

Nuclear fission cross sections induced by deuterons of 4 GeV

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Nuclear fission cross sections induced by deuterons of 4 GeV were measured by the solid state nuclear track detectors technique. The experiments were carried out at the Nuclotron accelerator of the Joint Institute for Nuclear Research (JINR) (LHE), Dubna. Heavy targets such as ^{232}Th , ^{235}U , and ^{238}U as well as ^{209}Bi and ^{197}Au were irradiated by 10^{11} deuterons. The cross sections for radioactive targets were estimated as $\sigma_f = 1153 (\pm 198)$, $1666 (\pm 430)$, $1453 (\pm 350)$ mb for ^{232}Th , ^{235}U , ^{238}U , while for ^{209}Bi and ^{197}Au as $\sigma_f = 206 (\pm 46)$, $92 (\pm 23)$, respectively. The comparison of these results with proton-induced fission systematics shows higher fission cross sections when deuterons are used as projectiles. The part of the reaction cross section that fission acquires is found to be much higher for actinide targets than for targets of lower atomic numbers.

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I. INTRODUCTION

After inelastic scattering and charge exchange reactions, in the next order of complexity are nucleon transfer reactions, in which one or more nucleons are transferred from the projectile to the target (stripping reactions) or from the target to the projectile (pickup reactions). A typical one-nucleon transfer reaction is the (d,p) reaction, which is essentially the transfer of a neutron from the projectile to an unfilled single-particle state of the residual nucleus [1]. For that reason, deuteron reactions have been studied since the 1950s.

Oppenheimer and Phillips were the first to use deuteron projectiles at energies of a few MeV. They found that reaction thresholds lie below the Coulomb barrier, which corresponds to the repulsion between deuterons and the target nucleus [2]. Due to these particular reactions, deuterons have been extensively used in the study of fission thresholds, especially when fission barriers are higher than binding energies [3].

Fission cross section data are used in nuclear physics basic research to provide information on the evolution of different mechanisms of nuclear reactions at GeV energies. At these energies, the reactions are understood as a two-step process: Primary collisions induce an excited hot prefragment which then deexcites by particle emission and fission [4–6]. The existing models are based on a mechanism of reaction including spallation-evaporation or spallation-fission residue. Fission could then be the ending stage of both mechanisms and therefore is influenced by both the excitation energy and the angular momentum transferred from the projectile to the target. While a lot of experiments have been performed over the last 20 years with proton projectiles, the replacement of the proton beam by deuterons (or heavier beams) allows studying fission under increasing heat of the target. The increase of excitation

energy has led to the study of the dependence of fission cross sections on projectile energy and has been proven to be a useful tool for the study of fission barriers [6], the quantity of energy and angular momentum transferred to the target, and finally the part of the reaction cross section attributed to fission cross sections [7].

In recent years, deuteron reactions have attracted new attention for their applications associated with accelerator-driven sources (ADS) including extrapolations to nuclear waste transmutation and nuclear energy production. Experiments using protons at few GeVs, deuterons, and alpha particles as projectiles have shown an enhanced neutron production when irradiating thick targets [8–12]. Recent calculations of the Energy+Transmutation collaboration [13] proved that in ADS, a beam of 1.5 GeV deuteron projectiles results in a higher neutron production rate than a proton beam and therefore a higher energy gain [14].

A well-ordered database is an essential requirement for the nuclear physics research community. Such credible databases also act as a bridge between science and technology. An enrichment of the nuclear data sources for each reaction, especially in nuclear fission data, is needed, because they can be used as input to Monte Carlo calculations or for the comparison of calculations with experimental data [15]. Therefore, reliable data on fission cross sections are welcomed, at GeV energies, especially for heavy targets proposed for spallation sources and nuclear waste transmutation. Such a target is ^{232}Th , which is the most abundant isotope in nature, and for this reason it was proposed by Rubbia *et al.* for energy production by ADS [16]. To date there have been a number of experiments using $^{\text{nat}}\text{Pb}$ and ^{209}Bi targets [17, 18].

