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Physical aspects of lead as a neutron producing target for accelerator transmutation devices

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- **Part 3.9: Analysis of the high energy data (Work Packages 8 and 10)**

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Study of reactions on Pb at intermediate energies (200-2500 MeV)

Theory

3.9. Theoretical analysis of high energy data (WP8 and WP10)

3.9.1. Introduction

Production of neutrons and protons in high energy collisions is amenable to a microscopic description by multiple scattering theory embodying all the way down to the formation and subsequent decay of an equilibrated remnant (Cugnon 2000). The most popular and the most efficient form of these theories is the intranuclear cascade (INC) plus evaporation model. Residue cross-sections can also be described simultaneously by this kind of model. Other data, like composite production yields, may require a more complex approach. The open questions, from the practical point of view, are :

1. Does a proper description of data require an extension of the INC + evaporation model? The division of the whole process between a cascade stage and an evaporation stage may look arbitrary and often arguments are given for the introduction of an intermediate stage, usually called pre-equilibrium.
2. Can the parameters of the INC + evaporation model be determined by fitting to some selected set of data ? The INC models have basically two free (or quasi free) parameters : the depth of the average nuclear potential well and a parameter governing the coupling between the INC and evaporation stages. As for the evaporation model, the most important parameters are the capture cross-sections and the level density parameters. For heavy targets, one should add the relative fission probability and other parameters determining fission properties.
3. Are quantum effects important ? In these models, some quantum effects are mocked up by classical or stochastic means and some other ones are neglected. The goal is to improve the description of the first ones and assess the importance of the second ones.

The practical viewpoint prevailing nowadays is to build an improved INC + evaporation model which can describe experimental data reasonably well by fitting additional parameters to selected benchmark measurements. Even though progress has been made recently, this task is far from being completed. In the following, a progress report is presented concerning mainly the cascade stage at high energy, using the Pb data discussed in the previous sections.

Beyond this empirical approach, the INC + evaporation model should be assessed theoretically. Two main questions arise. What is the theoretical foundation of the division of the whole collision process in two quite different stages? What is the domain of validity of the model ? These questions are just briefly commented in this report.

3.9.2. The situation at the beginning of the Concerted Action

At the beginning of the Concerted Action, most of the practitioners were using the BERTINI INC code (Bertini 1963), often in conjunction with the transport code LAHET (Prael and Lichtenstein 1989), or the ISABEL code (Yariv and Fraenkel 1979). These two codes are rather untransparent and not very amenable to systematic improvements. At about the same time, a practical version of the Liège INC model became available (Cugnon *et al* 1997a). The performances of these models, concerning Pb data, are examined in the next section. Before, it is worthwhile to briefly present the main differences between these codes.

In the BERTINI and ISABEL codes, the target is viewed as a continuum providing the travelling particles with a mean free path, related as usual to the total particle-nucleon cross-section. The free path is determined stochastically with an exponential function, after which the travelling particle can scatter with a nucleon promoted from the continuum as a new travelling particle. The procedure is resumed for the new travelling particle, and so on. Particles can escape from the nucleus whenever they hit the surface with a sufficiently large energy. The process is stopped when the kinetic energy of all particles inside the nucleus is below a given value (usually taken as 20 MeV above the Fermi energy). Each cascade is followed individually. In the ISABEL code however, a cascade influences the other ones by an average modification of the density of the continuum. The Liège model, on the contrary, possesses a time structure and follows the fate of all the particles. At the beginning, nucleons are positioned in space and momentum space according to spherical uniform distributions. Particles travel freely until two of them reach their minimum distance of approach d_{min}. They can scatter if $\pi d_{\min}^2 \leq \sigma_{tot}$. The process is resumed until some stopping time, t_{stop}, determined by the model itself : on the average (over events), the target excitation energy E* first increases drastically, then decreases rapidly for a while and further decreases on a much slower pace, on an evaporation-like pattern. This is illustrated in Fig. 3.9.1 for a typical case. The quantity t_{stop} is defined as the time when the slope of E* changes its value (i.e. at the knee of E*-plot). This time is however not sharply defined.

