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PROTON-INDUCED FISSION CROSS SECTION CALCULATION WITH THE LANL CODES CEM2K+GEM2 AND LAQGSM+GEM2

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ABSTRACT

The improved Cascade-Exciton Model code CEM2k and the Los Alamos version of the Quark-Gluon String Model code LAQGSM, previously merged with the Generalized Evaporation Model code of Furihata (GEM2) were further modified to provide reliable proton-induced fission cross sections for applications. By adjusting two parameters in GEM2 for each measured reaction, we were able to describe very well with CEM2k+GEM2 and LAQGSM+GEM2 all available experimental fission cross sections induced by protons with energies from 20 MeV to 10 GeV both for subactinide and actinide targets. We also successfully tested our approach on several reactions induced by neutrons, pions, and photons.

Introduction

In recent years, an improved version of the Cascade-Exciton Model (CEM), contained in the code CEM2k [1] and the Los Alamos version of the Quark-Gluon String Model, implemented in the high-energy code LAQGSM [2] have been developed at the Los Alamos National Laboratory for a number of applications. CEM2k is intended to describe nucleon-, pion-, and photon-induced reactions at incident energies up to about 5 GeV, while LAQGSM describes both particle- and nucleus-nucleus reactions at energies up to about 1 TeV/nucleon. Originally, both CEM2k and LAQGSM were not able to describe fission reactions and production of light fragments heavier than ^{4}He, as they had neither a high-energy-fission nor a fragmentation model. Recently, we addressed these problems [3, 4] by further improving our codes and by merging them with the Generalized Evaporation Model code GEM2 developed by Furihata [5].

GEM2 is an extension by Furihata of the Dostrovsky *et al.* [6] evaporation model as implemented in LAHET [7] to include up to 66 types of particles and fragments that can be evaporated from an excited compound nucleus plus a modification of the version of Atchison's fission model [8] used in LAHET. It was found [3, 4] that if we were to merge GEM2 with CEM2k or LAQGSM without any modifications, the new code would not describe correctly the fission cross section (and the yields of fission fragments). This is because Atchison fitted the parameters of his fission model when it was coupled with the Bertini Intra-Nuclear Cascade (INC) [9] which differs from our INC. In addition, Atchison did not model preequilibrium emission. Therefore, the distributions of fissioning nuclei in A, Z, and excitation energy E^* simulated by Atchison differ significantly of the distributions we get; as a consequence, all the fission characteristics are also different. Furihata used GEM2 coupled either with the Bertini INC [9] or with the ISABEL [10] INC code, which also differs from our INC, and did not include preequilibrium particle emission. Therefore the real fissioning nuclei simulated by Furihata differ from the ones in our simulations, and the parameters adjusted by Furihata to work the best with her INC will not be the best for us. To get a good description of fission cross sections (and fission-fragment yields) we need to modify at least two parameters in GEM2 (see details in [3, 4]). This problem was solved both for CEM2k+GEM2 and LAQGSM+GEM2 in the present work.

Calculation of σ_f in GEM2

A comprehensive description of GEM2 was published by Furihata [5], some details may be found in our papers [3, 4], therefore we recall here only how fission cross sections are calculated by GEM2, as we need to modify them here. The fission model used in GEM2 is based on Atchison's model [8], often referred in the literature as the Rutherford Appleton Laboratory (RAL) model, which is where Atchison developed it. There are two choices of parameters for the fission model: one of them is the original parameter set by Atchison [8] as implemented in LAHET [7], and the other is a parameter set evaluated by Furihata [5], used here as a default of GEM2.

The Atchison fission model is designed to only describe fission of nuclei with $Z \geq 70$ (we extended it in our codes down to $Z \geq 65$). It assumes that fission competes only with neutron emission, *i.e.*, from the widths Γ_j of n, p, d, t, ^{3}He, and ^{4}He, the RAL code calculates the probability of evaporation of any particle. When a charged particle is selected to be evaporated, no fission competition is taken into account. When a neutron is selected to be evaporated, the code does not actually simulate its evaporation, instead it considers that fission may compete, and chooses either fission or evaporation of a neutron according to the fission probability P_f. This quantity is treated by the RAL code differently for the elements above and below $Z = 89$.

