

Experimental estimation of the “scission” neutron yield in the thermal neutron induced fission of ^{233}U and ^{235}U

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Abstract. The analysis of angular and energy distributions of prompt neutrons from the thermal neutron induced fission of ^{233}U and ^{235}U measured recently in the WWR-M research reactor (Gatchina, Russia) have been performed. The yield of “scission” neutrons has been estimated by comparing the measured distributions with calculations within the model of emission of neutrons from completely accelerated fragments. Besides taking into account “scission” neutrons, for the best description of measured angular and energy distributions of fission neutrons, the calculation should be performed under the assumption that neutrons with a higher (7-9%) probability are emitted along the fission axis in the center-of-mass system of fission fragments.

1 Introduction

Up to now many theoretical and experimental works were performed to investigate the low energy nuclear fission. A special attention was given to the details of the prompt fission neutron (PFN) emission: spectra and multiplicities, their dependence on fission fragment (FF) characteristics and all possible correlations between reaction products. These data are used widely for the construction of nuclear reactors and applied for the development of non-destructive methods of nuclear safety and for the control of non-proliferation of nuclear materials. Despite considerable progress achieved in description of the properties of PFNs [1]-[3], some discrepancies still exist between the experimental data and results of theoretical calculations. The observed differences are related to both the limitations of theoretical models used to describe the properties of PFNs and the absence of necessary experimental data.

Since for theoretical descriptions it is usually assumed that PFNs are emitted from the fission fragments fully accelerated in the mutual Coulomb field, a special attention deserves the question of the existence of scission neutrons emitted from fissioning nucleus during its evolution from equilibrium deformation to the scission point. The search for scission neutrons and investigations of their properties are complicated by impossibility of discriminating in an experiment between these neutrons and neutrons emitted from the fully accelerated fission fragments. These investigations can only be conducted by comparing measured distributions of the PFNs and model calculations performed by assuming that all PFNs are emitted from fully accelerated fragments. In this case, it can be found the yield of neutrons whose emission mechanism differs from evaporation of neutrons from fully accelerated fragments (for example, emission of neutrons before or at the

time of scission of a fissioning nucleus or in the process of acceleration of produced fission fragments), such neutrons are called as “scission” neutrons. A final conclusion about validity of any mechanism of such neutron emission can be done only after detailed comparison of predictions made in the framework of different theoretical models with the dependences reliably observed in the experiment.

Experimental studies dedicated to search for “scission” neutrons are limited to spontaneous fission of ^{252}Cf and thermal neutron-induced fission of ^{235}U . Estimates of the contribution of “scission” neutrons obtained from the analysis of independent experimental data range from 1 to 20% of the total number of neutrons per fission event (for example, see ref. [4]). In most of these works only the final conclusions are given without presenting all used input parameters of the model calculation and measured data in digital format.

In 2008-2015, at the NRC KI - PNPI, a series of investigations was carried out, intended to study the PFN angular and energy distributions in thermal-neutron induced fission ^{233}U , ^{235}U , ^{239}Pu and spontaneous fission ^{252}Cf . The results of these investigations are given in publications [3]-[7], while the numerical data obtained are presented in the EXFOR data base [8]. Note that before these works were started in PNPI, there were only two reported sets of data [9],[10] for $^{252}\text{Cf}(s.f)$ and one [11] for $^{235}\text{U}(n_{th},f)$ that can be used for joint analysis.

In the present report are given the results of analysis of the PFN angular and energy distributions from thermal-neutron induced fission ^{233}U and ^{235}U measured in NRC KI - PNPI. During this analysis, to calculate model PFN distributions under an assumption that all neutrons are emitted from the fully accelerated fragments, it was used a method free of any assumptions about the PFN properties analogous to those used in refs [6],[12].

