Fast proton-induced fission of ^{238}U from threshold to 40 MeV

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Abstract. Fast proton-induced fission of ^{238}U has been investigated from the threshold up to 40 MeV. Fission variables such as cross-sections, mass distributions, and prompt neutron emission were evaluated. For the analysis, Talys and programs written by authors were used to describe the fission process using Brosa model. As a result, we estimate the contribution of different nuclear reaction mechanisms (direct, pre-equilibrium, compound nucleus) to cross-sections, prompt neutrons emission, and other fission parameters. In the interaction of fast protons with target nuclei, excited residual nuclei by mean of different (p,p'), (p,xn), (p,xp), (p,xα) (x=1,2,...,n) and other processes are formed, due to different nuclear reaction mechanisms. In the case of ^{238}U, excited residual nuclei can also fission, contributing in this way to the investigated variables and isotope production. Theoretical results were compared with previous experimental data found in the literature. The comparison allowed us to extract parameters of the optical potential, fission barrier height and width, and type of nucleus deformation. Cross sections and isotope yields (as Mo, I, Xe and other fragments) obtained in fission process induced by gamma and neutrons were compared with protons case and with available data from the EXFOR database.

1 Introduction

It was demonstrated in theory and experiment that in the fast neutron induced fission proton spectra are obtained. Protons produced by fast neutron reactors with energies up to 20 MeV produce large amount of isotopes with very high neutron cross sections which will affect its functioning and will lead to the activation of walls and vessels around nuclear facility [1,2].

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In fission process of ^{238}U by protons with energy higher than 20 MeV becomes of interest the production of radioisotopes with applications in medicine, electronics, industry and other fields of human activities [3,4]. Fission process induced by fast protons up to 40 MeV energy on ^{238}U was analyzed. Fission observables as cross sections, fragments mass distribution, yields and cross sections of some nuclides of interest, prompt neutrons emission and others observables, characterizing ^{238}U fast protons induced fission were theoretically evaluated and compared with existing experimental data from literature. The present study represents a research proposal for fast protons induced fission investigations and isotopes production at basic facilities from Europe and abroad.

2 Codes and elements of theory

Evaluations of fission variables were realized mainly with Talys code. Talys represents a free software dedicated to nuclear reactions and structure of atomic nuclei calculations, working under Linux. In the mentioned program are implemented all nuclear reaction mechanisms, together with a large database containing spin, parity, energy of nuclear levels, parameters of nuclear states density and of optical nuclear potential (Wood-Saxon) with volume (V), surface (d) and spin-orbit (SO) components respectively, each with real and imaginary part [5]. Compound processes are described by Hauser-Feshbach formalism [5,6], direct mechanism by Distorted-Wave-Born-Approximation (DWBA) [5,7] and pre-equilibrium one by two-component exciton model [5,8]. Protons induced fission can be described by compound processes in the frame of statistical model of nuclear reactions. In this formalism, cross section is [5,6]:

$$\sigma_{ab} = \frac{T_a T_b}{\sum_c T_c} W_{ab} \tag{1}$$

where T are transmission (penetrability) coefficients; a, b are incident and emergent channels respectively; {c} are all open emergent channels; W_{ab} is the width fluctuation correction factor.

Transmission coefficients can be calculated using approximate methods or applying quantum mechanical approach based on reflection factor [9]. In previous works the authors developed codes for the evaluation of transmission coefficients for neutral and charged particles with no approximation, using integral form of regular and irregular Coulomb functions in (n,α) and (n,p) processes [10,11]. For fission process T coefficients are calculated applying the formalism from [5,12]. The sum is take over all open emergent channels.

Widths fluctuation correction factor represents the correlation between incident and emergent channels. At incident energies lower than 0.5 MeV compound processes are dominant, the compound nucleus “forgets” how it was formed. As result correction factor is $W_{ab} = 1$. With the increasing of incident particles, width fluctuation correction factor is slowly decreasing. This factor has a complicated form and in this work it was calculated applying approach from [5,13].

The advantage of using Talys is that all nuclear reaction mechanisms are enabled in calculation. This is important at higher incident energies when other channels like (p,xn), (p,xp), (p,xα) and other (x = 1,2,…,n) are open. These channels can produce significant amount of fissionable excited nuclei which can undergo to fission and in consequence to contribute to the general mass distribution of fission fragment, total fission cross section, neutron emission, isotope production and other fission variables.