The present work deals with fission cross section experimental data deriving from deuteron-induced fission at 4 GeV. Actinide targets such as ^{232}Th , ^{235}U , and ^{238}U targets were used. ^{209}Bi and ^{197}Au , having higher fission barriers than actinides, were also studied. A comparison of deuteron results with available data for proton beams is presented. The part of the reaction cross section due to fission is discussed. A

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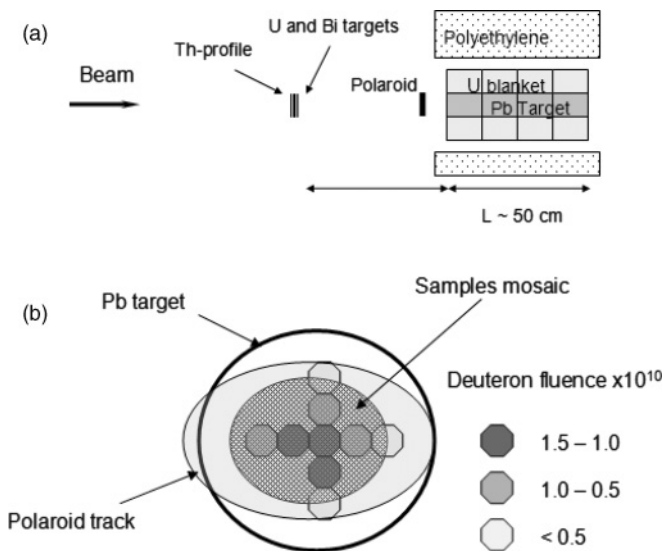


FIG. 1. (a) The experimental setup used during irradiation of E+T assembly with deuteron beam; (b) the mosaic of ^{232}Th samples placed in front of the Pb-target for beam profile in comparison with the beam track obtained using Polaroid film. The deuteron fluence determined by each ^{232}Th sample used for the beam profile is presented.

systematic trend of the fission cross section as a function of the target Z^2/A is investigated and empirical conclusions on the total fraction of the reaction cross section that is due to fission are presented.

II. EXPERIMENTAL

A deuteron beam with energy 4 GeV and intensity of the order of 10^{11} ions was delivered by the Nuclotron accelerator, Joint Institute for Nuclear Research (JINR), Dubna. This experiment was part of the ADS studies in the frame of the international collaboration “Energy plus Transmutation” (E+T) [13]. The E+T assembly consisted of a lead target, 50 cm in length and 8 cm in diameter, surrounded by a natural uranium-blanket fuel (Fig. 1).

Cross section determination was achieved by using fissionable targets such as ^{232}Th , ^{238}U , ^{235}U , ^{209}Bi , and ^{197}Au . The targets had a diameter of 1.2 cm and were manufactured at CNRS, Strasbourg, France, by evaporation of the target on Lexan foils [19]. Lexan sheets were also used for the detection of the fission fragments. The mass of the radioactive targets was measured using alpha and gamma spectrometry at the Laboratory of Nuclear Physics of the Aristotle University of Thessaloniki, while the mass of ^{209}Bi and ^{197}Au targets was measured using Rutherford backscattering analysis at the Tandem Van de Graaff accelerator facility of the Institute of Nuclear Physics, NCSR Demokritos, Athens. Mass determinations by α and γ spectrometry were very close to each other (Table I). The uncertainties given in Table I were estimated taking into account the contributions due to the calibration of the source (3.7%), counting statistics (2%–5%) and ray intensity (1% for γ and 2%–4% for α rays). The high uncertainty observed in the case of γ -spectrometry measurement of ^{238}U is due to the uranium content in the

TABLE I. Mass determination (μg) for a typical sample of each radioactive target.

	γ spectroscopy	α spectroscopy			
		^{232}Th	^{238}U	^{235}U	^{234}U
^{232}Th target	134 ± 6	126 ± 6	7 ± 1		
^{238}U target	95 ± 12		91 ± 7		
^{235}U target	104 ± 6		<9	99 ± 14	1.3 ± 0.2

background spectrum. In the α -spectrometry measurement of ^{235}U the uncertainty is mostly due to computational errors when a fitting process is applied to the triple peak of the measured α spectrum. For the same reason, the minimum detectable value of ^{238}U impurity in the ^{235}U targets is also high. The percentage of ^{238}U in ^{232}Th targets was around 5% and in ^{235}U targets less than the minimum detectable value of 9%. In ^{238}U targets the admixtures of ^{235}U were 0.4% according to data given by the providers. Since the determined thickness is considerably lower than the range of the heaviest fission fragments in the given targets, they are suitable for fission cross section determination.

