Neutron-Induced Light-Ion Production from Iron and Bismuth at 175 MeV

RICCARDO BEVILACQUA
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Abstract


Radioactive waste management is one of the key issues in sustainability of nuclear energy production. Geological repositories represent today the most appropriate solution to long-term management of high-level radioactive waste. Strategies such as Partitioning and Transmutation of spent nuclear fuel may offer a positive impact on geological repositories, by reducing the mass of transuranium elements to be disposed and the time scale for their radioactivity. In this scenario, Accelerator Driven Systems (ADS) may play a relevant role as dedicated burners towards sustainable nuclear energy.

The NEXT project at Uppsala University contributes to a European effort to improve nuclear data knowledge for transmutation, providing the first experimental neutron induced data in the 100 to 200 MeV energy region. This thesis presents measurements of double-differential cross sections for production of light-ions in the interaction of 175 MeV quasi-monoenergetic neutrons with Fe and Bi. Results are compared with model calculations obtained with state-of-the-art nuclear reaction codes; TALYS-1.2, a modified version of JQMD, and MCNP6. Special focus in this work is given to pre-equilibrium emission of composite light-ions. A new energy dependence in the mechanisms described by the Kalbach systematics used in TALYS to account for composite particle emission in the pre-equilibrium stage is proposed. Data show also the need to include multiple pre-equilibrium emission of composite particles, a mechanism now included in TALYS only for protons and neutrons. The JQMD code was recently modified to include a surface coalescence model in the quantum molecular dynamics description of the formation of composite particles. Comparisons of the measured data with results from this modified JQMD code confirm the importance of coalescence mechanisms for the description of the emission spectra of composite particles. Finally, the neutron-induced data are compared with MCNP6 calculations, to contribute to the process of validation and verification of the code.

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urn:nbn:se:uu:diva-149999 (http://urn.kb.se/resolve?urn=urn:nbn:se:uu:diva-149999)
I haven’t got any special religion this morning. My God is the God of Walkers.
If you walk hard enough, you probably don’t need any other god.
(Bruce Chatwin, In Patagonia)

Our battered suitcases were piled on the sidewalk again;
we had longer ways to go. But no matter, the road is life.
(Jack Kerouac, On the road)
List of Papers

This thesis is based on the following papers, which are referred to in the text by their Roman numerals.

A scientific collaboration is a thermodynamic system, in which the contribution of each participant is in some way inseparable from the one of others. I had some primary responsibilities in this work, indicated for each paper as “My contribution”. However, my participation is not limited to the listed items, nor would it be fair to assume that I have solely merit for any part of this research. When using the first singular person “I” in this list and in the discussion of the Thesis, I acknowledge the scientific contribution of all my colleagues and co-authors.


My contribution: I wrote the paper and carried out most of the analysis described in it. I had an active role in all measurements for light-ion production with Medley since February 2007.


My contribution: I wrote the paper and performed most of data reduction and analysis of Fe and Bi data. I had an active role in the measurements. I have produced TALYS calculations.


**My contribution:** I wrote the paper and performed model calculations. I had a leading role in the study reported in this work and I presented it at the International Nuclear Physics Conference (INPC2010), in Vancouver (Canada).


**My contribution:** I wrote the paper, performed TALYS calculations and led the work on Kalbach systematics. I presented this paper at the 2010 Symposium on Nuclear Data of the Atomic Energy Society of Japan, in Fukuoka (Japan).


**My contribution:** I have been technically responsible person and shift leader in the experimental campaign. I have performed TALYS calculations presented in the paper.


**My contribution:** I had an active role in the experimental run.

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List of papers related to this Thesis, but not included in the comprehensive summary. I am first author or co-author of all listed papers. I have participated in the experimental activities and/or collaborated to data analysis of the results presented in these works.


15. S. Pomp et al. (2011) A Medley with over ten years of (mostly) light-ion production measurements at The Svedberg Laboratory, EFNUDAT - Proceedings of the Scientific Workshop on Measurements and Models of Nuclear Reactions, *EPJ Web of Conferences*.


<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>ADS</td>
<td>Accelerator Driven System</td>
</tr>
<tr>
<td>CEM</td>
<td>Cascade-Exciton Model</td>
</tr>
<tr>
<td>EM</td>
<td>Exciton Model</td>
</tr>
<tr>
<td>HPRL</td>
<td>High Priority Request List</td>
</tr>
<tr>
<td>ICM</td>
<td>Ionization Chamber Monitor</td>
</tr>
<tr>
<td>INC</td>
<td>Intra-Nuclear Cascade</td>
</tr>
<tr>
<td>KO</td>
<td>Knock-Out</td>
</tr>
<tr>
<td>NT</td>
<td>Nucleon Transfer</td>
</tr>
<tr>
<td>PE</td>
<td>Pre-Equilibrium</td>
</tr>
<tr>
<td>P&amp;T</td>
<td>Partitioning and Transmutation</td>
</tr>
<tr>
<td>QMD</td>
<td>Quantum Molecular Dynamics</td>
</tr>
<tr>
<td>QMN</td>
<td>Quasi-Monoenergetic Neutron</td>
</tr>
<tr>
<td>SCM</td>
<td>Surface Coalescence Model</td>
</tr>
<tr>
<td>TFBC</td>
<td>Thin Film Break-down Counter</td>
</tr>
<tr>
<td>TOF</td>
<td>Time Of Flight</td>
</tr>
<tr>
<td>TSL</td>
<td>The Svedberg Laboratory</td>
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1. Introduction

Apples from Chernobyl are good. If you bury the core deep enough.
(Anonymous Ukrainian farmer)

I really don’t believe in magic.
(J. K. Rowling)

Sustainability is today the major driver in nuclear reactor research. Dr. Joachim Knebel from the Karlsruhe Institute of Technology, at the recent 7th European Commission conference on Euratom research and training in reactor systems, affirmed that

"The major issue in sustainability is the issue of waste."

Generation II nuclear reactors (now in operation) are inefficient when considering utilization of uranium: fuel rods need to be replaced when only a small fraction of the uranium is consumed. Whereas Generation III reactors will make a more efficient use of the nuclear fuel, only the future Generation IV will be able to make use of most of the uranium and to reduce the production of high-level radioactive waste. Generation IV reactors will enter in operation not earlier than 30 years from now. Even though this new generation of reactors will be able to burn off a relevant amount of minor actinides and plutonium, until Generation IV will enter in operation there is a need to provide a solution for the present production of spent nuclear fuel.

According to a report published in 2008 by the European Commission [1], 70% of the population in the EU believes that there is no safe way to deal with radioactive waste. A main finding of the report is that

"More than 9 in 10 (93%) Europeans on average see an urgent need to finding a solution to the nuclear waste issue now, rather than leaving it unresolved for later generations."

The Swedish Nuclear Fuel and Waste Management company (SKB, Svensk Kärnbränslehantering AB) has submitted an application to build a deep geological repository for residual high-level waste from Swedish nuclear power plants. Geological repositories represent today the most appropriate solution

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to long-term management of high-level radioactive waste [2]. However, public perception of nuclear energy can be improved by proving the existence and viability of a technical methodology able to reduce the amount of high-level radioactive waste to be disposed in the repository.

Partitioning and Transmutation (P&T) is a methodology that may reduce the mass of transuranium elements from spent nuclear fuel to be disposed in the geological repository by more than two orders of magnitude. Applying P&T, the radiotoxicity of nuclear waste may reach the level corresponding to uranium ore within 1000 years, whereas at least 100000 years would be needed to reach the same level for unprocessed spent fuel. The main advantage of P&T consists in the possibility to obtain a reduction of the thermal load of processed spent fuel [3]; this heat load reduction would ease the disposal of larger amounts of waste in the same repository. P&T may also contribute to reduce the potential proliferation interest of geological repositories. An overview on the P&T impact on high-level nuclear waste management is given by González-Romero [4].