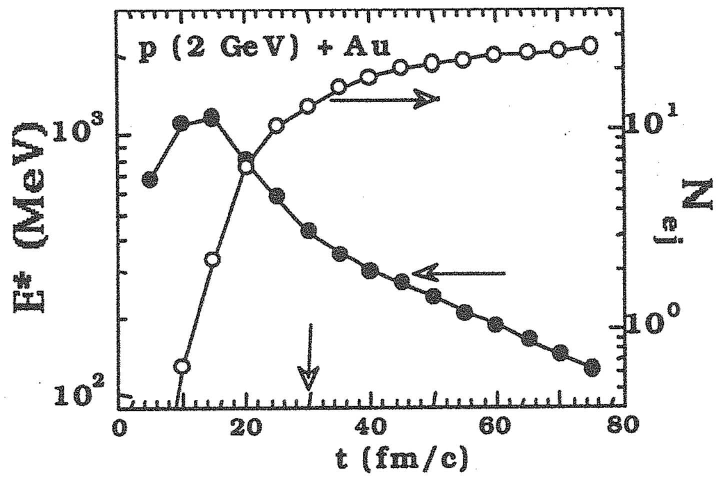

Fig. 3.9.1: *Evolution of the excitation energy (full dots, left scale) and of the number of ejectiles (open dots, right scale) in central p + Au collisions at 2 GeV. The vertical arrow indicates the value of the stopping time.*

Other differences pertain to the description of the nuclear surface, to the treatment of the Pauli blocking and to the parametrization of the nucleon-nucleon cross-sections. In the Liège INC code, referred to as *incl2* (Cugnon 1997a), a sharp nuclear surface is introduced, whereas the BERTINI and ISABEL models include a diffuse surface, described by a series of step functions. In these codes, the Pauli blocking refers to the original Fermi distribution, possibly depleted in the ISABEL code. In the Liège INC code, the Pauli blocking is introduced stochastically taking account of actual phase space population around the colliding particles, and not simply the original one. In other words, not only the depletion of the original Fermi distribution is taken into account, but also the population of higher states. Up-to-date parametrizations of the relevant hadron-hadron cross-sections (Cugnon *et al* 1996), especially the inelastic ones, are included in the Liège INC code. The latter is extensively described in (Cugnon *et al* 1997a).

3.9.3. Analysis of high-energy data

Neutron cross-sections

We will mainly concentrate on the comparison between the performances of the BERTINI and the Liège codes. We refer in the second case to the *incl3* version. The latter, developed in the course of this Concerted Action, differs slightly from the *incl2* version, as it is explained in section 3.9.4. We first consider the data recently obtained at SATURNE (Ledoux *et al* 1999) and described in section 3.6. A comparison of these data with the BERTINI and the Liège codes is given in Fig. 3.6.3. In both cases, the same Dresner evaporation model (Dresner 1980) has been used. In both cases also the same total reaction cross-section, taken as the experimental one (1720 mb), is adopted (see a discussion of this point in section 3.9.4). The theoretical predictions thus restrict to the distribution of the various observables. One can see an overall satisfying agreement, strongly indicating that the right dynamics is picked up in these models, namely the collision regime, characterized by a sequence of binary collisions, separated in time and occurring basically as in free space. The theoretical predictions differ nevertheless in some detail. There is a slight underestimation of the quasi elastic peak, i.e. the rightmost peak in the energy distribution at forward angles, a little more pronounced for the Liège cascade. The reason, in the last case, is very likely coming from the introduction of a sharp surface. A diffuse surface favours the importance of quasi-free events, i.e. events where the incoming proton changes charge and transforms into a neutron with very low momentum transfer, which escapes without further interaction.

In the BERTINI model, the quasi-inelastic peak, i.e. the peak located at the incident energy minus roughly 300 MeV, appears pathologically overestimated, whereas it is more or less reproduced in the Liège model. The difference can be traced back to the unrealistic parametrization of the inelastic differential $NN \rightarrow N\Delta$ cross-section used in the BERTINI code. On the contrary, the parametrization of the same quantity in the Liège model rests on the fitting of good recent measurements of the $np \rightarrow p+X$ cross-section (Cugnon *et al* 1997b).

A careful analysis of the multi collision component (i.e. between 20 MeV and 800 MeV in this particular case) reveals the following trends : (i) at very small angles, both models slightly overestimate the data (ii) at intermediate angles, the Liège model is slightly better and compares rather well with the data (iii) at large angles, both models underestimate the data. The last deficiency is probably inherent to the cascade approach: it is known for a long time that backscattering is enhanced by collisions with clusters inside the nucleus (Fujita 1977). The other deficiencies can perhaps be cured by refining the INC model, in particular by changing the NN angular distributions. This is legimate as in-medium effects are expected. However, these distributions cannot be changed at will, as there are theoretical constraints on these in-medium corrections, at least in the elastic channel (Cugnon *et al* 1987, Li and Machleidt 1994).