A mosaic of ^{232}Th samples was placed directly into the beam at a distance of about 50 cm from the E+T assembly. An identical mosaic of ^{235}U , ^{238}U , ^{209}Bi , and ^{197}Au samples was positioned back to back to the ^{232}Th samples. The mosaics were adjusted according to the Pb-target center (see Fig. 1). Thorium samples were also used in order to study the profile of the deuteron beam. The experimental results matched the data obtained using a Polaroid film (see Fig. 1). The integrated deuteron fluence was measured by the Nuclotron operators, using ionization chambers. The Lexan detectors after irradiation were properly etched and the track density was measured under an optical microscope.

III. RESULTS AND DISCUSSION

The experimental values of ^{235}U and ^{238}U fission cross sections are based on about 10^4 binary events registered in three different samples positioned at various places on the mosaic. About 10^3 events were counted from nine samples in the case of ^{232}Th , used for the beam profile. ^{209}Bi and ^{197}Au fission cross sections were obtained by counting about 400 fission fragments in total for each target. The measurements were corrected for registration efficiency, which for fission fragments in Lexan is 96%. The bulk material removed from the detector’s surface during etching was $0.5 \mu\text{m}$ and subsequently a loss of 4% of tracks was added to the total tracks counted.

Another correction of the results for fissions induced by the backscattered neutrons from the spallation target was applied. Fission events measured were coming from (d, f) reactions and the contribution of any other fission, induced by gamma, neutron, proton, and elementary particles (π , μ) backscattered from the E+T assembly. Track density caused by the E+T assembly backscattered particles (>97%) was attributed mainly to neutron-induced fission. Typical calculated neutron spectra emitted by the assembly using

TABLE II. Contribution of the relative uncertainties (%) involved in fission cross section estimations.

Source	^{232}Th	^{238}U	^{235}U	^{209}Bi	^{197}Au
Mass of ^{232}Th	4	4	4	4	4
Target mass		7	6	9–12	11–15
Number of tracks (^{232}Th) ^a	9–21	9–11	17–21	11–19	13–19
Number of tracks (target)		14–21 ^a	11–17 ^a	5–7	5–7
Beam fluence	9				
$\sigma_{\text{Th}(d,f_{\text{total}})}$		17	17	17	17
Overall uncertainty of σ target (d,f)	19–27	24–29	27–30	22–28	24–29

^aThe uncertainties in measured track number include the correction for fissions induced by backscattered neutrons and fissions induced by deuterons in impurities.

the DCM-DEM code [20,21] show an ascendancy of neutrons emitted in the energy range from 0.1 up to 300 MeV (peaking around 1 MeV) with a mean energy of 6.5 MeV. The percentage of fast-superfast neutrons ($E_n > 2$ MeV) was 18.4% of the total neutrons emitted, while neutrons with energies higher than 30 MeV were less than 0.4% [22]. Considering the geometrical factor of the experimental setup, corrections to track density due to neutron contribution were applied to ^{232}Th , ^{238}U , and ^{235}U measurements. In the cases of ^{209}Bi and ^{197}Au no corrections were made, since their fission starts practically at ~ 60 MeV. The contribution of the backscattered neutrons above 60 MeV was estimated to be around 0.1% [22].

An additional correction of the fission events measured was applied for fissions produced in impurities of ^{238}U nuclei in ^{232}Th and ^{238}U in ^{235}U . The number of fission events induced by deuterons on ^{238}U nuclei was estimated taking into account the deuteron fluence determined by the beam profile [see Fig. 1(b)] and a cross section of 1.5 b. In both targets, 2%–6% of the measured fission was due to the ^{238}U impurities. Those contributions are smaller than the track counting uncertainty and comparable to the uncertainty in mass determination. A detailed analysis of the uncertainties involved in fission cross section calculations is presented in Table II. In the case of ^{232}Th , the reported fission cross sections are the mean values of nine samples weighted by the uncertainty of each sample. For the other targets, the reported values of fission cross section were estimated relative to the $\text{Th}^{232}(d,f)$ cross section, taking into account the ratio of fission events measured per target nuclei to ^{232}Th nuclei. The determined values are the weighted mean values of two to three samples.

The reported cross sections in Table III are inclusive values of (d,f) as well as of (d,pf), ($d,d'f$), etc., reactions, and are thus referred to as “total fission cross sections.” The nuclear track technique does not allow distinguishing the (d,f) process from all other reactions such as (d,pf), ($d,d'f$), etc. However, it could be possible to correct fission cross sections for the contribution of (d,pf), ($d,d'f$), etc., reactions if the cross sections of these reactions were known at the region of energies studied.

