Success and deficiencies of high energy nuclear models regarding spallation neutron and residue data

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The detailed design of Spallation Neutron Sources or Accelerator-Driven Systems requires reliable computational tools able to properly predict the production of particles and nuclides produced in the spallation process. During the last years, a significant experimental effort has been undertaken in several places to collect new elementary cross-sections to probe the basic nuclear models. Different models, widely used in transport codes or recently improved ones, are compared with the results of experiments regarding the production of neutrons, measured at SATURNE, and residual nuclei, measured by the reverse kinematics technics at GSI. Conclusions on the success and deficiencies of these models are drawn and implications on applications are discussed.

I. INTRODUCTION

The detailed design of Spallation Neutron Sources or Accelerator-Driven Systems requires reliable computational tools in order to optimize their performances in terms of useful neutron production and to properly assess specific problems likely to happen in such systems. Among those problems are the radioactivity induced by spallation reactions, radiation damage in target, window or structure materials, additional required shielding due to the presence of high energy neutrons, etc... Generally, one uses high energy transport codes, in which elementary cross-sections are generated by nuclear physics models above 20 MeV (or 150 MeV), coupled for lower energies to a neutron transport codes like MCNP [1] that reads evaluated data files. Although the spallation mechanism has been known for many years, the models used in such codes, intra-nuclear cascade followed by evaporation-fission, have never been really validated on experimental data and large discrepancies remain both between experimental data and model predictions and between different models.

During the last years, a significant experimental effort has been undertaken in several places to collect new data regarding neutron production [2–7], light charged particles [8] and heavy residues [9–12]. Therefore, it is now possible to make an extensive comparison of some of the models widely used inside high-energy transport codes with the bulk of available experimental data in order to assess their prediction capability. Improved or new models are also being developed that can be tested and validated against these new data. In Europe, all the work done in this domain is coordinated within the HINDAS project, presented in this conference [13]. In this paper, we present the first conclusions of a work dedicated to the comparison of several models with neutron double differential data measured at SATURNE [6,14] and isotopic distributions of residual nuclei measured by the reverse kinematics technics at GSI [10,11]. Results with improved models, in particular the new version of the Liège intra-nuclear cascade, are also shown. These models have been recently implemented into a transport code and possible implications on applications are discussed.

II. COMPARISON WITH EXISTING MODELS

Spallation reactions are generally described by a two step mechanism: a first stage of individual nucleon-nucleon collisions, generally described by intra-nuclear cascade models, then, the decay of the excited remnant nucleus by evaporation-fission. Some authors [25], introduce a pre-equilibrium stage between intra-nuclear cascade and de-excitation. In high energy transport codes, the most widely used INC model is the old Bertini [16] one dating from

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1963. However, several other models are available, among which the Isabel [17] and the Liège [18,19] INCL models, which have brought some improvements in the physics. The most widely used evaporation model in the domain of spallation reaction is the Dresner model [20], usually associated with the Atchison [21] fission model.

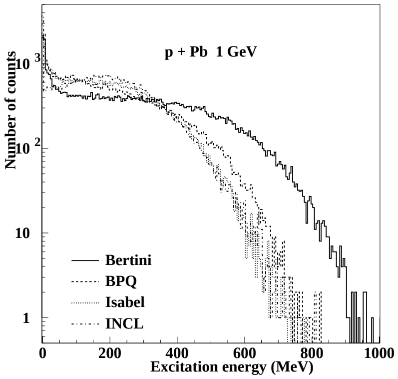

FIG. 1. Excitation energy distribution in the p (1 GeV) + Pb reaction found with the Bertini (solid line), Bertini+pre-eqilibrium (dashed line), Isabel (dotted line) or INCL (dashed-dotted line) intra-nuclear cascade models.

