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^{233}U(n,F) Prompt Fission Neutron Spectra

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Simultaneous analysis of measured data for ^{235}U(n,F), ^{239}Pu(n,F) and ^{233}U(n,F) maintains stronger justification for the predicted prompt fission neutron spectra (PFNS) of ^{233}U(n,F). Pre–fission neutrons influence the partitioning of fission energy between excitation energy and total kinetic energy of fission fragments. For the reactions ^{233}U(n,F) and ^{235}U(n,F) shape of prompt fission neutron spectra strongly depends on relative positions of (n,xnf) and (n,xn) reaction thresholds. The correlation of these peculiarities with emissive fission contributions (n,xnf) to the $\sigma_{n,F}$ and competition of reactions ($n,n\gamma$) and $(n,xn)^{1,..x}$ is established. Exclusive neutron spectra $(n,xnf)^{1,..x}$ are consistent with $\sigma_{n,F}$ of ^{233}U(n,F) and ^{232}U(n,F). Initial model parameters for ^{233}U(n,F) PFNS are fixed by description of PFNS of ^{233}U(n_{th},F) and ratios of PFNS of ^{233}U(n_{th},F)/ ^{235}U(n_{th},F) and ^{239}Pu(n_{th},F)/ ^{233}U(n_{th},F). We predict the ^{233}U$(n,xnf)^{1,..x}$ exclusive pre-fission neutron spectra, exclusive neutron spectra of ^{233}U$(n,xn)^{1,..x}$ reactions, total kinetic energy TKE of fission fragments and products, observed and partials of average prompt fission neutron number and observed PFNS of ^{233}U(n,F).

INTRODUCTION

Fissile nuclides ^{233}U may build-up in breeder or hybrid reactors. Nuclear data for ^{233}U+n interaction, with the exception for $\sigma_{n,F}$ data, are scarce, especially as regards PFNS $S(\varepsilon,E_n)$ of ^{233}U(n,F) in the range of ^{233}U$(n,xnf)^{1,..x}$ reaction. Data on PFNS $S(\varepsilon,E_n)$ at E_n~14.3 MeV [1] remained the only available before long. Since model analysis in [1] was then over-simplified, relative contributions of pre- and post-fission neutrons in [1] disagree with later predictions [2, 3]. Measured data at $E_n \sim E_{th}$ [4], E_n~0.55 MeV [5], and data [1] as well, were abandoned in all versions of BROND/ROSFOND, ENDF/B, JEFF and JENDL data libraries. The data of recent PFNS measurements for ^{233}U(n_{th},f) [6], ^{235}U(n_{th},f) and ^{239}Pu(n_{th},f) [6, 7] are discrepant with the data of [4]. Lumping [4, 6, 7] data in a spline fitting procedure of [8] would change predicted PFNS shapes of ^{233}U(n_{th},f), ^{235}U(n_{th},f) and ^{239}Pu(n_{th},f) drastically (see Fig. 1).

In differential PFNS data for ^{235}U and ^{239}Pu for E_n~1.5–20 MeV and ε~0.01–10 MeV [9–11] strong variations of average PFNS energies $\langle E \rangle$ were observed around (n,xnf) thresholds. $\langle E \rangle$ is rough signature of PFNS, however it was established in [9–11] that the relative amplitude of $\langle E \rangle$ variation in case of ^{239}Pu(n,F) reaction is much weaker than in case of ^{235}U(n,F). That is due to influence of (n,xnf) reactions on fission observables when fission is preceded by pre-fission neutrons. In case of ^{233}U(n,F) reactions similar variations of $\langle E \rangle$ were predicted in [2, 3, 8]. We intend to predict PFNS of ^{233}U(n,F) at E_n~ E_{th}–20 MeV.

