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FISSION OF ^{209}Bi, ^{232}Th, ^{235}U, ^{238}U, AND ^{237}Np IN A SPALLATION NEUTRON FIELD

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The spatial-angular distributions and total yields of the fission reactions of ^{209}Bi, ^{232}Th, ^{235}U, ^{238}U, and ^{237}Np induced by spallation neutrons of the extended lead target $\varnothing$ 20 × 60 cm bombarded with 1–3.7 GeV protons and deuterons are analysed.

The investigation has been performed at the Laboratory of High Energies, JINR.

Деление ^{209}Bi, ^{232}Th, ^{235}U, ^{238}U и ^{237}Np в поле spallation нейтронов

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Анализируются пространственно-угловые распределения и полные выходы реакций деления ядер ^{209}Bi, ^{232}Th, ^{235}U, ^{238}U и ^{237}Np, вызванные spallation нейтронами протяженной свинцовой мишени $\varnothing$ 20 × 60 см, бомбардируемой протонами и дейтронами с энергией 1–3,7 ГэВ.

Работа выполнена в Лаборатории высоких энергий ОИЯИ.

1. Introduction

A high flux of neutrons with a hard energy spectrum generated in an extended heavy element target by hydrogen-ion beam at an intermediate energy is often considered as one of the most promising opportunities to transmutate nuclear materials and to create more safe and effective nuclear energetics. The ionization loss for beam ions becomes a minimum at approximately a constant value of the inelastic cross section over an energy range of 1–3 GeV. So, at such energies one can convert the kinetic energy of ions to the neutron production in targets of electronuclear facilities with a maximum efficiency.

The substantiation of nuclear technologies based on high intensity accelerators needs experimental data, especially for target and blanket materials (W, Pb, Bi, Th, U, Np, etc.). The base of such a kind of data is very poor, and it is very important to expand experimental activities in this field of research for making progress in the technology applications.

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In this paper, the authors present the experimental results on the fission reaction yields for ^{209}Bi, ^{232}Th, ^{235}U, ^{238}U, and ^{237}Np in the spallation neutron field of an extended lead target $\varnothing 20 \times 60$ cm bombarded with 1–3.7 GeV protons and deuterons. The fission reaction is one of the most important channels of the heaviest mass nuclei disintegration in neutron-nucleus collisions, and it takes a large fraction of the interaction cross section, especially for actinides. The extended lead target used in our measurements is a good approximation to reproduce the spallation neutron field under ideal and simple experimental conditions. So, the results obtained by the authors can be considered as initial and reference data for planning a new more complicated research.

The spatial-angular distributions of fission reaction yields, the dependence of their shape and total yields on the type and energy of beam ions were studied by the solid state nuclear track detector method, SSNTD. The first results were reported in Ref.1. A comparison of the experimental results measured with proton and deuteron beams at the same energy shows that the deuteron produces approximately a 25–30 % higher fission reaction yield for ^{235}U and ^{237}Np than the proton, but this difference is very small for the other studied actinides. The situation is opposite for bismuth: protons are not of much benefit to the fission reaction yield in comparison with deuterons. For all nuclides, more than 80 % of the fission reaction yield occurs on the side surface of the target with a maximum in the first half of the lead target. The distribution shapes have a small dependence on the type and energy of beam ions, and the total fission reaction yields are a linear function of the beam energy, rising with increasing energy. The measurements have been carried out at an external beam of the Dubna Synchrophasotron.

2. Experiment

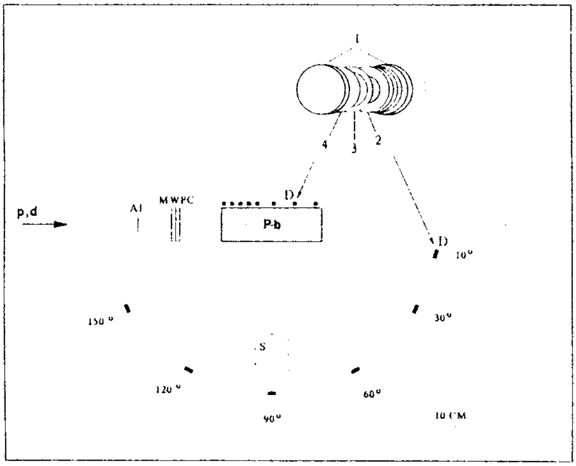

Fig.1. A layout of the experimental set-up and fission detector construction: Pb — lead target, D — fission detector, Al — aluminium disc of beam monitor, S — concrete cone, 1 — cadmium capsule, 2 — backing with a fissionable layer, 3 — SSNTD, 4 — polyethylene disc

