Письма в ЭЧАЯ. 2016. Т. 13, № 2(200). С. 382–390

НЕЙТРОННАЯ ФИЗИКА

FISSION OF ^{232}Th IN A SPALLATION NEUTRON FIELD

V. I. Yurevich [a, 1], *V. A. Nikolaev* [b], *R. M. Yakovlev* [b]

[a] Joint Institute for Nuclear Research, Dubna

[b] V. G. Khlopin Radium Institute, Saint Petersburg, Russia

The spatial distributions of the thorium fission reaction rate in a spallation neutron field of a thick lead target bombarded by protons or deuterons with energy between 1.0 and 3.7 GeV were measured. An approximately linear dependence of the thorium fission rate on the beam energy is observed. The mean fission cross section of ^{232}Th is $\langle\sigma_f\rangle \approx 123$ mb and it does not depend on the energy and type of the beam particles.

Измерены пространственные распределения скорости деления тория в поле нейтронов расщепления толстой свинцовой мишени, бомбардируемой протонами или дейтронами с энергией от 1,0 до 3,7 ГэВ. Наблюдается приблизительно линейная зависимость скорости деления тория от энергии пучка. Среднее сечение деления ^{232}Th $\langle\sigma_f\rangle \approx 123$ мб, и оно не зависит от энергии и типа частиц пучка.

PACS: 25.85.Ec; 29.25.Dz

INTRODUCTION

Thanks to the progress in nuclear and accelerator technologies, the idea to use ADS concept for energy production in sub-critical reactors becomes realistic in the near future. Nowadays experts consider thorium as a viable alternative nuclear fuel to uranium. The advantages of thorium are: (i) reduction of the toxicity and longevity of nuclear waste, (ii) a thorium sub-critical reactor operates far from criticality and is inherently safer, (iii) low amounts of weapons-grade materials required for producing a bomb, (iv) can be extracted from many parts of the globe and in greater quantities than uranium, etc. Recently a new international Thorium Energy Committee (iThEC) has been organized with a goal to raise global awareness of the many possibilities thorium offers for solving mankind's growing energy needs [1]. The iThEC is an organization bringing together scientific and technical knowledge from the world's leading research institutes, political decision makers, and industrial partnerships. The important event on this way is organization of international Thorium Energy Conference (ThEC 2013) at CERN in October 2013 [2].

The concepts of sub-critical thorium reactors [3, 4] use an intense beam of GeV-protons incomes vertically in the central part of the reactors bombarding a lead target. The neutrons produced in the spallation lead target hit a thorium blanket located around the spallation area.

[1] E-mail: yurevich@jinr.ru

The history of neutrons in a thorium blanket depends on energy of the neutrons. The energy spectrum of spallation neutrons overlaps a wide energy range from thermal to hundreds of MeV. The high-energy neutrons induce (*n, xnf*) reactions in thorium increasing the neutron multiplicity [5, 6]. The low-energy neutrons are captured and used in the thorium cycle ($^{232}\mathrm{Th} + n - {}^{233}\mathrm{U}$; $^{233}\mathrm{U} + n - \mathrm{fission} + 2.3n$).

Despite an importance of experimental information about reaction rates in thorium induced by spallation neutrons for practical realization of the sub-critical thorium reactor project, these data are very limited. Some information about the fission reaction rates of ^{232}Th around a spallation source based on a thick lead target irradiated by a beam of GeV protons or deuterons was obtained in our experiment [7]. The aim of the experiment was to study spatial-energy distribution of neutrons generated in the thick lead target by the p/d beam. The measurements were carried out by the threshold detector method where the detector set consisted of thin layers of various actinides with different thresholds of fission and one of them was thorium. The primary experimental information was fission reaction rates which were used for restoration of energy distributions of the spallation neutrons.

In this paper, the fission reaction rate distributions of ^{232}Th measured in the spallation neutron field of thick lead target and dependence of the rates on beam energy and ion type are discussed in detail.