1) $70 \leq Z_j \leq 88$. For fissioning nuclei with $70 \leq Z_j \leq 88$, GEM2 uses the original Atchison calculation of the neutron emission width Γ_n and fission width Γ_f to estimate the fission probability as

$$P_f = \frac{\Gamma_f}{\Gamma_f + \Gamma_n} = \frac{1}{1 + \Gamma_n/\Gamma_f}. \qquad (1)$$

Atchison uses [8] the Weisskopf and Ewing statistical model [11] with an energy-independent pre-exponential factor for the level density and Dostrovsky's [6] inverse cross section for neutrons and estimates the neutron width Γ_n as

$$\Gamma_n = 0.352\big(1.68J_0 + 1.93A_i^{1/3}J_1 + A_i^{2/3}(0.76J_1 - 0.05J_0)\big),$$

where J_0 and J_1 are functions of the level density parameter a_n and $s_n(= 2\sqrt{a_n(E - Q_n - \delta)})$ as

$$J_0 = \frac{(s_n - 1)e^{s_n} + 1}{2a_n},$$

$$J_1 = \frac{(2s_n^2 - 6s_n + 6)e^{s_n} + s_n^2 - 6}{8a_n^2}.$$

The RAL model uses a fixed value for the level density parameter a_n, namely

$$a_n = (A_i - 1)/8.$$

The fission width for nuclei with $70 \leq Z_j \leq 88$ is calculated in the RAL model and in GEM2 as

$$\Gamma_f = \frac{(s_f - 1)e^{s_f} + 1}{a_f},$$

where $s_f = 2\sqrt{a_f(E - B_f - \delta)}$ and the level density parameter in the fission mode a_f is fitted by Atchison to describe the measured Γ_f/Γ_n as

$$a_f = a_n\Big(1.08926 + 0.01098(\chi - 31.08551)^2\Big), \qquad (2)$$

and $\chi = Z^2/A$.

2) $Z_j \geq 89$. For heavy fissioning nuclei with $Z_j \geq 89$ GEM2 follows the RAL model and does not calculate at all the fission width Γ_f and does not use Eq. (1) to estimate the fission probability P_f. Instead, the following semi-empirical expression obtained by Atchison by approximating the experimental values of Γ_n/Γ_f published by Vandenbosch and Huizenga [12] is used to calculate the fission probability:

$$\log(\Gamma_n/\Gamma_f) = C(Z_i)(A_i - A_0(Z_i)), \qquad (3)$$

where $C(Z)$ and $A_0(Z)$ are constants dependent on the nuclear charge Z only. The values of these constants are those used in the current version of LAHET [7] and are tabulated in Table 1 (note that some adjustments of these values have been done since Atchison's papers [8] were published).

Table 1. $C(Z)$ and $A_0(Z)$ values used in GEM2

Z	$C(Z)$	$A_0(Z)$
89	0.23000	219.40
90	0.23300	226.90
91	0.12225	229.75
92	0.14727	234.04
93	0.13559	238.88
94	0.15735	241.34
95	0.16597	243.04
96	0.17589	245.52
97	0.18018	246.84
98	0.19568	250.18
99	0.16313	254.00
100	0.17123	257.80
101	0.17123	261.30
102	0.17123	264.80
103	0.17123	268.30
104	0.17123	271.80
105	0.17123	275.30
106	0.17123	278.80

Prokofiev's Approximation of σ_f

We choose not to use in the present work experimental fission cross sections directly as they are published in the literature. Fig. 1 (kindly provided by Dr. Prokofiev) explains well the reason: The point is that for intermediate- and high-energy reactions, where our codes are supposed to be used, the experimental data on proton-induced fission cross sections are sparse and not as precise as for low-energy reactions measured for reactor applications. Intermediate- and high-energy experimental fission cross sections induced by neutrons, pions, and other projectiles are even more sparse than the ones measured with protons. As one can see from Fig. 1, fission cross sections measured at such energies in different experiments differ so significantly from each other that it is difficult to use such data in development and validation of models and codes, without a special analysis of all details of every measurement. Fortunately, this has been done by Prokofiev [13] so we use here his results. Prokofiev spent many years on compiling proton-induced measured fission cross sections and on analyzing the details of each experiment. As a result, he divided all measurements into three categories: 1) the highest, where obtained data are very reliable and can be used without any mistrust; 2) high-quality data, reliable, but requiring some normalization; 3) data of low reliability, that would be better not used. Then, using only measurements from the first group and data from the second group after a corresponding re-normalization, Prokofiev developed systematics for proton-induced fission cross sections for all preactinide and actinide nuclei for which he was able to find enough data [13, 14]. At our energies, we consider Prokofiev's systematics as the most reliable "experimental" fission cross sections and prefer to use them to develop and test our codes instead of using experimental values published in original publications by different authors.