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2 Description of the PNPI's experiment

The experiments have been done at the radial neutron channel N7 of the research reactor WWR-M (PNPI). The spectra of PFN were measured simultaneously for 11 angles between the direction of emission of a neutron and the direction of motion of a light fragment. Taking into account the real geometry and angular resolution of experimental set-up, these angles were 8.8°, 19.9°, 36.8°, 54.5°, 72.2°, 90°, 107.8°, 125.5°, 143.2°, 160.1°, and 171.2°. The energies of PFNs and velocities of fission fragments were determined using the time-of-flight (TOF) technique. The detailed description of measurement technique and data processing has been given elsewhere [12],[13].

At present, the commonly accepted international standard for measuring PFN spectra is established by the total PFN spectrum of spontaneous ^{252}Cf fission [15]. For this reason, measurements of the PFNs distributions from thermal neutron-induced fission of ^{233}U and ^{235}U were performed in comparison with the spectra of spontaneous ^{252}Cf fission under identical experimental conditions. The measurements were carried out in repeated cycles. Each cycle included sequential measurements of the PFNs distributions from $^{233,235}\text{U}(n_{th},f)$ and $^{252}\text{Cf}(s.f)$ reactions. The ^{252}Cf target was located in the reaction chamber at the same position as the target under investigation.

3 Model

The yield of “scission” neutrons is estimated by comparing the measured distributions of PFNs with model calculations under the assumption that all PFNs are emitted from fully accelerated fragments. Within this model, the spectra of PFNs in the laboratory system can be calculated using the spectra of PFNs in the center-of-mass system of a fission fragment for all possible combinations of fragment masses and kinetic energies. Nevertheless, to construct model spectra of PFNs, it is possible to use the approximation of two fragments according to which PFNs are emitted from two fully accelerated fragments with fixed average masses and energies (see [3], [4]).

In this work the spectra of PFNs in the center-of-mass system of fission fragment were obtained in the approximation of two fragments using the data obtained in PNPI. The spectra of neutrons measured for six selected detection angles relative to the light fragment escape direction $\Omega = 8.8^\circ, 19.9^\circ, 36.8^\circ$ ($\Omega < 40^\circ$) and $\Omega = 141.3^\circ, 160.1^\circ, 171.2^\circ$ ($\Omega > 140^\circ$) were used. In this case, it is possible to obtain almost unlimited (in the low-energy range) PFN spectrum in the center-of-mass system of fission fragment and, hence, to minimize the uncertainty of model calculations related to uncertainty of the shape of spectra and the number of PFNs emitted from light and heavy fragments.

The main advantage of the discussed model is that, after the average energy per nucleon is determined, the model includes only one varying parameter A_2 responsible for the anisotropy of the angular distribution of PFNs in the center-of-mass system of the fission fragment. The analysis of ^{252}Cf data showed that the best description of the total spectrum of PFNs is achieved simultaneously

with the best description of the partial spectra of PFNs at $A_2 = 0.04$ [4]. For example, Fig. 1 shows the calculated total spectrum of PFNs in a comparison with standard spectrum. In order to indicate more clearly the existing differences, the spectra of PFNs are presented as the ratio to the Maxwell distribution $M(T_M, E)$ ($T_M = 1.42$ MeV):

$$\mu(E) = \frac{\Phi(E)}{\bar{\nu}_p \cdot M(T_M, E)} = \frac{\Phi(E) \cdot \sqrt{\pi T_M^3}}{2\bar{\nu}_p \sqrt{E} \cdot \exp(-E/T_M)} \quad (1)$$

Here $\bar{\nu}_p$ is an average number of PFN per fission event.

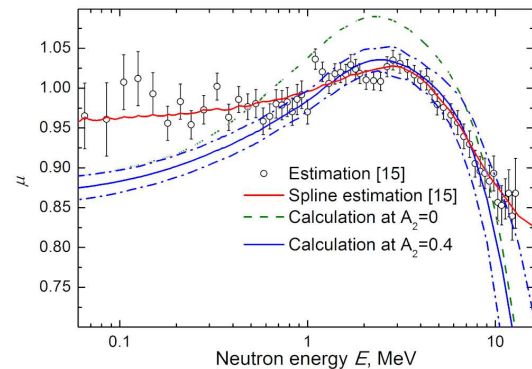


Figure 1. Ratio of the total PFN spectrum of $^{252}\text{Cf}(s.f)$ to the Maxwell distribution: circles and red curve passes through them show the standard spectrum from [15], green dash curve shows the result of calculations at $A_2 = 0$, and blue curve (uncertainties are limited by blue dash-dot curves) shows the result of calculation at $A_2 = 0.04$.