A. Neutron double-differential cross-sections

A program aimed at measuring the production of neutrons induced by protons and deuterons on various targets, was carried out at the SATURNE accelerator in Saclay. Two different experimental techniques were used for these measurements [22,23]: for the low energy part of the neutron spectrum (namely from 2 to 400 MeV) time-of-flight was employed between a neutron sensitive detector and a plastic scintillator detecting the incident particle. High energy neutrons, from 200 MeV to the beam energy, were measured using (n,p) scattering on a liquid hydrogen target and reconstruction of the proton trajectory in a magnetic spectrometer. Measurements were performed with protons and deuterons between 0.8 and 1.6 GeV on various thin targets representative of different parts of the periodic table of elements and corresponding to materials used in targets or in structures of ADS.

The high energy part of the neutron spectra allows to directly probe the INC models. Low energy neutrons, which are the majority of the neutrons produced in spallation reactions, are emitted during the evaporation process. However, their number mainly depends upon the INC stage since it is determined by the excitation energy of the decaying hot residue at the end of the cascade stage.

Results on lead targets, at 800, 1200 and 1600 MeV, were presented in [6] together with calculations performed with the TIERCE [24] code system developed at Bruyères-le-Châtel using either the Bertini or the Liège INCL model with the same evaporation-fission model (based on Dresner-Atchison model). The actual thicknesses of the targets (a few cm) are taken into account in all the calculations presented in this section. It was shown that, at the three energies, the Bertini model was largely overpredicting the experimental data (in particular for the low energy neutrons) while INCL was giving a rather good agreement. This was ascribed to the higher excitation energy, E^*, obtained at the end of the cascade stage with the Bertini calculation than with INCL. This can be verified in fig. 1 where the corresponding E^* distributions (respectively solid and dashed-dotted lines) are shown. Similar conclusions of a too high excitation energy in Bertini were also drawn by other authors on experiments regarding light charged particle [8] and residue [10] production. Several reasons can explain the difference in E^* between both models : first, INCL lead to the emission of more pions than Bertini. However, the difference in average E^* due to the energy carried away by the pions is only around 30 MeV. Second, as mentioned in [6], the Pauli blocking is treated in a different way. In Bertini, only collisions of nucleons with momentum larger than the Fermi momentum are allowed while, in INCL, the actual phase space occupation rate is taken into account. This leads to a less stringent condition, therefore more cascade particles can escape and make the energy remaining in the nucleus lower.

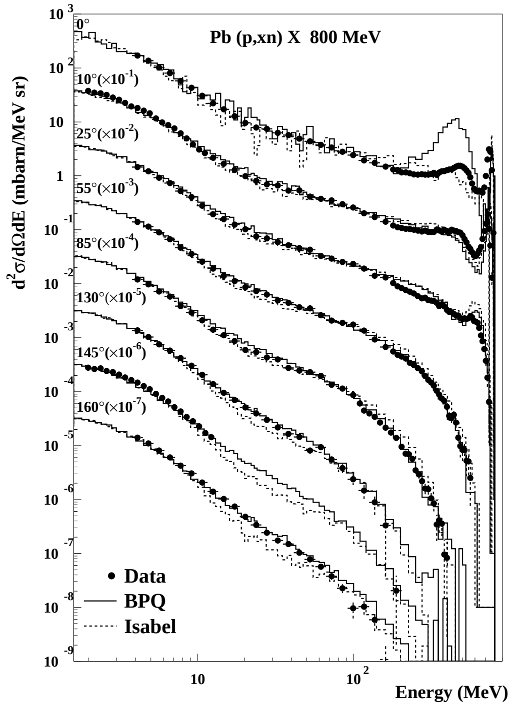

FIG. 2. Experimental p (800 MeV) + Pb neutron double-differential cross-sections compared with calculations performed with LAHET using either Bertini plus pre-equilibrium ((solid line) or Isabel (dashed line) intra-nuclear cascade model.