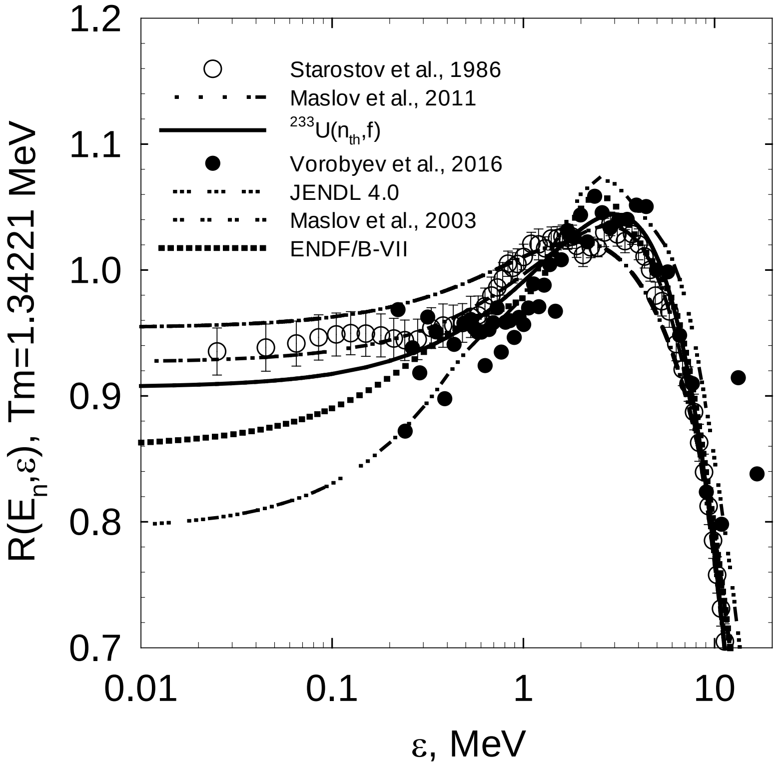

Fig.1. Prompt fission neutron spectra of ^{233}U(n_{th},f) relative to Maxwellian, $\langle E \rangle$ = 2.0564 MeV.

^{233}U(n,f) PROMPT FISSION NEUTRONS

Recent PFNS data for ^{233}U(n_{th},f) [6] added even more controversy: at range 0.02<ε<5 MeV they support the evaluations of ENDF/B–VII [12] and JENDL-4.0 [13], while PFNS of both libraries disagree with data [4]. Data [4] are presented as spline approximation of [8], which summons empirical features of consistent analysis of ^{233}U(n_{th},f), ^{235}U(n_{th},f), ^{239}Pu(n_{th},f) and ^{252}Cf(sf) measured PFNS data [4]. In the energy range 5<ε<11 MeV data of [6] for ^{233}U(n,f) support the evaluation of [8], which is based on data [4] fitting at 0.02<ε<9.3 MeV.

The comparison of PFNS measured data [4, 6, 7, 9–11] for ^{233}U(n,f), ^{235}U(n,f) and ^{239}Pu(n,f) in the range E_{th} <E_n< E_{nnf} shows that enhanced soft neutron yield, $\varepsilon \lesssim 1$ MeV is a common feature except PFNS of [6, 7]. In [6, 7] PFNS of ^{233}U(n_{th},f), ^{235}U(n,f) and ^{239}Pu(n,f) were measured relative to spontaneous fission neutron spectra (SFNS) of ^{252}Cf(sf). After various correction are applied to get absolute PFNS values, a number of systematic errors/uncertainties may appear, while the uncorrected cross ratios of various ^{233}U(n_{th},f), ^{235}U(n_{th},f) and ^{239}Pu(n_{th},f) PFNS pairs might be quite sterile in that respect.

