High-energy nuclear data for ADS

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Abstract. Accelerator-driven sub-critical reactors use intense neutron fluxes produced through spallation reactions in a heavy metal target. During the last years, new high-quality experimental data have been collected leading to a better understanding of the spallation reaction mechanism and to the development of more reliable spallation models. Examples of recent experimental data and model improvements are presented. The impact on target design, radioactivity production and material damage are discussed. The importance of benchmarking the available models against well chosen experimental data is also emphasized.

1. Introduction

In accelerator-driven sub-critical reactors (ADS) an intense neutron flux is produced through spallation reactions in a heavy metal target and used to drive the sub-critical core. Specific nuclear data related to the high-energy interactions occurring in the spallation target and in its surroundings are needed to optimize the target and assess the induced radioactivity and material damages.

A spallation reaction is generally described as a two step mechanism: first, a succession of individual nucleon-nucleon collisions, which leads to the ejection of a few energetic particles, and the deposition of part of the incident energy into the target nucleus. The excited remnant then decays by evaporating particles, mainly neutrons, or by fission in the case of heavy nuclei. Some of the nucleons ejected during the spallation reactions have sufficient energy to induce further spallation reactions with neighbouring nuclei inside a thick target, leading to a multiplication of the emitted neutrons. It is therefore possible to produce intense neutron fluxes with high-intensity proton beams.

Simulation codes used for ADS design generally consist of a high-energy transport code, in which nuclear models give the production yields and characteristics of all the nuclei generated in spallation reactions down to 20 (sometimes 150) MeV, and evaluated nuclear-data files are used below. To provide reliable predictions, it is therefore necessary to have models describing correctly all the features of the spallation reactions, validated on appropriate experimental data. During the last years, new high-quality experimental data have been collected, leading to a better understanding of the reaction mechanism and allowing testing the currently used codes [1, 2]. This has resulted in the development of more reliable models.

This paper summarizes the progress made recently and, when possible, the degree of reliability that can be expected from the model predictions. It points out the importance of benchmarking the available models against well-chosen experimental data and shows that further work is still necessary.

2. Specificities of spallation reactions

In a spallation reaction, high-energy nucleons are ejected during the cascade stage. Their number is rather small, around 2 neutrons and 2 protons above 20 MeV per reaction in the case of Pb at 1 GeV, but they carry out the largest part (about 90%) of the energy and induce secondary reactions in a thick target. During the de-excitation of the excited remnant nucleus low energy (below 20 MeV) particles, mostly neutrons (around 15 in Pb) are emitted. In an ADS, the total number of produced neutrons is related to the efficiency of the driving source. The high-energy neutrons should be properly predicted for radioprotection issues and because they can reach the fuel and structural materials.

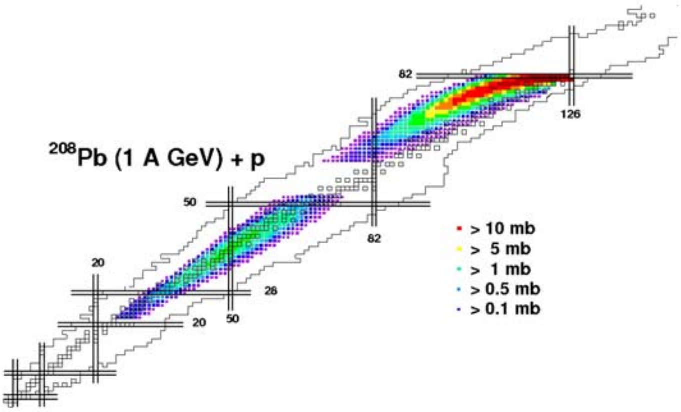

FIG. 1. Nuclide production cross sections measured at GSI for the reaction p+Pb at 1 A GeV on a chart of the nuclides [3]. The colors indicate the production cross sections.

An important feature of spallation reactions is the large number of different residues that are produced. The example of proton on lead at 1 GeV, measured using the reverse kinematics technique [3], is shown in FIG. 1 left. The cross-sections of the thousand measured isotopes are represented on a nuclear chart, the empty squares indicating the stable nuclei. It can be seen that most of the residues are radioactive.