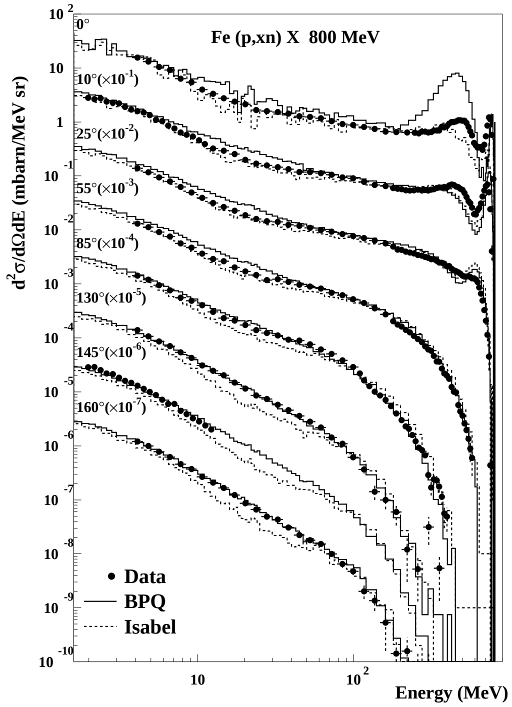

FIG. 3. Experimental p (800 MeV) + Fe neutron double-differential cross-sections compared with calculations performed with LAHET using either Bertini plus pre-equilibrium (solid line) or Isabel (dashed line) intra-nuclear cascade model.

In the LAHET code system [15] it is possible to add after the intra-nuclear cascade stage a pre-equilibrium stage which is expected to reduce the excitation energy of the nucleus by emission of intermediate energy particles prior to the evaporation. In the following, Bertini plus pre-equilibrium will be referred to as BPQ. Also available is the Isabel model which can be used only up to 1 GeV. Calculations have been performed with both models with the same Dresner-Atchison evaporation-fission and compared with SATURNE data on Pb and Fe target [6,14] at 800 MeV. They are shown in Figs. 2 and 3. It can be observed that, for Pb, the BPQ calculation reproduces very well the data, except at very forward angles and high neutron energies where the peak corresponding to the excitation of the Δ resonance appears much too high. This is a deficiency of Bertini INC model, already pointed out in [27] as due to a bad parameterisation of the $NN \to N\Delta$ reaction angular distribution. The problem does not exist with Isabel which, on the other hand, underestimates cross-sections at backward angles in the intermediate energy region. Both models correctly predict the low energy part of the spectra. This can be understood by the respective excitation energy distributions similar in their extension to the one found with INCL (see dashed and dotted lines in fig. 1). For iron, Isabel presents the same features as for lead while BPQ now overpredicts low and intermediate energy neutron production at forward angles, indicating that the angular distribution of pre-equilibrium neutrons is probably too much forward-peaked.

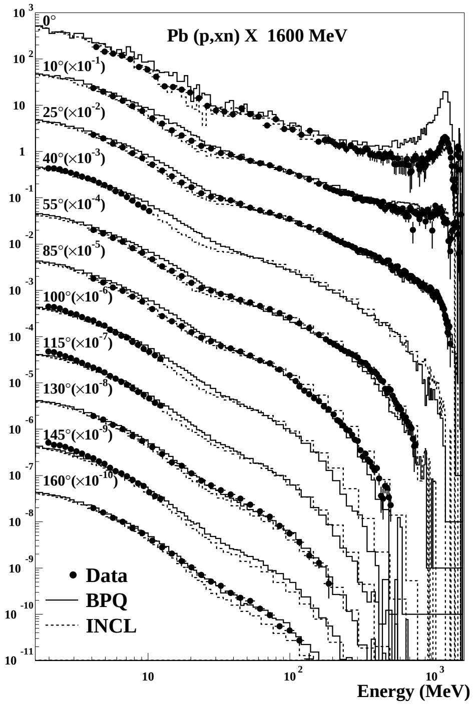

FIG. 4. Experimental p (1600 MeV) + Pb neutron double-differential cross-sections compared with calculations performed with LAHET using either Bertini plus pre-equilibrium (solid line) or INCL (dashed line) intra-nuclear cascade models.