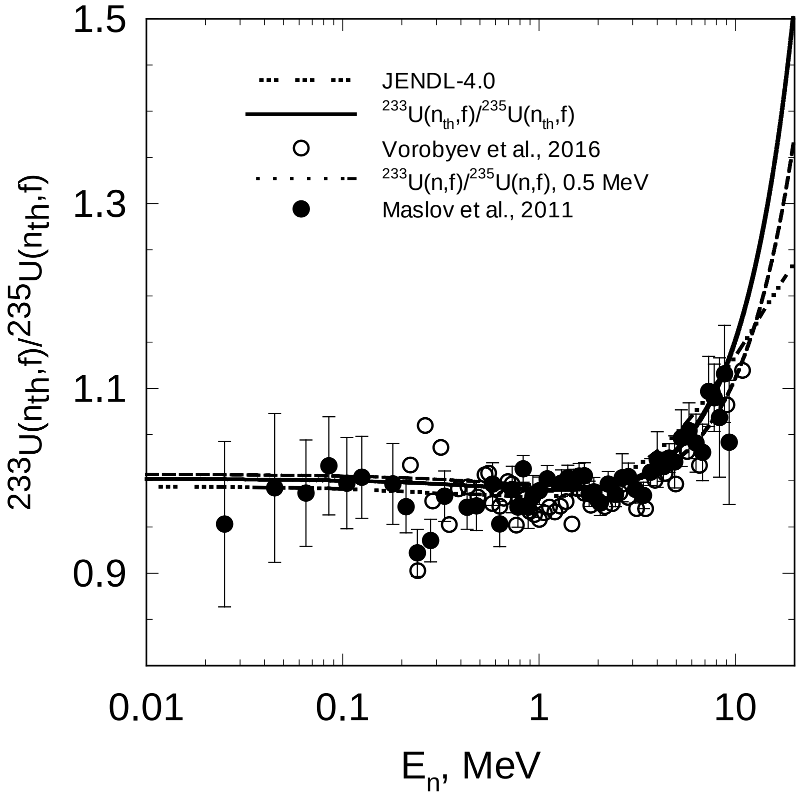

Fig. 2. Ratio of PFNS of ^{233}U(n_{th},f) and ^{235}U(n_{th},f) for thermal neutron-induced fission.

The ratios of PFNS for ^{239}Pu(n_{th},f)/^{233}U(n_{th},f) and ^{233}U(n_{th},f)/^{235}U(n_{th},f) [6, 7] and [4], contrary to absolute PFNS of ^{233}U(n_{th},f), ^{235}U(n_{th},f) and ^{239}Pu(n_{th},f), quite agree with each other (Fig. 2). At E_n~E_{th} and E_n~0.5 MeV the ratios of calculated PFNS of ^{239}Pu(n,f) and ^{235}U(n,f) in the range 0.01<ε <10 MeV weakly depend on the E_n. Calculated PFNS ratios of [2, 3, 8, 14], as well as present calculation, at E_n~E_{th} and E_n ~0.5 MeV almost coincide with PFNS ratios ^{239}Pu(n_{th},f)/^{233}U(n_{th},f) and ^{233}U(n_{th},f)/^{235}U(n_{th},f) of [4, 6, 7]. It might be concluded that the hardest prompt fission neutrons are emitted in ^{239}Pu(n_{th},f) reaction, while the softest PFNS is that of ^{235}U(n_{th},f), PFNS of ^{233}U(n_{th},f) takes intermediate position. Renormalization of model parameters at E_n~E$_{th}$ after fitting data on total kinetic energy of fission fragments [15] amounts to rather small changes of PFNS: for ^{239}Pu(n, f) a decrease by ~2–3% at ε <1 MeV, for ^{235}U(n, f) and ^{233}U(n,f) PFNS - shifts by ~1–2% [2, 3].

^{233}U(n,xnf) PROMPT FISSION NEUTRONS

Pre–fission neutrons emerging when $E_n \gtrsim E_{nnf}$, influence the PFNS $S(\varepsilon,E_n)$ shape, total kinetic energy of fission fragments E_F^{pre} and fission products E_F^{post} , prompt fission neutron number $\nu_p(E_n)$, mass distributions and other fission observables. PFNS $S(\varepsilon,E_n)$ is a superposition of exclusive spectra of pre-fission neutrons, $(n,nf)^1$, $(n,nf)^1$, $(n,2nf)^{1,2}$, $(n,3nf)^{1,2,3}$–$d\sigma^k_{nxnf}(\varepsilon,E_n)/d\varepsilon$ (x=0, 1, 2, 3; k=1,…,x), index x denotes the fission chance of $^{234\text{-}x}$U and spectra of prompt fission neutrons, emitted by fission fragments, $S_{A+1\text{-}x}(\varepsilon,E_n)$:

$$
\begin{aligned}
&S(\varepsilon,E_n)=\tilde{S}_{A+1}(\varepsilon,E_n)+\tilde{S}_A(\varepsilon,E_n)+\tilde{S}_{A-1}(\varepsilon,E_n)+\tilde{S}_{A-2}(\varepsilon,E_n)=\\
&\nu_p^{-1}(E_n)\cdot\{\ \nu_{p1}(E_n)\cdot\beta_1(E_n)S_{A+1}(\varepsilon,E_n)+\nu_{p2}(E_n-\langle E_{nnf}\rangle)\beta_2(E_n)S_A(\varepsilon,E_n)+\\
&+\beta_2(E_n)\frac{d\sigma^1_{nnf}(\varepsilon,E_n)}{d\varepsilon}+\nu_{p3}(E_n-B_n^A-\langle E^1_{n2nf}\rangle-\langle E^2_{n2nf}\rangle)\beta_3(E_n)S_{A-1}(\varepsilon,E_n)+\beta_3(E_n)\cdot\\
&\left[\frac{d\sigma^1_{n2nf}(\varepsilon,E_n)}{d\varepsilon}+\frac{d\sigma^2_{n2nf}(\varepsilon,E_n)}{d\varepsilon}\right]+\nu_{p4}(E_n-B_n^A-B_n^{A-1}-\langle E^1_{n3nf}\rangle-\langle E^2_{n3nf}\rangle-\langle E^3_{n3nf}\rangle)\cdot\\
&\beta_4(E_n)S_{A-2}(\varepsilon,E_n)+\beta_4(E_n)\left[\frac{d\sigma^1_{n3nf}(\varepsilon,E_n)}{d\varepsilon}+\frac{d\sigma^2_{n3nf}(\varepsilon,E_n)}{d\varepsilon}+\frac{d\sigma^3_{n2nf}(\varepsilon,E_n)}{d\varepsilon}\right]\}.
\end{aligned}
\qquad (1)
$$

In equation (1) $\tilde{S}_{A+1-x}(\varepsilon,E_n)$ is lumped contribution of *x*-chance fission to the observed PFNS $S(\varepsilon,E_n)$, $\langle E^k_{nxnf}\rangle$ – average energy of exclusive pre-fission neutron of $(n,xnf)^{1,..x}$ reaction, spectra $S(\varepsilon,E_n)$, $S_{A+1-x}(\varepsilon,E_n)$ and $d\sigma^k_{nxn}(\varepsilon,E_n)/d\varepsilon$ are normalized to unity, $\beta_x(E_n)=\sigma_{n,xnf}(E_n)/\sigma_{n,F}(E_n)$ is the contribution of *x*-th fission chance $\sigma_{n,xnf}(E_n)$ to $\sigma_{n,F}$, $\nu_p(E_n)$ is the average number of prompt fission neutrons, $\nu_{px}(E_{nx})$ – average number of prompt fission neutrons, emitted by $^{234-x}$U nuclides. PFNS, of neutrons, emitted from the fragments, $\tilde{S}_{A+1-x}(\varepsilon,E_n)$, as proposed in [16], were approximated by the sum of two Watt [17] distributions with different temperatures, the temperature of light fragment being higher.