The measurements were made at five energies of protons and six energies of deuterons over a range of 1–3.7 GeV. A layout of the experiment and the fission detector construction are presented in Fig.1. The detectors were placed on the side surface of the lead target along its axis and sometimes on the front and back surfaces and at a radius of 1 m from the target center at various angles of 10°, 30°, 60°, 90°, 120°, and 150° to the beam trajectory. For the detectors located on the target surface, the distance between the surface and the fissionable layers was on the average 1–2 mm. The fission detectors consisted of two groups: (1) ^{232}Th, ^{235}U, ^{238}U, ^{237}Np and (2) only ^{209}Bi layers. For each group of detectors we carried out two separate measurements: (1) on the target surface and (2) at different angles. Such detector sets and measurements were undertaken to get optimum statistics for all nuclides and detector positions. A small set of thin-film breakdown counters, TFBC, [2] coupled to the fission layers was used for an operative control and the determination of an optimum fluence of beam ions. To reject the thermal neutron background, all the fission detectors were placed inside the cadmium capsules. An additional experiment with a concrete cone was carried out to estimate the contribution of background neutrons. For angular measurements, the neutron background corrections were 21 ± 4, 12 ± 3, 7 ± 2 and $1 \pm 1\%$ for ^{237}Np, ^{238}U, ^{232}Th, and ^{209}Bi, respectively. The correction for ^{235}U was much higher, and these detectors were used only in measurements on the side surface of the target.

3. Method

The fission events were registered by detecting fission fragments with the SSNTDs made of 6 μm polyethyleneterephtalate film, PETP, and placed close to the fissionable layers. The layers made on aluminium backings were 1 mg/cm^2 thick and 11.3 mm ($S = 1.0$ cm^2) in diameter. The inhomogeneity of the layer thickness was smaller than 10 % and the layer mass uncertainty was 2–3 %.

After the fission detectors had been irradiated in the neutron field of the extended lead target, the SSNTDs were etched in KOH solution at a temperature of 60 ± 0.1 °C for 60 minutes. The next step was a track counting by an automated spark counter [3]. The linearity range extended up to 4000 counts/cm^2, and the maximum fission fragment fluence was 20000 fragments/cm^2. The total methodical error of fragment counting was found as 6–8 %. The critical angle of fission fragment detection by the SSNTD measured with a ^{252}Cf source, $\theta_c = 29°$, corresponded to the registration efficiency $\eta = 0.515$ for isotropic emission. Moreover, additional experiments gave a threshold value of nuclear fragment charge, $Z_{\mathrm{th}} \cong 5$, above which the nucleus could be detected. This value is much smaller than the fission fragment charges, and one can assume the independence of the registration efficiency of the kind of studied fissionable nuclei and the variation of the mass distribution and kinetic energy of fission fragments with increasing neutron energy. At the same time, such a value of Z_{th} is high enough to allow a large fraction of light-mass fragments from hydrogen to boron to be rejected. These fragments have a maximum yield in inelastic interactions of intermediate energy hadrons with nuclei of detector materials, fissionable layers and SSNTD film. Moreover, even for light-mass nuclear fragments with $Z > Z_{\mathrm{th}}$, the registration efficiency continues to be very low due to a small energy deposition for the largest fraction of fragments, $dE/dx < (dE/dx)_{\mathrm{th}}$. Taking into account this fact and also some special methods of decreasing a nuclear fragment share in a detector count (orientation of fissionable layers to a neutron source and the use of polyethylene foils between one side of the SSNTDs and the other detector materials), we optimized experimental conditions and practically rejected this source of background.

