Achievements and deficiencies of nuclear models used for the design of spallation sources

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Abstract

High-energy transport codes are used in a large number of domains as spallation neutron sources or ADS design, space or medical applications. In these codes, nuclear models describing spallation reactions generate the necessary elementary cross-sections and characteristics of all the reaction products. During the last years, new high-quality experimental data have been collected leading to the development of more reliable spallation models, as the INCL4-ABLA combination of intranuclear cascade and de-excitation models, now available in MCNPX. However, remaining deficiencies have been identified, which can be of important consequences for applications, for instance in the prediction of damage. They generally originate from not well understood physics mechanisms. To solve these problems more constraining experiments are now being carried out, which allow a deeper understanding of the reaction mechanisms, in particular for the de-excitation stage. Recent improvements of INCL4 are presented. Impact for applications is discussed.

KEYWORDS: Spallation, Nuclear reaction models, High-energy transport codes, Accelerator-driven systems

1. Introduction

The recent interest for spallation neutron sources and Accelerator-Driven Systems (ADS) for nuclear waste transmutation has raised the need for reliable high-energy transport codes. These codes are used, for example, to calculate the neutron flux produced by the spallation target, the radioactivity induced by the spallation residues or the damages in the window and structure materials surrounding the target due to produced particles. All these quantities are mainly depending on the high-energy reactions of the incoming proton beam in the spallation target. In transport codes, for reactions above 150 MeV (sometimes 20 MeV), elementary cross-sections and characteristics of all the reaction products are calculated by nuclear models, contrary to low energies at which data libraries are used. To provide reliable predictions it is therefore necessary to have models describing correctly all the features of spallation reactions, validated on appropriate experimental data. Generally, spallation is modelled as a two step mechanism, a fast stage of nucleon-nucleon collisions, called intranuclear cascade (INC), leading to an excited nucleus, followed by a slower de-excitation stage of particle evaporation, with a possible competition with fission for heavy systems.

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During the last years, new high-quality experimental data have been collected, leading to a better understanding of the spallation reaction mechanism and allowing the testing of the currently used high-energy models. A large part of this work has been done in the framework of the HINDAS European FP5 program [1]. Also an important effort has been devoted to the development of more reliable spallation models, as the INCL4 [2] - ABLA [3] combination of intranuclear cascade and de-excitation models in Europe or CEM2k in USA [4], both now available in MCNPX [5]. In [1] a comprehensive comparison between the predictions of mainly INCL4-ABLA and Bertini-Dresner [6, 7] (default option of MCNPX) and the whole set of available experimental data has been carried out, with always the same set of (default) parameters in the models. Other comparisons [8] have been performed with other INC or de-excitation models. Although some important differences between the predictions of the models are found, some general trends can be observed.

As a result of these works, it is now possible to draw some conclusions on the degree of reliability of the predictions of quantities relevant for spallation sources. In this paper, we first present, in section 2, the quantities that can be assessed with a rather good confidence, sometimes provided that the right choice of nuclear models inside MCNPX is done. In section 3, we identify domains in which the predictions of the transport codes are expected not to be very reliable since persisting discrepancies in the comparison between the available models and basic experimental data have been found. Actually, it is not always easy to determine which part of the model (INC or de-excitation stage) is responsible for the deficiency. To answer this question, more constraining experiments are now being carried out, which should allow a deeper understanding of the reaction mechanisms. Section 4 presents some preliminary results, which provides clue for future improvements of de-excitation models, and recent progresses made in the development of INCL4 and their potential impact for applications.