The use of Isabel in LAHET being limited to 1 GeV, data obtained at 1200 MeV on Al, Fe, Zr, W, Pb and Th and at 1600 MeV on Fe and Pb were compared in [6,14] with Bertini, BPQ and INCL calculations only. Whatever the target, Bertini always yields too many low energy neutrons, emphasizing that it leads to too high excitation energies. For heavy targets, both BPQ and INCL models give a reasonnable agreement with the data, although BPQ tends to slightly overestimates the production of intermediate energies neutrons. As the target becomes lighter, this trend is amplified and BPQ begins to also overpredict low energy cross-sections. At 1600 MeV, the discrepancies grow worse: even for Pb, shown in fig. 4, the agreement is not very good between 10 and 40 MeV. Since the high energy part of the spectra is always rather well reproduced (except at 0°), this seems to point out a wrong dependence of the pre-equilibrium emission with incident energy. This is an indication that the addition of a pre-quilibrium stage after INC to decrease the too large excitation energy found in Bertini may not be the proper solution: in fact, it seems difficult to obtain the correct evaporation neutron cross-sections without overestimating intermediate energy ones, i.e. those produced by pre-equilibrium. On the contrary, INCL reproduces quite well the results for all the targets, proving that the model has a correct mass and energy dependence. Only for the Fe targets at very backward angles, the high energy neutron production is underpredicted.

In summary, we can conclude that the Bertini model followed by pre-equilibrium is found to be an improvement compared to Bertini alone since it leads to lower excitation energies. Yet, while it works perfectly for Pb at 800 MeV, it fails as the energy is increased and the target gets lighter. Isabel, tested only at 800 MeV, seems to be not so good for light targets. On the contrary, INCL is able to globally reproduce the bulk of data, with some slight discrepancies in the angular distributions. However, it has to be mentioned that the INCL model does not predict a correct total reaction cross-section mainly because the diffuseness of the nuclear surface is not taken into account. This means that all the INCL calculations had to be renormalized to the experimental total reaction cross-sections values from [26].

B. Residual nuclide isotopic distributions

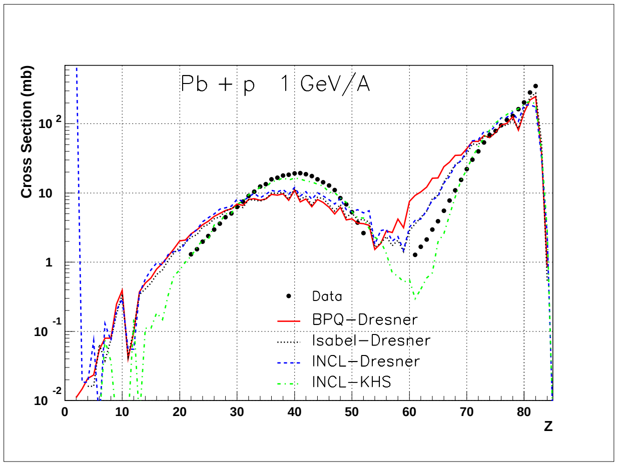

FIG. 5. Element distribution measured in the 1 GeV/A Pb on H2 reaction [10,11]. The solid, dotted and dashed curves were calculated with the BPQ, Isabel and INCL intra-nuclear cascade models, respectively, followed by the Dresner-Atchison evaporation-fission model while the dashed-dotted line is for INCL followed by the ABLA model from [29].