P&T is a two step methodology: in a first stage elements in the spent fuel are chemically separated (in particular actinides from lanthanides), and in a second stage the actinides are irradiated with neutrons to obtain short-lived fission products. Transmutation and its effects on the fuel cycle are discussed in several works (see e.g. Salvatore [5]).

Accelerator Driven Systems (ADS) are subcritical nuclear reactors coupled to a proton accelerator and a spallation target. ADS may be used as dedicated burner reactors for transmutation, however the system may also be designed for production of energy. Designing a roadmap for accelerator driven transmutation of nuclear waste in Europe, the European Technical Working Group on ADS, lead by Nobel Laureate Carlo Rubbia, identified the advantages of a sub-critical system over a critical reactor [6]:

"Critical reactors, however, loaded with fuel containing large amounts of minor actinides pose safety problems caused by unfavourable reactivity coefficients and small delayed neutron fraction. [...] ADS is particularly favorable and allows a maximum transmutation rate while operating in a safe manner. An advantage of accelerator driven systems is that, since there is no criticality condition to fulfill, almost any fuel composition can be used in the system."

In a report of the Nuclear Science Committee/Nuclear Energy Agency, Koning et al. [7] identify a nuclear data High Priority Request List (HPRL). Here, measurements of neutron and proton production cross sections for energies up to 200 MeV, from a selected number of target materials, are indicated as most relevant for ADS applications.
Table 1.1: List of target nuclides included in the HPRL list [7]. Bi is included in the Japanese HPRL [8], but not in the European one.

<table>
<thead>
<tr>
<th>Target nuclides</th>
<th>application / motivation</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{16}\text{O}$</td>
<td>medical applications, dosimetry</td>
</tr>
<tr>
<td>$^{27}\text{Al}$, $^{56}\text{Fe}$, $^{58}\text{Ni}$</td>
<td>structural materials</td>
</tr>
<tr>
<td>$^{90}\text{Zr}$</td>
<td>&quot;nuclear model&quot; material</td>
</tr>
<tr>
<td>$^{100}\text{Mo}$</td>
<td>fission product</td>
</tr>
<tr>
<td>$^{184}\text{W}$, $^{208}\text{Pb}$, $^{209}\text{Bi}$</td>
<td>candidate target materials in ADS</td>
</tr>
<tr>
<td>$^{232}\text{Th}$, $^{238}\text{U}$</td>
<td>actinides</td>
</tr>
</tbody>
</table>

The HPRL indicates the need to obtain complete sets of data:

"This is of critical importance to nuclear model calculations. If elastic and total (reaction) cross sections for both neutrons and protons [...] as well as a complete set of (p,xn,), (p,xp), (n,xn) and (n,xp) cross sections are available, code developers can narrow down the uncertainties in their calculations, which has an immediate, positive impact on predictions for other nuclides where no experimental data exists."

A request for neutron-induced and proton-induced production cross sections of composite light ions (deuteron, triton, $^{3}\text{He}$ and $\alpha$ particle) is also included in the HPRL. The target nuclei indicated by the HPRL are presented in Table 1.1.

In Table 1.2 are listed the experimental facilities in the world offering suitable quasi-monoenergetic (QMN) neutron beams for cross section measurements with incident energies above 20 MeV. One of these facilities is located in Uppsala: the The Svedberg Laboratory (TSL), where a QMN beam line with peak neutron energy up to 175 MeV is in operation.

The NEXT (Neutron data EXperiments for Transmutation) project was initiated in 2006 by the Department of Neutron Research$^2$, Uppsala University, and supported as a research task agreement by the Swedish Nuclear Power Inspectorate$^3$, SKB and Ringhalsverket AB. The aim of the NEXT project was to provide neutron-induced data, in the 100 to 200 MeV region, relevant for ADS-based transmutation of spent nuclear fuel. NEXT was following previous research projects by the same research group, where neutron-induced data have been measured for energies up to 100 MeV.

The experimental activity was primarily carried out at TSL, where the Medley spectrometer and the SCANDAL setup were installed. Medley is devoted

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$^2$Since January 2008, Division of Applied Nuclear Physics
$^3$Since July 2008, Swedish Radiation Safety Authority
Table 1.2: *Experimental facilities offering quasi-monoenergetic neutron beams with peak energies above 20 MeV.*

<table>
<thead>
<tr>
<th>Facility</th>
<th>Country</th>
<th>Neutron energies (MeV)</th>
<th>notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>The Svedberg Laboratory</td>
<td>Sweden</td>
<td>20 to 175</td>
<td></td>
</tr>
<tr>
<td>iThemba LABS</td>
<td>South Africa</td>
<td>25 to 200</td>
<td></td>
</tr>
<tr>
<td>CYRIC at Tohoku University</td>
<td>Japan</td>
<td>20 to 90</td>
<td></td>
</tr>
<tr>
<td>TIARA (JAEA)</td>
<td>Japan</td>
<td>40 to 90</td>
<td></td>
</tr>
<tr>
<td>Ring Cyclotron facility at RCNP</td>
<td>Japan</td>
<td>100 to 400</td>
<td></td>
</tr>
<tr>
<td>Cyclotron Research Centre at UCL</td>
<td>Belgium</td>
<td>25 to 70</td>
<td>not active</td>
</tr>
</tbody>
</table>

to the measurement of neutron-induced light-ion production cross sections, whereas SCANDAL is used to measure neutron elastic scattering.

Light-ion production from C, O, Si, Fe, Bi and U at 175 MeV QMN was measured with the Medley setup at TSL, from 2007 until 2009, in the frame of the NEXT project and in collaboration with Kyushu University (Japan), Chiang Mai University (Thailand), Université de Caen (France) and KTH - Royal Institute of Technology (Sweden). The experimental activity with Medley included also the characterization of the QMN spectrum and of the white neutron source ANITA (Atmospheric-like Neutrons from thick Target).

These measurements represent the first data available in the 100 to 200 MeV energy region for neutron-induced light-ion production, whereas several measurements are available for proton-induced reactions in the same energy interval (e.g. [9, 10, 11]).

The present work

In this Thesis, I present double-differential cross sections for light-ion production from Fe and Bi at 175 MeV QMN. These results are relevant for development of accelerator driven transmutation of nuclear waste. Fe is an important construction material in nuclear reactors and accelerators, and Bi has been considered as spallation target material in several ADS designs. Measurements on Fe and Bi are important for the development of nuclear models and reaction codes: the most abundant isotope of iron is $^{56}$Fe, an even-even nucleus with high binding energy (∼8.8 MeV/A); bismuth is naturally mono-isotopic, $^{209}$Bi, has a *magic number* of neutrons (126) and differs by just one proton from the double-magic $^{208}$Pb.

I describe the experimental setup, data reduction procedures and characterization of the QMN spectrum at TSL. Measured double-differential cross-sections are presented in comparison with model calculations; these were ob-
tained with state-of-the-art nuclear reaction codes: TALYS-1.2 [12], a modified version of JQMD [13] and MCNP6 [14]. Special focus in this work is given to pre-equilibrium emission of composite light ions.

To investigate discrepancies between TALYS results and neutron-induced data, TALYS calculations are compared also with proton-induced data at 120, 160, 175 and 200 MeV, for target nuclides close in mass to Fe and Bi. I discuss a possible energy dependence in the mechanisms described by Kalbach systematics [15] to account for composite particle emission in the pre-equilibrium stage. Multiple pre-equilibrium emission of composite particles is also discussed.

I present comparison of Fe data at 175 MeV with the JQMD code, modified by Watanabe and Kadrev [13]. The modified code includes a surface coalescence model to complement quantum molecular dynamics production of composite particles.

Comparison of MCNP6 code calculations with our experimental results are presented as part of the validation and verification process of the CEM03.02 event generator of this Monte Carlo transport code.

