Advanced Modelling of $^{238}\text{U}(\text{n},\text{f})$ in a Fast Reactor Application

Cristiana Oprea^{1,2}, Alexandru Mihul^{2,3} and Alexandru Oprea¹

¹Joint Institute for Nuclear Research (JINR), Frank Laboratory of Neutron Physics (FLNP), 6 Joliot Curie, 141980 Dubna, Moscow Region, Russia

²Romanian National Authority for Scientific Research, Mendeleev 21-25, 010362 1 Bucharest, RO

³CERN, CMS, CH-1211 Genève 23, Switzerland

Abstract. Fast neutron reactors, as a possible future solution on energy demand of human society, based on fission process of ^{238}U , request new and reliable nuclear data necessary for new generation reactors design. Fission process induced by fast neutrons on ^{238}U was investigated. Fission observables like cross sections and their uncertainties, fission fragment mass distribution, prompt neutrons emission, isomer ratios and other parameters were obtained by using Talys computer code or programs realized by authors. Then the production of isotopes like $^{135,133}\text{Xe}$, ^{99}Mo , ^{131}I , ^{89}Y as well as yields of fissile nuclei were evaluated. Obtained theoretical evaluations are compared with existing experimental data.

1 Introduction

Forecast for world energy demand predicts an increasing with about order of magnitude for the year 2100 in comparison with 2015. In the '80 years of the last century the scientists demonstrated that hydrocarbs based energy will end in a few decades. Almost the same situation is for nuclear energy produced by ^{235}U fission. Alternative new sources using solar or wind energy are still expensive and not effective [1]. The most appropriate solution for future energy challenge can be considered the fission of ^{238}U (^{8}U) nucleus [2].

^{8}U fission process in power generation IV reactors is important for a wide range of fundamental and applied researches issues [3,4]. For basic investigations, ^{8}U fission offers new information about configuration of fissionable systems near scission point, anisotropy, emitted gamma, ground states of fission product. ^{8}U fission represents also a source of new isotopes for specific applications in medicine, electronics, nuclear technologies etc.

In this paper fission of ^{8}U nucleus induced by fast neutrons starting from 7 up to 25 MeV was detailed investigated. Cross sections, fragment mass distributions, yields of some nuclides of interest, average prompt neutrons multiplicity, prompt neutrons multiplicity distributions and isomer ratios were obtained with the extensively - used reaction code Talys [5], version 1.9. The isomer ratios were calculated with an author's computer program by using cross sections from Talys. The present evaluations of a large set of fission variables are necessary in the planning and analysis of future experiments to be realized at FLNP - JINR basic facilities.

2 Codes and elements of theory

Fast neutron induced fission process on ⁸U can be described in the frame of statistical model; then cross sections can be evaluated by using of Hauser - Feshbach approach [5,6]. For fission, transmissions are calculated using Hill-Wheeler formulas [5, 7].

Fragment mass distribution, isotopes relative yields and production cross sections, average prompt neutrons multiplicity and prompt neutrons distributions were modeled by Brosa model [5,8].

⁸U isomer ratios knowledge allows extracting the spin distribution of fission fragments, levels density dependence on angular momentum and other important parameters. The loss of charged particles in simple case of thin target can be neglected, than the experimental isomer ratios are defined as:

$$R = Y_m \cdot [Y_g]^{-1} = \int_{E_{thr}}^{E_{max}} N_0 \phi_n(E) \sigma_{nf}^m(E) dE \cdot \left[\int_{E_{thr}}^{E_{max}} N_0 \phi_n(E) \sigma_{nf}^g(E) dE \right]^{-1}$$
 (1)

where: $Y_{m,g}$ = yields of isotope in isomer (m) and ground (g) states, respectively; N_0 = number of target nuclei; ϕ_n = flux of incident beam; $\sigma^{m,g}$ = cross section production of m and g states, respectively; E_{thr} = threshold energy of emergent particle; E_{max} = maximum energy of incident beam.

For default Talys run isomer and ground states production cross sections are not given for ⁸U fission as in the case of other nuclear reactions. In this case, according to Huizenga approach, it is supposed that the relative yields of isomer and ground states are proportional with spin distribution [9 - 12]. It has the form [9]:

$$P(J) = (2J + 1) (2\sigma^2)^{-1} \exp \left[- (J + 0.5)^2 (2\sigma^2)^{-1} \right]$$
 (2)

where: J = spin; σ = spin cut - off parameter.

3 Results and discussions

Results for fast neutron induced fission of ⁸U were obtained by varying a large number of Talys input data like parameters of Wood-Saxon potential (volume and surface types) and spin-orbit potential, both with real and imaginary part. A double humped potential barrier was considered. Coupled channels calculations were enabled considering rotational band and deformations available in Talys database. For residual nuclei 10 discrete levels were considered in evaluations and 40 levels for target nucleus and formed fissile nuclei. Experimental fission barriers suggested by Talys were considered [5]. The most important parameters of $n + ^8\text{U}$ channel are given in Tables 1 and 2.

