NEW DATA AND MONTE CARLO SIMULATIONS ON SPALLATION REACTIONS RELEVANT FOR THE DESIGN OF ADS

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Abstract

The main European experimental programs to characterise spallation reactions used as neutron sources are reviewed. The neutron production is described in terms of the multiplicities, spatial and energy distributions. Experiments to determine the residual nuclei production in the spallation target are also discussed. These data are used to benchmark existing nuclear model calculations.

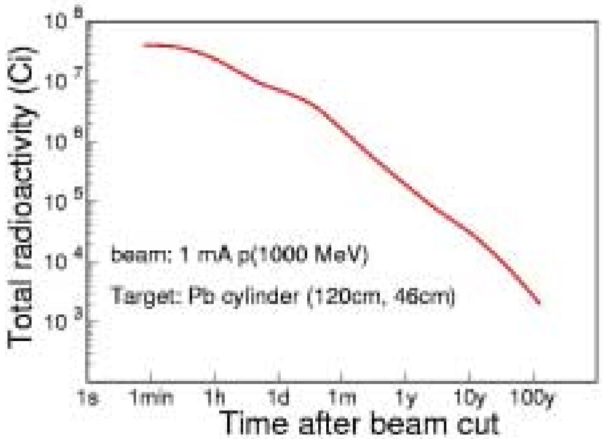
1. Introduction

Nowadays it is well established that spallation reactions constitute an optimum neutron source to feed a sub-critical reactor in an accelerator driven system (ADS). However, the present knowledge about this reaction mechanism is not accurate enough for any technical application. Two main aspects will play a major role in the design and construction of the target assembly of the spallation neutron source used in an ADS: the neutron yields and the residual nuclei produced in the reaction.

The neutron production should be characterised in terms of the neutron multiplicity and their spatial and energy distributions. The neutron multiplicity will determine the intensity of the proton-driver accelerator while their energy and spatial distribution should be considered to design the geometry of the spallation target and the shielding to high-energy neutrons.

Spallation reactions do not only produce neutrons but also residual nuclei. Most of these nuclei are radioactive, therefore, activation problems should be considered in the design of the target. In Figure 1 we report an example of the simulated activity induced in a cylindrical lead target by a 1 mA proton beam. As it is showed in the figure, both the cooling time and the total activity induced in the target are not negligible. In addition the residual nuclei will contribute to the corrosion of the target and to the radiation damages in the target, accelerator window and structural materials.

Figure 1. **Calculated radioactivity induced in a cylindrical lead target (120 cm long and 46 cm diameter), by a 1 GeV proton beam of 1 mA after one year of irradiation. Calculations done with the Lahet Code system by D. Ridikas [1].**



Although spallation reactions are understood qualitatively, they are not known with the degree of accuracy needed for any technical application. In this sense, most of the existing codes used to describe these reactions have a limited predictive power. Therefore a large experimental program has been initiated in Europe few years ago in order to improve our knowledge on these reactions. These experiments will provide accurate data to benchmark more reliable model calculations. In the following sections we will describe some of these experiments.

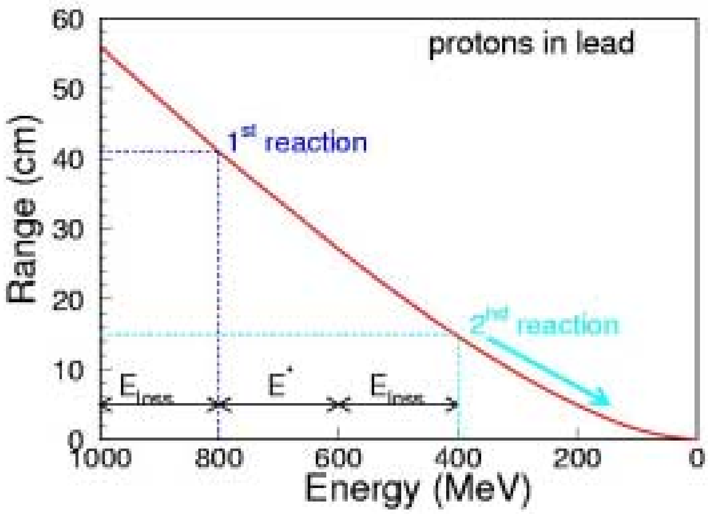
2. General considerations on spallation reactions