The results presented in this Thesis will provide benchmark points for theoretical models and other nuclear reaction codes, and will help to produce reliable evaluated data needed for the development of ADS.
Part I:
Materials and methods

In Chapter 2, I give a short description of the Medley experimental setup, installed at the The Svedberg Laboratory, in Uppsala (Sweden). Medley was initially designed for measurements of light-ion production induced by neutrons with energies up to 100 MeV, and it is now used for measurements with neutrons up to 175 MeV. The upgrade of Medley is described in Paper I.

In Chapter 3, I summarize procedures for data reduction and analysis, for measurements with Medley. Calibration and response function of CsI(Tl) scintillators, reduction of the low-energy tail contribution from the QMN beam, analysis of background and normalization to absolute cross section of measured data are discussed in Paper I. Thick target correction and neutron spectrum measurements are described in Paper VI.

In Chapter 4, I introduce model calculations used in comparison with measured data. These calculations, reported in Paper II, were obtained with the TALYS reaction code, with a modified version of JQMD and with the Monte Carlo particle transport code MCNP6. Model calculations are also reported in Papers III, IV and V.
2. Experimental setup

Computer, compute to the last digit the value of $\pi$.

*(Mr. Spock, Star Trek)*

2.1 The Svedberg Laboratory

The experimental work presented in this Thesis has been conducted at the The Svedberg Laboratory (TSL), located in Uppsala (Sweden).

TSL is a university facility established in 1986 from the scientific tradition of the former Gustav Werner Institute. A cyclotron has been in operation in Uppsala since the end of 1940s. A quasi-monoenergetic neutron (QMN) beam line was commissioned and operated at TSL since late 1980s until 2003 [16], when the QMN line underwent a major upgrade. A new QMN line is now in operation and provides neutrons with energies up to 175 MeV.

The QMN beam line at TSL is described by Prokofiev et al. [17]. Neutrons are produced via the $^7\text{Li}(p,n)$ reaction: protons are accelerated in a cyclotron, extracted and focused in a beam; the proton beam is transported to the experimental hall (Blue Hall), where it interacts with a $^7\text{Li}$ target. The thickness of the 99.99% enriched $^7\text{Li}$ target can be chosen depending on the energy of the incident proton beam and the experimental requirements. 175 MeV QMN are obtained from 180 MeV protons interacting with a 23.5 mm-thick $^7\text{Li}$ target.

Neutrons shaped in a beam are transported to the irradiation area. The residual proton beam is deflected by a bending magnet to a beam dump. The proton current is monitored integrating the dumped beam in a Faraday cup. The neutron beam intensity is monitored by an ionization chamber monitor (ICM) and by a thin film break down counter (TFBC); both these neutron monitors utilize neutron-induced fission of $^{238}\text{U}$.

The layout of the Blue Hall at TSL is presented in Fig. 2.1. In my Licentiate Thesis [18] I describe the collimation system and provide detailed drawings of the neutron line.
2.2 Medley

Medley is a spectrometer system for light-ion detection in neutron-induced reactions. Medley has been semi-permanently installed at TSL for the last ten years. An overview of the activities with Medley at TSL is given by Pomp et al. [19].

An extensive campaign of measurements with QMN at 96 MeV has been conducted with the Medley setup and is now completed: inclusive double-differential cross-sections $\sigma(E,\theta)$ for production of protons, deuterons, tritons, $^3$He and $\alpha$ particles from C [20], O [21], Si [22], Fe, Pb and U [23] have been published; data are now available on the EXFOR database. Dangtip et al. [24] provides a detailed description of the Medley spectrometer in the configuration for the measurements at 96 MeV QMN.

In 2007 Medley has been upgraded to detect light-ions produced in neutron-induced reactions at 175 MeV; since then, Medley was used to measure inclusive $\sigma(E,\theta)$ for production of light ions from C [25], O [Paper V], Si [26], Fe [Paper II], Bi [Paper II] and U at 175 MeV QMN.

Figure 2.1: Layout of the Blue Hall at TSL, where the QMN beam line is installed (D-line). The cyclotron is situated in the upper left side, outside the image.
2.2.1 The spectrometer system

The Medley spectrometer consists of eight three-elements telescope detectors. The telescopes are installed in an evacuated scattering chamber, positioned symmetrically downstream the neutron beam. A reaction target is positioned at the center of the chamber. The arrangement of the telescope detectors and of the reaction target in the Medley chamber are in Fig. 2.2 (a).

Each telescope consists of two fully depleted silicon surface barrier detectors and a CsI(Tl) scintillator. Particle identification and total kinetic energy measurement are based on the ΔE-E technique. The chosen configuration provides good particle separation, with low detection threshold (∼2 MeV for protons, ∼9 MeV for α particles) and wide dynamic range (up to 170 MeV). Construction details of the telescope detectors are presented in Fig. 2.2 (b).

2.2.2 Silicon detectors

The first silicon detector (Si₁) is 50 to 60 μm-thick; the thickness of Si₁ determines the energy threshold for identification of detected light ions. The second silicon detector (Si₂) of each telescope has been upgraded to 1000 μm-thick elements, replacing the 500 to 600 μm-thick ones previously installed in Medley. Whereas for measurements of light-ion production induced by neutrons with peak energy of 96 MeV the configuration with thinner Si₂ detectors was acceptable, the new 1000 μm-thick Si₂ detectors are required for measurements involving higher energies.

In Fig. 2.3: energy deposition in Si₂ (ΔE₂) for protons and deuterons is plotted against energy deposition E in the CsI(Tl) scintillator: results are presented for a 1000 μm-thick Si₂ (a) and for a 500 μm-thick Si₂ (b). The new thick detectors allow identification of particles over all the energy spectrum, whereas it is not possible to resolve the ambiguity above ∼80 MeV with the thin Si₂. The energy deposition of protons and deuterons in the detectors was calculated with the SRIM code [27] and plotted for comparison. The informa-
Figure 2.3: Contour plot of $\Delta E_2$ versus $E$ for protons (p) and deuterons (d); data from a 1000 $\mu$m-thick Si2 are plotted in the left panel (a), whereas in the right panel (b) a 500 $\mu$m-thick Si2 was used. Calculations with the SRIM code are plotted for comparison (solid lines).

2.2.3 CsI(Tl) scintillators
The third element of each telescope detector installed in the Medley spectrometer is a CsI(Tl) scintillator. This detector is 100 mm-long, has a diameter of 50 mm and it is designed to fully stop all the light-ions produced in the measured reactions at 175 MeV QMN. Calculations with the Geant code [28] and with the PHITS code [29] show that the chosen CsI(Tl) detector size and geometry are appropriate to measure the kinetic energy of incident protons up to 170 MeV.

2.2.4 Readout, electronics and data acquisition system
I described readout and electronics associated to the Medley setup in my Licentiate Thesis [18]; there I report also the general scheme of the electronics. The configuration used in the experiments at 175 MeV QMN is essentially the same as used in previous experiments with Medley at 96 MeV QMN. Information on the SVEDAQ data acquisition system used at TSL is available on-line [30].
Table 2.1: Some reaction targets installed in the Medley spectrometer.

<table>
<thead>
<tr>
<th>material</th>
<th>thickness (mm)</th>
<th>mass (mg)</th>
<th>notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si</td>
<td>0.96</td>
<td>1132 (±1)</td>
<td></td>
</tr>
<tr>
<td>Fe</td>
<td>0.375</td>
<td>1959.6 (±0.1)</td>
<td></td>
</tr>
<tr>
<td>Bi</td>
<td>0.5</td>
<td>3130.1 (±0.2)</td>
<td></td>
</tr>
<tr>
<td>CH₂</td>
<td>1.0</td>
<td>461.55 (±0.01)</td>
<td></td>
</tr>
<tr>
<td>CH₂</td>
<td>1.0</td>
<td>1017 (±1)</td>
<td>reference target for Fe and Bi runs</td>
</tr>
<tr>
<td>CH₂</td>
<td>5.0</td>
<td>2293 (±1)</td>
<td>reference target for Si runs</td>
</tr>
</tbody>
</table>

2.2.5 Reaction targets

A reaction target is installed at the center of the scattering chamber. A mechanic system allows to exchange targets without breaking the vacuum. Reaction targets installed in Medley are fully covered by the incident neutron beam. The thickness of the target is a compromise between a reasonable count rate, and the minimization of particle- and reaction-losses in the target itself. Thickness and mass of several targets installed in the Medley spectrometer are reported in Tab. 2.1.
3. Data reduction and analysis

*Do everything by hand, even when using the computer.*

*(Hayao Miyazaki, Anime Director)*

3.1 Particle identification

Particle identification and total kinetic energy measurements are based on the $\Delta E$-$E$ technique.