The reverse kinematics technique, together with the fragment separator FRS has been used at GSI to measure and isotopically identify residual nuclei produced in spallation reactions. Experiments with ^{197}Au [12], ^{238}U and ^{208}Pb [10,11] beams between 0.5 and 1 GeV/A on liquid hydrogen and deuterium targets have been carried out. The element distribution measured in the Pb (1 Gev/A) + p reaction is compared in fig.5 with three calculations: BPQ (solid line), Isabel (dotted line) and INCL (dashed line) models followed by the same Dresner-Atchison evaporation-fission model. It can be seen that the slope of the Z-distribution for evaporation residues (Z>61) is quite different for BPQ and the two other models. Since these calculations use the same evaporation, this is related to the difference in the excitation energy distribution and in the mass and charge of the remnant nucleus at the end of the cascade. In fact, BPQ leads to a remnant with a mass significantly smaller than Isabel and INCL, because of the emission of pre-equilibrium particles, and its excitation energy distribution extends to slightly higher values, as observed in fig 1. This explains why it produces lighter nuclei and therefore a flatter slope. In [10], it was shown that Bertini alone leads to an even flatter slope. None of these calculations, which were giving a reasonable agreement with the low energy neutron cross-sections, is able to reproduce these data. This is also true for the fission fragment element distributions which are predicted too wide compared to the experimental data and the total fission cross-section which is found too small. However, contrary to the evaporated neutron spectra which depend only on little on the evaporation model, residue cross-sections appears to be very sensitive to its details. This can be verified in fig. 5 if one compares the INCL-Dresner calculation with the dashed-dotted curve obtained with the same INCL model but with the ABLA evaporation-fission model, presently being developed at GSI [29]. The main differences between this and the Dresner-Atchison models are: the level density parameter which tends towards $\sim A/11$ for high excitation energies rather than $A/8$ for Dresner, the barriers for charged particle emission not depending on excitation energy and the treatment of fission in which friction is introduced and angular momentum is taken into account. Here, the agreement with the data, although far from being perfect, is better for evaporation residues and quite good for the fission fragment element distributions.

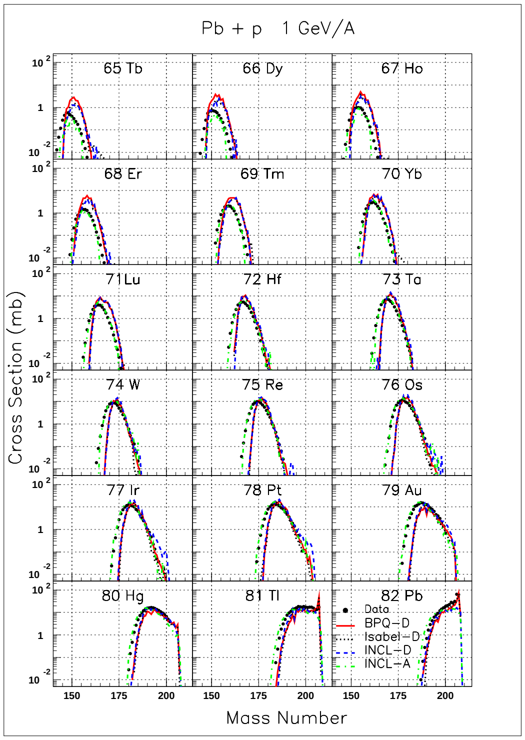

FIG. 6. Isotopic distributions of elements between Z=82 and 65, in the 1 GeV/A Pb on H2 reaction from [10,11] compared with the same calculations as in fig.5.

The same calculations can also be compared to the isotopic distribution of the residues. Fig.6 shows the comparison with the isotopic distributions of evaporation residues. It can be seen that, if the same evaporation-fission model is used, whatever the intra-nuclear cascade model, the shape of these distributions are exactly identical. Only the height of the peak changes a little, following the trends of fig.5. The distribution shapes differ significantly from the experimental ones: they are shifted with respect to the data towards the neutron-rich side. This is ascribed to the fact that the prediction of the neutron/proton/composite particle competition in the Dresner code is not satisfying, likely because the charged particle emission barriers are not correct as suggested by Enke et al. [8]. The use of the ABLA evaporation-fission model, on the contrary, together with the INCL model leads a fairly good agreement with the shapes of the isotopic distributions. However, the main defect of this calculation is the underproduction of isotopes very close to the projectile, also visible on the element distribution, which actually represent a major part of the total cross-section. This is due to the sharp surface approximation in the INCL model which leads to a bad description of the most peripheral reactions.