Spallation reactions are collisions induced by light-energetic projectile on a heavy-ion target. These reactions can be described as a two-stage process. First the incoming projectile induces quasi-free nucleon-nucleon collisions with the nucleons of the target nucleus. These collisions lead to the prompt emission of few neutrons and protons. A fraction of the kinetic energy of the incoming projectile will be transferred to the target nucleus as excitation energy, e.g. a 1 GeV proton deposits on average 200 MeV in the target nucleus. The rest of the energy will be shared between the prompt emitted nucleons. This emission of fast nucleons will play an important role in the development of an inter-nuclear cascade process inside the target.

In a second step the residual nuclei produced in the collisions will de-excite by evaporation of low energy protons and neutrons or fissioning. In principle, neutron evaporation is favoured since to evaporate protons or to fission the system needs extra energy to overcome the Coulomb barrier. The energy of the evaporated nucleons is determined mainly by the temperature reached by the residual nucleus in the collisions and will be in the range of a few MeV.

To describe the full interaction of a relativistic projectile with a target material we should consider that the most probable interaction of this projectile with the target material will be governed by electromagnetic processes. The main consequence of the electromagnetic interaction of the projectile with the electrons of the target material will be the slow down of the projectile and heat load of the target.

Figure 2. **Range of protons in lead as a function of their energy**



The nuclear interaction between the projectile and the target is determined by the total reaction cross-section. In the case of the reaction proton on lead at 1 GeV the reaction cross-section corresponds to a mean free path of protons on lead around 15 cm. In contrast, the mean free path for electromagnetic interaction is much shorter, consequently the incoming projectile will be slowed down before any nuclear interaction. The electromagnetic interaction can be characterised in terms of the range of the incoming particle in the traversed medium. In Figure 2 we represent the range of protons in lead as a function of their energy. As can be seen the range of a proton with 1 GeV in lead is around 55 cm.

In order to describe the inter-nuclear cascade inside the target we should estimate the energy balance in the interaction of the projectile with the target. If we consider that on average the nuclear interactions take place at 15 cm, the mean energy loss of the incoming projectile before the reaction will be 200 MeV. In addition the energy dissipated in the first spallation reaction is around 200 MeV. This excitation energy leads to a large population of different residual nuclei. The remaining kinetic energy ≈ 600 MeV will be shared between the four or five prompt nucleons emitted during the first stage of the reaction. These nucleons will lead to secondary reactions in the target (inter-nuclear cascade).

The maximum energy of the prompt emitted nucleons is expected to be lower than 300 MeV. According to Figure 2, at this energy the range of protons in lead is few centimetres, therefore most of the secondary protons will be stopped before they induced any secondary reactions. The inter-nuclear cascade will be then induced mainly by neutrons with energies lower than 300 MeV. At this energy the spallation reaction is less violent and only few nucleons will be produced with an energy range lower than 100 MeV. Consequently the final reaction residues will be very close in mass and atomic number to the target nucleus.

In summary we can conclude that an incoming proton at 1 GeV on a lead target will induce on average two spallation reactions. The first one at high energy will determine mostly the residual nuclei produced in the target. The second reaction at lower energy will contribute to the multiplication and moderation of the neutrons.

3. Neutron production in spallation reactions

The neutron yield produced in spallation reactions will depend strongly on the projectile-target combination. In principle the heavier the target nucleus the larger the neutron excess leading to a larger neutron yield. Nevertheless the gain factor between heavy and light targets is not larger than a factor of five. In contrast, the radiotoxicity induced in the spallation target can be drastically reduced when using lighter targets as discussed in [2].

In addition to the neutron yields, reliable information on the energy and spatial distributions of the neutrons is required. This information can be used in the design of the spallation-target assembly geometry or the shielding to high-energy neutrons.

