Nuclear Reaction Mechanisms of Neutron Induced Fission of ^{237}Np Nucleus

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Abstract.

For a few years at FLNP, JINR Dubna has already started new prospects for development of new neutrons facilities that will replace the IBR-2 neutron pulsed research reactor, which will finish its activities in 2032 year. Some of the agreed projects are using fission process induced by neutrons on neptunium based fuels. In the present research the neutron fission process on ^{237}Np nucleus was investigated. Various parameters during the neutron induced fission of ^{237}Np nucleus such as; fission cross-section, mass distributions, prompt neutrons emissions, isotopes production and neutrons spectra, were analyzed. The evaluations of the above observables were done by using TALYS – 9.1 software for incident neutron energy ranging from 0.4 MeV up to 25 MeV. The exact value of cross-section was measured as 5 – 5.5 MeV in the present work, whereas from literature, it was found up to 10 MeV. For neutrons spectra, a separation of the contribution of different nuclear reaction mechanisms was obtained. Some important yields and cross-section production of isotopes have been recorded and were found within good agreement with literature data.

Keywords: neutron fission, cross sections, fission yields, nuclear reaction mechanisms, isotopes

1 Introduction

In the modern sciences neutrons are successfully used in the fundamental researches for the studies of nuclear interactions, symmetry issues and structure of atomic nuclei. In applied researches, neutrons are used in condensed state physics, molecular biology, structural chemistry, material sciences, nondestructive control of volume objects and industrial products. Neutron researches become more effective with the increasing of the neutron flux intensity because the time of experiments is reduced, the precision of measurements enhances, new possibilities open in the studies of complex, small objects in neutron scattering experiments (Aksenov et. al., 2017, A), (Aksenov, 2017, B). Recent progresses of applied and fundamental researches in sciences necessitate new neutrons sources with highly improved intensity.

1.1 **Neptunium-237**

Neptunium is a silver-gray metal and a good conductor of heat and electricity. His melting point is 637 °C and that of boiling 4175 °C (Thomassin et al., 2001). It is part of the actinide elements and can exist in five oxidation states, from III to VII. Neptunium is the first one of the transuranic elements; its twenty two isotopes are ranging from ^{225}Np to ^{242}Np. All are radioactive and most of them have very short radioactive half-lives.

The ^{237}Np is the most significant neptunium isotope; it is a radioactive artificial nucleus with atomic mass *A=237* and proton number *Z=93*. It has the longest radioactive half-life about 2.144×10^6 years and mass activity of 2.6×10^7 Bq.g^{-1}.

Throw α disintegration ^{237}Np gives the Protactinium-233, itself β emitter, and have a half live of 27 days. ^{237}Np is produced in significant quantities in nuclear reactors mainly during the combustion of ^{238}U. As known, ^{237}Np is produced by neutron irradiation of uranium according to the reactions (Audi et al., 2003, A), (Audi et al., 2003, B):

$$^{238}U\left(n,2n\right)\,^{237}U \xrightarrow{\beta}\,^{237}Np \tag{1}$$

$$^{235}U(n,\gamma)\,^{236}U(n,\gamma)\,^{237}U \xrightarrow{\beta}\,^{237}Np \tag{2}$$

Its contribution consists in long-term high-level waste (HLW) radiotoxicity of nuclear spent fuel. In nuclear reactor ^{237}Np is synthesized

1.2 **Neutron induced fission on Neptunium-237**

Neptunium-237 is present in nuclear spent fuel in considered amount. This long-lived minor actinide can be incinerated in nuclear reactors by neutron fission process and eventually used as nuclear fuel. The ^{237}Np has fission cross section of a threshold type in comparison with traditional ^{235}U and ^{239}Pu fuels. The effective fission threshold for ^{237}Np is situated around *0.4* MeV which is about *0.2* MeV lower than in the case of ^{238}U. This property is very important in the evaluation of the critical mass (Shabalin et al., 2017, A), (Shabalin et al., 2017, B). Time of life of delayed neutrons produced in fast neutron induced fission of ^{237}Np is with three orders of magnitude lower than of ^{238}U and ^{239}Pu. Mentioned above properties of ^{237}Np, recommends this long-lived actinide isotope as an eventually fuel for future research nuclear reactors (Shabalin et al., 1966), (Aksenov et al, 2017, A), (Aksenov et al, 2017, C).