At low energy transmission coefficients are calculated for discrete states. With the increasing of energy, number of discrete states is also increasing and therefore it is necessary to consider levels density. This is the case of continuum states, when in relation (1) effective transmission coefficients must be used [5].

$$T_{eff}(U) = \int_{E\min}^{Emac} T(E)\rho(E)dE \tag{2}$$

where U is the excitation energy; ρ is the levels density with models given in [10].

3 Results and discussions

Theoretical and experimental total fission cross section of ^{238}U induced by fast protons up to 40 MeV is represented in Fig. 1.

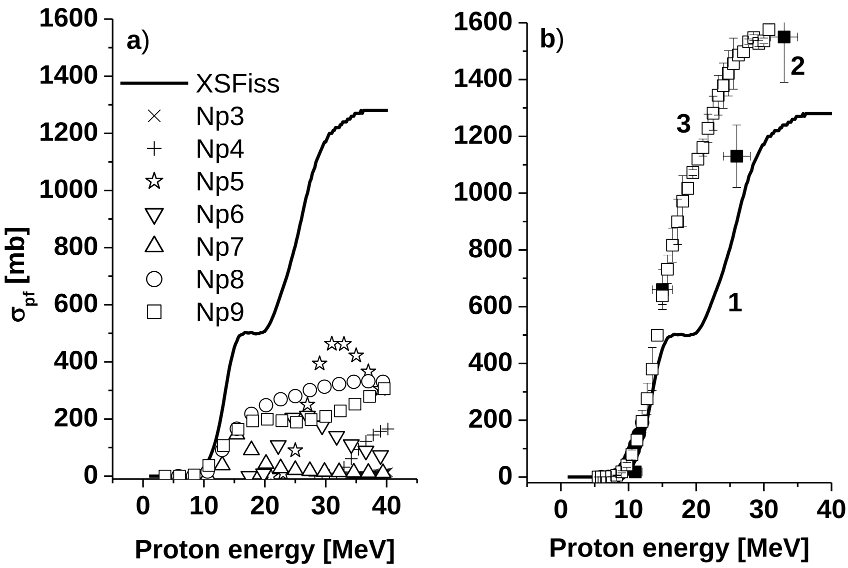

Fig. 1. Fast proton induced total fission cross section of ^{238}U a) Theory. $^{238}U(p,f)$. Line – Total; "x" – ^{233}Np; "+" - ^{234}Np; Star - ^{235}Np; Down triangle -^{236}Np; Up triangle - ^{237}Np; Circle -^{238}Np; Square - ^{239}Np. b) Theory and Experiment. 1 – Theory; 2, 3 – Experiment

In Fig. 1.a total fission cross section of ^{238}U induced by fast protons with energy up to 40 MeV is represented. In this process excited states of Np and U are formed due to the (p,xn) and (p,pxn) processes, respectively, and these excited states will fission. Fission cross section of only $^{238}U(p,f)$ is of order of 15 mb at 40 MeV energy and of $^{238,\,239}Np$ is about 320 mb. Theoretical results from Fig. 1.a have shown that fission cross section of U isotopes is with order of magnitude lower than cross sections of Np nuclei which are giving the main contribution to the total fission cross section.

In Fig. 1.b two sets of experimental data [14,15] are compared with the theoretical evaluation. At low energies it is a very good agreement between theory and experiment. With the increasing of proton energy can be described very well only one set of experimental data. The difference can be explained by presence of a large number of open channels with participation of deuterium, tritium, ^{3}He, alpha particles and ions with mass A > 4, ions which will act as a background.

Considering a good description of total fission cross section experimental data other fission observables were evaluated. Mass distributions (yields and cross sections) of fission fragment, prompt neutron emissions and isotope production (yields, cross sections) for many nuclides were calculated from 0.5 MeV up to 40 MeV. In Figs. 2 and 3, mass distributions of fission fragment, for protons energy E_p = 5 and 35 MeV, respectively, are represented. Yields are shown in Fig. 2a, Fig. 3a and cross sections in Fig. 2b, Fig. 3b.

