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$^{225-232}$Th symmetric/asymmetric neutron-induced fission up to 200 MeV

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Abstract. The transition from asymmetric to symmetric fission as a function of the excitation energy and the nucleon composition of fissioning Th nuclei is investigated for the ^{232}Th(n,f) reaction. Observed neutron-induced fission cross section is described in a fission/evaporation approximation. A separate relatively high outer fission barrier E_{fB}^{SL} with significant curvature was assumed for the symmetric (SL) mode, while the inner one ($E_{fA}^{SL(AS)}$) was assumed to be the same for symmetric and asymmetric (AS) modes. Axial asymmetry and mass symmetry is assumed for the outer saddle of the SL-mode, as distinct from the AS-mode. A drop of ($E_{fA}^{SL} - E_{fA}^{AS}$) = 3.5 MeV to 1–1.5 MeV for Th nuclei with A ≤ 226 is responsible for the increase of the symmetric fission contribution to ^{225,229,230,231,232}Th(n,f) fission cross sections with neutron energy up to E_n = 200 MeV. It is shown above 80 MeV the ^{229,230,231,232}Th(n,f) fission cross sections are no longer dependent on the Th target nuclide fissility.

1 Introduction

The transition from asymmetric to symmetric fission as a function of the excitation energy and the nucleon composition of fissioning nucleus is a long-standing problem of nuclear fission [1]. The U and Np nuclei near the beta-stability line demonstrate mostly asymmetric first-chance fission [2]. The contribution of the symmetric scission in that excitation energy range is never more than a few percents [3,4]. In the case of ^{238}U(n,f) reaction the competition of symmetric/asymmetric splits was investigated by Zoller et al. [5] up to E_n = 500 MeV. The analysis of the TKE-A distributions revealed rather fast increase of the symmetric contribution $r^{SL} = \sigma_{nF}^{SL}/(\sigma_{nF}^{SL}+\sigma_{nF}^{AS})$, up to ~0.5 at E_n = 200 MeV. In the emissive fission domain fission observables are composed of partial contributions of the ensemble of uranium nuclei, which emerge after emission of x pre-fission neutrons (up to $x \sim 20$ at E_n = 200 MeV) [6,7]. Statistical-model calculations consistently reproduce up to E_n = 200 MeV the observed fission cross sections of ^{233}U(n,f), ^{235}U(n,f), ^{238}U(n,f) and ^{237}Np(n,f) alongside with the symmetric-to-observed fission ratio $r^{SL}(E_n)$ in ^{238}U(n,f) reaction [6,7]. It was possible only with the assumption that the most contribution to the observed fission cross section at high incident neutron energies is coming from the neutron-deficient U or Np nuclides, which emerge after pre-fission (pre-saddle) neutron emission. The small differences of $r^{SL}(E_n)$ for the ^{233,235,238}U and ^{237}Np target nuclides might be explained by different (n,$x$nf) emissive fission chances contributions to the observed fission cross sections for targets with different fissilities.

In case of ^{232}Th(n,f) reaction, since Th nuclei fission probabilities are much lower than those of U or Np nuclei, the influence of preferential (n,$x$nf) emissive fission contributions to the σ_{nF} is much more pronounced than in case of U or Np targets [8,9]. The properties of the potential energy surface of the ensemble of neutron-deficient fissioning nuclei, which contribute to the fission observables in a multiple-chance fission, may strongly depend on the (Z, N)-composition of the fissioning nucleus. Henceforth, we will not make a distinction between Standard 1 ($S1$) and Standard 2 ($S2$) modes [2], denoting a lumped asymmetric mode as AS. The relative heights of symmetric (denoted as SL(superlong) [2]) and asymmetric saddles ($E_{fB}^{SL} - E_{fB}^{AS}$) for the neutron-deficient (A ≤ 226) Th nuclides may vary in compliance with the observed distinct increase of the symmetric fission yield [10–12]. However, the experimental estimates [10–12] of the symmetric-to-observed fission yields for the low excited ^{226}Th nuclide (U ≲ 26 MeV) are rather controversial. That might be explained by the differences of the excitation energies of an ensemble of fissioning Th nuclei, which contribute to the observed yield. This may lead to different contributions of (n,$x$nf) reactions to the observed fission fragment yields in the ^{208}Pb(^{18}O,f) [11, 12] reaction and in fission of relativistic heavy-ions after electromagnetic interaction with lead nuclei [10]. Nonetheless, it was concluded in [10], that the contribution of the symmetric fission to the observed neutron-induced fission yield (in the first-chance fission domain) increases with the decrease of the neutron number N for Th fissioning nuclides with A ≤ 226. For the ^{232}Th(n,f) reaction that peculiarity may lead to the appreciable increase of the symmetric fission yields due to ^{232}Th(n,$x$nf) reactions at E_n = 50–200 MeV, as compared to observed symmetric fission yield in ^{238}U(n,f) reaction. The pronounced increase of the symmetric fission contribution to the observed fission cross section of ^{229,230,231}Th(n,f) reactions might happen for still lower E_n. That peculiarity could be attributed to the lower contribution of the highly excited neutron-rich fissioning Th nuclides to the fission observables in the emissive fission domain. Previously, for the ^{232}Th(n,f) reaction, the ratio of symmetric-to-observed fission in [9], obtained based on systematics of the fission saddle heights for symmetric and asymmetric splits of U nuclides [8], was predicted to be lower than for the ^{238}U(n,f).

