

Monte Carlo Simulation of Spallation and Fission Fragment Distributions for ADS-Related Nuclear Reactions

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Abstract

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Preamble

Monte Carlo Simulation of Spallation and Fission Fragment Distributions for ADS-Related Nuclear Reactions

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Monte Carlo simulations with the CRISP code were conducted to study spallation and fission fragment distributions induced by intermediate- and high-energy protons and photons on actinide and pre-actinide nuclei. The model accounts for intranuclear cascade, pre-equilibrium, and evaporation-fission competition, enabling consistent treatment of both residues and fission products. Comparisons with experimental data show good agreement in mass and charge distributions, with minor deviations for light fragments. The results highlight the reliability of Monte Carlo approaches for predicting residual nuclei and fragment yields

under accelerator-driven system (ADS) conditions. This work provides nuclear data relevant to ADS design, safety, and transmutation analysis.

Keywords: Monte Carlo simulation; Spallation and fission fragments; Accelerator-driven system (ADS)

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1. Introduction

The accelerator driven system (ADS) is an innovative reactor being developed as a dedicated burner in a double strata fuel cycle to incinerate nuclear waste [1–4]. The ADS system consists of a subcritical assembly driven by an accelerator delivering a proton beam onto a target to produce neutrons via spallation reactions. The spallation target constitutes simultaneously the physical and functional interface between the accelerator and the subcritical reactor. For this reason, it is probably the most innovative component of the ADS, and its design is a key issue for ADS development. The performance of the reactor is characterized by the number of neutrons emitted per incident proton, the mean energy deposited in the target per neutron produced, the neutron spectrum, and the spallation product distribution [5].

The detailed design of spallation neutron sources or accelerator-driven systems requires reliable computational tools to optimize their performance in terms of useful neutron production and to properly assess specific problems likely to occur in such systems. Among these problems are the radioactivity induced by spallation reactions and the problem of shielding [6] the radiation due to energetic particles generated in the reaction, the changes in the chemical composition and radiation damage in target, window, or structure materials [7], and induced radiotoxicity within the target [8] due to the production of several nuclides.

Radiation damage can arise from gas production that causes embrittlement of structural materials and from atomic displacements (DPA) which fragilize the various components of the spallation source. Modifications of the chemical composition of these materials may result in problems of corrosion or alloy cohesion and modification of mechanical properties because of the appearance of compounds not existing initially in the materials.

It is important to emphasize that, at the present status in the development of ADS, it is still necessary to study the best technological options for variables such as the material used for the target and the energy of incident particles. For this reason, the most important aspect in the calculations of yields of nuclides is the confidence one has in the results obtained. More important than accuracy, at this moment, may be the reliability of the calculation method. One should consider methods which take into consideration the correct mechanisms of reactions as more important than those methods that have many free parameters to fit experimental data. These are of great importance to interpolate experimental data when they are available but are of restricted use when there is no

data.

This paper is organized as follows: Section 2 describes the main mechanisms for fragment production in nuclear reactions and gives a short description of the intranuclear cascade process, which is relevant for the description of residual nucleus formation. In Section 3, the evaporation mechanism is described. This is the most important mechanism for the study of spallation. In Section 4, the fission process is described, this decay channel being relevant for the formation of fission fragments. In Section 5, we present and discuss the results obtained with the CRISP code, and in Section 6, we show our conclusions.

2. Production of Nuclides in Nuclear Reactions

The production of nuclides is associated with the different mechanisms through which the nuclear reaction evolves. Depending on the target, the probe, and the energy, different mechanisms are more or less relevant. On the other hand, the distribution of fragments generated when the reaction finishes is strongly dependent on the channels open to the system and on the respective branching ratios.

As the reaction energy increases, more channels become available and the complexity of the reaction increases. This high complexity is the main reason to adopt Monte Carlo (MC) methods in calculations of nuclear reaction results. Indeed, the large number of particles, the large number of available channels, and the fact that the reaction may be understood as a Markovian sequence of steps are features that perfectly match the characteristics of Monte Carlo calculations. In this work, we adopt the MC method and use the implementation for nuclear reaction calculations developed in the CRISP code [12], which has already been used in ADS studies [13–15]. This implementation is described in some detail below.