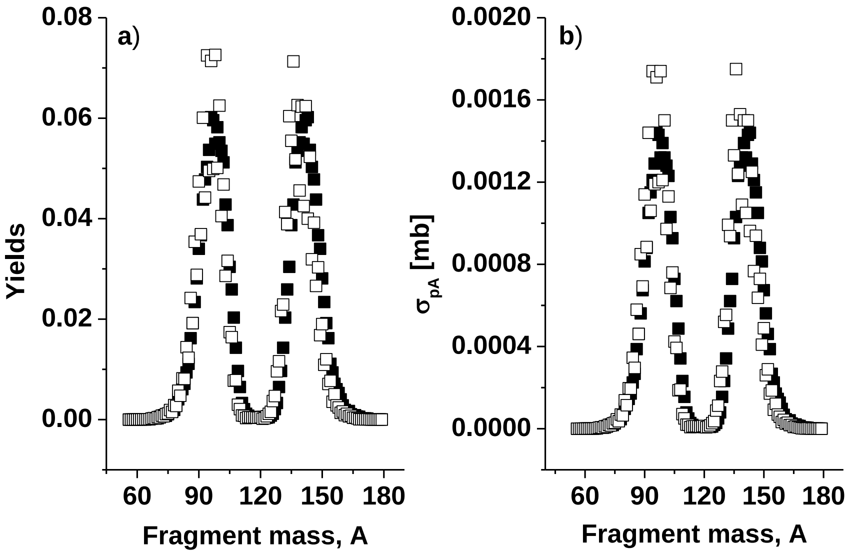

Fig. 2. ^{238}U(p,f) fission fragment mass distribution. Proton energy E_p = 5 MeV. a) Yields b) Cross section. Neutron emission. Squares: full – Pre; Empty - Post

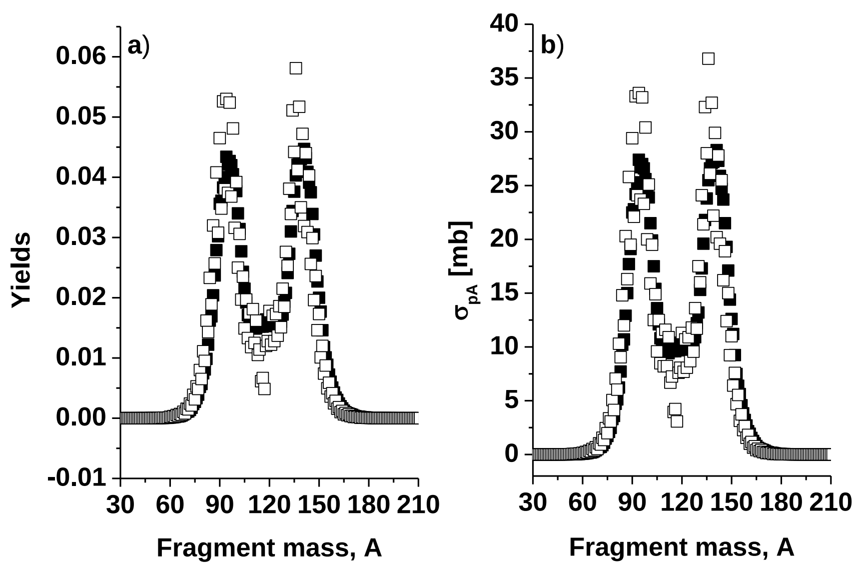

Fig. 3. ^{238}U(p,f) fission fragment mass distribution. Proton energy E_p = 35 MeV. a) Yields b) Cross section. Neutron emission. Squares: full – Pre; Empty – Post