III. NEW MODELS

As we have seen above, the INCL intra-nuclear cascade model gives encouraging results, especially when coupled to the ABLA evaporation-fission models, but still suffers from severe deficiencies, due in particular to the fact that the difuseness of the nuclear surface is not taken into account. Recently, a new version of this model, called INCL4 [28], has been developed in which a smooth nuclear surface is introduced. The shape of the matter density is now described with a two-parameter Fermi model, the parameters being obtained from electron scattering data. Other improvements have also been made which mainly concern:

- the treatment of the Pauli blocking: the statistical procedure, which is one of the originality of the INCL model, is still used but the final acceptance of a collision is submitted to the condition that this collision does not lead to a negative excitation energy;

- the collision between spectator nucleons which are now forbidden, but spectators are still moving in the nuclear potential;
- the intrinsic angular momentum of the residual nuclei which is now calculated from angular momentum conservation and which is of some importance for the evaporation-fission stage of the spallation;
- the pion emission which has been reduced by increasing absorption.

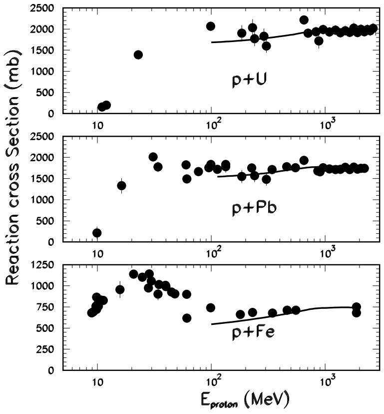

FIG. 7. Total reaction cross-sections obtained with INCL4 and compared to experimental data from [26].

One of the interesting consequences of the introduction of the smooth nuclear surface is that the calculated reaction cross-sections are now in good agreement with the measurements. This is illustrated in fig. 7, where the predictions (full lines) are compared to a compilation of measurements [31,26] for protons on iron, lead and uranium.

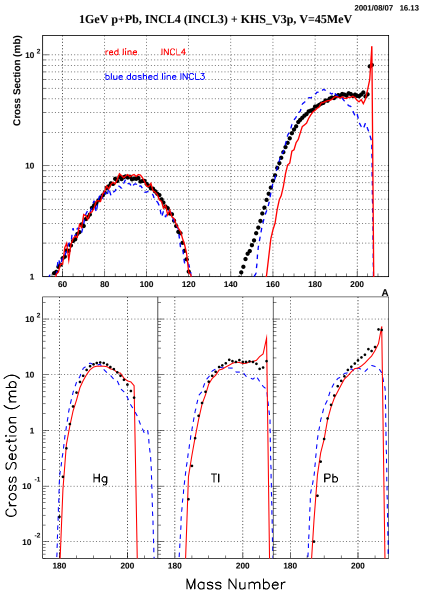

FIG. 8. New (INCL4) and original versions of the INCL model compared to residual nucleus production cross-sections from [10,11] for the 1GeV/A Pb +p reaction

A significative improvement is also obtained on the cross-section of residual nuclei close to the projectile. This can be seen for proton-lead interaction at 1 GeV from [10,11] on fig. 8 where calculations performed with both INCL4 and the old version coupled with the ABLA evaporation-fission code are compared to the measured mass distribution (top) and isotopic distributions of Pb, Ti and Hg (bottom). The realistic density is here crucial for heavy residues produced in peripheral reactions. Note also that the fission part is very well reproduced with the default value of the friction parameter and the intrinsic angular momentum as computed at the end of the cascade. The mean value of this momentum is about a factor 2 larger than the one previously used which was estimated from abrasion-ablation codes developed for heavy ion collisions [30].