2 Fission cross section of ^{232}Th

The detailed description of the statistical model calculations of the symmetric/asymmetric fission competition in the emissive fission domain is given elsewhere [4,8,9]. The energy dependence of symmetric fission cross section turned out to be quite sensitive to the symmetry of the outer saddle of a double humped fission barrier. A separate rather high outer fission barrier with significant transparency was assumed for

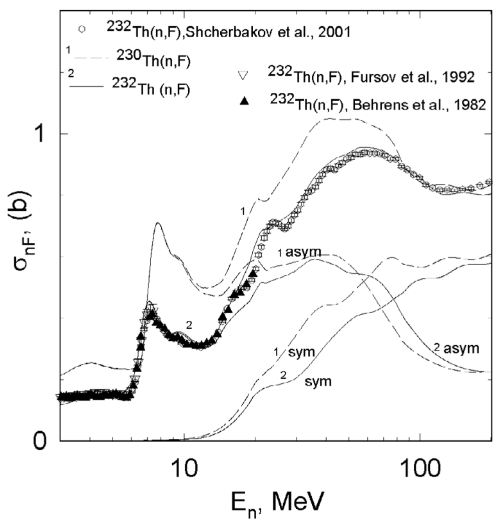

Fig. 1. Fission cross sections of ^{232}Th(n,f) and ^{230}Th(n,f). Measured data points for ^{232}Th(n,f) only.

the SL-mode, while the inner one was the same for the symmetric and the asymmetric modes. Axial asymmetry and mass-symmetry are assumed for the outer saddle of the SL-mode, as distinct from the asymmetric AS-mode, for which outer saddle is axially symmetric and mass-asymmetric. In summary, it was found that to the mass symmetric fission corresponds rather thin but high outer barrier E^{SL}_{fB}, as compared to the mass-asymmetric outer fission barrier E^{AS}_{fB}, actually $(E^{SL}_{fB} - E^{AS}_{fB}) =$ 3.5 MeV was obtained. In the emissive fission domain the observed fission cross section of ^{238}U(n,f) and the ratio of symmetric-to-observed fission yields $r^{SL}(E_n)$ are described under assumption that more fissions come from the neutron-deficient U nuclei via (n,$x$nf) fission chances with high number x of pre-fission neutrons [8,9].

After the pre-saddle neutron emission the $^{233-x}$Th ($x \sim 1 \div 20$) nuclides contribute to the observed fission cross section of ^{232}Th(n,f) reaction. Light charged particle (LCP) emission is assumed to be negligible. For incident neutron energies $E_n \leq 20$ MeV the ^{232}Th(n,f) fission chances partitioning could be defined quite unambiguously. Statistical-model calculations [13, 14] consistently reproduce the ^{232}Th(n,f), ^{231}Th(n,f), ^{230}Th(n,f) and ^{232}Th(n,2n) cross section data and prompt fission neutron spectra of ^{232}Th(n,f) reaction. The deformed optical potential describes the n+^{232}Th total cross section data up to $E_n = 200$ MeV [9]. The calculated total cross section $\sigma_T = \sigma_R + \sigma_{SE}$ is the sum of the reaction cross section σ_R and the scattering cross section σ_{SE}. The preequilibrium neutron spectrum is fixed by simultaneous description of the ^{232}Th(n,f) and ^{232}Th(n,2n) cross sections [9] and prompt fission neutron spectra [14]. Finally, the value of σ_R can not be lower than the fission cross section of the higher fissility targets, like ^{233}U.

