

Protactinium neutron-induced fission up to 200 MeV

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Abstract. The theoretical evaluation of $^{230-233}\text{Pa}(n,F)$ cross sections is based on direct data, $^{230-234}\text{Pa}$ fission probabilities and ratios of fission probabilities in first-chance and emissive fission domains, surrogate for neutron-induced fission. First chance fission cross sections trends of Pa are based on consistent description of $^{232}\text{Th}(n,F)$, $^{232}\text{Th}(n,2n)$ and $^{238}\text{U}(n,F)$, $^{238}\text{U}(n,xn)$ data, supported by the ratio surrogate data by Burke et al., 2006, for the $^{237}\text{U}(n,F)$ reaction. Ratio surrogate data on fission probabilities of $^{232}\text{Th}(^6\text{Li},^4\text{He})^{234}\text{Pa}$ and $^{232}\text{Th}(^6\text{Li},d)^{236}\text{U}$ by Nayak et al., 2008, support the predicted $^{233}\text{Pa}(n, F)$ cross section at $E_n=11.5-16.5$ MeV. The predicted trends of $^{230-232}\text{Pa}(n, F)$ cross section up to $E_n=20$ MeV, are consistent with fissilities of Pa nuclides, extracted by $^{232}\text{Th}(p,F)$ (Isaev et al., 2008) and $^{232}\text{Th}(p,3n)$ (Morgenstern et al., 2008) data analysis. The excitation energy and nucleon composition dependence of the transition from asymmetric to symmetric scission for fission observables of Pa nuclei is defined by analysis of p-induced fission of ^{232}Th at $E_p=1-200$ MeV. Predominantly symmetric fission in $^{232}\text{Th}(p,F)$ at $E_{n(p)}=200$ MeV as revealed by experimental branching ratios (Dujvestijn et al., 1999) is reproduced. Steep transition from asymmetric to symmetric fission with increase of nucleon incident energy is due to fission of neutron-deficient Pa ($A\leq 229$) nuclei. A structure of the potential energy surface (a drop of symmetric and asymmetric fission barriers difference ($E_{SYM}^f - E_{ASYM}^f$) from ~ 3.5 MeV to ~ 1 MeV) of N-deficient Pa nuclides ($A\leq 226$) and available phase space at outer fission saddles, are shown to be responsible for the sharp increase with $E_{n(p)}$ of the symmetric fission component contribution for $^{232}\text{Th}(p,F)$ and $^{230-233}\text{Pa}(n, F)$ reactions. That is a strong evidence of emissive fission nature of moderately excited Pa nuclides, reliably quantified only up to $E_{n(p)}\sim 20(30)$ MeV. Predicted fission cross section of $^{232}\text{Pa}(n,F)$ coincides with that of $^{232}\text{Th}(p,F)$ at $E_{n(p)}\geq 80$ MeV, that means that entrance channel dependence of fission cross section with increase of nucleon incident energy diminishes.

1 Introduction

Neutron-induced cross sections of $^{231}\text{Pa}(n,F)$ and $^{233}\text{Pa}(n,F)$ data [1–3] when complemented with surrogate fission data, measured in reactions $^{232}\text{Th}(^3\text{He},d)^{233}\text{Pa}$, $^{231}\text{Pa}(d,p)^{232}\text{Pa}$, $^{230}\text{Th}(^3\text{He},d)^{231}\text{Pa}$ and $^{230}\text{Th}(^3\text{He},t)^{230}\text{Pa}$ at excitation energies 6–11.5 MeV [4] and fission probabilities measured in [5] via reactions $^{232}\text{Th}(^3\text{He},p)^{234}\text{Pa}$, $^{232}\text{Th}(^3\text{He},d)^{233}\text{Pa}$ and $^{232}\text{Th}(^3\text{He},t)^{232}\text{Pa}$ at excitation energies 6–15 MeV pose a number of severe problems for consistent theoretical description. In an emissive fission domain data by Petit et al. [5], as well as older indirect data by Birgul et al. [6] provoke assumption of steep decrease of the first-chance fission cross sections of $^{233}\text{Pa}(n,F)$ and $^{231}\text{Pa}(n,F)$ and systematically lowered fission probabilities of relevant Pa nuclides [7]. At excitations near fission threshold surrogate data [4, 5] are model-dependent [8, 9]. Above emissive fission threshold the sensitivity to the angular momentum may again increase. Recently developed surrogate ratio method [10] largely removes the uncertainty, imposed by pre-equilibrium effects and different angular momentum spectra of excited and fissioning states in (n,F) and transfer reactions. The theoretical approach, which would be employed for the $^{229-233}\text{Pa}(n,F)$ cross section predictions, was independently supported by the ratio surrogate data [10]