1. EXPERIMENT

A lead target 20 cm in diameter and 60 cm thick, irradiated by a beam of protons or deuterons of the Dubna Synchrophasotron, was a source of spallation neutrons that interacted with ^{232}Th nuclei in a set of samples used in our experiment [7]. The thickness of the target more than three times exceeds the nuclear interaction length in lead and such a target may be considered as the target of total absorption of beam.

Besides the thorium, the detector samples included 1-mg/cm^2 fissile layers of ^{235}U, ^{237}Np, ^{238}U, ^{209}Bi, and fragmenting layers of Cu and Cd. The fragments produced by neutrons in the fission and fragmentation reactions were detected with 6-μm PETP SSNTD. This set of various layers with different thresholds of fragment production, neutron detector, was used for determination of energy distribution of neutrons in a wide energy range from thermal to GeV.

The intervals of maximum sensitivity of the fission threshold detectors, corresponding to 90% and 60% contributions to the detector responses, were calculated for the spectrum of spallation neutrons escaping from the target, and the results are given in Table 1. The fission of thorium is effectively induced by the neutrons in a wide energy range from threshold to ~ 155 MeV.

Table 1. **Energy intervals with 90% and 60% contributions to responses of the fission threshold detectors**

Threshold detector	ΔE (90%), MeV	ΔE (60%), MeV
$^{235}\mathrm{U}(n, f)$	0.16–35.4	0.47–4.30
$^{237}\mathrm{Np}(n, f)$	0.58–39.3	0.98–5.31
$^{238}\mathrm{U}(n, f)$	1.50–113	2.06–25.8
$^{232}\mathrm{Th}(n, f)$	1.50–155	2.54–48.5
$^{209}\mathrm{Bi}(n, f)$	66.5–443	101–291

The detectors with various fissile layers were placed on the side surface of the target along its axis z and on the front and back surfaces. For each beam energy, additional measurements were carried out with the detectors placed at six different angles to the beam direction and at a distance of 1 m from the target center. In the latest measurements a larger fluence of beam particles on the target was required.

More detailed information about the experiment can be found elsewhere [7].

2. BEAM

In the measurements the energy of proton and deuteron beams was varied from 1.0 to 3.7 GeV. The beam structure was one burst per 9 s with an intensity of 10^9–10^{10} particles/burst. The fluence of beam particles on the lead target was determined by means of monitor reactions $^{27}\mathrm{Al}(p,X)^{24}\mathrm{Na}$ or $^{27}\mathrm{Al}(d,X)^{24}\mathrm{Na}$, and it was 10^{10}–10^{11} and 10^{11}–10^{12} for surface and angular measurements, respectively. The aluminum disc of the beam monitor was placed at a distance of 40 cm upstream from the front surface of the target. The beam profile was measured with MWPC in vertical and horizontal directions, and it was good fitted by Gaussian distribution with FWHM $\approx 1.5-2$ cm.

3. MEASUREMENTS

The measurements were carried out at five energies of protons and six energies of deuterons between 1.0 and 3.7 GeV listed in Table 2.

In addition we made some measurements with detectors at different distances from the target and behind a concrete block for estimation of reaction rates induced by background neutrons. For the thorium layers at 1 m from the target center this contribution was $(7 \pm 2)\%$.

Besides neutrons, some contribution to fission reaction rates was added by charged particles emitted from the target. The estimation showed that for thorium this effect is a few percent less.

Table 2. **List of measurements with p/d beams**

Beam ion	Energy, GeV	Number of measurements
p	0.99	2
p	2.00	2
p	2.55	1
p	3.17	2
p	3.65	3
d	1.03	1
d	1.49	1*
d	1.98	1
d	2.55	1*
d	3.00	1*
d	3.76	1

*Systematical error $\geqslant 30\%$.

4. FIELD OF SPALLATION NEUTRONS

The neutron energy distributions were obtained from the results of integral measurements by an iteration procedure proposed in [8]. The results of TOF measurement of neutron energy spectra with lead target 20 cm in diameter and 20 cm thick [7] were taken as an initial approximation in the iteration method.