Calculated (n,$x$nf) contributions to the observed fission cross sections are largely defined by the level density parameters a_f and a_n for fissioning (f) and residual (n) nuclides [8], as well as the damping of the rotational modes contributions to the level densities

$$\rho(U, J, \pi) = K_{rot}(U)\, K_{vib}(U)\, \rho_{qp}(U, J, \pi). \qquad (1)$$

Nuclear level density $\rho(U, J, \pi)$ is represented as the factorized contribution of the quasiparticle and collective states [15]. Quasiparticle level densities $\rho_{qp}(U, J, \pi)$ were calculated with a phenomenological model by Ignatyuk et al. [16], $K_{rot}(U, J)$ and $K_{vib}(U)$ are factors of the rotational and vibrational enhancement. At saddle and ground state deformations factor $K_{rot}(U)$ is defined by the deformation order of symmetry, adopted from the shell correction model calculations [17], except outer saddles for the symmetric scission. For calculation of the fission probabilities of $^{233-x}$Th nuclides we use double-humped fission barrier model, since possible splitting of the outer fission barrier hump is not important in the present context. Damping of the rotational mode contributions to the $\rho(U, J, \pi)$ was assumed, as anticipated by Hansen and Jensen [18], but at much lower excitations. The damping might be different for the axially symmetric and triaxial shapes [19].

The ^{232}Th target nuclide exhibits the lowest fissility among the actinide nuclides investigated with the neutron-induced fission reactions up to $E_n = 200$ MeV [20]. The observed fission cross section of ^{232}Th(n,f) reaction [20–22] could be reproduced up to $E_n = 200$ MeV only under assumption that more fissions come from the neutron-deficient Th nuclei [8,9]. The ratio of symmetric-to-observed fission yields, for ^{232}Th(n,f) cross sections in [9] was obtained based on the assumption that the difference of heights of symmetric and asymmetric saddle points $(E^{SL}_{fB} - E^{AS}_{fB}) = 3.5$ MeV [8,9] is independent on the neutron number of fissioning nucleus. That estimate is lower, than $(E^{SL}_{fB} - E^{AS}_{fB}) \sim 5$ MeV lowering of the reflection-asymmetric outer saddle for ^{232}Th and ^{234}Th, obtained within a Hartree-Fock and BCS pairing approach by Bonneau et al. [23]. However, the pronounced isotopic dependence of E^{SL}_{fB} and E^{AS}_{fB} may be the case for Th nuclides with $A \leq 226$, i.e., the symmetric fission yield may have a tendency to increase for neutron-deficient nuclides [10–12]. That may lead to the increase of the symmetric fission yields in ^{232}Th(n,f) reaction due to ^{232}Th(n,$x$nf) fission reactions at $E_{n(p)} = 50$–200 MeV. Figure 2 shows the sharing of the ^{232}Th(n,f) observed fission cross section [20–22] to SL- and AS-mode contributions. For that $(E^{SL}_{fB} - E^{AS}_{fB}) = 1.5$ MeV for Th nuclides with $A \leq 226$, which is achieved by ~1 MeV decrease of E^{SL}_{fB} and ~1 MeV increase of E^{AS}_{fB} of outer fission barriers, as compared to the barrier values, used in [9]. This is generally consistent with the outer fission barrier estimates by Ohtsuki et al. [24], based on fission yield analysis for proton-induced fission of Th, U, Np, Pu and Am nuclei at E_p = 8-16 MeV. The sharp increase of the ^{232}Th(n,f) lumped symmetric fission yield at $E_n \geq 80$ MeV (see fig. 2) is due to appreciable increase of the ^{232}Th(n,$x$nf) contributions of relatively low excited neutron-deficient Th nuclides. The predicted sharing into σ^{SL}_{nF} and σ^{AS}_{nF} is shown on figure 1 for the ^{230}Th(n,f) reaction as well.

3 Branching ratio of symmetric-to-observed fission events

The branching ratio of the symmetric to observed fission events $r^{SL}(E_n)$ for ^{232}Th(n,f), shown on figure 2, is higher than that observed for the ^{238}U(n,f) reaction [5,8]. The calculated ratio r^{SL} is much dependent on the (n,$x$nf) fission chances distribution. Because the fissilities of Th nuclides with