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for the $^{237}\text{U}(n,F)$ reaction [11, 12]. Recent ratio surrogate data on fission probabilities of $^{232}\text{Th}(^6\text{Li}, ^4\text{He})^{234}\text{Pa}$ and $^{232}\text{Th}(^6\text{Li}, d)^{236}\text{U}$ by Nayak et al. [13], relevant for the $E_n = 11.5-16.5$ MeV, support our prediction [14]. The shell effects either in fission fragments or saddle configurations define the fission observables at relatively low excitation energies U . It is generally believed that with increase of U the shell effects diminish and fission observables are dominated by the macroscopic nuclear properties. Pre-fission neutron emission decreases the excitation energy of the fissioning nuclei, which may influence the competition of the symmetric and asymmetric fission modes [15, 16], decreasing the contribution of the symmetric one. At the other hand, the neutron-deficient Pa nuclides, in $^{232}\text{Th}(p,xf)$ or $^{232}\text{Pa}(n,xf)$ reactions, might be more susceptible to symmetric fission even at low excitations [17, 18]. Interplay of these two trends would define the fission observables at higher excitations.

2 Direct and surrogate data for $^{230-233}\text{Pa}(n,F)$

The surrogate data [5] are appreciably higher than direct $^{233}\text{Pa}(n,F)$ data [2, 3] both around (n,f) and second-chance (n,nf) fission thresholds (see Fig.1). Discrepancies of di-

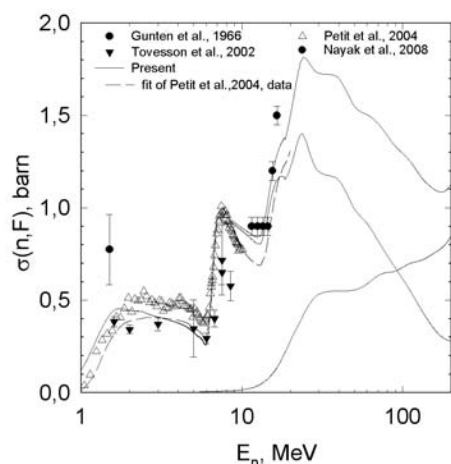


Fig. 1. $^{233}\text{Pa}(n,F)$ cross section

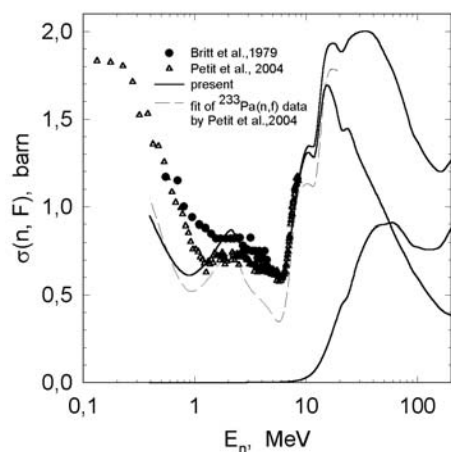


Fig. 2. $^{232}\text{Pa}(n,F)$ cross section

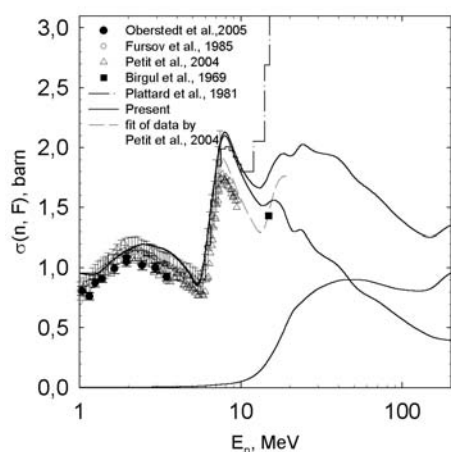
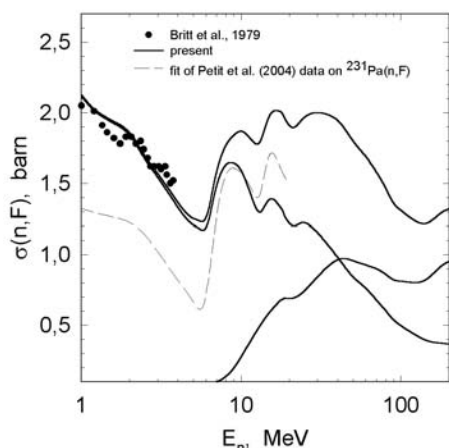
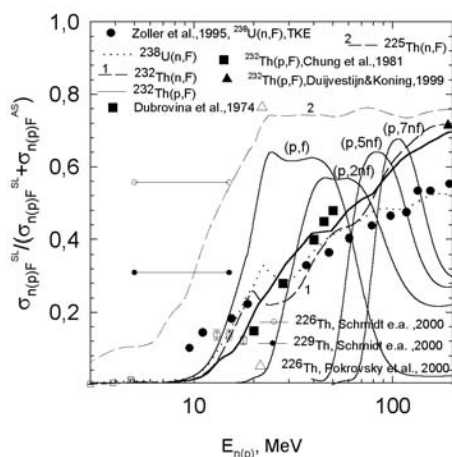