Some examples of the distributions of neutrons with energies above 0, 1, 6, 20, and 100 MeV obtained for two energies of proton beam 0.99 and 3.65 GeV are given here. The neutron fluxes per incident proton on the side surface of lead target are shown in Figs. 1 and 2 for these two beam energies, respectively. The polynomial approximation was applied to fit the data. All the curves have a broad maximum and its position a bit moves from the front surface of the target with increasing neutron energy. A comparison of the distributions

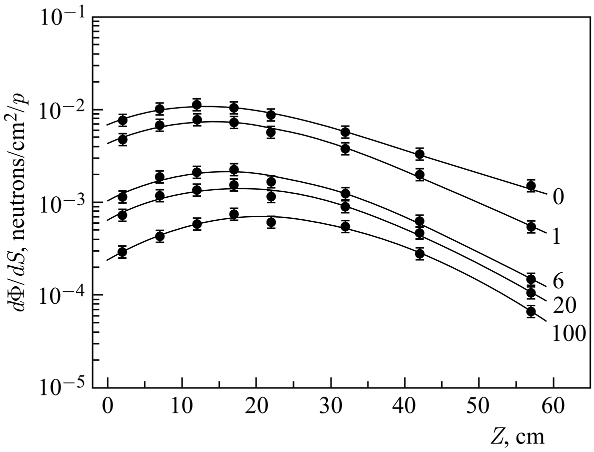

Fig. 1. Neutron distributions over the side surface of lead target obtained for a proton energy of 0.99 GeV: the points — the experimental data; the curves — the polynomial fits; the numbers — the minimal neutron energy in MeV

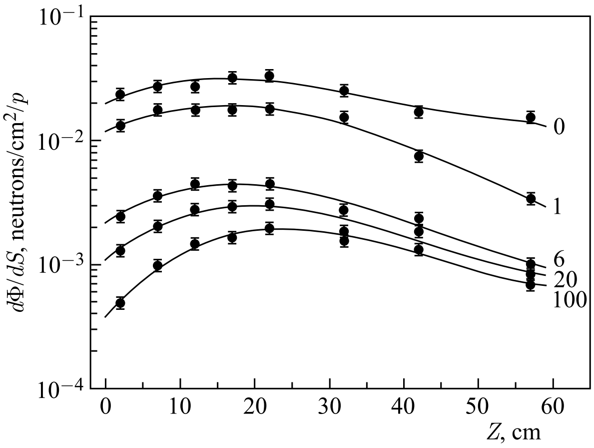

Fig. 2. The same as in Fig. 1 for a proton energy of 3.65 GeV

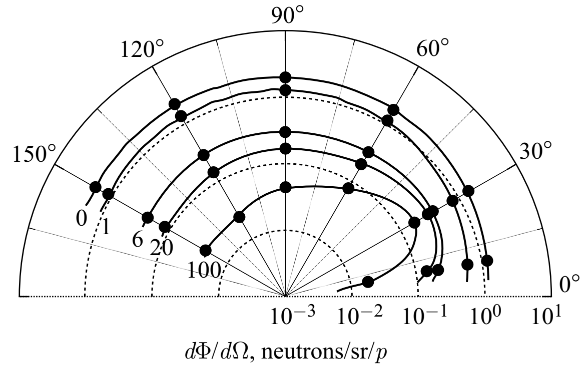

Fig. 3. Angular distributions of neutrons for different energy groups obtained for a proton energy of 0.99 GeV: the points — the experimental data; the curves — the polynomial fits; the numbers — the minimal neutron energy in MeV

