Braz J Phys (2016) 46:415–423
DOI 10.1007/s13538-016-0430-7

GENERAL AND APPLIED PHYSICS

Deexcitation Modes in Spallation Nuclear Reactions

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Received: 23 February 2016 / Published online: 21 June 2016

Abstract Spallation nuclear reactions in the range of 0.2 to 1.2 GeV are studied using the CRISP code. A new approach for the deexcitation stage of the compound nucleus was introduced. For the calculations of the level densities, this approach is based on the Back-shifted Fermi gas model (BSFG), which takes into account pairing effects and shell corrections, whereas the calculation of the fission barriers were performed by means of the Extended Thomas-Fermi plus Strutinsky Integral (ETFSI) method, which is a high-speed approximation to the Hartree-Fock method with pairing correlations treated as in the usual BCS plus blocking approach. This procedure is more appropriate to calculate level densities for exotic nuclei. Satisfactory results were obtained and compared with experimental data obtained in the GSI experiments. As another important result, we highlight some directions for the development of a qualitatively superior version of the CRISP code with the implementation of more realistic and suitable physical models to be applied in stable and exotic nuclei that participate in the process. This new version of the code includes several substantial changes in the decay of the hot compound nucleus which allow satisfactory agreement with the experimental data and a reduction of the adjustment parameters.

Keywords Level densities · Fission barrier · Spallation reactions · Neutron multiplicities · Energy correction

1 Introduction

Although spallation reactions have been studied since the mid 50s of the last century, it is in the last decades of this century that they have become a research field which strongly attracts the attention of the scientific community related to the nuclear physical and its applications. This motivation is driven, perhaps, primarily by the wide applications encountered in this type of reactions in different technological fields including the accelerator-driven systems (ADS) [1] for energy production, the radioactive waste treatment, and a lot of medical applications. On the other hand, it is well known that the spallation reactions are more efficient for the production of nuclei in a broad spectrum of excitation energies [2, 3] without significant modifications of their nucleonic composition. These hot nuclei do not exhibit strong collective excitations at their initial states as it happens in heavy systems collisions. Besides, due to the fact that only a nucleus takes part in the reaction, the experimental research is fully determined without ambiguity. Therefore, the spallation reactions constitute a way to study the properties and behavior of nuclei at high temperature. This kind of nuclear reaction has been treated by a

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mechanism which includes two stages: a sequence of binary collisions initiated by the incident nucleon, known as the intranuclear cascade (INC), followed by the decay, through several channels, of the excited compound nucleus. In general, the second step of almost all the spallation reaction models takes into account the evaporation-fission competition, and the evaporation has been treated by the approach due to Dostrosvsky, Fraenkel, and Friedlander [4] which can be considered an extension of the Weisskopf-Ewing model. Normally, this evaporation model in the framework of the spallation reaction might seem naive if one takes into consideration the inadequate description of the multiplicities of neutrons and protons among other observables. This aspect is not yet clearly explained. The question is whether this problem is related only to the fission or evaporation channels, or if it also includes the first phase which provides pre-equilibrium particles in a faster process than the evaporation. From the point of view of the neutron emission, one has to consider the different channels which contribute to the final multiplicities. In this regard, it is worth to mention that for a comprehensive understanding of the neutron production in spallation, it is necessary to clarify which part of the model is responsible for the description of the available data. That is so because the neutron production mechanism includes four possible contributions: the INC, the multi fragmentation, the evaporation from these excited fragments (residues), and the evaporation from the hot remnant nucleus after the INC. There are still opportunities for further developments, in particular for the prediction of residues and composite light charged particles. It has to be stressed that all models have their strength and weaknesses, and because of the complexity of the spallation reaction one should be careful with any kind of extrapolation. On the other hand, from the overall analysis of the results, some general conclusions about the physics of the models can be drawn. For instance, it has been found that, although the hypotheses inherent to INC models are not valid below 150 MeV, INC + deexcitation models give acceptable results. Therefore, they can be used in transport codes when evaluated libraries are not available and as long as one is not looking at collective or detailed structure effects. In some codes, a pre-equilibrium stage is added between the INC and deexcitacion stages. It can be concluded that there is not a clear advantage of having this additional stage or not, therefore, the scientific community must continue working to improve the evaporation-fission models. In several evaporation codes [5–7], γ-radiation is not included as a possible channel because the particle decay channels dominate above the particle-emission threshold. However, in the last deexcitation step of the evaporation cascade, gamma emission becomes competitive to particle decay for heavy compound nuclei. Normally, the emission of gamma is much less probable than the particle decay (about 105 times less favorable). Since the level density depends on the mass (heavier nuclei have a greater density levels), the number of levels between the ground state and the particle separation energy of a heavy nucleus can be as high as 105 or even exceed this value. If the excitation energy of the compound nucleus is slightly higher than its particle separation energy, it can decay only into the ground state or into the first excited states of the daughter nucleus (if the daughter nucleus is an even-even nucleus, then only the ground state is energetically accessible due to the pairing gap). In this situation, gamma emission and particle decay can become two competitive channels. This paper deals mainly with the study of the evaporation-fission phase of the spallation nuclear reaction searching for an improvement of the models included in the CRISP code [8]. In Section 2, we describe the basic features of the model and the principal assumptions incorporated for the calculations. Section 3 is devoted for the discussion of the results. In the last section, some final remarks and some ideas are pointed out for the further development of the model.