The analysis of the evaporation part (below ~20 MeV) is less obvious, as it also involves the evaporation module. However, using the same evaporation module with different INC codes allows a comparison between these codes. Clearly, the BERTINI model produces more evaporation neutrons, indicating that the average excitation energy left in the remnant after the cascade stage is much larger in the BERTINI model than in the Liège model. It seems that the BERTINI model is stopped earlier (or with a too high energy cutoff, see above) or is less able to evacuate energy by high energy particles. The self-consistent procedure used in the Liège approach seems more satisfactory and more accurate. Fig. 3.6.3 indicates that the Liège cascade is more successful than the BERTINI one, which overestimates the evaporation yield substantially. This does not allow however to conclude that the Liège code is superior. It only allows to conclude that the combination Liège + Dresner is rather accurate for the evaporative part of the neutron emission. A detailed study of the dependence of the results upon the evaporation code remains to be done It is also worth to mention that all the results for the Liège model have been obtained with the standard value of t_{stop}., as defined by the procedure described in Fig. 3.9.1.
A comparison of the same data with the ISABEL cascade (keeping the same evaporation model) is provided in Fig. 3.6.4. The predictions of ISABEL are quite similar to these of the Liège code. Slight differences are however noticeable. The ISABEL results are slightly better at forward angles, presumably because of the introduction of a diffuse surface. On the other hand, ISABEL underestimates the double differential cross-section at large angles and intermediate energy. This might originate from using less satisfactory parametrizations of the elementary cross-sections.

Examination of the cascade model results yields the same conclusions for energies at 800 and 1600 MeV when investigated by the SATURNE group. Other incident energies are much less documented. However, the success of the codes is similar for the data at 597 MeV (Amian *et al* 1992), 256 MeV (Stamer *et al* 1993) and 113 MeV (Meier *et al* 1989), although the agreement at small angles deteriorates when the incident energy decreases (Cugnon *et al* 1997a).

Neutron multiplicity

It is also instructive to analyze the neutron multiplicity data deduced from the differential data of (Ledoux *et al* 1999) by integrating over angle and energy. They are given in Table 3.6.1. Unfortunately, the experimental data of multiplicities in the

0-2 MeV range are not known. They are probably close to the usual extrapolation of the Maxwellian distribution fitted in the 2-20 MeV range. In the latter, neutron multiplicities are rather well predicted by the Liège model, in agreement with the observation above. Both the BERTINI and ISABEL models overestimate the neutron multiplicities. In the cascade range (20 MeV to beam energy), the Liège model reproduces the multiplicities rather well. The difference with the BERTINI and ISABEL predictions is not as important as in the 2-20 MeV range, but the latter are systematically smaller. These considerations are reinforced by the analysis of the neutron energy yield (right part of table 3.6.1), i.e. the integrated product of the neutron yield and the neutron kinetic energy. In this case, it appears more clearly that the BERTINI and ISABEL models are unable to evacuate energy by emission of fast particles and that the remaining excitation energy at the end of the cascade stage is too large (note however that this statement is subject to an assessment of the Dresner evaporation code in the excitation energy range under consideration here, i.e. for $< E^* > \approx 100-150 MeV, E^*_{max} \approx 300-400 MeV$). The possible explanation of this observation is not obvious. It may originate from the less reliable parametrization of the nucleon-nucleon cross-sections in these two codes but it most probably pertain to the stopping criterion. This issue is however difficult to settle, as this criterion is completely different in these two codes from the one used in the Liège model. An interesting discussion concerning this point can be found in (Cugnon *et al* 1997a). It suggests that the criterion used in the ISABEL and BERTINI codes amounts roughly to stop the cascade in the Liège model earlier than the standard stopping time. In addition, the criterion used in the first two codes depends upon the history of the actual event and is thus subject to large fluctuations. This might explain the higher value of the excitation energy reached in these models.

It is also worthwhile mentioning that the observed neutrons carry only about one fourth of the available energy. If one adds the separation energy (for neutrons), the fraction of the available energy rises to a value between one third and one half. It would thus be highly desirable to have precise information on the way the remaining energy is distributed, i.e. on how this energy is distributed among the possibly outgoing incident proton, the emitted light charged particles and the produced pions.

