

HIGH AND INTERMEDIATE ENERGY NUCLEAR DATA FOR ACCELERATOR DRIVEN SYSTEMS – THE HINDAS PROJECT

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Abstract

The HINDAS project (High and Intermediate energy Nuclear Data for Accelerator driven Systems) is a three years project supported by the European Commission under the Fifth Framework Program. The gathering of 16 partners, both experimentalists as theoreticians, allows to measure the wealth of new nuclear reaction cross-sections in the energy range between 20 MeV and 2 GeV on 3 elements of crucial importance for ADS systems: Pb as a target element, U as an actinide and Fe as a shielding element. The new experimental data will help to benchmark the existing theoretical models or to improve them. The assembly of nuclear data tables on those elements will allow interpolating to other elements appearing to be important in the design and the construction of an European ADS demonstrator.

1. Introduction

The HINDAS project (High and Intermediate energy Nuclear Data for Accelerator driven Systems) is supported by the European Commission under the Fifth Framework Program (September 2000-August 2003) and involves 16 European laboratories. Its general objective is to obtain a complete understanding and modelling of nuclear reactions in the 20-2 000 MeV region, in order to build reliable and validated computational tools for the detailed design of the spallation module of an accelerator driven system. This essential goal can only be accomplished by means of a well-balanced combination of basic cross-section measurements, nuclear model simulations and data evaluations.

Therefore, three nuclides, Fe, Pb and U have been chosen which provide a sufficiently broad coverage of the periodic table and are representative of the target, structure and core materials of the ADS. Hence, not only a few of the top-priority materials are chosen but, more importantly, with detailed theoretical and experimental knowledge of these particular elements, the nuclear models present in the foreseen simulation codes of this project will be fine-tuned. These will be employed to generate nuclear codes and data libraries for the materials that are requested by the ADS community.

The measurements will be performed at six nuclear physics laboratories in Europe, where beams of proton, neutron and heavy ion (in conjunction with inverse kinematics) as well as relevant measurement are available.

There appear to be a transition region around 200 MeV for the theoretical models. In the 20-200 MeV region, the theoretical calculations include direct interaction, pre-equilibrium, fission and statistical models, all with many uncertainties. Above 200 MeV, the theoretical analysis includes the intra-nuclear cascade model together with fission and evaporation models. A similar transition appears at about the same energy in the experimental facilities and in the measuring techniques.

The HINDAS project is therefore divided in experimental as well as theoretical work packages, according to this energy limit. The detection techniques differ also substantially for neutrons, protons and residual nuclide production, which has motivated a further division of the work packages according to the detected nuclides. This paper presents these work packages with the different results that would be available at the end of the project.

2. Experimental work between 20 and 200 MeV

The experimental work in the intermediate energy range from 20 MeV to 200 MeV is condensed in 3 parts. The measurements of cross-sections of nuclear reactions induced by protons and neutrons on Fe, Pb and U targets cover 2 work packages and the measurement of residual nuclide production is the object of the third part. Those experiments will be performed at 4 European accelerators which allows to cover the entire energy range from 20 to 200 MeV for the proton beams and two of those facilities can produce monoenergetic neutron beams with time of flight facilities.

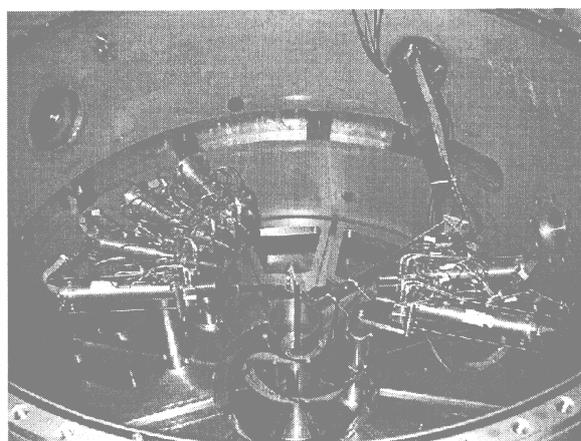
2.1 *Light charged-particle production induced by neutrons or protons between 20 and 200 MeV (WP1)*

