Nuclear data for accelerator-driven transmutation


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This report concerns a study which was conducted for SKB. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.
Contents

1 Background 4

2 Introduction 4

3 Experimental setup and techniques 5
  3.1 The TSL neutron beam facility 5
  3.2 The MEDLEY setup 6
  3.3 The SCANDAL setup 7

4 Results and analysis 9
  4.1 Elastic scattering 9
  4.2 (n,xlcp) reactions 10

5 International activities 12
  5.1 Collaboration 12
  5.2 Meetings and conferences 14

6 Administrative matters 14
  6.1 Personnel and PhD students 14
  6.2 Reference group 15

References 15

Appendices:


IX. Minutes of the 23rd Meeting of the International Nuclear Data Committee (INDC), IAEA, Vienna, May 24 – 26, 2000, *IAEA Report INDC/P(00)-15.*
1 Background

The present project, supported as a research task agreement by Statens Kärnkraftsinspektion (SKI), Svensk Kärnbränslehantering AB (SKB), Barsebäck Kraft AB (BKAB) and Vattenfall AB, started 1998-07-01. From 1999-01-01 the project also receives support from Försvarets forskningsanstalt (FOA). The primary objective from the supporting organizations is to promote research and research education of relevance for development of the national competence within nuclear energy.

The aim of the project is in short to:

• promote development of the competence within nuclear physics and nuclear technology by supporting licenciate and PhD students,

• push forward the international research front regarding fundamental nuclear data within the presently highlighted research area “accelerator-driven transmutation”,

• strengthen the Swedish influence within the mentioned research area by expanding the international contact network,

• constitute a basis for Swedish participation in the nuclear data activities at IAEA and OECD/NEA.

The project is run by the Department of Neutron Research (INF) at Uppsala University, and is utilizing the unique neutron beam facility at the national The Svedberg Laboratory (TSL) at Uppsala University.

In this document, we give a status report after the second year (1999-07-01–2000-06-30) of the project.

2 Introduction

Transmutation techniques in accelerator-driven systems (ADS) involve high-energy neutrons, created in the proton-induced spallation of a heavy target nucleus. The existing nuclear data libraries developed for reactors of today go up to about 20 MeV, which covers all available energies for that application; but with a spallator coupled to a core, neutrons with energies up to 1 – 2 GeV will be present. Although a large majority of the neutrons will be below 20 MeV, the relatively small fraction at higher energies still has to be characterized. Above ~ 200 MeV, direct reaction models work reasonably well, while at lower energies nuclear distortion plays a non-trivial role. This makes the 20 – 200 MeV region the most important for new experimental cross section data.

Very little high-quality neutron-induced data exist in this energy domain. Only the total cross section (Finlay et al., 1993) and the np scattering cross section have been investigated extensively. Besides this, there are data on neutron elastic scattering from UC Davis at 65 MeV on a few nuclei (Hjort et al., 1994). Programmes to measure neutron elastic scattering have been proposed or begun at Los Alamos (Rapaport and Osborne) and IUCF (Finlay et al., 1992), with the former resulting in a thesis on data in the 5° – 30° range on a few nuclei.

The situation is similar for (n,xp) reactions, where programmes have been run at UC Davis (Ford et al., 1989), Los Alamos (Rapaport and Sugarbaker, 1994) and TRIUMF
(Alford and Spicer, 1998), but with limited coverage in secondary particle energy and angle. Better coverage has been obtained by the Louvain-la-Neuve group up to 70 MeV (Slypen et al., 1994).

Thus, there is an urgent need for neutron-induced cross section data in the region around 100 MeV, which is an area where very few facilities in the world can give contributions. By international collaboration within an EU supported Concerted Action, which will be followed by the full scale project HINDAS, the level of ambition for the present project has been increased, and the potential of the unique neutron beam facility at The Svedberg Laboratory in Uppsala can be fully exploited.

3 Experimental setup and techniques

3.1 The TSL neutron beam facility

At TSL, quasi-monoenergetic neutrons are produced by the reaction $^7\text{Li}(p,n)^7\text{Be}$ in a $^7\text{Li}$ target bombarded by 50–180 MeV protons from the cyclotron, as is illustrated in Fig. 1 (Condé et al., 1990, Klug et al., 2000). After the target, the proton beam is bent by two dipole magnets into an 8 m long concrete tunnel, where it is focused and stopped in a well-shielded Faraday cup, which is used to measure the proton beam current. A narrow neutron beam is formed in the forward direction by a system of three collimators, with a total thickness of more than four metres.