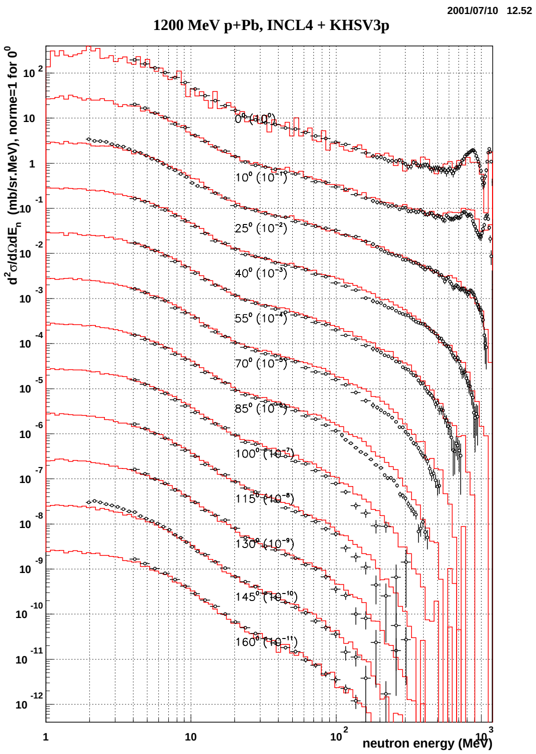

FIG. 9. Comparison of the INCL4-ABLA calculation with the (p,xn) double-differential cross-sections measured on Pb at 1200 MeV.

The neutron double-differential cross-sections measured at SATURNE have also been compared to the INCL4 model. As an example, fig. 9 presents the results obtained for Pb(p,xn)X at 1200 MeV. The quasi-elastic peak and the region of the Δ excitation are slightly improved (compared to the old version) due to the taking into account of the diffuseness of the nuclear surface. The overall agreement is satisfactory above and below 20 MeV, especially if we note that the thickness of the target (2cm) is not taken into account. In fact, this leads to a slight de-population of the high energy to the benefit of the low energy part (30% increase below 3 MeV) [14]. The evaporative part gives an agreement comparable to the previous calculations shown above and done with the Dresner evaporation. The same quality of agreement is obtained for the other targets at 1200 MeV and the other energies measured at SATURNE. Data at lower energies (597, 256 MeV) and (p,xp) cross-sections are also reasonably reproduced [28].

Finally, as it is shown in [28], INCL4 coupled to the ABLA evaporation-fission code developed at GSI, is able to reproduce reasonably well a large set of recent data (neutron and proton production, residual nuclei for a wide set of incident energies from 200 to 1.6 GeV) without any change of the parameters. The possibility to include light fragments as projectiles has also been implemented and comparison with recent deuteron datafrom [32] appears very encouraging.

IV. IMPLICATIONS FOR APPLICATIONS

The final goal of all the present efforts in collecting new experimental data and developing new physics models is to end up with reliable high energy tranport codes for the design of spallation targets. Although we have not yet succeeded to find models able to perfectly reproduce all the available experimental data, the work described above already permits to draw conclusions on the validity of the simulations done with the existing codes.