Mass distributions of fission fragment is enlarging and become more symmetric with the increasing of proton energy (Fig. 2.a and Fig. 3.a). Cross section dependence of fission fragment mass is also increasing, becomes larger and symmetrical (Fig. 2.b and Fig. 3.b).
Prompt neutron emission can be described by two fission observables. First is the average prompt neutron multiplicity (APNM) depending on the mass of fission fragment and the second is prompt neutron multiplicity distribution (PNMD). Theoretical evaluations of APNM for 5, 15, 35 MeV are represented in Fig. 4.a and of PNMD in Fig. 4.b. Prompt neutron fission variables were evaluated from 0.5 up to 40 MeV but these results were not represented in this work. In the case of proton-induced fission of ^{238}U experimental data are very poor in comparison with neutrons. As is expected for both observables, number of emitted neutrons by fission fragments and their distribution is increasing with incident proton energy. The parameter describing APNM and PNMD is the so-called nu-bar prompt representing the average number of emitted prompt neutrons as functions of incident energy. It is an important parameter in nuclear engineering. Calculated values of nu-bar prompt in fast protons induced fission, for 5, 15 and 35 MeV are $\bar{\nu}_{pr}(5MeV) = 3.95$, $\bar{\nu}_{pr}(15MeV) = 4.93$ and $\bar{\nu}_{pr}(35MeV) = 6.77$ neutrons, respectively.

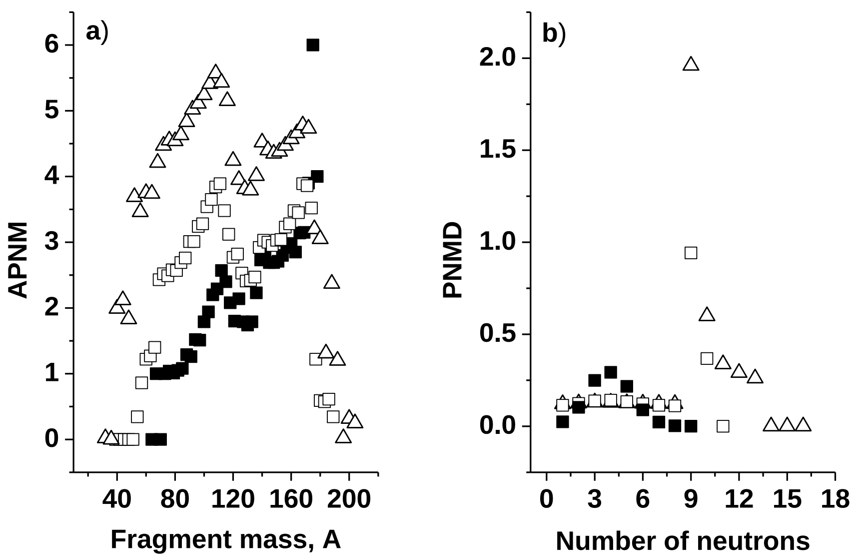

Fig. 4. Prompt neutron emission by fission fragments. a) APNM; b) PNMD. Full squares – 5 MeV; Empty squares – 15 MeV; Empty triangles – 35 MeV

An interesting effect can be observed in Fig. 4. At 5 MeV proton energy number of emitted neutrons is higher than for 15 and 35 MeV for fragments mass around A = 200. This result may be related to the structure of excited nuclei but the evaluations must be verified in experiment.
Fission process is a source of new isotopes for many applications. In fast proton induced fission of ^{238}U were evaluated yields and cross section production for a large number of artificial nuclei. Further will be presented results for isotopes of Mo, I, Xe, and Y which are important for medicine, engineering, nuclear technology and other fields of activities.
In Figs. 5.ab, Fig. 6.ab and Fig. 7.ab are represented yields and cross section of ^{99}Mo, ^{131}I and ^{133}Xe isotope production, respectively. Nucleus ^{99}Mo is of interest in medicine in cancer therapy. ^{131}I isotope is a fission product of Uranium and Plutonium, very important in

radiobiological protection, nuclear medicine, industry (as tracer). ^{133}Xe nucleus is used in medicine applications.