In the general case, the angular distribution of fission fragments is not isotropic and depends on the type of nuclides and the energy of neutrons. Our measurements in the spallation neutron field for different orientations of the fissionable layers showed a maximum of the anisotropy effect for bismuth layers placed at small angles to the beam axis, where the energy spectrum of neutrons was the hardest one. The ratio of counts for forward, N_F, and backward, N_B, fission fragment emission was obtained as

$$\alpha = N_F / N_B = 1.33 \pm 0.07.$$

Coming to a large angle range, the difference between N_F and N_B became only 10–15 %. For the other nuclides, ^{232}Th, ^{238}U, and ^{237}Np, the ratio values of α were 1.13 ± 0.07, 1.10 ± 0.06 and 0.95 ± 0.05, respectively in all angular range. Moreover, it was found that for all the studied nuclides the counts for the layers orientated at 90° between the layer normal and the incident neutron trajectory were approximately equal to half the sum of the values of N_F and N_B. The study of detector counts on the side surface of the lead target did not show any difference between N_F and N_B within experimental errors of 6–9 %. Therefore, the influence of angular anisotropy on fission detector counting was usually small and could be taken into account by a simple formula used to calculate the number of fission reactions in 1 mg of nuclide

$$N_f = (N_F + N_B) / 2\eta = N_B (1 + \alpha) / 2\eta,$$

where the experimental values of N_F and N_B were taken for 1 mg fissionable layer. The experimental method is described in more detail in Ref.4.

4. Beam Monitoring

A number of incident ions was obtained by means of monitor reactions $^{27}\mathrm{Al}(p, X)^{24}\mathrm{Na}$ and $^{27}\mathrm{Al}(d, X)^{24}\mathrm{Na}$. The cross section of the first reaction in a few GeV energy region of protons is known with an uncertainty of 5 % [5, 6] and was taken as 10.8 mb at $E_p = 1.0$ GeV and 10.0 mb at higher energies. For deuterons in the same energy region, the cross section of ^{24}Na production was measured only in Ref.7, where the authors gave a value of 15.25 ± 1.50 mb at $E_d = 2.33$ GeV. Our relative measurements with deuterons showed the energy dependence of the cross section value similar to that obtained with protons. The experimental error of proton beam monitoring was the smallest one: 2 and 3.5 % at $E_p = 2.0$ GeV and 3.17, 3.65 GeV, respectively, and it increased to 10 % at $E_p = 1.0$ GeV. For the deuteron beam, this error was on the average larger: ~ 5 % at $E_d = 2.0$ and 3.76 GeV and up to 20% at other energies.

The aluminium discs were placed 40 cm upstream the target. The beam was focussed by means of two MWPCs, and the beam profile was Gaussian in shape with a width of ~ 30 mm at a 0.1 maximum level.

5. Results

The results of measurements with the fission detectors on the side surface of the extended lead target ∅ 20 × 60 cm at different energies of proton and deuteron beams are shown in Figs. 2—4. The data are presented as a number of fission reactions in a 1 mg/cm^2 layer with a square of 1 cm^2 per incident ion. Basic conclusions are the following:

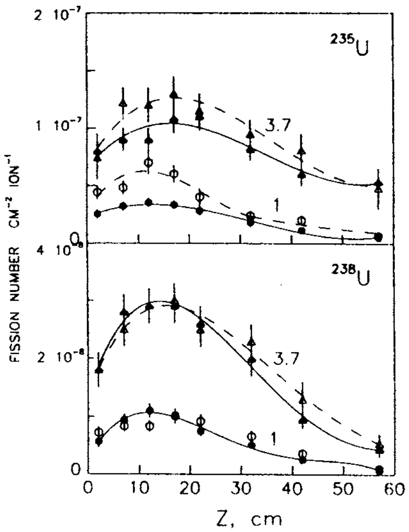

Fig.2. Fission reaction distributions along the side surface of the target for ^{235}U and ^{238}U measured with 1.0 and 3.7 GeV protons (close symbols) and deuterons (open symbols). The curves are a polynomial fit

