https://doi.org/10.15407/ujpe67.1.11

C. OPREA,[1,2] M.A. AHMAD,[3] J.H. BAKER,[3] A.I. OPREA[1]

[1] Frank Laboratory for Neutron Physics (FLNP) Joint Institute for Nuclear Researches (JINR)
(6, Joliot Curie Str., Dubna 141980, Moscow Region, Russian Federation)
[2] Romanian National Agency for Scientific Research
(21-25 Mendeleev, 010362, Sector 1. Bucharest; e-mail: istina@nf.jinr.ru)
[3] Physics Department, Faculty of Science
(P.O.Box 741, University of Tabuk, 71491, Saudi Arabia; e-mail: mayaz.alig@gmail.com)

MATHEMATICAL MODELING OF NEUTRON INDUCED FISSION OF ^{237}Np NUCLEUS

Recent progress of applied and fundamental researches in nuclear physics necessitates new neutron sources with highly improved intensity. For a few years at JINR (Dubna) the development of new neutron facilities that will replace the IBR-2 neutron pulsed research reactor, which will finish its activities in 2032, is carried on. Some projects use the fission process induced by neutrons in neptunium-based fuels. In the present research, we will study the neutron-induced fission of ^{237}Np nucleus. The cross-section, mass distribution, yields of isotopes of interest, average number of emitted prompt neutrons, neutron fission spectra, and other parameters are obtained. The mathematical modeling is done partially by using the theoretical models implemented in Talys software (TALYS-1.2) and by computer codes realized by the authors. The presented results are compared with the available data and are of interest in the JINR projects for the design of new neutron facilities destined for researches.

Keywords: neutron fission, cross sections, yields, reaction mechanism, production of isotopes.

1. Introduction

Due to the technologically advancement in nuclear science and nuclear engineering for nuclear data, the study of neutron induced fission of ^{237}Np nucleus has a great interest. The ^{237}Np nucleus is the most important and interesting isotopes for experimental investigation with neutrons at different energies. This ^{237}Np generated (and accumulated) in the nuclear reactor core during reactor operation [1, and references therein]. Recently, some considerable effort, both experimental and theoretical, has been focused at increasing the knowledge of such nuclear reactions (neutron induced fission). Comparatively few of the results can be found in compilations of general availability, such as those published by a new experimental setup TANGRA has been constructed at the Frank Laboratory of Neutron Physics of the Joint Institute for Nuclear Research in Dubna [2]. The TANGRA, (Tagged Neutrons and Gamma Rays) setup consists of: a portable ING-27 neutron generator with in-build position sensitive detector for alpha-particles [3], an array of BGO detectors of gamma rays, 2 Stilbene detectors for neutron spectroscopy and PC based 32 channels ADC for signal processing [4].

The ^{237}Np isotope, is a radioactive artificial nucleus with atomic mass and atomic number ($A = 237$ and $Z = 93$ respectively) synthesized in nuclear reactor during the combustion of Uranium (^{238}U) with a half life time $\tau = 2.144 \times 10^6$ years [5, 6]. The relative to nuclear fuel, the ^{237}Np has a threshold type of fission cross-section (C–S) 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 critical mass [7, 8]. Another interesting feature of ^{237}Np nucleus used as fuel is that the time of life of delayed neutrons is with three orders of magnitude lower than ^{238}U and ^{239}Pu fuels. All the mentioned above of ^{237}Np characters recommends this nucleus as a possible candidate for fuels for future research nuclear reactors [9, 10, 11].

In the modern sciences, neutrons are successful used in the fundamental researches for the studies of nuclear interactions, symmetry issues, structure of atomic nuclei. Related to applied researches, neutrons are used more widely in the 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 intensity because is reduced the time of experiments, enhances the precision of measurements, opens new possibilities in the studies of complex, small objects in neutron scattering experiments [11, 12].

In the present study, some physical parameters of interest in the neutrons induced fission like cross sections, mass distributions, prompt neutrons emission, and isotopes productions were evaluated. Evaluations were realized with Talys computer codes (i.e. TALYS-1.2) [13, 14] which represent friendly user software, working mainly under Linux, dedicated to nuclear reactions and structure of atomic nucleus calculations [14, 15]. Finally, the findings of our present research work were found within good agreements with others [1–15] working in the field of in nuclear science and nuclear engineering.

