accelerated particle and 1 GeV of beam energy.

Table 2

Total number of ^{239}Pu nuclei and total number of fissions in uranium target of "QUINTA"

Isotope	Energy AGeV	^{239}Pu	^{nat}U(n,f)
d	2	11.3 ± 0.6	9.6 ± 0.7
	4	10.5 ± 0.6	9.5 ± 0.7
^{12}C	2	8.7 ± 0.7	7.5 ± 0.8
	4	7.8 ± 0.7	7.7 ± 0.8

In total five Runs of irradiation by deuterons with four energies (0.5, 1, 2 and 4 AGeV) [6] and ^{12}C nuclei with two energies (2 and 4 AGeV) have been held during the research of neutron-physical characteristics of uranium target of assembly "QUINTA". Fig. 2 shows the results of total number of produced ^{239}Pu nuclei and total number of fissions for four deuteron energies and two ^{12}C nuclei energies per 1 accelerated particle and 1 GeV of beam energy. Data for deuterons are averaged over the results of several Runs.

We can see that with increasing of primary particle energy the number (per unit of primary beam power) of radiative capture reactions and fissions does not change for deuterons and for ^{12}C nuclei within the experimental error. It should be noted that in case of ^{12}C nuclei the total number of fissions and especially the total number of captures is markedly lower than in case of deuteron irradiation. Perhaps this is due to usage of understated values of monitor reactions cross sections for ^{12}C nuclei.

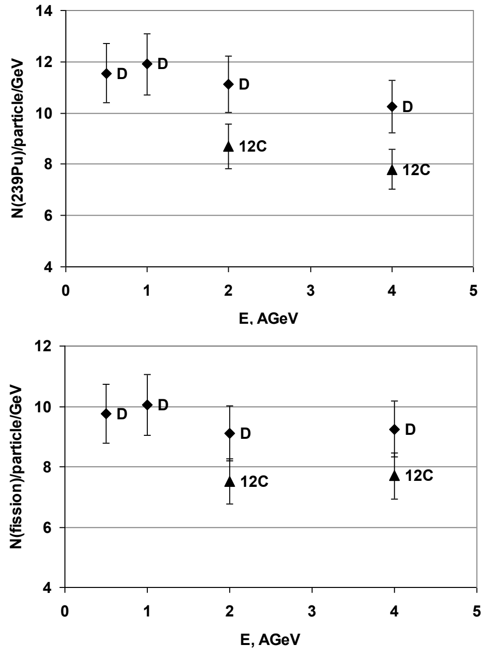

Fig. 2. Total number of ^{239}Pu nuclei (top) and total number of fissions (bottom) in the uranium target of assembly "QUINTA" at deuteron energy E_d = 0.55, 1, 2, 4 AGeV and ^{12}C nucleus energy E_{12C} = 2 and 4 AGeV

Thus, we do not observe the neutron yield increase with growth of energy of primary accelerated particles predicted by V. Yurevich. Presumably this is due to insufficient size of uranium target, which is not quasi-infinite.

Fig. 3 shows the radial distributions of total number of ^{239}Pu production and uranium fissions (per unit of beam power) for each of the five sections of the uranium target and for the whole target at different energies of ^{12}C ion beam.

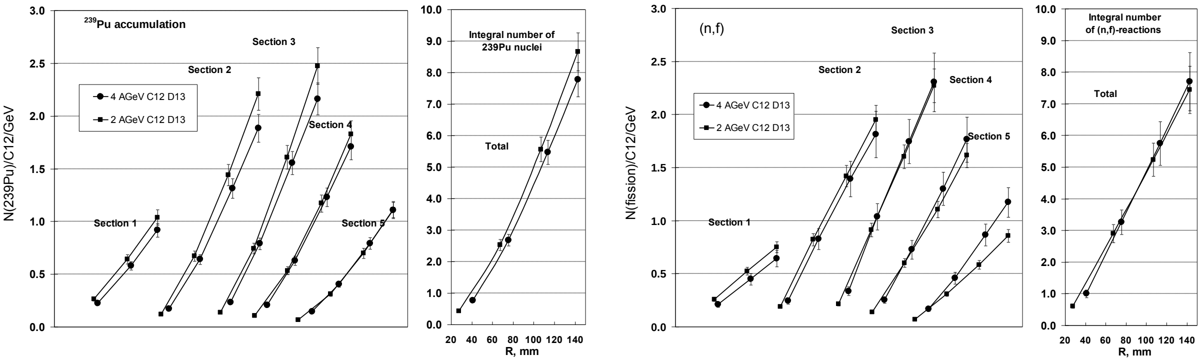

Fig. 3. Dependence of integral distributions of reactions (n, γ) – (left) and (n, f) – (right) on the uranium target for ^{12}C nuclei with energies of 2 and 4 AGeV

