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Fission at intermediate nucleon energies

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Abstract. In the present work Monte Carlo calculations of fission of actinides and pre-actinides induced by protons and neutrons in the energy range from 100 MeV to 1 GeV are carried out by means of a recent version of the Liège Intranuclear Cascade Model, INCL++, coupled with different evaporation-fission codes, in particular GEMINI++ and ABLA07. Fission model parameters are adjusted on experimental (p, f) cross sections and used to predict (n, f) cross sections, in order to provide a theoretical support to the campaign of neutron cross section measurements at the n_TOF facility at CERN.

1. Introduction

Fission induced by nucleons at intermediate energies is important in both fundamental and applied nuclear physics. From a fundamental viewpoint a theory able to describe nucleon-induced fission over a broad range of target nuclei and a wide interval of projectile energies is still lacking; therefore, Monte Carlo simulations able to reproduce the sparse data on (p, f) and (n, f) reactions at intermediate energies constitute a valuable integration.

From an applied viewpoint, the above mentioned data are very important for energy production, radioactive waste transmutation and radiation shield design for accelerators. In particular, high-accuracy data of neutron-induced fission cross sections are essential to the design of Generation IV reactors and, as far as neutron intermediate energies are concerned, of accelerator-driven subcritical systems (ADS). This is the reason why fission cross sections in the unprecedented range from thermal energies up to about 1 GeV are measured at the n_TOF facility [1] at CERN, exploiting the neutron beam produced by 20 GeV/c protons from the PS accelerator impinging on a lead spallation target.

Fission cross sections for natural lead and bismuth [2], forming the eutectic system that should act as a spallation target and a coolant in an ADS, as well as for actinides of the U-Th cycle and minor actinides [3], have been measured relative to the $^{235}\text{U}(n, f)$ or the $^{238}\text{U}(n, f)$ cross sections, which are considered fission standards at low energy.

In order to obtain absolute fission cross sections of actinides and pre-actinides in the whole energy range measured at n_TOF, experimental ratios have been normalized to an evaluated $^{235}\text{U}(n, f)$ cross section, taken from the ENDF/B-VII library [4] up to 30 MeV and from the JENDL/HE-2007 library [4] from 30 MeV to 1 GeV. It is then natural to investigate whether the choice made for the normalization in the intermediate energy range, from about 100 MeV to 1 GeV, is compatible with recent absolute data of the (p, f) reaction, since it is expected



that at energies well above the fission barrier neutrons and protons have a similar behaviour and the corresponding cross sections a similar energy trend. In Ref. [5], (p, f) cross sections for several pre-actinides and actinides, including, of course, ^{235}U , have been measured at nine proton energies in the range from 200 MeV to 1 GeV and are used to calibrate the fission model parameters for our (n, f) calculations.

2. The models

In the energy range of interest, fission induced by nucleons can be seen as a two-stage process: a fast cascade stage, initiated by the high energy projectile, and representable as a succession of two-body collisions, with emission of fast nucleons, light clusters, pions, etc, leaving an excited residual nucleus, and the slow decay stage, where the remnant decays by evaporation, fission, or other mechanisms. In our system of codes, the intranuclear cascade is described by a recent C++ version of the Liège Intranuclear Cascade Model, INCL++ [6], the evaporation-fission model by a C++ version of GEMINI, GEMINI++ [7, 8], or a Fortran version of ABLA07 [9].

INCL++ [6] is a time-like intranuclear cascade model. At the beginning of the cascade stage, the incident nucleon is located with its own impact parameter on the surface of a working sphere, centered on the target nucleus with a radius $R_{max} = R_0 + 8a$, where R_0 and a are the radius and the diffuseness of the target nucleus, respectively. Particles move along straight-line trajectories between collisions in the working sphere and are divided into participants and spectators in the usual sense. Participants that leave the working sphere are considered as ejectiles. Inside the working sphere, nucleons feel a potential that depends on energy and isospin. The depth of the potential well decreases linearly with increasing energy, from ordinary values at the Fermi level to zero at about 200 MeV. The isospin dependence is such that neutron and proton Fermi levels have the same energy.