2 Model

We use the Colaboração Rio-Ilhéus-São Paulo (CRISP) [8] for the study of the deexcitation modes in spallation reaction. This model has being developed for the last decades, starting from the intranuclear cascade part, which is implemented in the MCMC code [9] and the evaporation fission process, implemented in the MCEF code [10, 11].

Both the intranuclear cascade and the evaporation-fission parts have been under continuous study and development. In the case of cascade, the main milestones are as follows:

1. The introduction of the multicollisional description of the cascade performed in [9].
2. The introduction of photon-induced reactions from [12].
3. The introduction of kaon photo-production from [13].
4. The inclusion of vector-meson photo-production developed in [14].

The main developments in the evaporation-fission part are as follows:

1. The introduction of evaporation of neutrons, protons, and alpha-particles in [11].
2. The interpretation of the non-saturation of the fissility observed experimentally from [15].
3. The simultaneous description of fission and spallation for different energies and target nuclei according to [16].
4. The introduction of the random-neck-rupture model to describe the formation of fission fragments developed in [17, 18].

5. The introduction of evaporation process in the fission fragments from [18].

In the development of the CRISP model, special care is taken to avoid the use of many free parameters, what is done through the introduction of more realistic descriptions of the physical processes. In this way, good and reliable calculations of cross sections for different channels are obtained over a wide range for nuclei with masses going from $A = 6$ up to $A = 240$. All these results are obtained with a single set of parameters.

The realistic description of the physical processes, including the multicollisional cascade [9] and a strict observation of the Pauli's principle [8, 16] is behind the initial success of the model, allowing some interesting features, as the elimination of some undesirable technical problems in the Monte Carlo calculations [19], and the inclusion of the Λ non-mesonic decay inside the nucleus with the inclusion of final state interactions [20]. With the most recent developments, the production of J/Ψ and ω vector-mesons in ultraperipheral collisions is being investigated and the super-asymmetric fission was identified [18]. Applications to the study of nuclear reactors were attempted.

Despite all efforts already made, there are many aspects in the CRISP model that deserves improvements. The study of fission of pre-actinides and their fragment mass distributions evidenced that the calculation results give fragment mass distributions shifted to lower values when compared to experimental data [21], while for heavier target nuclei the results are in good agreement with experiments. Moreover, the discrepancy with experiment depends on the incident particle (photons, protons, and deuteron were studied), being more evident for heavier incident particles. On the other hand, neutron multiplicity correctly reproduces the experimental data [22]. Thus, we are led to believe that some additional physical mechanism is needed in our model.

The main hypothesis we use in the present work is that the energy transfer to collective degrees of freedom, as nuclear oscillations and rotation. The main motivations to this hypothesis is the fact that evaporation of nucleons from fission fragments seems to be overestimated. In order to test this hypothesis, we add a parameter, $W(Z_i, A_i)$, which corresponds to the energy carried by collective degrees of freedom. Such collective energy will not contribute to evaporation or fission [23] as given by Weisskopf formulas.

With the introduction of collective degrees of freedom, we also hope to reduce the number of free parameters by introducing additional and more realistic descriptions of some aspects of the nuclear structure. The evaporation and fission channels regulate the decay of the compound nucleus. One of the important issues in the calculation is the inclusion of adequate level densities. In this work, we choose a new level density parametrization, which improves the employed physical models in the code, leading to a more direct evaluation of fission barriers, fission ratios, and mass distributions. This parametrization was adopted from the work of Bucurescu and Egidy [24], where the authors used the back-shifted Fermi gas (BSFG) [25, 26] and the model of constant temperature (CT) to obtain an adjustment at low excitation energies for more than 300 nuclei. These authors also proposed some simple formulas, based on experimental atomic mass isotopes tables [27] to describe the main features of the empirical data. Besides we take, as a basis, the data of more consistent physical models to evaluate the level densities, as such as the microscopic model performed by Demetriou and Goriely [28] based on deformed Hartree-Fock-BCS model. Moreover, this kind of microscopic model can be recommended as a reliable extrapolation to unknown nuclei as is the case of some nuclei produced in the intranuclear cascade.