2. Theoretical Background

The mass and charge distributions are very much significant parameters of the fission process in nuclear reactions. Study on the mass distribution provide vital informations about the potential energy landscape of the fissioning nucleus and the mechanism involved [16, 17]. The main interest in the heavy ion induced fission at medium energy (into MeV) is to study the effect of entrance channel parameters namely, projectile energy, angular momentum and entrance channel mass asymmetry, on the fission process. An analysis of the data on the variance of the mass distribution over a wide range of the fissility of the compound nucleus was reported by various worker [18, 19].

Neutrons-induced fission cross-sections (including multichance) can be calculated in software (TALYS-1.2) [13] using phenomenological and microscopic approach taking into account various nuclear quantities. Levels densities are taken from statistical formalism of nuclear reactions, developed mainly from Fermi gas model and combinatorial approaches [20]. Levels densities are necessary for the evaluations of transmission coefficients and they represent the probability of a particle to tunnel a barrier potential. For fission process these coefficients are calculated according to Hill–Wheeler expressions [15, 21]. In the evaluations a double – humped (class II) potential is considered. In the case of statistical models of nuclear reactions the cross section has a Hauser–Feschbach form [22], which has been represented by following mathematical relation:

$$q_{nf}(E_n) = g\pi\lambda_n^e T_n T_f \left[\sum_c T_c\right]^{-1}, \quad (1)$$

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

The abbreviations, used in above equations (1 and 2) are such as; T, for transmission coefficient, g, is the statistical factor, λ_n is the wave number, c is channel, and E_x is the excitation energy, $E_{\rm th}$ is threshold energy, ε is current energy, J is spin, Π is parity and ρ is the density state. The factor "f" might be equal to 1, if spin and parity will be conversing, otherwise the value of "f" will be zero.

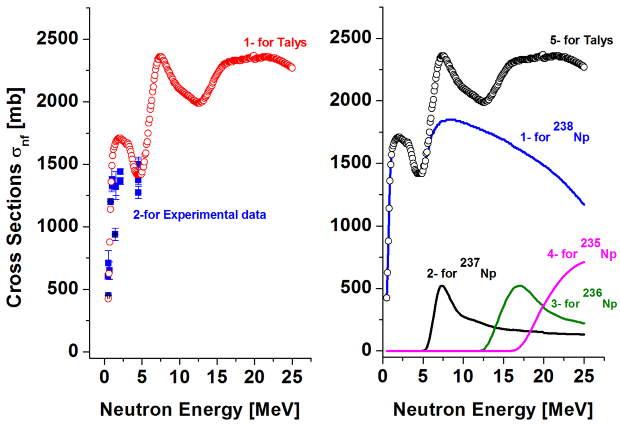
Brosa *et al.* [23] proposed the random neck rupture model (RNRM) for the calculation of post-fission parameters such as mass distribution, kinetic energy distribution and neutron multiplicity. According to this model, the pre-scission shape of the fissioning nucleus utters the post-fission parameters. This model has been successful in explaining the width of the mass distribution in low- as well as medium energy fission [23]. The mass and charge distribution, yields of some isotopes of interest for different application were evaluated using the Brosa model [23]. Also the neutrons spectra were evaluated considering the evaporation of fission fragments and taking into account different reactions mechanism in the incident neutrons energy interval.

According to the Brosa model [23], during the motion of the fissioning nucleus towards scission, a lump is build up in the neck region of the fissioning nucleus, which is expanded by the capillary force, finally leading to fission. The curvature of the fissioning nucleus changes from positive to negative in the motion towards scission [19 and references therein]. During this transition when the neck becomes flat, there can be a large shift in the dent without sizeable physical mass motion, which finally leads to large mass fluctuations in fission.

2.1. Results and Discussions

The cross-sections (C–S) in the fission process induced by neutrons with threshold energies range from (≈0.4 MeV) up to 25 MeV in ^{237}Np nucleus have been measured. This evaluation was done with software (TALYS-1.2) [13] and the results have been shown in Fig. 1, *a, b*. The measured cross-section in the present work has been compared with the theoretically estimated in the literature [24, 25] to get the more validity of present data. The dependence of cross-section (into mb) on the neutron energy [MeV] of ^{237}Np nucleus has been represent in Fig. 1, *a* and the symbolic indication is such as; *1* – represent to (TALYS-1.2) whereas the *2* – represent to present experimental data. It is clearly observed in Fig. 1, *a*, that the experimental data exist only below 10 MeV energy. The exact value of cross-sections has been reported around approximate 5–5.5 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 [1–3, 24, 25]. The experimental values of the ross-sections estimated in the present work with the choice of (TALYS-1.2) were found within good agreement with the corresponding theoretical literature [24, 25].