Also specific to high-energy reactions is the large production cross-sections of light charged particles (protons and composites particles) which increases with increasing incident energy up to a plateau around 1 GeV as can be seen in FIG. 7 right from [21]. Actually, most of them are produced in the de-excitation stage because of rather high excitation energies. Composite particle spectra exhibit a high energy tail, which has to be explained either by coalescence of cascade particles or by pre-equilibrium emission. Globally, this leads to a production rate of gases, hydrogen, helium and tritium, much higher than in conventional reactors.

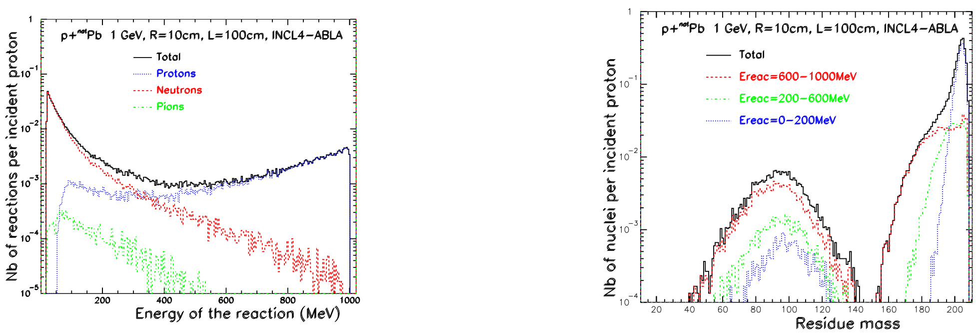

FIG. 2. Left: Respective and total numbers of reactions per incident proton due to the interactions of the different particles involved in a thick Pb target bombarded by a 1 GeV beam. Right: Number of nuclides per incident proton as a function of their mass for different energy bins of the interaction.

In a thick target, because the reaction cross-section of a high energy proton is large (1.7 barns on Pb) a large fraction of the primary proton beam interacts in the first centimetres of the spallation target before slowing down. This explains that the maximum of the energy deposition in a spallation target is located close to the entrance of the beam. FIG. 2 left shows the respective numbers of reactions per incident proton as a function of the energy of the reaction for the different types of particles involved in the interaction of a proton with a thick target. The large number of low energy interactions is due to secondary particles and is dominated by neutron reactions because evaporated particles are mostly neutrons and low energy charged particles have a large probability to stop without interacting.

In a thick target, a lot of residues are also produced by the secondary reactions. Actually, 3.5 isotopes per incident proton are created in average in a Pb target bombarded by a 1 GeV beam. The right panel in FIG. 2 shows the mass distribution of the residues produced in the different interactions, divided into several bins in energy of the reaction, occurring in a thick lead target. It can be observed that the heaviest residues are created mostly in secondary low-energy (between 20 and 200 MeV) interactions while the fission products originate essentially from high-energy primary ones.

3. Progress achieved during the last years

3.1. Neutron production

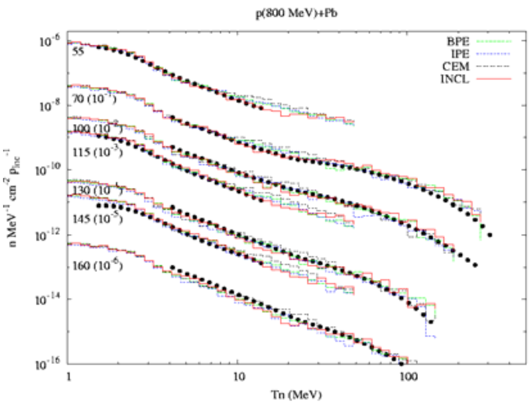

FIG. 3: Energy spectra of neutrons emitted from a thick Pb target irradiated by 800 MeV protons, compared to different models. From [4].

A large amount of neutron data, double-differential cross-sections and multiplicity distributions, are available. Most of the spallation models generally predict reasonably well the elementary as well as thick target data. This is illustrated in FIG. 3 from [4] which shows the comparison of different models with neutron energy spectra from a thick lead target. Similar conclusions were drawn by [5] in a recent benchmark for EURISOL. Consequently, the total number of produced neutrons in a thick target as well as their energy and angular distributions can be considered as reliably predicted, with a precision of the order of 15 to 20%.