In our calculations, the level density parameters were chosen according to the formulation by [24]. It means the state density of a nucleus with a given excitation energy U was obtained by Bethe on the basis of the Fermi-gas model as,

$$\rho(U) = \frac{\sqrt{\pi}}{12a^{1/4}U^{5/4}} \exp\left(2\sqrt{aU}\right), \tag{1}$$

where $U = E - E_1$, and E_1 is the energy back shift. The level density parameter a reads,

$$a = A\left[p_1 + p_2 S'(N,Z) + p_3 A\right], \tag{2}$$

with $p_1 = 0.127$, $p_2 = 4.98 \times 10^{-4}$, $p_3 = -8.95 \times 10^{-5}$ and $S'(N,Z) = S(N,Z) - \Delta$, where $S(N,Z) = M_{exp} - M_{LD}$ is the mass difference, being M_{exp} the experimental mass, and M_{LD} is the mass coming from the liquid drop model. The Δ value and the E_1 energy back shift are in Table 1. The Θ-function presented in Table 1 is

$$\begin{aligned}\Theta &= \frac{dS(N,Z)}{dA}, \\ &= [S(Z+1, N+1) - S(Z-1, N-1)]/4,\end{aligned}$$

Table 1 Δ value and the E_1 energy back shift from the BFSG of [24]

	Even Z-even N	Odd A	Odd Z-odd N
Δ	$0.5P_d$	0	$-0.5P_d$
E_1	$b_1 - 0.5P_d + b_4\Theta$	$b_2 - 0.5P_d + b_4\Theta$	$b_3 + 0.5P_d + b_4\Theta$

Here, $b_1 = -0.468$, $b_2 = -0.565$, $b_3 = -0.231$, and $b_4 = 0.438$. The functions Θ and P_d are defined in the text and P_d is called the deuteron pairing:

$$P_d = \frac{(-1)^{Z+1}}{4}[S_d(A+2, Z+1) - 2S_d(A, Z) + S_d(A-2, Z-1)],$$

where S_d is the deuteron separation energy.

For the mass formula, we used the corresponding to [29], which allows to take into account the shell effects. The used mass formula has a strong influence on the results obtained in the parametrization of the level densities. This is noteworthy, since it resorts to various factors used in the calculation such as the deuteron energy separation. These data are consistent with those available in ref. [29]. As it was already mentioned, the description of level density parameters based on the Fermi gas model offers a number of limitations to properly consider the shell effects and the influence of pairing correlations. This limitation in the description of the nuclei by this model does not generate a satisfactory treatment of the level densities at all excitation energies.

For fission, the absence of a dependence with microscopic variables in this model can lead to some wrong parameterizations to evaluate the level densities of the transition states. Moreover, the parameters of these phenomenological models are performed for nuclei equilibrium states. It means that they could not be used to describe deformation. These deformations correspond to extreme points on the fission path, where the nucleus suffers significant changes in its nuclear structure.

Then, it is necessary a method to describe properly the fission due to the complexity of interactions happening in this stage. The microscopic model extended Thomas-Fermi plus Strutinsky integral (ETFSI) complies well with that purpose [30, 31]. Therefore, we adopted a method based on ETFSI model to describe the fission barrier, which offers a more realistic approach to the estimation of fission barriers. This is an approximation of a high standard for the Hartree-Fock (HF) method which includes pairing correlations treated in the standard BCS approximation with blocking. In this way, we propose a new parametrization for the barriers, which is valid for $A \geq 200$. The form of the fission barrier under this approach is defined by a function of several terms with three regions of validity. The first one corresponds to the interval $200 \leq A \leq 220$ where the fission barrier formula reads:

$$B_f = -14.1871 - 21.8005Z + 27.4098A - 24.1243\frac{Z^2}{A}. \quad (3)$$

For $A \geq 221$, the fission barriers are calculated by:

$$B_f = 1241.88 + 1.56788Z - 15.7209A + 13.8798\frac{Z^2}{A} - 0.177254Z^2 + 0.0724926A^2 + 0.000752518Z^3 - 9.8343 * 10^{-5}A^3. \quad (4)$$

The case $A < 200$ is considered using the standard parameterizations included in the CRISP code:

$$B_f = f_0 E_S + C(A)\exp[-0.40(Z - Z_0)^2], \quad (5)$$

where $Z_0 = A/(1.98 + 0.015A^{2/3})$.

This parametrization is based on the work of Möller and Nix [32, 33]. Here, f_0 is function of the critical fissility and also another parameter that take into account the competition between nuclear and Coulomb forces, E_S defines the surface energy of the nucleus, and $C(A)$ is introduced to include the shell effects.

3 Results

We performed the calculation for the reactions p + ^{208}Pb and p + ^{197}Au at 1 GeV and 800 MeV, respectively. In order to evaluate the role of the INC, we analyzed the results of the simulations of the isotopic distribution corresponding to the remnant nuclei formed at the end of the intranuclear cascade. It can be used as an interesting tool for discussing the evolution of the systems at different critical points. In particular, to get some important aspects on the particle emission mechanism and the composition of the mass distribution at the end of the spallation process.