2. Achievements of the high-energy transport codes

2.1 Neutron production

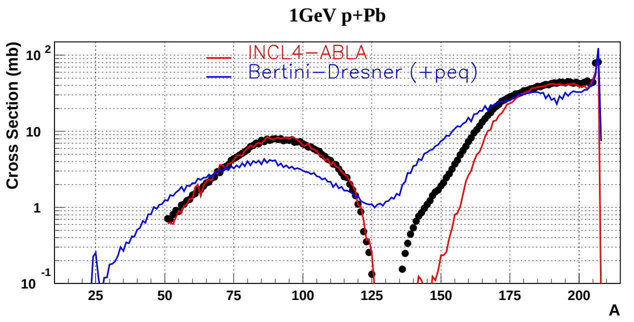
As regards neutron production, which is of primary importance for neutron source design, the situation is rather clear. Thanks to the existing complete and coherent set of experimental data on double-differential cross-sections and multiplicities on both thin [9] and thick targets [10], it can now be stated that total neutron production in an individual spallation reaction as well as in a realistic ADS target can be predicted with a precision of 10-15%, whatever the combination of intranuclear cascade and de-excitation models used in MCNPX. This may be due to the fact that, neutron production being of major importance for applications and the amount of data being rather large, most models have been first adjusted to reproduce this quantity. The precision of the model predictions corresponds also roughly to the uncertainties of the experimental data, meaning that it cannot be much improved. General trends of energy, angular or (thick) target geometry dependence are also well understood, although, locally, discrepancies could reach several tens of percent [8, 11].

2.2 Spallation residues

The precise knowledge of spallation residues is important for the assessment of radioactivity and damage in a spallation target. The use of the reverse-kinematics technique at the Fragment

Separator (FRS) in GSI has brought an extensive set of high-quality data and lead to the measurements of thousands of fully identified isotopes, in particular in p+Pb reactions at 1 and 0.5 GeV [12, 13]. These data showed that the INCL4-ABLA combination of models generally gives a satisfying agreement with the isotopic production cross-sections of evaporation residues not too far from the target nucleus and in the region of fission. On the contrary Bertini-Dresner was found not to predict correctly the fission fragment production and, generally, the isotopic distributions, except for isotopes very close to the target nucleus. The comparison of the mass distribution of p+Pb at 1 GeV measured at GSI with both models is shown in Fig. 1. Excitation functions measured by gamma- or mass-spectrometry in direct-kinematics experiments on Pb and Bi targets showed that these conclusions are generally valid in a wide range of incident energies [14, 15].

Figure 1: Comparison of INCL4-ABLA and Bertini-Dresner predictions with the production cross-sections (mb) as a function of the residue masses measured at GSI with the reverse kinematics technique for the p+Pb system [12].



2.2.1 Activity of a spallation target

The global activity of a spallation target is the sum of a large number of individual isotope contributions. Actually, as it was shown in [16], the main contributors are isotopes very close to the target nucleus. These isotopes are produced in very peripheral reactions in which only few cascade nucleons are emitted and little excitation energy is deposited. Generally, all models well predict their production cross-sections, the differences appearing for more inelastic collisions. This is illustrated in Fig. 2 which shows the production cross-sections of some isotopes in Pb and Bi targets as a function of the proton incident energy, measured by [15]. These isotopes appear to be the main contributors to the activity of a Pb-Bi target. Both INCL4-ABLA and Bertini-Dresner well reproduce the data. Consequently and as shown in [17], the total activity of a spallation target does not depend on the model used in the transport code. This has been confirmed by [8] for other choices of models, in particular CEM2k. Furthermore, thanks to the validation on the elementary data, it can be stated that predictions can be relied on within a few tens of percent.

2.2.2 Volatile fission products

Although fission products contribute only little to the total activity, they are important in the case of a liquid metal target. Indeed, some of the fission products are volatile gases, krypton, iodine and xenon, of which some isotopes are radioactive and can be a concern for radioprotection in case of a containment failure. It is therefore important to assess reliably their production rates.

Figure 2: Production cross-sections (mb) of a few isotopes measured in p+Pb or p+Bi by γ-spectroscopy [15] as a function of the incident energy, compared to the predictions of Bertini-Dresner (blue) and INCL4 - ABLA (red).