During the nuclear fission reactions, some other fissionable nuclei are formed, and these isotopes contribute to the total cross-section (C–S). The contribution of total cross-section (into mb) of fissionable nuclei derived from ^{237}Np nucleus has been depicted in Fig. 1, *b*. There are many curve like shapes in Fig. 1, *b*, one can expected that the most important part for total cross-section (C–S) is given by the compound nucleus of Neptunium (^{238}Np), where as beyond the energy approximate 7–8 MeV some other isotopes of Neptunium (Np) influence the value of total cross-section and curve like shapes become change slightly. It has been found clearly in Fig. 1, *b*. One can see this special behavior for the curves/shapes *2, 3, 4* in Fig. 1, *b*. The curve shape number (5) of Fig. 1, *b* is the same pattern as curve like shape of Fig. 1, *a*, because both them were estimated by software (TALYS-1.2) [13]. Based on the above explanation of Fig. 1, *a* and *b*, we found that the Neptunium (Np) isotopes produced in significant quantity during the nuclear fission process by neutron induced reaction, on the other hand, in the fission process of Uranium (^{235}U) and Protactinium (^{231}Pa) the isotopes



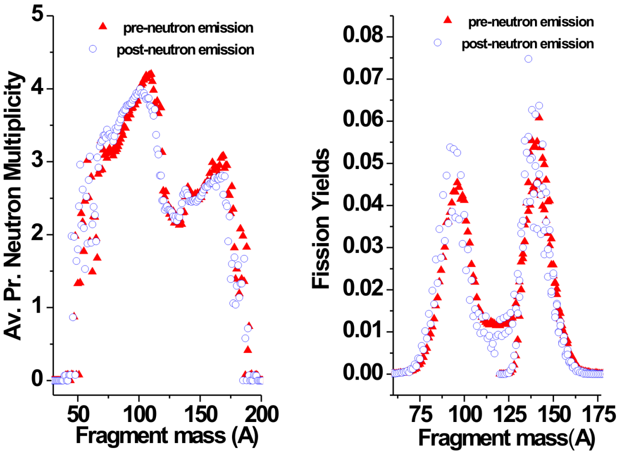
a *b*

Fig. 1. The dependence of cross-section (mb) on Neutron Energy [MeV] of ^{237}Np nucleus (*a, b*); *1* – represent to (TALYS-1.2) whereas the *2* – indicated to the present experimental data (*a*). The contribution of total cross-section (into mb) of fissionable nuclei derived from ^{237}Np nucleus and the symbolic representations are such as: *1* – for ^{238}Np; *2* – for ^{237}Np; *3* – for ^{236}Np; *4* – for ^{235}Np and the 5-indicated to (TALYS-1.2) (*b*)

produced but their contribution to the total cross-section (C–S) were neglected. This separation of the total cross-section (C–S) due to different isotopes is very useful in the evaluation of neutrons field and spectra in certain cases.

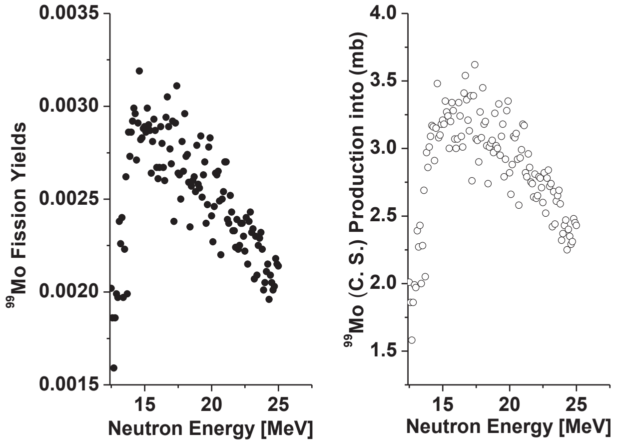
Further, other significant observables such as the mass distributions have been studied for the fission process in nuclear reactions and results reported graphically in Fig. 2, *a, b*. In this figure, first part (Fig. 2, *a*), represent the average prompt neutron emission, whereas (Fig. 2, *b*) represent the mass distribution of the fission fragments during the neutron induced reaction of ^{237}Np nucleus. The incident neutron energy (E_n) was 20 MeV during the experiment. In both figures the solid red triangles indicate to the “before neutron emission” and the blue open circles indicate to the “post neutron emission” of the fission fragments.