The data on ^{237}Np (n,f) can be obtained from several sources. The experimental data can be downloaded from EXFOR database and the evaluated ones can be obtained from the nuclear data libraries as ENDF/B-VII.1, JENDL-4.0 JEFF3.2, CENDL 3.1, and ROSFOND-2010, which present a good source for data especially when no experimental data exist.

In the present investigation nuclear reaction mechanisms and fission variables, like cross sections, mass distributions, prompt neutrons emission, isotopes productions, as well as neutrons spectra, were evaluated in $^{237}Np(n,f)$ process induced by fast neutrons with energies from *0.4* MeV up to *25* MeV.

2 Computer codes and Nuclear Reaction Mechanisms

Evaluations of fission variables under different reaction mechanisms were realized with Talys and programs realized by authors.

2.1 Talys code

TALYS is software for the analysis and prediction of nuclear reactions that involve neutrons, photons, protons, deuterons, tritons, ^{3}He- and alpha-particles, in the 1 keV - 200 MeV energy range and for target nuclides of mass 5 u.a. and heavier. To achieve this, a suite of nuclear reaction models has been implemented into a single code system. This enables to evaluate nuclear reactions from the unresolved resonance range up to intermediate energy (Koning et al., 2007), (Koning et al., 2008).

TALYS can be used as nuclear physics tool for the analysis of nuclear reaction experiments according to nuclear models and also as nuclear data generator when no measurements are available (Koning & Rochman, 2012).

The data that can be produced in a calculation with the TALYS code are typically the following: Total cross sections, elastic and non-elastic cross sections, Elastic scattering angular distributions, Inelastic scattering angular distributions and cross sections to discrete states, Cross sections of exclusive channels, i.e. (n,γ), (n,2n), (n,np),.., with their energy spectra and double differential spectra, Gamma ray production for discrete and continuum states, Production cross sections of isomeric states, Fission cross sections, fission fragments and product yields, Production cross sections of residual nuclei, and Total particle emission cross sections, i.e., (n,xn), (n,xp),.., with their energy spectra and double differential spectra (Koning & Rochman, 2011).

2.2 Nuclear reaction mechanisms. Elements of theory

Fission induced by fast neutrons with energies up to *25* MeV can be described by compound processes using Hauser Feshbach formalism. In this case fission cross section has the form (Hauser & Feshbach, 1952), (Koning et al., 2007):

$$\sigma_{nf}(E_n) = g\pi\lambda\!\!\!^{-}{}_n^2 T_n T_f \left[\sum_c T_c\right]^{-1} \tag{1}$$

$$T_f^{J,\Pi}(E_x) = \sum_i T_f(E_x,\varepsilon_i) f(i,j,\Pi) + \int_{E_{th}}^{E_x} \rho(\varepsilon,j,\Pi) T_f(E_x,\varepsilon) d\varepsilon \tag{2}$$

where T are the transmission coefficients; g is the statistical factor; $\lambda\!\!\!^{-}{}_n$ is the reduced neutron wave number; c are the channels; $E_x, E_{thr}, \varepsilon$ are the excitation, threshold and current energies, respectively; J,Π are the spin and parity of the compound nucleus; ρ represents the nuclear states density.

Transmission coefficients, *T*, from relation (2), are the probabilities of particles to pass a potential barrier and can be evaluated for discrete and continuum states, respectively. In relation (2) discrete states are represented by the sum and continuum ones by the integral (Koning et al, 2007). Levels density is taken from statistical formalism of nuclear reactions, developed mainly from the Fermi gas model and combinatorial approaches (Koning et al., 2008). For fission process transmission coefficients are calculated according to Hill – Wheeler expressions (Koning et al, 2007).

Further fission variables, like mass and charge distribution of fission fragments, prompt neutrons distributions, isotopes yields fission product are obtained using Brosa model (Brosa et al., 1990). In the frame of Brosa approach it is supposed that the fission process is a succession of non stable nuclear states (instant Rayleigh type of instability) where the break of the neck has an instant character. The scission is produced when the neck is flat and the place of the scission is random therefore giving the possibility of the multichannel fission.