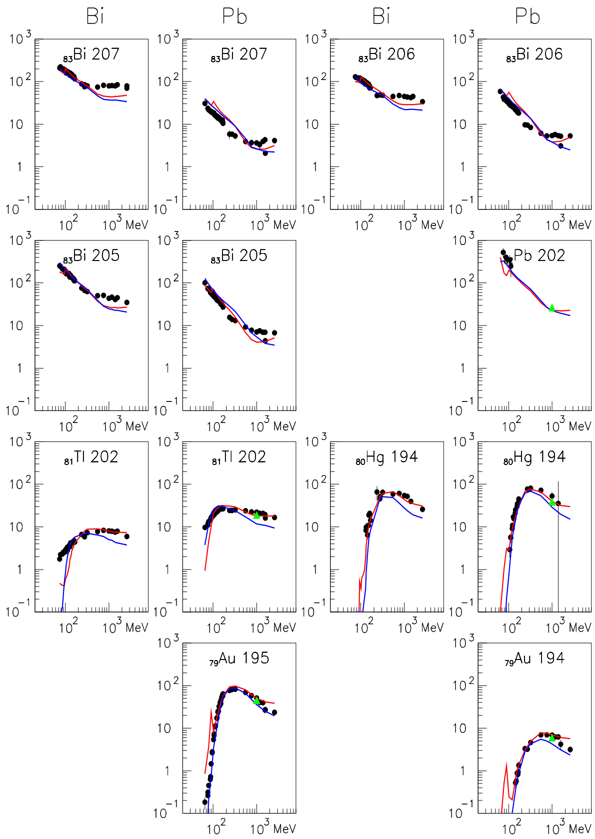
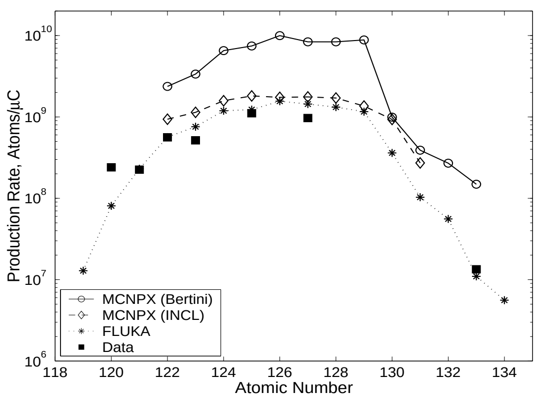


Figure 3: Production rates of Xenon isotopes from a thick Pb-Bi target, bombarded with 1400 MeV protons, measured at ISOLDE, compared with different calculations: FLUKA, MCNPX with either Bertini-Dresner or INCL4-ABLA models. From Zanini et al. [18].



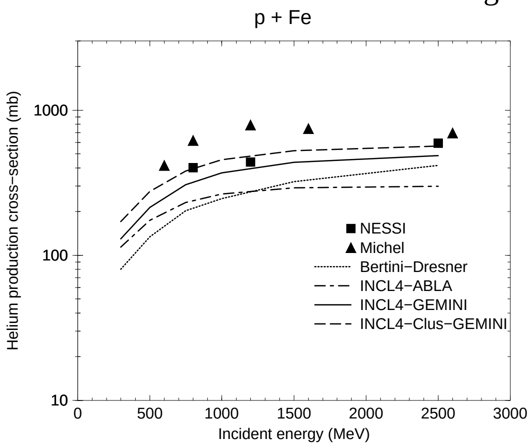
In the case of fission fragments, the differences between models are significant. As seen in Fig. 1, Bertini-Dresner is totally unable to correctly predict fission fragment distributions while INCL4-ABLA gives very good predictions. This means that for any prediction involving fission products, calculations done with Bertini-Dresner (which is the default option in MCNPX) should not be relied on. On the contrary, predictions of INCL4-ABLA can be trusted for targets with Pb or Bi (unfortunately the situation is not as clear for W or Ta). An experimental check of this has recently been brought by an experiment performed at ISOLDE [18] at CERN on a thick Pb-Bi target bombarded by 1.4 GeV protons, in which the production rates of Xenon isotopes have been measured. As shown in Fig. 3, Bertini-Dresner in MCNPX overpredicts the production rates sometimes by a factor larger than 4. INCL4-ABLA, but also FLUKA [19], give a reasonable agreement with the data.