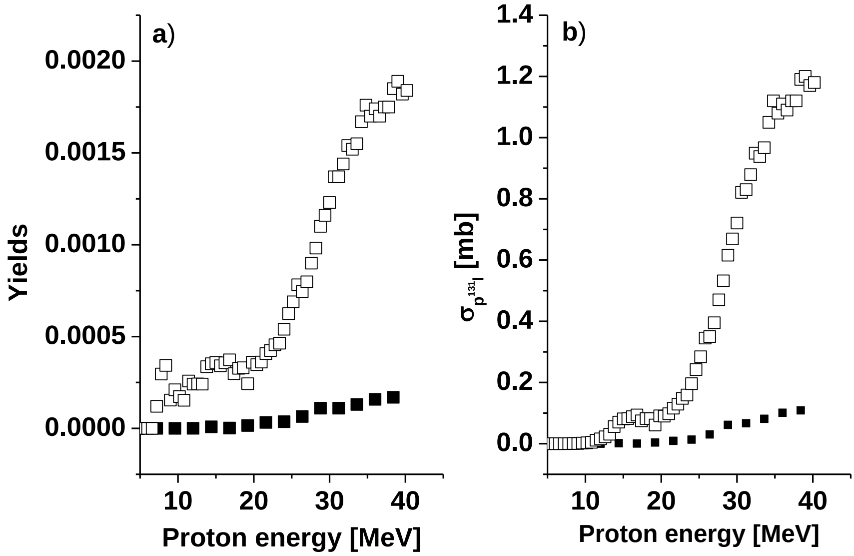

Fig. 6. ^{131}I production in ^{238}U(p,f). a) Yields; b) Cross section. Neutron emission. Full squares – Pre; b) Empty squares – Post

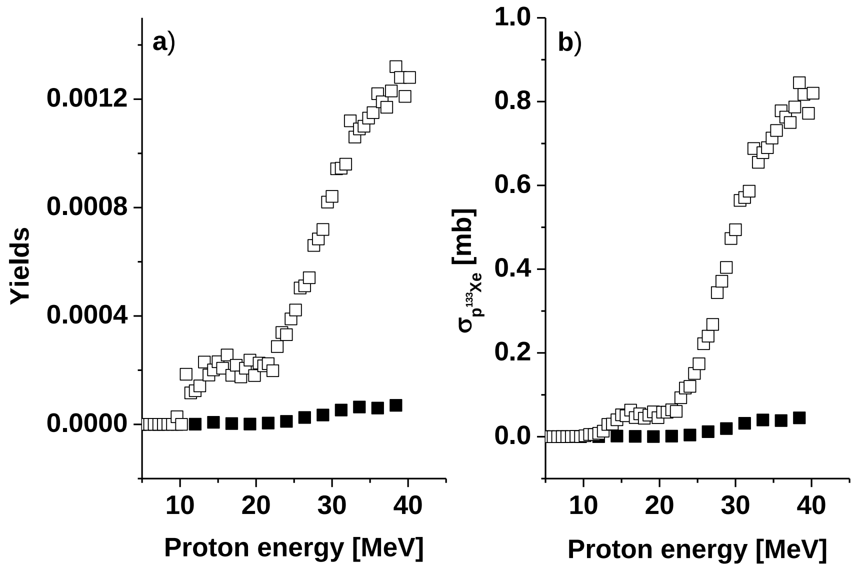

Fig. 7. ^{133}Xe production in ^{238}U(p,f). a) Yields; b) Cross section. Neutron emission. Full squares – Pre; b) Empty squares – Post

Isotopes ^{99}Mo. ^{131}I, ^{133}Xe are produced in very low amount in proton induced fission of ^{238}U. In Talys it is necessary to enhance the precision of calculations of yields and cross sections in order to obtain the results from Figs. 5, 6 and 7. With a default Talys run the upper evaluations cannot be obtained. Production of the same isotopes in fast neutron induced process of ^{238}U is with one or two orders of magnitude lower [21]. In the case of fast neutron induced fission of ^{233}U, production of the three mentioned isotopes is much higher. For example cross section production of ^{99}Mo, ^{131}I and ^{133}Xe at 20 MeV for $^{233}U(n,f)$ are 1,11 mb, 3.43 mb and 3.61 mb, respectively. For fast protons, for the same incident energy and isotopes, cross sections are: 0.4 mb, 0.1 mb and 0.05 mb, respectively. It is necessary to remark that cross sections of isotopes production mentioned before, for proton, gamma and neutron induced fission of Uranium isotopes were considered after prompt neutron emission of fission fragments. In all studied cases, isotope production is considerably increasing in comparison with pre-neutron emission stage.