Fission barrier parameters were taken from experimental data (Koning et al., 2007) and calculated by authors in accordance with (Mamdouh et al., 2001) where sets of double – humped parameters are defined for a large number of isotopes obtained from Extended Tomas – Fermi and Strutinski integral calculations (Strutinsky, 1968).

Mass distribution (relative yields) is calculated applying the following relation (Koning et al., 2007):

$$Y\left(A_{FF};Z_{FS},A_{FS},E_x\right)=\sum_{FM}W_{FM}\left(Z_{FS},A_{FS},E_x\right)Y_{FM}\left(A_{FF};Z_{FS},A_{FS},E_x\right) \quad (3)$$

where A_{FF} is the fragment mass coming from a fissionable system (nucleus) with mass A_{FS} and charge Z_{FS}; W_{FM} is the weight of a fission mode of a nucleus with mass A_{FS} and charge Z_{FS}; $Y_{FM}\left(A_{FF};Z_{FS},A_{FS},E_x\right)$ are the relative yields of a fission mode; E_x is the excitation energy.

The production XS of the fission fragment with given mass (A_{FF}) and charge (Z_{FF}) is (Koning et al, 2007):

$$\sigma_{prod}\left(Z_{FF},A_{FF}\right)=\sum_{Z_{FS},A_{FS},E_x}\sigma_F\left(Z_{FS},A_{FS},E_x\right)Y\left(A_{FF};Z_{FS},A_{FS},E_x\right)Y\left(Z_{FF};A_{FF},Z_{FS},A_{FS},E_x\right) \quad (4)$$

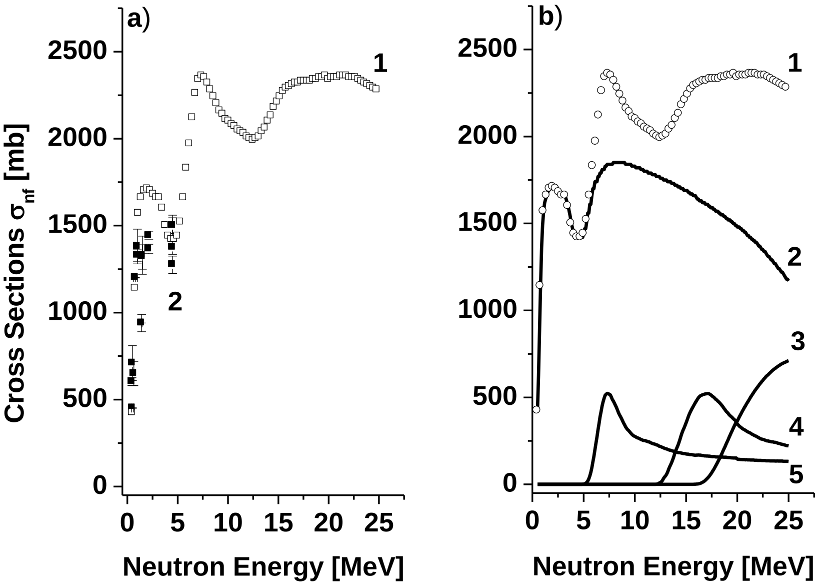
where $Y\left(Z_{FF};A_{FF},Z_{FS},A_{FS},E_x\right)$ represent the relative yields with charge Z_{FF} and mass A_{FF} coming from fissionable nucleus with charge Z_{FS} and mass A_{FS}.

Neutron spectra were evaluated considering neutron evaporation of fission fragments and neutrons obtained from other possible channels. Neutron evaporation of excited fission fragments is enabled by compound processes. Further, in the interaction of neutrons with fissionable nuclei beside fission, are open also elastic, inelastic, capture or other channels, involving emission of light charged particles or clusters. In this case, neutrons in the spectra are produced not only by compound mechanisms characteristic to fission and evaporation but also by direct and pre-equilibrium ones of other open channels (Koning et al., 2007).

3 3. Results and Discussions

In Figure 1 are represented cross sections (XS) in the fission process induced by the neutrons with energies from *0.4* MeV up to *25* MeV on ^{237}Np nucleus.