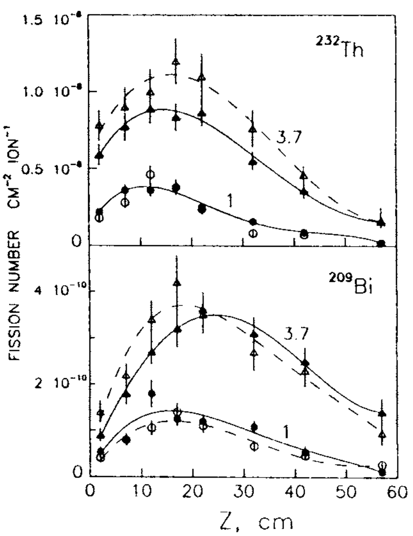

Fig.3. The same as in Fig.2 but for ^{232}Th and ^{209}Bi

1. A small shift of a maximum position from the front surface of the target is observed for all nuclides with increasing energy;

2. In comparison with protons, deuterons give a higher yield of the fission reaction for ^{235}U and ^{237}Np (~ 20–30 %) over all the energy range;

3. For actinides, the distribution shape weakly depends on the type of beam ions and ion energy.

The measurements of fission reaction yields on different surfaces of the target showed that for the studied actinides approximately 80 % of the total number of fission reactions took place in the layer on the side surface independently of the energy of a proton beam. The share given by the layer on the front surface decreases from ~ 19 to ~ 13 % with increasing proton energy in the investigated region, and simultaneously the contribution of the back surface layer goes up to 3–4 %. For bismuth, a relative contribution of the layer surrounding the side surface is even higher and the share of the back surface layer increases stronger than for actinides with rising proton energy (Figs. 5, 6). The angular dependence of fission reaction yields in 1 mg/cm^2

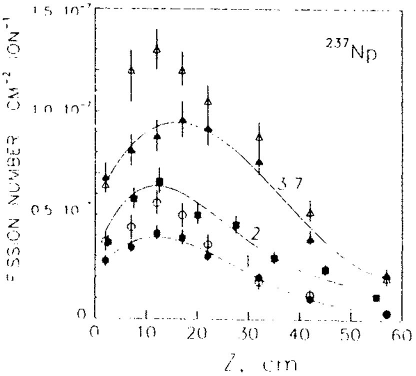

Fig.4. The same as in Fig.2 but for ^{237}Np and additional energy of protons 2.0 GeV

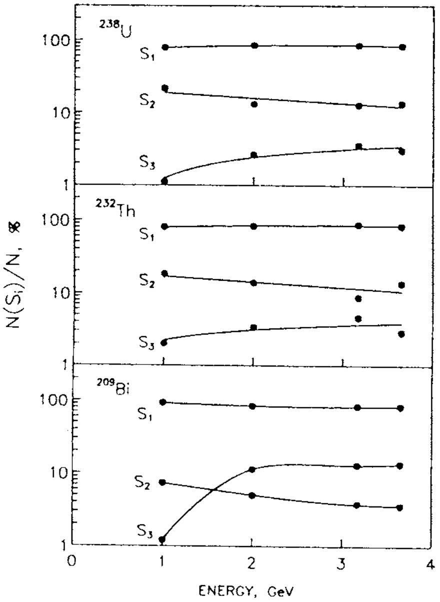

Fig.5. Relative fission reaction yields on different surfaces of the lead target for 1 mg/cm^2 layers of ^{209}Bi, ^{232}Th, and ^{238}U as a function of proton energy: S_1, S_2, S_3 — side, front and back surfaces, respectively

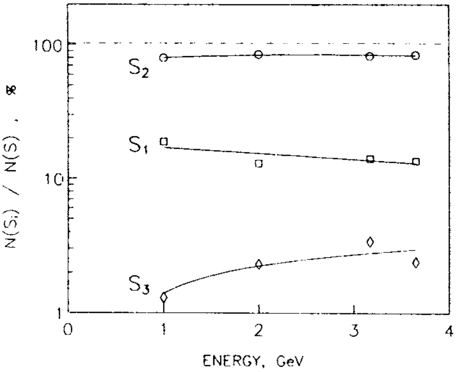

Fig.6. The same as in Fig.5 but for ^{237}Np

of a spherical layer has a small deflection from the isotropic distribution for actinides, Figs.7, 8. But for ^{209}Bi having a high energy threshold of fission, the reaction yield has a strong maximum at small angles as shown in Fig.7 for a few values of ion energy. The shape of angular distributions weakly depends on the type of beam ions.

