AVERAGE HIGH-ENERGY NEUTRON FLUX DISTRIBUTION IN THE QUINTA SUB-CRITICAL ASSEMBLY IRRADIATED WITH PROTON BEAM OF 0.66 GeV ENERGY APPLYING THE ACTINIDE SPECTRAL INDEX METHOD*

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The paper presents the results of investigations of nuclear-physical characteristics of neutron fields generated in a massive uranium target irradiated by protons with an energy of 0.66 GeV. Twenty three natural uranium samples were spatially arranged in a sub-critical assembly Quinta (at the Joint Institute for Nuclear Research, Dubna, Russia). The samples were irradiated with spallation neutrons. We have processed the experimental data based on gamma-ray spectrometry in order to reach (obtain) the number of neutron induced fissions and neutron captures in the detector foils. The try-and-error method was applied to obtain the neutron energy for which the ratio of the fission cross section to the capture cross section of the natural uranium, from the nuclear database, is equal to the measured ratio of the spectral indexes. The retrieved distinct fission and capture cross sections for the distinct neutron energy from the nuclear database describe the average values which enabled us to evaluate the average neutron flux and neutron fluency distribution in the assembly.

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1. Introduction

The idea of the actinide spectral index method is to search the neutron energy (E_{d}) for the ratio ($\alpha(E_{\mathrm{d}})$) of fission cross section ($\sigma_{\mathrm{f}}(E_{\mathrm{d}})$) to cap-

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ture cross section ($\sigma_{\mathrm{c}}(E_{\mathrm{d}})$) of the selected actinide isotope from the nuclear database that is equal to the measured ratio (α_{m}) of fissioned ($N_{y\mathrm{f}}$) and captured ($N_{y\mathrm{c}}$) actinide isotopes (spectral indexes) [1, 2]

$$\alpha(E_{\mathrm{d}}) = \frac{\sigma_{\mathrm{f}}(E_{\mathrm{d}})}{\sigma_{\mathrm{c}}(E_{\mathrm{d}})} = \alpha_m = \frac{N_{y\mathrm{f}}}{N_{y\mathrm{c}}} = \frac{\bar{\sigma}_{\mathrm{f}}}{\bar{\sigma}_{\mathrm{c}}}\,.$$

Since the measured spectral indexes are defined as the ratio of average fission and capture cross sections, the retrieved distinct fission and capture cross sections for the distinct neutron energy from the nuclear database describe the average values.

Having the average fission and capture cross section values, we can evaluate the average neutron flux in the location of the actinide sample using the measured amount of fissioned and captured actinide isotopes. Two different equations for fissioned ($N_{y\mathrm{f}}$) and captured ($N_{y\mathrm{c}}$) actinide isotopes should give the same average neutron flux value which is a proof for correct measurement.

The number of neutron induced fissioned ($N_{y\mathrm{f}}$) and neutron captured actinide isotopes ($N_{y\mathrm{c}}$) in the actinide sample of volume V_p can be expressed by equations

$$N_{y\mathrm{f}} = V_p\bar{\Phi}N\bar{\sigma}_{\mathrm{f}}t\,, \tag{1}$$
$$N_{y\mathrm{c}} = V_p\bar{\Phi}N\bar{\sigma}_{\mathrm{c}}t\,, \tag{2}$$

where $\bar{\Phi}$ — average neutron flux in the place of actinide sample location [$n/\mathrm{cm}^2\,\mathrm{s}$], N — number of actinide isotopes in volume unit [cm^{-3}], $\bar{\sigma}_{\mathrm{f}}; \bar{\sigma}_{\mathrm{c}}$ — average microscopic cross section for the reactions (n, f) or (n, γ) [barns], t — irradiation time.

The precision of this method is estimated to be around 25%.

1.1. Metallic natural uranium as activation detector foil

Since metallic natural uranium consists of U-238 (99.2752%), U-235 (0.7202%), and a very small amount of U-234, the irradiated detector foil introduces an additional error in the measurement of the average neutron flux and neutron fluency. The number of U-235 fissions is significant comparable to the number of U-238 fissions for the high-energy neutron. This complicates the measurement and reduces the accuracy of the average neutron energy determination and, consequently, the average fission cross sections and in turn the neutron fluency.

