HINDAS A EUROPEAN NUCLEAR DATA PROGRAMME FOR ACCELERATOR-DRIVEN SYSTEMS

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Abstract

In the HINDAS programme, nuclear data in the 20-2 000 MeV range are evaluated by means of a combination of nuclear models and well-selected intermediate- and high-energy experiments. A panoply of European accelerators is utilised to provide complete sets of experimental data for iron, lead and uranium over a large energy range. Nuclear model codes are being improved and validated against these new experimental data. This should result in enhanced ENDF-formatted data libraries up to 200 MeV, and cross-sections for high-energy transport codes above 200 MeV. The impact of the new data libraries and high-energy models will be directly tested on some important parameters of an accelerator-driven system (ADS). Here, we report the recent progress of the various experimental and theoretical activities in HINDAS.

Introduction

HINDAS (High and Intermediate energy Nuclear Data for Accelerator-driven Systems) is a project supported by the European Commission, from September 2000-August 2003, and involves 16 European laboratories. Its objective is to obtain a thorough understanding and complete 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. To achieve this, an ambitious experimental and theoretical programme has been launched.

Six European facilities, listed in Table 1, are participating in the measurement of the following observables:

- double-differential $(p, xn...x\alpha)$ and $(n, xn...x\alpha)$ cross-sections;
- residual nuclide production, by activation and inverse kinematics techniques, and residual kinetic energies;
- neutron elastic scattering angular distributions;
- neutron and charged-particle multiplicity distributions;
- thick target neutron spectra.

The diversity of the used accelerators is such that the complete energy region between 20 and 2 000 MeV is covered. A suitable coverage of the periodic table of elements is obtained with the choice of target elements: Fe, Pb and U. Lead and iron are representative of materials used in ADS, while uranium represents the actinide region.

Parallel to the measurement programme, theoretical models are under development. For energies from zero up to 200 MeV, the new nuclear model code TALYS has been designed. It includes the optical model, direct, pre-equilibrium, fission and statistical models and thereby gives a prediction for all the open reaction channels. In addition, for energies up to 2 000 MeV, a new intranuclear cascade model is under development. During the measurement programme, the nuclear model codes are directly benchmarked against the new experimental data. The intermediate energy models will be used to create ENDF-libraries up to 200 MeV, while the high-energy nuclear models will be implemented in high-energy transport codes. Finally, calculations of several quantities important in the design of ADS target or window, such as activity, radiation damage, gas production, etc., will be performed in order to assess the improvements brought by the new data and models.

The HINDAS project is divided into 8 work-packages (WP), which together aim to cover all aforementioned objectives. In this contribution, a general description of the programme and the first results will be presented.

Institute	Accelerator	Energy range (MeV)
UCL-Louvain-la-Neuve	CYCLONE	20-70 (p and n)
UU-Uppsala	TSL	20-180 (p and n)
KVI-Groningen	AGOR	130-190 (p)
PSI-Villigen	cyclotron	45-70 (p)
FZ-Jülich	COSY	50-2 500 (p)
GSI-Darmstadt	SIS	300-1 000 (MeV/A)

Table 1. European facilities participating in HINDAS

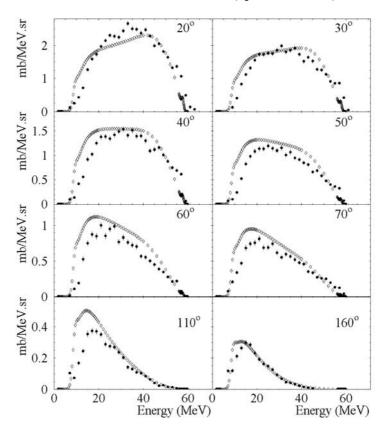
Experimental programme up to 200 MeV

Light charged-particle production induced by neutrons and protons (WP1)

The main objective of WP1 is to measure double-differential production cross-sections for light charged particles (*lcp*), i.e. protons, deuterons, tritons, Helium-3, alpha's, in nuclear reactions induced by protons and neutrons in the incident energy range 20-200 MeV. The goal is to provide precise experimental data over a complete angular range (20 to 160 degrees) with an overall uncertainty better than 15%.

