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Production of heavy clusters (up to $A=10$) by coalescence during the intranuclear cascade phase of spallation reactions

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Abstract. Spallation reactions are generally considered to proceed in two stages: a cascade stage followed by an evaporation stage. Light charged particle (lcp) spectra indicate that such particles are produced by both stages. The mechanism of production in the cascade stage is still not fully understood. Using the improved versions of the Liège Intra-Nuclear Cascade model and of the ABLA evaporation model, we have recently shown that lcp's are presumably produced by some kind of dynamical coalescence process, by which a fast particle of the cascade drags a few other nucleons. In the very recent years, precise measurements of the production of heavier clusters have been performed. We improved and generalized our production models for the heavier clusters, up to $A=10$ and we reached good agreement with experiment. These results strongly suggest that the dynamical coalescence mechanism applies to heavy clusters. The importance of these data for spallation neutron sources and accelerator-driven systems is underlined.

1. Introduction

Spallation reactions are high-energy reactions induced by nucleons and/or light nuclei. In the recent years, a renewed interest has emerged for proton-induced reactions in the GeV incident energy range, mainly triggered by the contemplated possibility of transmuting nuclear waste in ADS. In such devices, a high energy proton beam bombards a spallation target inside a subcritical nuclear reactor core. The neutrons which are copiously produced by spallation reactions, are multiplied in the reactor core and are used for transmuting nuclear waste, mainly by fission (see [1] for a review). The first project of this kind, the MYRRHA project, has recently been launched, in Belgium [2]. Accordingly, we will limit ourselves in this paper to the proton-induced reactions below 3 GeV.

In spallation reactions, neutrons are produced abundantly. This emission is accompanied by the much smaller (by one order of magnitude, typically) production of light charged particles (lcp). With still a lesser frequency, so-called intermediate mass fragments (IMF) are also emitted.

There is a general consensus among physicists on the idea that spallation reactions proceed in two stages, a fast stage, characterized by the emission of fast particles, followed by a slow stage, in which an excited, but more or less equilibrated, remnant emits slow particles by evaporation-like

processes. Likewise, the basic theoretical tool consists in the coupling of an intranuclear cascade (INC) model, for the first stage, to an evaporation/fission model, for the second stage. In such models, nucleons are emitted in both stages, and light clusters, like alpha particles, are emitted only in the evaporation stage. But measurements clearly indicate that lcp's can be emitted with a kinetic energy of 100 MeV or more, much larger than the typical evaporation energies ($\leq \sim 20$ MeV), very likely during the cascade stage. To cure the deficiency of the models, the emission of the lcp's, up to α 's, has been introduced in the cascade stage, during the recent years. In ref. [3], some of the authors of this paper have implemented a kind of dynamical coalescence model in the Liège Intra-Nuclear Cascade (INCL) model: an unbound nucleon crossing the surface can drag other nucleons to form a cluster, provided these other nucleons are sufficiently close in phase space. This model is successful in reproducing the gross features of the double differential cross sections for emissions of lcp's, up to α 's [3]. In the latter reference, old versions of the INCL and ABLA codes were used. Recently, calculations have been redone with the latest versions of the models (see later for detail) and results were improved. Especially, the excitation functions of the total production cross sections of tritium and helium isotopes are well reproduced on a broad domain of incident energy [4]. Other authors have also used simpler coalescence models. See ref.[5] for more information. In the meantime, new measurements, especially by the PISA collaboration [6], have shown that heavier clusters can be emitted with large kinetic energy. For our convenience here, we will call "light clusters", those which are not heavier than the alpha particle, and "heavy clusters", those which are heavier, up to $A=10$. It is the purpose of this paper to investigate whether the production of the heavy clusters can be described with the same theoretical model as for the light ones. This question presents both theoretical and practical interests. If it is clear that light clusters can be produced by both the cascade and the evaporation stages, it may be not so for heavier clusters like ${}^9\text{Be}$ or even less so for clusters like ${}^{22}\text{Ne}$, a question which has been repeatedly raised by R. Michel [7]. On the practical side, many of these (both light and heavy) isotopes are unstable (like ${}^3\text{H}$ and ${}^7\text{Be}$) or volatile, and may pose problems of radioprotection and/or damages of materials around spallation sources and ADS.

