Basic Nuclear Data at High and Intermediate Energy for Accelerator-Driven Systems

P. Armbruster[a], *J. Benlliure*[b]

[a]Gesellschaft für Schwerionenforschung, Planckstr. 1, D-64291 Darmstadt, Germany
[b]Universidad de Santiago de Compostela, E-15706 Santiago de Compostela, Spain

1 General considerations on spallation reactions needed for ADS

Nowadays it is well established that spallation reactions constitute an optimum neutron source to feed a subcritical 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 current and beam energy of the proton-driver accelerator while their energy and spatial distribution shapes 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. Figure 1 represents the simulated activity induced in a cylindrical lead target by a 1 mA proton beam after one year of irradiation. As can be seen in the figure, both the cooling times and the total activities induced in the target are not easy to be handled. In addition the residual nuclei will contribute to the corrosion and to the radiation damages in the target, accelerator window and structural materials.

The main consequence of the present qualitative understanding of spallation reactions is that 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 are expected to provide accurate data to benchmark more reliable model calculations. In the following sections we will describe the highlights of this experimental program.

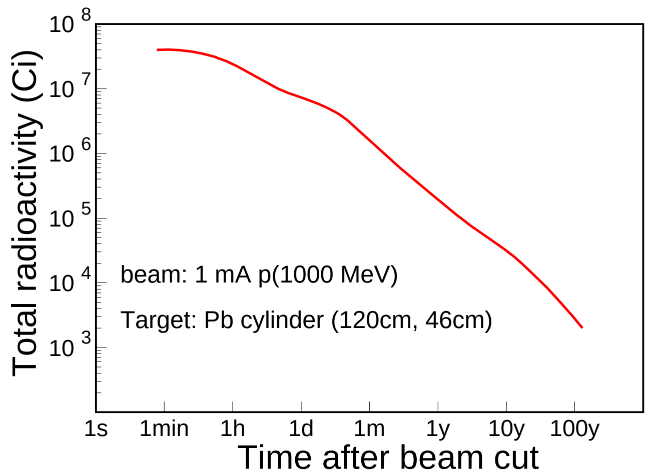

Figure 1: *Calculated radioactivity induced in a cylindrical lead target (120 cm long and 46 cm diameter), by a 1 GeV proton beam of 1 mA after one year of irradiation. Calculations done with the Lahet code system [1].*

2 Interaction of relativistic protons with thick targets.

Spallation reactions are induced by light projectiles at relativistic energies impinging on a heavy target. These reactions can be described as a two stage process. First the incoming projectile interacts in quasi-free nucleon-nucleon collisions with the nucleons of the target nucleus. These collisions lead to the prompt emission of few high energy nucleons while a fraction of the kinetic energy of the incoming projectile is transferred to the target nucleus as excitation energy (e.g. a 1 GeV proton is expected to deposit on average 200 MeV in the target nucleus leading to the emission of around five prompt nucleons). These 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 deexcite by evaporation of low energy neutrons and protons or by fission. Neutron emission is favoured since to evaporate protons or to fission extra energy is needed to overcome the Coulomb or fission barriers. Around 10 additional neutrons are expected to be produced in this second stage of the reaction. The energy of the evaporated nucleons is determined mainly by the temperature reached by the residual nucleus in the primary 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 recall that the most probable interaction with the target material will be governed by electromagnetic processes. The main consequence of this electromagnetic interaction will be the slowing down of the projectile and the heat load of the target.

The nuclear interaction between the projectile and the target is determined by the total nuclear reaction cross section. The total reaction cross section for collisions induced by 1 GeV protons on lead corresponds to a mean free path of protons in lead of about 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 is characterised in terms of the range of the incoming particle in the traversed medium. The range of a proton with 1 GeV in lead is around 55 cm.