Mass distributions for gold and lead are showed in Fig. 1. The generated mass numbers spread out over a wide range of values. The difference between the highest and lowest mass can reach 48 nucleons, as it can be seen for ^{208}Pb.

As it was already discussed above, the second stage of the spallation reaction is well described through evaporation-fission models, which basic ingredients are the level densities and the fission barriers. Usually, the simulation of this stage is performed by means of Monte Carlo methods under the assumption of a statistical equilibrium arising from a previous intranuclear cascade, which generates hot compound systems. This final state of the INC constitutes the initial conditions for the statistical branch of the calculations. Some other approaches, namely dynamical ones, have been explored for the decay of these compound hot nuclei, but till now they are not included in a comprehensive treatment of the spallation nuclear reactions.

On the other hand, the traditional approaches do not seem to include all the physical aspects of the process. For instance, one can assume a more active role of collective degrees of freedom during the decay of excited nucleus because there is enough time for their evolution, as it can be the case of the shape coordinates. Therefore, with these ideas in mind, it is possible to consider some energy exchange between collective coordinates and the nucleonic (or internal) ones. The results of such coupling should be manifested in a decrease of the available energy for evaporation of neutrons. Notwithstanding of a satisfactory treatment

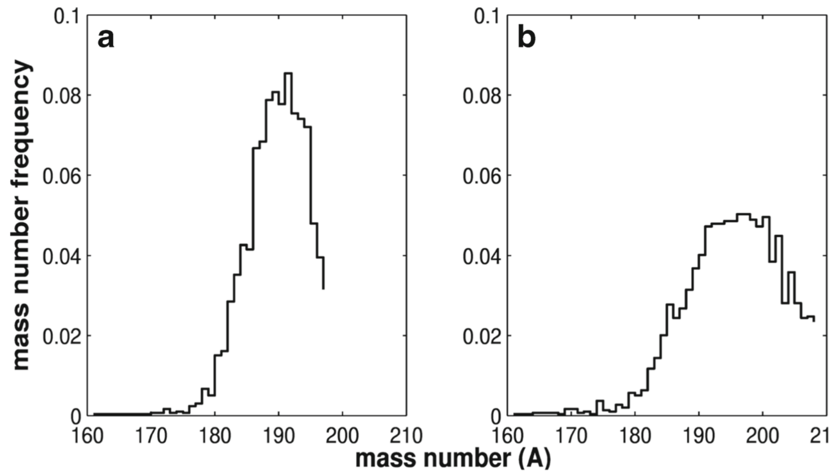

Fig. 1 Mass distribution at the end of the cascade for **a** p + ^{197}Au at 800 MeV and **b** p + ^{208}Pb at 1000 MeV

of the level densities and a best way of calculations of neutron binding energies, the consideration of the asymmetry in the distribution of the remnant nuclei after the cascade should be carefully considered within the energy balance in each step of the evaporation process.

The excitation energy of the nucleus after the evaporation of a particle can be written as:

$$E^*_{i+1} = E^*_i - B_i - \epsilon_i - V_i, \qquad (6)$$

where E^*_{i+1}, E^*_i, B_i, ϵ_i, and V_i describe the final and initial excitation energy of the nucleus, the separation energy, the kinetic energy in each step of the evaporation sequence, and the Coulomb potential barrier if the particle is charged ($V_i = 0$ in other case), respectively.

For the calculations, a modification of (6) is performed:

$$E^*_{i+1} = E^*_i - B_i - \epsilon_i - V_i - W(Z_i, A_i). \qquad (7)$$

In this equation, a phenomenological parameter, $W(Z_i, A_i)$, is included in order to take into account all the collective energy contributions which are not considered in (6). The arguments to incorporate such phenomenological energy correction in the calculations are based on the fact that,