For subactinide nuclei from ^{165}Ho to ^{209}Bi and incident proton energies above 70 MeV, Prokofiev proposed [13] the following universal parameterization for the proton-induced fission cross section, $\sigma_f(E_p)$ [mb]:

$$\begin{aligned}\sigma_f(E_p) &= P_1\{1-\exp[-P_3(E_p-P_2)]\} \\ &\times (1-P_4 \ln E_p),\end{aligned} \tag{4}$$

where E_p is the incident proton energy [MeV] and P_1, P_2, P_3, and P_4 are fitting parameters. P_4 was fitted as

$$P_4(Z^2/A) = \begin{cases} 0 & \text{if } Z^2/A \leq 32.32, \\ Q_{4,1} + Q_{4,2} Z^2/A & \text{if } Z^2/A > 32.32, \end{cases} \tag{5}$$

where fitting parameters Q_{ij} are given in Table 2. Parameters P_1, P_2, and P_3 were fitted as

$$P_i(Z^2/A) = \exp[Q_{i,1}+Q_{i,2}(Z^2/A)+Q_{i,3}(Z^2/A)^2]. \tag{6}$$

Table 2. Parameters Q_{ij} in the $P_i(Z^2/A)$ systematics for target nuclei from Ho to Bi [13]

i	$j=1$	$j=2$	$j=3$
1	119.0	-7.852	0.1332
2	9.979	-0.1847	0
3	-27.40	0.6792	0
4	-1.140	0.0352	0

For actinide nuclides from ^{232}Th to ^{239}Pu and incident proton energies above 20 MeV, Prokofiev found [13] $P_2 = 12.1$, $P_3 = 0.111$, $P_4 = 0.067$, and

$$P_1(Z^2/A) = R_{11}\{1-\exp[-R_{13}(Z^2/A - R_{12})]\}, \tag{7}$$

where $R_{11} = 2572$, $R_{12} = 34.99$, and $R_{13} = 2.069$. Numerical values of all P_i parameters of the nuclear targets fitted by Prokofiev together with the energy interval of fitting are published in Tab. 4 of Ref. [13].

In Ref. [14], Prokofiev extended his systematics to describe fission cross sections of preactinide nuclei from ^{197}Au to ^{209}Bi in the energy region from 35 to 70 MeV and to predict fission cross sections for nuclei between ^{209}Bi and ^{232}Th, where not a single data point is available at present. It was found [14] that one can approximate fission cross sections of preactinides between Au and Bi at proton energies between 35 and 70 MeV with the formula

$$\sigma(E_p) = \sigma_0 \exp\Big[-\frac{(E_p-E_0)^2}{2w^2}\Big], \tag{8}$$

where $E_0 = 76.3$ MeV. Parameters w and σ_0 depend on the fissioning system and characterize, respectively, the steepness and the absolute scale of the fission excitation functions and are approximated as following:

$$w(A,Z) = a + b(Z^2/A) + c\,\delta W_{gs}(A,Z), \tag{9}$$

where δW_{gs} is the shell correction to the ground-state mass of the fissioning nucleus calculated using the systematics of Myers and Swiatecki [15], and $a = -33.667$, $b = 1.5699$, and $c = 0.30069$. Parameter σ_0 was fitted as