The (p, xlcp) reactions on Pb and U have been measured by the partners of UCL-Louvain-la-Neuve, SUBATECH-Nantes and LPC-Caen at 65 MeV at the CYCLONE cyclotron (UCL), and the same reactions on Fe, Pb and U at 135 MeV are planned by the partners of SUBATECH, LPC and KVIGroningen at KVI. For these measurements, 8 triple telescopes (Si-Si-CsI) allow to measure the light charged-particles from a low energy threshold up to 160 MeV.

Figure 1. Double-differential cross-sections for 62.7 MeV Pb(*n*, *xp*), measured (closed circles) and calculated with TALYS (open diamonds)



The (*n*, *xlcp*) reactions on Fe, Pb and U will be measured at 65 MeV at the CYCLONE cyclotron (UCL). 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 to reconstruct, event by event, the energy spectra for each ejectile. Double-differential cross-sections are obtained for the

neutron monoenergetic peak (~63 MeV) and also for energies from the continuum of the neutron energy spectrum [2] (from 30 to 57 MeV). In Figure 1, the recently obtained 62.7 MeV Pb(n, $x\rho$) cross-sections [1] for the whole angular range are shown. Results for the other nuclides are still under analysis.

The $(n, xlc\rho)$ reactions on Fe and Pb at 100 MeV will be measured at the Svedberg cyclotron (Uppsala), with a similar detection set-up as for the proton induced-reactions. Partners of SUBATECH, LPC and UU-Uppsala are involved. [3]

In addition, a series of experiments devoted to the measurement of light-charged particle multiplicity distributions from proton-induced reactions for projectile energies between 130 and 200 MeV will be performed at KVI.

Neutron production induced by neutrons and protons (WP2)

Elastic neutron scattering angular distributions at 100 MeV for Fe and Pb will be measured [4] in Uppsala by UU-Uppsala and LPC-Caen. 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, 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. Optical models developed in WP7 will be used to analyse the measurements.

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.

This set-up will also be used, together with a new converter (CLODIA), to study the feasibility of (n, xn) experiments at 100 MeV. CLODIA has a much higher conversion efficiency, which will be needed for measurements of secondary neutron spectra.

Also, the measurement of double-differential spectra from (ρ , *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 NE-213 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. Note that the energy is chosen such that a data set consistent with that of WP1 is formed.

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

WP3 comprises measurements of integral cross-sections for the production of residual nuclides from Fe and U by protons with energies up to 2.6 GeV and 70 MeV, respectively, [5] and by neutrons with energies between 30 and 170 MeV, [6] measurements of the ^{235,238}U, ²⁰⁹Bi and ^{nat}Pb fission cross-sections for neutrons with energies from 30 MeV to 150 MeV [7] and measurement of production cross-sections of long-lived radionuclides from Fe and Pb after chemical separation.

Irradiation experiments with protons [8] are performed at PSI using the stacked-foil technique and off-line γ -spectrometry. Long-lived radionuclides are measured after chemical separation via AMS at the PSI/ETH Tandem AMS at ETH.

Experiments with quasi-monoenergetic neutrons produced by the ⁷Li(p,n)⁷ Be reaction are performed at UCL, [9] Uppsala [10,11] and NAC-Cape Town, South Africa. [12] Residual nuclides are measured by off-line γ -spectrometry. Neutron cross-sections are determined by unfolding methods from the experimental response integrals determined in a series of irradiation experiments with different neutron energies. Fission cross-sections are measured using parallel-plate fission chambers (PPFC). While the U- and the Bi-PPFC's were already used in earlier experiments, the ^{nat}Pb-PPFC was specially developed within the HINDAS project.

The data to be determined in this work package will provide an experimental basis to calculate inventories of the spallation target, of shielding and structural materials for an accelerator-driven system after shut-down. Also, it will aid in validating theoretical work which is needed for calculations on 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.