The inter-nuclear cascade inside the target will be determined by the energy balance of the interaction of the projectile with the target. Considering that on average the nuclear interactions take place at 15 cm inside the target, the mean energy loss of the incoming particle 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 $\approx$ 600 MeV will be shared between about five prompt nucleons emitted during the first stage of the reaction. The prompt neutrons mainly will lead to secondary reactions in the target (inter-nuclear cascade). Consequently, a 1 GeV proton impinging 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 $\approx$ 200 MeV will produce residues very close in mass and atomic number to the target nucleus, but will contribute to the multiplication of the neutrons and soften their spectrum.

3 Neutron production in spallation reactions.

The neutron fluxes produced in spallation reactions will depend strongly on the projectile-target combination. In principle the heavier the target nucleus the larger the neutron excess and the larger the neutron yield. The gain factor between heavy and light targets is around a factor of five. However, the radiotoxicity induced in the spallation target could be drastically reduced when using lighter targets [2]. In addition to the neutron yields, reliable information on the energy and spatial distributions of the neutrons is required. Different experimental devices are needed to characterise the neutron production in spallation reactions.

3.1 Measurement of neutron yields.

Neutron multiplicities can be investigated using liquid-scintillator based detectors with a large angular acceptance. Clear examples are 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 de-

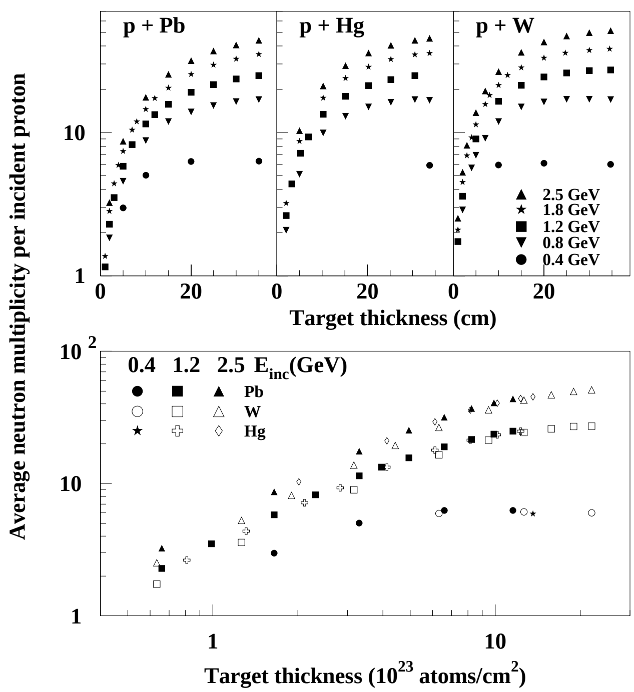

Figure 2: *Average neutron multiplicity per incident proton as a function of target thickness and beam energy for Pb, Hg and W materials obtained by the NESSI collaboration [3].*

termine the neutron yields produced in thin and thick targets for a large range of primary projectiles and energies. To fulfil this program experiments were done at GANIL (France) [4], COSY (Germnay) [3] and CERN [5].

Figure 2 shows representative results obtained by this collaboration at Jülich with the BNB detector. This figure shows the measured average neutron multiplicity per incident proton as a function of the target thickness and beam energy for Pb, Hg and W as target materials. For the different target materials, the neutron multiplicity saturates at a given target thickness which increases with the proton energy. This saturation, at about 30 cm, underlies the previous conclusion that an incident proton of 1 GeV originates on average two nuclear collisions.

3.2 Energy and spatial distribution of neutrons.

Specific experimental setups are needed to measure the spatial and energy distribution of the neutrons produced in spallation reactions. The experiments performed by the "transmutation" collaboration at Saturne (France) constitute a clear example. 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 below 400 MeV was based on a measurement of their time of flight, the time difference between the incident proton, tagged by a plastic scintillator, and a signal from 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 to determine the angular distribution of the neutrons.

The neutron production in reactions induced by protons with energies between 0.8 and 1.6 GeV on thin and thick lead targets was investigated [8]. Figure 3 shows the results obtained on a two centimetres thick lead target with a 1.2 GeV proton beam, characterising the spallation process to be applied in an ADS. 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 will provide information about the inter-nuclear cascade.