The differential measured PFNS at $E_n \gtrsim E_{nnf}$ are also susceptible to systematic errors of various origin. In ratios of PFNS, especially of draft PFNS data, before corrections for the backgrounds, etc., these errors may be partially canceled [18, 19]. Figure 3 shows the ^{239}Pu(*n,F*)/^{235}U(*n,F*) and ^{233}U(*n,F*) /^{235}U(*n,F*) ratios of PFNS for E_n~ 7÷8 MeV. The averaged ^{233}U(*n,F*)/^{235}U(*n,F*) ratio is very much different from that of ^{239}Pu(*n,F*)/^{235}U(*n,F*). The ^{239}Pu(*n,F*)/^{235}U(*n,F*) ratios of differential PFNS at E_n~ 7, 7.5 and 8 MeV mildly, bur significantly, fluctuate around averaged value. The respective ^{233}U(*n,F*)/^{235}U(*n,F*) ratios of PFNS also deviate from those of ^{239}Pu(*n,F*)/^{235}U(*n,F*). That is due to differing shapes of exclusive pre-fission (*n,nf*) spectra and $\beta_x(E_n)$ values. In the energy range of E_n~ 6÷7 [19] the fluctuations of PFNS at E_n ~ 6, 6.5 and 7 MeV are more pronounced for both ^{239}Pu(*n,F*)/^{235}U(*n,F*) and ^{233}U/^{235}U(*n,F*) ratios, since competition of (*n,nf*), (*n,2n*) and (*n,nγ*) depends on excitation energy. The observed PFNS of ^{233}U(*n,F*) and ^{235}U(*n,F*) are similar, as the increase of contribution of ^{233}U(*n,nf*) is accompanied by decrease of ^{233}U(*n,f*) reaction contribution $S_{A+1}(\varepsilon,E_n)$.

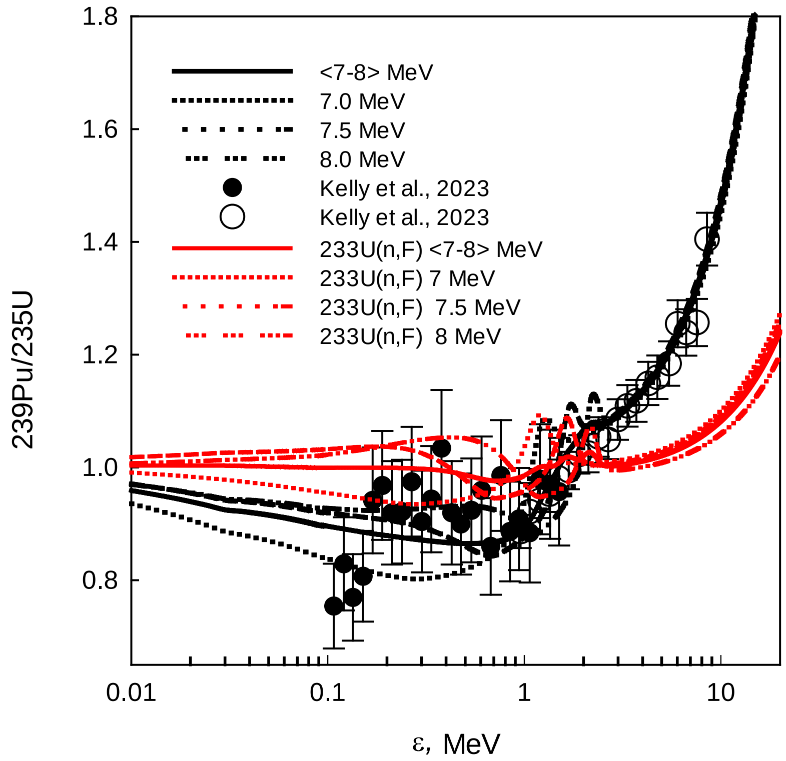

Fig.3. Ratios of PFNS ^{233}U(n,F)/^{235}U(n,F) and ^{239}Pu(n,F)/^{235}U(n,F) at E_n~6÷7 MeV.