The analysis of the total yield of fission reactions as a function of ion energy has shown that for all the investigated nuclides, the yields can be described by a linear dependence on the energy of bombarding particles, Figs.9, 10. For actinides, deuterons

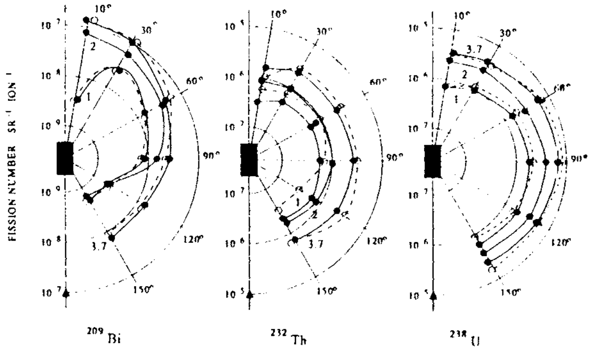

Fig.7. Angular distributions of fission reaction yields for 1 mg/cm^2 layers of ^{209}Bi, ^{232}Th, and ^{238}U measured with 1.0, 2.0, 3.7 GeV protons and 1.0, 3.7 GeV deuterons: the symbols are the same as in fig.2, and the curves are a polynomial fit

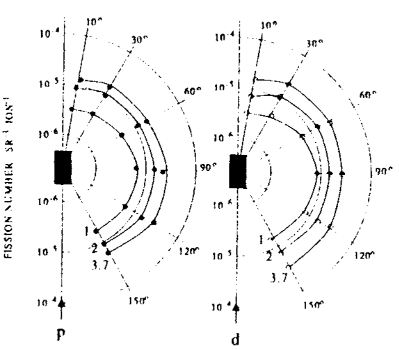

Fig.8. The same as in Fig.7 but for ^{237}Np and 1.0, 2.0 and 3.7 GeV protons and deuterons

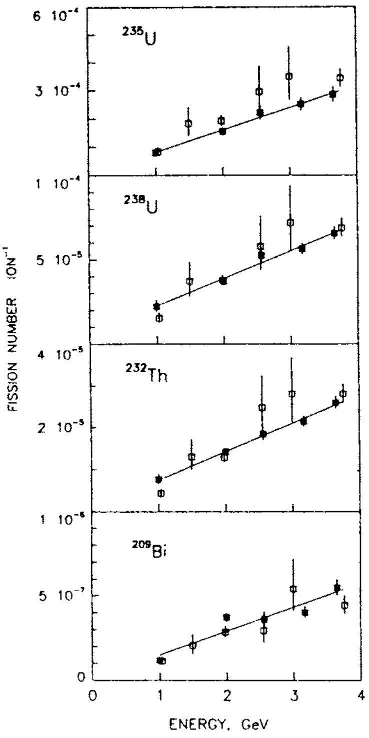

Fig.9. Total fission reaction yields in 1 mg/cm^2 spherical layers of ^{209}Bi, ^{232}Th, ^{238}U, and ^{235}U per incident ion as a function of ion energy: closed symbols — for the proton beam, open symbols — for the deuteron beam; the curves are a linear fit to the proton data