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 make them 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

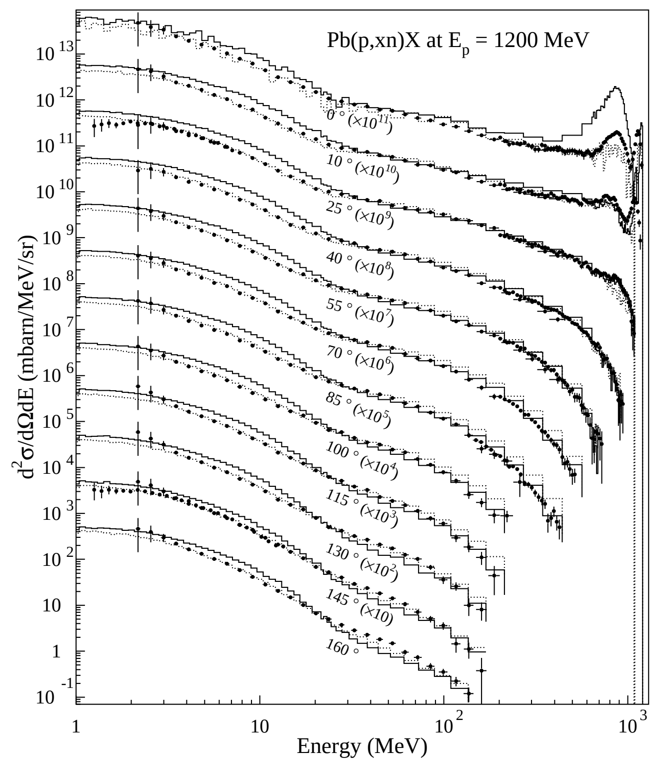

Figure 3: *Neutron production double-differential cross sections measured in 1.2 GeV proton induced reactions on a 2 cm thick Pb target [8]. The histograms represent calculations using the Bertini INC code [9] while the dotted lines corresponds to calculation done with the Cugnon INC code [10].*

most of the residues the measurement is done after β decay and consequently only isobaric identification is possible.

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

4.1 Measurement of residue production in inverse kinematics.

Outstanding experiments were performed by a German-French-Spanish collaboration at GSI (Germany). The technique used in these experiments takes advantage of the inverse kinematics and gives the full identification in mass and atomic number of the reaction residues as well as their velocities.

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. At this energy all residues of the reaction are predominantly fully stripped, bare ions. The achromatic high resolution magnetic spectrometer FRS [11] equipped with an energy degrader, two position sensitive scintillators and a multisampling ionisation chamber allowed to identify in atomic and mass number all the reaction residues with half lives longer than 200 ns. Resolving powers of $A/\Delta A \approx 400$ and $Z/\Delta Z \approx 150$ were achieved with this technique. The final production cross sections are evaluated with an accuracy not far from 10%. In addition, the high resolving power of the magnetic spectrometer allows to determine the recoil velocity of the reaction residues. This information is relevant for the characterization of the damages induced by the radiation in the accelerator window or the structural materials. More details about these experiments can be found in references [12, 13, 14, 15].

In Fig. 4, all residues measured in the reaction ^{208}Pb(1 A GeV)+p are presented on top of a chart of the nuclides. More than 850 different nuclei were identified in this reaction. As can be seen in this figure, the spallation residues populate two different regions of the chart of the nuclides. The upper region corresponds to the spallation-evaporation residues which populate the so called evaporation-residue corridor. The second region corresponds to medium-mass residues produced in spallation-fission reactions. Both reactions mechanisms, fission and evaporation, inherently different, contribute to the production of residues.

The measured isotopic production cross sections for some selected elements are presented in figure 5. This figure shows clearly the quality of the measured data that can be used to benchmark any model calculation.