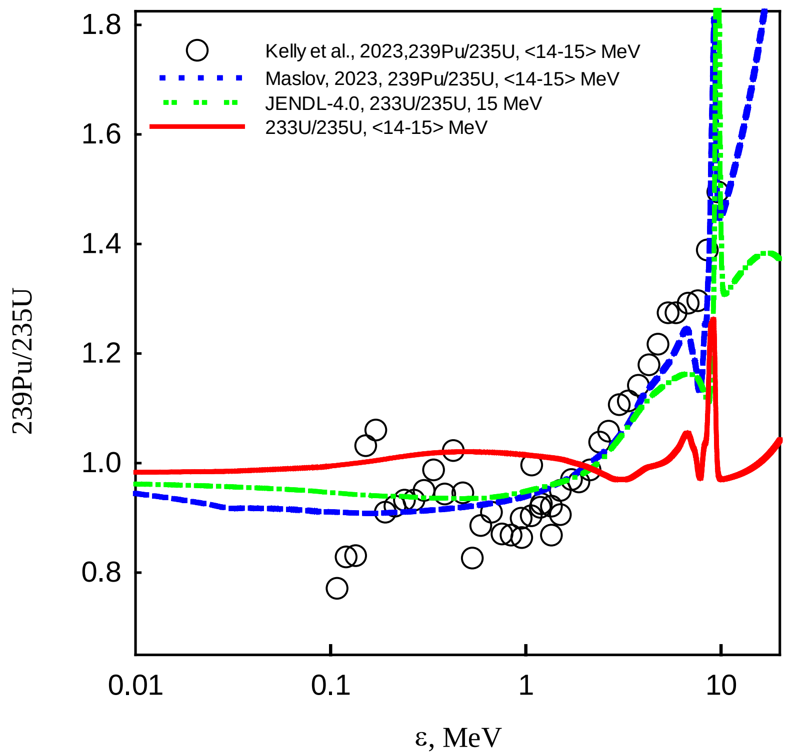

Fig.4. Ratios of PFNS ^{233}U(n,F)/^{235}U(n,F) and ^{239}Pu(n,F)/^{235}U(n,F) at E_n~14÷15 MeV.

At $E_n \gtrsim E_{n2nf}$ integral emission spectrum of $(n,nX)^1$ reaction, $d^2\sigma^1_{nnx}(\varepsilon,E_n)/d\varepsilon$, could be represented as a sum of compound and weakly dependent on neutron emission angle pre-equilibrium components, and phenomenological function, modelling energy and angle dependence of neutron spectra, relevant for the ^{233}U excitations of 1~6 MeV. Angle-averaged $\langle\omega(\theta)\rangle_\theta$ function, $\omega(\theta)$ [20], is approximated as $\langle\omega(\theta)\rangle_\theta \approx \omega(90^o)$, as described in [21]. Figure 4 compares calculated [20] and measured ratios of PFNS ^{239}Pu(n,F)/^{235}U(n,F) [20] and ^{233}U(n,F)/^{235}U(n,F) ratios at E_n~13÷14 MeV. The latter calculated present ratio is much discrepant with that of JENDL-4.0 [13], which just follows the shape of ^{239}Pu(n,F)/^{235}U(n,F) of [13].

TKE values of E_F^{pre} are superposition of TKE for $^{234-x}$U nuclides contributing to the observed fission cross section:

$$E_F^{pre}(E_n) = \sum_{x=0} E_{fx}^{pre}(E_{nx})\beta_x(E_n) \ . \tag{2}$$

The excitation energy E_{nx} of $A,\ldots\ A+1-x$ nuclides, formed after emission of $(n,xnf)^{1,..x}$ pre-fission neutrons, depends on their average energies $\langle E^k_{nxnf}\rangle$:

$$E_{nx} = E_r - E_{fx}^{pre} + E_n + B_n - \sum_{x=0,1\le k\le x}\left(\left\langle E^k_{nxnf}\right\rangle + B_{nx}\right). \tag{3}$$

Kinetic energy E_F^{post} of fission products, which emerge after emission of pre-fission neutrons, but before β⁻-decay, is defined as

$$E_F^{post} \approx E_F^{pre}\left(1-\nu_{post}/\left(A+1-\nu_{pre}\right)\right). \tag{4}$$