Neutron detection is not an easy task since neutrons only feel the strong interaction. This is the reason why different experimental devices are needed to characterise the neutron production in spallation reactions. In the following we will consider two examples.

3.1. Measurement of neutron yields

Neutron multiplicities can be investigated using liquid-scintillator based detectors with a large angular acceptance. A clear example is the detectors BNB (Berlin Neutron Ball) [3] and ORION [4] used by the NESSI collaboration (Berlin-Ganil-Jülich). This collaboration has performed a large experimental program to determine the neutron yields produced in thin and thick targets for a large range of primary projectiles and energies. To fulfil this programme experiments were done at GANIL (France) [4], Jülich (Germany) [3] and CERN (Switzerland) [5].

Figure 3. **Average neutron multiplicity per incident proton as a function of target thickness and beam energy for Pb, Hg and W materials obtained by the NESSI collaboration [3]**

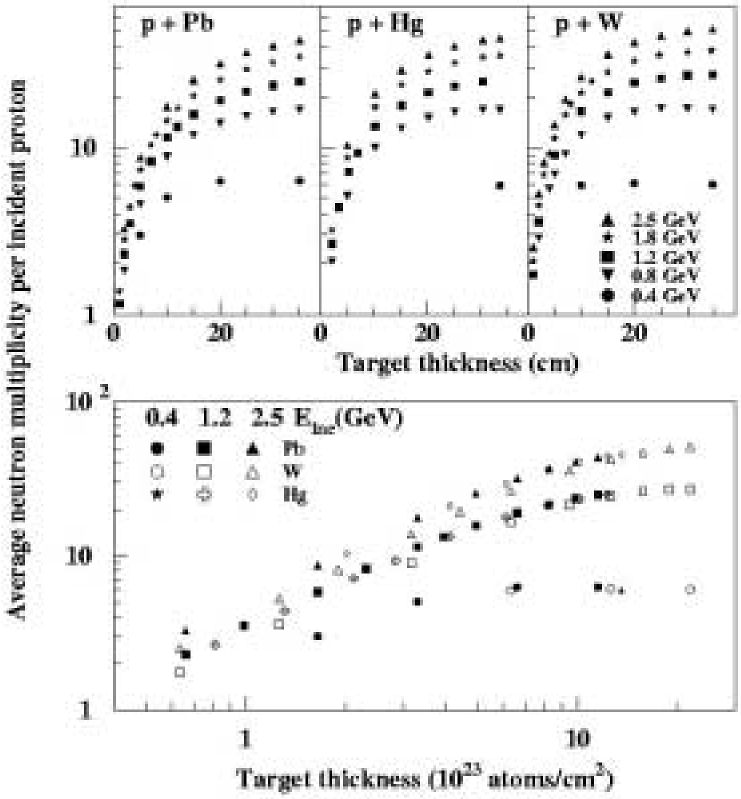


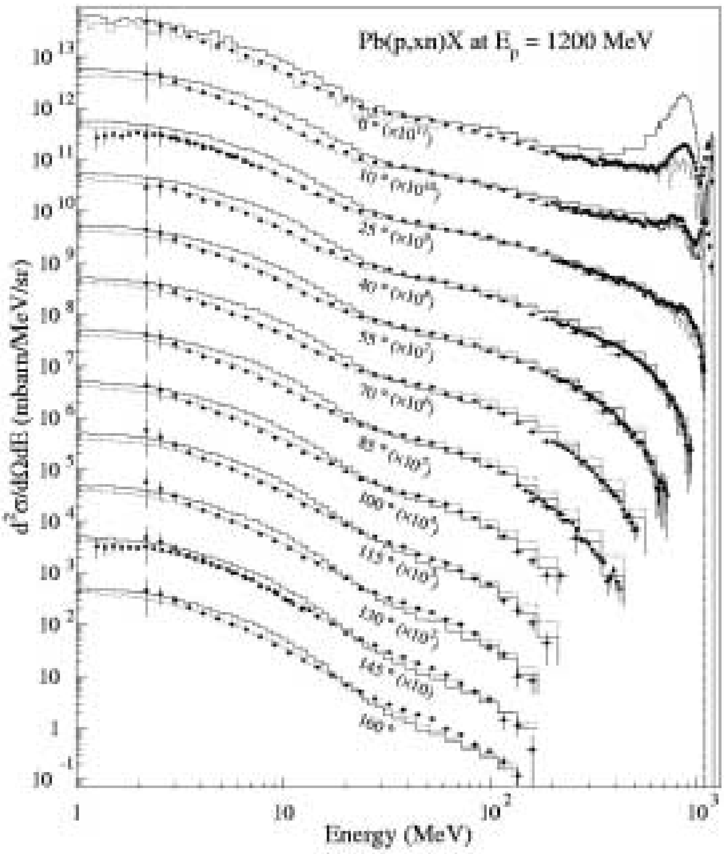
Figure 3 shows some representative results obtained by this collaboration at Jülich with the BNB detector. This figure represents the measured average neutron multiplicity per incident proton as a function of target thickness and beam energy for Pb, Hg and W materials. As can be seen, for the different target materials, the neutron multiplicity saturates at a given target thickness which increase with the proton energy. This saturation corresponds to the previous picture where every incident proton originates on average two collisions with a mean free path of 15 cm.

3.2. Energy and spatial distribution of neutrons

Specific experimental set-ups are needed to measure the spatial and energy distribution of the neutrons produced in spallation reactions. A clear example is the experiments performed by the "transmutation" collaboration at Saturne (France). These measurements use two different experimental techniques to cover the full energy range of the neutrons produced in the reaction. The detection of neutrons with energies lower than 400 MeV was based in a measurement of their time of flight between the incident proton beam, tagged by a plastic scintillator, and a neutron-sensitive liquid scintillator [6]. Neutrons with higher energies were measured using (n,p) scattering on a liquid hydrogen converter and reconstruction of the proton trajectory in a magnetic spectrometer [7]. An additional collimation system allowed determining the angular distribution of the neutrons.