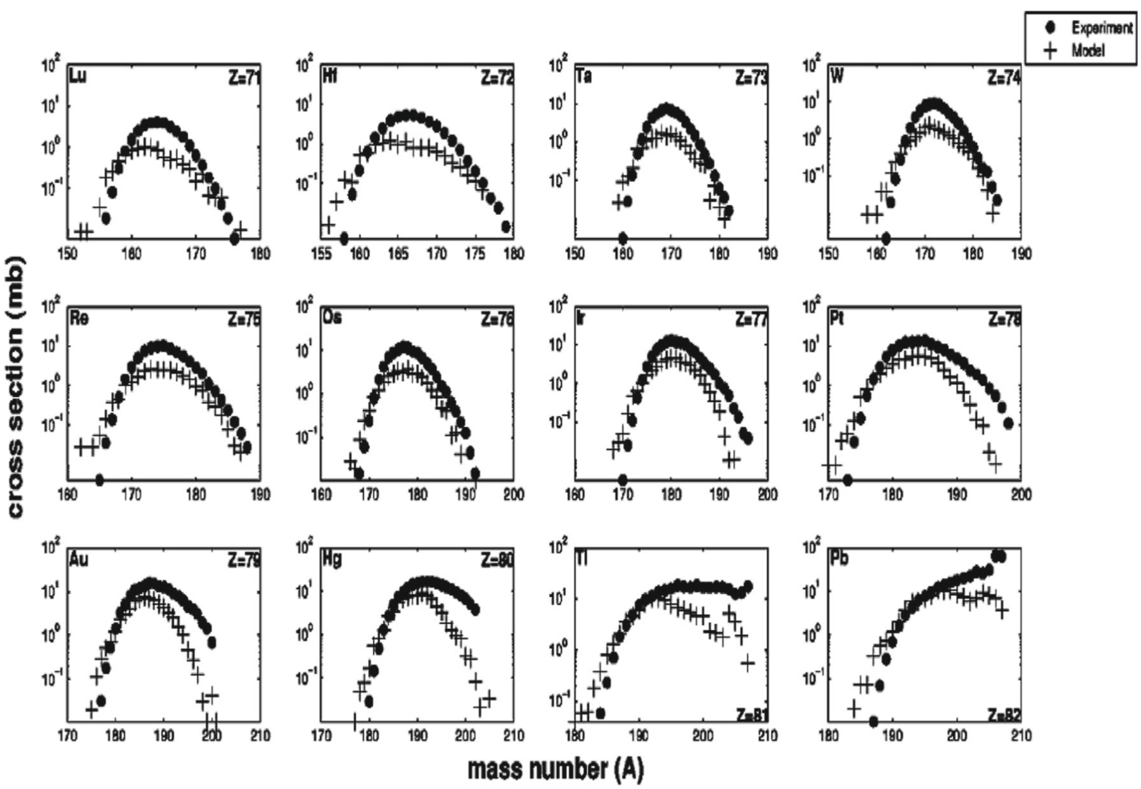

Fig. 2 Isotopic distribution for the reaction p+^{208}Pb at 1000 MeV, compared with experimental data from [34]

commonly, in almost all the statistical approaches for the decay of a fissioning system, the initial state is considered without taking into account the excitation of all the collective degrees of freedom of the system. On the other hand, in the evolution of the system to the final states, one has to incorporate the dynamical aspects in which a freely energy exchange between collectives and internal degrees of freedom occurs ([38]). It means that the combination of both effects, namely energy distributed among collective degree of freedoms at the beginning and, energy transfer from the internal to the collective degree of freedoms, diminish the available energy for the evaporation/fission process. Therefore, an estimation for the $W(Z, A)$ should include both effects. Typical values for the energy exchange between collectives and internal degree of freedoms are around 4 MeV, same as the point of view of the work of [39].

As a first step, we assumed W to be independent on nuclear mass and charge, and determined an averaged value of 4.50 MeV for this energy correction to perform the calculations for the two selected nuclei. The results for the isotopic distribution are shown in Figs. 2 and 3.

Figure 2 shows a rather well agreement with the experimental data for almost all the cases. Some discrepancy with the experiment is observed for $Z = 79$ in which the theoretical calculations underestimate the production of residual nuclei. The trend for the isotopes with atomic number close to the target shows a light disagreement with the experiment for the heavy residues, which could be an indication of the role of the cascade.

The results for the reaction p+^{197}Au are shown in Fig. 3. A satisfactory agreement with the experimental results is obtained. Only in the distribution for $Z = 78$, the calculations show a pocket around $A = 192$, which corresponds to the stable even-even nucleus ^{192}Pt.

In order to explore the role of the energy correction, we will remove its contribution to the calculations. The comparison for the same targets is shown in Figs. 4 and 5. Both figures show a clear deviation of the calculations from the experimental data along almost all the residues. From our point of view, this discrepancy indicates the need for including in a comprehensive way the dynamical aspects in the description of evaporation-fission decay modes. Finally, we remark that in all the analyzed cases, the numerical value of the energy correction was the same one.

The present calculations were performed considering that fission barrier does not depend on the excitation energy of the remnant nucleus. In order to evaluate the influence of this approach in the isotopic distribution at the end of the process, the ratio between Γ_f/Γ_n was calculated. This ratio describes the competition between fission and neutron