In the data reduction process it is only possible to identify light-ions with sufficient kinetic energy to reach $Si_2$. If a light-ion is fully stopped by $Si_2$, this detector is acting as $E$ detector, whereas $Si_1$ is acting as $\Delta E$ detector. When a light-ion has enough kinetic energy to reach the CsI(Tl) scintillator, both the silicon detectors act as $\Delta E$ detectors. The total kinetic energy is measured as sum of the energy deposition in all three detectors. A thin $Si_1$ provides a low energy threshold for particle identification; a thick $Si_2$ offers good particle separation, as seen in Fig. 2.3.

Particle selection was performed with graphical cuts around the characteristic bands in the two-dimensional ADC-channel spaces, obtained plotting the events as recorded in $Si_1$, in $Si_2$ and in the scintillators.

Energy deposition of protons, deuterons, tritons, $^3He$ and $\alpha$ particles in $Si_2$ plotted against their respective energy deposition in $Si_1$ is presented in Fig. 3.1 (a), whereas energy deposition of the same light-ions in the CsI(Tl) scintillator plotted against energy deposition in $Si_2$ is plotted in Fig. 3.1 (b). Only events selected by the graphical cut around the characteristic particle bands are plotted. Events below the identification threshold are excluded from the plot. Energy deposition in the detectors calculated with the SRIM code [27] is plotted for comparison.

3.2 Energy calibration

Energy calibration of the detectors is obtained from the data. The same calibration for all light-ions is applied to silicon detectors, whereas CsI(Tl) scintillators are calibrated independently for each light-ion.

Energy deposition of light-ions in each silicon detector, as function of the incident kinetic energy, is calculated with SRIM. Punch-through energy for each kind of light-ion and for each detector is determined from these calcu-
Figure 3.1: Two dimensional scatter plot of energy deposition in Si$_2$ versus energy deposition in Si$_1$ (a), and energy deposition in the CsI(Tl) scintillator versus energy deposition in Si$_2$ (b); experimental data are compared with energy deposition calculations obtained with the SRIM code (solid lines).

Light output of CsI scintillators is dependent on the mass of the detected particle [31, 32] and shows a non-linear response to energy deposition [33]. To calibrate hydrogen isotopes, I follow Horn et al. [32] and the approximation used by Dangtip et al. [24], assuming a quadratic relationship between energy deposition and light output. Data are fitted against the equation:

$$E = b(H - a_o) + cb^2(H - a_o)^2$$  \hspace{1cm} (3.1)

where $E$ is the energy deposition, $H$ the ADC channel, $a_o$ the offset, $b$ and $c$ the fitting parameters.

The offset $a_o$ is determined for each scintillator from the ADC channel corresponding to zero energy deposition. To determine the energy deposition $E$ in the scintillator, I have computed the residual kinetic energy of the light-ion associated to each measured event from the energy loss in the Si$_2$ detector. Moreover, (n,p) elastic events from peak neutrons measured with a CH$_2$ target provide a reference point at high energies, where the uncertainties associated with the energy loss in Si$_2$ are too large to offer a reliable calibration. Calibra-
tion is finally obtained by applying Eq. 3.1 to the data, where b and c are now constants, as determined from the fit.

To calibrate helium isotopes it is necessary to include the effect of quenching at very dense ionization: this effect gives a bending of the calibration curve at low energy [32]. Data are fitted against the equation:

\[ E = b(H - a_o) + c \ln \left| 1 + d(H - a_o) \right| \]  

(3.2)

where b, c and d are the fitting parameters, and \( a_o \) is the offset. As in the case of hydrogen isotopes, calibration is obtained by applying Eq. 3.2 to the data, with b, c and d constants determined from the fit.

### 3.3 Time-of-flight

A time-of-flight (TOF) gate is applied to registered events. This gate reduces the contribution of low-energy neutrons in the QMN spectrum.

TOF of detected light-ions is measured as the time difference between signal in \( \text{Si}_1 \), or in \( \text{Si}_2 \), and next signal from the cyclotron radiofrequency (RF). The TOF of each light-ion is associated to the measured total kinetic energy; considering mass and kinetic energy, TOF from the Medley reaction target to the triggering silicon detector is subtracted from the total TOF. The resulting TOF depends only on the kinetic energy of the incident neutron.

In principle, selecting events associated to the appropriate neutron TOF, would allow to select only light-ions produced by neutrons with a specific energy. However, there are limitations due to the experimental time resolution. The TOF FWHM of the \((n,p)\) elastic peak at 20 degrees in the laboratory system, measured from a \( \text{CH}_2 \) target with 175 MeV QMN, is equal to 8.8 ns. The repetition time between beam micropulses is of the order of 45 ns. In these experimental conditions the time resolution measured with the Medley setup does not allow to resolve events induced by 175 MeV peak neutrons from events induced by low-energy neutrons down to 80 MeV.

In Fig. 3.2, neutron TOF is plotted against the kinetic energy of protons from a 1 mm-thick \( \text{CH}_2 \) target. The peak at 153 MeV corresponds to \((n,p)\) elastic events from 175 MeV peak neutrons. The 153 MeV elastic peak is fitted by a Gaussian in the TOF space, and a gate with amplitude of \( 2\sigma \) is applied to all events. It is possible to observe bands corresponding to events induced by frame overlap neutrons.

The neutron spectrum at TSL has been measured with the proton recoil technique, at 20 degrees in the laboratory system. Presented results are obtained from a 5 mm-thick \( \text{CH}_2 \) target. The carbon contribution is removed from the experimental data obtained with Medley [34]. In Fig. 3.3, the measured neutron spectrum (open circles) is plotted in comparison with calculations following the Prokofiev systematics [35]. Calculations have been
Figure 3.2: Two dimensional contour plot of neutron TOF plotted against proton kinetic energy (linear scale). Data were measured at 20° in the laboratory system, from a CH₂ reaction target, at 175 MeV QMN. The solid line represents the TOF gate selecting accepted events, to reduce the low energy tail contribution.

folded with the experimental energy resolution; integrals of experimental data and calculations are normalized to unity. The fraction of neutrons in the measured 175 MeV peak is 39% of the total, in agreement with prediction from Prokofiev systematics; data are consistent with measurements by Pomp et al. [36] at TSL with lower peak-energies.

The accepted neutron spectrum in Fig. 3.3 (filled circles) is obtained applying the 2σ TOF cut described above. The TOF gate reduces the low energy contribution; the fraction of neutrons in the low energy tail, including a 2% contribution from frame overlap, accounts for 50% of the spectrum.

3.4 Response function of CsI(Tl)
3.4.1 Reaction losses

Measured data need to be corrected for energy losses in the CsI(Tl) scintillator due to nuclear reactions. Measurements at TSL have been conducted by Hayashi et al. [34] to investigate the response function of the scintillators installed in Medley to 160 MeV protons. Results show that ~80% of the protons deposited all their kinetic energy in the scintillator, whereas the others are
3.3 Experimental 175 MeV quasi-monoenergetic neutron spectrum at TSL (filled circles) in comparison with calculations with systematics by Prokofiev et al. [35] (solid line); total area normalized to unity. Accepted neutron spectrum after TOF gating (open circles) is plotted in the same picture.

registered in a low-energy reaction tail. Experimental results are in agreement with calculations with the PHITS code [29]. Reaction losses at other proton incident energies have been calculated by PHITS, whereas reaction losses for composite light-ions were computed by Hirayama et al. [25] from non-elastic cross-sections obtained with the ECIS code [37, 38].