Neutron multiplicities have also been measured directly with the Berlin neutron ball (Enke *et al* 1999), as reported in Section 3.8. Analysis is still in progress, but final results at 1.2 GeV agree with those of SATURNE (see tables 3.6.1 and 3.8.2), as they do with the calculated values in the Liège model, associated with either the Dresner or the GEMINI evaporation codes. On the other hand, they disagree with the neutron multiplicity predicted by LAHET (with the BERTINI code).

Light charged particle emission

Very few data exist in the high energy range. For a Pb target, double differential cross-sections have been measured at 800 MeV and 2100 MeV (Tanihata *et al* 1981) at a few angles and covering only the high energy part of outgoing energy. Recently, multiplicity measurements have been made by the NESSI Collaboration (Enke *et al* 1999), as reported in Section 3.8. Comments will be made only on the H and He yields. They are reproduced in table 3.9.1, for the case of 1.2 GeV incident energy and compared with theoretical predictions, on which the experimental filter has been applied. The analysis of the comparison is instructive.

Table 3.9.1: *Particle multiplicities for p (1200 MeV) + Pb reactions.*

	Exp	Liège + GEMINI	LAHET
n	17.5 ± 2.5	17.4	17.5
H	1.31 ± 0.1	1.61	3.6
He	0.7 ± 0.1	0.70	1.61

For the LAHET predictions, the BERTINI code is used, with the Dresner evaporation module and a preequilibrium module (Prael and Bozoian 1988). The H and He yields are clearly overestimated while the predictions of the Liège code agree quite well with the experimental data. This clearly indicates that particle emission is too much enhanced by the preequilibrium stage (although some of the differences can be traced back to the use of different Coulomb barriers in the two calculations). This was already pointed out in (Cugnon *et al* 1997a). However, in the Liège + GEMINI approach light charged particles are emitted during the evaporation stage only. This clearly contradicts the ubiquitous observation of so-called preequilibrium composites with energy larger than typical evaporation energies. From the measurements of (Enke *et al* 1999), one can evaluate the preequilibrium component for He production, as the difference, on the high energy side, between the experimental and theoretically predicted (Liège + GEMINI) He spectra, after integration over angles. One finds $\sigma^{He}_{p-eq}/\sigma^{He}_{tot} \approx 0.12$. This gives an idea of the contribution of "preequilibrium" particles. Note however that the same ratio for energy-integrated spectra, which is perhaps more indicative, is equal to roughly twice this value.

Residue production

Most of the existing data, discussed in Section 3.2 are based on radiochemical methods. Even though they are of paramount importance for radioprotection or other topics, they do not provide definite and easily exploitable constraints for dynamical models. First, stable isotopes are not detected by these methods. Second, what is basically measured is the cumulated yield, integrating automatically the decay of parent isotopes with lifetimes smaller than a few hours at least. The situation has changed drastically, during this Concerted Action, with the inverse kinematics measurements described in Section 3.7. The latter basically provide the isotope yields after the evaporation stage, before further $\beta-$ and $\gamma-$decays.

The analysis and the comparison with codes are still in progress and it is premature to draw conclusions. Nevertheless, first results are given in (Mustapha 1999). In this reference, it is shown that although the Liège model gives a good overall description of the data, it definitely underpredicts the peak in the isotopic yield, for the isotopes close to the target (see Fig. 3.7.8). This should be attributed to the lack of a diffuse surface in the Liège model, as we already mentioned in section 3.9.2.

3.9.4. Theoretical progress

Most of the theoretical work in this Concerted Action has been devoted to the evolution of the Liège INC from the *incl2* version, extensively described in (Cugnon *et al* 1997a), to the *incl3* version, for which a detailed description can be found in (Cugnon and Henrotte 2000). Here only a brief account of the main modifications is given.

The first one deals with the excitation energy. In the Liège INC model, the excitation energy is computed as the difference between the actual kinetic energy contained in the target volume and some reference energy, basically the energy of a cold Fermi gas contained in this volume. However, the Pauli principle is implemented stochastically. This means that, as long as the evaluated phase space density is not unity, a collision is always possible. It is then understandable that, due to fluctuations in the phase space distributions of nucleons and due to this stochastic handling of the Pauli blocking, nucleons may accumulate in the potential well with energy smaller than the nominal value for a Fermi gas, although this occurs rather unfrequently. In the *incl3* version, the excitation energy is checked at any time and the event is terminated prior to t_{stop} when E* vanishes. As a consequence of this modification, the E* spectrum is slightly harder. It turns out that this is sufficient to improve the comparison with the SATURNE experimental data.