The total fission cross sections deduced by this experiment are given in Table III. Their comparison to ^{232}Th cross sections is presented in column 2 in order to show the low fissility of

TABLE III. Total fission cross section of U, Th, and Bi, Au targets induced by 4 GeV deuteron beam.

Nucleus	$\sigma_{(d,f)}$ (mb)	$\sigma_{(d,f)}/\sigma_{(d,f)\text{ofTh}}$	$\sigma_{(p,f)}$ (mb) ^a
^{238}U	1453 ± 350	1.26 ± 0.22	1000
^{235}U	1666 ± 430	1.44 ± 0.28	1100
^{232}Th	1153 ± 198	1	800
^{209}Bi	206 ± 46	0.18 ± 0.03	160
^{197}Au	92 ± 23	0.08 ± 0.02	80

^aValue corresponding to Prokofiev’s systematics for proton beams (Ref. [28]).

subactinides. The fission cross sections of ^{209}Bi and ^{197}Au at the energy range in which they were studied are small compared to ^{232}Th , ^{238}U , and ^{235}U fission cross sections, and they confirm the observation that they are of the order of one tenth of the corresponding ^{232}Th cross section [3]. In particular, their low cross sections are connected to the difference of fission barrier B_f , relative to the neutron binding energy, S_n . Indeed, the heights of the fission barriers for ^{209}Bi and ^{197}Au are about 22 MeV, while those for actinides are about 6 MeV [23].

Even though cross sections for the elements studied in the present work are useful for their applications in nuclear theory and ADS configuration, limited experimental data are available in the literature at GeV energies. Limited data are presented for ^{209}Bi , ^{232}Th , and ^{238}U at lower energies, yet close to those of the current experiment ($E_d = 1.0\text{--}2.1$ GeV) [3,4,24–27]. Furthermore, there are no published fission cross section results at the same region of energy for ^{235}U and ^{197}Au . The results of the present work can only be compared in the case of ^{209}Bi with recent published data of deuteron-induced fission at 4 GeV [26] and are found to be in good agreement.

Fission cross sections induced by deuterons present higher values than those obtained for protons (Table III, column 3). Due to the abundant experimental data existing for proton-induced fission, a representative value was taken for comparison with deuterons, according to the fitting applied by Prokofiev [28] for the same projectile energy range. The difference between proton and deuteron fission cross sections varies from about 50% to 15% depending on the target-projectile system. Larger differences of about 60%–70% have been observed in ^{238}U targets at lower projectile energies (protons-deuterons) [29]. Higher fission cross sections have been measured for heavier-than-deuteron projectiles as early as the 1950s for energies up to about 200 MeV [30,31]. The same observations were reported at higher energies by bombarding heavy targets with alpha particles [3,31–33] or ^{12}C [34,35]. This behavior was interpreted within the context of the excitation energy transferred to the target, which is a function of projectile mass and energy. Fission barriers can easily be penetrated as a consequence of the higher excitation energies transferred by heavier projectiles [36]. The data collected up to now provide evidence that at high projectile energies, there is an upper limit to the excitation energy transferred to the target, depending on the projectile mass. Similar behavior is observed for the angular momentum transfer [3].

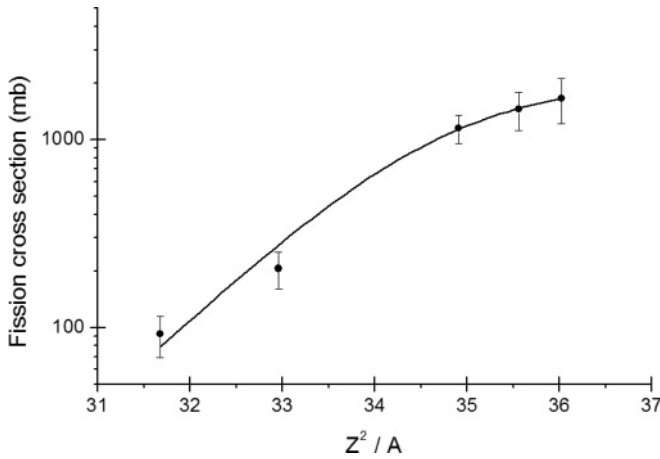


FIG. 2. Fission cross sections induced by 4 GeV deuterons versus the Z^2/A of the target. The full line corresponds to the empirical equation (1).