2. Theoretical tools

2.1. *The INCL4.5+ABLA07 theoretical model*

We use here the updated versions of the Liège INC model [8], denoted as INCL4.5, and of the ABLA de-excitation model [9,10], known as ABLA07. These updated versions have not been published yet, but good accounts can be found in refs. [11] and [12], respectively. It is sufficient for our purpose here to remind the salient features of the models. The INCL4.5 model is a time-like INC model, following the fate of all particles in space-time. Particles are travelling along straight-line trajectories, until either two of them reach a sufficiently small minimum distance of approach, in which case a collision is realized, or if a particle hits the nuclear surface, in which case it is either reflected or transmitted according to transmission probability on the nuclear potential surface, or if a particle (a Delta resonance) decays. Inelasticity is taken into account by explicitly introducing pion and Delta degrees of freedom. Nucleons are moving in a nuclear potential well and collisions are subject to Pauli blocking. Special care is exercised for soft collisions, which makes the model to work well even down to 50 MeV (see ref [11] for detail). As we said, in INCL4.5, a new module is introduced for the emission of clusters. It is described in some detail below. When the cascade is stopped (at a time which is determined self-consistently, a unique feature of INCL [8]), the properties of the remnant nucleus are transmitted to the ABLA07 model, which describes the subsequent de-excitation. In this model, neutron, proton and any stable nucleus up to half of the mass of the remnant can be emitted, on the basis of the Weisskopf-Ewing formalism, even if simple estimates of the emission widths are used for the largest clusters ($Z>3$). On the other hand, parametrizations of inverse cross sections and

of the level density, especially concerning the pairing effects, have been improved. Similarly, a simplified procedure has been used in order to take account of the emission of angular momentum in course of the successive emission. The ABLA code is known for including a sophisticated fission module: fission width is determined by the Bohr-Wheeler formula using realistic level densities at the barrier, mass partition is based on microscopically calculated energy surfaces, including realistic shell effects, and time delay is introduced to take account of viscosity in the collective motion toward scission. In ABLA07, the latter is replaced by a time-dependent fission width, the dependence being parametrized on solutions of detailed Fokker-Planck equations. In this new version, if the temperature of the remnant exceeds a certain value, which depends upon the mass, another exit channel, namely multifragmentation, is accommodated. Finally, thermal expansion of the nucleus is also introduced. These last two features are of minor importance in the context of this paper. The INCL4.5+ABLA07 code has been “validated” in a recent intercomparison of numerical codes for spallation reactions, organized by the IAEA [5]: it has been recognized as one of the best codes, for all kind of observables: double differential cross sections, multiplicities, mass and charge spectra for residues, excitation functions, etc.

2.2. The cluster formation model inside INCL4.5

As we said in the Introduction, the production of clusters in INCL4.5 relies on the idea that a outgoing nucleon hitting the surface can drag along particles which are sufficiently close to it, elaborating on the original idea of Butler and Pearson [13]. Here are some details about the procedure.

- when an unbound nucleon, denoted as the leading nucleon, is at a radial distance $r = R_0 + h$ (where R_0 is the half-density radius), all potential clusters of mass $A=2$ to A_{max} are constructed, by selecting $A-1$ nucleons which, with the leading nucleon, are sufficiently “packed” in *phase space*. Actually, they have to satisfy the following criteria:

$$r_{i,[i-1]} p_{i,[i-1]} \leq \pi, \quad i = 2, 3, \dots, A, \quad (1)$$

where $r_{i,[i-1]}$ and $p_{i,[i-1]}$ are the Jacobian coordinates of the i -th nucleon, i.e. the relative spatial and momentum coordinates of this nucleon with respect to the subgroup constituted of the first $[i-1]$ nucleons, the leading nucleon corresponding always to $i=1$. The quantity π is a function of A .

- the “most bound” cluster is selected: if \sqrt{s} is the total energy of a constructed cluster and if $B(A, Z)$ is its nominal binding energy, the cluster corresponding to the lowest value of $(\sqrt{s} - Am_N - B(A, Z))/A$ is selected.
- this cluster is emitted provided (i) its energy (in the target system) is above the threshold for emission (ii) the test for transmission through the Coulomb barrier is positive (iii) the emission angle is not too tangential; if this angle θ is defined as the angle between the direction of emission (determined by the total momentum of the cluster) and the radial outward direction at the location of the leading nucleon, it is required that $\theta < 45^\circ$; the idea beyond this condition is that the longer a cluster stays in the surface region, the more it is expected to get dissolved; if one of these conditions is not met, the leading nucleon is emitted, provided it can tunnel to its Coulomb barrier, otherwise it is reflected;
- all known clusters up to $A_{max} = 10$ are considered; unstable clusters of very short lifetime, such as ${}^8\text{Be}$, are forced to decay at the end of the cascade stage;
- the parameter h is equal to 1.0 fm; we could not obtain good results, especially at low energy, with a single “proximity” parameter, contrarily to what was done for light clusters, at high incident energy at least [3]. We let π depend upon the mass A and used (in $\text{MeV} \times \text{fm}/c$ units): $\pi(d) = 424$, $\pi(t) = 300$, $\pi({}^3\text{He}) = 300$, $\pi(4\text{He}) = 300$ and for $A > 4$,

$\pi(A) = 210A^{1/3}$. These values have been obtained by fitting on experimental data in a few illustrative cases.