Table 1. First and second barriers properties

	Height [MeV]	Width [MeV]	Type of axiality
First barrier	6.35	0.7	Triaxial + left - right asymmetry
Second barrier	5.80	0.5	Left - right asymmetry

Table 2. Optical Wood-Saxon potential parameters. Central. Surface. Spin-orbit

	Volume			Surface			Spin - orbit		
	U_V [MeV]	r_V [fm]	a_V [fm]	U_d [MeV]	r_d [fm]	a_d [fm]	U_{so} [MeV]	r_{so} [fm]	a_{so} [fm]
Real	46.56	1.245	0.660	0	0	0	5.716	1.121	0.590
Im	0.05	1.284	0.594	1.690	1.208	0.614	0.003	1.121	0.590

Total fission cross section of ^8U nucleus induced by neutrons with energy from 7 MeV up to 25 MeV is shown in Fig. 1. Results with Talys default run are in Fig. 1.a). Due to (n,xn), (n,xp) and other reactions the following excited fissile nuclei are formed: ^{236}U , ^{237}U , ^{239}U , ^{236}Pa , ^{237}Pa , ^{238}Pa , ^{232}Th , ^{233}Th , ^{234}Th , etc. The contributions of ^{236}U , ^{237}U , ^{238}U , ^{239}U fissile nuclei to the total fission cross section are shown in Fig.1.a). Fission cross sections of ^{236}Pa , ^{237}Pa , ^{238}Pa , ^{232}Th , ^{233}Th , ^{234}Th have low values and therefore are not shown in Fig 1.a). In Fig. 1.b) the total fission calculated cross section is compared with experimental data [13]. Knowledge of the contribution of the above fissile nuclei to total fission cross section is obvious important in nuclear technology. Agreement between theoretical calculations and experimental data was obtained by varying parameters of Wood-Saxon and spin-orbit optical potentials together with levels density. The experimental statistical uncertainties reduction was done by Talys using a Bayesian Monte Carlo procedure based on the EXFOR database and they were in fair agreement with the standards [14,15]. For incident energies higher than 20 MeV, the height of first and second fission barriers of $^{236,237,238,239}\text{U}$ nuclei was modified from 10% up to 30%.

Fig. 1. Total cross section (TXS). a) TXS and contribution of fissile nuclei: 1- ^{239}U ; 2- ^{238}U ; 3- ^{237}U ; 4- ^{236}U ; 5- 1+2+3+4. b) Comparison between: 1) Experiment [13]; 2) Talys (Theory)

Relative yields of fission fragment mass distribution were obtained for a large incident neutron energy up to 25 MeV applying Brosa model [5,8]. In Fig. 2 mass distribution results are shown for neutron energy, $E_n=10, 18, 25$ MeV, respectively. In Fig. 2.a) the pre-neutrons emission case is represented and in Fig. 2.b) the post-neutrons ones. With the increasing of neutron energy the asymmetry of mass distribution is reducing. Furthermore, with neutron energy, the mass distribution is enlarging, including new light and heavy fragments (see Fig 2.a) and 2.b)). In Fig. 2.c) calculations were verified for 5.5 MeV neutron energy for which the experimental data are well described [16].

Results related to prompt neutron emission are presented in Fig. 3. In this figure are selected 10, 18 and 25 MeV neutrons energy cases also. In Figs. 3a) and 3b) the dependence of average neutron multiplicity on fission fragment mass are represented before

and after neutron emission. As it is expected the number of emitted neutrons by fission fragments is increasing with incident energy. As seen in Fig. 2 with the increasing of neutron energy the mass distribution is enlarging with the fragment mass A .

Fig. 2. Mass distribution (MD). Neutron emission: a) Pre; b) Post; c) MD for 5.5 MeV; 1 - 10 MeV; 2 - 18 MeV; 3 - 25 MeV; t – Talys evaluation; e – experimental data

In Fig. 3c) prompt neutron multiplicity distributions are shown for the same energy as in Figs. 3a) and 3b). Multiplicity is rising with incident neutron energy and is limited by excitation energy. Further parameter extracted is the average prompt neutrons (nubar-prompt), $\bar{\nu}_{pr}$. For 10, 18 and 25 MeV, the neutron induced fission cross section is $\sigma_{nf} = 1020, 1406, 1482$ mb, and average prompt neutrons, $\bar{\nu}_{pr} = 4.15, 5.11, 5.86$ neutrons, respectively.

Fig. 3. Prompt neutrons. a) + b) Average prompt neutron multiplicity (APNM); a) Pre b) Post; c) Prompt neutron multiplicity distribution (PNMD). 1 - 10 MeV; 2 - 18- MeV; 3 - 25 MeV

From the large number of nuclides obtained in fission process, ^{99}Mo , ^{131}I , ^{133}Xe and ^{135}Xe isotopes were selected. The ^{99}Mo nucleus has applications of great interest in cancer treatment. The ^{131}I isotope is important in radiological protection, nuclear medicine and also can be used as industrial tracer. The last nuclide, ^{133}Xe , has many applications in medicine.