3.4.2 Efficiency correction
To correct for the nuclear reaction losses, I have modified the method described by Bertrand et al. [39] to account for particle identification selection cuts applied in the energy space. Due to particle selection and the bending at low energies in the ΔE2-E space, only a fraction of events from the reaction-tail will contribute to low energy events. I correct the data with the expression:

$$R(E_i) = \frac{M(E_i) - \sum_{j=i+1}^{w_i} \left( R(E_j) \frac{1 - \epsilon_j}{j-1} \right)}{\epsilon_i}$$

(3.3)

where \(R(E_i)\) are the corrected counts and \(M(E_i)\) the measured number of counts in the scintillator. The efficiency correction factor \(\epsilon_i\) corresponds to the computed fraction of events that fully deposit their kinetic energy in
Figure 3.4: Histogram in energy of the signal counts (open area), the background counts (dark-shadow area) normalized to the same neutron fluence, and the net events (light-shadow area). Experimental data correspond to proton production at 20° in the laboratory system from a bismuth target, at 175 MeV QMN.

the scintillator. The expression in Eq. 3.3 differs from the one proposed by Bertrand et al. [39] since now the sum is limited to an energy bin $w_i$. This limit should be adapted to the curve of the particle band in the $\Delta E_2$-E space, to include only reaction-tail events that contribute to the measured counts.

From Medley data, I have estimated that the method by Bertrand et al. [39] underestimates the counts in low energy bins up to 15%, and gives an overall 4% underestimation of the total number of counts. The proposed modification has a maximum systematic uncertainty of 1 to 2%, depending on the width of the particle identification cut.

3.5 Background subtraction

Instrumental background is measured in a target-out configuration. Background counts for each characteristic particle-band and for each energy-bin are subtracted from target-in counts, after normalization to the same neutron fluence. An example of background subtraction for proton data measured with Medley is presented in Fig 3.4.

The overall signal-to-background ratio measured with Medley at 175 MeV QMN is 4:1 for protons, 5:1 for deuterons, 8:1 for tritons and 11:1 for $\alpha$ particles.
3.6 Thick target correction

To achieve a reasonable count rate, relatively thick reaction targets are installed in the Medley scattering chamber. The typical thickness for a reaction target is about 0.3 to 5 mm. Whereas energy loss by incident neutrons is negligible, light-ions produced in the Medley reaction target lose a fraction of their kinetic energy in the target itself. This energy loss causes a distortion in the measured spectrum for all light-ions. Moreover, light-ions with low kinetic energy can be fully stopped in the target. Hence, not only the shape of the measured spectrum is distorted, but the total number of light-ions reaching the telescope detectors is lower than the number of particles produced at each given emission angle. This thick target effect is more important for heavier particles and lower energies: in the case of $\alpha$ particles, the combined effect of particle absorption and energy loss (such that the kinetic energy of the particle falls below the detection threshold), can account for up to one order of magnitude more than the number of measured events.

To correct for this effect, Pomp and Tippawan [40] developed the TCORR code. This method applies an iterative procedure to find the inverse response functions of the system for a given light-ion and a defined target thickness, starting from a first guess function, and calculating separately energy and particle loss in the target. Cross-sections measured with the Medley setup are processed through the TCORR code to obtain final data.

Paper VI discusses the validation of the TCORR correction method for measurements with the present configuration of the Medley setup. Monte Carlo particle transport simulations are performed with the PHITS code [29]: the corrected light-ion energy spectrum is provided as input and particles are transported through a volume having the same thickness and composition of the reaction target. Results show that the spectrum obtained from PHITS calculations is in perfect agreement with the measured one.

3.7 Cross-section normalization

Absolute cross-section normalization of the net counts for each light-ion is obtained from relative measurements of a reference cross-section.

$H(n,p)$ elastic scattering events are measured at 20 degrees in the laboratory system using a CH$_2$ reference target. The TOF gate is applied to the data to reduce the contribution of the low-energy tail. Proton events produced from a C target are measured and subtracted from the CH$_2$ target after appropriate normalization, as shown in Fig. 3.5 (a). The proton peak at 153 MeV corresponds to $H(n,p)$ elastic events induced by 175 MeV neutrons, at 20 degrees in the laboratory system. The integral I$_H$ of a Gaussian fit of this elastic peak provides the measured number of protons induced by 175 MeV neutrons.
Figure 3.5: (a) Net proton spectrum at 20° in the laboratory system, produced in the interaction of 175 MeV QMN with a CH₂ target (open histogram) and with a C target (shadow histogram), after normalization. (b) Proton spectrum from H (open histogram) is obtained subtracting the carbon contribution from the CH₂ spectrum; the solid line represents a Gaussian fit of the (n,p) elastic peak at 153 MeV.

In Fig. 3.5 (b) are plotted the net H(n,p) counts from the accepted neutron spectrum presented in Fig. 3.3; the Gaussian fit selects peak energy events.

Reference cross-section data are available on-line from the SAID Partial-Wave Analysis Facility, based at the George Washington University, Virginia (USA) [41]; data for the emission of a neutron in the center of mass system are converted to cross-section values for the emission of a proton at 20 degrees in the laboratory system:

$$\sigma_H = \frac{d\sigma \left( H(n,p) \right)}{d\Omega \vert_{\text{lab,}\theta=20\,\text{deg}}} = 19.9 \, \text{mb sr}^{-1}$$

(3.4)

Absolute double-differential cross-sections $\sigma_x$, for a reaction target $x$ and a given light-ion, are obtained from net counts $N_x$, applying the following expression:

$$\frac{\sigma_x}{N_x} = \frac{\sigma_H \, 2A_x \, m_{CH_2} \, \Phi_{CH_2} \, \Omega_{CH_2} \, 1}{I_H \, A_{CH_2} \, m_x \, \Phi_x \, \Omega_x \, \Delta E}$$

(3.5)

where $A_x$ and $A_{CH_2}$ are respectively the atomic weight of the reaction target and of the CH₂ target, $m_x$ and $m_{CH_2}$ are the target masses, $\Phi_x$ and $\Phi_{CH_2}$ are the neutron fluences measured with one of the neutron monitors, $\Omega_{CH_2}/\Omega_x$ is the ratio between the solid angle seen by the telescope at 20 degrees and the telescope used for the measurement, and $\Delta E$ is the energy bin width.
Before applying the normalization formula, \( I_H \) and \( N_x \) are corrected for the respective dead time of the acquisition system and for the efficiency of the CsI(Tl) scintillator.

### 3.8 Reference target and cross-section normalization

*It can only be attributable to human error.*

(HAL 9000, 2001: A Space Odyssey)

Preliminary results for light-ion production from Fe and Bi as reported in [18, 42, 43, 44, 45, 46] were obtained under the erroneous assumption that the reference CH\(_2\) target used for normalization was 1 mm-thick, with a diameter of 25 mm and mass of 461.55 mg. However, further investigation proved that the reference CH\(_2\) target used in the January/February 2009 experimental runs, when Fe and Bi data were measured, was 1 mm-thick, but with a diameter of 37 mm and a mass of 1017 mg.

This erroneous assumption lead to an underestimation of the preliminary cross-section data by a factor \( \sim 2.2 \); final corrected data are presented in this Thesis, and in the most recent Paper II.

A method to verify \emph{a posteriori} the mass of the CH\(_2\) installed in the Medley chamber is described here, based on the notes by Pomp [47].