Fig. 3. $^{231}\text{Pa}(n,F)$ cross section.

rect and surrogate fission data were addressed by Arthur [19], it was proved to be a consequence of the spin population differences in transfer and (n,f) reactions [19,20]. Much larger discrepancy is observed at the onset of the $^{233}\text{Pa}(n,nf)$ reaction, the σ_{nf} data being much lower (see Fig.1). It could be traced back to oversimplified procedure of obtaining surrogate data as $\sigma_{nf} = \sigma_{CN} \times P_f^{exp}$ in the emissive fission domain, pre-equilibrium emission sensitivity and influence of spectroscopic properties of transition states of ^{233}Pa . The latter effect for o-e nuclide ^{233}Pa should be excluded, unlike the observed discrepancy of direct and surrogate $^{230}\text{Th}(^4\text{He}, ^3\text{He})^{231}\text{Th}$ data for $^{230}\text{Th}(n,F)$, which might be traced back to properties of e-e nuclide ^{230}Th , fissioning in $^{230}\text{Th}(n,nf)$ reaction. Wide peak around $E_n \sim 8$ MeV, observed in [21], is described by lowering the negative parity octupole band of ^{230}Th due to the mass-asymmetry of outer saddle deformations [22,23]. It might be concluded that the discrepancy of surrogate [5] and direct fission data for ^{230}Th target nuclide above (n,nf) fission threshold is of systematic character and might be pronounced in case of Pa nuclides. Above emissive fission threshold contributions to the observed fission cross section coming from (n,xnf), $x = 1, 2, 3 \dots X$, fission of equilibrated Pa nuclei, are calculated as

$$\sigma_{nF}(E_n) = \sigma_{nf}(E_n) + \sum_{x=1}^X \sigma_{n,xnf}(E_n). \quad (1)$$

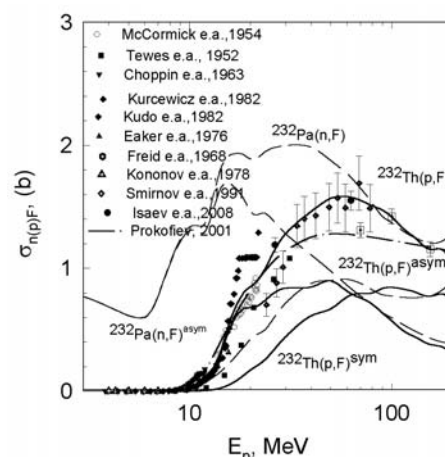
For $^{233}\text{Pa}(n,F)$ emissive fission contributions $\sigma_{n,xnf}(E_n)$ are calculated using fission probability estimates of $^{234-x}\text{Pa}$ nuclides from $^{232}\text{Th}(^3\text{He},d)^{233}\text{Pa}$ [4,5], $^{231}\text{Pa}(d,p)^{232}\text{Pa}$ [5] reactions. Overall consistency of $^{232}\text{Th}(^3\text{He},d)^{233}\text{Pa}$ fission data measured in [4,5] is demonstrated on Fig. 2. The first-chance fission probability of the $^{232,233,234}\text{Pa}$ nuclides depends mostly on the level density of fissioning and residual nuclei (see for details [24,25]). The $^{233}\text{Pa}(n,f)$ fission cross section shape of [2,3] needs extremely low contribution of the second chance fission reaction $^{233}\text{Pa}(n,nf)$ to the $^{233}\text{Pa}(n,F)$. Consequently, calculated $^{232}\text{Pa}(n,f)$ cross section would be drastically discrepant with the indirect data [4] on $^{232}\text{Pa}(n,f)$ at $E_n \sim 0.5 - 5$ MeV (see Fig.2). Shape of the $^{233}\text{Pa}(n,F)$ calculated cross section, obtained by fitting $^{232}\text{Pa}(n,f)$ data by Britt and Wilhelmy [4] in $E_n \sim 2-5$ MeV energy range in [14] is supported by ratio surrogate data of $^{232}\text{Th}(^6\text{Li},^4\text{He})^{234}\text{Pa}$ and $^{232}\text{Th}(^6\text{Li},d)^{236}\text{U}$ by Nayak et al. [13], relevant for the $E_n \sim 11.5-16.5$ MeV. In case of $^{231}\text{Pa}(n,f)$ reaction (see Fig.3) discrepancy of calculated cross section, based on direct data [26,27], and surrogate data is quite similar to that observed in case of $^{233}\text{Pa}(n,f)$. Data point by Birgul et al. [6] at $E_n \sim 14$ MeV is compatible with the surrogate data trend. That trend and data by Birgul et al.[6] could be reproduced if the contribution of the $^{231}\text{Pa}(n,nf)$ is much lower, than predicts fission probability from $^{230}\text{Th}(^3\text{He},d)^{231}\text{Pa}$ reaction [4]. Fig. 4 compares $\sigma_{n,F}(E_n)$ of $^{230}\text{Pa}(n,F)$ with surrogate data [4]. The predicted trend of $^{231}\text{Pa}(n,F)$ cross section up to $E_n \sim 20$ MeV, which is similar to that of $^{233}\text{Pa}(n,F)$, is consistent with fissilities of Pa nuclides, stemming from consistent analysis of $^{232}\text{Th}(p,F)$ and $^{232}\text{Th}(p,3n)$ database, enlarged by new data on $^{232}\text{Th}(p,F)$ [28] and $^{232}\text{Th}(p,3n)$ [29], sensitive to