Figure 6 shows the average kinetic energy in the center-of-mass frame of frag-

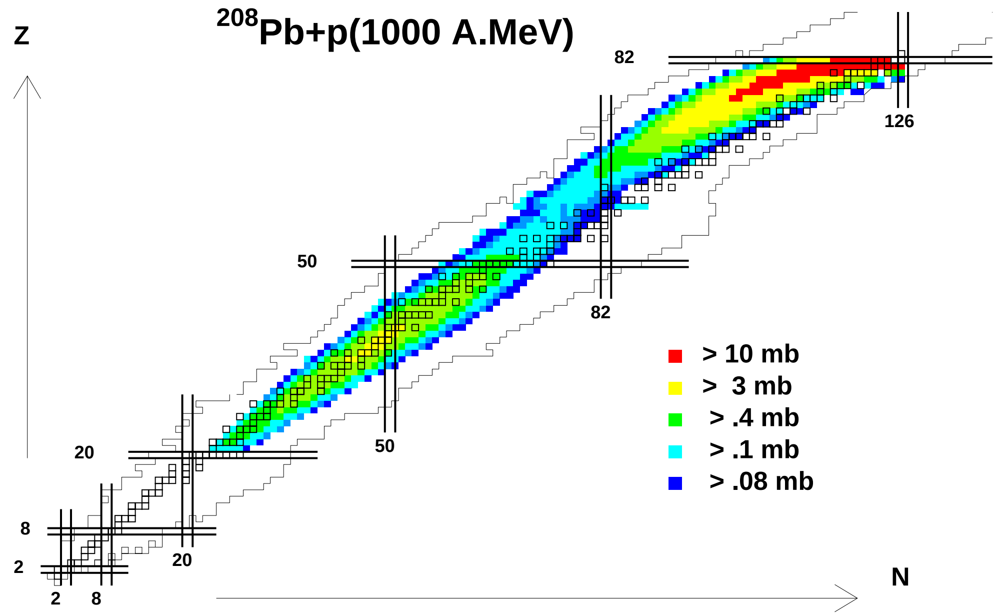

Figure 4: *Two-dimensional cluster plot of the isotopic production cross sections of all the spallation residues measured at GSI in the reaction* ^{208}*Pb(1 A GeV)+p shown on top of a chart of the nuclides [15].*