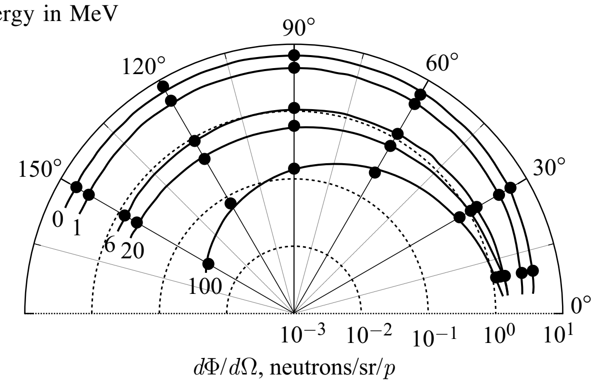

Fig. 4. The same as in Fig. 3 for a proton energy of 3.65 GeV

obtained for different energies of bombarding protons shows more effective neutron production in the second part of the lead target at $z > 30$ cm for the higher proton energy. The maxima of curves shift to higher magnitudes of z with the beam energy. We note that the target thickness is about equal to path of 0.99-GeV protons in lead and in this case one cannot expect an essential contribution to the neutron production at $z > 30$ cm. The situation strongly changes with increasing proton energy to 3.65 GeV where due to hadronic shower in lead the whole target takes part in production and multiplication of neutrons.

The angular distributions of neutrons in different energy groups corresponding to these two energies of protons are shown in Figs. 3 and 4, respectively. The energy spectrum of neutrons becomes harder with decreasing angle of observation, which is especially clearly seen from the results for 3.65-GeV protons.

5. THORIUM FISSION REACTION RATE

In this section the results on fission reaction rate of ^{232}Th in the spallation neutron field of lead target are presented and discussed.

The measured distributions of fission reactions in 1-g/cm^2 thorium layer over the side surface of target per proton for two beam energies of 0.99 and 3.65 GeV are shown in Fig. 5.

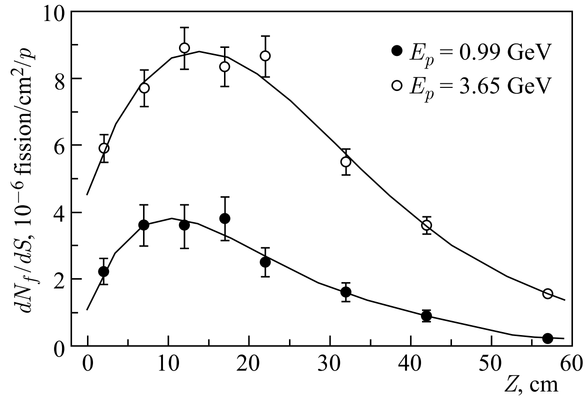

Fig. 5. Distributions of fission reaction rates of ^{232}Th in 1-g/cm^2 layer over the side surface of target for two proton energies of 0.99 and 3.65 GeV: the points — the experimental data; the curves — the polynomial fits

Both the distributions have a broad maximum at a distance from the front surface 10–15 cm and then the reaction rate falls with the distance. But for higher proton energy the reaction rate at $30 < z < 60$ cm is still rather large. As expected, 3.65-GeV protons give much larger yield of fission reactions per incident beam particle than 0.99-GeV protons.

The fission reaction yields on the side, front, and back surfaces are compared in terms of relative contribution to the total yield. The dependence of these contributions R_S, R_F, and R_B on the proton energy is shown in Fig. 6. The relative reaction rates on the side and back surfaces slowly increase with the beam energy. The results obtained for R_F on the front surface of lead target show an opposite tendency.

Two examples of the angular distributions of thorium fission reaction rate measured with 0.99- and 3.65-GeV protons in our experiment are shown in Fig. 7. The magnitudes are given for thorium layer with a thickness of 1 g/cm^2. We remind that in the measurements the