The measurement of double differential cross-sections of light charged-particles p, d, t, ^3He and α induced by protons or neutrons on the different chosen targets will be obtained by measuring the energy spectra of each charged-particle over a large angular range from 15° to 160° . By integration over the angle, energy differential cross-sections are obtained at each angle, and by integration over the energy, angular differential cross-sections of the produced particle are deduced. The information contained in the double differential cross-sections is very stringent for theoretical models of nuclear

reactions, since the pre-equilibrium reactions have to be taken into account, in addition to the direct interaction contribution at the high energy part of the spectrum and the statistical evaporation component at the low energy side of the spectra.

The (p,xlcp) reactions on Pb and U will be measured by the partners of UCL, Subatech and LPC-Caen at 65 MeV at the CYCLONE cyclotron (UCL, Louvain-la-Neuve), and the same reactions on Fe, Pb and U at 135 MeV by the partners of Subatech, LPC-Caen and RuG at the KVI cyclotron (Groningen, The Netherlands) (see also N. Marie, this meeting). For these measurements, 8 triple telescopes (Si-Si-CsI) allow to measure the light charged-particles over their entire energy range (with low energy thresholds). Figure 1 shows a picture of the reaction chamber with the triple telescopes.

Figure 1. **The reaction chamber and the triple telescopes used in the (p,xlcp) reactions**



The (n,xlcp) reactions on Fe, Pb and U will be measured at 65 MeV at the CYCLONE cyclotron (UCL, Louvain-la-Neuve) (see also M. Kerveno, this meeting). Six ΔE -E telescopes (NE 102 plastic scintillator – CsI(Tl) detector) detect the charged particles produced by the neutrons on the target. The information from the telescopes coupled to the time of flight method with excellent time resolution (less than 1 ns) allows reconstructing, event by event, the energy spectra for each ejectile. Double differential cross-sections are obtained for the neutron mono-energetic peak (~63 MeV) and also for energies from the continuum of the neutron energy spectrum [1] (from 30 to 57 MeV).

The (n,xlcp) reactions on Fe and Pb at 100 MeV will be measured at The Svedberg cyclotron (UU, Uppsala) with a similar detection setup as for the proton induced-reactions. Partners of Subatech (Nantes), LPC-Caen and UU (Uppsala) are involved in these measurements ([2] and F.R. Lecolley, this meeting).

Finally, charged-particles multiplicities will be measured in proton-induced reactions on Fe, Pb and U in the energy range between 130 and 200 MeV by partners from RuG (Groningen).

2.2 Neutron production induced by neutrons and protons (WP2)

Partners of UU and LPC-Caen study elastic neutron scattering (n,n), at 100 MeV for Fe and Pb [2]. Such data are important to determine the nuclear optical potential to high precision in an energy range where data are essentially lacking. With this model at hand, cross-sections for elastic scattering, which is the most important reaction channel in the moderation and transport of the source neutrons, can be

calculated. Moreover, the optical potential is a necessary component in the description of many other reaction channels, since it accounts for the behaviour of a neutron entering or emerging from a nucleus.

The measurements will be performed using a recently developed detector set-up, consisting of two identical detector sets, which can be arranged to cover, e.g., the 10-50 and 30-70 degree ranges. Each detector set consists of a front veto scintillator, a 1 cm thick plastic scintillator for conversion into recoil protons, two drift chambers with x-y position sensitivity for proton tracking, and an array of 12 large CsI detectors for proton energy measurement. Absolute cross-sections will be determined by comparison with the reasonably well-known neutron-proton scattering cross-section.

Furthermore it is proposed to study the feasibility of (n, xn) reactions on Pb at 100 MeV. Such experiments are difficult to perform, but information is of great importance to understand and improve quantum-mechanical multi-step direct and classical pre-equilibrium models, as well as statistical models built on multiple Hauser-Feshbach emission. The measurements will make use of part of the previously described set-up, together with an active target for conversion of the emitted neutrons into recoil protons. The active conversion target will be positioned outside the neutron beam, but close to the Pb target, to obtain a large solid angle for neutrons. The recoil protons will be traced by a couple of drift chambers, and finally the energy will be determined in the CsI detector array.