Collisions are, of course, governed by Pauli blocking, treated in a different way in the first and in the subsequent collisions. The nucleons involved in the first collision are subject to a strict blocking: after the collision, both of them should lie outside the Fermi sphere. In subsequent collisions, the blocking is applied stochastically, with a probability given by the product of final-state blocking factors. A careful definition of the latter allows one to account for surface effects and for the depletion of the Fermi sphere during the evolution of the cascade.

An important novelty of the present version of the code is the introduction of a coalescence model based on phase space, which permits the emission of light clusters, with mass $A \leq 8$, during the cascade stage, in keeping with experimental evidence.

Pions are produced in inelastic nucleon-nucleon collisions through the excitation and subsequent decay of Δ resonances, which sets an upper limit of the order of 3 GeV to the incident nucleon energy for the mechanism of pion production to be valid. The lower energy limit is given by the requirement that the de Broglie wave length of relative motion be much smaller than the range of nuclear forces, which in turn is smaller than the average distance of neighbouring nucleons and is commonly set to 200 MeV, although the model performs reasonably well even at lower energies, as shown in fission calculations of the following section.

An important characteristic of the model is the self-consistent determination of the stopping time of the cascade, which can be simply parametrized as $t_{stop} = 29.8A_T^{0.16}$ fm/c, with A_T the mass of the target nucleus. At $t = t_{stop}$ many physical quantities, such as the excitation energy of the target nucleus and the average kinetic energy of the ejectiles, do not undergo a rapid variation any more, but only a much slower change. Thanks to this choice of the stopping time, it is not necessary to introduce a pre-equilibrium model describing the intermediate stage between the fast cascade and the evaporation-fission decay.

GEMINI++ [7, 8] is a statistical-model code which follows the decay of a compound nucleus by a series of sequential binary decays until such decays are forbidden by energy conservation or become improbable because of gamma-ray competition. Differently from most statistical-model

codes, light-particle evaporation is described by the Hauser-Feshbach formalism, which strictly conserves angular momentum, at the price of higher computational time with respect to the more common Weisskopf-Ewing formalism. An important ingredient of the decay width is the nuclear level density as a function of excitation energy U and angular momentum J , described by a Bethe formula

$$\rho(U, J) \sim (2J + 1) \exp \left[2\sqrt{a(U, J) U} \right] \quad , \quad (1)$$

where the level density parameter $a(U, J)$ includes the damping of shell correction δW according to Ignatyuk [10]

$$a(U) = \tilde{a}(U) \left[1 + \frac{1 - e^{-\eta U}}{U} \delta W \right] \quad , \quad (2)$$

with $\frac{1}{\eta} = 18.5$ MeV and the asymptotic level density parameter $\tilde{a}(U)$ is taken of the form

$$\tilde{a}(U) = \frac{A}{k_\infty - (k_\infty - k_0) \exp \left(-\frac{\gamma}{k_\infty - k_0} \frac{U}{A} \right)} \quad , \quad (3)$$

with $k_0 = 7.3$ MeV, $k_\infty = 12$ MeV and $\gamma = 0.00493 \exp(0.03332A)$, with A the mass number.

The decay width for symmetric fission, dominant at high excitation energy, is given by the well-known Bohr-Wheeler formula, while the decay width for asymmetric fission is derived from Moretto's formalism [11], based on the concept of conditional fission barrier, i.e. a saddle point configuration with fixed asymmetry of mass and charge of the prefragments. Liquid drop barriers are calculated by means of Sierk's finite-range model [12], with shell and pairing corrections taken from Ref. [13].

An important adjustable parameter in the code is the ratio of the asymptotic level density parameter at the saddle point, \tilde{a}_f , to the same quantity at ground-state deformation, \tilde{a}_n , with a default value of 1.036 [8].