The second modification is related to the description of inelastic np scattering. The recent data obtained at SATURNE concerning $np \rightarrow p+X$ allowed to extract the angular distribution of the $np \rightarrow p+\Delta^0$ cross-section (Cugnon *et al* 1997b). The latter has been parametrized, in the forward hemisphere, as

$$\frac{d\sigma}{d\Omega} \propto e^{B_{in}t}$$

where t is the squared momentum transfer. The extracted parameter B_{in} varies with the neutron incident momentum, as given in Fig. 3.9.2. The non zero value indicates a forward peaked distribution. In comparison, the distribution for the same reaction is taken in the BERTINI model as totally concentrated at zero degree. In the presentation of Fig. 3.9.2, this would correspond to an infinite B_{in} parameter.

Another effort aimed at understanding the boundaries of the domain of validity of the INC + evaporation model, especially on the low energy side. An indicative discussion of this topic can be found in (Cugnon 2000). In parallel, an empirical approach has been pursued, looking simply at the performances of the model at lower and lower energy. It was found that the predictions compare surprisingly well with neutron double differential cross-sections at incident energy well below 200 MeV, although the agreement is not as impressive as in Fig. 3.6.3. An example is given in Fig. 3.9.3 at 80 MeV incident energy. A comment is however in order. In this calculation, the Pauli principle is handled as in the BERTINI model, i.e. all collisions leading to a momentum smaller than the Fermi momentum are forbidden. The ordinary implementation of the Pauli principle deteriorates the description at the smallest angle for high outgoing energy.

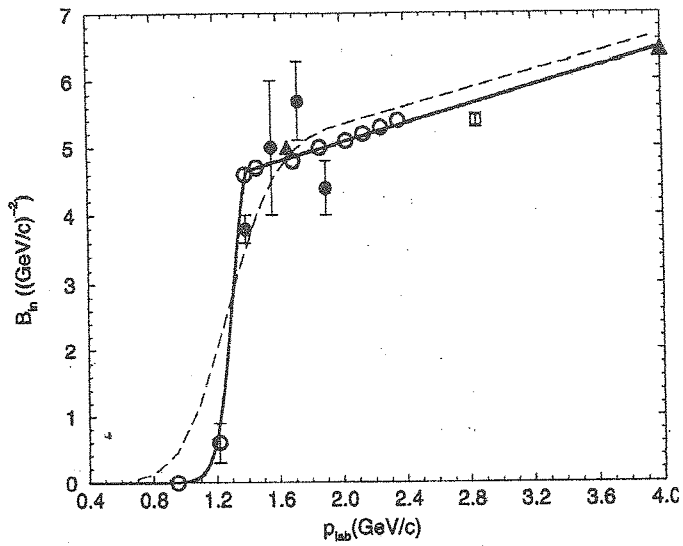

Fig. 3.9.2: *Slope parameter B_{in} for the angular distribution of the Δ production in np collisions. Symbols indicate data from (Laville 1976, black dots), (Bugg et al 1964, triangle), and (Cugnon et al 1997b, open symbols). The full line corresponds to the parametrization used in the Liège incl3 code.*

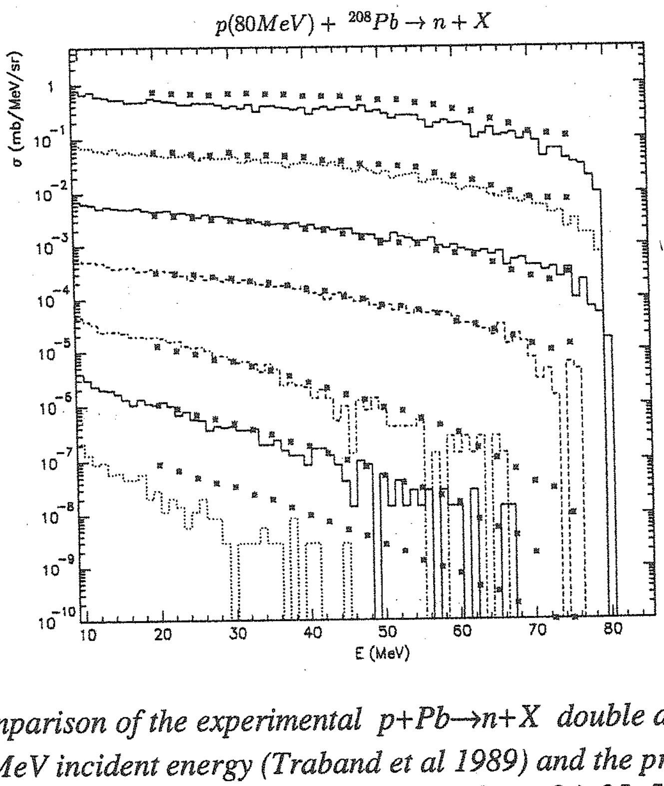

Fig. 3.9.3: *Comparison of the experimental p+Pb→n+X double differential cross-section at 80 MeV incident energy (Traband et al 1989) and the predictions of the Liège + Dresner model (Henrotte 2000). The data refer to 24, 35, 56, 69, 95, 106 and 144 degrees, from top to bottom.*