Finally, the measurement of double-differential spectra from (p, xn) reactions in Pb and U, using a 65 MeV proton beam will be performed by partners of LPC-Caen, Subatech and UCL. In these experiments the emitted neutrons will be detected by well-shielded NE213 neutron detectors, placed around the scattering centre to measure angular distributions. The neutron energy distribution will be determined using time-of-flight techniques. Neutrons will be distinguished from gamma-rays using the pulse shape discrimination properties of this kind of detectors.

2.3 Residual nuclide production induced by neutrons and protons and production of long-lived radionuclides (WP3)

Reliable cross-sections for the production of residual nuclides by medium-energy proton- and neutron-induced reactions are essential for ADS to calculate the radioactive inventories of the spallation target, of structural materials and of ambient matter. Production of residual nuclides by GeV protons in thick or massive targets are a complex phenomenon the modelling of which needs to follow in detail the inter- and intra-nuclear cascades, the production and transport of primary and secondary particles. The spectra of primary and secondary particles strongly depend on the material irradiated as well as on geometry and depth inside the target. To calculate activation rates and radioactive inventories such calculated spectra have to be folded with the energy-dependent cross-sections of the underlying nuclear reactions for energies from thresholds up to the initial energy of the primary particles. Presently, there is no model or code available to predict the required cross-sections with an accuracy of better than a factor of two on the average. Therefore, one has to rely for the important nuclides on experimental cross-sections. Such experimental cross-sections are also needed if one tries to improve models and codes as a basis for validation.

Due to the importance of nuclear reactions of secondary particles, neutron-induced reactions will dominate the radionuclide inventory of the spallation target though the high-energy primary protons will significantly contribute. As a consequence, one needs cross-sections for both proton- and neutron-induced reactions for a reliable modelling of residual nuclide production over the entire energy range.

The data to be determined in this section will provide an experimental basis to calculate such inventories of the spallation target, of shielding and structural materials for an accelerator driven system a few minutes after shut-down as well as to validate theoretical work which is needed to

calculate the very short-lived radionuclides which make up an essential part of the spallation target during operation of a facility. With respect to the long-term behaviour and the final disposal of spallation targets and structural materials the precise modelling of long-lived radionuclides will be essential. Up to now, there are no inventory calculations which take into account long-lived radionuclides, mainly due to the lack of respective cross-sections.

For the modelling of radionuclide inventories it will be sufficient as a first approximation to have neutron-cross-sections up to 200 MeV. Measurements of residual nuclide production induced by neutrons between 30 and 180 MeV are foreseen. For proton-induced reactions one needs the complete excitation functions up to the energy of the primary beam. The latter do exist from recent work of our collaboration for most relevant target elements [3]. Measurement of production cross-sections of long-lived radionuclides via accelerator mass spectrometry (AMS) after chemical separation (partners ZSR and ETHZ) will be performed between 40 and 75 MeV.

3. Experimental work between 200 and 2 000 MeV

The aim of this work will be to collect high quality data and compare them with the state-of-the-art nuclear models. Data will be either measured in the framework of the project or have already been measured by the partners but not yet fully interpreted. In any case, they will be delivered as a ready-to-use file to be included in international data banks.

Particular attention will be paid to the impact of the new data for applications. Calculations of several quantities important in the design of ADS target or window will be performed using standard High Energy Transport Codes. In these codes the elementary cross-sections generated by the old nuclear models will be replaced either by the most recent version of models from J. Cugnon validated on data from our collaboration or, when possible and if models are not yet reliable enough, directly by the measured cross-section. Errors or uncertainties due to the use of the standard codes will be assessed.