3.2. Residues

The experimental programme using the reverse kinematics technique at the Fragment Separator at GSI [6] has permitted a considerable breakthrough in the knowledge of spallation residues. It has allowed for the first time the measurement of the complete isotopic distribution of most of the elements produced in spallation reactions (FIG. 1). This has brought severe constraints on the models and, for instance, has led to rule out the old Bertini-

Dresner model used as the default option of many transport code, especially in the region of fission fragments.

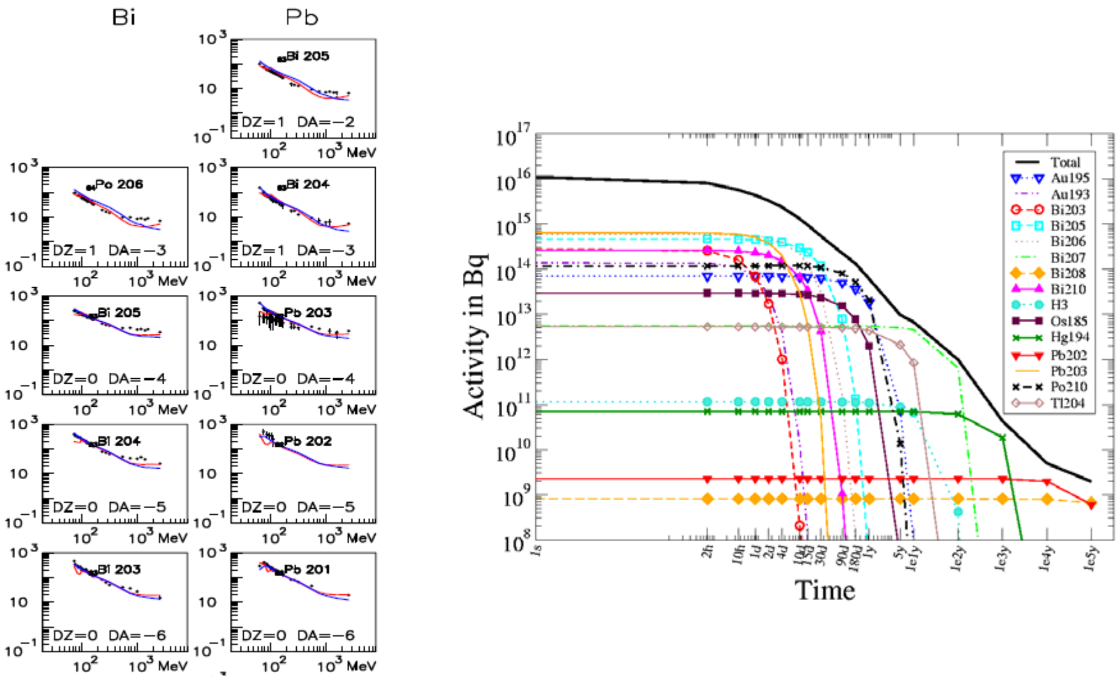

FIG. 4: Left: Production cross-sections (mb) of a few isotopes measured in p+Pb or p+Bi by γ-spectroscopy [7] as a function of the incident proton energy compared with predictions of INCL4-ABLA [8, 9] and Bertini-Dresner. Right: Main contributors to the activity of the liquid lead-bismuth MEGAPIE target as a function of cooling time after 123 days of irradiation with 575 MeV protons at 0.947 mA, calculated with INCL4-ABLA. From [10].

Since in a thick target secondary reactions are responsible for the production of more than two-third of the residues, it is necessary to also have production cross-sections all over the energy range of the possible reactions. Therefore, excitation functions measured for specific isotopes are needed to check that the models behave correctly at all energies. FIG. 4 (right) shows the total activity and the main contributors in the case of the MEGAPIE target [10]. As expected from FIG. 2 right, the isotopes contributing the most to the activity are radioactive nuclides close to the target elements (Pb and Bi here). Generally, the models used to simulate the reaction are well established for small energy transfers as shown in [11] in comparisons to measurements [7] of excitation functions. Therefore, their predictions for these isotopes are very similar and reliable in a wide range of incident energies.