The energy spectrum of the neutron beam consists of a high-energy peak, having approximately the same energy as the incident proton beam, and a low-energy tail. About half of all neutrons appear in the high-energy peak, while the rest are roughly equally distributed in energy, from the maximum energy and down to zero. The thermal contribution is small. The low-energy tail of the neutron beam can be reduced using
time-of-flight (TOF) techniques over the long distance between the neutron source and
the reaction target (about 8 m).

The relative neutron beam intensity is monitored by integrating the charge of the
primary proton beam, as well as by using thin film breakdown counters, placed in the
neutron beam, measuring the number of neutron-induced fissions in $^{238}$U (Prokofiev et
al., 1999).

Two multi-purpose experimental setups are semi-permanently installed at the neu-
tron beam line, namely MEDLEY and SCANDAL. These will be described below.

### 3.2 The MEDLEY setup

The MEDLEY detector array (Dangtip et al., 2000), shown in Fig. 2, is designed for
measurements of neutron-induced light-ion production cross sections of relevance for
applications within ADS and fast-neutron cancer therapy and related dosimetry. It
consists of eight particle telescopes, installed at scattering angles of 20° — 160° with
20° separation, in a 1 m diameter scattering chamber, positioned directly after the last
neutron collimator. All the telescopes are fixed on a turnable plate at the bottom of the
chamber, which can be rotated without breaking the vacuum.

Each telescope (Fig. 3) is a $\Delta E - \Delta E - E$ detector combination, where the $\Delta E$
detectors are silicon surface barrier detectors with thicknesses of 50 or 60 \( \mu \text{m} \) and 400
or 500 \( \mu \text{m} \), respectively, while the $E$ detector is a 50 mm long inorganic CsI(Tl) crystal,
tapered over the last 20 mm to fit the 18 x 18 mm photodiode readout. $\Delta E - \Delta E$ or
$\Delta E - E$ techniques are used to identify light charged particles (p, d, t, $^3$He, $\alpha$). The
chosen design gives a sufficient dynamic range to distinguish all charged particles from
a few MeV up to more than 100 MeV.

The solid angle of the telescopes is defined by active collimators, designed as thin
hollow plastic scintillator detectors, mounted on small photomultiplier tubes. A signal
from such a detector is used to veto the corresponding event, thereby ensuring that only particles that pass inside the collimator are registered.

Energy calibration of the silicon detectors is performed by determining the pulse height for the various particles at the point where they start to punch through the detectors, with the assumption of linear correspondence between pulse height and energy. Alpha particles from a $^{241}$Am source are used to check the calibration curve for the first, thin $\Delta E$ detector. For the CsI detectors, the linear response assumption is no longer valid, and the calibration curve is determined particle by particle by plotting the calculated energy, derived from the energy deposited in the second silicon $\Delta E$ detector and standard stopping power data, versus the measured pulse height. The obtained calibration is checked by comparing with pulse heights of resolved states in, e.g., $^{12}$C(n,p) and (n,d) reactions, for which the energies are known. Adding the energy losses in the three detectors gives the incident energy for each charged particle. The energy resolution is typically about 2 MeV at 80 MeV.

Absolute cross section normalization is obtained by comparison with free $np$ scattering, using a CH$_2$ target. After proper subtraction of target-out and $^{12}$C(n,xp) background contributions, the cross section per count can be determined from the $np$ scattering peak, using data previously taken at a similar energy (Rönnqvist et al., 1992, Olsson et al., 2000). The normalization coefficient is then applied to the data for the target under study to get the absolute cross section.

### 3.3 The SCANDAL setup

The SCANDAL setup (Klug et al., 2000) is primarily intended for studies of elastic neutron scattering, i.e., (n,n) reactions. Neutron detection is accomplished via conversion to protons by the H(n,p) reaction. In addition, (n,xp) reactions in nuclei can be studied by direct detection of protons. This feature is also used for calibration, and the setup has therefore been designed for a quick and simple change from one mode to the other.

The device is illustrated in Fig. 4. It consists of two identical systems, in most cases located on each side of the neutron beam. The design allows the neutron beam to pass
through the drift chambers of the right-side setup, making low-background measurements close to zero degrees feasible.