The study of fission cross sections could be used to learn more about their correlation with the fissility parameter. The fissioning nucleus is not a precisely known system since it depends on the mechanism of its formation. As it has been established over recent years, the reaction mechanism takes place in a two-step process at high energies [4,24]. Primary collisions induce the rapid formation of an excited target prefragment, which then deexcites by particle emission and/or fission. This leads to either a spallation-evaporation or a spallation-fission residue. The usual presentation of fission cross sections as a function of Z^2/A is based on the liquid drop theory, which is well established for low energy fissions [2], and remains until today a characteristic classification of fission events. In addition, it is difficult to separate the dependence of fission cross sections on Z^2/A from E^* , but there is evidence that Z^2/A is the predominant factor. Therefore in Fig. 2 the fission cross sections are presented as a function of target Z^2/A , and correspond to a fissility parameter of 0.63–0.71 for the targets of this study. The characteristic groups of actinides and subactinides are clearly distinguished, indicating a drop of fission cross sections for targets of lower fissility. The experimental data could be fitted by an empirical equation such as

$$\sigma_f(\text{mb}) = \sigma_{\text{max}} \frac{1}{1 + \exp[-k \cdot (\frac{Z^2}{A} - C)]}. \quad (1)$$

The derivative of Eq. (1) with respect to Z^2/A presents a critical value at $C = 34.7 \pm 0.2$ and reveals the different behavior of subactinides. This result is compatible with data available since the early days of nuclear physics, where a separation of nuclei characteristics was set at $Z = 90$ examining nuclear fission for $Z > 90$ and $Z < 90$ [2]. The constant k is equal to 1.1 ± 0.1 and the maximum value of the fission cross section is 2091 ± 231 mb. Comparing the results deduced when deuterons are used as projectiles with proton ones available in the literature [37,38], it is concluded that they have a similar behavior.

One of the fundamental observables in nuclear physics is the reaction cross section, which includes all inelastic

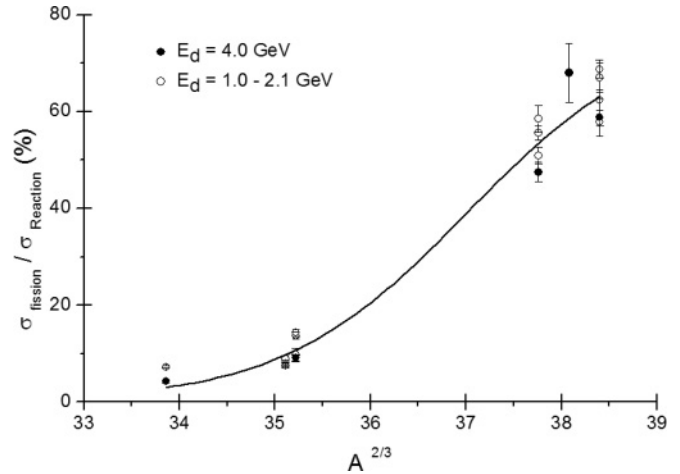


FIG. 3. The ratio of fission-to-reaction cross sections induced by 4 GeV deuterons versus the $A^{2/3}$ of the target nuclei (full circles). Open circles correspond to the data available in the literature for the same range of energies [3,4,24–27]. The full line is the fitting result by the empirical equation (1).

processes. The section part that the nuclear fission contributes to the inelastic channel is of great interest, as the rest of the inelastic channel occurring to fragmentation mechanisms has the tendency to increase with increasing projectile energy. For fixed energy, heavier targets reveal higher fission cross sections and take the largest part of the reaction cross section. This observation is presented in Fig. 3 as the ratio of fission to reaction cross section. In this figure, fission cross section results from this work are presented together with all data available in literature at energies close to 4 GeV [3,4,24–27]. The reaction cross section was calculated according to the soft spheres model as $\sigma_R = \pi r_o^2 (A_T^{1/3} + A_p^{1/3} - b_o)$, with $r_o = 1.48$ fm and $b_o = 1.32$ fm [39,40], where A_T and A_p indicate the target and the projectile mass, respectively.