Fission products are also important for radioprotection since some of them are gaseous elements, as Kr, Xe or I, which are released from the liquid metal during operation. Comparison of models to GSI elementary data have shown that Bertini-Dresner is not able to predict properly the fission residues (see FIG. 5 left) while INCL4-ABLA gives a very good agreement. The same is true in a thick target as shown in FIG. 5 right, in which the production rates of Xe from a Pb-Bi target irradiated by 1400 MeV protons measured at Isolde [12] are compared to different calculations. INCL4-ABLA, but also FLUKA [13], gives a reasonable agreement with the data while Bertini-Dresner overestimates the production rates by nearly one order of magnitude. This confirms that Bertini-Dresner should be avoided to calculate fission residues.

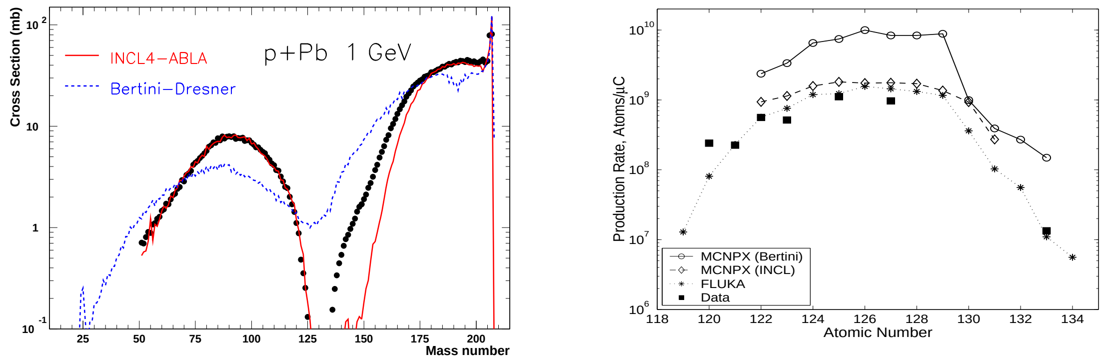

FIG. 5. Left: Comparison of INCL4-ABLA and Bertini-Dresner predictions with the residue mass distribution measured [7] for the p+Pb system. Right: Production rates of Xe isotopes from a thick Pb-Bi target, bombarded with 1.4 GeV protons, measured at Isolde [12], compared to different Bertini-Dresner, INCL4-ABLA and FLUKA [13] calculation.

FIG. 5 left also shows that a systematic misprediction of light evaporation residues is observed with INCL4-ABLA and other standard models [14]. This is even more pronounced in light systems as iron, shown in FIG. 6 left from [15]. 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 some impurities in a given material. In addition, very light fragments with charge between 3 and 10, often called intermediate mass fragments, observed in a lot of experiments [7, 18, 19] are generally not predicted by the models [11]. Among those are for instance ^{7}Be and ^{10}Be, which are a concern for radioprotection.