Figure. 1. Total fission XS of ^{237}Np. a) 1–Talys; 2–Experimental Data. b) Contribution to the XS of fissionable nuclei derived from ^{237}Np.1-Talys; 2–^{238}Np; 3–^{237}Np; 4-^{236}Np; 5–^{235}Np



In the Figure 1.a the theoretical Talys calculations were compared with experimental data from literature (Diakaki et al., 2016), (Jiacoletti et al., 1972). From Figure 1.a it can be noted that experimental data exist only up to *10* MeV and a good agreement between theory and experiment was obtained below 10 MeV energy. The exact value of cross-sections has been reported around approximate 5 MeV in the present work. However, over the neutron energy range 14 to 16 MeV, only a few values of cross-sections have been reported by other workers (Diakaki et al., 2016), (Jiacoletti et al., 1972), (Ruskov et al., 2018).

During fission process other fissionable nuclei are formed and these isotopes contribute to the total XS (Figure 1.b). The most important part is given by the compound nucleus ^{238}Np, but after *7-8* MeV other isotopes of *Np* influence the value and the shape of total XS (seen in curves 2, 3, 4 from Figure 1.b). Curve 5 from Figure 1.b is the same with curve 1 from Figure 1.a. In fission process are obtained isotopes of *U* and *Pa* but their contributions to the XS is neglected. Separation of XS due to different isotopes is useful in the evaluation of neutron field in nuclear reactor.

In the Figures 2 and 3, the fission fragment mass distribution and the average prompt neutron multiplicity, respectively, are shown. In each figure are represented two curves corresponding to the pre- and post-neutron emission of the excited fragments. Average prompt neutron multiplicity and mass distribution are obtained by applying Brosa model. Prompt neutrons emission is described by evaporation process depending on nuclear temperature, density levels and nuclear reaction mechanisms (Brosa et al., 1990). Average prompt neutrons multiplicity and mass distribution are enlarging in mass fragment with the increasing of neutrons incident energies. Also, symmetrical fission and prompt neutrons emission are increasing with the neutrons incident energy as predicted by theory (see Figure 2.a - 2.b and Figure 3.a – 3.b).

Figure 2. Fission fragment mass distribution. A = mass fragment. Incident neutrons energy, E_n. a) E_n= 1 MeV; b) E_n= 20 MeV. Neutrons emission: Empty triangles – Pre; Black squares - Post

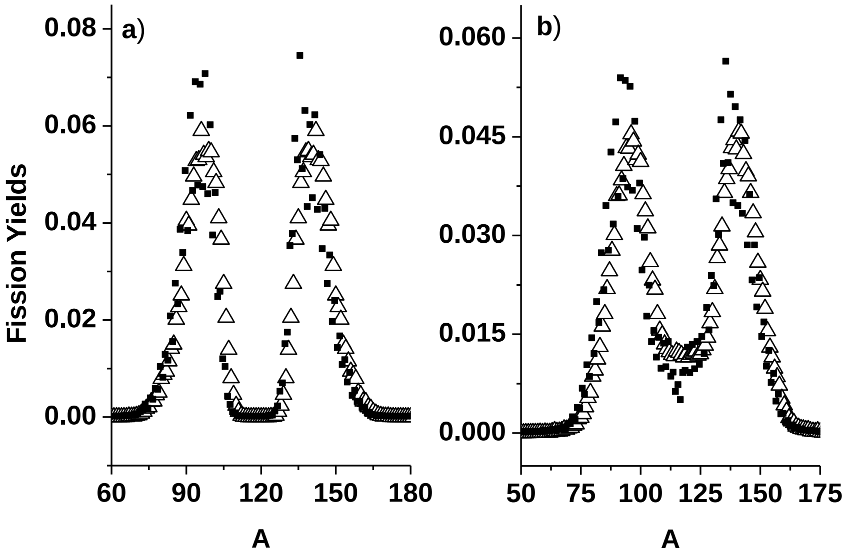
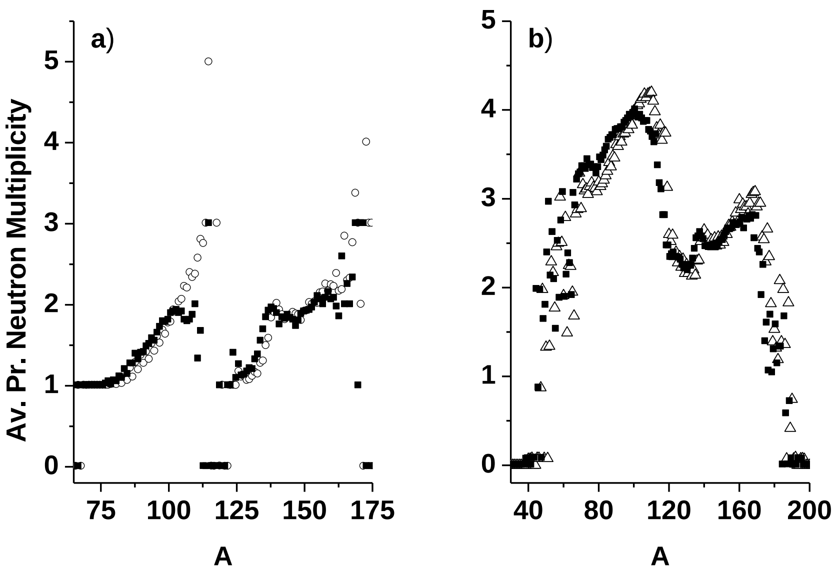