Since the measurements of the amount of fissions in the irradiated natural uranium foil constitute the sum of U-238 and U-235 fissions, Eq. (1) must be modified. In contrast, the measurement of neutron captures is based on

the measurement of the amount of Pu-239 produced (see Eq. (3)), so the Eq. (2) does not have to be modified since neutron captures do not occur for U-235

$$^{238}\mathrm{U}(n,\gamma)^{239}\mathrm{U}\frac{\beta}{23.5\,\mathrm{min}}{}^{239}\mathrm{Np}\frac{\beta}{2.36\,\mathrm{day}}{}^{239}\mathrm{Pu}\,. \tag{3}$$

In order to include this, we take into account the number of neutron fission induced (N_{yfs}) which is the sum of U-238 (N_{yf8}) and U-235 (N_{yf5}) fissions and neutron captured actinide isotopes (N_{yc8}) in the actinide foil of volume V_p which is expressed by the following equations:

$$\begin{aligned} N_{yfs} &= N_{yf8} + N_{yf5} = V_p\bar{\Phi}N_8\bar{\sigma}_{\mathrm{f8}}t + V_p\bar{\Phi}N_5\bar{\sigma}_{\mathrm{f5}}t \\ &= V_p\bar{\Phi}N_8t\left(\bar{\sigma}_{\mathrm{f8}} + \frac{N_5}{N_8}\bar{\sigma}_{\mathrm{f5}}\right), \end{aligned} \tag{4}$$

$$N_{yc8} = V_p\bar{\Phi}N_8\bar{\sigma}_{\mathrm{c8}}t\,, \tag{5}$$

where N_8 — number of U-238 atoms in volume unit of actinide foil $[\mathrm{cm}^{-3}]$, N_5 — number of U-235 atoms in volume unit of actinide foil $[\mathrm{cm}^{-3}]$, $\bar{\sigma}_{\mathrm{f8}}$ — U-238 average microscopic cross section for the reactions (n,f) [barns], $\bar{\sigma}_{\mathrm{f5}}$ — U-235 average microscopic cross section for the reactions (n,f) [barns], $\bar{\sigma}_{\mathrm{c8}}$ — U-238 average microscopic cross section for the reactions (n,γ) [barns], $\bar{\Phi}$ — average neutron flux in the place of actinide sample location $[n/\mathrm{cm}^2\,\mathrm{s}]$, $N_5/N_8 = 0.7202/99.2752 = 0.00725 = 7.25\times10^{-3}$.

The quotient of Eqs. (4) and (5) gives the measured spectral index of the irradiated sample on the left, and on the right we get the expression (α_{m85}) which becomes equal to the measured index when we find the neutron energy applying try-and-error method from the nuclear database for which the relevant fission and capture cross section of the U-238 and U-235 fulfill the equation

$$\alpha_{\mathrm{m85}}(E_{\mathrm{d}}) = \frac{N_{yfs}}{N_{yc8}} = \frac{\bar{\sigma}_{\mathrm{f8}}}{\bar{\sigma}_{\mathrm{c8}}} + \frac{N_5}{N_8}\frac{\bar{\sigma}_{\mathrm{f5}}}{\bar{\sigma}_{\mathrm{c8}}}\,. \tag{6}$$

2. Experimental part

2.1. Sub-critical assembly Quinta

Quinta is a deeply sub-critical nuclear assembly with k_{eff} on the level of 0.22. Its target-blanket consists of a total of 512 kg of natural uranium in five sections. Each section is 114 mm long and is separated by a 17 mm air gap, where samples mounted onto sample plates may be easily placed (Fig. 1 (left)).

The fission and activation samples of interest (natural uranium) were mounted on these plates at different radial distances from the Z-axis. More details about the Quinta sub-critical assembly can be found in [2].