ABLA07 [9] is a statistical code describing the de-excitation of a nucleus in thermal equilibrium by particle evaporation, fission, or, above a prescribed excitation energy per nucleon, multifragmentation. Particle evaporation is treated in an extended Weisskopf-Ewing formalism, where a distribution of orbital angular momenta in the emission of nucleons and clusters is evaluated in semiclassical approximation, based on phase space arguments. An essential ingredient of the decay formalism is the nuclear level density as a function of excitation energy and angular momentum, described by a constant temperature formula at low energy and by a Bethe formula (1) at high energy. The asymptotic level density parameter, \tilde{a} (see formula (2)), is energy-independent and given, in MeV^{-1} , by the original prescription of Ref. [10]

$$\tilde{a} = 0.073A + 0.095B_s A^{2/3} \quad , \quad (4)$$

where B_s is the ratio of the surface of the deformed nucleus to that of a spherical nucleus with the same mass number A . The level density contains a collective enhancement factor, due to nuclear rotations and vibrations, depending on excitation energy. The approach to fission contains elements of dynamics: the time evolution of the fission degree of freedom is treated as a diffusion process, determined by the interaction of collective degrees of freedom with a heat bath formed by the individual nucleons and described by a Fokker-Planck equation whose solution leads to a time-dependent fission width, $\Gamma_f(t)$. An analytical approximation to such a solution and, consequently, to the time dependence of the fission width makes the problem computationally tractable. At low excitation energy, the code uses the standard model of a two-humped fission barrier, whose penetrability is computed in the approximation of full damping of the vibrational resonances in the intermediate well. Like in the GEMINI++ code, liquid drop barriers are computed in the frame of the finite-range model [12] and shell and pairing corrections are taken from Ref. [13].

3. Cross section calculations

In keeping with the philosophy of the authors [6], no parameters have been modified in the INCL++ model, which was already optimized by reproducing a large amount of experimental data connected with the cascade stage, such as total reaction cross sections, double-differential spectra of emitted nucleons and light clusters, etc, while we have taken the freedom to adjust fission parameters in both GEMINI++ and ABLA07. There are essentially two crucial parameters: the height of the fission barrier and the asymptotic level density parameter at the saddle point, \tilde{a}_f , or its ratio to the level density parameter at ground state deformation, \tilde{a}_n . The barrier height is expected to play a main role at low energy, the level density is important in the whole energy range, in particular at high energy.

While reliable experimental values of absolute (p, f) cross sections in the energy range up to 1 GeV exist for a number of pre-actinides and actinides, (n, f) cross section measurements above 200 MeV are relative to $^{235}\text{U}(n, f)$ or $^{238}\text{U}(n, f)$, which are not experimentally known, but computed by means of some theoretical model. Even if the measured absolute (n, f) cross sections below 200 MeV are systematically lower than the (p, f) cross sections corresponding to the same target nuclei, the effect being more significant for pre-actinides than for actinides, it is expected that, with increasing incident energy, the behaviour of neutrons and protons becomes more and more similar; consequently, (n, f) and (p, f) cross sections are expected to show a similar energy trend and comparable absolute values. Thus, the basic strategy of the present work is to reproduce (p, f) and (n, f) data for the same target with the same model parameters, or, at least, as close as possible in the proton and neutron channels. Where only (p, f) data exist, the model parameters that reproduce them are used to predict (n, f) data.