3. Deficiencies

3.1 Gas production

A correct prediction of the production yields of hydrogen and helium isotopes is important for damage assessment in solid spallation targets, structure materials and window separating the accelerator from the target. Helium production, in particular, is expected to lead to swelling and consequently embrittlement in the window. Tritium production can also be important for radioprotection issues.

Figure 4: Helium production measured by NESSI [20, 22] and Michel et al. [14] compared to the predictions of Bertini-Dresner and INCL4 coupled either to ABLA or GEMINI. The dashed curve is obtained with the version of INCL4 allowing helium cluster formation.



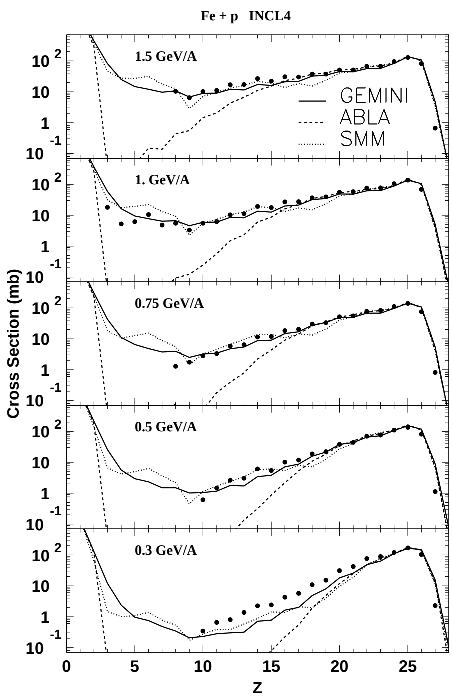
Comparisons of data [14, 20-22] obtained with different measurement techniques with codes have revealed severe deficiencies in most of the currently used models. This is illustrated in Fig. 4, for the production of helium in iron. It can be observed that both Bertini-Dresner (dotted line) and INCL4-ABLA (dashed-dotted line) systematically underpredict the experimental data. It can be also noticed that the replacing of ABLA by another de-excitation model, GEMINI [23], improves largely the situation. This will be discussed more precisely in Section 4. Comparisons of hydrogen and helium production for different target and energies have been done in [24] with different models, including FLUKA calculations, and led to the same conclusions. Since GEMINI is not available in any transport code used for applications, it can be stated that, for the moment, gas production cannot be reliably predicted.

3.2 Spallation residues

3.2.1 Light evaporation residues

As it can be already seen in Fig. 1, a systematic misprediction of light evaporation residues has been observed with INCL4-ABLA and other standard models [2, 4]. This is even more pronounced in light systems as iron, as shown in Fig. 5 (dashed line), which shows the charge distribution for iron measured by [25] at GSI at different energies. This means that production rates of isotopes far from the target nucleus cannot be predicted with a good precision. This can be important for the assessment of impurity yields in a given material (although the major ones are well calculated).

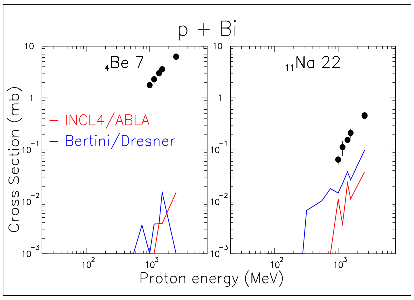
Figure 5: Charge distributions in p+Fe at five energies [25], compared with the predictions of the INCL4 model coupled with the de-excitation codes ABLA, GEMINI or SMM.



3.2.2 Intermediate mass fragments

The production of intermediate-mass residual nuclei is important since some nuclei, such as ^{7}Be or ^{10}Be, have half-lives worrying for radioprotection issues, as maintenance operation or final disposal. Production cross-sections has been measured on a wide energy range [15] and found generally underpredicted by orders of magnitude by the nuclear models used in transport codes. Fig. 6 shows the example of ^{7}Be and ^{22}Na from a Bi target compared with INCL4-ABLA and Bertini-Dresner. This can be ascribed to a production mechanism not yet well understood.