Figure 3. Prompt average neutron multiplicity. A = mass fragment. Incident neutrons energy, E_n. a) E_n= 1 MeV; b) E_n= 20 MeV. Neutrons emission: Empty triangles – Pre; Black squares - Post

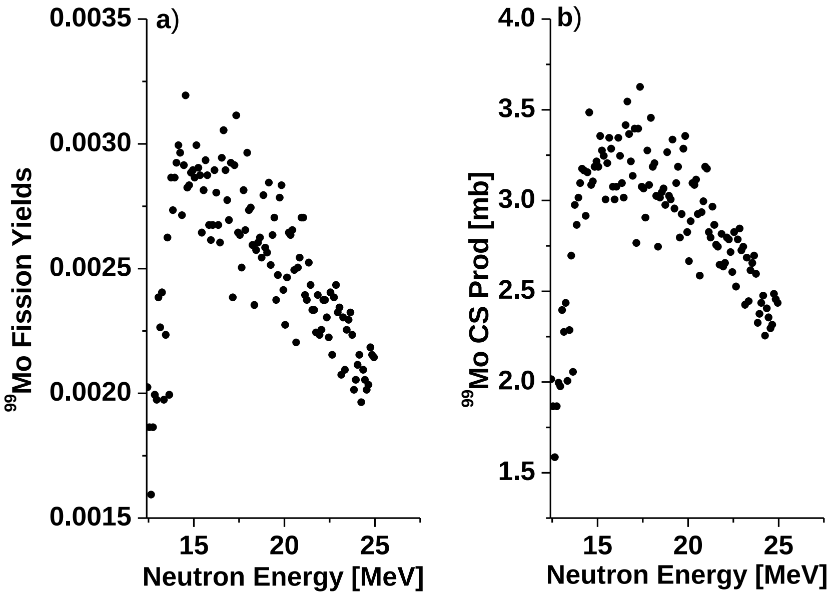


Talys gives also the possibility to evaluate the production of isotopes for different application like ^{133}*Xe*, ^{131}*I*, ^{99}*Mo* and other. The ^{133}*Xe* nucleus is a high neutrons absorber and very important for functioning of nuclear reactor. The ^{131}*I* nuclide is of interest in radioactive pollution and ^{99}*Mo* is used in medicine. In Figure 4, the dependence of ^{99}*Mo* production by incident neutron energy is represented. In Figure 4.a the relative yields of ^{99}*Mo* are shown and in Figure 4.b the corresponding production XS. In both cases results are obtained after neutrons emission of the fission fragments. The theoretical evaluations showed that the production of ^{99}*Mo* isotope in ^{237}*Np* neutron fission is with order of magnitude higher than in the case of the fast neutrons induced fission of ^{238}*U* nucleus.