Experimental programme above 200 MeV

Light charged-particle production (WP4)

Light charged-particle production data are important to probe the high-energy nuclear models. In the latter, the competition between neutrons and charged-particles and the emission of composite particles (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, see Figure 2. So far, experiments have been performed at 0.8, 1.2, 1.8 and 2.5 GeV on several targets. [13] These measurements are also performed in coincidence with neutron multiplicity distributions using the neutron detector mentioned in WP5 (see below). This allows to study the production rates of protons and alphas as a function of the excitation energy in the nucleus remaining at the end of the intranuclear cascade stage. All these data will be analysed and compared to highenergy nuclear models. A careful analysis of the light charged-particle production data at high energies has shown that experimental data are scarce and often limited to a small range of energy. In particular, the high energy part of the out-going proton spectrum, which contributes as much as neutrons to the propagation of the inter-nuclear cascade inside a thick target, is not well known. In WP4, the partners will join to develop a new magnetic spectrometer able to measure, with a high resolution, double-differential cross-sections for the production of light charged-particles over a broad energy range. This apparatus will also be used in coincidence with a neutron scintillator detector measuring low energy neutron multiplicities. This will permit to disentangle the intra-nuclear cascade and the de-excitation stages in the reaction mechanism. These second generation experiments will provide a deeper understanding of the mechanisms that will be necessary to definitively tune all the parameters of the nuclear models. Simulations of the expected results with the high-energy nuclear models from WP8 will be done.

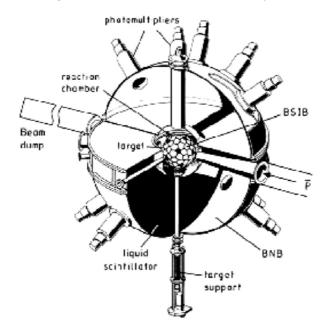


Figure 2. The Berlin ball detector system

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

In this work-package, different types of neutron production data measured in both thin and thick targets will be collected, intercompared and compared with models. Neutron energy spectra and complete angular distributions using two complementary experimental techniques have been measured: 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 (ρ , *xn*) reactions with a high resolution from 2 MeV to the incident energy, on several targets at 800, 1 200 and 1 600 MeV. [14] The same apparatus was used to measure neutron energy spectra from thick targets with different length and diameters.

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 has been measured. This was performed with a 4π liquid scintillator detector able to measure event-wise. The neutron multiplicity distribution in thin targets reflects the excitation energy distribution of the nucleus left at the end of the intranuclear cascade stage and is therefore important to understand the reaction mechanism.

Results will be used to assess the remaining deficiencies in the codes to be improved in WP8. Simulation of thick target results will also be realised using a 200 MeV evaluated data file instead of the standard 20 MeV file. Direct applications of the thick target experiments such as average neutron multiplicities or high energy neutron leakage for shielding estimation will be investigated.

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 methods are used in WP3 of HINDAS and 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 intercomparison 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 crosssections emerging from the nuclear reaction is required.

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 to identify 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, [15] an information of highest importance for estimating radiation damages in structure materials 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 loose 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 ²⁰⁸Pb, ²³⁸U and ⁵⁶Fe at 1 A GeV and partly at 500 A MeV. It is expected that the full isotopic distributions and kinetic energies obtained in inverse kinematics provide sufficient information to develop substantially improved nuclear-reaction models.

Theoretical/Evaluation programme

Nuclear data libraries and related theory (WP7)

1. TALYS. TALYS [17] is a computer code system for the simulation and analysis of nuclear reactions, created by NRG Petten and CEA Bruyères-le-Châtel. The basic objective behind the construction of TALYS is the ability to give a complete description of nuclear reactions that involve neutrons, photons, protons, deuterons, tritons, ³He- and alpha-particles, for target nuclides of mass 12 and heavier. To achieve this, a suite of nuclear reaction models has been implemented into a single code system. This enables to evaluate nuclear reactions from the unresolved resonance region up to intermediate energies. As specific features of TALYS we mention:

- In general, a non-approximative implementation of many nuclear models for direct, compound, pre-equilibrium and fission reactions.
- A continuous, smooth description of reaction mechanisms over a wide energy range (0.001-200 MeV).