Figure 1 compares the (p, f) and (n, f) cross sections of $^{235,238}\text{U}$ calculated with the INCL++ + GEMINI++ chain with selected experimental data and with the JENDL/HE-2007 evaluations. The fission model parameters adjusted in GEMINI++ so as to reproduce the (p, f) data of Ref. [5] are the \tilde{a}_f/\tilde{a}_n ratio and the height of the liquid drop fission barrier. For ^{235}U we have chosen $\tilde{a}_f/\tilde{a}_n = 1.050$ and reduced the height of the fission barriers of all residual nuclei by the same amount, $\Delta B_f = -0.2$ MeV, for ^{238}U we have adopted $\tilde{a}_f/\tilde{a}_n = 1.038$ and $\Delta B_f = -0.3$ MeV. The parameters adjusted on the (p, f) cross sections have been used to compute the corresponding (n, f) cross sections: as shown by the figure, the (n, f) data in the energy range from 100 to 200 MeV are satisfactorily reproduced and the predicted cross sections at higher energy are rather close to the corresponding (p, f) cross sections. It is worthwhile to stress that the (n, f) data from Refs. [15, 16] can be considered as absolute cross sections, since the neutron flux is determined by simultaneous measurement of (n, p) scattering events and knowledge of the differential (n, p) cross section in the energy range of interest, while the data of Ref. [17] are relative to the $^{235}\text{U}(n, f)$ cross section, normalized in the intermediate energy range according to the recommendations of Ref. [18].

As for the comparison with JENDL/HE-2007, it is worthwhile to point out that the (p, f) evaluations do not take into account the data of Ref. [5], but are consistent with older data, in conflict with Ref. [5], used by Prokofiev [19] for his (p, f) systematics, which exhibit a steady decrease with increasing energy in the range from 500 MeV to 1 GeV, at variance with Ref. [5], whose data show a large plateau reproduced in our calculations. Two arguments supporting our choice of basic reference data [5] should be mentioned: the responsible for the JENDL evaluations, Tokio Fukahori, co-authored Ref. [5] and reproduced those data in good agreement with our calculations in the range from 200 MeV to 1 GeV; a recent $^{238}\text{U}(p, f)$ measurement [14] in inverse kinematics yields two additional points, shown in figure 1, in excellent agreement with the data of Ref. [5].

As for (n, f) cross sections, JENDL/HE-2007 reproduces the data below 200 MeV very well, but shows in the range from 500 MeV to 1 GeV a fast decrease with increasing energy which is not consistent with the present experimental evidence for (p, f) cross sections. Our calculations