The next obtained results are the neutron spectra obtained in $^{237}Np(n,f)$ process for each incident energy. In Figure 5 the cases for incident neutrons energy equal with *1* and *20* MeV, respectively, are represented.

In Figure 5.a, for incident energy, $E_n = 1$ *MeV*, compound processes produced neutrons up to *0.5* MeV. At higher energies, until *2.5* MeV neutrons are given by direct processes. These neutrons are coming mainly from other nuclear reaction mechanisms *(n,n)*, *(n,n')*, *(n,γ)* and less from the fission.

Fig. 4. Energy dependence of ^{99}Mo isotopes production. a) Yields; b) XS; Post neutron emission



In the Figure 5.b, at *20* MeV neutrons incident energies, up to *8* MeV the spectrum is given by compound process due to evaporation. In comparison with the Figure 5.a, neutrons are provided by multiemission pre-equlibrium processes at the beginning of the spectra. Furthermore, the pre-equilibrium mechanism is present in the whole energy range. For neutrons with energies higher than *20 MeV* direct processes become dominant. In all cases, preequilibrium and multipreequilibrium mechanisms contribute mainly at low energy when they compete with compound processes. Neutrons spectra from Figure 5.b were calculated taking into account all possible exit channels in the interaction of neutrons with ^{237}Np nucleus. The theoretical calculations have demonstrated that the most important channels are fission, gamma and neutrons. Emergent channels like protons, alpha particles, deuterium, and tritium can be neglected due to the height of Coulomb barrier. Then, the neutrons from Figure 5.b are coming not only from excited fragments obtained in fission, but also from excited nuclei resulted in the inelastic, capture and *(n,xn)* interactions.

Figure 5. Reaction mechanisms contribution to neutron spectra for incident neutrons energy a) 1 MeV; 1 – compound; 2 - . direct – 3 – total; b) 20 MeV; 1 – compound; 2 – direct, 3 – pre-equilibrium; 4 – multi pre-equilibrium; 5 - total

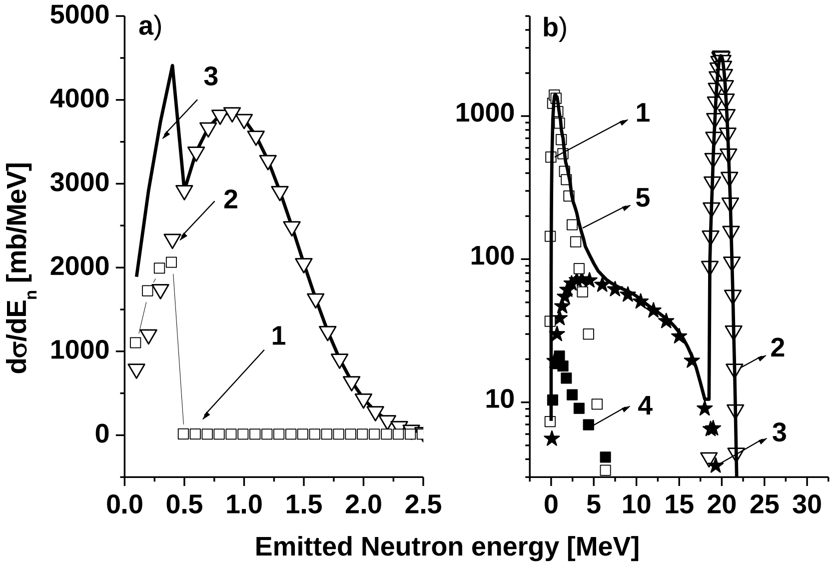
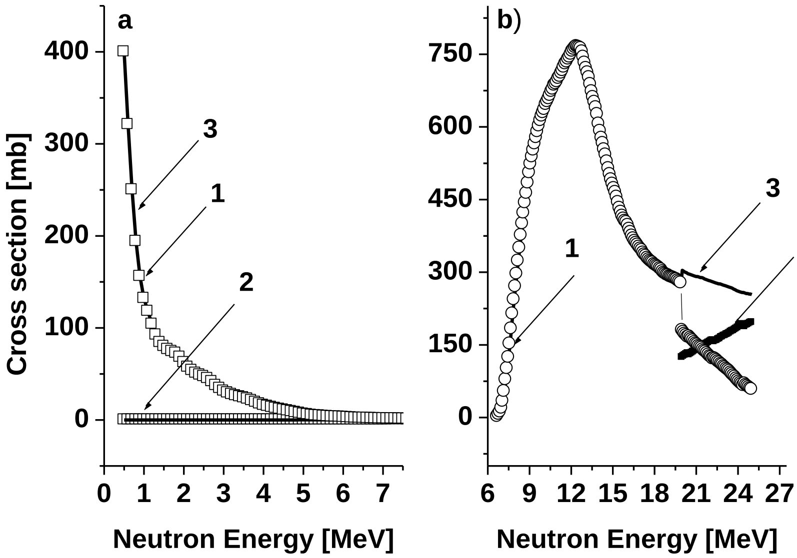