$$\sigma_0 = \sigma_b \exp\Big[\frac{(E_b-E_0)^2}{2w^2}\Big], \tag{10}$$

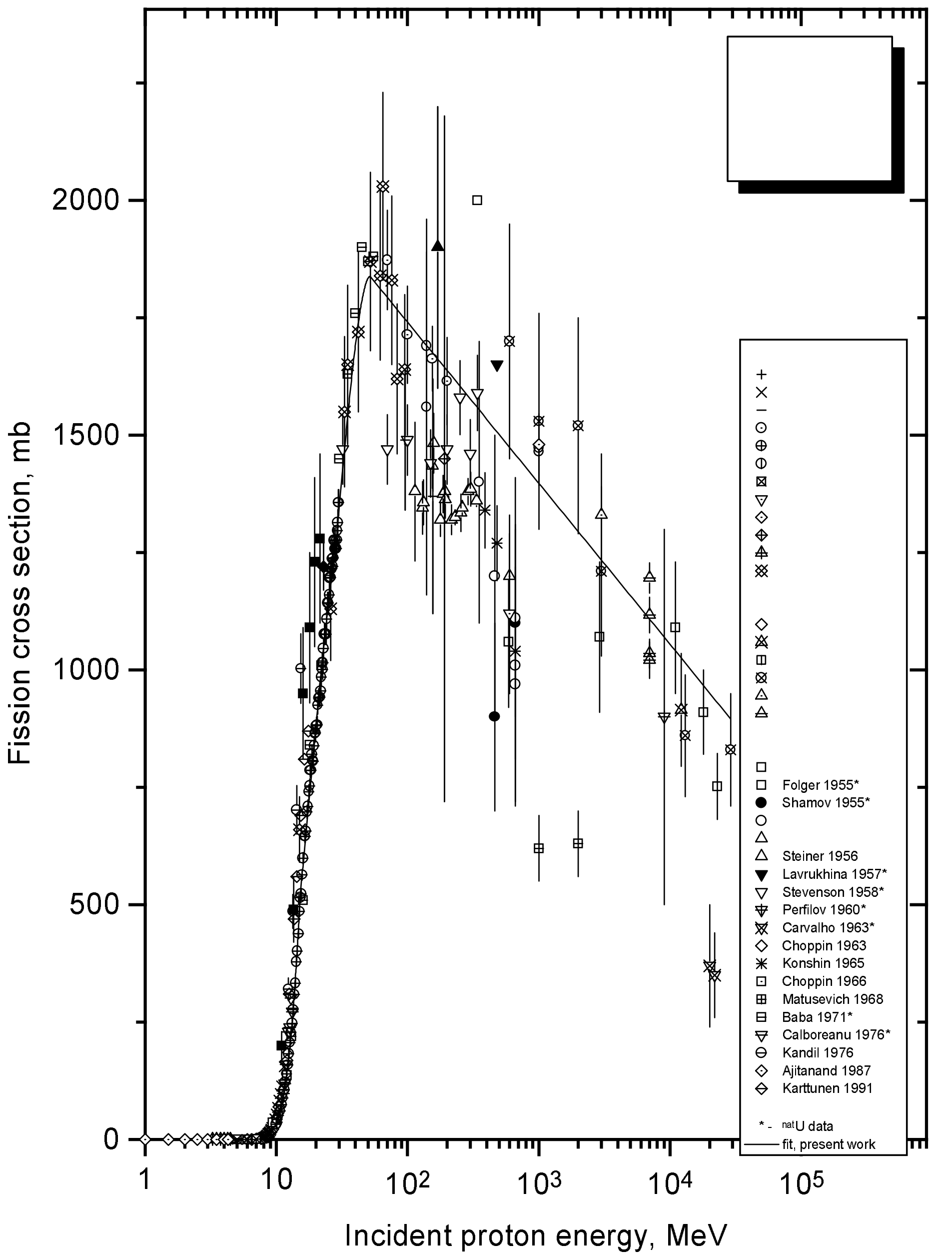
where $E_b = 70$ MeV and $\sigma_b = \sigma(E_b)$ is calculated according to the high-energy systematics given by Eq. (4).