This experimental technique was used to investigate the neutron production in reactions induced by protons with energies between 0.8 and 1.6 GeV on thin and thick lead targets. In Figure 4 we report some of the results obtained with a 1.2 GeV proton beam on a two centimetres thick lead target. This kind of measurements allow to characterise the spallation process. High energy neutrons emitted at low angles are representative of the first stage of the collision while low energy neutrons emitted isotropically correspond to the evaporation phase. Measurements done with thicker targets are representative of the inter-nuclear cascade leading to the multiplication and moderation of neutrons.

Figure 4. **Neutron production double-differential cross-sections measured in 1.2 GeV induced reactions on a 2-cm thick Pb target [8]. The histograms represent calculations using the Bertini INC Code [9] while the dotted lines corresponds to calculation done with the Cugnon INC Code [10].**



4. Residue production in spallation reactions

Residue production in spallation reactions can be investigated using two different experimental approaches. In the standard one, the reaction is induced in direct kinematics, the light-energetic projectile hits a heavy target. In this case the recoil velocity of the residues produced in the reaction is not sufficient to leave the target and γ-spectroscopy or mass spectrometry techniques are used to identify those residues. The main limitation of this technique is that for most of the residues the measurement is done after β decay and consequently only isobaric identification is possible.

Better suited seems to be the measurement of the spallation residues in inverse kinematics. In this case the heavy nucleus is accelerated at relativistic energies and impinges a light target. Due to the kinematical conditions, the reaction residues leave easily the target and can be identified in a short time using the appropriate technique.

4.1 Measurement of residue production in inverse kinematics

One of the most outstanding experiments are the ones performed by a German-Spanish-French collaboration at GSI. The technique used in these experiments takes advantage of the inverse kinematics and the full identification in mass and atomic number of the reaction residues by using a magnetic spectrometer.