Figure 6: Production cross-sections (mb) of ^{7}Be and ^{22}Na isotopes measured in p+Bi [15] as a function of the incident energy compared to the predictions of Bertini-Dresner (blue) and INCL4 - ABLA (red).



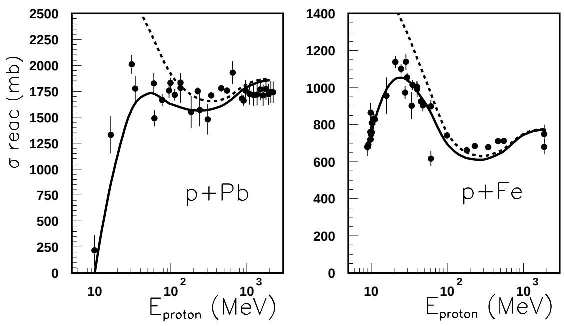
4. Recent results that could help solving the identified deficiencies

4.1 Intra-nuclear cascade model

In the description of spallation reactions, the INC stage determines the initial conditions for the de-excitation process, that are the charge, mass, excitation energy and angular momentum of the excited nucleus. It is therefore important to have the best possible INC model to be sure that the discrepancies with the data are due to the de-excitation stage and not, for instance to a wrong excitation energy of the remnant nucleus. This can be ensured only if all the characteristics of all the particles emitted during the cascade are correctly predicted.

Several deficiencies have been identified in INCL4. One of them was the overprediction of pion production compared to available experimental data. This has recently been cured by adding a potential for pions in the model [26]. This, together with the inclusion of an energy and isospin dependence of the nucleon potential, improves the prediction of particle spectra, including pions, and leads to slightly larger excitation energies.

Figure 7: Total reaction cross-sections of p+Pb and Fe as a function of energy calculated with the last version of INCL4 [29] with (solid line) or without (dashed line) Coulomb repulsion.



Another deficiency of INCL4, and all other INC models, concerns the emission of energetic composite particles observed experimentally and not explainable by any de-excitation process. A mechanism of surface clusterization has been added in INCL4 [27] to solve this problem. It allows for instance to reproduce the general trends of particle spectra measured by [28]. This has a non negligible impact on helium production, as can be seen in Fig. 4, where the dashed curve is a calculation with INCL4-GEMINI with this clusterization mechanism. The enhancement of the total helium production reaches 30% at 300 MeV. For tritium, the relative effect is even larger, since it is little produced in evaporation, the enhancement being of the order of a factor 2.5 in Pb at 1 GeV. This shows the importance of correctly treating composite particle emission in INC models. As the production ratios between the different types of particles are not yet satisfactory, further work is in progress to refine the details of the clusterization model.

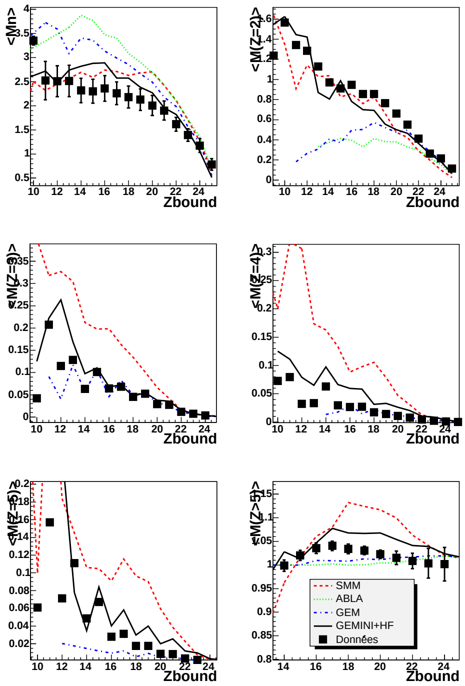
In a transport calculation a large part of the interactions occurs at energies much lower than the incident one. It is therefore important to ensure that the models can be used safely down to the energy at which libraries will take over, i.e. 150 MeV for a certain number of isotopes but 20 MeV for the other ones, although INC models are in principle out of their range of validity at these energies. The first requirement is to have a correct total reaction cross-section at all energies. This was not the case in INCL4 [2]. A forced renormalisation was imposed below 100 MeV, which had spurious effects as the kick observed in Fig. 2 around 100 MeV in the excitation functions of isotope production. Recent improvements of the momentum distribution of nucleons near the surface and of the Coulomb repulsion have had a drastic effect on the total reaction cross-section at low energies. As shown in Fig. 8, INCL4 is now in perfect agreement with available data.