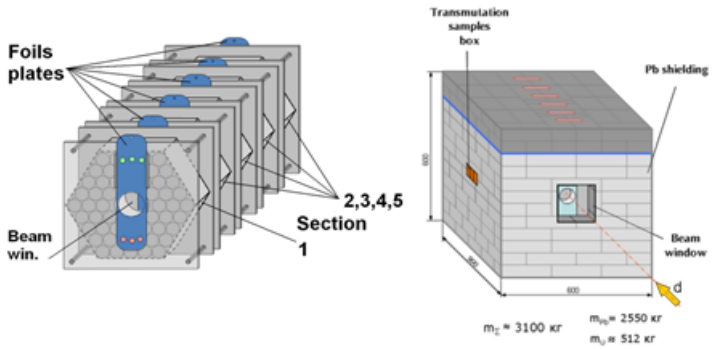

Fig. 1. Schema of Quinta assembly. On the left, there is a view on the uranium target with supporting structures and plastics used for sample placement (detector's plates), on the right, there is a view on the lead shielding enfolding the target with marked the transmutation samples box (window) for the actinides sample location in the shielding.

2.2. Location of activation detector foils in the Quinta sub-critical assembly

The positions of the activation detector foils are in the center of the target, and at the horizontal positions of 40, 80 and 120 mm above the target center behind all five sections and also in front of the first section at the horizontal positions at 40, 80 and 120 mm. The foils were fixed at the plates 2 mm in thickness and arranged to the 17 mm air gaps, which is between the hexagonal sections.

The location coordinates of all of 23 uranium detectors are shown in Table I relative to the axis of the target (along the radius R of the uranium target and along the axis of the target Z).

TABLE I

The location coordinates of all of 23 uranium detectors.

R/Z, mm	Foil plates					
R — vertically	1	2	3	4	5	6
Z — horizontally	0	123	254	385	516	647
0		U_{11}	U_{21}	U_{31}	U_{41}	U_{51}
40	U_{02}	U_{12}	U_{22}	U_{32}	U_{42}	U_{52}
80	U_{03}	U_{13}	U_{23}	U_{33}	U_{43}	U_{53}
120	U_{04}	U_{14}	U_{24}	U_{34}	U_{44}	U_{54}

2.3. Measurements

2.3.1. Irradiation details

The Quinta target was irradiated with a pulsed proton beam of 0.66 GeV energy extracted from the Phasotron accelerator, located at the JINR. The total number of protons of the irradiation is equal to 8.5×10^{14} during the time of irradiation equal to 20 580 seconds (5 h 43 min). Prior to the irradiation, several Polaroid films were placed on the front of Quinta to ensure the proton beam was striking in the centre of the beam window.

2.3.2. Results

After the end of irradiation, the uranium foils were taken out from the target to measure γ-spectra using HPGe detectors. Measurement of gamma-ray spectra of irradiated foils was performed in 4 hours after the end of irradiation (more than 10 half-lives of U-239). In this period, 99.9% of U-239 nuclei have decayed to Np-239.

The gamma-ray spectrum analysis was carried out in a well-established manner. The number of fissions was determined by yield of gamma-lines 743.36 keV (93%), 364.49 keV (81.5%), 529.9 keV (87%), and 293.3 keV (42.8%) of fission fragments ^{97}Zr — 5.7%, ^{131}I — 3.6%, ^{133}I — 6.3%, ^{143}Ce — 4.3%, respectively [3, 4].

The number of neutron radiation capture reactions was determined by the yield of-line with the energy of 277.6 keV γ-line ($I = 14.44\%$) accompanying decay of ^{239}Np (see Eq. (3)) [3, 4].

Spatial distributions of $^{238}\mathrm{U}(n, f)$ reaction rate (fission rate) and Pu-239 production rate (capture rate) were performed.

Having the measured number of fissions and captures in the natural uranium foils, we get (obtain) the spectral indexes.

Having, in turn, the measured spectral index equal to the ratio of average fission and average capture cross section, we can evaluate the average neutron flux in the location of the actinide sample using the measured amount of fissioned and captured actinide isotopes.

This is done by applying the try-and-error method where we look for the neutron energy for which the ratio of fission cross section to capture cross section of the selected actinide isotope from the nuclear database is equal to the measured ratio of fissioned and captured actinide isotopes.