The experiments have been performed at the SIS synchrotron at GSI. Primary beams of ^{197}Au, ^{208}Pb and ^{238}U accelerated up to an energy of 1 A GeV impinged on a liquid hydrogen or deuteron target. The achromatic spectrometer FRS [11] equipped with an energy degrader, two position sensitive scintillators and a multisample ionisation chamber allowed to identify in atomic and mass number all the reactions products with half lives longer than 200 ns and with a resolving power of $A/\Delta A \approx 400$. Figure 5 represents an example of the resolution achieved with this experimental technique. The final production cross-sections are evaluated with an accuracy around 10%. In addition, the magnetic spectrometer allows determining the recoil velocity of the reaction residues. This information is relevant for the characterisation of the damages induced by the radiation in the accelerator window or the structural materials. More details about these experiments can be found in [12-15].

Figure 5. **Example of identification matrix obtained with the Fragment Separator at GSI [13]**

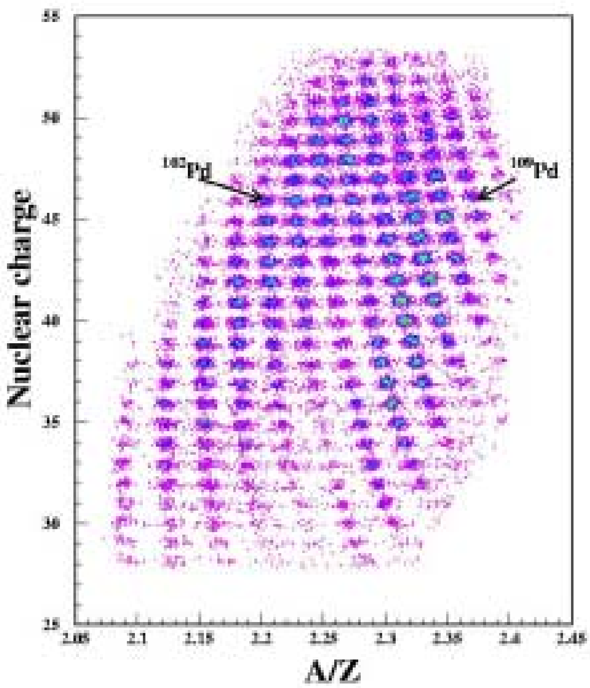
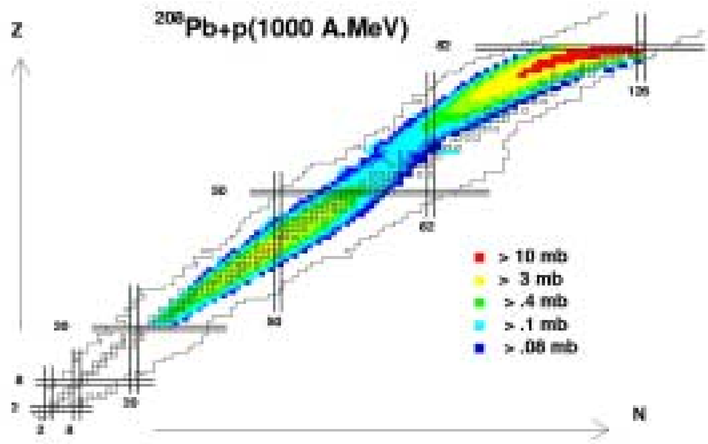