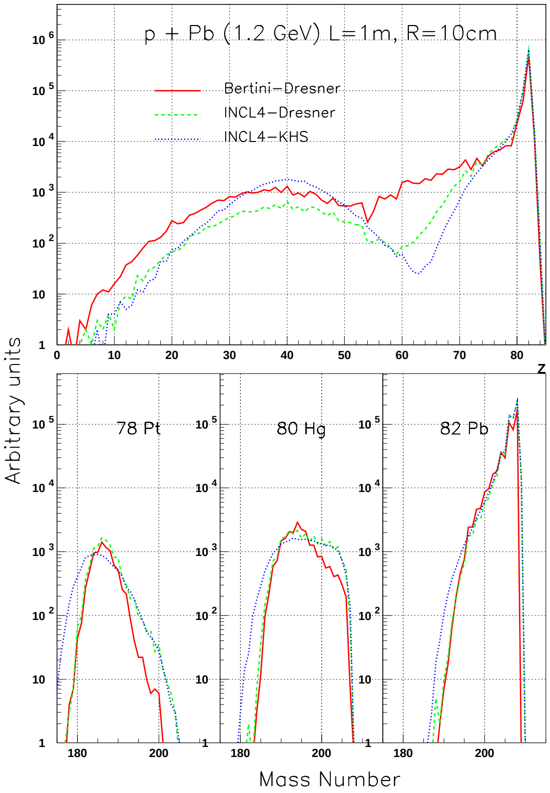

FIG. 10. Element production and isotopic distributions for 3 selected elements in a thick (1m meter length, 10 cm radius) lead target bombarded by a 1.2 GeV proton beam, calculated with LAHET3 code using different intra-nuclear cascade and evaporation-fission models.

For instance, replacing the Bertini by the INCL intranuclear cascade model in a thick lead target simulation, inside the TIERCE high energy transport code [24] was found to decrease by 20% the total number of emitted neutrons [34]. Actually, this is confirmed by results on neutron energy spectra coming out of thick targets [33] which were obtained at SATURNE with the same experimental set-up as the one used for the thin target measurements. In all cases, INCL agrees very well with the low energy part of the neutron spectra while the Bertini model generally leads to an overprediction.

As we have shown in the preceeding sections, probably the major deficiency of the Bertini-Dresner combination of models (which are generally those used by default in most of the high-energy transport codes) appears in the prediction of the residual nuclide cross-sections. It is therefore interesting to estimate the consequences on the residues produced, for instance, in a lead spallation target. Since INCL4-ABLA was the calculation which gave the closest results to the experimental thin target data, both models have very recently been implemented into the LAHET3 [35] code system. We show in fig.10 preliminary calculations of the residual nuclide primary (before radioactive decay) element distribution in a 1 m meter thick, 10 cm radius, natural lead target irradiated by a 1.2 GeV proton beam. The solid, dahed and dotted curves were obtained using respectively Bertini-Dresner, INCL4-Dresner and INCL4-ABLA models. The production of the elements close to the target charge is not very sensitive to the choice of the model. This is due to the fact that these nuclei are mostly produced in reactions induced by low energy secondary particles for which the differences in the model predictions vanish. On the other hand, large differences appear for lighter evaporation residues and fission fragments which exactly reflects the trends observed for thin targets. In fact, it can be verified that these nuclei are essentially produced by the interaction of the incident proton.

The isotopic distributions of a few elements close to the target nucleus are also displayed. It can be seen that even when the total element production is similar with the three models, significative differences can show up in isotopic distributions that can have consequences in the prediction of final element composition and radioactivity predictions. For Z=78 (and lighter elements) it is the change of the evaporation model rather than the change of intra-nuclear cascade which produces the main differences in the isotopic distribution, as already observed for thin targets. First attempts have been made to calculate the differences expected in radioactivity induced in a thick target. Calculations using LAHET3 with Bertini or INCL4 but the same Dresner model led to little variance on the total activity due to the large number of contributing nuclides [36]. Calculations with ABLA evaporation model are under progress.

V. CONCLUSION

In this paper, we have shown a comprehensive comparison of recent data with some of the high energy models commonly used in high energy transport codes. Severe deficiencies have been pointed out, in particular, for the Bertini-Dresner combination which is generally the default model in these codes. The new version of the Liège intranuclear cascade model, INCL4, coupled to the ABLA evaporation-fission model developed at GSI, has been tested rather successfully on the same set of data without changing the parameters. The recent implementation of these two models into the LAHET3 code system makes it now possible to assess the implications for the detailed design of spallation neutron sources.

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