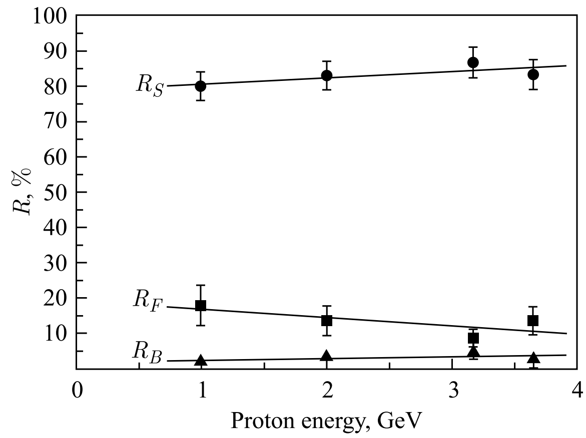

Fig. 6. Dependence of relative fission reaction yields on the side (R_S), front (R_F) and back (R_B) surfaces on proton energy: the points — the experimental data; the curves — the linear fits

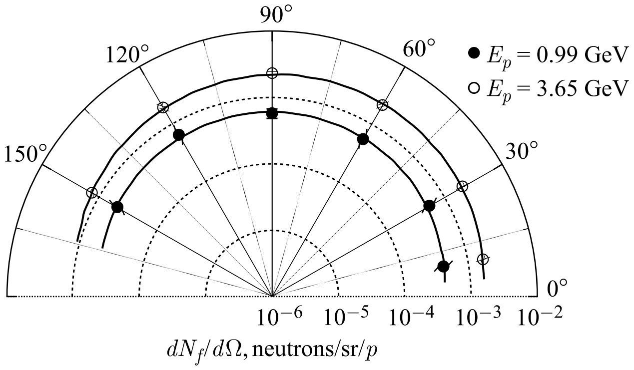

Fig. 7. Angular distributions of thorium fission reaction rate measured with 0.99- and 3.65-GeV protons for thorium layer 1 g/cm^2: the points — the experimental data; the curves — the polynomial fits

detectors were located at a distance of 1 m from the target center. In both cases the maximal fission reaction rate is observed between 60 and 120° and it slightly falls at smaller and larger angles.

Integration of the measured angular distributions over solid angle gives total fission reaction rate of ^{232}Th in 1-g/cm^2 spherical layer around the lead target. The dependence of this value on the proton energy is shown in Fig. 8. The total fission reaction rate demonstrate a linear increasing with the beam energy between 1.0 and 3.7 GeV.

The obtained total fission reaction rates can be used for calculation of the mean fission cross section of ^{232}Th in the spallation neutron spectrum by the formula

$$\langle \sigma_f \rangle = \frac{N_f}{nY_n},$$

where N_f — fission reaction rate per incident proton found by integration over spherical surface with radius 1 m and thickness of thorium layer of 1 g/cm^2; n — number of thorium nuclei per cm^2 in the layer; Y_n — total neutron yield per proton [7].

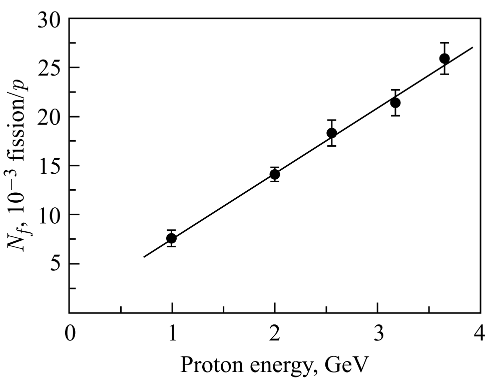

Fig. 8. Total fission reaction rate of ^{232}Th in 1-g/cm^2 spherical layer around the lead target: the points — the experimental data; the curve — the linear fit

Table 3. **The values of N_f, Y_n, and $\langle\sigma_f\rangle$ obtained for three energies of the proton beam bombarding the lead target**

E_p, GeV	N_f, fission/p	Y_n, neutron/p	$\langle\sigma_f\rangle$, mb
0.99	$(7.58 \pm 0.83) \cdot 10^{-3}$	24.1 ± 2.9	121 ± 19
2.0	$(1.41 \pm 0.07) \cdot 10^{-2}$	44.2 ± 5.3	123 ± 16
3.65	$(2.59 \pm 0.16) \cdot 10^{-2}$	80.6 ± 9.7	124 ± 17

The values of N_f, Y_n, and $\langle\sigma_f\rangle$ are given in Table 3. As one can see, the magnitudes of the mean fission cross section of ^{232}Th do not depend on the beam energy and $\langle\sigma_f\rangle \approx 123$ mb.