- Completely integrated optical model and coupled-channels calculations through the ECIS code, with incorporation of new (global and local) optical model parameterisations for many nuclei. [18]
- Total and partial cross-sections, energy spectra, angular distributions, double-differential spectra and an exact modelling of exclusive cross-sections and spectra. Excitation functions for residual nuclide production, including isomeric cross-sections.
- Automatic reference to nuclear structure parameters as masses, discrete levels, resonances, level density parameters, deformation parameters, fission barrier and gamma-ray parameters, mostly from the RIPL library. [19]
- Various phenomenological and microscopical level density models, such as Gilbert-Cameron, Ignatyuk and combinatorial state densities built on Hartree-Fock-Bogoliubov based single-particle states.
- Semi-classical and quantum-mechanical (multi-step direct/compound) models for preequilibrium reactions.
- Use of systematics if an adequate theory for a particular reaction mechanism is not yet available or implemented, or simply as a predictive alternative for nuclear models.
- Automated construction of ENDF-6 data libraries.

TALYS is used for the analysis of all sub-200 MeV experiments in HINDAS. Figure 1 shows a prediction of the double-differential data measured in WP1. In Figure 3, residual production cross-sections calculated by TALYS are compared with experimental data. For both calculations, pre-equilibrium reactions were modelled with a two-component exciton model (and Kalbach systematics), while multiple compound emission was treated with the Hauser-Feshbach model. As two important ingredients we mention nucleus-tailored optical potentials and energy-dependent shell effects in the level density.

2. Tamura-Udagawa-Lenske model. In this part of WP7 the quantum-mechanical multistep direct reaction model according to Tamura-Udagawa-Lenske (TUL) is used to describe nucleon-induced reactions at intermediate energies. It will be applied for the calculation of the double-differential cross-sections measured in WP1 and WP2. Furthermore, it will be used for predictions of unmeasured data in this energy range. For ADS applications it is important that the magnitude of the calculated cross-sections is free of any adjustable parameters and is strictly related to known physical parameters and quantities such as effective interactions and applied optical potentials. Consequently, knowledge of dedicated optical potentials for the different isotopes is extremely important. Another objective is to include this option in TALYS.

3. Quasi-particle code DYWAN. DYWAN has been optimised by Univ. Nantes for nucleon and charged composite particle production in the 20-200 MeV range. In this code, the quasi-particles can occupy a set of discrete single-particle states, with some probability. The phase space extension of these single-particle states is described with the help of a wavelet basis, in a way which minimises the loss of information contained in the whole quantal wave-function. Equations of motion for the relevant degrees of freedom have been derived and solved numerically. The code is now in a state which allows a comparison with the measurements of WP1.

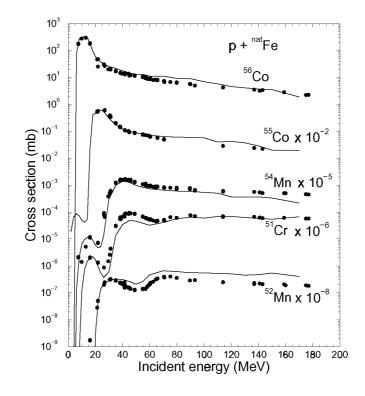


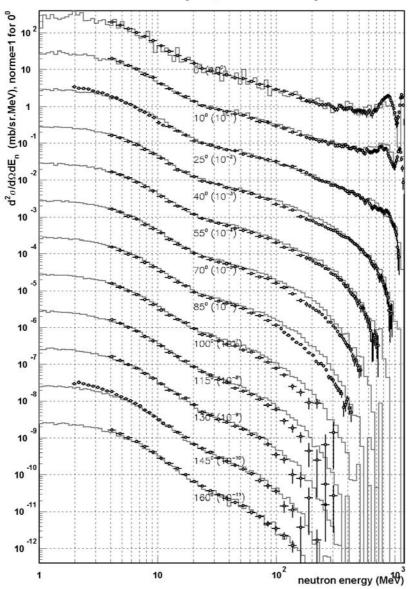
Figure 3. TALYS-0.49 calculation for residual production cross-sections of $p + {}^{nat}Fe$, compared with experimental data [20]