It should be emphasized that the description of PFN distributions observed in the experiment is not achieved with models (program codes) proposed for the numerical description of PFN characteristics [3]. Fig. 2 presents the results of calculations performed using some of these codes for the total PFN spectrum of $^{235}\text{U}(n_{th},f)$ with the same set of input parameters.

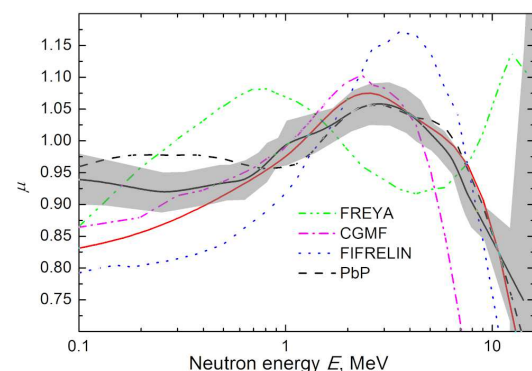


Figure 2. Ratio of the total PFN spectrum of $^{235}\text{U}(n_{th},f)$ to the Maxwell distribution ($T_M = 1.32$ MeV): black curve corresponds to evaluated data (GMA-approximation [3], uncertainty is shaded by grey color); FREYA, CGMF, FIFRELIN, and PbP are models (program codes) from [3] used for calculation of PFN properties; red curve shows the result of calculation at $A_2 = 0.04$.

4 Results

In this work the preliminary processing and the determination and applying of corrections were done in a similar way for $^{235}\text{U}(n_{th},f)$, $^{233}\text{U}(n_{th},f)$ and $^{252}\text{Cf}(s,f)$ [4]. The parameters of the model used to calculate the angular and energy distributions of PFNs by assuming that they were emitted from the fully accelerated fragments were optimized to produce the best description of all data obtained in NRC KI - PNPI [3].

The total PFN spectra for $^{235}\text{U}(n_{th},f)$ and $^{233}\text{U}(n_{th},f)$, as well as the total PFN spectrum of $^{252}\text{Cf}(s,f)$, are presented in Figs. 1, 3, 4 reveal some differences between the shape of experimental (estimated) PFN spectra and that of the model calculation in the range of PFN energies below 0.6 MeV. Also for neutron detection angle $\Omega = 90^\circ$, a neutron excess is observed relative to the model calculations: $4.6 \pm 2.7\%$, $5.9 \pm 2.8\%$, and $7.6 \pm 2.8\%$ for $^{233}\text{U}(n_{th},f)$, $^{235}\text{U}(n_{th},f)$, and $^{252}\text{Cf}(s,f)$, respectively. The observed deviations can be treated as evidence of the existence of “scission” neutrons and, hence, their yield and spectrum can be determined.

As an example, Fig. 5 shows the angular dependence of the yield of “scission” neutron from $^{235}\text{U}(n_{th},f)$ fission calculated by Carjan et.al. [16], [17] in a framework of dynamical scission model under different initial condition and that obtained in this work as the difference between yields of PFNs measured and one calculated under assumption that all PFNs was emitted from fully accelerated fragments. In Fig. 6 it is presented the spectrum