Povh et al. [48] describes the geometric reaction cross-section in scattering experiments as

\[
\sigma = \frac{\text{reaction rate}}{\text{incident flux} \times \text{number of scattering centers}} \tag{3.6}
\]

In Section 3.7 we have seen how it is possible to measure, with Medley, \( N_H \) the number of protons produced in the H(n,p) elastic scattering at a given angle (or the equivalent integral \( I_H \)).

The neutron flux \( \Phi_n \) is given by the ICM or TFBC neutron monitors; \( \Phi_n \) can be also calculated from the integrated proton beam current in the beam dump. Neutron monitors and beam bump readings are saved with the data. Correction factors to actual neutron flux are provided by TSL. The uncertainty associated to the neutron flux is 10% for ICM and 30% for TFBC; when the neutron flux is obtained from the proton current, the uncertainty is 10% (Anders Hjalmarsson, private communication). Since the neutron monitors have virtually no dead-time, whereas the Medley acquisition system has a life-time of \( \sim 80\% \), a correction factor should be introduced: \( t_{life} \) is the active data acquisition time in seconds.

Knowing the elastic \( \sigma(E,\theta) \) in the laboratory system, the solid angle covered by the detector \( \Delta\Omega \) and the incident neutron flux, it is possible to obtain
Table 3.1: Expected $H(n,p)$ counts from 175 MeV peak neutrons for three CH$_2$ targets with different masses.

<table>
<thead>
<tr>
<th>material</th>
<th>thickness (mm)</th>
<th>mass (mg)</th>
<th>$N_{H}^{\text{expected}} \times 10^3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH$_2$</td>
<td>1.0</td>
<td>461.55 ($\pm$0.01)</td>
<td>4.1 ($\pm$0.6)</td>
</tr>
<tr>
<td>CH$_2$</td>
<td>1.0</td>
<td>1017 ($\pm$1)</td>
<td>9.1 ($\pm$1.4)</td>
</tr>
<tr>
<td>CH$_2$</td>
<td>5.0</td>
<td>2293 ($\pm$1)</td>
<td>20.7 ($\pm$3.1)</td>
</tr>
</tbody>
</table>

the number of scattering centers (SC) of the measured target with the expression:

$$SC = \frac{N_{H}}{t_{life} \times \sigma(E, \theta) \times \Delta \Omega \times \Delta E \times \Phi_{n} \times 10^{27}} \quad (3.7)$$

The conversion from mb$^{-1}$ to cm$^{-2}$ is given by the factor $10^{27}$.

However, we have only three possible candidates as CH$_2$ targets; from their masses is possible to obtain the number of protons, i.e. the number of scattering centers SC. Calculating the expected number of proton counts $N_{H}^{\text{expected}}$ for each target, and comparing it with the measured number of counts $N_{H}$ allows to determine which target was in use. Expected counts for the available CH$_2$ targets are reported in Table 3.1.

The number of H(n,p) counts from 175 MeV peak neutrons measured with the Medley setup in the January/February 2009 runs, is $7.5 \times 10^3$. This result confirms that the CH$_2$ target installed in Medley at the time of the experimental runs was the one with mass of 1017 mg; thus this target is used as reference for the normalization to absolute cross-section values.

3.9 Summary of systematic uncertainties

The main sources of systematic uncertainties are the number of H(n,p) counts from the CH$_2$ target (5%), the relative uncertainty from the neutron beam monitors (2 to 3%), the uncertainty on the reference cross-section (2%) and the correction for the thick-target effect (1 to 20%). In low-energy bins, the uncertainty due to the thick-target correction is 10% for protons and up to 20% for $\alpha$ particles, whereas it accounts for a few percents for high-energy bins. Tippawan et al. [22] analyzes systematic uncertainties in the analysis of Medley data at 96 MeV; in the present analysis, the systematic uncertainty associated to the absolute cross-section normalization (5 to 10%), is larger than at 96 MeV due to the contribution of the low-energy tail in the QMN spectrum.
4. Theoretical models

A good model can advance fashion by ten years.
(Yves Saint-Laurent, 1936-2008)

4.1 TALYS

TALYS is a nuclear reaction code developed by Koning et al. [12, 49]. The aim of the code is to predict a wide range of nuclear reactions involving photons, neutrons, protons, deuterons, tritons, $^3$He and $\alpha$ particles for energies from 1 keV to 200 MeV and for target nuclei with mass number $A \geq 4$.

TALYS includes state-of-the-art nuclear models and it is widely used by the nuclear data community for the analysis of experiments and to generate evaluated data for applications. Calculations made with TALYS have been used to build the TENDL-2010 [50] evaluated data library. Since data presented in this Thesis are the first measurements for neutron-induced light-ion production in the 100 to 200 MeV incident energy region, they offer the first opportunity to verify and validate TALYS and the reliability of the evaluated data in the TENDL-2010 library for neutron-induced reactions in this energy range.

TALYS describes the pre-equilibrium (PE) emission of single particles following the two-component exciton model (EM) described by Koning and Duijvestijn [51]. However, the EM does not include direct-like production mechanisms as nucleon transfer (NT) or knock out (KO) of preformed clusters. Such direct-like mechanisms play an important role in the production of composite particles in the energy range of interest. To account for these mechanisms, TALYS complements the EM with the systematics proposed by Kalbach [15].

The Kalbach phenomenological model is based on proton-induced data with incident energies up to 90 MeV, and on neutron-induced data with incident energies up to 63 MeV. TALYS calculates the PE production of composite light-ions extending the energy dependence proposed by Kalbach for incident energies up to 200 MeV.

TALYS calculates the total PE cross-section as sum of the EM, NT and KO contributions:

$$\frac{d\sigma^{PE}}{dE} = \frac{d\sigma^{EM}}{dE} + \frac{d\sigma^{NT}}{dE} + \frac{d\sigma^{KO}}{dE} \quad (4.1)$$

To control how the NT and KO contributions are added to the EM cross-section, TALYS provides three adjustable parameters. The \textit{preeqcomplex pa-}
rameter allows to turn off the NT and KO contribution, thus only the EM is used to calculate the PE emission, for all particles. \textit{cstrip} and \textit{cknock} are two adjustable parameters: their values may vary between 0 and 10 (default is 1) and they act as multiplication factors respectively for the NT and for the KO terms in Eq. 4.1. \textit{cstrip} and \textit{cknock} may be applied independently to calculate PE emission of each particle.

TALYS calculates the total reaction cross-section and then distributes its value according to the branching ratio of different reaction channels and different reaction mechanisms. Hence, varying the PE emission of a particle, via the \textit{cstrip} and/or \textit{cknock} parameters, produces a variation in the cross-section of other mechanisms for emission of the same particle and in the production cross-sections of other particles.

The use of \textit{cstrip} and \textit{cknock} is discussed in Papers III and IV, where an energy dependence for these two parameters is proposed in order to improve the predictive power of TALYS for energies above 100 MeV.

Other adjustable parameters may be set in TALYS to modify how the code computes particle production in the PE stage. I have performed several calculations with TALYS to identify the relevant parameters for nucleon-induced reactions in the 100 to 200 MeV region: the parameters used are listed in Paper III. No significant difference emerged when calculations where performed with default or modified parameters.

\section*{4.2 JQMD}

QMD is a semiclassical simulation method that gives a microscopic description of the time evolution of the nucleon many-body system \cite{52,53,54}. Each nucleon propagates in the nuclear mean field formed by all other nucleons. Interactions among nucleons are described by stochastic two-body collisions. At the end of a given evolution time (of the order of \(10^{-22}\) s), conditions for the emission of single and composite particles are verified. The generalized evaporation model is used to describe particle emission when the compound system reaches equilibrium.