To predict fission cross sections for nuclei between ^{209}Bi and ^{232}Th at proton energies above 70 MeV were there are no data, it was suggested [14] that parameters P_i of Eq. (4) can be found by interpolation of the systematics [13] predictions. The logarithmic interpolation scheme was chosen [14]:

$$\ln P_i = C_{i1} + C_{i2}\,x, \tag{11}$$

where the constants C_{ij} $(i = 1 \ldots 4, j = 1,2)$ are calculated as following:

$$C_{i1} = \frac{x_{Th}\ln P_i(x_{Bi}) - x_{Bi}\ln P_i(x_{Th})}{x_{Th}-x_{Bi}}, \tag{12}$$


Fission cross section, mb
Incident proton energy, MeV
0
500
1000
1500
2000
1
10
10^2
10^3
10^4
10^5
Folger 1955*
Shamov 1955*
Steiner 1956
Lavrukhina 1957*
Stevenson 1958*
Perfilov 1960*
Carvalho 1963*
Choppin 1963
Konshin 1965
Choppin 1966
Matusevich 1968
Baba 1971*
Calboreanu 1976*
Kandil 1976
Ajitanand 1987
Karttunen 1991
* - ^{nat}U data
fit, present work

tivity of results to these parameters is much higher than to fission barriers used in calculation or other parameters of the model. Therefore we choose to adjust only these two parameters in our merged CEM2k+GEM2 and LAQGSM+GEM2 codes. We do not change the form of systematics (2) and (3) derived by Atchison. We only introduce here additional coefficients both to a_f and $C(Z)$, replacing $a_f \to C_a \times a_f$ in Eq. (2) and $C(Z_i) \to C_c \times C(Z_i)$ in Eq. (3) and fit C_a and C_c both for CEM2k+GEM2 and LAQGSM+GEM2 codes for all nuclei and incident proton energies where Prokofiev's systematics apply. No other parameters in GEM2 or our CEM2k and LAQGSM were changed. For preactinides, we had to fit only C_a. The values of C_a found by fitting our results to Prokofiev's predictions are close to one and change smoothly with changing the proton energy and the charge or mass number of the target. Such finding gives us confidence in our procedure, and allows us to interpolate or extrapolate the values of C_a for nuclei and incident proton energies not covered by Prokofiev's systematics. For actinides, as described in [3, 4], we have to fit both C_a and C_c. The values of C_a we find are also very close to one, while the values of C_c are more varied, but both of them change smoothly with the proton energy and Z or A of the target, that again allows us to interpolate and extrapolate them for nuclei and energies outside Prokofiev's systematics.

We fixed the fitted values of C_a and C_c in data blocks in our codes and complemented them with routines for their interpolation/extrapolation outside the region covered by Prokofiev's systematics. We believe that such a procedure provides quite a reliable fission cross section calculation by our codes, at least for proton energies and target-nuclei not too far from the ones covered by Prokofiev's systematics. Our results by CEM2k+GEM2 for preactinides are shown in Fig. 2, and for actinides, in Fig. 3. Results by LAQGSM+GEM2 are very similar, almost coinciding with the ones shown in Figs. 2 and 3, therefore we do not duplicate them here. One can see that after fitting C_a and C_c, the fission cross sections calculated by our codes reproduce very well all the experimental data covered by Prokofiev's systematics.

To see how this approach works for reactions induced by other projectiles, we tested our codes on several reactions induced by neutrons, pions, and photons, without any more changes or fitting. Fig. 4 shows several examples of such results. We see that our codes describe them from quite well to very well, although experimental data on pion-induced fission cross sections are not so rich and precise, and it is difficult to draw conclusions from a comparison to this data. The fact that we give such fits to fission induced by other probes gives us confidence in the value of the fitting procedure we performed in our CEM2k+GEM2 and LAQGSM+GEM2 codes.