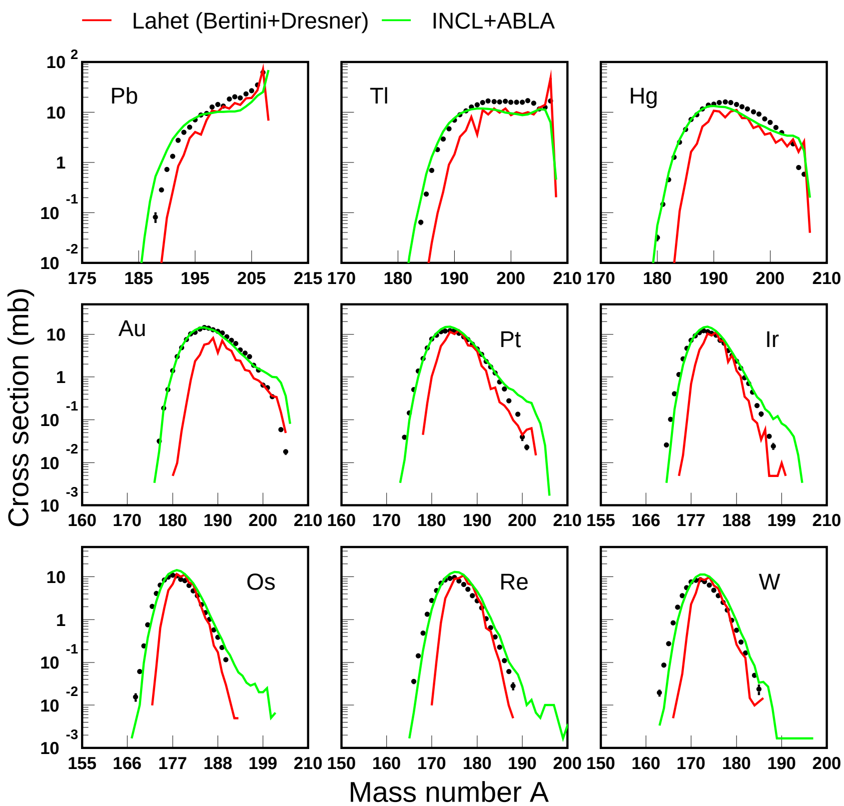

Figure 5: *Isotopic production cross sections for some of the elements produced in the reactions* $^{208}Pb+p$ *at 1 A GeV measured at GSI [15]. The data are compared with two model calculations, the red lines correspond to the results obtained with the Lahet code [16] while the green lines were obtained with the intra-nuclear cascade model of Cugnon [10] coupled to the evaporation-fission code ABLA from GSI [17, 18].*

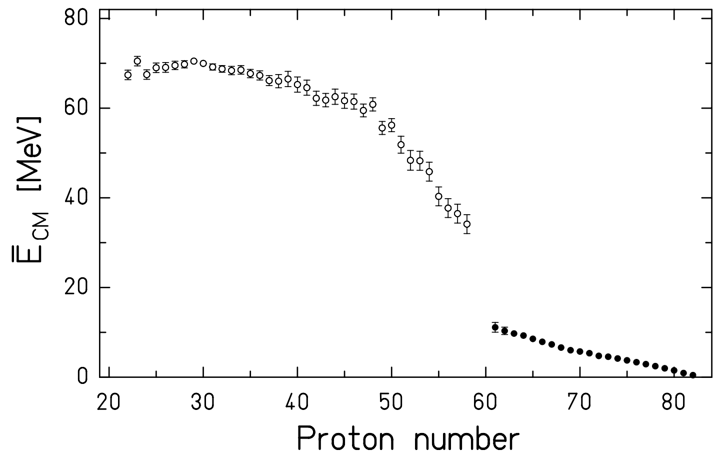

Figure 6: *Average kinetic energy in the center-of-mass frame of fragmentation (closed symbols) and fission (open symbols) residues produced in the reaction ^{208}Pb+p at 1 A GeV as a function of their atomic number [15].*

mentation (closed symbols) and fission (open symbols) residues produced in the reaction ^{208}Pb+p at 1 A GeV as a function of their atomic number [15]. These results clearly show an increase of the recoil velocity of the fragmentation residues for the most violent collisions leading to the production of lighter residues. The large kinetic energies of the fission residues are a key parameter to evaluate the heat load of the spallation target.

4.2 Measurement of residue production in direct kinematics.

γ-ray spectroscopy allows to investigate the production of spallation residues in direct kinematic reactions. Although this method is restricted to isobaric identification after β-decay, it is possible for some shielded isotopes to determine their primary production cross sections. This technique consumes less beam time than the inverse kinematics method. Therefore, full excitation functions can be established for selected isotopes, as shown in Fig. 7. The method can also be applied to thin and thick targets at low and high energies and in this sense, it is complementary to the inverse kinematics technique.

Presently there are two main experimental programs in Europe using this technique. The group of R. Michel at the University of Hannover performed experiments mainly at Saturne (France) and PSI (Switzerland). In these experiments different target material were irradiated with protons in the energy range 20 - 2000 MeV. Figure 7 shows representative results of these investigations. In this figure, excitation functions of the production of some shielded isotopes in collisions induced by protons on lead are shown [21]. These results clearly indicate