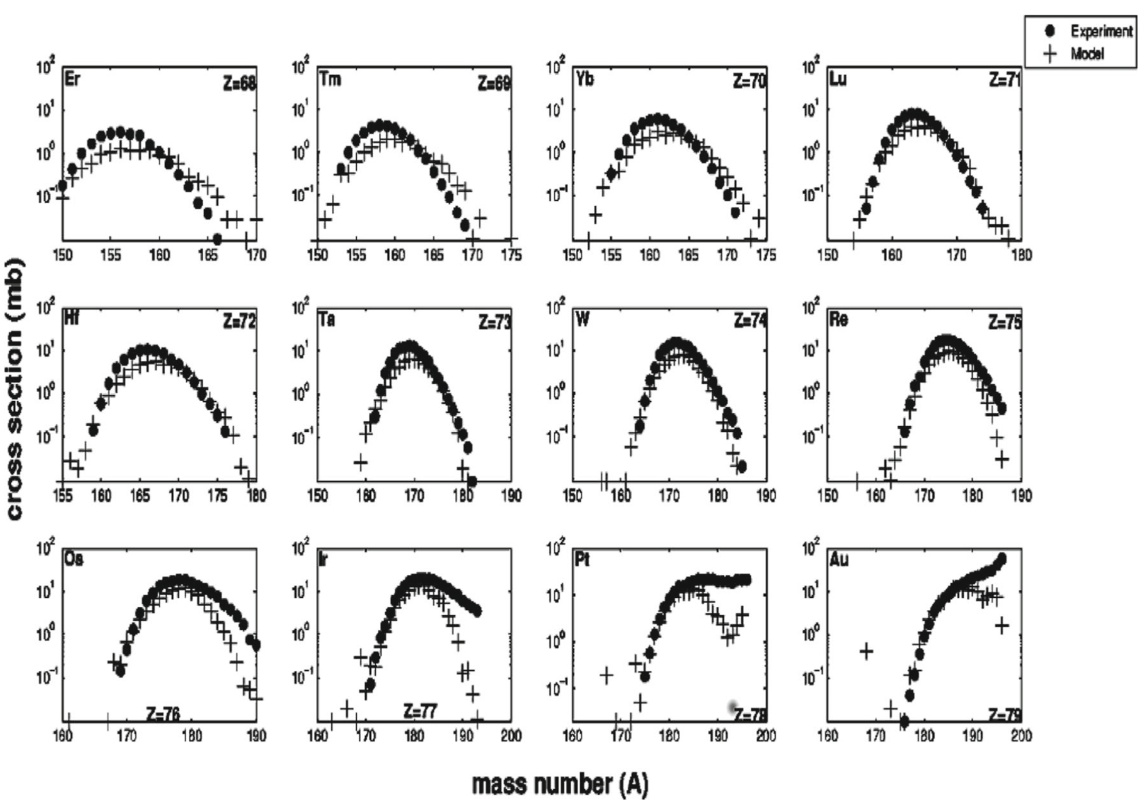

Fig. 3 Isotopic distribution for the reaction p+^{197}Au at 800 MeV, compared with experimental data from [35]

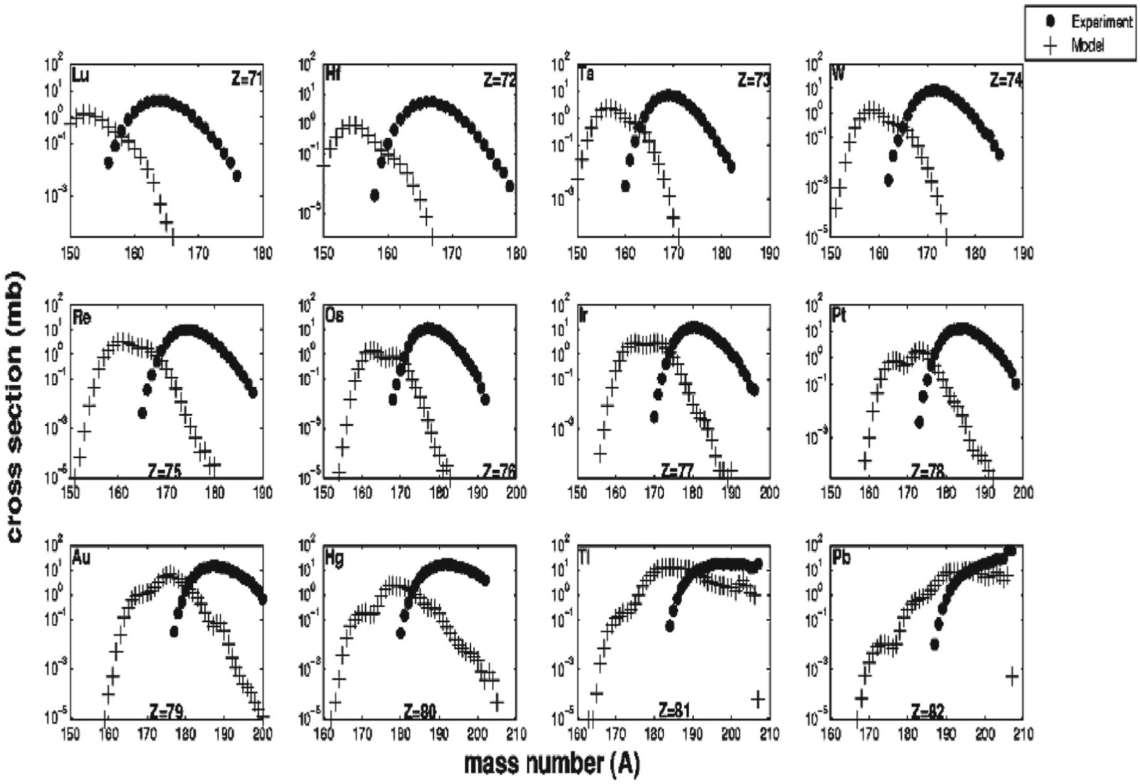

Fig. 4 Isotopic distribution for the reaction p+^{208}Pb at 1000 MeV without energy correction, compared with experimental data from [34]