Fig. 4. $^{230}\text{Pa}(n,F)$ cross sectionFig. 5. Symmetric yield r^{SL}

the fission probabilities of ^{231}Pa and ^{230}Pa (see [25]). Fit of $^{232}\text{Th}(p,F)$ and $^{232}\text{Th}(p,3n)$ cross sections data corresponds to present description of $^{230-233}\text{Pa}(n,F)$.

3 p+ ^{232}Th and n+ $^{231-233}\text{Pa}$ reactions

The excitation energy and (Z,N)-dependence of the transition from asymmetric to symmetric scission of fissioning Pa nuclei are probed in $^{232}\text{Th}(p,F)$ reaction at $E_{n(p)}$ up to 200 MeV. That is the mass and excitation energy range where the transition to predominantly symmetric fission was observed in $^{232}\text{Th}(p,F)$ reaction [30,31]. The data on the symmetric yield $r^{SL} = \sigma_{pF}^{SL} / (\sigma_{pF}^{SL} + \sigma_{pF}^{AS})$ for $^{232}\text{Th}(p,F)$ [30–32] are interpreted to be due to steep transition from asymmetric to symmetric fission for neutron-deficient Pa ($A \leq 229$) nuclei, predicted in [17] (see Fig.5). At $E_{n(p)} \sim 200$ MeV about 20 neutron-deficient nuclides might contribute to the fission observables [23], while in [17] $\nu_{pre} = 1-2$. Symmetric/asymmetric (p(n),xnf) contributions to observed fission cross sections are largely defined by the level density parameters a_f and a_n for fissioning and residual nuclides and damping of the rotational modes contri-