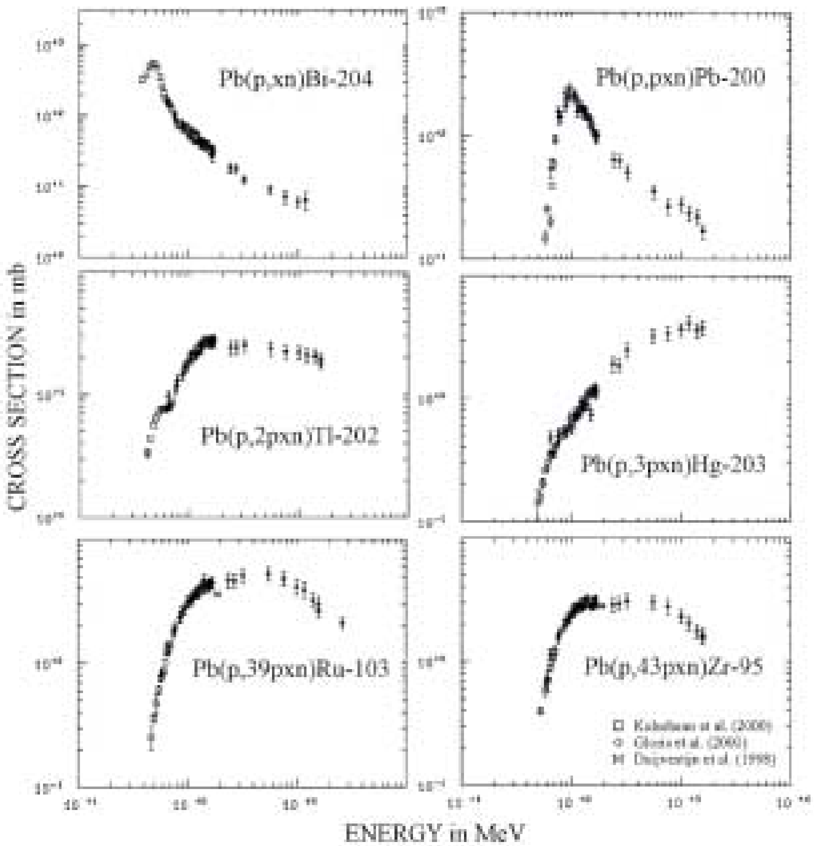

Figure 7: *Excitation functions for some selected isotopes produced in the interaction of protons with lead measured using γ-spectroscopy techniques [21].*

that the low energy reactions produce mainly residues close to the target nucleus, while most of the reaction residues further away from the target are produced by energetic particles. Similar experiments are also performed at the ITEP in Moscow [20].

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. As discussed, these reactions play a major role in the multiplication and moderation of the neutrons. The energy dissipated in the target nucleus leads to the emission of few nucleons and consequently only to residual nuclei close in mass and atomic number of the target material.

The experiments performed in this energy range intend to measure the double-differential production cross sections of neutrons and light-charged particles in reactions induced by protons and neutrons with energies in the range 20-200 MeV. Fission is also investigated. Measurements of the total fission cross sections and mass distributions of fission residues for specific targets are also in progress. These measurements will allow to extend ENDF-libraries up to 200 MeV.

An important number of European laboratories contribute to this experimental program which takes 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). Most of them contribute to the HINDAS project of the Fifth Framework program of the European Commission [22]. This European project also includes the measurements of residue production in inverse kinematics done at GSI and light charged-particle production experiments in COSY, both in the 200-2000 MeV region.

Another important program supported by the European Commission is the n_TOF project. This project is based on a time-of-flight neutron facility recently set up at CERN. After moderation, the neutrons produced in a spallation lead target fed with 20 GeV protons from the PS cover an energy range between 1 eV and 200 MeV [23]. The experimental program foreseen for this facility intends to cover a large number of experiments related to neutron capture and neutron-induced fission reactions. The expected neutron fluxes will allow to use radioactive targets for some of these measurements. Results from these experiments are expected in the coming years.

6 Model simulations.

The primary interaction of relativistic protons in a spallation target is mainly described in terms of semiclassical nucleon-nucleon collisions (intra-nuclear cascade) followed by the statistical deexcitation of the hot residue. The main inputs of the intra-nuclear cascade (INC) are the elastic and inelastic nucleon-nucleon cross sections and the distribution in position and momentum space of the nucleons inside the target nucleus. The statistical evaporation of particles is generally based on the Weisskopf formalism while fission should be described considering dissipation effects leading to the hindrance of that channel at high excitation energy. In this case the main parameters are the description of the level densities, the barriers for charged-particle emission and the fission barriers. Another critical parameter is the coupling time between the intra-nuclear cascade and the deexcitation stage.