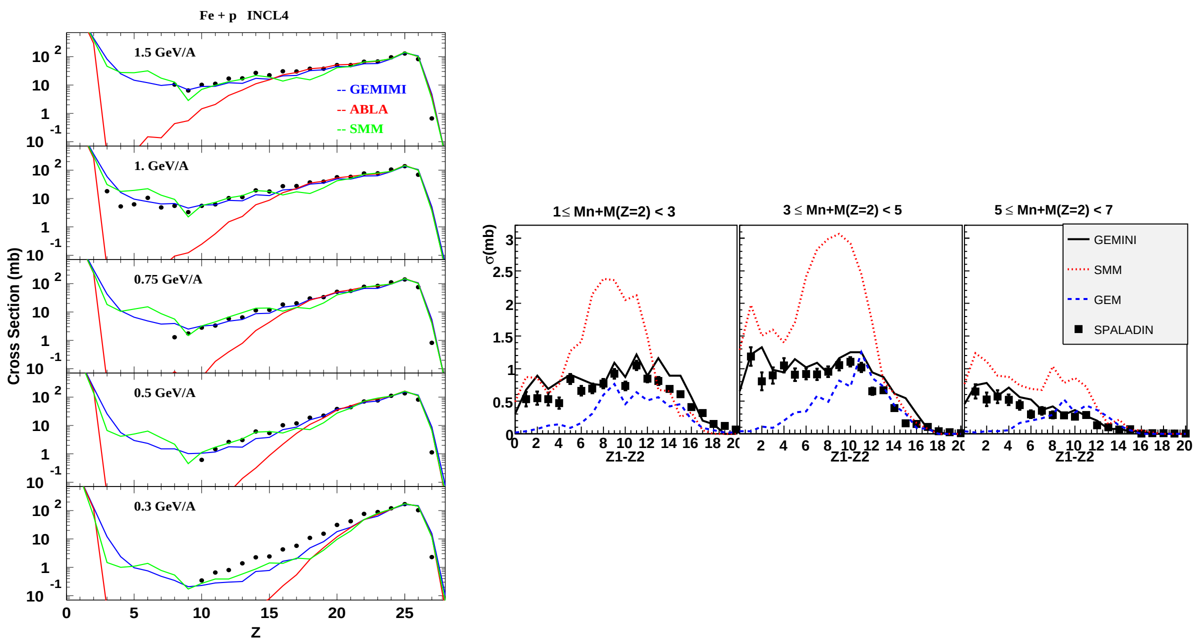

FIG. 6. Left: Charge distributions in p+Fe at five energies [15], compared with the predictions of the INCL4 model coupled with the de-excitation codes ABLA [9], GEMINI [16] or SMM [17]. Right: SPALADIN experiment: charge difference between the 2 heaviest fragments detected in multifragment events, for 3 bins in the multiplicity of neutrons and Z=2 particles. From [20].

The SPALADIN experiment, performed in GSI, on p+Fe at 1 GeV [20], in which residues, neutrons and light charged particles have recently been measured simultaneously, brings a new insight on the reaction mechanism. It has been possible, for instance, to measure in each event the correlation between a variable related to the excitation energy at the end of the cascade stage, the multiplicity of neutrons and Z=2 particles, and the charge difference between the 2 heaviest fragments. In FIG. 6 right, the data are compared with three different de-excitation models, ABLA [9], SMM [17] and GEMINI [16]. 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. This shows that, contrary to inclusive data (FIG.6 left) from which no clear conclusion could be drawn, more exclusive experiments could bring much more severe constraints on models.

3.3. Light charged particles

Reliable estimations of light charged particle production are important for spallation neutron sources: for instance, helium is a concern in structural materials, in particular the window separating the accelerator vacuum, because it can lead to swelling and embrittlement; tritium can escape from the liquid target and cause problems of radioprotection.

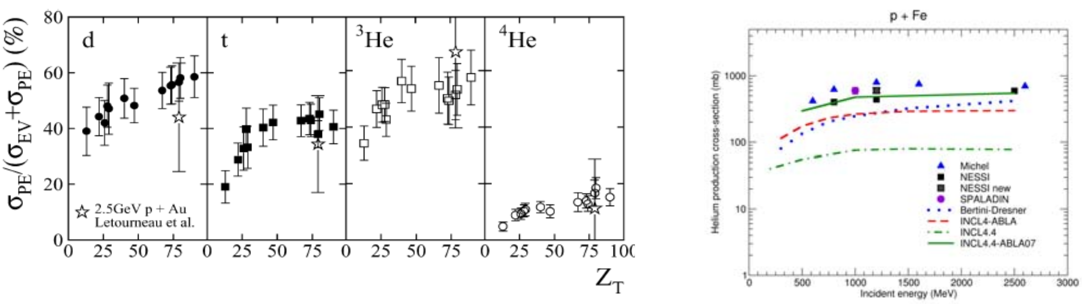

FIG. 7. Left: Contributions of the pre-equilibrium emission relative to the total yield of light composite particles measured for reactions with 1.2 GeV protons as a function of the atomic target number Z_T. From [18]. Right: Helium production cross-sections in iron calculated with the new versions INCL4-ABLA07 compared to experimental data and to calculations with the standard INCL4-ABLA and Bertini-Dresner models. From [21].