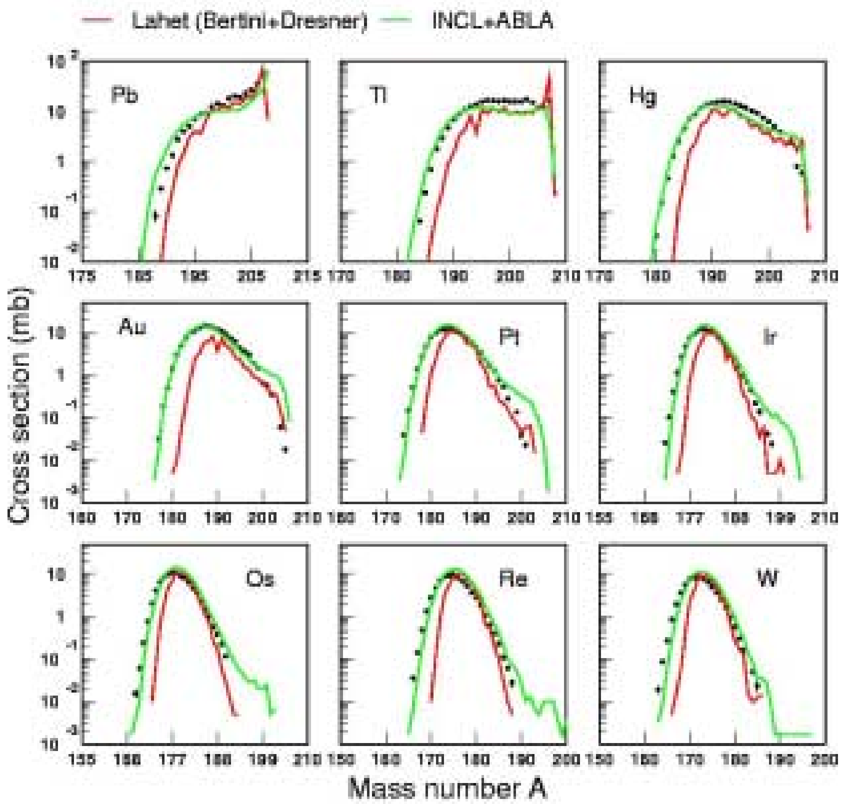
Figure 6. **Two-dimensional cluster plot of the isotopic production cross-sections of all the spallation residues measured at GSI in the reaction ^{208}Pb(1 A GeV) + p shown as chart of nuclides [15]**



In Figure 6 we present in a chart of the nuclides all the residues measured in the reaction ^{208}Pb(1 A GeV) + p. More than 1 000 different spallation residues were identified in this reaction. As can be seen in this figure, the spallation residues populate two different regions of the chart of the nuclide. The upper region corresponds to the spallation-evaporation residues which populate the so-called evaporation-residue corridor. The second region populates medium-mass residues produced in spallation-fission reactions. Both reactions mechanism, fission and evaporation, should be considered to describe the production of spallation residues in these reactions.

The measured isotopic production cross-sections for some selected elements are presented in Figure 7. This figure shows clearly the quality of the measured data that can be used to benchmark any model calculation.

Figure 7. **Isotopic production cross-sections for some of the elements produced in the reactions ^{208}Pb + p at 1 A GeV [15]. The data are compared with two model calculations, the dark line correspond to the results obtained with the Lahet Code [16] while the hell line was obtained with the intra-nuclear cascade model of Cugnon [10] coupled to the evaporation-fission Code ABLA from GSI [17,18].**