Fig. 4. Relative yields isotope production as function of incident neutron energy: a) ^{99}Mo ; b) ^{131}I ; c) ^{133}Xe ; Neutron emission: 1- Pre; 2- Post

Production of ^{99}Mo , ^{131}I and ^{133}Xe isotopes in fast neutron induced fission of ^{235}U is very low and then not calculated with a default Talys run. In a default Talys run, for reducing computing time, cross sections lower than 10^{-7} mb are not considered. For obtaining results from Fig. 4 it is necessary to extend the limit for cross section calculations up to 10^{-10} mb. In Figs. 4.a - 4.c) are represented the energy dependences of relative yields for production of ^{99}Mo , ^{131}I and ^{133}Xe isotopes before and after neutron emission.

Fig. 5. ^{135}Xe production. Energy dependence of: a) Relative yields; b) Cross section; Neutron emission: 1 - Pre; 2 - Post.

Another isotope of interest for nuclear technology is ^{135}Xe nucleus, which is a major fission product and an important neutron absorber. In Fig. 5 the energy dependence of relative yields and cross section production are represented before and after neutron emission. The results from Fig. 5 are obtained with default Talys precision. Relative yields for ^{99}Mo , ^{131}I , ^{133}Xe nuclei (Figs. 4.a - c) are much lower than in the case of ^{135}Xe (Fig. 5.a). The same situation is in the case of cross section production. Cross section for ^{135}Xe is of order of millibarns (Fig. 5.b) and for ^{99}Mo , ^{131}I and ^{133}Xe isotopes is about 10^{-3} mb or lower.

The relative yields and cross sections evaluated with Talys for a large number of isotopes were further used to calculate isomer ratios.

Isomer ratios in $^8\text{U}(n,f)^{133m,g}\text{Xe}$ process with neutrons from 7 up to 20 MeV were evaluated. Fission cross sections are taken from Talys. Spin distributions are evaluated using expression (2) with parameters from Talys database. The flux of incident neutrons is considered proportional with $(1/E_n)^{0.9}$, characteristic for pulsed neutron source available at FLNP. Spin, parity, time of life of $^{133m,g}\text{Xe}$ states are taken from [18]. Using the procedure from [11,12], the calculated isomer ratio is:

$$R = 3.95 \pm 0.42 \quad (3)$$

The error from (3) comes from the fact that integrals are sums with a 0.2 MeV step. The obtained isomer ratio is in agreement with similar experimental data [18].

Another nucleus of interest is Yttrium because it has no natural isotopes [19,20]. Artificial Yttrium isotopes with masses from 87 up to 111 can be produced in ^8U fission with fast neutrons. Their production depends on the incident neutron flux and therefore natural ^{89}Y nucleus can be used in fast neutron threshold detectors. For the calculation of relative yields and cross sections of Yttrium isotopes, the cross section lower limit was extended from 10^{-7} mb to 10^{-10} mb. For example, production cross section of ^{89}Y after neutron emission is $\sigma_{n\gamma} = 5.36 \times 10^{-4}$ mb for $E_n = 25$ MeV. Relative yields and cross section production of ^{98}Y are shown in Fig 6.a) and 6.b). Analysis of $^{89}\text{Y}(n,xn)$ processes with fast neutrons was done in [21] and $^{89}\text{Y}(n,xn)$ cross section results are represented in Figs. 6.c).

Fig.6. Yttrium isotopes production in $^8\text{U}(n,f)$. a) Relative yields of ^{98}Y ; b) Production cross section of ^{98}Y . Neutron emission: 1 - pre; 2 - post. $^{89}\text{Y}(n,xn)$, $x=1,2,3,4$. Production of: 1 - ^{89}Y ; 2 - ^{88}Y ; 3 - ^{87}Y ; 4 - ^{86}Y . c) Cross section production of Yttrium isotopes in (n,xn) reactions [21]. 1 - ^{89}Y ; 2 - ^{88}Y ; 3 - ^{87}Y ; 4 - ^{86}Y

4. Conclusions

Fast neutron induced fission observables on ^8U were investigated. The theoretical modeling and calculations were performed for cross sections, mass distributions, average prompt neutron multiplicity, prompt neutron multiplicity distributions, and isotopes production necessary in applications for neutrons with energy from 7 MeV up to 25 MeV were obtained. The isomer ratios using fission cross sections evaluated with Talys and the statistical approach from [11] were calculated. Yttrium isotopes production in neutron induced fission of ^8U nucleus as well as $^{89}\text{Y}(n,xn)$ reactions necessary for fast neutron threshold detectors were analyzed.

This paper stated the need for new fission experimental data, improvement of the analyses of existing ones and computer modeling of fission measured observables. In this respect future measurements are planned to be realized at JINR Dubna basic facilities at the new neutron source IREN from FLNP and Microtron MT-25 from Flerov Laboratory for Nuclear Reactions.

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