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

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Шута М. и др. E1-2020-33

Распределение среднего потока нейтронов высокой энергии в подкритической сборке «Квинта», облученной протонным пучком с энергией 0,66 ГэВ с применением метода спектрального индекса актинида

Для оценки распределения нейтронных потоков высокой энергии в подкритической сборке «Квинта» (Объединенный институт ядерных исследований, Дубна) были пространственно распределены двадцать три образца природного урана. Образцы облучались с помощью нейтронов расщепления. Когда соотношение поперечного сечения деления и поперечного сечения захвата природного урана для выбранной из базы ядерных данных нейтронной энергии равно измеренному соотношению спектральных индексов актинида, тогда можно оценить среднее распределение нейтронного потока в сборке.

Работа выполнена в Лаборатории физики высоких энергий им. В. И. Векслера и А. М. Балдина ОИЯИ.

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Average High Energy Neutron Flux Distribution in the Quinta Subcritical Assembly Irradiated with Proton Beam of 0.66-GeV Energy Applying the Actinide Spectral Index Method

To evaluate the high energy neutron flux distribution, twenty three natural uranium samples were spatially arranged in the Quinta subcritical assembly (at the Joint Institute for Nuclear Research, Dubna). The samples were irradiated with spallation neutrons. When the ratio of fission cross section to capture cross section of the natural uranium for the distinct neutron energy from the nuclear data base is equal to the measured ratio of spectral indexes, it enables us to evaluate the average neutron flux distribution in the assembly.

The investigation has been performed at the Veksler and Baldin Laboratory of High Energy Physics, JINR.

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INTRODUCTION

The idea of the actinide spectral index method is to search for the neutron energy (E_d) for the ratio ($\alpha(E_d)$) of fission cross section ($\sigma_f(E_d)$) to capture cross section ($\sigma_c(E_d)$) of the selected actinide isotope from the nuclear data base that is equal to the measured ratio (α_m) of fissioned (N_{yf}) and captured (N_{yc}) actinide isotopes (spectral indexes) [1, 2]:

$$\alpha(E_d) = \frac{\sigma_f(E_d)}{\sigma_c(E_d)} = \alpha_m = \frac{N_{yf}}{N_{yc}} = \frac{\overline{\sigma}_f}{\overline{\sigma}_c}.$$

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 data base 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_{yf}) and captured (N_{yc}) actinide isotopes should give the same average neutron flux value which is a proof for correct measurement.

The number of neutron-induced fissioned (N_{yf}) and neutron-captured (N_{yc}) actinide isotopes in the actinide sample of volume V_p can be expressed by equations

$$N_{yf} = V_p \overline{\Phi} N \overline{\sigma}_f t, \tag{1}$$

$$N_{yc} = V_p \overline{\Phi} N \overline{\sigma}_c t, \tag{2}$$

where

$\overline{\Phi}$ — average neutron flux in the place of actinide sample location, $\mathrm{cm}^{-2} \cdot \mathrm{s}^{-1}$,

N — number of actinide isotopes in volume unit, cm^{-3},

$\overline{\sigma}_f; \overline{\sigma}_c$ — average microscopic cross section for the reactions (n,f) or (n,γ), b,

t — irradiation time.

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

Metallic Natural Uranium as Activation Detector Foil. Since metallic natural uranium consists of ^{238}U (99.2752%), ^{235}U (0.7202%), and a very small amount of ^{234}U, the irradiated detector foil introduces an additional error in the measurement of the average neutron flux and neutron fluency. The number of ^{235}U fissions is significant comparable to the number of ^{238}U fissions for the high energy neutron. And this complicates the measurement and reduces the accuracy of the average neutron energy determination and, consequently, the average fission cross sections, and this, in turn, the neutron fluency.

Since the measurements of the amount of fissions in the irradiated natural uranium foil constitute the sum of ^{238}U and ^{235}U fissions, Eq. (1) must be modified. In contrast, the measurement of neutron captures is based on that of the amount of ^{239}Pu produced (see Eq. (3)), so Eq. (2) does not have to be modified since neutron captures do not occur for ^{235}U

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

In order to include this, we take into account the number of neutron-induced fission (N_{yfs}) which is the sum of ^{238}U (N_{yf8}) and ^{235}U (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:

$$N_{yfs} = N_{yf8} + N_{yf5} =$$

$$= V_p\overline{\Phi}N_8\overline{\sigma}_{f8}t + V_p\overline{\Phi}N_5\overline{\sigma}_{f5}t = V_p\overline{\Phi}N_8t(\overline{\sigma}_{f8} + \frac{N_5}{N_8}\overline{\sigma}_{f5}), \tag{4}$$