A ≤ 233 are lower, than those of U nuclei with A ≤ 239, the contribution of the first few fission chances is lower in case of ^{232}Th(n,f) reaction. That means the contribution of the fission reactions of neutron-deficient $^{233-x}$Th nuclides with low intrinsic excitation energies would be relatively high. That might lead to the lowering of r^{SL} for the ^{232}Th(n,f) reaction, but this effect could be more than compensated, since for Th nuclei the relative heights of the symmetric and asymmetric outer fission barriers ($E_{fB}^{SL} - E_{fB}^{AS}$), as was already mentioned, might change in favor of symmetric fission contribution [10–12,25–27]. The experimental estimates of the branching ratios r^{SL} for Th nuclides with A ≤ 226 by Itkis et al. [11], Pokrovsky et al. [12] and Schmidt et al. [10] correspond to different excitation energies of composite nuclei. Different contributions of emissive fission reactions to the observed fission fragment yields in ^{208}Pb(^{18}O,f) reaction [11] and in peripheral relativistic heavy-ion reaction [10] would follow, since the intrinsic excitations of the ensemble of fissioning nuclides also differ. The excitation energies of the fissioning nuclides ^{229}Th and ^{226}Th [10], shown on figure 2 (∼11 MeV), correspond to two-phonon excitation of GDR. The experimental estimate of the symmetric fission contribution to the observed fission fragment yield in ^{208}Pb(^{18}O,f) reaction [11] is shown for the equivalent incident neutron energy E_n ∼ 20 MeV. Obviously, the contribution of symmetric fission of Th nuclides with A ≤ 226 is rather sensitive to the value of fission barrier splitting ($E_{fB}^{SL} - E_{fB}^{AS}$). The estimate of the relative symmetric fission yield [29] for the ^{232}Th(p,f) reaction amounts to ∼0.715, which is quite compatible with the present estimate for the ^{232}Th(n,f) reaction. Figure 2 shows also the calculated contribution of symmetric fission events to the observed fission yield for the ^{225}Th(n,f) reaction up to E_n ∼200 MeV. In case of ^{225}Th(n,f) the contribution of the neutron-deficient Th nuclei to the fission observables is much higher, it seems to be compatible with the experimental estimates by Pokrovsky et al. [12] and Schmidt et al. [10].

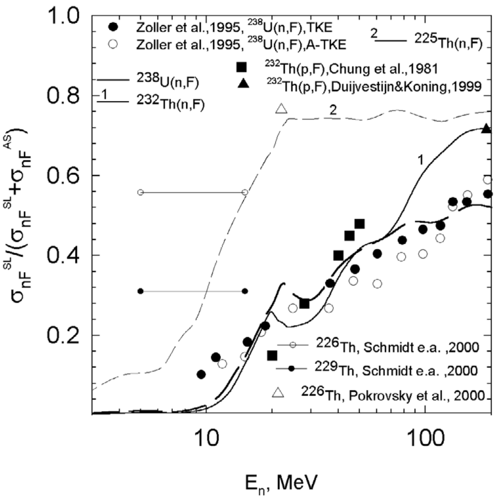

Fig. 2. Branching ratio r^{sym} for ^{238}U(n,f), ^{232}Th(n,f), ^{232}Th(p,f) and ^{225}Th(n,f).

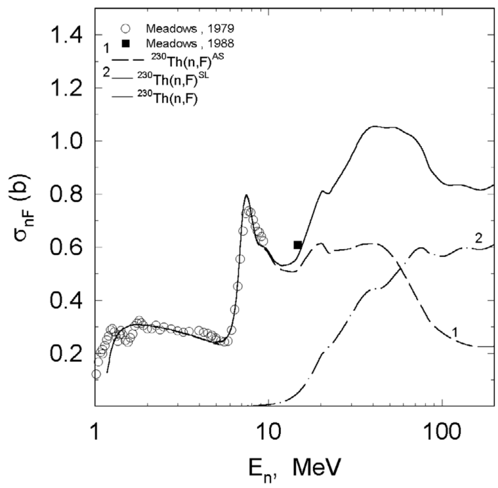

Fig. 3. Fission cross section of ^{230}Th(n,f).

4 Fission cross section of Th nuclei

The description of the ^{232}Th(n,f) fission cross section and $r^{SL}(E_p)$ [28,29] for ^{232}Th(p,f) [30] fixes virtually all the parameters to predict the neutron-induced fission cross sections of Th nuclides with $A < 232$. The observed fission cross sections of ^{230}Th(n,f) [31,32], ^{231}Th(n,f) [33,34] and ^{229}Th(n,f) [35,36] reactions are predicted at 1–200 MeV (figs. 1, 3), 0.001–200 MeV (fig. 4) and 0.001–200 MeV (fig. 5) energy ranges, respectively. The predicted cross section shape of ^{229}Th(n,f) reaction is quite consistent with the shape of the data [36] and data [35] below E_n ∼ 10 keV. For ^{231}Th(n,f) [13] and ^{229}Th(n,f) reactions the theoretical estimate of the fission cross section below E_n ∼ 1 MeV is defined by the lowering of the $K^{\pi} = 0^-$ octupole band for the outer fission barrier quadrupole deformations of even-even fissioning nuclides ^{232}Th and ^{230}Th, respectively. That is consistent with the fission probability estimates of ^{232}Th and ^{230}Th nuclides, extracted by the ^{232}Th(n,f) and ^{230}Th(n,f) cross section description above the ^{232}Th(n,nf) and ^{230}Th(n,nf) reaction threshold, respectively (see figs. 1 and 3). A similar approach was followed in case of the ^{237}U(n,f) reaction [37]. Recently, a theoretical estimate of ^{237}U(n,f) cross section, based on ^{238}U(n,f) cross section description above the ^{238}U(n,nf) reaction threshold [14], was assured by the surrogate techniques measurements by Burke et al. [38]. The first chance ^{238}U(n,f) fission cross section of ^{238}U(n,f) reaction in no case could be a steeply decreasing function of energy, as is still frequently assumed. The same applies in case of ^{232}Th(n,f) reaction. It seems fission data measured with surrogate techniques would appear soon on ^{229}Th(n,f) and ^{231}Th(n,f) for E_n up to ∼20 MeV.