Figure 3 illustrates the decreasing role of fission in the reaction cross section as one proceeds to lower Z targets. In the case of actinides, fission accounts for 60%–70% of the reaction cross section, while in the case of ^{209}Bi , ^{208}Pb , and ^{197}Au for only 10%–4%. Comparing to the data given in literature for lower energies, we realize that the part of the reaction cross section which is taken by fission of heavy targets is lower than observed at lower energies, at the MeV range [41], while for subactinides it is almost the same. This means that the influence of excitation energy that increases with bombarding energy plays an important role mainly in the case of heavy targets. In subactinides the residual nuclei left after the cascade stage are lighter, while the fission barriers are higher. As a consequence, the probability of their fission decreases instead of increasing.

Fitting the data for σ_f/σ_R versus $A^{2/3}$, a similar empirical expression to Eq. (1) can be derived. The constant k is equal to 1.0 ± 0.1 and the maximum observed value of the total fraction of the reaction cross section due to fission is $79 \pm 11\%$. The turning point separating nuclei that “like fission and don’t like fission” [39] is presented at a critical value of $C = 37.2 \pm 0.3$. This value corresponds to A values of 225 ± 3 . Taking into

consideration the critical value estimated in Eq. (1), the Z value should be 88–89, corresponding to the natural radionuclide of actinium and radium. For those isotopes, the fission cross section should be half of the maximum value determined [Eq. (1)], i.e., around 1050 ± 150 mb. Indeed, similar values for ^{226}Ra fission were determined experimentally using deuteron and alpha particles as projectiles [42].

The behavior of the fission-to-reaction cross section ratio given in Fig. 3 is very similar to the one observed when protons were used as projectiles [39], indicating that fission induced by deuterons has no qualitative differences relatively to fission induced by protons, the only difference being the absolute value of the cross sections observed. Although Eq. (1) is an empirical approximation, some remarks, interesting for their physical content, can be made, such as the saturation of fission process for $Z > 92$ and the threshold of rapid increment after the double magic nuclei of ^{208}Pb .

IV. CONCLUSION

Cross sections of deuteron-induced fission were measured for actinides and lighter nuclei. Systematic studies of fission cross sections at GeV energies are of importance for nuclear reaction theory as well as for their applications in accelerator-driven systems. Although there is an abundance of published results regarding proton-induced fission at high energies, very few studies involve deuteron-induced fission, probably because of the limited availability of deuteron beams. The findings of the present work provide data concerning fission cross sections at 4 GeV deuterons for ^{238}U , ^{232}Th , and ^{209}Bi , while at the same time the values for fission cross sections of ^{235}U and ^{197}Au are presented, to the best of our knowledge, for the first time in the literature.

Total fission cross sections induced by deuterons at 4 GeV feature the same characteristics as fission induced by protons of the same energy. Fission induced by deuterons exhibits higher cross sections compared to that induced by protons. The observed difference is higher for actinides than for subactinides. As the fission production is attributed to a residue of target left after a two-step mechanism, in the case of subactinides the residual nuclei left after the cascade stage are lighter, while their fission barriers are higher. As

a consequence, their fission probability decreases instead of increasing. For the same reason, the part of the reaction cross section corresponding to the fission process indicates that deuteron projectiles affect mainly the heavy targets, while in the case of subactinides it remains almost the same. Comparing the results of the present work with previous fission studies at lower energies, it is concluded that fission at lower energies contributes larger parts to the reaction cross section than at higher energies. Taking into account that inelastic cross sections, which are practically equal to the reaction cross section, are almost constant at GeV energies it is concluded that when increasing the beam energy, other reaction mechanisms such as fragmentation reactions tend to cover the difference between fission and reaction cross section.

It is useful to note at this point that a deuteron interacts mostly with either a proton or neutron due to its low binding energy. Although deuteron inelastic cross sections are the same as those of protons and neutrons [22,25,43], fission cross sections induced by protons present higher fission cross sections than those induced by neutrons [28]. A significant difference is observed between proton- and neutron-induced fission cross sections for both ^{209}Bi and ^{208}Pb at energies of tenths of MeV [44]. An explanation for this observation is given in the same publication as “the fission probability depends strongly on the charge of the fissioning nucleus in Pb-Bi-Po region and the increase of the mean charge of the fissioning nuclei caused by the incident proton results in a fission probability which is higher than for the incident neutron” [44]. In contrast, at high energies, at GeVs, proton and neutron fission cross sections approach each other, as concluded by the n -TOF collaboration experiments for ^{208}Pb and ^{209}Bi [45]; a similar result is presented for ^{232}Th fission by the same collaboration experiments [46].

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