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

where
N_8 — number of ^{238}U atoms in volume unit of actinide foil, cm^{-3},
N_5 — number of ^{235}U atoms in volume unit of actinide foil, cm^{-3},
$\overline{\sigma}_{f8}$ — ^{238}U average microscopic cross section for the reactions (n,f), b,
$\overline{\sigma}_{f5}$ — ^{235}U average microscopic cross section for the reactions (n,f), b,
$\overline{\sigma}_{c8}$ — ^{238}U average microscopic cross section for the reactions (n,γ), b,
$\overline{\Phi}$ — average neutron flux in the place of actinide sample location, $\mathrm{cm}^{-2}\cdot\mathrm{s}^{-1}$,
$N_5/N_8 = 0.7202/99.2752 = 0.00725 = 7.25\cdot10^{-3}$.

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 the try and error method from the nuclear data base for which the relevant fission and capture cross sections of ^{238}U and ^{235}U fulfill the equation

$$\alpha_{m85}(E_d) = \frac{N_{yfs}}{N_{yc8}} = \frac{\overline{\sigma}_{f8}}{\overline{\sigma}_{c8}} + \frac{N_5}{N_8}\frac{\overline{\sigma}_{f5}}{\overline{\sigma}_{c8}}. \tag{6}$$

EXPERIMENTAL PART

The Quinta Subcritical Assembly. Quinta is a deeply subcritical nuclear assembly with k_{eff} on the level 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 separated by a 17-mm air gap, where samples mounted onto sample plates may be easily placed (Fig. 1, *a*).

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

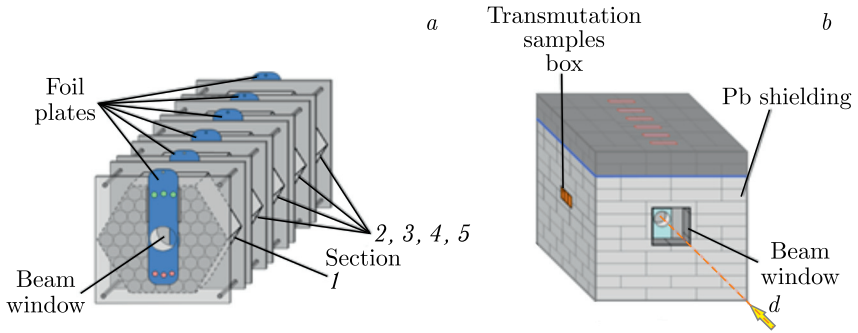

Fig. 1. Schema of the Quinta assembly. A view on the uranium target with supporting structures and plastics used for sample placement (detector's plates) (*a*), and a view on the Pb shielding enfolding the target with marked transmutation samples box (window) for the actinide sample location in the shielding (*b*)

Location of Activation Detector Foils in the Quinta Subcritical Assembly. The positions of the activation detector foils are in the centre of the target and in the horizontal positions of 40, 80, and 120 mm above the target centre behind all five sections, as well as in front of the first section in 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 between the hexagonal sections.

The location coordinates of all of 23 uranium detectors are shown in Table 1 relative to the axis of the target (along the radius R of the uranium target and along the Z-axis of the target).

Table 1. **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}

Measurements. ***Irradiation Details.*** The Quinta target was irradiated with a pulsed proton beam of 0.66-GeV energy extracted from the Phasotron accelerator located at JINR. Total number of protons of the irradiation is equal to $8.5 \cdot 10^{14}$ during the time of irradiation of 20 580 s (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.

Results. After the end of irradiation, the uranium foils were taken out from the target to measure γ spectra using high-purity germanium (HPGe) detectors. Measurement of gamma-ray spectra of irradiated foils was

performed in 4 h after the end of irradiation (more than 10 half-lives of ^{239}U). In this period, 99.9% of ^{239}U nuclei have decayed to ^{239}Np.

Gamma-ray spectrum analysis was carried out in a well-established manner. The number of fissions was determined by yield of γ lines with 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 γ line with energy of 277.6 keV ($I = 14.44\%$) accompanying decay of ^{239}Np (see Eq. (3)) [3, 4].

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

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

Having in turn the measured spectral index equal to ratio of average fission and capture cross sections, 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 data base 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 data base (ENDF/B-VII.1) describe the average values.

Associating the evaluated average neutron energy (see Table 1) 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).

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

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

Distribution of 23 natural uranium samples in the whole volume of deeply subcritical Quinta assembly allows us also to determine the volumetric distribution of average neutron flux of a specified average energy which, in turn, makes it possible to determine the optimal place in the assembly for incineration of the minor actinides. So far, such measurements have not been performed in the world.