Weak variations of TKE values [15, 22], of both E_F^{pre} and E_F^{post}, in the vicinity of ^{233}U(n,xnf) reaction thresholds are due to the decrease of excitation energy of ($A+1-x$) fissionning nuclides after emission of x pre-fission neutron [23]. Contribution of $\sigma_{n,nf}$ to the $\sigma_{n,F}$ of ^{233}U(n,F), is larger than that of ^{235}U(n,nf) to the $\sigma_{n,F}$ of ^{235}U(n,F) [20, 24], nonetheless the local bumps in TKE around ^{233}U($n,2nf$) and ^{233}U(n,nf) reaction thresholds are weaker. That might be due to rather flat dependence on excitation energy of TKE for ^{232,233,234}U, opposite to the case of TKE for ^{235,236,237,238,239}U fissionning nuclides. To reproduce the observed dependence of E_F^{pre} on E_n in ^{233}U(n,F) reaction one may assume linear dependence of first-chance fission TKE $-E_{f0}^{pre}(E_n)$ (Fig. 5).

Average energy of prompt fission neutron spectra is its rather rough signature. Figure 6 evidence that the shapes of $\langle E\rangle(E_n)$ in cases of ^{233}U(n,F) and ^{235}U(n,F) [20] are similar. Values of $\langle E\rangle$ are presented here in the interval ε~0.01–10 MeV. Our estimate of $\langle E\rangle(E_n)$ for ^{235}U(n,F) [20] reproduces the estimate of $\langle E\rangle$ based on measured PFNS data [9, 26], especially around thresholds of ^{235}U(n,nf) and ^{235}U($n,2nf$) reactions.

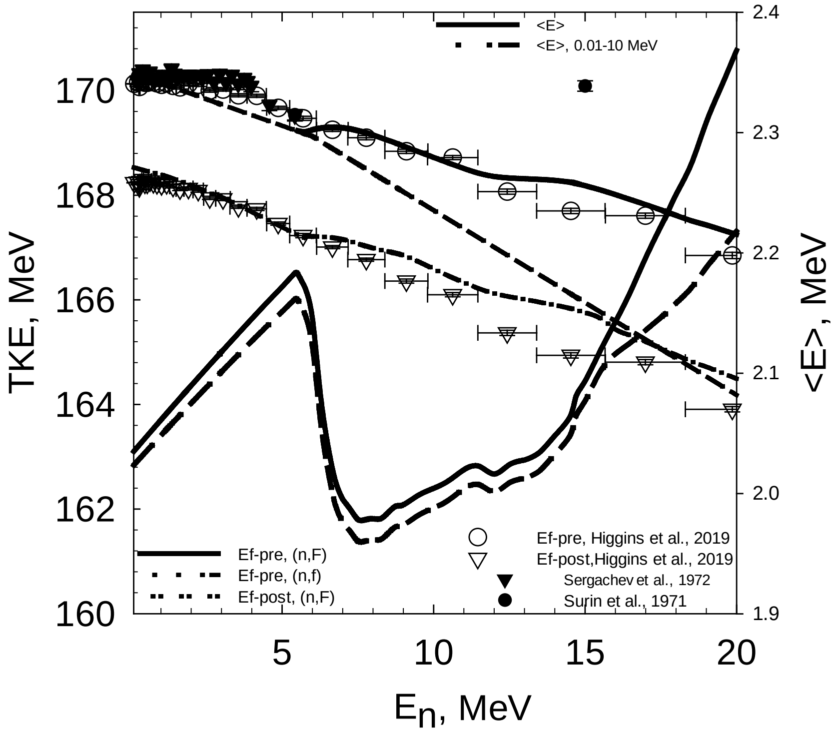

Fig. 5. Total kinetic energy TKE of ^{233}U(n,F).