Compared to our previous model for cluster production, which was restricted to light clusters, the main difference deals with the selection of the cluster: in the previous model, the largest possible cluster was selected. We had also only one single proximity parameters for all clusters.

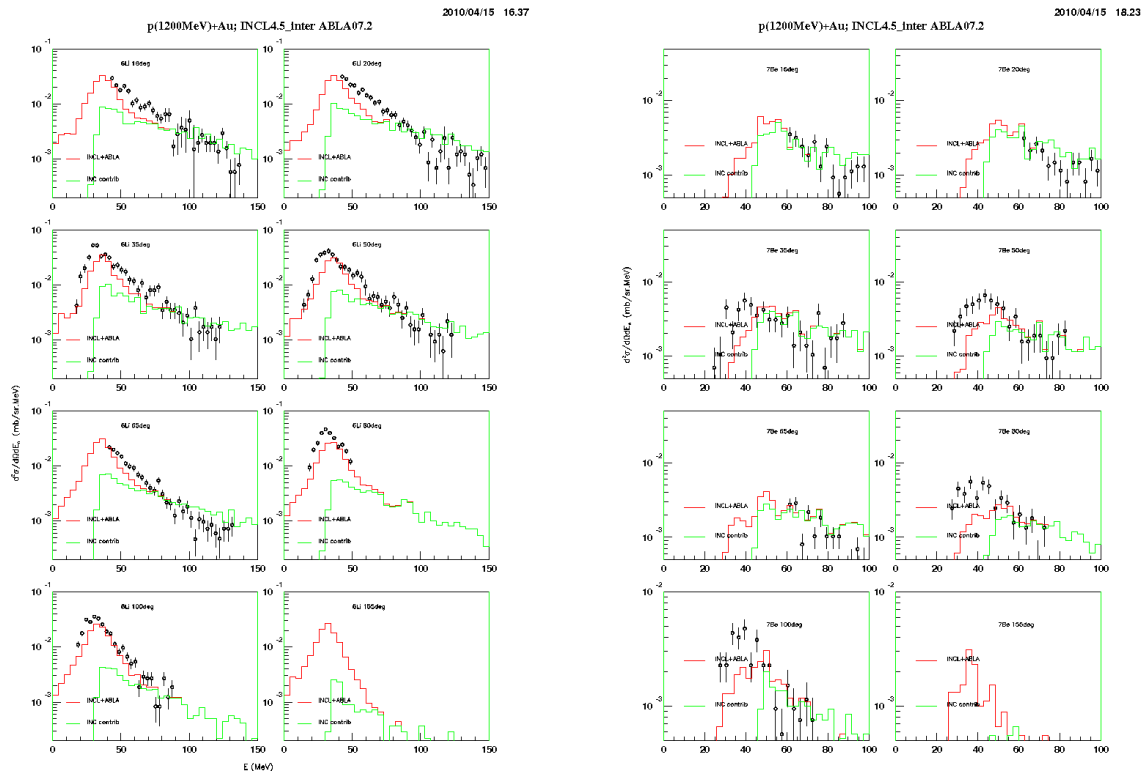


Figure 1. Double differential cross section of ${}^6\text{Li}$ (left panel) and of ${}^7\text{Be}$ (right panel) production in p(1200MeV)+Au reactions. The corresponding angles are given in the various subpanels. Comparison of our predictions, using the INCL4.5+ABLA07 model (red histograms), with the experimental data (heavy dots) of [14]. The cascade contribution is given by the green histograms.

3. Results

We want to mention first that, for light clusters, our new model generates, most of the time, better results than before. Actually, for low incident energy, we get now quite good results. This applies to experimental data for proton energy ranging from 62 to 2500 MeV and targets ranging from Al to Bi. Results can be found in refs [4] and [5].