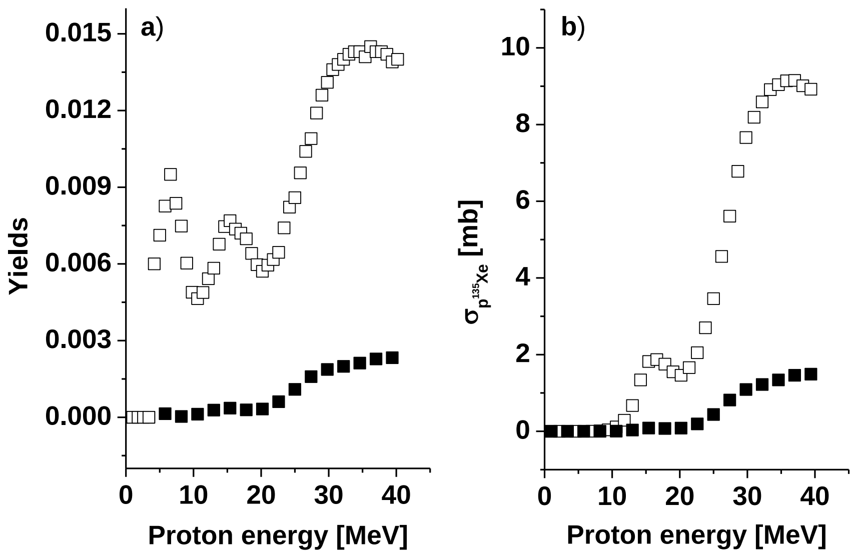

Fig. 8. ^{135}Xe production in $^{238}U(p,f)$. a) Yields; b) Cross section. Neutron emission. Full squares – Pre; b) Empty squares – Post

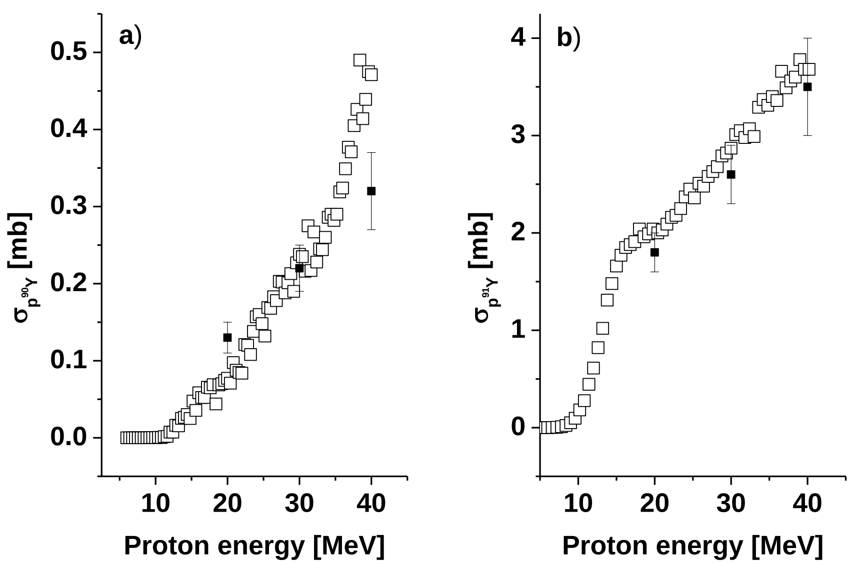

Fig. 8. ^{135}Xe production in $^{238}U(p,f)$. a) Yields; b) Cross section. Neutron emission. Full squares – Pre; b) Empty squares – Post

In the neutron induced fission ^{135}Xe nucleus is a major fission product, is a high neutron absorber, which affects the function of nuclear reactor. Cross section production of ^{135}Xe is around 12 mb at 20 MeV in the neutron-induced fission [16]. Results of production of ^{135}Xe isotope for protons are shown in Fig. 8.ab and are much smaller than neutrons case.
In the literature there are experimental data for ^{135}Xe production in $^{238}U(p,f)$ process but there are large differences at the same energies and in many cases the results are affected by large errors. Cross section theoretical evaluation at E_p = 9.4 MeV is σ_{pf} = 0.037 mb and in experiment where obtained σ_{pf} = (0.03 ± 0.02) mb and (0.104± 0.04) [17]. The authors of [18] had shown also other experimental cross sections at the same energies with large differences. These differences in the measured cross section production can be explained by the high number of produced isotopes and difficulties in the separation of their amounts.