High energy models and codes (WP8)

1. Intranuclear cascade (INC) code. An enhanced INC code, INCL4, is under construction at the University of Liège and CEA Saclay. Although the previous version, INCL3, [21] gave reasonable predictions for proton-induced neutron cross-sections and residual mass distributions, the model suffered from some shortcomings: (i) it systematically underestimates the intensity of the quasi-elastic peak in neutron spectra (ii) it underestimates the cross-section for the production of residues with mass close to the target mass. This was attributed to the neglect of the diffuseness of the nuclear surface in INCL3. Currently, a density distribution with a smooth surface, of the Wood-Saxon type, is taken into account. Some additional newfeatures have been implemented in INCL4: (i) consistent dynamical Pauli blocking: The baryon-baryon collisions are allowed stochastically according to the estimated occupation of the phase space around the outgoing particles. In addition, the possible final state of a binary collision is checked for the energy content in the original Fermi sphere. This eliminates spurious emission due to the otherwise stochastic treatment of the Pauli blocking, (ii) improvement of the pion dynamics. The lifetime attributed to the delta particles has been lengthened for small delta mass, reflecting the small decay probability governed by the reduction of the phase space in pionnucleon channel, (iii) the angular momentum of the remnant is delivered by the code. This offers a microscopic alternative to phenomenological formulae like the one of de Jong et al., [22] (iv) inclusion of incident light ions: d, t, ³He, ⁴He, (v) The stopping time has been revisited. It is still determined by the observed changes of rate in the time variation of the average values of some key physical quantities, like the target excitation energy. However, the impact parameter and incident energy dependences are considerably weakened when a smooth surface is introduced.

A complete and usable version of INCL4 is under construction. [23] As an example, Figure 4 shows that the quasi-elastic peak in neutron spectra is satisfactorily reproduced.

Figure 4. Neutron production double differential cross-sections for 1 200 MeV protons *Pb*. Comparison between the Liège INC model plus the KHSV3p evaporation-fission model (histograms) with the experimental data (dots) of WP5 [14]



1200 MeV p+Pb, INCL4 + KHSV3p

2. Evaporation code. The latest version of the KHS evaporation code, [24] KHSv3p, has been completed by GSI and CEA Saclay. It differs from the preceding versions on three points. First, effective barriers for proton and alpha evaporation have been preferred to potential ones in order to account for penetration effects and have been adjusted to reproduce properly the isotopic distributions of residues. Second, the fission barriers have been adjusted from those of Ref. [25] by a factor $f_B=0.9$ to account for the reduction of the fission barrier with increasing temperature. Finally, dissipative effects are simulated by forbidding fission until some transient time has passed. This model, together with the INCL4 code, satisfactorily describes the residue production data from WP6.

Conclusions

The HINDAS project is now well underway. High quality experimental data, representing all important channels (for neutron, light charged particle and residual nuclide production) of proton- and neutroninduced reactions above 20 MeV on Fe, Pb and U are measured and compared with the state-of-the-art nuclear models. Data will be measured at the European facilities that are the best equipped for the reaction under consideration. Theoretical nuclear models will be developed (optical model, pre-equilibrium, fission, direct and statistical models at energies in the 20-200 MeV region; intra-nuclear cascade, fission and evaporation models above 200 MeV) and then benchmarked against the new experimental data. Intermediate energy models will be used to create evaluated data libraries up to 200 MeV, both for incident neutrons and protons, whereas the high energy nuclear models will be implemented in transport codes to generate the necessary cross-sections. The combination of these new high- and low energy transport codes will be probed on integral experiments.

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