We now turn to heavy clusters. In Fig. 1, we show typical results for double differential cross sections. Clearly, the high energy tails of the spectra, say above 50 MeV, are entirely due to the cascade and are well reproduced by our calculation. It is interesting to note that the ${}^6\text{Li}$ isotope is predominantly produced by evaporation, whereas the situation is roughly reversed for

${}^7\text{Be}$, which is mainly produced in the cascade stage. Due to lack of space, we cannot give here the results of all our calculations. We have calculated the production of ${}^6\text{He}$, ${}^6\text{Li}$, ${}^7\text{Li}$, ${}^7\text{Be}$, ${}^8\text{Li}$, ${}^9\text{Li}$, ${}^9\text{Be}$, ${}^{10}\text{Be}$ and ${}^{10}\text{B}$ isotopes and compared with the experimental data of ref [14], for p+Au reactions at 1.2, 1.9 and 2.5 GeV, of ref [15], for $p + \text{Au}$ reactions at 2.5 GeV, and of ref [16], for reactions of 1.2 GeV protons on 13 targets ranging from *Al* to *Th*. Other interesting data are contained in Refs. [17-20]. We did not compare with these works because the data are less systematic or are more or less similar to those of Refs. [14-15]. Of course, not all calculated isotopes have been measured in all cases, and in some cases [15] heavier isotopes, such as *N*, *O* and *F* have been measured. We summarize our results:

- our model provides an overall good description for all calculated clusters (not only the heavy ones), corresponding to integrated production cross sections extending typically from 1 b for *d* down to less than 1 mb for the heaviest clusters
- for most of the heavy clusters, the quality of the predictions concerning the double differential cross sections is similar to the one shown by Fig. 1 for ${}^6\text{Li}$ and ${}^7\text{Be}$
- there are some observed deviations: for instance, the production of ${}^9\text{Be}$ is underestimated by the cascade (by a factor 2 or 3) and it is also underestimated in the evaporation; production of *Li* isotopes is somehow underestimated at the largest incident energy
- the cross section for the production of high energy ${}^6\text{He}$ clusters (the cascade contribution) is surprisingly good, in spite of the complex structure of this isotope
- the cascade contribution is fading out, in comparison with the evaporation one, for $A=9-10$. The ABLA07 contribution for *C*, *N*, *O* isotopes in $p(2.5\text{GeV}) + \text{Au}$ reactions [15] is particularly good.

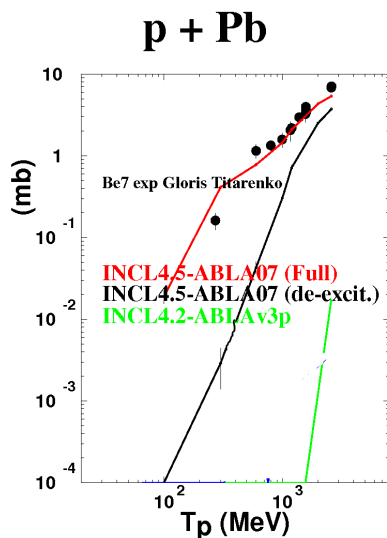


Figure 2. Total ${}^7\text{Be}$ production cross section in p+Pb reactions. Comparison of our predictions, using the INCL4.5+ABLA07 model (red curve), with the experimental data (black dots) of [21,22]. The de-excitation contribution is given by the black curve. The predictions of the previous version of our model are given by the green curve.

We have to notice that, even if our results are very encouraging, they have to be taken with some care. First, we have presented here results with including up to $A=10$ clusters. Moving the limit from $A=8$ to $A=10$ have changed the predictions for $A \leq 8$ clusters. Of course, the predictions for light clusters are very stable, but the yield for those isotopes close to the limit may change by a few percent to a few tens of percent. Actually, we cannot raise the limit very much further up, because of increasing computation time, due to the huge combinatorics involved in the construction of the potential clusters. There is also some uncertainties concerning evaporation

of large clusters, for which Coulomb barrier properties are not well known. Comparison with similar calculations with the GEM evaporation code [23,24] shows discrepancies of factors 2 or 3 for special isotopes.

We also paid attention to the excitation functions and to the target mass dependence. An example of excitation function is given in Fig.2. One can also see from this figure that ${}^7\text{Be}$ is predominantly produced by the cascade at low energy. Above 1 GeV, it is produced in almost equal contributions by cascade and evaporation.

4. Conclusion

We have presented our model INCL4.5+ABLA07 for spallation reactions, as well as the new module for generating heavy clusters in the cascade stage. Preliminary and promising results have been shown. In general, the shape and the amplitude of the spectra are well described. We noticed that some isotopes are more produced in the cascade and some other ones are essentially emitted in the evaporation. The reasons for this apparently complex behavior will be analyzed in a forthcoming publication.

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