6. COMPARISON OF RESULTS FOR PROTON AND DEUTERON BEAMS

Neutron emission from lead targets bombarded by high-energy protons and deuterons was carefully studied by means of TOF and threshold detector methods in our experiments in Dubna [7]. The form of energy spectra and spatial distributions of spallation neutrons are not much different for proton and deuteron beams with the same energy. The most essential differences are: (i) deuteron gives higher neutron yield by a factor of 1.10–1.15 than proton with the same energy and (ii) the deuteron generates the neutrons with smaller mean energy in comparison with the proton. As a result, these ions with equal energy in the range from 1.0 to 3.7 GeV produce approximately the same fission reaction rate for ^{232}Th in the neutron field of the lead target.

CONCLUSIONS

The energy spectrum of neutrons produced in the lead target 20 cm in diameter and 60 cm thick by GeV protons or deuterons overlaps a very wide energy range up to beam energy. The large fraction of these neutrons can induce fission of ^{232}Th because their energy exceeds the fission threshold. We observe effective fission of thorium in a wide angular range between 10 and 150° for beam energies from 1.0 to 3.65 GeV. The mean fission cross section of ^{232}Th does not depend on the energy and type of beam particles, and it has rather large magnitude of 123 mb.

We note that there are some arguments to use proton beam with higher energy than 1.0 GeV in the sub-critical thorium reactor project. A single proton with an energy of 3.65 GeV produces in the lead target only a bit less number of neutrons than four protons with an energy of 1 GeV. But the mean energy of spallation neutrons is higher in the first case [7] and it leads to approximately linear dependence of thorium fission reaction rate on the beam energy in the range from 1.0 GeV to 3.65 GeV. Moreover, due to the increasing mean energy one can expect a higher neutron multiplication in a thorium blanket for 3.65-GeV protons. Here, the number of neutrons increases by (n, xnf) reactions and the estimations [5, 6] give the mean multiplicity $\nu \approx 3.3$ and 10 neutrons for the thorium fission at incident neutron energies 10 and 100 MeV, respectively. Additionally, the thickness of lead target volume involved in effective production of neutrons increases with the beam energy, and it leads to more uniform distribution of energy deposition in lead along the beam axis and to wider distribution of thorium fission reaction rate on the blanket surface. Another important point is the magnitude of energy deposition of beam ions in a beam window of sub-critical thorium reactor. The rise of beam energy allows a significant decrease of this magnitude.

REFERENCES

1. http://www.ithec.org/
2. http://indico.cern/ch/event/thec13
3. *Hashemi-Nezhad S. R.* Accelerator Driven Sub-Critical Nuclear Reactors // Austral. Phys. 2006. V. 43. P. 90–95.
4. *Rubbia C.* Sub-Critical Thorium Reactors // Intern. Conf. Energy 2050, Stockholm, Oct. 2009. http://energy2050.se/?type=static&id=35&mo=41.
5. *Kornilov N. V. et al.* Neutron Multiplicity for Neutron Incident Energy from 0 to 150 MeV. IAEA, INDC(CCP)-437. 2003.
6. *Svirin M. I.* Systematics of the Mean Energy and the Mean Multiplicity of Prompt Neutrons Originating from ^{232}Th Fission // Phys. At. Nucl. 2012. V. 75. P. 1466–1480.
7. *Yurevich V. I. et al.* Investigation of Neutron Emission in the Interaction of Relativistic Protons and Deuterons with Lead Targets // Phys. Part. Nucl. Lett. 2006. V. 3. P. 169–182.
8. *Tarasko M. Z., Kramer-Ageev E. B., Tikhonov E. B.* Issues of Dozimetry and Radiation Protection. M.: Atomizdat, 1970. V. 2. P. 125 (in Russian).

Received on April 24, 2015.