Watanabe and Kadrev \cite{13} compare QMD calculations with neutron-induced light-ion production measured with Medley at 96 MeV \cite{21,22}. Calculations were performed with the JQMD code \cite{54}. The experimental data for proton production from O and Si are well reproduced by the JQMD code, however production of composite particles is largely underestimated by calculations. To account for this underestimation, Watanabe and Kadrev introduce a surface coalescence model (SCM) for the formation of clusters: the main hypothesis is that, before the system reaches the equilibrium, composite particles may be formed by coalescence to a leading nucleon, in the surface region of the nucleon many-body system. Calculations with the modified code, including a SCM, show good agreement with data at 96
MeV at intermediate and low emission energies. At high emission energies, where direct-like mechanisms (NT, KO) play an important role, data are still underestimated by calculations.

The SCM is dependent on three parameters: they have been optimized to describe the analyzed neutron data at 96 MeV. A complete discussion is presented in Ref. [13]. Experimental light-ion production from Fe at 175 MeV QMN presented in Paper II, and in the Part II of this Thesis, is compared with preliminary calculations obtained with this modified version of the JQMD code; the parameters are kept at the same values used for the 96 MeV data.

These preliminary calculations do not include information on the Q-value of the reaction. An integration of the modified QMD model with the PHITS code is currently under development.

4.3 MCNP6

MCNP6 [14] is the latest Los Alamos National Laboratory (LANL) transport code representing a merger of MCNP5 and MCNPX. MCNP6 uses the cascade-exciton model (CEM) as the default event generator for reactions induced by nucleons, pions, and photons at energies below several GeVs. The code has been validated and verified against a variety of experimental data [55], but has not been released yet. The data presented in this Thesis represent an interesting challenge for MCNP6.

CEM assumes that nuclear reactions occur generally in three stages. The first stage is the intranuclear cascade (INC), in which primary particles can be re-scattered and produce secondary particles several times prior to absorption by, or escape from, the nucleus. When the cascade stage of a reaction is complete, CEM uses the coalescence model to create high-energy deuterons, tritons, $^3$He, and $\alpha$ particles via final-state interactions among emitted cascade nucleons, already outside of the target. The subsequent relaxation of the nuclear excitation is treated in terms of an improved version of the modified exciton model of PE decay followed by the equilibrium evaporation/fission stage of the reaction. However, if the residual nuclei after the INC have atomic numbers with A<13, CEM uses the Fermi breakup model to calculate their further disintegration instead of using the PE and evaporation models. The lecture by Mashnik et al. [56] provides an extensive discussion on the CEM event generator.

Experimental Fe and Bi data in Paper II, and in Part II of this Thesis, are compared with MCNP6 calculations where the CEM03.02 event generator is used.

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1MCNP6 will be made available to users outside LANL via RSICC at Oak Ridge, TN, USA in September 2011
Part II:

Experimental results

In Chapter 5, I present double-differential cross-sections for light-ion production from Fe and Bi in 175 MeV QMN-induced reactions. The experimentally obtained cross-sections are plotted in comparison with model calculations described in Chapter 4. Model calculations are folded with the accepted neutron spectrum presented in Fig. 3.3. Complete results at eight angles in the laboratory system, from 20 degrees to 160 degrees, are presented in the Appendix. These results are discussed in Paper II.

I have performed the TALYS calculations, whereas QMD calculations with the modified JQMD code were done by Y. Watanabe, and MCNP6 calculations by S. Mashnik.
5. 175 MeV QMN data

I've watched C-beams glitter in the dark, near the Tannhauser Gate.
(Blade Runner, 1982)

5.1 Light-ion production from Fe

Proton production
QMD calculations are the most successful in describing proton production (Fig. 5.1), except for PE emission at the most forward angle. As mentioned in Section 4.2, information on the Q-value of the reaction is not included in the QMD calculations, thus we observe a high-energy contribution in the QMD results. TALYS underestimates proton production at all emission angles. MCNP6 describes the emission of protons at 20 and 40 degrees better than at larger angles.

All the models overestimate the evaporation peak.

Deuteron production
PE production of deuterons (Fig. 5.2) is overestimated by TALYS, except at the most backward angles (140 and 160 degrees). In Chapter 6, I discuss proton-induced production of deuterons at 175 MeV from Ni [11, 57] in comparison with TALYS: here a similar overestimation of the data is observed. TALYS PE emission of deuterons and other composite light-ions is discussed in Chapter 7.

QMD calculations underestimate the PE and direct production of deuterons at forward angles; this underestimation is larger at 20 degrees, and for high emission energies. At backward angles QMD is in agreement with the data. Direct-like reactions, as pick-up of an incident proton by the incoming neutron, are not included in the QMD description; the contribution of direct-like mechanisms is important at forward angles and high emission energies, corresponding to few nucleon-nucleon interactions. The absence of this reaction mechanism may explain the observed underestimation of deuteron production. Watanabe and Kadrev [13] show a similar underestimation of deuteron data in angle-integrated differential cross-sections measured from Si and O at 96 MeV neutrons; angle-integrated 96 MeV data show also an overestimation by QMD calculations of the evaporation peak: this overestimation is present at all angles in the 175 MeV QMN data.
Calculations with MCNP6 show an underestimation of the data over a wide energy range in the emission spectrum. As in the QMD case, the coalescence model included in CEM does not seem sufficient to predict emission of deuterons at high emission energies. This confirms the importance of direct-like mechanisms for the production of deuterons, at least at forward angles. This result is in agreement with the findings by Mashnik et al. [56], comparing MCNP6 calculations with neutron-induced production of deuterons from Fe at 96 MeV measured by Blideanu et al. [23] with the Medley setup.

As observed for proton production, all models overestimate compound emission of deuterons.

Triton production

PE emission of tritons (Fig. 5.3) is overestimated by TALYS at forward angles, whereas at large angles model predictions are inside the statistical uncertainties of the experimental data. At 20 degrees, QMD and MCNP6 calculations are in agreement, but show an underestimation of triton data. Direct-like mechanisms seem to play a role also in the production of tritons, at high emission energies. QMD calculations reproduce PE emission of tritons at other
emission angles, but overestimate the low-energy emission. MCNP6 underestimates the experimentally observed PE triton production at forward angles.

Triton data at backward angles are affected by poor statistics and large uncertainties.

\( {^3}He \) production

TALYS largely overestimates PE emission of \( {^3}He \) (Fig. 5.4); this is also observed in the case of (p,\( {^3}He \)) reactions presented in Chapter 6.

QMD calculations are in agreement with experimental data at forward angles, but for high-energy emission of \( {^3}He \) at 20 degrees. QMD description of PE reactions does not include direct-like mechanisms, such as nucleon transfer: this may explain the discrepancy observed at high emission energies, at most forward angles. However, at backward angles QMD overestimates the measured data.

MCNP6 overestimates low-energy emission of \( {^3}He \) at most forward angles, whereas underestimates high-energy production. At larger angles, MCNP6 is in agreement with the measured data.
Figure 5.5: Experimental $\sigma(E,\theta)$ for the Fe(n,$\alpha$) reaction at 175 MeV QMN. Calculations: default TALYS calculations (solid black line); QMD with surface coalescence model following Watanabe and Kadrev (short-dashed blue line); MCNP6 calculations with the CEM03.02 event generator (long-dashed red line).

$\alpha$ particle production

Production of $\alpha$ particles at 20 and 80 degrees in the laboratory system is presented in Fig. 5.5. TALYS overestimates the PE production of $\alpha$ particles at all emission angles, except at 160 degrees (see Appendix). We observe the same pattern in proton-induced reactions, presented in Chapter 6.

PE emission of $\alpha$ particles is underestimated by QMD calculations at 20 degrees, as observed for production of other composite light-ions, whereas QMD reproduces the experimental cross-sections at larger angles.

MCNP6 overestimates the low-energy PE emission of $\alpha$ particles at 20 and 40 degrees, as observed for the emission of $^3$He; however, MCNP6 reproduces the data at high emission energies. At larger angles, MCNP6 and QMD show similar results for production of $^3$He and $\alpha$ particles; however, MCNP6 favors forward emission over backward emission, in comparison to QMD calculations. Mashnik et al. [56] compared MCNP6 calculations with proton-induced $\alpha$ particle production from Co at 160 MeV measured by Cowley et al. [10]: the comparison shows the same pattern observed in the neutron data presented here.