It is well established that intermediate- and high-energy reactions follow a two-step mechanism, as proposed by Bohr. In the first one, usually called intranuclear cascade, the energy and momentum of the incident particle are distributed among a few nucleons through baryon-baryon reactions which are mainly elastic, but at sufficiently high energies, nucleonic degrees of freedom can be excited. This step finishes when there are no nucleons with kinetic energy that is high enough to escape from the nuclear binding. Thereafter, the collisions among nucleons lead only to the system thermalization.

The second step depends on the excitation of the residual nucleus formed at the end of the intranuclear cascade. If the nuclear excitation energy is $E/A \leq 3$ MeV, a statistical competition between evaporation and fission takes place. For heavy nuclei ($A > 230$), fission is the dominant channel, and in most cases, the reaction ends with the formation of two fragments [16]. For nuclei with $A < 230$, fission is much less probable, and in most cases, the nucleus evaporates until there is no energy for evaporation of any particle (neutrons, protons, and

alpha particles are the most frequent). Then a spallation product is formed which is characterized by its mass and atomic numbers.

If the nuclear excitation energy is $E \geq 3$ MeV/A, an entirely different process may occur, namely, nuclear multifragmentation [17–19]. This process is much faster than the evaporation/fission competition, and it is characterized by the simultaneous production of a large number of fragments with very different mass and atomic numbers. In the following, we give a brief description of each process mentioned above.

2.1 Intranuclear Cascade

We now describe the nuclear mechanisms relevant in nuclear reactions at intermediate and high energies, which have been implemented in the CRISP code. The first thing we have to deal with is the intranuclear cascade (IC). As the incident particle enters the nucleus region, IC is the first mechanism of the reaction triggered by the collision of the probe with one of the nucleons in inelastic scattering. This collision, called elementary collision, will always generate secondary particles, which can be two nucleons in the case of elastic scattering, mesons, or resonance states of the nucleon. These secondaries have energies relatively high compared to the other nucleons in the Fermi sea and occupy high-energy single-state levels in the system [20–23]. We call them cascade particles.

The secondary particles generated in an elementary collision will propagate inside the nucleus and may interact, increasing the number of cascade particles, or may reach the nuclear surface. In this case, if it has energy higher than the nuclear binding energy, it escapes from the nucleus; otherwise, it is reflected back and continues its propagation inside the nucleus. In this way, as the intranuclear cascade continues, the number of cascade particles increases. The decision to stop the intranuclear cascade and start the second step of the reaction is based on energetic criteria, namely, when there is no bound particle that is in an excited state or has kinetic energy greater than its binding energy [21, 22, 24].

There are several important aspects in the intranuclear cascade calculations with the CRISP code. The first one is that it is a multicollisional simulation of the cascade, with all nucleons moving simultaneously. The time-ordered sequence of elementary collisions considers the probability of interactions among all those particles based on their respective cross sections. This is an important difference with respect to other Monte Carlo codes where only one cascade particle is considered, while all others are kept frozen in their initial state.

The multicollisional approach is an important improvement in the simulation of intranuclear cascade with respect to the MC method used in well-known codes like those from Barashenkov et al. or Bertini et al. [25, 26], since dynamical aspects such as nuclear density modifications or the evolution of occupancy numbers during the cascade are naturally taken into account in the former method but not in the latter [24].

Another important aspect is the Pauli blocking mechanism, responsible for keeping track of possible violations of the Pauli Principle. With the multicollisional method, it is possible to adopt a very precise method for verifying the availability of single-particle states for the fermions generated in the elementary collisions, eliminating possible violations of this important principle of quantum mechanics. It is because we use this method for Pauli blocking that we can use the energetic criteria to decide to end the intranuclear cascade [27].

After the cascade finishes, there is no particle with energy high enough to escape from the nucleus. Then a sequence of elementary collisions will distribute the excitation energy remaining in the nucleus among all nucleons. This is called the thermalization process. The main characteristics of the nucleus do not change during this step, and the mass number, the atomic number, and the excitation energy at the end of the thermalization step are the same as at the end of the intranuclear cascade.

In a CRISP simulation, the reactions can be initiated by intermediate- and high-energy protons [21] or photons [21, 22, 27, 28]. It has been used for energies up to 3.5 GeV [24], where it was shown that the CRISP code can give good results for total photonuclear absorption cross sections from approximately 50 MeV, where the quasi-deuteron absorption mechanism is dominant, up to 3.5 GeV, where the so-called photon-hadronization mechanism is dominant, leading to a shadowing effect in the cross-section. Recently, the CRISP code has been used to study final state interactions for the nonmesonic weak decay of hypernucleus [29]. The results show that it can be used even for light nuclei, such as ^{12}C , and relatively low energies, as in the case of hypernucleus decay.