The inter-nuclear cascade in the target assembly will lead to reactions induced by light particles, mainly neutrons, at energies below 200 MeV. At these energies the semi-classical approach of the intra-nuclear cascade is not valid any more and codes based in preequilibrium models and direct reactions are mainly used.

The last model intercomparison done by NEA [24] 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 high quality data when the codes where developed. The new data provided by the present experimental programs will help to improve this situation. In figure 5, 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] are compared with two model calculations. In this figure, the red lines correspond to the results obtained with the Lahet code (Bertini + Dresner) [16] while the green lines were obtained with the intra-nuclear cascade model of Cugnon [10] coupled to the new evaporation-fission code ABLA from GSI [17, 18]. As can be seen, the new INC and evaporation models provide a much better description of the experimental data.

Similar improvements are obtained with the new codes developed in the energy region below 200 MeV. The TALYS computer code system [25] created by NRG Peten and CEA Bruyères-le-Châtel represents one of the most promising tools including nuclear models for direct, compound, preequilibrium and fission reactions.

7 Outlook

The present experimental programs investigating the nature of spallation reactions are expected to provide accurate data on both neutron and residual nuclei production. Although the high energy interaction of protons with lead has been intensively investigated during the last years, experiments at lower energies are still in progress. In that sense programs like HINDAS or n_TOF will provide interesting data during the incoming years.

The new sets of data will allow to extend ENDF-libraries up to 200 MeV but also will contribute to develop new simulation codes. These new codes and libraries will constitute a key input in the design of an ADS target or window. In particular, target activity, radiation damage or gas production could be calculated with high accuracy. In order to accomplish these aims coordination programs between the different communities contributing to the design of an ADS are in progress like the French GEDEON project in a national scale or the BASTRA cluster in a European context.

References

[1] D. Ridikas, thesis, University of Caen, 1999

[2] D. Ridikas and W. Mittig, Nucl. Instr. and Methods A 414 (1998) 449

[3] A. Letourneau et al., Nucl. Instr. and Methods B 170 (2000) 299

[4] B. Lott et al., Nucl. Instr. and Methods A 414 (1998) 117

[5] D. Hilscher et al., Nucl. Instr. and Methods A 414 (1998) 100

[6] F. Borne et al., Nucl. Instr. and Methods A 385 (1997) 339

[7] E. Martinez et al., Nucl. Instr. and Methods A 385 (1997) 345

[8] X. Ledoux et al. Phys. Rev. Lett. 82 (1999) 4412

[9] H.W. Bertini et al., Phys. Rev. 131 (1963) 1801

[10] J. Cugnon et al., Nucl. Phys. A 620 (1997) 475

[11] H. Geissel et al., Nucl. Instr. Methods B 70 (1992) 286

[12] W. Wlazlo et al., Phys. Rev. Lett. 84 (2000) 5736

[13] J. Benlliure et al., Nucl. Phys. A 683 (2001) 513

[14] F. Rejmund et al., Nucl. Phys. A 683 (2001) 540

[15] T. Enqvist et al., Nucl. Phys. A 686 (2001) 481

[16] R.E. Prael et al., Los Alamos National Laboratory, report LA-UR-89-3014

[17] A.R. Junghans et al., Nucl. Phys. A 629 (1998) 635

[18] J. Benlliure et al., Nucl. Phys. A 628 (1998) 458

[19] R. Michel et al. Nucl. Instr. and Methods B 129 (1997) 153

[20] Y.E. Titarenko et al., Nucl. Instr. and Methods A 414 (1998) 73

[21] M. Gloris et al., Nucl. Instr. and Methods A 463 (2001) 593

[22] J.P. Meulders, Proc. of the 6^{th} Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Madrid, Spain, December 2000

[23] C. Rubbia et al., CERN/LHC/98-02(EET)

[24] R. Michel, P. Nagel, International Codes and Model Intercomparison for intermediate energy activation yields. NSC/DOC(97)-1, NEA/P&T No 14

[25] A. Koning et al., contribution to the International Conference on Nuclear Data for Science and Technology, Tsukuba, Japan, October 2001