In Fig. 2, *a, b*/graph, the average prompt neutrons multiplicity and the mass distribution (Fission Yields) on Y-axis has been plotted as a function of fragment mass (A) on X-axis. These quantities are enough important for the description of nuclear fission process. In both the graphs, two curves with the solid red triangles and the blue open circles represent the cases of pre-neutron and post-neutron emission of the excited fragments. The mass distributions were



a *b*

Fig. 2. A pictorial representation of mass distributions of nuclear fission process (*a, b*); (*a*) – represent to average prompt neutrons multiplicity and (*b*) – represent the mass distribution (Fission Yields) as a function of fragment mass (A). In both figures the solid red triangles indicate to "before neutron emission" and the blue open circles indicate to "post neutron emission" of the fission fragments



a *b*

Fig. 3. The energy dependence of ^{99}Mo isotopes production in neutron emission; (*a*) Fission Yields and (*b*) cross-section production on the neutron energy in MeV (*a, b*)

obtained with the prediction of Brosa model [23] and neutrons emission is described by evaporation process depending on nuclear temperature, density levels and nuclear reaction mechanisms [23]. These results spots out, the scission point configuration, which includes the excitation energy at scission point and the scission point deformation plays significant role in deciding width of mass distribution.

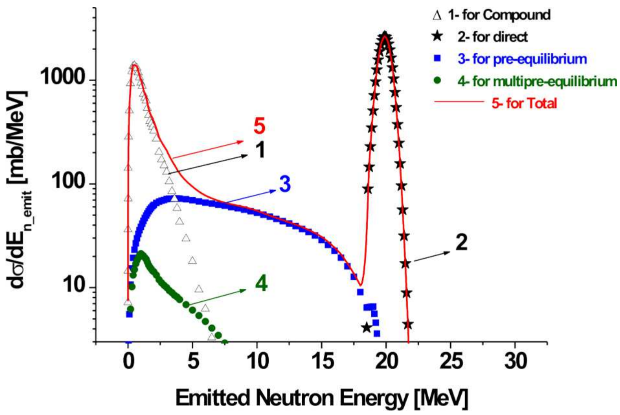


Fig. 4. The fission neutron spectra (FNS) from induced neutron at energy $E_n = 20$ MeV, shows the dependence $d\sigma/dE_n$ (into mb/MeV) as function of emitted neutron energy (MeV). The symbolic representation of separation of reaction mechanism such as; *1* – for compound, *2* – for direct, *3* – for pre-equilibrium, *4* – for the-multi-pre-equilibrium and the *5* – for overall total

The software (TALYS-1.2) [13] gives the possibility to extract data on isotopes production of interest for fission reactions chain (^{133}Xe), radioactive pollution (^{131}I) or medicine applications (^{99}Mo). The incident neutron energy dependence of the production of ^{99}Mo has been presented in Fig. 3, *a, b*. In this graph, Fig. 3, *a* represent to the ^{99}Mo, fission yields as a function of the neutron energy in MeV and Fig. 3, *b* has been plotted to get the behavior of ^{99}Mo cross-section production on the neutron energy in MeV. In both cases the results were obtained after neutrons emission of excited fission fragments.

The fission neutron spectra (FNS) are significant for nuclear reactor applications. Many workers [26] in both the fields of experiment and theory, have been drawn the spectra mainly from major actinides, such as ^{233,235,238}U(n,f) and ^{239}Pu(n,f), and also from the spontaneous fission of ^{252}Cf as a standard. For ^{237}Np, calculation of the neutron fission spectra based on the multi-mode fission model by Brosa *et al.* [23] was also reported by T. Ohsawa *et al.*, [27]. From these points of view, experimental fission spectrum data are important also for the verification of fission models.

The present studies give the neutron spectra at incident neutrons energy of 20 MeV e in the ^{237}Np(n,f) process. The fission neutron spectrum of ^{237}Np(n,f) on said energy has been depicted in Fig. 4. It is clear from the same figure that the experimental data agree fairly well with curves distributions. A separation of the contribution of different reactions mechanism was also obtained. From, Fig. 4, it is worth meaning that the main part of the spectra is given by compound process up to 8 MeV due to evaporation. And the direct nuclear mechanism was dominant during the high energies. In rest of all cases, pre-equilibrium and multi-pre-equilibrium mechanisms contribute especially at low energy when they compete with compound processes.