3. Evaporation

The thermalization is followed by the evaporation process, when nucleons or small clusters are emitted carrying part of the nuclear excitation energy. This process continues while there is enough energy in the nucleus to allow the evaporation of any particle. It consists of a sequence of emissions of particles by a nucleus, each one governed by the Weisskopf theory. Here the evaporation regime is governed by the relative probabilities of different particle emission channels [27, 28, 30].

These probabilities are obtained from the particle emission width, Γ , calculated according to the well-known Weisskopf evaporation model [31] in such a way that for proton emission we have:

$$\Gamma_p = \frac{2m_p}{\pi^2 \hbar^2} \int_0^{E_p} dE \sigma_{inv}(E) \rho(E_f)$$

and for alpha particle emission:

$$\Gamma_{\alpha} = \frac{2m_{\alpha}}{\pi^2 \hbar^2} \int_0^{E_{\alpha}} dE \sigma_{inv}(E) \rho(E_f)$$

where E corresponds to the nuclear excitation energy after emission of a particle of kind k , with $k = p, n, \alpha$, which is calculated as:

$$E_n = E - B_n, \quad E_p = E - B_p - V_p, \quad E_{\alpha} = E - B_{\alpha} - V_{\alpha}$$

where B_n , B_p , and B_{α} are the separation energies of neutrons, protons, and alpha particles, and V_p and V_{α} are the Coulomb potentials for protons and alpha particles, respectively.

These Coulomb potentials are given by the expressions:

$$V_p = \frac{Z_d e^2}{r_d}, \quad V_{\alpha} = \frac{2Z_d e^2}{r_d}$$

where B is the nuclear binding energy. The level density parameters a_k are calculated from Dostrovsky's empirical formulas [32]:

$$a_k = \alpha_k A + \beta_k$$

Table 1: Values for the relevant parameters in semiempirical mass formula.

Parameter	Value
a	-15.0175 ± 0.000013
a	15.5981 ± 0.000032
a_c	-7.09740 ± 0.00067
a_a	144.764 ± 0.0022

Table 2: Values for the relevant parameters in Dostrovsky's empirical formulas.

Parameter	Value
α	18.81302 ± 0.000097
β	1.30001 ± 0.000097
α	18.670295 ± 0.000097
β	4.23501 ± 0.000097
α	18.89 ± 0.19
β	24.82 ± 2.18

To evaluate the evaporation probability, one assumes that it is proportional to the corresponding width, that is, [15, 30]:

$$P_k = \frac{\Gamma_k}{\sum_i \Gamma_i}$$

While enough energy is available for particle evaporation, other emissions are processed. The evaporation phase ends when the excitation energy of the nucleus is smaller than all the separation energies B_n , B_p , and B_α . At this point, a nucleus that can be completely different from the initial one is formed. This nucleus is called a spallation product.

4. Fission

The CRISP code can also evaluate the fission probability [36]. Fission is a process that competes with evaporation in the sense that each nucleus in the evaporation sequence can undergo fission, forming two fragments with masses around one-half of the fissioning nucleus. This process can be easily included in the framework of the evaporation process by including the fission branching ratio, Γ_f , in the evaporation formula, in such a way that it now reads:

$$\Gamma_{total} = \Gamma_n + \Gamma_p + \Gamma_\alpha + \Gamma_f$$

The fission branching ratio is calculated according to the Bohr-Wheeler fission model [37], by:

$$\Gamma_f = \frac{1}{2\pi\rho(E)} \int_0^{E-B_f} dE' \rho(E')$$

For the mass formula, we considered the semiempirical one proposed in [33] for the nuclear binding energy so that the nuclear masses are calculated according to:

$$M(N, Z) = Nm_n + Zm_p - B(N, Z)/c^2$$

where N is the number of neutrons. The formula was fitted to the compilation of Audi et al. [34] by the method of least squares with the MINUIT package [35]. The values for each parameter corresponding to the best fit are shown in Table 1.

Table 3: Values for some of the relevant parameters in the multimode formula for the fission-fragment mass distributions.