These distributions were obtained by integrating the corresponding radius of the target. That is, each point on the graph (see Fig. 3) shows the total number of fissions or the number of (n, γ) reactions in the cylindrical volume of the corresponding radius of the section of uranium target. It should be noted that those values have substantially linear dependence in radial direction. In case of deuterons similar curves are observed [6]. It is clear that such dependence should go to the plateau with increasing of the uranium target radius. This condition corresponds to quasi-infinite target condition. Thus, obtained dependencies confirm that uranium target of assembly "QUINTA" is not quasi-infinite.

As result of γ-spectra processing of irradiated Co activation detectors the yields of different ^{59}Co(n,x) reactions were obtained.

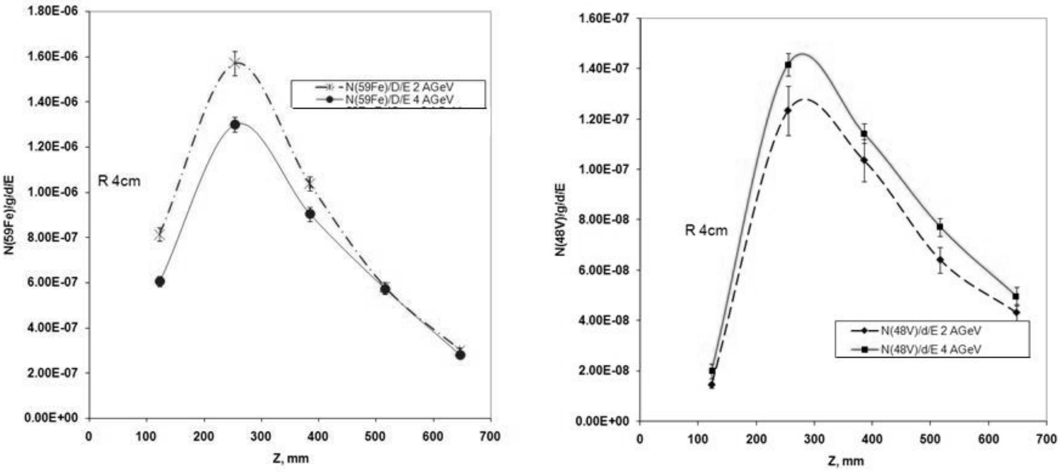

Fig. 4. Axial distribution of reaction rates $^{59}Co(n,p)^{59}Fe$ (left) and $^{59}Co(n,x)^{48}V$ (right) for deuteron energies E_d = 2 and 4 AGeV

As an example, Fig. 4 shows the axial distributions of reaction rate ^{59}Co(n,p)^{59}Fe and ^{59}Co(n,x)^{48}V for deuterons with energy E_d = 2 and 4 AGeV. The maximum cross section of ^{59}Fe production is E_{max} ~ 13 MeV [10]. While the maximum cross section of ^{48}V production is E_{max} ~ 160 MeV [11]. The figure analysis shows, that the number of neutrons with energies above 100 MeV increases with increasing of deuteron energy from 2 to 4 AGeV, but at the same time the number of neutrons with energies less than < 30 MeV decreases.

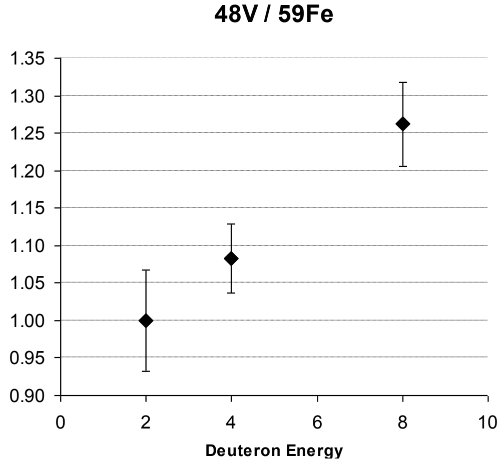

Fig. 5. Dependence of the reaction rate ratio $^{59}Co(n,x)^{48}V/^{59}Co(n,p)^{59}Fe$ from deuteron energy

This fact is indirect confirmation of the fact that neutron spectrum becomes harder with increasing of deuteron energy in uranium target of assembly "QUINTA". Similar results were also obtained in previous target irradiation by three deuteron energies 1, 2, 4 AGeV.