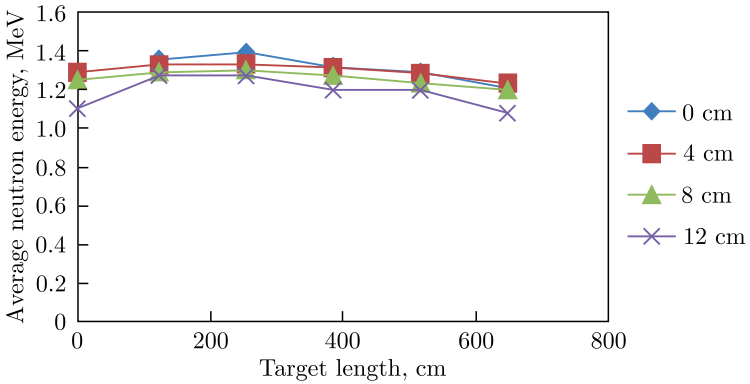

Fig. 2. Average neutron energy distribution versus target length for four different radii

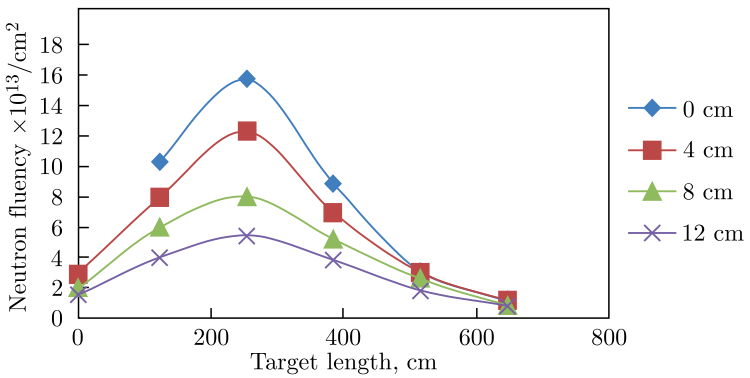

Fig. 3. Neutron fluency distribution versus target length for four different radii

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 ^{237}Np actinides can be applied as high energy neutron fluencies detectors.

In the case of irradiation of ^{241}Am under the conditions described above, the spectral index should reach above 4, while for ^{237}Np 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 2 where the mentioned parameters are

Table 2. **The fission/absorption ratios in function of neutron energy for ^{237}Np and ^{241}Am**

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

collected for these neutron energies extracted from the ENDF/B-VII.1 data base.

REFERENCES

1. *Szuta M., Kilim S., Strugalska-Gola E., Bielewicz M., Zamyatin N. I., Shafronovskaya A. I., Tyutyunnikov S.* Comparison of Two Fast Neutron Fluence Measurement Methods Based on ^{237}Np Fission-to-Capture Ratio Measurement (Spectral Index) and a Reverse Dark Current Measurement in Planar Silicon Detector // Proc. of XXIII Intern. Baldin Seminar on High Energy Physics Problems "Relativistic Nuclear Physics & Quantum Chromodynamics", Dubna, Russia, Sept. 2016; Eur. Phys. J. Web Conf. 2017. V. 138. P. 10006; https://doi.org/10.1051/epjconf/201713810006.
2. *Szuta M., Kilim S., Strugalska-Gola E., Bielewicz M., Zamyatin N. I., Shafronovskaia A. I., Tyutyunnikov S.* Impact of Average Neutron Energy on the Fast Neutron Fluency Measurement by ^{237}Np Fission-to-Capture Ratio and Reverse Dark Current of Planar Silicon Detector Methods // Proc. of XXIV Intern. Baldin Seminar on High Energy Physics Problems "Relativistic Nuclear Physics & Quantum Chromodynamics", Dubna, Russia, Sept. 2018; Eur. Phys. J. Web Conf. 2019. V. 204. P. 04002; https://doi.org/10.1051/epjconf/201920404002.
3. *Artiushenko M. Yu., Voronko V. A., Husak K. V., Kadykov M. G., Petrusenko Yu. T., Sotnikov V. V., Irzhevskyi D. A., Tyutyunnikov S. I., Furman W. I., Chilap V. V.* Investigation of the Spatial and Energy Distributions of Neutrons in the Massive Uranium Target Irradiated by Deuterons with Energy of 1…8 GeV // Probl. Atomic Sci. Technol. 2013. No. 6(88). P. 170–174.
4. *Artiushenko M. Yu., Baldin A. A., Berlev A. I., Chilap V. V., Dalkhajav O., Sotnikov V. V., Tyutyunnikov S. I., Voronko V. A., Zhadan A. A.* Comparison of Neutron-Physical Characterictics of Uranium Target of Assembly "Quinta" Irradiated by Relativistic Deuterons and ^{12}C Nuclei // Probl. Atomic Sci. Technol. 2016. No. 3(103).

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