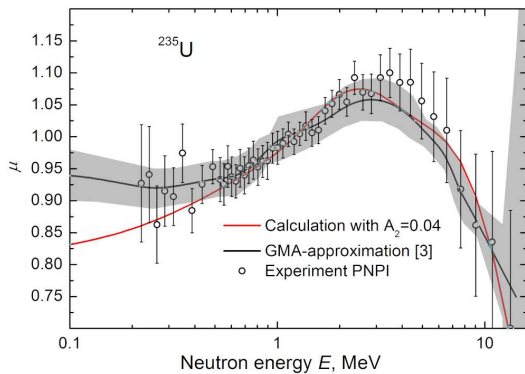


Figure 3. Ratio of the total PFN spectrum of $^{235}\text{U}(n_{th},f)$ to Maxwell distribution ($T_M = 1.32$ MeV): circles present data of measurements [13]; (black solid curve) estimated data (GMA-approximation [3]) with uncertainty area indicated by gray shade; (bottom [red] curve) the result of calculation at $A_2 = 0.04$.

of $^{235}\text{U}(n_{th},f)$ “scission” neutrons obtained in two different ways. In the first, the desired spectrum was defined as the difference between the measured and model neutron spectra for angles Ω close to 90° ($\Omega = 72.2^\circ, 90^\circ$ and 108.8°). In the second, the total spectrum of “scission” neutrons was defined as the difference between the reference total PFNs spectrum (GMA-approximation [3]) and model calculation. To compare the two estimates, the “scission” neutron spectrum obtained in the first way was multiplied by 4π (it was assumed that the distribution of “scission”

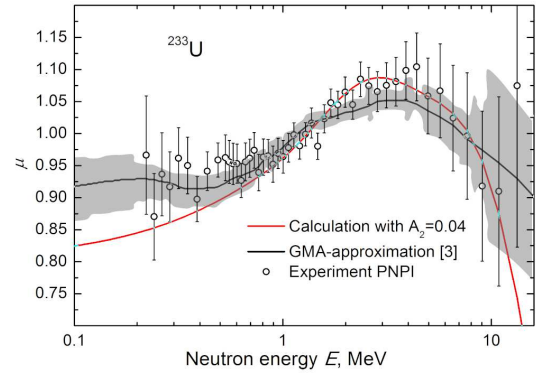


Figure 4. Ratio of the total PFN spectrum of $^{233}\text{U}(n_{th},f)$ to Maxwell distribution ($T_M = 1.32$ MeV). Denotations are the same as in Fig. 3. Circles present data of measurements [14].

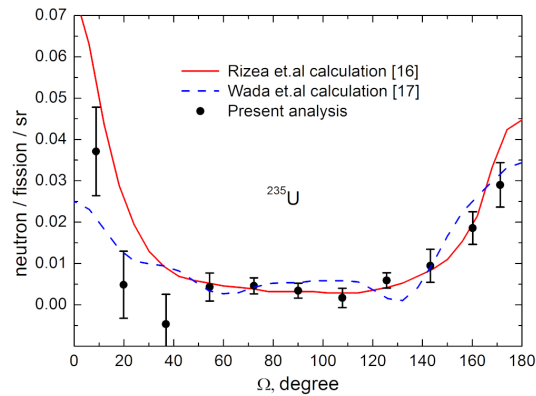


Figure 5. Angular dependence of the neutron yield for $^{235}\text{U}(n_{th},f)$: points - the difference between the measured and model neutron yields, the errors include statistical and model parameters uncertainties. Straight curve - calculation [16] performed on a sphere of radius $R=30$ fm and for time T after the moment of scission is $2 \cdot 10^{-21}$ sec. Dash line - calculation [17] - $R=50$ fm, $T=4 \cdot 10^{-21}$ sec and the effects of the scattering and re-absorption by the fission fragments on the angular distribution of scission particles were included into calculation.

neutrons in the laboratory system was isotropic). A comparison of the spectra obtained in this manner shows the agreement (within the errors of the experimental data) between the results from estimates performed in different ways.

Since the relative contribution from “scission” neutrons should be largest at angles Ω close to 90° , the yield of these neutrons from the fission of the investigated nuclei was estimated using the spectrum obtained in the first way with least squares fit by functions:

$$p_S(E) = p_0 \cdot \frac{E}{T_0^2} \cdot \exp\left(-\frac{E}{T_0}\right) \quad (2)$$

$$p_S(E) = p_0 \cdot \frac{E}{T_0^2} \cdot \exp\left(-\frac{E}{T_0}\right) + p_1 \cdot \frac{E}{T_1^2} \cdot \exp\left(-\frac{E}{T_1}\right) \quad (3)$$

The results of these approximations are given in Table 1.