In neutron detection mode, each arm consists of a 2 mm thick veto scintillator for fast charged-particle rejection, a neutron-to-proton converter which is a 10 mm thick plastic scintillator, a 2 mm thick plastic scintillator for triggering, two drift chambers for proton tracking, a 2 mm thick $\Delta E$ plastic scintillator, which is also part of the trigger, and an array of 12 large CsI detectors for energy determination. The trigger is provided by a coincidence of the two trigger scintillators, vetoed by the front scintillator. The compact geometry allows a large solid angle for protons emitted from the converter. Recoil protons are selected using the $\Delta E$ and $E$ information from the plastic scintillators and the CsI detectors, respectively.

The response of the equipment has been carefully studied using carbon (March 1999) and CH$_2$ (November 1999) targets. The energy resolution is about 3.7 MeV (FWHM), which is sufficient to resolve elastic and inelastic scattering in several nuclei. The angular resolution is calculated to be about 1.4° (rms) when using a cylindrical scattering sample of 5 cm diameter.

When SCANDAL is used for (n,xp) studies, the veto and converter scintillators are removed. A multitarget arrangement can be used to increase the target content without impairing the energy resolution, which is typically 2.5 MeV (FWHM). This multitarget box allows up to seven targets to be mounted simultaneously, interspaced with multiwire proportional counters (MWPC). In this way it is possible to determine in which target layer the reaction took place, and corrections for energy loss in the subsequent targets can be applied (Thun et al., 2000). In addition, different target materials can be studied simultaneously, thus facilitating absolute cross section normalization by filling a few of the multitarget slots with CH$_2$ targets. The first two slots are normally kept empty, and used to identify charged particles contaminating the neutron beam.

The response in (n,xp) mode was tested in March 2000 by measuring $np$ scattering, using CH$_2$ targets. The result is shown in Fig. 5, where it can be compared to a previous measurement of that cross section with a magnetic spectrometer (Rönnqvist
et al., 1992, Olsson et al., 2000). As can be seen, the data sets agree well within the statistical uncertainties.

4 Results and analysis

4.1 Elastic scattering

Elastic scattering of neutrons on $^{208}\text{Pb}$ was studied in May, 2000. Since natural lead only contains about 50% $^{208}\text{Pb}$, we acquired an amount of thorium ore, highly enriched in $^{232}\text{Th}$, on the international market. In this radioactive ore, $^{208}\text{Pb}$ is the end product of $^{232}\text{Th}$ decay. To extract a pure sample of lead, a contract was signed between STCU and Institute of Colloid and Water Chemistry in Ukraine, and Uppsala University, in which the Ukrainian partners took on responsibility to process the ore. Thus, in early May we received about 400 g of lead, enriched to 88% in $^{208}\text{Pb}$. The material was casted in the shape of a cylinder, to be useful in the scattering measurements.

Data were collected for about one week of beam time, which was used for measurement with the lead target, but also CH$_2$ and carbon targets were used for absolute cross section determination. In addition, sample-out background measurements were performed. The two arms of SCANDAL were placed to cover the angular ranges 10° — 50° and 30° — 70°, respectively.

The analysis of the scattering data has recently started, and angular distributions are not yet available. However, a preliminary energy spectrum of neutrons scattered at 9° is shown in Fig. 6. The tail on the right side of the elastic peak is not only the result of inelastic scattering, but is also affected by the response of the converter scintillator. The dotted curve illustrates the sample-out background, normalized to the same incident neutron flux as the lead data. As can be seen, this background is very small.

Some work remains before a complete angular distribution in the region 10° — 70° can be presented. Since the cross section falls off several orders of magnitude within this
interval, it might be necessary to complement the data with a new measurement for the largest angles. It will be very interesting to see how well these data can be described by recent optical model representations (Koning).

4.2 \( (n,xlcp) \) reactions

In April 1999 and February 2000, we performed experiments to measure double differential cross sections \( \frac{d^2\sigma}{d\Omega dE} \) for protons and other light charged particles \((d, t, ^3He, \alpha)\) emitted in reactions of 100 MeV neutrons on enriched \(^{208}\text{Pb}\) targets. The charged particles were detected using MEDLEY, which allowed to measure continuum energy distributions in the forward direction \((10^\circ - 80^\circ)\). At larger angles, in view of the relatively low intensity of the neutron beam and of the estimated small cross sections, only the low-energy part of the spectra could be measured \((E_p < 40 \text{ MeV at } \theta = 160^\circ)\). To improve the counting rate at backward angles, at least for protons, we also used the multitarget arrangement together with the two arms of SCANDAL, which covered the angular range \(10^\circ - 140^\circ\) in two settings. With this setup, the high-energy part \((E_p > 30 \text{ MeV})\) of the proton spectra could be measured also at backward angles.