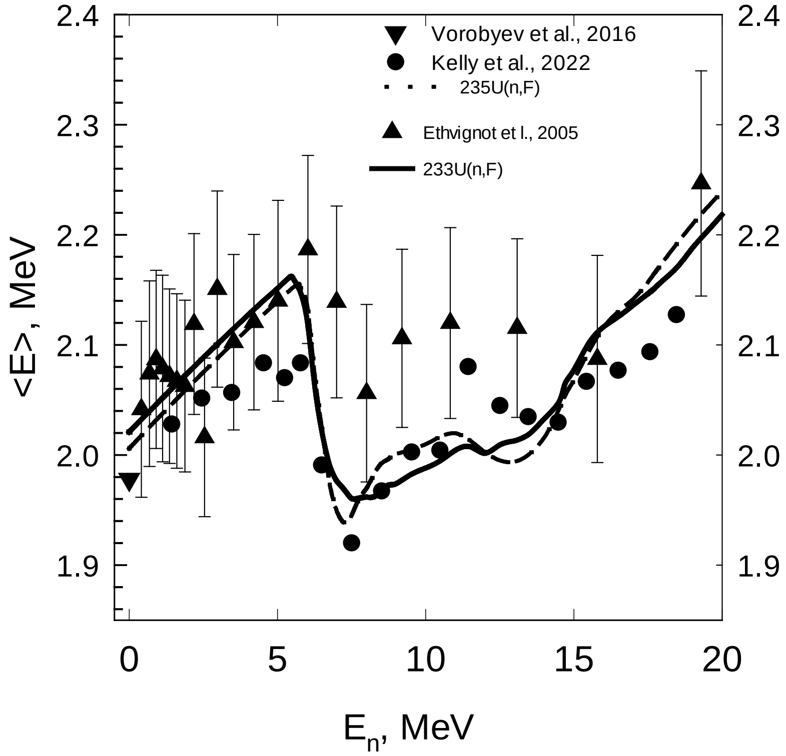

Fig.6. Average energy $\langle E \rangle$ of ^{233}U(n,F) and ^{235}U(n,F) PFNS.

CONCLUSION

A number of observed peculiarities in PFNS, TKE, $\nu_p(E_n)$ correlate with the emission of pre-fission (*n,xnf*) neutrons, as predicted for the ^{233}U(*n,F*) and ^{233}U(*n,xnf*) and earlier for ^{235}U(*n,F*) and ^{235}U(*n,xnf*) [20, 24]. Cross ratios of PFNS of ^{233}U(*n,F*), ^{235}U(*n,F*) and ^{239}Pu(*n,F*) reactions are compatible with measured data [11–13, 18, 19]. The correlation of PFNS shape and emissive ((*n,xnf*)) fission contribution to the observed fission cross section for ^{233}U(*n,F*) and ^{235}U(*n,F*) reactions is established. The net effect of these peculiarities is the occurrence of dips in $\langle E \rangle$ in the vicinity of (*n,nf*) and (*n,*2*nf*) reaction thresholds and bumps in both E_F^{pre} and E_F^{post}. Amplitude of dips in $\langle E \rangle$ of ^{233}U(*n,F*) PFNS is quite similar to that observed in PFNS of ^{235}U(*n,F*) reaction, notwithstanding the appreciable differences of ^{233}U(*n,xnf*) and ^{235}U(*n,xnf*) reaction contributions to the observed fission cross sections ^{233}U(*n,F*) and ^{235}U(*n,F*), respectively. That is explained by relatively large contributions of $\nu_{px}(E_{nx})$ as compared with $\nu_{pre}(E_n)$ for the reaction ^{233}U(*n,F*). PFNS of ^{233}U(*n,F*) are more hard than those of ^{235}U(*n,F*) PFNS, but softer than those of ^{239}Pu(*n,F*). Difference of average energies of PFNS $\langle E \rangle$ of ^{233}U(*n,F*) and ^{235}U(*n,F*) amounts to 1~3 %. At incident energies higher than (*n,2nf*) reaction threshold the observed PFNS may seem similar, though the partial contributions of ^{233}U(*n,xnf*) and ^{235}U(*n,xnf*) to the observed PFNS are quite different. It might be argued that correct estimate of the exclusive pre-fission (*n,xnf*) neutron spectra and modelling of spectra of neutrons emitted from excited fission fragments gives a robust prediction of PFNS for ^{233}U(*n,F*) for incident neutron energies E_n~ E_{th}–20 MeV with a precision and reliability comparable to those attained for ^{235}U(*n,F*) PFNS.

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