Parameter	Low-energy
K (mb)	10.0 ± 2
A	135.0 ± 1
Γ	3.75 ± 2
K_1 (mb)	141.0 ± 2
A_1	5.0 ± 1
Γ_1	-

Table 4: Values of the relevant parameters found for the best agreement between simulated and experimental data for the fission of ^{208}Pb induced by 500 MeV protons. Errors indicated represent superior limit for uncertainties.

Parameter	Value
b_1	0.01 ± 0.05
b_2	121.68 ± 0.05
b_3	0.23 ± 0.05
b_4	125.66 ± 0.05
b_5	14.93 ± 0.05
b_6	3.97 ± 0.05
b_7	5.21 ± 0.05

The fission probability, P_f , at each step of the evaporation/fission competition process is calculated as:

$$P_f = \frac{\Gamma_f}{\Gamma_n + \Gamma_p + \Gamma_\alpha + \Gamma_f}$$

Whenever the fission channel is chosen, two fragments are formed [39, 40], the heaviest one having mass and atomic numbers, A_H and Z_H , respectively, is sorted according to a probability distribution given by the statistical scission model (SSM) from Brosa et al. [41]. The lighter fragment has mass and atomic numbers given respectively by:

$$A_L = A_f - A_H, \quad Z_L = Z_f - Z_H$$

The SSM accounts for the collective effects of nuclear deformation during fission through a liquid-drop model and includes single-particle effects through microscopic shell-model corrections. The microscopic corrections create valleys in the space of elongation and mass number, each valley corresponding to one different fission mode.

Fission cross section results from the incoherent sum of the contributions of each channel, $\sigma_i(A, Z)$, which are usually written in the form:

$$\sigma(A, Z) = \sum_i K_i \exp \left[-\frac{(A - A_i)^2}{2\Gamma_i^2} \right] \exp \left[-\frac{(Z - Z_i(A))^2}{2\Gamma_Z^2} \right]$$

where the summation runs over the asymmetric modes. The parameters for the symmetric mode are K_S , A_S , and Γ_S , while $K_H(L)$, $A_H(L)$, and $\Gamma_H(L)$ are the parameters for the heavy (light) fragment produced in the asymmetric mode i . For the atomic number distribution, the parametrization used is:

$$Z_i(A) = \mu_1 + \mu_2 A, \quad \text{for the most probable atomic number of the fragment}$$

$$\Gamma_Z = \nu_1 + \nu_2 A, \quad \text{for the width of the atomic number distribution}$$

where μ_1 , μ_2 , ν_1 , and ν_2 are fitting parameters.

It is important to stress that the evaporation process has as input the distribution of nuclei obtained at the end of the intranuclear cascade; therefore, the fissioning nucleus may be different from the cascade residual nucleus because some neutrons, protons, and/or alpha particles are allowed to evaporate before fission takes place. The effects of mass and energy distribution of the fissioning nucleus have been discussed elsewhere [40].

It is important to notice that all the parameters used in the CRISP code are fitted to a large number of experimental data for many different nuclei in different energy ranges. Also, the probes can be as different as protons, tagged photons, Bremsstrahlung photons, or electrons. In this way, the method is reliable because it is not an ad hoc fitting to a specific nucleus, reaction, or energy.

5. Results and Discussions

We used the CRISP code to calculate cross sections for the formation of fragments in nuclear reactions. As explained above, the main sources of fragments in the reactions studied here are spallation and fission processes, and in this section, we show the cross sections for fission fragments and for spallation products.

5.1 Fission Reactions

The relevant parameters in Dostrovsky's empirical formulas for the evaporation process are shown in Table 2. This set of parameters was used for all reactions studied in this work. The parameters for fission fragment calculations are shown

in Table 3. For ^{241}Am , ^{237}Np , and ^{238}U cases, the values for width and position of the fission modes were taken from a low-energy systematics [9]. The relative intensities of each fission mode for ^{241}Am and ^{237}Np were considered fixed, with their values given in Table 3. In the case of ^{238}U , the relative intensities were calculated according to Gaussian expressions depending on the fissioning system mass numbers.