Predicted increase of the symmetric fission yield, which becomes higher than that of the asymmetric fission yield at E_n ≥ 80 MeV in ^{232}Th(n,f) reaction, is due to the increased symmetric fission of neutron-deficient Th nuclei. Figures 1, 3, 4 and 5 demonstrate also that with increase of the fissility of the ^{229,230,231,232}Th target nuclides, the symmetric fission yield tends to be higher than that of the asymmetric yield at lower E_n, at E_n ≥ 60 MeV in case of ^{230}Th(n,f) reaction, for instance. Figures 1, 3, 4 and 5 show also that at

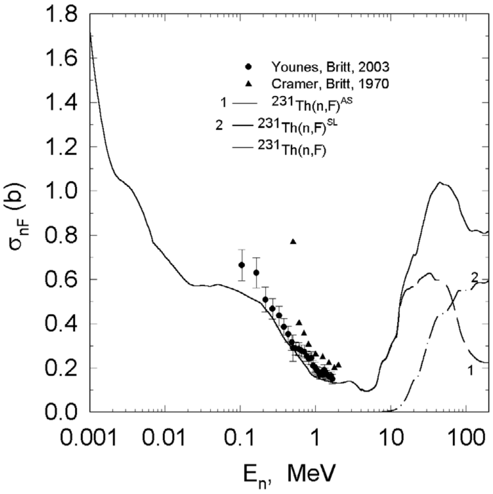

Fig. 4. Fission cross section of ^{231}Th(n,f).

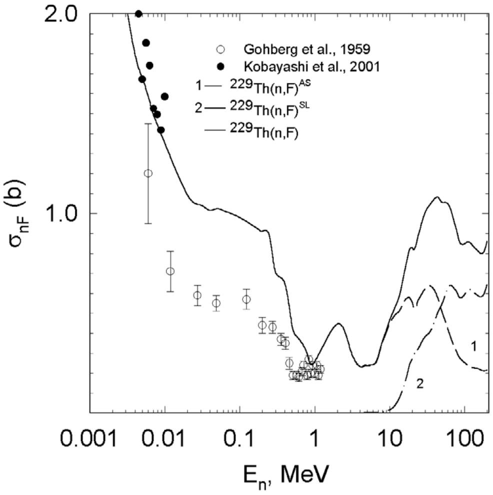

Fig. 5. Fission cross section of ^{229}Th(n,f).

$E_n \gtrsim 80$ MeV the ^{229,230,231,232}Th fission cross sections are no longer dependent on the target nuclide fissility.

5 Conclusion

The description of the observed fission cross sections of ^{232}Th(n,f) reaction up to $E_n \sim 200$ MeV was achieved under assumption of preferential contribution of fission of neutron-deficient Th nuclides. The fission chances distribution was obtained by the consistent description of the observed fission cross section and symmetric fission branching ratio for the ^{238}U(n,f) reaction. Sharp increase of the symmetric fission yield for the ^{232}Th(n,f) reaction above $E_n \gtrsim 50$ MeV is predicted due to the similar behavior of Th neutron-deficient nuclides. For fission reactions of ^{226}Th and ^{228}Th nuclides enhanced symmetric fission yields were observed previously [10–12] at excitation energies of $U \sim 10$–26 MeV. The calculated neutron-induced fission cross sections of ^{229,230,231,232}Th target nuclides remain much lower than the neutron absorption cross section and at $E_n \geq 80$ MeV, they are virtually independent upon the target nuclide fissility. Calculated cross sections of ^{229,230,231}Th(n,f) reactions are much different from those currently adopted for major data libraries, the transparent physics of fission reasoning is provided.

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