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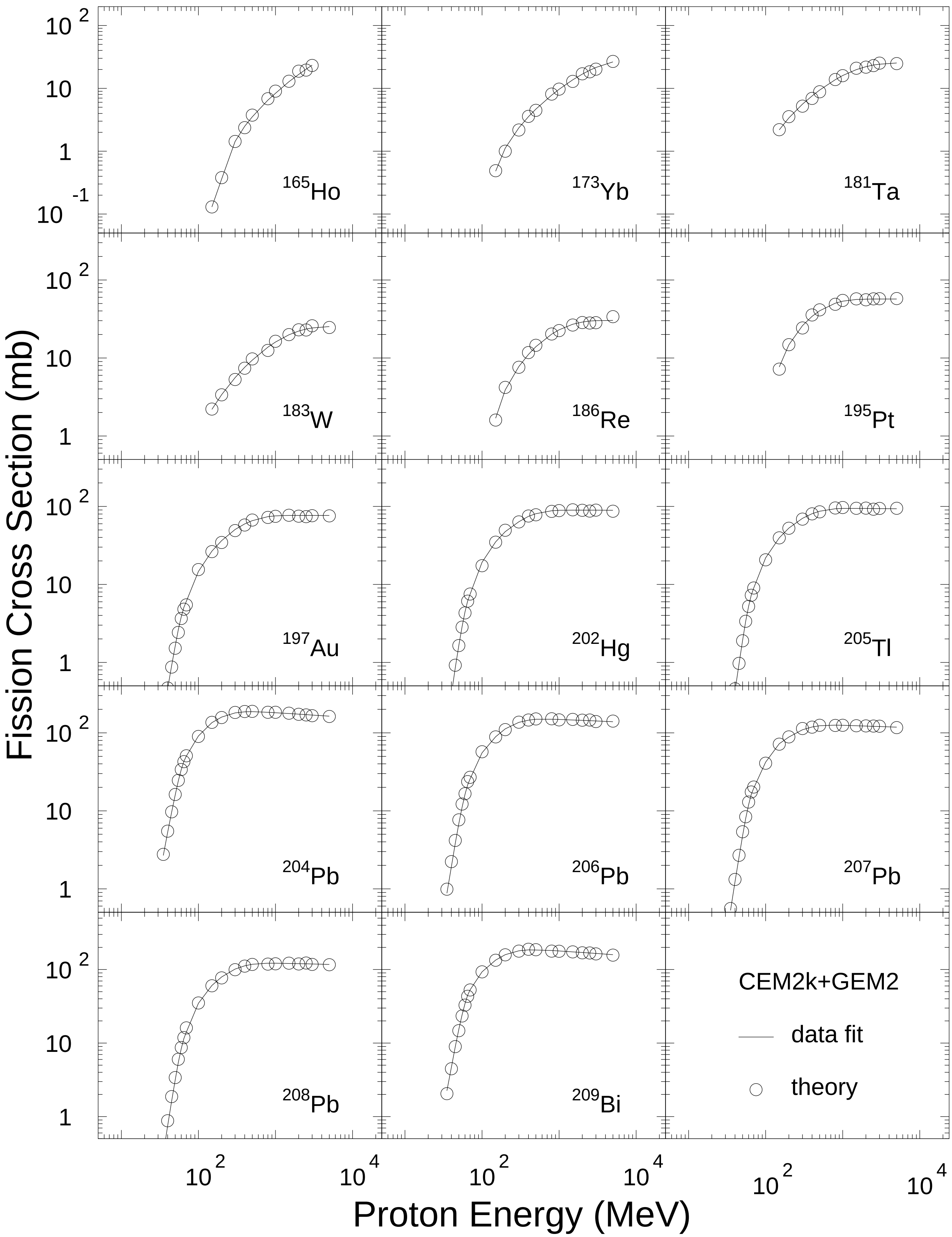

Figure 2: Comparison of Prokofiev's [13, 14] sytematics of experimental (p,f) cross sections of ^{165}Ho, ^{173}Yb, ^{181}Ta, ^{183}W, ^{186}Re, ^{195}Pt, ^{197}Au, ^{202}Hg, ^{205}Tl, ^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb, and ^{209}Bi nuclei (lines) with our present CEM2k+GEM2 calculations (circles).