3. Conclusions and Final Remarks

The findings of the present work are useful in the evaluating nuclear data. This study might be very useful in modern science and technology as well as works like a bridge in between the nuclear physics and nuclear engineering worldwide. Based on the above explanation one can draw the following conclusions:

• Various parameters during the neutrons induced fission of ^{237}Np nucleus such as; cross-section (C–S), mass distributions, prompt neutrons emissions, isotopes production and neutrons spectra, were analyzed. For this study, it was necessary to design an intense neutron source based on ^{237}Np fuel.

• The evaluations of the above said observables were done with software (TALYS-1.2) for incident neutrons energy from 0.4 MeV up to 25 MeV. The exact value of cross-sections was measured approximate 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 reactions mechanism was obtained. Some important applications yields and cross-section (C–S) production of isotopes have been recorded with enough satisfactory and were found within good agreement with others.

Finally, the future experimental data obtained at LNF JINR (Dubna) for various observables such as the mass distribution, isotopes production, neutron spectra, *etc.* will help to understanding the new physics of nuclear fission mechanism. The present work will be continued also with neutron spectra theoretical modeling and experimental measurements.

The authors would like to acknowledge the keen support of this work by Frank Laboratory for Neutron Physics (FLNP) Joint Institute for Nuclear Researches (JINR), Moscow Region, Russian Federation and by the Department of Physics, Faculty of Science, University of Tabuk, Saudi Arabia.

1. I. Ruskov, A. Goverdovski, W. Furman, Y. Kopatch, O. Shcherbakov, F.J. Hambsch, S. Oberstedt, A. Oberstedt. Neutron induced fission of 237Np – status, challenges and opportunities. *EPJ Web of Conferences* **169**, 00021 (2018).
2. I. Ruskov, Yu.N. Kopatch, V.M. Bystritsky, V.R. Skoy, V.N. Shvetsov, F.J. Hambsch, S. Oberstedt, R. Capote Noy, P.V. Sedyshev, D.N. Grozdanov, I.Zh. Ivanov, V.Yu. Aleksakhin, E.P. Bogolubov, Yu.N. Barmakov, S.V. Khabarov, A.V. Krasnoperov, A.R. Krylov *et al.* TANGRA-swetup for the investigation of nuclear fission induced by 14.1 MeV neutrons. *Phys. Proc.* **64**, 163 (2015).
3. I. Ruskov, Y. Kopatch, (TANGRA Coll.); TANGRA – an experimental setup for basic and applied nuclear research by means of 14.1MeV neutrons. *EPJ Web of Conferences* **146**, 03024 (2017).
4. AFI ADCM, a digital pulse processing system for nuclear physics experiments; ADCM16- LTC, a 16-channel/14 bit/100MHz ADC board with signal processing core.
5. G. Audi, O. Berssilon, J. Blachot, A. H. Wapstra. The NUBASE evaluation of nuclear and decay properties. *Nucl. Phys. A* **729**, 3 (2003).
6. G. Audi, A.H., Wapstra, C. Thibault. The AME2003 atomic mass evaluation: (II), Tables, graphs and references. *Nucl. Phys. A* **729**, 337 (2003).
7. E.P. Shabalin, V.L. Aksenov, G.G. Komyshev, A.D. Rogov. Highly intense pulsed neutron source based on Neptunium. Preprint JINR Dubna, P13-2017-57 (2017).
8. E. P. Shabalin, M.V. Ryazin. Dynamics of power pulses in the Neptunium research Reactor, Preprint JINR Dubna, P-13-2017-69 (2017).
9. E.P. Shabalin, G.N. Pogodaev. *On optimization of fast neutrons impulse reactors* (JINR Dubna Communication, 1966) (in Russian).
10. V.L. Aksenov, V.D. Ananev, G.G. Komyshev, A.D. Rogov, E.P. Shabalin. On limit of neutron flux in pulsed neutron sources based on fission. Preprint JINR Dubna, P3-2016-90 (2016).
11. V.L. Aksenov, V.D. Ananev, G.G. Komyshev, A.D. Rogov, E.P. Shabalin. On the limit of neutron fluxes in pulsed sources based on the fission reactions. *Phys. Part. Nucl. Lett.* **14** (5), 788 (2017).
12. V.L. Aksenov. JINR Dubna, A 15-year forward look at neutron facilities in JINR. Preprint E3-2017-12 (2017).
13. TALYS-1.2 computer code. http: //www.talys.eu
14. EXFOR Nuclear Reaction Data. http: //www-nds.iaea.org/ exfor
15. A.J. Koning, S. Hilaire, M.C. Duijvestijn, "TALYS-1.0". *Proceedings of the International Conference on Nuclear Data for Science and Technology*, April 22-27, 2007, Nice, France. Edit. by O. Bersillon, F. Gunsing, E. Bauge,