3.1 Light charged-particle production (WP4)

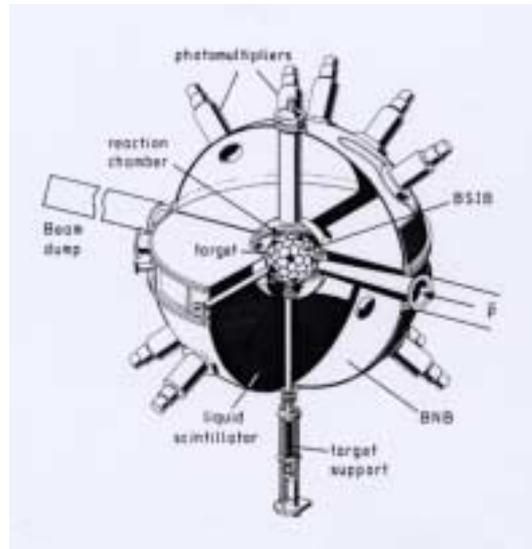
This part will be devoted to the collection of data concerning the production of light charged-particles. These data are important to probe the high-energy nuclear models in which the competition between neutrons and charged-particles, and the emission of composite nuclei (deuterons, alphas) are not yet treated satisfactorily. Moreover, the production yields of hydrogen and helium are essential for estimation of gas production in the window or structure materials of an ADS.

Production cross-sections for hydrogen and helium are being measured using a 4π silicon ball detector. So far, experiments have been performed at 0.8, 1.2, 1.8 and 2.5 GeV on several targets [4] and further experiments are foreseen. These measurements are also performed in coincidence with neutron multiplicity distributions. This allows studying the production rates of protons and alphas as a function of the excitation energy in the nucleus remaining at the end of the Intra-Nuclear Cascade stage. All these data will be analysed and compared to high-energy nuclear models.

Implications of the results from this experiment for gas production in some of the components of an ADS will be assessed, for instance on the lifetime of the window or on structure materials.

Moreover, a new magnetic spectrometer able to measure with a high resolution double-differential cross-sections for the production of light charged-particles (induced by protons) in coincidence with low energy neutrons will be designed.

Figure 2. The Berlin ball detector system



3.2 Neutron production induced by protons in thin and thick targets (WP5)

In this work-package, different types of neutron production data measured recently by the partners in both thin and thick targets will be collected, cross compared and compared with models.

Up to recently, very little high-quality data concerning double-differential cross-sections of neutron production were existing above 800 MeV and below there were significant discrepancies between different sets of data. Partners of CEA-Saclay, CEA-Bruyères and LPC-Caen have measured neutron energy spectra and complete angular distributions using two complementary experimental techniques: time-of-flight for the low energy part of the neutron spectrum and neutron-proton scattering on a liquid hydrogen converter with a magnetic spectrometer measuring the momentum of the recoiling proton for high energy neutrons. This has allowed to obtain energy spectra of (p,xn) reactions with a high resolution from 2 MeV to the incident energy, on several targets at 800, 1 200 and 1 600 MeV [5]. The same apparatus was used to measure neutron energy spectra from thick targets with different length and diameters.

Partner from FZJ has participated to a collaboration using a 4π liquid scintillator detector able to measure event-wise the multiplicity of neutrons up to 150 MeV on both thin and thick targets of different length and diameter for incident proton energies of 0.4, 0.8, 1.2, 1.8 and 2.5 GeV over a wide range of structural and target materials for ADS applications [4]. The neutron multiplicity distribution in thin targets reflects the excitation energy distribution of the nucleus remaining at the end of the Intra-Nuclear Cascade stage and is therefore important to understand the reaction mechanism. The average value of the neutron multiplicity distribution in thick targets is directly interesting for applications.

The measurements performed by FZJ and CEA-Saclay, CEA-Bruyères, LPC-Caen are complementary both for technical (energy range of the measurements) and physics reasons (high-energy neutrons test the intra-nuclear cascade stage while low energy neutrons probe the evaporation stage). So far, no coherent simultaneous analysis of both experiments has been done. This will be the goal of this work-package in which, for example, the average multiplicity distributions measured with the neutron ball will be compared to those inferred from the integration of the double-differential

cross-sections; the secondary reactions induced in the neutron scintillator detector will be assessed using results of the high energy neutron spectrum measured by the double-differential cross-section experiment, etc.... Comparisons with the same high energy nuclear models for thin targets, the same high-energy transport codes for thick targets, taking into account the rather complex experimental acceptance of both experiments, will be performed. Results will be used to assess the remaining deficiencies in the codes to be improved in the theoretical section of the HINDAS project. Simulation of thick target results will also be realised. Direct applications of the thick target experiments such as average neutron multiplicities or high-energy neutron leakage for shielding estimation will be discussed.