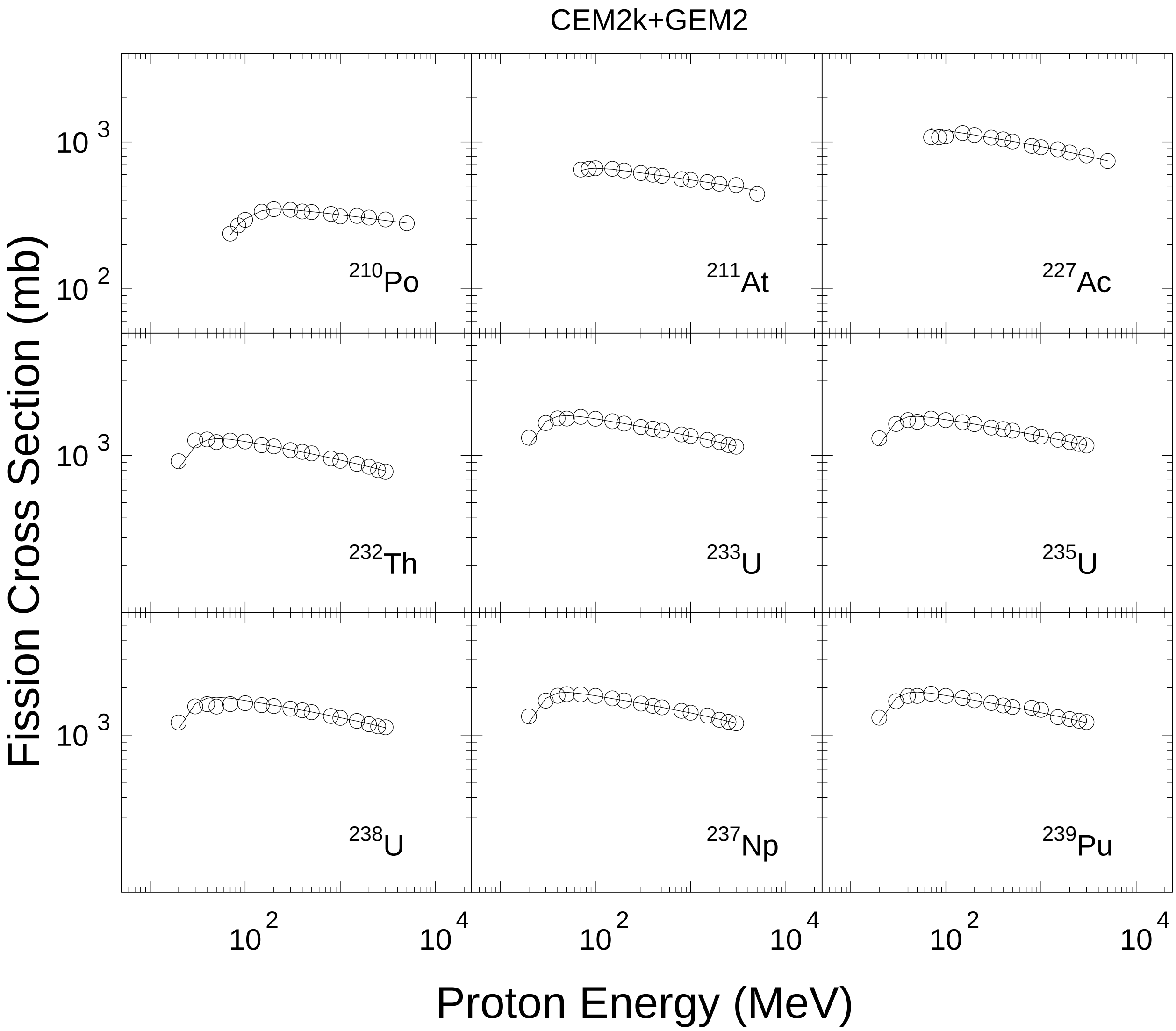

Figure 3: Comparison of Prokofiev's [13] sytematics of experimental (p,f) cross sections of ^{232}Th, ^{233}U, ^{235}U ^{238}U, ^{237}Np, and ^{239}Np nuclei and of predicted [14] (p,f) cross sections for ^{210}Po, ^{211}At, and ^{227}Ac targets (lines) with our present CEM2k+GEM2 calculations (circles).