For the MEDLEY measurements, we used a 25 mm diameter by 0.5 mm thick lead target, enriched to 88% in \(^{208}\text{Pb}\). Figs. 7a and b show typical \(\Delta E_1 - \Delta E_2\) and \(\Delta E_2 - E\) scatter plots, respectively, from 96.5 MeV neutron-induced charged-particle production reactions in lead at \(20^\circ\). The energy threshold of the telescopes was about \(2 - 3 \text{ MeV}\) for the hydrogen isotopes and about \(9 \text{ MeV}\) for the helium isotopes. Preliminary results from MEDLEY are presented in Figs. 8 and 9. The analysis is in progress. Fig. 8 shows the double differential cross section for protons emitted at \(20^\circ\) (filled circles) and \(40^\circ\) (open circles), while Fig. 9 displays the double differential cross section for protons (filled circles), deuterons (filled squares) and tritons (open circles) emitted at \(20^\circ\).

In the SCANDAL measurement the multitarget contained five lead foils (each 220
Figure 7: (a) $\Delta E$ vs. $\Delta E$ and (b) $\Delta E$ vs. $E$ scatter plots for $^{208}\text{Pb}(n,\alpha\gamma)$ reactions at 96.5 MeV.

Figure 8: Preliminary double-differential cross section for the $^{208}\text{Pb}(n,\alpha\gamma)$ reaction with $E_n = 96.5$ MeV, and at 20° (filled circles) and 40° (open circles).

$\mu$m thick), one carbon foil (360 $\mu$m) and one CH$_2$ foil (360 $\mu$m). The carbon target was used to subtract the background contribution of protons from quasi-free neutron scattering in the carbon of CH$_2$. In addition to normalization, the $np$ scattering data allowed to calibrate the CsI detectors using the well-defined two-body kinematics of that process. With the simultaneous measurement of the $np$ scattering and Pb($n,\gamma$) processes, the detection efficiency problem is in principle avoided. One needs, however, to evaluate the solid angle of the CsI detectors viewing the different finite targets. For
these evaluations, we performed a simulation of the setup using the GEANT code of CERNLIB. Preliminary results of proton production above 35 MeV in lead are given for the scattering angles 18°, 24°, 35° and 55° in Fig. 10.

Similar measurements have been performed for $^{56}$Fe(n,X) at 100 MeV, using both SCANDAL (February 2000) and MEDLEY (May 2000). These data will be analysed after the corresponding work on the lead data has been completed.

5 International activities

5.1 Collaboration

The Uppsala group participates since 1998-08-01 in a CEC supported two-year Concerted Action, called “Physical aspects of lead as a neutron-producing target for accelerator transmutation devices”. The aim of the project is to collect and structure available information on lead, and to make suggestions on what additional data are needed for this target material. The project is organized in ten work packages, of which our group is fully or partly involved in four.

The third and the fourth (and last) semi-annual meetings with the partners of the Concerted Action were held in Brussels 2000-01-28–29 and 2000-06-23–24, respectively, with Nils Olsson representing the Uppsala group. Progress reports were given by the various groups, and during the last meeting a final project report was drafted. This report will be delivered to the CEC in September.

The Concerted Action has been followed up by a proposal for a European collaboration on nuclear data for ADT, which was submitted to the 5th CEC program on October 4. The proposal, “High- and Intermediate Energy Nuclear Data for Accelerator-Driven Systems (HINDAS)”, involves 16 European institutions from Belgium, France, Germany,
Figure 10: Preliminary double-differential cross section for the $^{208}\text{Pb}(n,xp)$ reactions at $E_n = 96.5$ MeV and $\theta = 18^\circ$, $24^\circ$, $35^\circ$ and $55^\circ$.

The Netherlands, Spain, Sweden and Switzerland, and the experimental work will be performed at six European laboratories (UCL in Louvain-la-Neuve, TSL in Uppsala, KVI in Groningen, PSI in Villigen, COSY at Jülich and GSI in Darmstadt). Work on the theoretical interpretation of the experimental results is also included. The project is coordinated by Prof. Jean-Pierre Meulders, Louvain-la-Neuve, Belgium.