Fig. 6. $^{232}\text{Th}(p,F)$ cross section

butions to the level densities and saddle asymmetries [14, 23, 33]. Contribution of the higher emissive fission chances $^{232}\text{Th}(p,xfn) / ^{232}\text{Th}(p,F)$ becomes overwhelming with increase of E_p . The energy dependence of σ_{pF}^{SL} is defined by the symmetry of the outer saddle of a double humped fission barrier. For $^{232}\text{Th}(n,F)$ it was found that rather thin but high axially asymmetric outer barrier (E_{fB}^{SL}) corresponds to the mass symmetric fission, in contrary to the lower mass-asymmetric outer fission barrier (E_{fB}^{AS}). The $^{232}\text{Th}(n,F)$ measured fission cross section data also could be reproduced only for the fission chances distribution, corresponding to the preferential contribution of fission of neutron deficient Th nuclides [23] (see Fig. 6). The sharing of the $^{232}\text{Th}(p,F)$ fission cross section to SL- and AS-modes is obtained for $(E_{fB}^{SL} - E_{fB}^{AS}) = 1$ MeV for Pa nuclides with $A \leq 226$. That leads to the increase of symmetric fission yields in $^{232}\text{Th}(p,F)$ reaction due to $^{232}\text{Th}(p,xfn)$ at $E_{n(p)} \sim 50-200$ MeV. The measured symmetric fission yields for $E_p = 20-50$ MeV [32] and $E_p = 190$ MeV [30,31] provide an unambiguous evidence for the sharp increase of r^{SL} at $E_p \geq 30$ MeV. There is a strong evidence [17], that fission of $^{233-x}\text{Pa}$ nuclei ($x = 1-20$) in case of $^{232}\text{Th}(p,F)$ reaction would define competition of symmetric and asymmetric fission events at E_p up to 200 MeV. The estimate $(E_{fB}^{SL} - E_{fB}^{AS}) \sim 1.0$ MeV is used for Pa nuclei with $N \leq 135$. Cross section of $^{232}\text{Th}(p,F)$ is higher than that of $^{232}\text{Th}(n,F)$ at $E_{n(p)} \geq 18$ MeV [23]. That means in case of p+ ^{232}Th interaction the fissilities of Pa nuclei are relatively higher than those of respective Th nuclei for the n+ ^{232}Th interaction, which influences the observed fission cross section at $E_{n(p)} < 100$ MeV. In case of $^{232}\text{Th}(p,F)$ entrance channel plays a decisive role at $E_{n(p)} < 100$ MeV. Present estimate of $^{232}\text{Th}(p,F)$ cross section differs essentially from the phenomenological estimate [34].

Prediction of the optical potential for the incident protons was based on the optical potential for incident neutrons introducing into real and imaginary potential terms isoscalar and isovector components [35], which depend on the symmetry parameter $\eta = (N - Z)/A$, only in a real volume V_R^p and imaginary surface W_D^p potential terms [11, 39]. Values of V_R^p and W_D^p for incident protons are calcu-

lated as $V_R^p = V_R^n + 2\alpha\gamma$ and $W_D^p = W_D^n + 2\beta\gamma$, $\alpha = 16$ and $\beta = 8$ values, obtained by the description of the proton and neutron scattering data for a number of medium weight nuclei [35, 36]. The predicted proton absorption cross section $\sigma_R^p > \sigma_R^n$ at $E_n \gtrsim 50$ MeV is compatible [37] with the experimental data in the same way, as it was shown for the $p+^{238}\text{U}$ interaction [38, 39]. $^{230-233}\text{Pa}(n,F)$ cross sections, shown on Figs. 1-4 were calculated with fission barrier and level density parameters, fixed by $p+^{232}\text{Th}$ interaction data analysis. It might be argued that the uncertainty of $\text{Pa}(n,F)$ fission cross sections is similar to that of $^{232}\text{Th}(p,F)$.

4 Conclusion

The improved evaluation of $^{230-233}\text{Pa}(n,F)$ evaluated data is based on consistent description of fission probability data, coming from transfer reactions and $p+^{232}\text{Th}$ interaction data base. Recent ratio surrogate data on fission probabilities of $^{232}\text{Th}(^6\text{Li}, ^4\text{He})^{234}\text{Pa}$ by Nayak et al. [13] support the theoretical approach in case of $^{233}\text{Pa}(n,F)$ reaction. The predicted trend of $^{231}\text{Pa}(n,F)$ cross section up to $E_n=20$ MeV, which is similar to that of $^{233}\text{Pa}(n,F)$, is consistent with fissilities of Pa nuclides, stemming from analysis of $^{232}\text{Th}(p,F)$ and $^{232}\text{Th}(p,3n)$ data. The influence of the interplay of Pa fission barriers and entrance channel on the fission observables is shown to be different in case of $n(p)+^{232}\text{Th}$ and $n(p)+^{238}\text{U}$. For $p+^{232}\text{Th}$ interaction the Pa nuclei are responsible for $\sigma_{pF} > \sigma_{nF}$ at $18 \leq E_{n(p)} \leq 100$ MeV. For ^{232}Pa target σ_{nF} approaches σ_{pF} of ^{232}Th at $E_{n(p)} \sim 80$ MeV. In case of nucleon-induced fission of ^{232}Th the entrance channel plays a decisive role at $E_{n(p)} \geq 100$ MeV. The prediction of $^{231-233}\text{Pa}$ fission cross sections up to $E_n = 200$ MeV was achieved for preferential contribution of fission of neutron-deficient nuclides. The measured data on symmetric fission yield for $^{232}\text{Th}(p,F)$ are reproduced, similar increase of symmetric fission yield for $^{231,232,233}\text{Pa}(n,F)$ reaction is predicted at $E_n \sim 50$ MeV. Present estimate of $^{232}\text{Th}(p,F)$ cross section differs essentially from the phenomenological estimate [34].

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