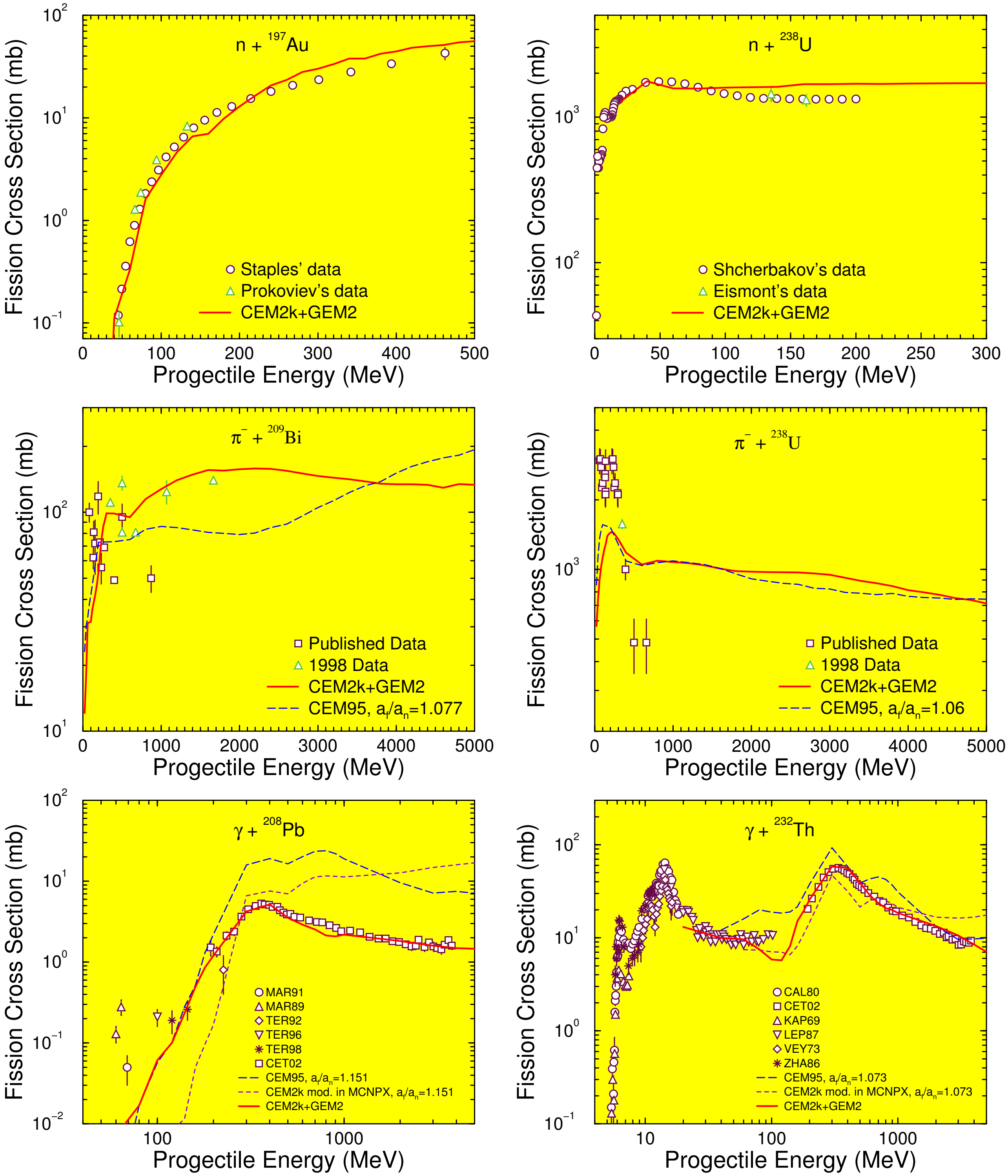

Figure 4: Comparison of calculated by the modified here CEM2k+GEM2 code fission cross sections induced by neutrons on ^{197}Au and ^{238}U, π^- on ^{209}Bi and ^{238}U, and γ on ^{208}Pb and ^{232}Th with experimental data and results by previous versions of CEM (see details and references in [1]), as indicated. Experimental data are from: 1) n: Staples [16], Prokofiev [17]; Shcherbakov [18], Eismont [19]; 2) π^-: [20]; 3) γ: MAR91 [21], MAR89 [22], TER92 [25], TER96 [23], TER98 [24], CET02 [26], CAL80 [27], KAP69 [29], LEP87 [28], VEY73 [30], ZHA86 [31].