Up to now, light composite particle production was rather poorly predicted by the different models implemented into MCNPX [22] or by FLUKA [23]. Often, in the models only the emission from the evaporation stage is considered and the observed forward-peaked high-energy tail is neglected. The relative contribution of this high-energy component, called pre-equilibrium in this paper, relative to the total yields of the different light composite particles, has been recently estimated in [18], from which FIG. 5 left is taken. It can be seen that, while ^{4}He is produced predominantly by evaporation, for the other light composite particles the so-called pre-equilibrium contribution is far from being negligible, reaching 40% for tritium and even 60% for ^{3}He on high Z targets. The level of this contribution shows the importance of being able to account for the high-energy tail with the models. This is the reason why the possibility to emit light composite particles through a mechanism of coalescence in phase space has been introduced in a new version of INCL4 [24].

The ABLA model in his standard version only allows the evaporation of n, p and ^{4}He. This means that tritium cannot be reliably predicted with INCL4-ABLA. This is a major deficiency as tritium appears, for instance, to be a major contributor to the radioactivity of the MEGAPIE target around 10 years after irradiation [10]. In order to solve this problem, ABLA has been recently modified to include the evaporation of all types of light particles and emission of intermediate mass fragments [25] with appropriate emission barriers.

FIG. 7 right shows the result (solid line) obtained with the new versions INCL4.4 coupled to ABLA07, for total helium production cross-section in iron as a function of proton incident energy. The comparison to the experimental data and to calculations with other models shows that obviously, the new models represent a progress compared to the previous one (dashed line) or to Bertini-Dresner (dotted line). Tritium production, not shown here, is now also reasonably predicted [21].

4. Forthcoming activities

As already pointed out, there is a need for reliable simulation tools in which the event generators used to compute the production yields and characteristics of the nuclei generated in the interactions are built on solid nuclear physics bases and validated against experimental data. The International Atomic Energy Agency (IAEA) and the Abdus Salam International Centre for Theoretical Physics (ICTP) have recently organised an expert meeting on model codes for spallation reactions [26]. The first goal of this workshop was to bring together the experts in spallation models in order to discuss the state-of-the-art of the models used in high-energy transport codes, in particular their validity and deficiencies, but also to consider other, often more sophisticated, models that could either be implemented in the future in transport codes or serve as reference calculations. Since it is of great importance to validate on selected experimental data the abilities of the various codes to predict reliably the different quantities relevant for applications, it has been agreed to organize an international benchmark of the different models developed by different groups in the world. The specifications of the benchmark [27], including the set of selected experimental data to be compared to models, have been fixed during the workshop. The objectives of this benchmark are: i) to assess the prediction capabilities of the spallation models used or that could be used in the future in high-energy transport codes; ii) to understand the reason for the success or deficiency of the models in the different mass and energy regions or for the different exit channels; iii) to reach a consensus, if possible, on some of the physics ingredients that should be used in the models.

Most of the well-known spallation model developers have agreed to participate to the benchmark and have presented preliminary results in the Satellite Meeting on Nuclear Spallation Reactions during this conference. Detailed comparisons of the model calculations with the experimental data set will be performed in the next months and a final workshop to discuss the results and draw conclusions is foreseen beginning of 2010.

The work devoted to solving the already identified deficiencies in the models and measuring more constraining experimental data has to be continued. A new FP7 project, ANDES, on nuclear data has recently been submitted, which contains a workpackage devoted to high-energy reactions. Actually, the goal is to cover the energy range between 150 and 600 MeV. This range has not been studied with the same level of accuracy than higher or lower energies, but on the other hand is the most probable for the first experimental/demonstration devices like the MYRRHA/XT-ADS project [28]. The project will focus on the validation of the high-energy nuclear models used in transport codes in this energy domain. The goal is first to assess and then to improve their predicting capabilities, with emphasis on the most quantities important for the ADS demonstrator, such as the target radioactivity, the gas production (tritium, helium and volatile radioactive elements produced in the liquid metal target), or the damage in the surrounding structural materials. Because there are still inconsistent or missing appropriate experimental data in the energy domain, a few specific experiments, focused on measurements of clear relevance for model validation, will be performed.

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