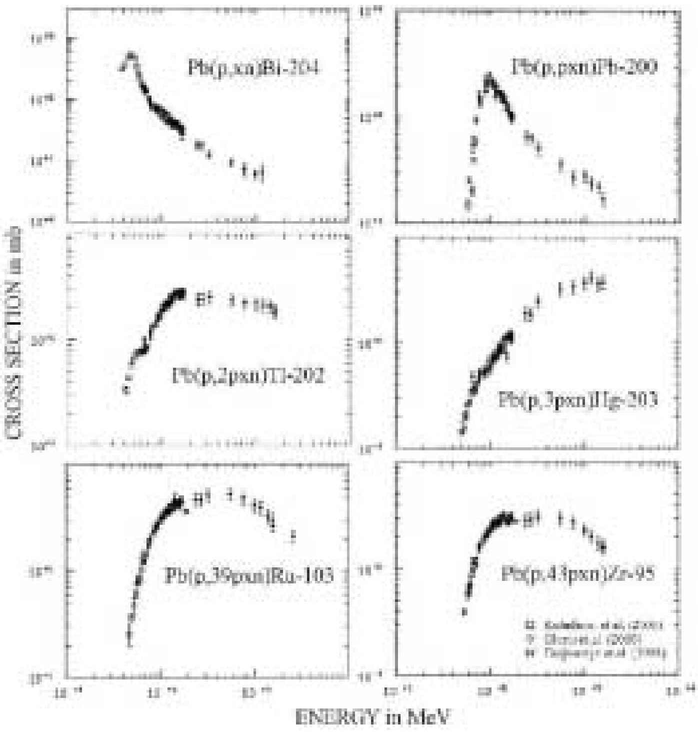


4.2 *Measurement of residue production in direct kinematics*

Although this method only allows isobaric identification after β-decay, for some shielded isotopes it is possible to determine their primary production cross-sections. In principle this experimental technique is less beam time consuming than the inverse kinematics. Therefore full excitation functions can be established for selected isotopes as shown in Figure 8. In addition this method can be applied to thin and thick targets.

From the results shown in Figure 8 we can conclude that the low energy reactions produced mainly residues close to the target nucleus, while most of the reaction residues populating a large part of the nuclear chart are produced by energetic particles. The two most important experimental programs in Europe using this technique are the ones performed by the group of R. Michel at the University of Hanover [19] and Y.E. Tiratenko at the ITEP in Moscow [20].

Figure 8. **Excitation functions for some selected isotopes produced in the interaction of protons with lead measured with γ-spectroscopy techniques [21]**



5 Reactions in the 20-200 MeV energy range

Reactions induced by neutrons and light-charged particles in the energy range between 20 and 200 MeV are representative of the inter-nuclear cascade in the spallation target. This reactions will play a major role in the multiplication and moderation of the neutrons. The energy dissipated in these reactions leads to the emission of few particles and consequently only residual nuclei close in mass and atomic number to lead will be produced.

These experiments intend to measure the double-differential production cross-sections of neutrons and light-charged particles. It is out to the scope of this work to review on all the experimental programs investigating these reactions. Most of them contribute to the Hindas project of the Fifth Framework Programme of the European Commission. The experiments take advantage of a large network of European facilities delivering protons and neutrons in the investigated energy range: KVI (Netherlands), Louvain-la-Neuve (Belgium) and Uppsala (Sweden). More detailed information about this program can be found in the contributions of N. Marie, F.R. Lecolley and J.P. Meulders to this conference.

6. Model simulations

Most of the existing models to simulate spallation reactions describe the first stage of the collision in terms of semi-classical nucleon-nucleon collisions (intra-nuclear cascade) and a statistical de-excitation of the hot residue. The main inputs of the intra-nuclear cascade are the elastic and inelastic nucleon-nucleon cross-sections and the distribution of the nucleons in the target nucleus in position and momentum space. The statistical evaporation of particles is generally based in the Weisskopf formalism while fission can be describe according to the prescription of Bohr. In this case the main parameters are the description of the level density and the Coulomb barriers for charged-particles emission or fission. Another critical parameter is the coupling time between the intra-nuclear cascade and the evaporation.

The last model intercomparison done by NEA [22] revealed important deficiencies in most of the existing codes to describe spallation reactions. In fact these deficiencies can be understood due to the lack of experimental information. The new data provided by the present experimental programs will help to improve this situation. In Figure 7 we compare the measured isotopic production cross-sections for some of the elements produced in the reactions ^{208}Pb + p at 1 A GeV at GSI [15] with two model calculations. In this figure, the dark line correspond to the results obtained with the Lahet Code (Bertini + Dresner) [16] while the hell line was obtained with the intra-nuclear cascade model of Cugnon [10] coupled to a new the evaporation-fission Code ABLA from GSI [17,18]. As can be seen, the new models provide a much better description of the experimental data.

7. Conclusions

Spallation reactions are considered as an optimum neutron source to feed an ADS. However this reactions are not known with the degree of accuracy needed for the design of such devices. This is the main justification for a large experimental program initiated in Europe few years ago to collect high quality data about neutron and residual nuclei production in these reactions. This experimental program takes advantage of most of the existing heavy-ion facilities in Europe in order to cover the full energy range involved in the interaction of light-energetic projectiles with heavy-ion targets. Most of these programs are supported by different programs of the European Commission like Hindas or the European Spallation Source (ESS).

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