3.3 Residual nuclide production in inverse kinematics (WP6)

In spallation reactions of heavy nuclei induced by protons of about 1 GeV, mostly short-lived radioactive nuclei are produced. The spallation residues are stopped inside the target. They decay towards stable isobars predominantly by beta decay. After irradiation, long-lived radioactive residues are identified in mass and atomic number by gamma spectroscopy and by accelerator mass spectrometry. These experiments provide reliable and comprehensive data on cumulative yields, from which long-lived activities and final element yields can be deduced. In addition, these techniques allow for measurements over a large range of bombarding energies. A previous inter-comparison with available data has revealed that the calculations with nuclear-reaction models are not realistic enough, but it is difficult to pin down the deficiencies of the models on the basis of cumulative yields. For this purpose, a complete systematic of isotopic production cross-sections emerging from the nuclear reaction is urgently needed.

In particular for proton energies above 200 MeV, a substantially different technique, based on the use of inverse kinematics, has been developed recently which allows identifying all short-lived radioactive nuclides produced as spallation residues prior to beta decay. Heavy nuclei are provided as projectiles, impinging on a liquid-hydrogen target. The spallation residues are identified in-flight in a high-resolution magnetic spectrometer. These experiments allow a much more direct insight into the reaction mechanism than experiments in normal kinematics and therefore are best suited to improve nuclear-reaction models which are known to be unable to reproduce available data. In addition, this technique allows to determine the kinetic energies of the spallation residues [6], an information of highest importance for estimating radiation damages in structure material of an ADS. That means that these experiments provide unique and valuable information which complements the results obtained in normal kinematics. Due to electronic interactions in the spallation target, the primary protons lose energy and induce nuclear reactions in a wide energy range. However, the higher energies are particularly important for residual-nuclide productions, since more than 75% of the primary protons of 1 GeV undergo nuclear reactions in the spallation source in an energy range above 700 MeV. Additional measurements with a liquid deuterium target are aimed to provide information on spallation reactions induced by neutrons.

The experiments in inverse kinematics and the data analysis being rather complex, only few projectile species and energies can be investigated. Therefore, the measurements are restricted to ^{208}Pb , ^{238}U and ^{56}Fe at 1 A GeV and partly at 500 A MeV. During the 3-year period of the project, final data on ^{208}Pb and ^{238}U will be available. It is expected that the full isotopic distributions and kinetic energies obtained in inverse kinematics in combination with detailed excitation functions of specific reaction products obtained in normal kinematics provide sufficient information to develop substantially improved nuclear-reaction models which can then be used in transport codes to predict realistic energy-integrated production yields in thick targets.

Finally, a new experimental technique will be developed to also measure neutrons and light charged-particles in inverse kinematics. This will allow establishing coincidences between these particles and the heavy residues, an information still more relevant for modelling the nuclear reaction correctly.

Calculations of the activities, radiotoxicities and element distributions in a realistic lead spallation target will be performed using transport and evolution codes. The elementary cross-sections generated by the old nuclear models will be replaced either by the most recent version of models from J. Cugnon, or directly by the measured production yields on Pb at 1 000 MeV, extrapolated at non-measured energies using the energy dependence of the excitation functions measured in WP3.

4. Theory and evaluation

For research on accelerator-driven systems, cross-sections for the important materials need to be known for ALL possible outgoing channels, outgoing energies and angles. This total amount of required information is so large that experiments alone can never cover the nuclear data needs for ADS. To fill this gap, the data are simulated computationally, with the help of theoretical reaction models. The development of this simulation is done in close correspondence with the experiments: adjustable parameters of the theoretical models are adjusted in such a way that the latter reproduce the measurements as closely as possible. The critical assumption is then that the models can also be used in areas where no measurements exist. Hence, the actual provision of nuclear data in a form usable for ADS design will be done in two work-packages

- Nuclear data libraries, improved and extended up to 200 MeV, based on nuclear models.
- Intra-nuclear cascade models and codes for the higher energies.