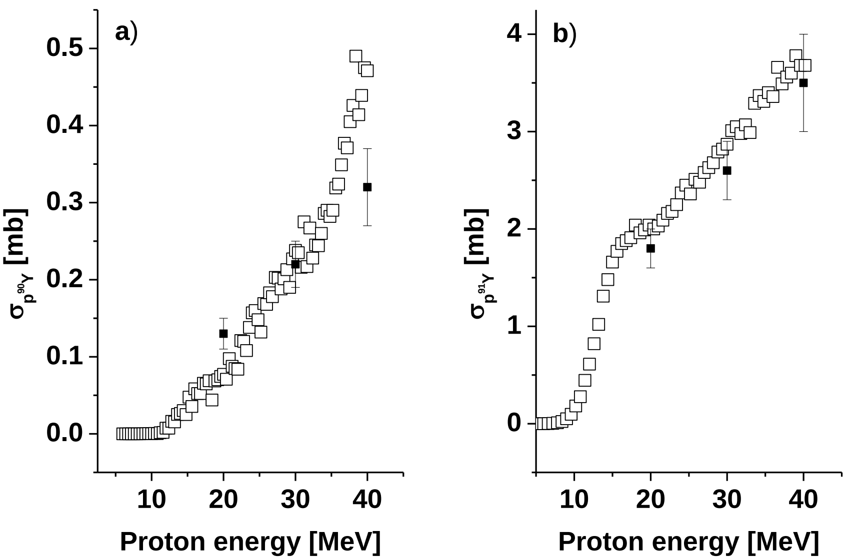

Fig. 9. Y isotope production in $^{238}U(p,f)$. Cross sections a) 90Y; b) 91Y. Squares: Empty – Theoretical evaluations; Full – Experimental data

A well description of production cross section was obtained for the isotopes $^{90, 91}Y$. Results are shown in Fig. 9.ab and experimental data are taken from [19,20]. Yttrium nucleus has just one isotope. Other Y isotopes are fission products and they can be used in medicine, superconductor materials, Lithium batteries etc.
Theoretical yields and cross sections production were obtained for a large number of isotopes but in this work were presented only results from Fig. 5 to Fig. 9.
Computer simulations, for isotope production of Radon, Actinium, Thorium, Protactinium, Uranium and Neptunium were realized. Activity, absolute and relative yields of mentioned fission products for different atomic masses A were obtained (but not shown here).
A sample of ^{238}U with 1 cm^2 transversal area of ^{238}U were irradiated by a proton beam with intensity 0,15 mA and energy 40 MeV. Thickness of the target was chosen in such a way that incident protons cannot emerge from the target and was equal to 1.85 mm. Energy loss in the target of fission fragments and the corresponding path were calculated with SRIM [21]. Irradiation and cooling time was the same, equal with 24h each. Density of ^{238}U is 19.1 g/cm^3 and initial number of ^{238}U nuclei in the target was $8.94 \cdot 10^{21}$. In Fig. 10.ab is represented the number of $^{239,238}Np$ nuclei as function of time (irradiation + cooling). It is easy to observe that the number of ^{238}Np nuclei is with three orders of magnitude higher than the number of

^{239}Np. In principle by activation method ^{238}Np can be easy detected in comparison with ^{239}Np which is at the limit of detection or beyond.