HINDAS was positively received by the CEC, and was approved at a level of 2.1 MEUR. Of this, 210 kEUR falls on the Uppsala partner, while the collaborators that use the TSL neutron facility get: Subatech, Nantes (150 kEUR), LPC, Caen (150 kEUR), ZSR, Hannover (150 kEUR), PTB, Braunschweig (36 kEUR). Most of the money is intended for PhD students or postdocs. This means an increasing engagement for the Uppsala group and TSL, but also more focus on the activities here.

The project will start 2000-09-01 with the kick-off meeting in Brussels 2000-10-09, and run over three years.

For the Uppsala partner, a substantial fraction of the grant will be used to employ a postdoc, who can act as liaison between the Uppsala group and the collaborating groups. However, he/she will also strengthen the experimental competence in Uppsala, and be a complement to the existing supervisors at INF.

To our judgement, the proposal is well organized and focused. It involves a major part of the competence and equipment available in Europe, and will also contribute to
the development of nuclear data activities in Europe, by bringing new scientists into this area.

During August and December 1999, Jan Blomgren and four students, including Cecilia Johansson and Joakim Klug, participated in an experiment at Indiana University Cyclotron Facility, located in Bloomington, Indiana, USA. The experiment concerns neutron-proton scattering, which is closely related to our own activities, but with a rather different experimental technique. Such journeys are very beneficial for the students, who learn a lot by participating in the setting up of a complicated experiment, and also get experience of work in an international environment.

5.2 Meetings and conferences

Accelerator-based research in Uppsala celebrated its 50th anniversary on December 8, with a half-day symposium and a dinner. The celebration attracted some 200 participants. The symposium included talks on historical views, but also five talks about present and future research. One of these, "Applied neutron physics", was given by Nils Olsson.

The International Nuclear Data Committee (INDC) of the Nuclear Data Section (NDS) at IAEA held its 23rd meeting in Vienna 2000-05-24-26. During the meeting, the performance of the NDS was reviewed, and proposals for Co-ordinated Research Projects (CRP) and other data development projects were discussed. Among the eight projects recommended by the INDC, one was related to creating a nuclear reaction data base for accelerator applications. During the meeting, Nils Olsson gave a report on the Nuclear Data Research Activities in Sweden.

6 Administrative matters

6.1 Personnel and PhD students

From 2000-01-01 the Department of Neutron Research (INF) got a new research group, specialized on studies of nuclear structure and gamma-ray detection. This group is headed by Johan Nyberg, who has received a six-year position as senior researcher from NFR. The group includes a junior researcher, Matthias Weiszvlog, also sponsored by NFR, and a PhD student. With this group, INF strengthens its competence in the mentioned areas of research.

In May, the professors chair in applied nuclear physics at INF was finally filled, after a vacancy period of five years. The new professor is Jan Källne, who is leading a large group in neutron diagnostics of fusion plasmas. This is a great step forward for the department, and it means that the subject of applied nuclear physics now is fully accepted and recognized at the university.

INF has had two PhD dissertations during the year, namely Marco Tardocchi who has worked with neutron diagnostics of fusion plasmas, and Anders Axelsson who spent his research efforts on nuclear structure. In September 2000, Somsak Dangtip will defend his thesis, which is related to cross section measurements of medical relevance, measured using MEDLEY.
The supervision within the present project is performed to limited extent by the project leader, Nils Olsson, and to a larger extent by Ayşe Ataç and Jan Blomgren. In the fall of 2000, a postdoc will also be employed using resources within the HINDAS project. Two PhD students are directly connected to and financed by the present project, namely Cecilia Johansson and Joakim Klug, which both are connected to the research school AIM (Advanced Instrumentation and Measurements). Two other students, Bel Bergenwall who is financed by AIM, and Udomrat Tippawan with a scholarship from Thailand, have tasks strongly related to the present project, and especially to the line of development emerging from the collaboration with the French groups within HINDAS.

Members of our group participate in several courses on nuclear physics as well as on energy technology. Some of these include problems related to transmutation. Also more outreach talks, seminars, articles and interviews related to this project have been given.

6.2 Reference group

The third and fourth reference group meetings, with participation by Per-Eric Ahlström (SKB), Benny Sundström (SKI), Thomas Lefvert (Vattenfall AB), Fredrik Winge (BKAB) and Anders Ringbom (FOA), were held at SKB in Stockholm 2000-01-14, and in Uppsala 2000-06-15, respectively. Scientific and administrative reports on the progress of the project were given at these meetings.

In addition to the meetings, the progress of the work is continuously communicated to the reference group members by short, written, quarterly reports.

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