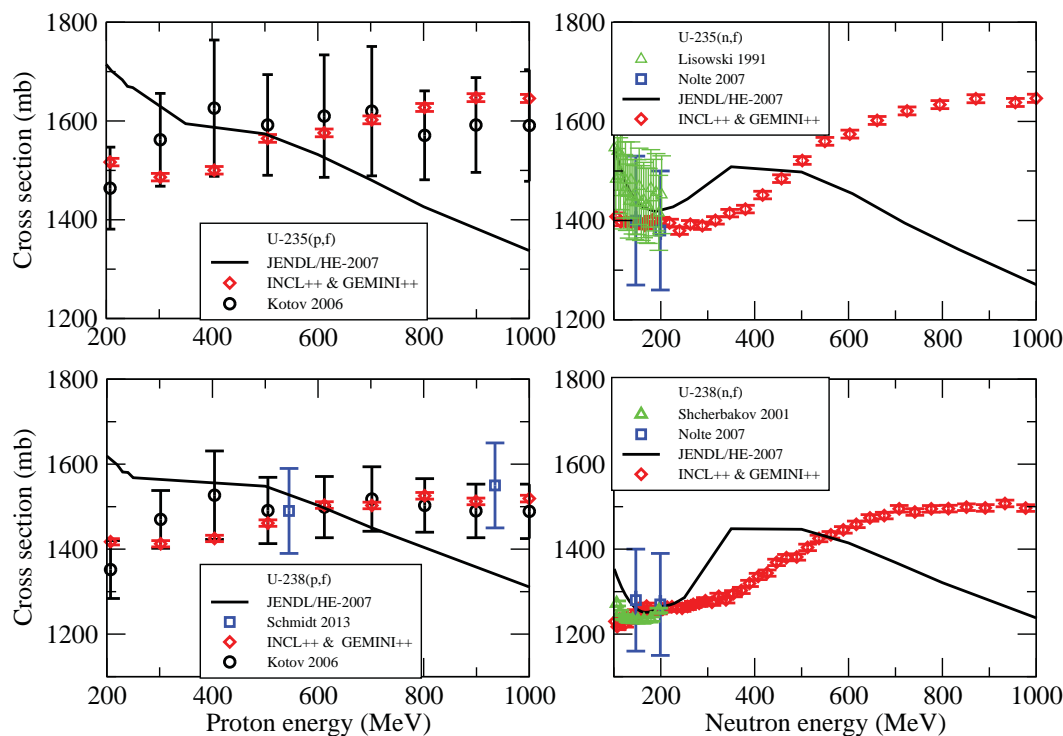


Figure 1. (Color online) Proton- and neutron-induced fission cross sections of ^{235}U and ^{238}U . $^{235}\text{U}(p, f)$: data from Ref. [5]; $^{238}\text{U}(p, f)$: data from Refs. [5, 14]; $^{235}\text{U}(n, f)$: data from Refs. [15, 16]; $^{238}\text{U}(n, f)$: data from Refs. [16, 17].

justify *a posteriori* the use of the intranuclear cascade model in the energy range from 100 to 200 MeV, where the formal conditions of applicability are not so well satisfied. At the present time, the main source of (n, f) data at intermediate energies is the n_TOF experiment [1], where the measurements are relative to the $^{235}\text{U}(n, f)$ and $^{238}\text{U}(n, f)$ cross sections. Final results for ^{234}U and ^{237}Np up to 1 GeV are published in Ref. [3], natural lead and ^{209}Bi are described in Ref. [2], while preliminary data are available for ^{232}Th and $^{233,238}\text{U}$. In the case of ^{234}U , since no (p, f) data exist in the energy range of interest, we have assumed in our GEMINI++ calculations the same model parameters of ^{235}U , namely $\tilde{a}_f/\tilde{a}_n = 1.050$ and $\Delta B_f = -0.2$ MeV. Figure 2 shows that theoretical and experimental $\sigma_{nf}(^{234}\text{U})/\sigma_{nf}(^{235}\text{U})$ ratios are in excellent agreement, while the absolute (n, f) cross sections diverge at high energy, owing to the normalization to the JENDL/HE-2007 cross section of ^{235}U , adopted in Ref. [3]. The fission model of ABLA07 is more sophisticated, but less flexible than the one of GEMINI++: the default parameters should be modified very carefully, in order not to spoil the internal consistency. In the present work we have changed only the most crucial quantity, \tilde{a}_f , by multiplying it by a constant factor for all fissioning nuclei. The price we pay is that, in general, it is not possible to reproduce (p, f) and (n, f) data of a given target nucleus with the same value of \tilde{a}_f : for instance, the best reproduction of $^{235}\text{U}(p, f)$ data from Ref. [5] is obtained by increasing the default \tilde{a}_f value (4) by 6%, while reproducing the $^{235}\text{U}(n, f)$ data below 200 MeV [15, 16] requires an increase of the order of 10%. It is however, expected that acting on other parameters of the fission model this inconvenience can be removed. The \tilde{a}_f values adopted in the proton and neutron channels are closer for pre-actinides than for actinides: for instance, $^{nat}\text{Pb}(p, f)$ data from Ref. [5] are reproduced at best with the default \tilde{a}_f value (4) and $^{nat}\text{Pb}(n, f)$ data from Ref. [2] by increasing it by 1%. The same increase by 1% is adopted in reproducing $^{209}\text{Bi}(p, f)$ [5] and $^{209}\text{Bi}(n, f)$ [2].

Figure 3 compares theoretical and experimental (p, f) cross sections of ^{nat}Pb and ^{209}Bi and figure 4 shows theoretical and experimental ratios to the $^{235}\text{U}(n, f)$ cross section as well as absolute (n, f) cross sections: for the former the agreement is satisfactory, for the latter the results diverge in the energy region from 500 MeV to 1 GeV because of the normalization to the JENDL/HE-2007 evaluation adopted in Ref. [2].