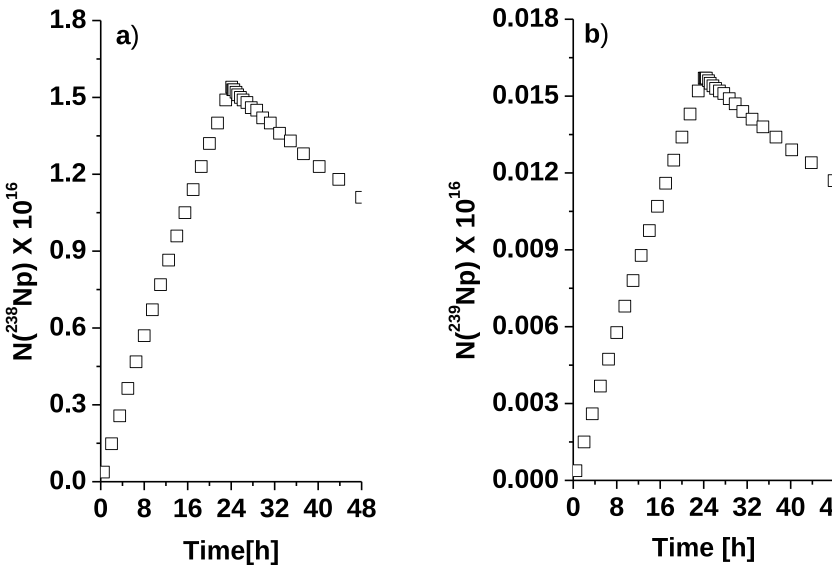

Fig. 10. Production of Np isotopes in ^{238}U(p,f). Absolute yields (number of nuclei, N). a) ^{239}Np; b) ^{238}Np

Results presented in Fig. 1 to Fig. 10 were obtained considering 30 levels of residual nuclei for elastic and inelastic scattering, 10 levels for reaction channels and 5 excited rotation levels. Parameters of Wood-Saxon in incident channel are shown in Table 1. Parameters shown in Table 1 are influencing the most cross section results.

Table 1. Parameters of Wood-Saxon potential in p+^{238}U channel

Volume						Spin-Orbit		
Real			Imaginary			Real		
V [MeV]	R_V [fm]	a_V [fm]	W [MeV]	R_W [fm]	a_W [fm]	V_{SO} [MeV]	R_{VSO} [fm]	a_{VSO} [fm]
55.12	1.245	0.66	0.1	1.248	0.594	5.66	1.121	0.590

General expression of Wood-Saxon potential with paameters used are given in [5].

$$U(r,E)=V\cdot f(r,r_V,a_V)+iW\cdot f(r,r_W,a_W)+4ia_dW_d\frac{df(r,r_d,a_d)}{dr}+V_{SO}k\frac{1}{r}\cdot\frac{df(r,r_{VSO},a_{VSO})}{dr}+ \quad (2)$$
$$+W_{SO}k\frac{1}{r}\cdot\frac{df(r,r_{WSO},a_{WSO})}{dr}$$

where $f(r,R,a)=\frac{1}{1+Exp\left(\frac{r-R}{a}\right)}$ is Wood-Saxon factor; a is diffuseness parameter; R is radius;

$k=\left(\frac{\hbar}{m_\pi c^2}\right)^2$; $\hbar$ is reduced Planck constant; c is speed of light in vacuum; m_π is pion mass.

Fission fragment yields and cross sections of isotopes produced were obtained using random neck rupture model [22]. In the evaluations were considered two fission barriers with parameters taken from experimental data [5]. The height of first barrier is 6.3 MeV, width is 1 MeV and triaxial left-right asymmetry type of axiality. The second barrier is 5.5 MeV height with 0.6 MeV width and left-right asymmetry type of axiality [5].

4 Conclusions

Fission of ^{238}U induced by fast protons up to 40 MeV was investigated. Fission observables like cross sections, mass distribution of fission fragments, prompt neutron emission, relative yields with cross section production and computer modeling of generated isotopes were obtained using dedicated and own codes for calculations. It was determined that total fission cross section is the sum of fission of excited fissionable nuclei obtained in the interaction of ^{238}U with fast protons. Theoretical evaluations were compared with experimental data. In this case, existing total fission cross section experimental data are in good agreement with theoretical results.

Cross sections and yields of a large number of isotopes produced in ^{238}U(p,f) process, were also calculated. In the present work was presented only a small part of produced isotopes of interest in applications. While theoretical and experimental data for most obtained isotopes are in good agreement, there are also synthesized nuclei with significant differences. Taking into account the agreement between theory and experiment of some fission observables new parameters of optical potential were obtained as well as parameters describing fission barriers (height, with, type of axiality, etc) were also extracted.

Fast proton induced fission was less investigated in comparison with neutrons and therefore are necessary new measurements of fission observables. It is necessary to improve data processing and further computer simulations of ^{238}U(p,f), considering the importance of this process for fundamental and applicative researches.

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