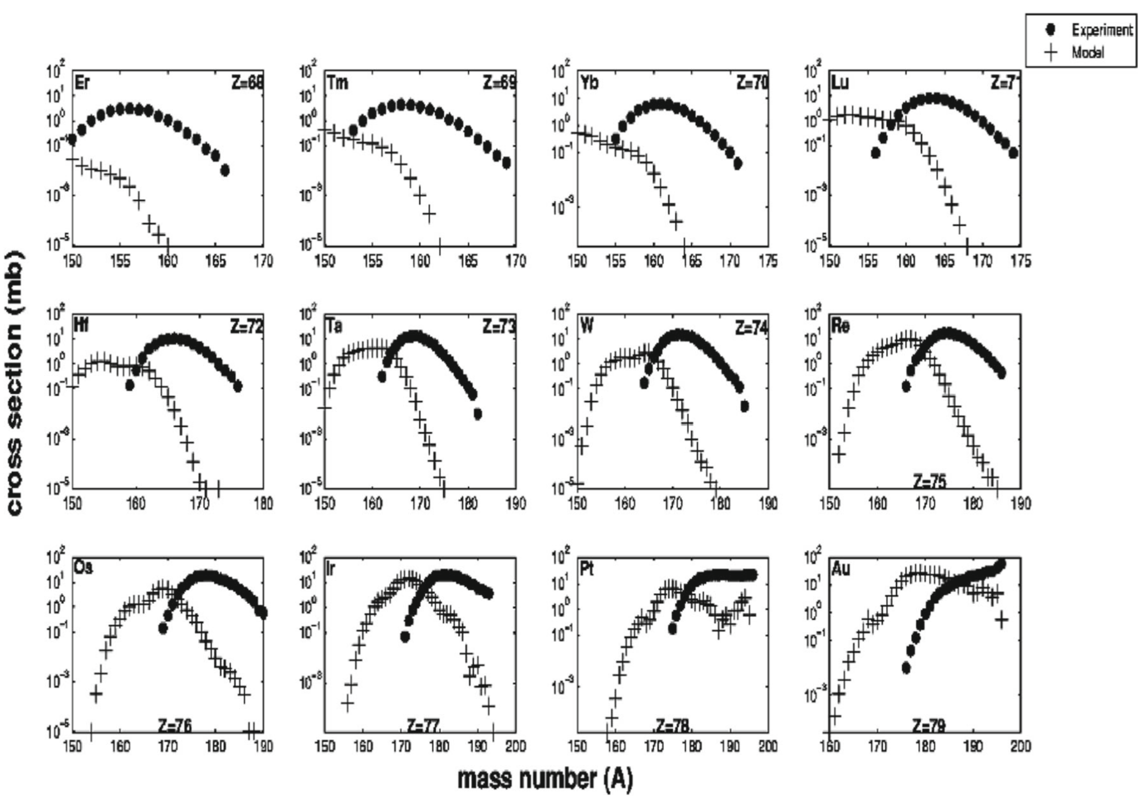

Fig. 5 Isotopic distribution for the reaction p+^{197}Au at 800 MeV without energy correction, compared with experimental data from [35]