4.1 De-excitation model

As already mentioned above, the GEMINI model, mostly used in heavy-ion physics, well predicts the production of hydrogen and helium contrary to the de-excitation models available in transport codes. It should be added that GEMINI is also the only one capable of reproducing the shape of the evaporation particle spectra [20, 22]. The success of this model may be due to the use of the Hauser-Feshbach formalism (which takes the angular momentum into account) rather than Weisskopf-Ewing, as most of the other ones, for evaporation. GEMINI coupled to INCL4 also reproduces very well the fragment distribution in iron measured by [25], as it can be seen in Fig. 5, in particular for the lightest nuclei. In GEMINI, these nuclei, which cannot be accounted for by standard evaporation models, are produced through an asymmetric fission mode, handled by the transition state formalism, competing with classical evaporation. However, another explanation of the data could be the onset of multifragmentation as treated in the Statistical Multifragmentation Model, SMM [30]. It can be seen in Fig. 5 that both calculations agree quite well with the whole data. Actually from these data it is not possible to be more conclusive and more constraining data are needed.

The SPALADIN experiment, performed in GSI, on p+Fe at 1 GeV [31], in which residues, neutrons and light charged particles have recently been measured simultaneously, brings a new insight of the situation. It has been possible to measure the correlation between a variable related to the excitation energy at the end of the cascade stage, Z_{Bound}, and the multiplicity of the different types of evaporated particles. Z_{Bound} is the sum of the charges of fragments with $Z\geq2$ in each event. Fig. 8 shows preliminary results for the multiplicities of neutrons, Z=2 to 5 and Z>5 particles [31]. The data are compared with the three different de-excitation models, ABLA, SMM and GEMINI. It can be seen that GEMINI is the only one which correctly predicts the whole set of data. This suggests that it has the right competition between the different types of particles and the correct dependence with excitation energy. It should be stressed that this agreement is obtained with the default parameters of GEMINI, except that Hauser-Feshback evaporation is extended up to Z=4.

Figure 8: Preliminary results of multiplicity of low energy neutrons, Z=2 to 5 and Z>5 particles, as a function of the total charge of fragments with Z≥2, Z_{Bound}, measured in the SPALADIN experiment. Calculations with INCL4 coupled either to ABLA (dashed-dotted line), SMM or GEMINI (full line) are also shown. From [31].



From all what precedes, it can be concluded that GEMINI seems the best de-excitation model as regards particle evaporation and IMF production, likely because of the use of the Hauser-Feshbach formalism and the possibility of asymmetric fission. It has to be noticed however, that the same GEMINI model is not able to reproduce data on heavy systems, in particular fission. An ideal de-excitation model would combine the fission of ABLA and the evaporation of GEMINI.

5. Conclusion

Thanks to the validation on the high-quality elementary data now available, the predicting capabilities of nuclear models implemented into widely used high-energy transport codes can now be assessed. It can be concluded that neutron production, heavy spallation residues and total activity of the spallation target can be calculated reliably by most of the models. As regards volatile fission element production, INCL4-ABLA should be used instead of the default option of MCNPX. For gas, light evaporation residue and intermediate mass fragment productions, the standard models are not yet reliable enough. From the most recent experimental data, in particular the more constraining coincidence experiments, the reaction mechanism can now be better understood and this indicates tracks to follow in order to improve the models. Improvements of both the INC and the de-excitation models are in progress and should help solving the identified deficiencies and providing reliable predictions for spallation source design.

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