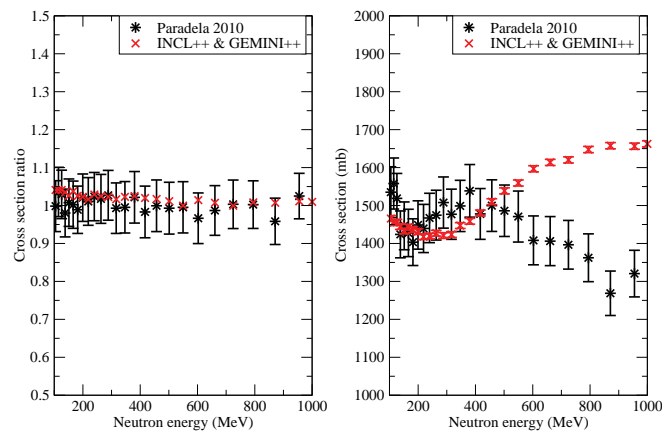


Figure 2. (Color online) Left panel: theoretical and experimental [3] ratio $\sigma_{nf}(^{234}\text{U})/\sigma_{nf}(^{235}\text{U})$. Right panel: theoretical $^{234}\text{U}(n, f)$ cross section compared with the experimental ratio normalized to JENDL/HE-2007.

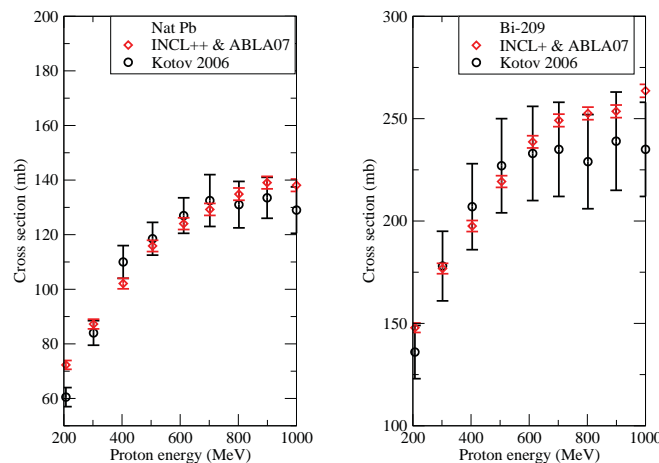


Figure 3. (Color online) (p, f) cross sections of ^{nat}Pb and ^{209}Bi . Experimental data from Ref. [5].

Summing up, experimental (n, f) ratios measured at n_TOF for pre-actinides and actinides are satisfactorily reproduced by using fission model parameters that are similar, or rather close, to the ones adopted in reproducing experimental (p, f) cross sections. Absolute cross sections are larger than those obtained by the n_TOF Collaboration in the energy range from 500 MeV to 1 GeV because of different normalization.