Fig. 5 shows dependence of reaction rate ratio ^{59}Co(n,x)^{48}V/^{59}Co(n,p)^{59}Fe from deuteron energy. This figure shows that reaction rate ratio increases with increasing of energy of primary deuteron beam. As for ^{12}C nuclei case, we could not determine the reaction yields ^{59}Co(n,x) reliably due to insufficient intensity of primary beam (see Table 1).

CONCLUSIONS

Based on the measured spatial distributions of density of radiative neutron capture and fission reactions, the total number of above reactions in the volume of uranium target of assembly "QUINTA" was obtained. Within experimental error, total number of reactions ^{238}U(n,γ) and total number of reactions ^{nat}U(n,f) do not change both for deuterons (energy range from 1 to 8 GeV) and for ^{12}C nuclei (24 and 48 GeV). That is we didn't note increase of the neutron yield as from thermal neutrons and above as from neutrons with energy above than 1 MeV. Probably, this is due to the fact that irradiated uranium target is not quasi-infinite (R ~ 15 cm and average density ~ 12 g/cm^3). Using activation cobalt detectors it was found that neutron spectrum hardening in the uranium target of assembly "QUINTA" was observed with increasing of deuteron energy. For this type of particles and their energy range such experiments were performed for the first time.

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СРАВНЕНИЕ НЕЙТРОННО-ФИЗИЧЕСКИХ ХАРАКТЕРИСТИК УРАНОВОЙ МИШЕНИ УСТАНОВКИ «КВИНТА» ПРИ ОБЛУЧЕНИИ РЕЛЯТИВИСТСКИМИ ДЕЙТРОНАМИ И ЯДРАМИ ^{12}C

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Проблема утилизации отработанного ядерного топлива и ограниченность запасов сырья на сегодняшний день являются двумя основными причинами, препятствующими широкомасштабному распространению атомной энергетики. Одним из путей решения данных проблем является использование ядерных релятивистских технологий, которые предлагают использование максимально жёсткого спектра нейтронов в массивных мишенях из природного или обеднённого урана, облучаемых пучками релятивистских частиц высоких энергий (2…10 ГэВ). Данная работа описывает исследование генерации нейтронов в протяжённой мишени из природного урана (установка "КВИНТА", $m_U \sim 500$ кг), облучаемой пучками релятивистских дейтронов и ядер ^{12}C с энергиями 2 и 4 ГэВ/нукл. на ускорителе «Нуклотрон» (ОИЯИ, Дубна). С помощью активационной методики были исследованы скорости реакций: ^{nat}U(n,f), ^{238}U(n,γ), ^{59}Co(n,x). Проведено сравнение полученных экспериментальных результатов в зависимости от энергии и вида налетающих частиц.

ПОРІВНЯННЯ НЕЙТРОННО-ФІЗИЧНИХ ХАРАКТЕРИСТИК УРАНОВОЇ МІШЕНІ УСТАНОВКИ «КВІНТА» ПРИ ОПРОМІНЕННІ РЕЛЯТИВІСТСЬКИМИ ДЕЙТРОНАМИ ТА ЯДРАМИ ^{12}C

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Проблема утилізації відпрацьованого ядерного палива та обмеженість запасів сировини на сьогоднішній день є двома основними причинами, що перешкоджають широкомасштабному поширенню атомної енергетики. Одним із шляхів вирішення даних проблем є використання ядерних релятивістських технологій, які пропонують використання максимально жорсткого спектра нейтронів у масивних мішенях з природного або збідненого урану, що опромінюються пучками релятивістських частинок високих енергій (2…10 ГеВ). Дана робота описує дослідження генерації нейтронів у протяжній мішені з природного урану (установка "КВІНТА", $m_U \sim 500$ кг), яка опромінювалася пучками релятивістських дейтронів та ядер ^{12}C з енергіями 2 та 4 ГеВ/нукл. на прискорювачі «Нуклотрон» (ОІЯД, Дубна). За допомогою активаційної методики були досліджені швидкості реакцій: ^{nat}U(n,f), ^{238}U(n,γ), ^{59}Co(n,x). Проведено порівняння отриманих експериментальних результатів залежно від енергії та виду налітаючих частинок.