Figure 6. *Np* isotopes production. a) ^{238}*Np*; b) ^{236}*Np*. Processes. 1_ compound; 2 – pre-equilibrium; 3 – total



In the interaction of fast neutrons with ^{237}*Np*, isotopes of *Np, U, Pa* and other fissionable nuclei are obtained. These nuclides are of interest in the design of nuclear reactor because they also will contribute to the fission chain reaction. The *U, Pa* and other isotopes obtained in the emission of light charged particles have much lower CS production values in comparison with *Np* isotopes and their contribution to the fission can be neglected.

In Figure 6.a and 6.b production cross sections of ^{238}*Np* and ^{236}*Np* isotopes are represented and contribution of nuclear reaction mechanism obtained. The ^{238}*Np* isotope is obtained in neutrons capture process given practically only by compound mechanism. The ^{236}*Np* nucleus is obtained in ^{237}*Np(n,2n)* reaction. In this case also, compound processes are dominant but at higher energies, compound processes are given pre-equilibrium multistep compound processes. In both cases presented in Figures 6.a and 6.b, direct mechanism has very low value in comparison with compound ones.

Theoretical evaluations of fission variables realized in the present research were obtained enabling the all nuclear reaction mechanisms. In the case of elastic and inelastic scattering *30* levels of the residual nucleus were taken into account and for reaction channels *10* levels. For excitation energies higher than the last discrete level, the residual nuclear states are considered in continuum. The fission calculations were realized considering a double humped barrier. The parameters of the first barrier are: height is *6* MeV, width is *0.6* MeV and triaxiality with left right asymmetry type of axiality. The parameters of the second barrier are: height = *5.25* MeV; width = *0.6* MeV and left – right asymmetry as type of axiality.

In Talys are implemented optical potentials with volume, surface and spin orbit components for incident and emergent channels. In the incident channel, "$n + {}^{237}Np$" parameters of optical potentials, are: real part of volume Wood – Saxon potential, $V_V = 46.52$ MeV, imaginary part of Wood = Saxon potential, $W_V = 0.08$ MeV, Imaginary part of surface potential, $W_d = 2.32$ MeV, Real part of spin – orbit interaction, $W_{SO} = 5.69$ MeV. Definition, notations, components and parameters of Wood – Saxon optical potential can be found in (Koning et al., 2007).

4. Conclusions

Fission observables, like cross sections, mass distributions, prompt neutrons emissions, isotopes production and neutrons spectra, necessary for designing of an intense neutron source based on ${}^{237}Np$ fuel, were analyzed. Evaluations were realized with Talys software and in addition with author's own calculations, for incident neutrons starting from *0.4* MeV up to *25* MeV. Existing XS experimental data from literature up to *10* MeV, are well described by our computed values. Using XS results, other fission observables were calculated, taking into account the lack of experimental data. For neutrons spectra, a separation of the contribution of different nuclear reaction mechanisms was obtained. Yields and XS production of more than 20 fission products of interest for applications were obtained as new theoretical data. From all isotopes produced in the fast neutrons fission of ${}^{237}Np$, ${}^{99}Mo$ nucleus was chosen and presented because theoretical evaluation had demonstrated that it is produced ten times more than in the fast neutrons fission of ${}^{238}U$.

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