3.9.5. Discussion of the accuracy of theoretical descriptions

We here address the question of whether the kind of agreement obtained above is sufficient or not to guarantee a useful utilisation of the theoretical models in technological applications, in particular to future ADS. This is not an easy question as many aspects of the spallation reactions are involved and as no firm requirement has been formulated from the engineers in charge of the conception of ADS or other devices. The discussion will here be restricted to neutron data.

The theoretical models are to be used in transport studies for the prediction of the neutron flux outgoing from the spallation target of the ADS. These results in turn are to be used to design the reactor of the ADS, in particular to foresee the value of the parameter k_{eff} , the distribution of the power inside the reactor, etc, with a reasonable accuracy. According to some experts (Salvatores 1996), this requires the knowledge of the integrated neutron flux going out of the spallation source with an accuracy better than 10 % and the distribution (angular, positional) of the flux everywhere better than 30 %. In turn, one may wonder what is the accuracy required in thin target calculations to guarantee this kind of accuracy for thick target transport calculations. There is no clear answer to this question at this moment, as no systematic sensitivity investigation is really available. Indications are however given in recent works. As for the total multiplicity, the new thick target data of (Letourneau *et al* 2000), with error bars of the order of 2 %, seem to be well described, for a large range of target thicknesses, by three transport codes with different high-energy modules (i.e. cascade + evaporation). This is perhaps not surprising since above say 20 cm thickness, all the neutrons produced in high-energy collisions are largely diffusing in the spallation target and their number is largely determined by the available energy and by the absorption properties of the material at low neutron energy. The theoretical description of the angular distribution of the outgoing neutrons, measured with a good accuracy at SATURNE (Ménard *et al* 1998), seem however more sensitive to the high energy module of the transport code (see Section 3.6 and Fig. 3.6.5), as explained in (Casoli 1999). In conclusion, one may consider, as a starting point, that demanding 10 % and 30 % accuracy for multiplicity and angular distribution in thick targets, respectively, requires the same kind of accuracy in thin target quantities. Notice that the use of liquid metal targets will likely allow to reduce their size and this statement is even more true for this choice.

If one considers the SATURNE measurements, the neutron multiplicity data are given with an error of 10 % and they are reproduced by the Liège cascade winthin this error bar. Note however that the energy weighted multiplicity is a more relevant quantity, since more energetic neutrons will produce more secondary neutrons. This quantity is then satisfactorily reproduced by the three calculations referred in Table 3.6.1. As for angular distributions, even if the agreement is quite impressive in Figs. 3.6.3 and 3.6.4 for the Liège and ISABEL models, one has to recognize that the 30 % accuracy criterion is not met everywhere. Let us also mention that the situation at incident energy lower than 800 MeV is by no means better.

3.9.6. Conclusion

The agreement of the INC + evaporation model predictions with the Pb data at high energy is quite impressive in view of the complexity of the spallation processes. It is however not satisfactory enough in view of the accuracy requirements in technological applications. The comparison of the Pb data with the INC models (Liège, ISABEL, BERTINI) seem to rule out the last one. The slightly better agreement reached by the Liège + Dresner evaporation, compared to ISABEL, can be interpreted as due to the better handling of Fermi motion and Pauli blocking and to a better parametrization of elementary cross-sections, in the *incl3* version of the Liège code, developed in the course of this Concerted Action. However, the lack of surface diffuseness in this version has been recognized as the main reason for a deficient description of the quasi-elastic region. The introduction of a surface in the Liège INC model will be performed in the frame of the HINDAS project. Let us also mention that the present study points toward a validity of the INC + evaporation approach at much lower incident energy than previously expected.

3.9.7. Publications related to the project

1. Cugnon J., Spallation Reactions, Ann. Phys. Fr.25 N° 2, 93

2. Cugnon J. and Henrotte P., The Liège Intranuclear Cascade Model, to appear in the Proceedings of the SARE5 Meeting, OECD Publications