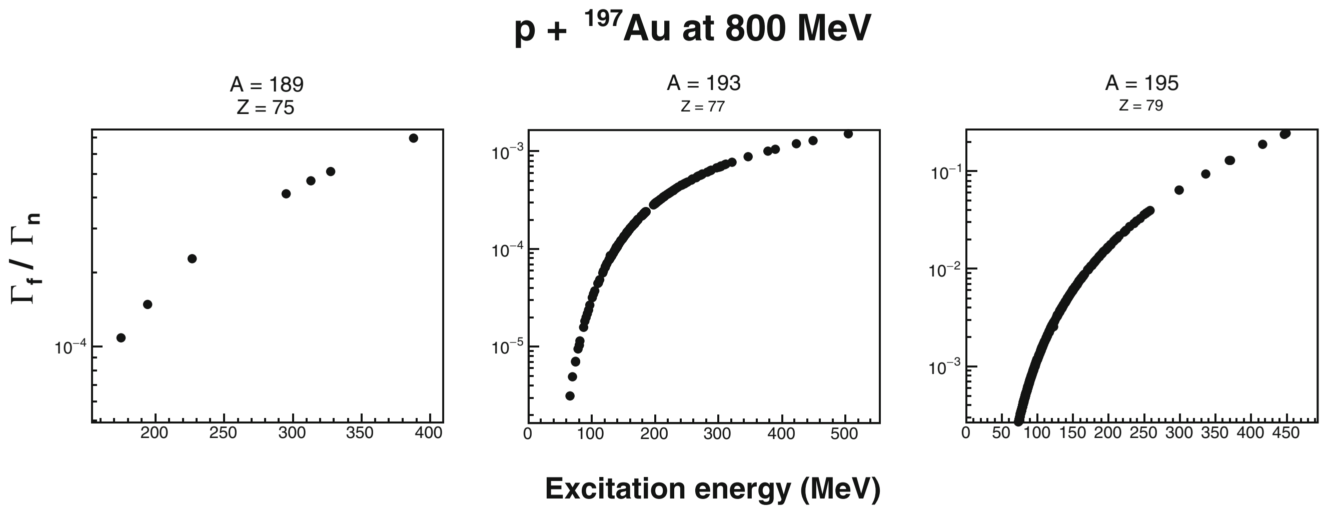

Fig. 6 Comparison of Γ_f/Γ_n for the reaction $p+^{197}$Au, in the statistical decay of three fissioning nuclei resulting of the intranuclear cascade (A = 189, 193, 195 and Z = 75, 77, 79) as function of the excitation energy of the fissioning nucleus

emission channels as function of the excitation energy of the fissioning nucleus.

Figure 6 shows the results of the calculations for reaction $p+^{197}$Au in three cases. In this example, the energy behavior of the ratio Γ_f/Γ_n for the statistical decay of three fissioning nuclei resulting of the intranuclear cascade (A = 189, 193, 195 and Z = 75, 77, 79) are compared. One can notice a growth of the ratio with the excitation energy as it is expected. The small absolute values of this magnitude are characteristics of competition fission-evaporation for these isotopes. On the other hand, it is also observed that the majority of events occurs at excitation energies between 100 and 300 MeV while the fission channel width shows values two to four times below of the orders corresponding to the neutron channel. In this way, it would justify the fact not having explicitly included a dependence between the height of the fission barrier and excitation energy. At this stage, it is worth to mention that at typical excitation energy reached after INC, the shell effects are vanished and, therefore, the two humped fission barrier can be described as a single one [41, 42] in the calculations considered in this work. However, the question of to take into account the temperature dependence of such single humped fission barrier is still open for the case of multiple fission channels, as well as the estimation of their contribution to the final states of the whole process.

4 Conclusions

In this paper, we present modifications to the model describing the evaporation-fission process in the CRISP code. The first modification was to incorporate a more advanced treatment for the statistical approach to the nuclear level densities, which reflects in a more consistent physical description for the excited nuclei.

Moreover, we discussed the improvements dealing with the calculation of the fission barrier. In this case, we used models that take into account microscopic approaches as such as shell correction and pairing effects. An advantage of the proposed improvements resides in the fact that by this way, the computation time in the Monte Carlo simulation was not increased. However, despite that these improvements are physically consistent reducing the number of free adjusted parameters, they alone do not ensure a good agreement with the experiment and therefore, it is necessary to examine to what extent should be taken into account the dynamic aspects of the process.

In a first attempt to evaluate this issue, we appeal to energy conservation and incorporate in a phenomenological way a correction energy term in every evaporation step. Our justification for this term is based on the fact that each step of the evaporation chain must be linked to different values of the deformation coordinates. This could result in the possibility of energy exchange between the collective and intrinsic degrees of freedom and therefore, a decrease of the available energy for evaporation. We believe that a comprehensive examination of the spallation reaction must take into account, besides the statistical description, the role of the nuclear dissipation as a necessary ingredient of the model. One possible way to perform such test is by means of the approach made by Fröebrich and Gontchar [36], which has been worked out in refs. [37, 40]. Studies along this line are in progress. The obtained results reproduce rather well the experimental data corresponding to the spallation parabola for reactions with Au and Pb [34, 35] induced by proton at energies corresponding to 800 and 1000 MeV, respectively.

Acknowledgments Part of this work was made in the frame of the academic cooperation agreement between UESC and InSTEC. O. Rodríguez and F. Guzmán would like to recognize the provisions of UESC for the conclusion of this work. F. Guzman would also like to thank the SBF (Sociedade Brasileira de Física) for the financial support of PLAF/SBF/CNPq in the frame of the *Catedra Itinerante* program. O. Tumbarell would like to thank the FAPESB (Fundação de Amparo à Pesquisa do Estado da Bahia) for the financial support. A.R. Samana thanks the financial support of AUXPE-FAPESB-3336/2014/Processo no: 23038.007210/2014-19 for the visit of F. Guzmán to the UESC-BA while performing this work. A. Deppman thanks the financial support of CNPq/305639/2010-2. D. A. Souza thanks the financial support of CAPES. And finally, the authors thank the financial support of Conselho Nacional de Desenvolvimento Cientfico e Tecnológico (CNPq).

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