R. Jacqmin, S. Leray, *EDP Sciences*, 211 (2008). URL: http: //www.talys.eu/
16. R. Vandenbosch, J.R. Huizenga. *Nuclear Fission* (Academic Press, Inc., 1973).
17. C. Waggemans. *The Nuclear Fission Process* (CRC, 1991).
18. A.J. Koning, D. Rochman. Modern nuclear data evaluation: Straight from nuclear physics to applications. *J. Korean Phys. Soc.* **59**, 773 (2011).
19. Y. Sawant, A. Saxena, R.K. Choudhury, B.K. Nayak, L.M. Pant, *et. al.* Temperature and fissility dependence of fragment mass variance in heavy ion induced fission. *Phys. Rev. C* **70**, 051602(R) (2004).
20. A.J. Koning, S. Hilaire, S. Goriely. Global and local level density models. *Nucl Phys. A* **810**, 13 (2008).
21. D.L. Hill, J.A. Wheeler. Nuclear constitution and the interpretation of fission phenomena. *Phys. Rev v* **89**, 1102 (1953).
22. W. Hauser, H. Feshbach. The inelastic scattering of neutrons. *Phys. Rev. v* **87**, 366 (1952).
23. U. Brosa, S. Grossmann, A. Muller. Nuclear scission. *Phys. Rep. Vol.* **197**, 167 (1990).
24. M. Diakaki *et al.* (HKS(JLab E05-115) Collaboration). High resolution spectroscopic study of ^{10}Be. *Phys. Rev. C* **93**, 034614 (2016).
25. R.J. Jiacoletti, W.K. Brown, H.G. Olson. Fission cross sections of neptunium-237 from 20 eV to 7 MeV determined from a nuclear-explosive experiment. *Nucl. Sci. Engin.* **48**, 412 (1972).
26. O. Iwamoto. Systematics of prompt fission neutron spectra. *J. Nuc. Sci. Technology* **45** (9), 910 (2008).
27. T. Ohsawa, *et al.* Proc. 1998 Symp. on Nuclear Data, Nov. 19–20, 1998, Tokai, JAERI, Japan, JAERI-Conf 99-002, p. 130 (1999).

Received 10.10.19

К. Опреа, М.А. Ахмед,
Дж.Х. Бейкер, А.I. Опреа

МАТЕМАТИЧНЕ МОДЕЛЮВАННЯ ІНДУКОВАНОГО НЕЙТРОНОМ ДІЛЕННЯ ЯДРА ^{237}Np

Успiхи останнiх рокiв у прикладних та фундаментальних дослiдженнях в ядернiй фiзицi потребують нових iнтенсивних джерел нейтронiв. Кiлька рокiв в ОIЯД (Дубна) ведеться розробка нових нейтронних установок для замiни реактора IBR-2, який припинить свою роботу у 2032 роцi. В деяких проектах використовується дiлення ядер, iндуковане нейтронами палива, що мiстить нептунiй. У цiй роботi ми розглянемо подiл ядра ^{237}Np. Ми знаходимо перерiз, розподiл мас, виходи iзотопiв, середню кiлькiсть випромiнених миттєвих нейтронiв, нейтроннi спектри подiлу i iншi параметри. У математичному моделюваннi частково використано теоретичнi моделi, реалiзованi у програмi TALYS-1.2, та програмнi коди, розробленi авторами. Представленi результати порiвнюються з наявними даними i становлять iнтерес для проектiв ОIЯД щодо проектування нових дослiдницьких експериментальних нейтронних установок.

Ключовi слова: подiл нейтронами, перерiзи, виходи, механiзм реакцiї, утворення iзотопiв.