4.1 Nuclear data libraries and related theory (WP7)

This part concerns nuclear model calculations for a theoretical analysis of the between 20 and 200 MeV and predictions for the unmeasured channels for energies up to 200 MeV. In combination, this will be used to construct complete nuclear data libraries for ^{56}Fe , ^{208}Pb and ^{238}U up to 200 MeV, which will show a clear improvement over all other existing nuclear data files and methods [7].

Theoretical calculations will be performed with a variety of nuclear models at NRG-Petten and at CEA-Bruyères-le-Châtel. The new model code system will be extended to include a proper treatment of all channels precisely. Coupled-channels optical models will be constructed for the simulation of the elastic and inelastic channels, not only for the total (angle-integrated) cross-sections, but also for the angular distributions. For the continuum reactions, complete outgoing energy and angular spectra will be included for all light particles. These will be predicted, and compared with the new measurements, using quantum-mechanical multi-step direct (MSD) and classical pre-equilibrium models that include novel models for microscopical particle-hole level densities and the optical models. Multiple pre-equilibrium emission beyond the second step will be included for the highest incident energies. Complete evaporation of the residual nuclides is accounted for by means of multiple Hauser-Feshbach emission that includes competition of all possible outgoing particle channels and fission, while conserving energy, angular momentum and parity. Simultaneously with the double-differential spectra, the calculated residual production cross-sections will be compared with the experiments as described in WP3. Both types of observables must be described by one and the same calculation. High-energy fission will also be included by means of an extension of the Brosa model.

All possible nuclear reactions will be evaluated simultaneously, in order to ensure flux conservation and energy balance. The results will be compared with the American GNASH code.

The calculated results will be processed automatically into the ENDF6-format. The results will be combined with the data below 20 MeV to come to one consistent final library. If the existing data file below 20 MeV turns out to be inadequate, the cross-sections will be improved in the low energy regime as well to ensure a smooth transition from low to high energies. All the nuclear data will be stored in the common ENDF-6 format and will be checked according to a standard QA-system. As basis for the new high-energy evaluations, the European JEFF library will be used.

4.2 High-energy models and codes (WP8)

The high energy codes, although globally rather successful, suffer from some deficiencies. Both their inter-comparison and the comparison with existing experimental data reveal, in some identified regime, discrepancies which are beyond the accuracy required by the engineers working on projects of ADS or spallation sources. These observations call for improvements of the physics already included in the cascade codes (in-medium corrections, Pauli principle, mean field dynamics,...), of the evaporation codes (level densities) and of fission codes (viscosity, evaporation-fission competition at high excitation energy) [8]. These improvements are part of the specific theoretical task in this project. They will be realised in successive steps at Ulg-Liège.

The first step will consist of improving the existing codes by including physics aspects not included so far and by refining some of the physics which is already implemented. For the most recent intra-nuclear cascade (INC) code, this concerns a proper description of the nuclear surface, an improvement of Pauli blocking, which present too much fluctuations, and refinements of the in-medium corrections. For the evaporation codes, the first step will involve a careful examination of the input data and an advanced development of the fission model at high excitation energy, taking advantage of the forthcoming measurement of the fission component in reverse kinematics (see WP6). The improvements will be inspired by the most recent theoretical progress in nuclear dynamics far from equilibrium.

The second step aims at a validation of the improved codes (and other standard codes). An extensive comparison with the neutron differential cross-sections measured at SATURNE (800 to 1 600 MeV) and with the neutron multiplicities and light charged-particle spectra measured at Jülich (WP5) will be performed, for both thin and thick target data. In addition, an extensive comparison with the experimental residue production data to be provided by WP6 will be realised.

As a third step, a new improvement of the codes will be undertaken, if necessary. This work will involve an adjustment of the introduced parameters to describe less well known physics aspects, like the parameters regulating the coupling between the INC and evaporation codes and some parameters of the fission model, especially viscosity.

The final goal will consist in the elaboration of a version of a high-energy transport code including these new simulation tools. This version could be tested on the thick target data generated by this project.

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