Since the measured ratio is defined as the ratio of average fission and capture cross sections, the retrieved distinct fission and capture cross sections for the distinct neutron energy from the nuclear database (ENDF/B-VII.1) describe the average values. Associating the evaluated average neutron energy (see Table I) with the fission-to-capture ratio spectral indexes, we can note that the average neutron energy is higher than 1 MeV in all the locations of the detector foils (see Fig. 2).

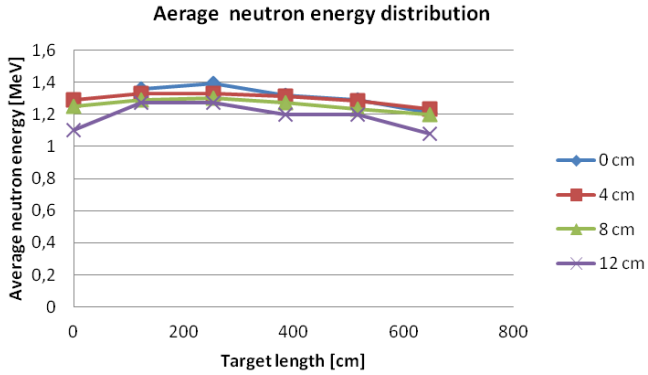

Fig. 2. Average neutron energy distribution *versus* target length for four different radii.

The obtained values for the average fission and capture cross sections let us evaluate the neutron fluencies distribution in the sub-critical assembly Quinta by the help of the equations (Eq. (4) or/and Eq. (5)). This is presented in Fig. 3.

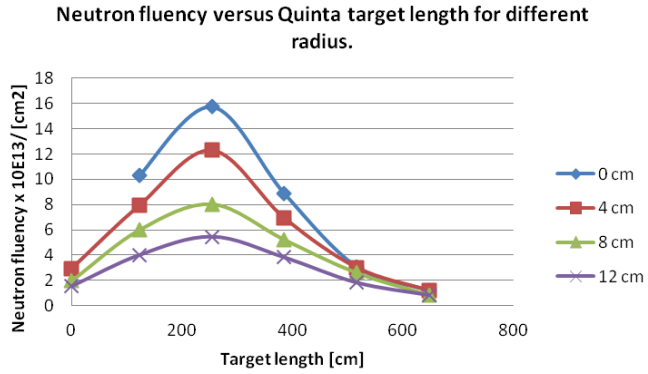

Fig. 3. Neutron fluency distribution *versus* target length for four different radii.

Two different equations (Eq. (4) and Eq. (5)) for fissioned and captured actinide isotopes gave the same neutron fluencies values what is a proof for correct measurement.

Distribution of 23 natural uranium samples in the whole volume of deeply sub-critical Quinta assembly let us also determine the volumetric distribution of average neutron flux of a specified average energy which, in turn, let us determine the optimal place in the assembly for incineration of the minor actinides. So far, such measurements have not been performed.

3. Conclusions

Actinide samples can be used as neutron fluency detectors especially in the high neutron energy range that is difficult to measure.

Both the natural uranium and the neptunium 237 actinides can be applied as high-energy neutrons fluencies detectors.

In the case of irradiation of Am-241 under the conditions described above, the spectral index should reach above 4, while for Np-237, it should reach above 8, which indicates that the incineration of these minor actinides will be effective.

It is widely known that the average neutron energy during the process of fission is about 2 MeV but during the process of spallation, it is about 3 MeV.

The quotient of cross sections for fission and capture for these neutron energies (1, 2 and 3 MeV) gives information on incineration of minor actinides. This is clearly seen in Table II where the mentioned parameters are collected for these neutron energies extracted from the database ENDF/B-VII.1

TABLE II

The fission/absorption ratios as a function of neutron energy for Np-237 and Am-241.

Neutron energy [MeV]	Fission cross section $\sigma(n,f)$ [barn]	Capture cross section $\sigma(n,\gamma)$ [barn]	$\sigma(n,f)/\sigma(n,\gamma)$
Np-237			
1.0	1.4587	0.17277	8.43213
2.0	1.7001	0.06090	27.9664
3.0	1.6609	0.032674	50.8339
Am-241			
1.0	1.2615	0.292707	4.30977
2.0	1.8498	0.07717	23.9701
3.0	1.85973	0.02145	86.6827

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