There is not any systematic study of the multimodal parameters in the mass region of Pb. To obtain those parameters, we used a sigmoid fit for K_S and Gaussian fits for K_1 and K_2 , all depending on the mass of the fissioning system. For heavy fragment distributions, the peaks for the asymmetric modes are obtained by:

$$A_1 = b_1 A_f + b_2, \quad A_2 = b_3 A_f + b_4$$

while the width for each mode is assumed to be constant:

$$\Gamma_S = b_5, \quad \Gamma_1 = b_6, \quad \Gamma_2 = b_7$$

The values for each of these parameters corresponding to the best fit are presented in Table 4.

In Figure 1, we show results obtained for proton-induced fission. We observe that the results obtained with the CRISP code give a good description of the experimental data, although the data are spread over a large range.

[Figure 1: see original paper]: Cross section of fragments produced in the fission of ^{241}Am (a) and ^{237}Np (b) as a function of fragment mass induced by 660 MeV protons. Values from the systematic study of [9] for the parameters corresponding to the three fission modes considered in this work (—) were used. Open symbols () are the experimental data [10].

Since CRISP can also be used for photon-induced reactions, we calculated the mass distributions of fragments for fission induced in ^{238}U by Bremsstrahlung photons with endpoint energies at 50 MeV and 3500 MeV. The results are presented in Figure 2. Also in this case, the data do not show good resolution, but we can observe that the results with CRISP give a good general description of the experimental data.

[Figure 2: see original paper]: Cross sections for fragments produced in the fission of ^{238}U induced by bremsstrahlung of 50 MeV (a) and 3500 MeV (b) endpoint energies as a function of fragment mass. Open circles () are experimental results [11].

A more precise experiment was performed for proton-induced fission on ^{208}Pb . In this case, the experimental results are very precise. In Figure 3, we show the experimental data compared to the calculation results. We observe a very good agreement between experiment and calculation with the CRISP code, especially

regarding the overall form of the distribution. Such agreement is better attested when the result is normalized to the data.

For all results shown in Figures 1, 2, and 3, the evaporation of fragments was performed. This was achieved by using the already presented expression to obtain the mass and atomic numbers of heavy and light fragments. As a first approximation, the calculated excitation energy of the fissioning system was divided between the fragments, which continued the evaporation process until the stopping criterion was reached again, as explained in Section 3.

5.2 Spallation Reactions

Nuclei with mass number $A < 220$ present low fission probabilities. In these cases, the most probable reaction channel is spallation, when the evaporation continues until the residual nucleus is cold, without fissioning. The nucleus at the end of the evaporation is then called a spallation product.

With the CRISP code, using the parameters for evaporation and fission competition as described above, we can also calculate spallation product distributions. In Figure 4, we show our results and compare them to experimental data.

In general, we observe a fair agreement with data, since the well-known spallation parabola as calculated by us shows position and width in good agreement with experimental results. For the most probable products, also the absolute cross sections are in good agreement with data, although for some spallation products the agreement is not good. Similar behavior occurs for the case of spallation on ^{197}Au , shown in Figure 5.

In fact, these results show that it is extremely difficult to obtain good results simultaneously for several different reactions on a wide range of target mass numbers and for quite different energies. The main difficulty comes from the fact that, at the end of the intranuclear cascade, we have in general excited residual nuclei that may be far from the line of stability. Since most of the nuclear models and their parameters are determined for cold stable nuclei, we do not have precise information on the structure of all nuclei that are formed during nuclear reactions at intermediate and high energies. Improvements on mass formula and on shell effects can lead to better agreement between calculations and experiments.

6. Conclusion

In this work, we addressed the problem of fragment production in nuclear reactions. This is a relevant issue in the development of ADS, as it is directly related to the study of damage induced in materials used in ADS. We discussed the importance of reliability of calculation and showed that the model implemented in the CRISP code is developed with a reduced number of free parameters and with care in reproducing accurately the physical processes that occur during the nuclear reaction. The parameters appearing in the mass formula were obtained

by fitting to nuclear mass data. The parameters for evaporation and fission were fitted simultaneously to several results for fission and spallation cross sections on several nuclei and with different probes at many different energies. For these reasons, the code can be used to calculate several observables in different reactions induced by probes like photons, electrons, protons, and neutrons in a large energy range on nuclei with mass from $A = 12$ to $A = 240$.

We used the CRISP code for calculating the production of nuclides in nuclear reactions induced by intermediate- or high-energy probes. Proton and photon reactions on actinide and pre-actinide nuclei were considered. The most important mechanisms of fragment production, spallation, and fission were studied in detail. We show that the results are in qualitatively good agreement with available experimental data.

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