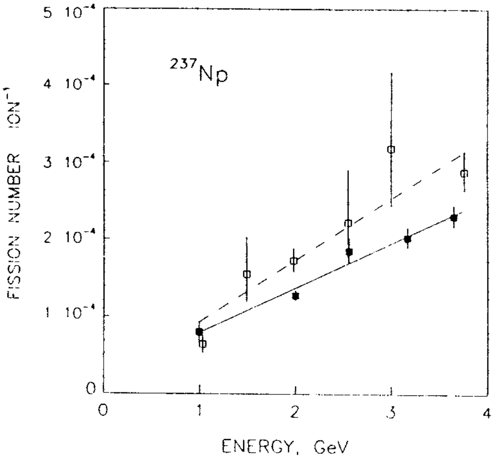

Fig.10. The same as in Fig.9 but for ^{237}Np layer

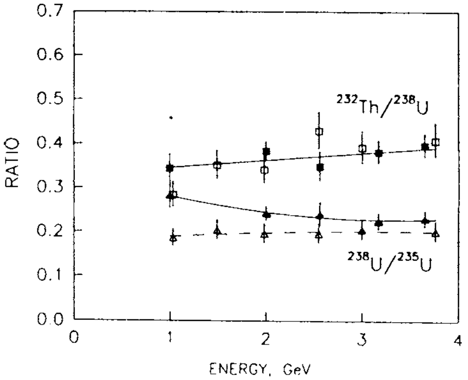

Fig.11. The energy dependence of the total fission reaction yield ratios ^{232}Th/^{238}U and ^{238}U/^{235}U: closed symbols — for the proton beam, open symbols — for the deuteron beam; the curves are a polynomial fit

produce, on the average, a higher reaction yield than protons at the same energy. For ^{235}U and ^{237}Np layers, this difference reaches ~ 25 and 30 %, respectively. At the same time, this relation becomes opposite for ^{209}Bi. The lowest level of experimental errors was in the measurements with 2 GeV protons and 2 GeV deuterons. The data obtained at 2 GeV are given in the Table (the error of beam monitoring has not been included). The ratios of the total fission reaction yields, ^{232}Th/^{238}U and ^{238}U/^{235}U, are given as a function of beam energy in Fig.11. The first ratio weakly depends on the type of ions slightly going up with increasing beam energy. However the second ratio obtained with the proton beam is larger than the same one measured with deuterons for all the studied energy region.

Table. Total fission reaction yields in 1 mg/cm^2 layers of fissionable nuclides surrounding the spallation neutron source per 2 GeV incident ion

Nuclide	p	d
^{209}Bi	$(3.70 \pm 0.27)\cdot 10^{-7}$	$(2.85 \pm 0.35)\cdot 10^{-7}$
^{232}Th	$(1.41 \pm 0.07)\cdot 10^{-5}$	$(1.28 \pm 0.11)\cdot 10^{-5}$
^{238}U	$(3.68 \pm 0.18)\cdot 10^{-5}$	$(3.75 \pm 0.30)\cdot 10^{-5}$
^{235}U	$(1.53 \pm 0.15)\cdot 10^{-4}$	$(1.92 \pm 0.18)\cdot 10^{-4}$
^{237}Np	$(1.27 \pm 0.07)\cdot 10^{-4}$	$(1.73 \pm 0.16)\cdot 10^{-4}$

6. Conclusion

The study of the fission reaction yields of ^{209}Bi, ^{232}Th, ^{235}U, ^{238}U, ^{237}Np in the spallation neutron field of the lead target $\varnothing 20 \times 60$ cm bombarded by a 1–3.7 GeV proton/deuteron beam has shown a small dependence of the spatial-angular distribution shape on the type and energy of beam ions with an approximately linear increase of the reaction yields with increasing ion energy. More than 80 % of the total fission reaction yield in a layer surrounding the target occurs on the side surface with a maximum at $Z = 12$–17 cm for actinides and at $Z = 17$–25 cm for bismuth. For actinides, the angular distributions have a small difference from the isotropic one, and for bismuth they have a strong maximum of the fission reaction yield at small angles. The use of a deuteron beam allows one to reach a larger reaction yield (25–30 %) for the actinides having a low energy threshold of the fission reaction, such as ^{235}U and ^{237}Np. But there is no benefit to use deuterons instead of protons for actinides with higher thresholds of fission as ^{232}Th and ^{238}U. For other heavy mass nuclides, for example, bismuth, protons generate higher yields of fission than deuterons of the same energy.

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