The obtained “scission” neutron spectrum for spontaneous ^{252}Cf fission in comparison with the calculation tak-

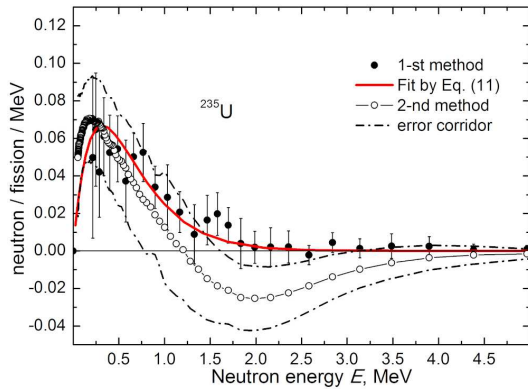


Figure 6. Spectrum of “scission” neutrons from the $^{235}\text{U}(n_{th},f)$ fission: points - first approach, the errors are statistical; circles - second approach (see text). The interval of errors arising from uncertainty of the reference PFN spectrum is limited by the dotted-and-dashed lines. The curve is the approximation by function (2).

Table 1. Main characteristics of “scission” neutrons

	$^{235}\text{U}(n_{th},f)$	$^{235}\text{U}(n_{th},f)$
Approximation using Eq. (2)		
Yield, %	1.5 ± 0.6	1.8 ± 0.6
Average energy, MeV	0.53 ± 0.08	0.47 ± 0.05
Approximation using Eq. (3)		
Yield, %	2.7 ± 0.8	2.6 ± 0.8
Average energy, MeV	1.7 ± 0.2	1.4 ± 0.2

ing into account non-adiabatic effects in the interaction of the single-particle degrees of freedom with fragment acceleration [18] is shown in Fig. 7. It is seen that the spectrum of “scission” neutrons can be adequately described, as well as, angular dependence of their yield by assuming that the neutron excess observed in the experiment was due to the dynamic effects in nuclear fission.

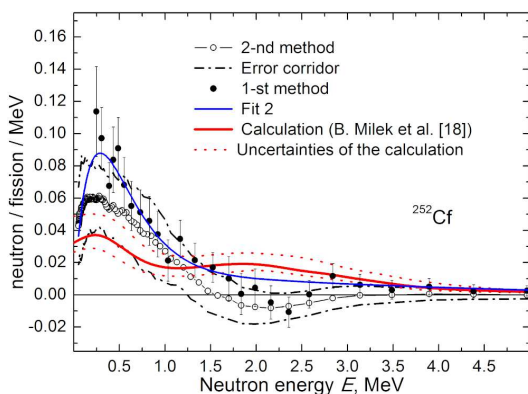


Figure 7. Spectrum of “scission” neutrons from the $^{252}\text{Cf}(sf)$ fission. Denotations are the same as in Fig. 6. The blue curve is the result of approximation using function (3). The solid line (red) is the scission neutron yield calculated in ref. [18]. The dotted line shows the errors of calculation [18].

5 Conclusion

Detailed analysis of the PNPI’s data shows that the model calculations must be performed using the anisotropy of prompt neutron emission in the centre-of-mass system of fission fragment (the PFNs are emitted along the fission axis with a higher probability than perpendicular to it, $\psi(0^\circ)/\psi(90^\circ) \approx 1.07-1.09$). Some difference between calculation within the model of neutron emission from fully accelerated fragments and experimental data is observed for partial PFNs distributions as well as for total PFNs spectra. This difference can be eliminated by assuming that there are 2-4% of “scission” neutrons. The nature of the observed neutron excess can be determined after a thorough comparison of the experimental data and the calculations using theoretical models that allow for possible PFN emission mechanisms in fission.

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