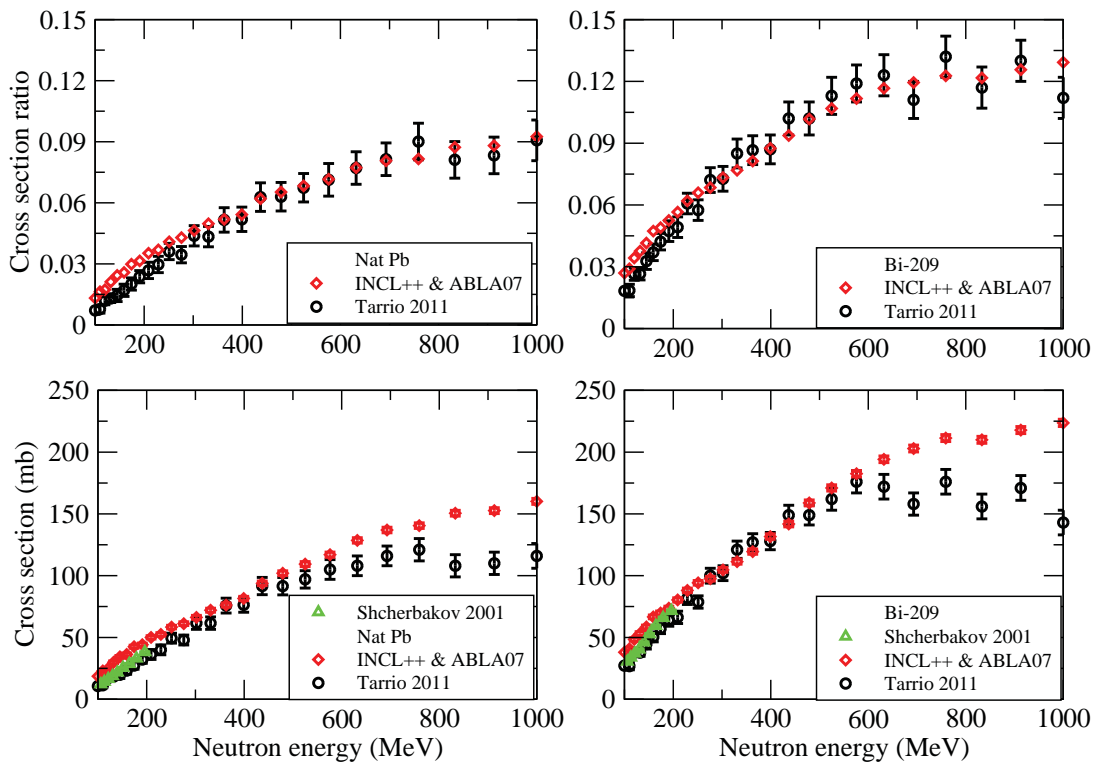


Figure 4. (Color online) (n, f) cross sections of ^{nat}Pb and ^{209}Bi . Upper panels: theoretical vs experimental [2] ratios to $^{235}\text{U}(n, f)$. Lower panels: absolute cross sections.

4. Conclusions and outlook

The preliminary calculations presented in this note indicate that the Liège Intranuclear Cascade Model, INCL, coupled with an appropriate evaporation-fission model like GEMINI or ABLA07, can reproduce the experimental data of fission of actinides and pre-actinides induced by protons and neutrons at intermediate energies with the same, or almost the same model parameters. As a consequence, predicted (n, f) cross sections are rather similar to (p, f) cross sections at high energies, while they are systematically lower than (p, f) cross sections below 200 MeV, in agreement with experiments.

The present study is also intended as a theoretical support to the (n, f) measurements at intermediate energies performed at the n_TOF facility at CERN: in particular, it offers a valid alternative to the normalization of experimental ratios to the JENDL evaluation of the $^{235}\text{U}(n, f)$ cross section adopted in Refs. [2, 3], which does not seem to be consistent with recent data of the (p, f) cross section in the same energy range.

Our preliminary results encourage us to carry out systematic calculations of (p, f) and (n, f) reactions for actinides and pre-actinides and compare our theoretical results with the most recent and reliable experimental data. As a by-product of such a systematic study, it will be of interest to examine the dependence of model parameters that simultaneously reproduce (p, f) and (n, f) data on characteristics of target nuclei, such as the fissility parameter, proportional to Z^2/A . In particular, it has been shown [20] that the $\sigma_{p,f}/\sigma_{n,f}$ ratio at $E = 180$ MeV decreases exponentially with increasing Z^2/A for a number of pre-actinides and actinides and the experimental result has been interpreted in terms of ratios of fission probabilities, depending, in turn, on neutron and proton fission barriers, B_f^n and B_f^p . In pre-actinides, $B_f^p < B_f^n$, resulting in $\sigma_{p,f}/\sigma_{n,f} > 1$, while, in actinides, $B_f^p \cong B_f^n$, yielding $\sigma_{p,f}/\sigma_{n,f} \sim 1$. Thus, it will be of interest to check this

interpretation on the basis of more recent data at other intermediate energies.

Finally, it is our intention to make (n, f) predictions for other isotopes whose measurements are already planned at CERN, not only of fission cross sections, but also of angular distributions of fission fragments, which are also measurable at the n_TOF facility.

Acknowledgments

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