The evaporation peak is well reproduced by all the three models considered.

5.2 Light-ion production from Bi

Proton production

TALYS calculations underestimate proton production (Fig. 5.6) at all emission angles; the relative discrepancy with measured data is larger at low-emission energies and increases with the angle.

MCNP6 reproduces the low-energy emission of protons at 20, 40 and 60 degrees; MCNP6 reproduces also the high-energy part of the emission spectrum at 20 degrees. At other emission angles and energies, MCNP6 underestimates
the data. The discrepancy between MCNP6 calculations and experimental data is greater than the discrepancy given by TALYS calculations. Both TALYS and MCNP reproduce the left shoulder of the proton spectrum.

**Deuteron production**

PE emission of deuterons (Fig. 5.7) is underestimated by TALYS at forward angles, whereas calculations well describe low-energy production. At larger angles, TALYS reproduces the high-energy part of the emission spectrum, but underestimates the low-energy PE production of deuterons. This underestimation may be due to multiple PE emission, i.e. to the emission of a low-energy deuteron after a first particle has already been emitted in the dynamic reaction process; TALYS treats this production mechanism only for protons and neutrons, but not for composite light-ions.

At the most forward angles, MCNP6 shows the same pattern in comparison with deuteron data observed from Fe. The low-energy part of the emission spectrum is overestimated by MCNP6, whereas, the high-energy PE emission is largely underestimated. The underestimation of the high-energy emission spectrum is present also at other angles, whereas MCNP6 describes the low-energy emission of deuterons better than TALYS.
Triton production

Production of tritons (Fig. 5.8) is reproduced by TALYS in the high-energy part of the PE emission spectrum, with better agreement at large angles than at forward angles. TALYS calculations underestimate the wide low-energy emission peak. This peak is larger than the expected evaporation peak; as in the deuteron emission case, this peak may be explained with multiple PE emission of composite light-ions, a mechanism not included in TALYS.

MCNP6 well describes the wide low-energy peak at forward angles, but underestimates it at larger emission angles. As observed in the emission of protons and deuterons, MCNP6 underestimates the high-energy PE emission of tritons.

$^3$He production

TALYS underestimates the PE production cross-section of $^3$He (Fig. 5.9) at intermediate energies, for forward angles, whereas it reproduces $^3$He production in the high-energy end of the spectrum and at low-emission energies. In Chapter 6, I compare TALYS calculations with proton-induced data at 120, 160 and 200 MeV for production of $^3$He from Au [9]; these comparisons show a similar pattern to the one observed in the Bi case. Calculations with
MCNP6 underestimate the high-energy emission spectrum at forward angles, whereas reproduce low-energy emission; at other angles, data are reproduced by MCNP6.

At large emission angles, low statistics result in large uncertainties and the prediction of both models are included in the error bars of the measured data.

**α particle production**

Low-energy production of α particle (Fig. 5.10) is largely underestimated by TALYS calculations, whereas high-energy emission is well described by the same code. This pattern differs from the one observed in α particles production from Fe: there the low-energy evaporation peak was well described by TALYS, and the PE emission was overestimated. When considering proton-induced data at 160 and 200 MeV from Au [10], we observe a third pattern: the underestimation of the evaporation peak and the overestimation of the PE emission.

MCNP6 calculations underestimate the production of α particles over all the emission spectrum; the agreement improves at 140 and 160 degrees where low statistics produce large statistical uncertainties. However, Mashnik et al. [56] compared MCNP6 calculations with 160 MeV proton-induced production of α particles from Au measured by Cowley et al. [10]: in this comparison, data are in good agreement with MCNP6 calculations.

### 5.3 Summary

*Light-ion production from Fe at 175 MeV QMN*

Comparison with the models showed that no code is able to reproduce all the measured data. QMD calculations proved to be most successful in describing proton production from Fe; QMD reproduces fairly well also production of tri-
Table 5.1: Coalescence (C) versus direct-like (D) description of PE emission of composite light-ions from Fe at 175 MeV-QMN. "C< D" indicates that production data are overestimated by a direct-like description, but underestimated by coalescence predictions. "C+D" indicates that low-energy emission is described by coalescence, but high-energy emission requires a direct-like description. "C=D" indicates data that are equally described by both mechanisms. Parenthesis indicates a smaller contribution.

<table>
<thead>
<tr>
<th>Emission angle (deg)</th>
<th>20</th>
<th>40</th>
<th>60</th>
<th>80</th>
<th>100</th>
<th>120</th>
</tr>
</thead>
<tbody>
<tr>
<td>Emitted particle</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>deuteron</td>
<td>&lt; D</td>
<td>C &lt; D</td>
<td>C &lt; D</td>
<td>C &lt; D</td>
<td>C &lt; D</td>
<td>C = D</td>
</tr>
<tr>
<td>triton</td>
<td>D</td>
<td>C (&lt; D)</td>
<td>C (&lt; D)</td>
<td>C = D</td>
<td>C = D</td>
<td>C = D</td>
</tr>
<tr>
<td>$^3$He</td>
<td>C+D</td>
<td>C(+D)</td>
<td>C</td>
<td>C</td>
<td>C</td>
<td>C</td>
</tr>
<tr>
<td>$\alpha$ particle</td>
<td>C</td>
<td>C</td>
<td>C</td>
<td>C</td>
<td>C</td>
<td>C</td>
</tr>
</tbody>
</table>

tons, $^3$He and $\alpha$ particles at all emission angles, except at 20 degrees. The importance of the surface coalescence model by Watanabe and Kadrev [13] is described in Paper IV, where default QMD calculations are compared with the modified version of the code, to account for the formation of clusters, by coalescence to a leading nucleon, in the surface region of the nucleon many body system. This comparison shows a large discrepancy between default QMD calculations and experimental data, for production of composite light-ions, whereas proton production is well described.

When the coalescence mechanism is included, data are fairly well reproduced at large emission angles. However, high energy emission of composite light-ions is still underestimated at forward angles by QMD calculations including the SCM. Watanabe and Kadrev [13] observe an underestimation of high energy emission of composite light-ions, in angle-integrated neutron-induced cross-sections at 96 MeV: there they explain this underestimation in terms of direct-like production mechanisms, as direct pick-up of one or more nucleons by the incident neutron, not included in the description given by the QMD model. Comparison with double-differential data from Fe shows that the underestimation of composite light-ion production is dependent on emission angle and on emission energy, thus confirming the picture drawn by Watanabe and Kadrev [13]. Pick-up of a proton by a neutron, to form a deuteron, is favored compared to pick-up of two or three nucleons, hence this mechanism is less important for production of tritons, $^3$He and $\alpha$ particles, as confirmed by the data.

Light-ion production in MCNP6 is described with the CEM code. Production of $^3$He and $\alpha$ particles from Fe is in agreement with QMD results including a SCM. The formation of composite particles by coalescence to high-energy nucleons already outside the nucleus seems to be the dominant production mechanism for PE emission of helium-isotopes from Fe.
Table 5.2: Coalescence (C) versus direct-like (D) description of PE emission of composite light-ions from Bi at 175 MeV-QMN. Description as in Table 5.1.

<table>
<thead>
<tr>
<th>Emission angle (deg)</th>
<th>20</th>
<th>40</th>
<th>60</th>
<th>80</th>
<th>100</th>
<th>120</th>
</tr>
</thead>
<tbody>
<tr>
<td>Emitted particle</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>deuteron</td>
<td>D</td>
<td>D</td>
<td>D</td>
<td>D</td>
<td>D</td>
<td>D</td>
</tr>
<tr>
<td>triton</td>
<td>D</td>
<td>D</td>
<td>D</td>
<td>D</td>
<td>D</td>
<td>D</td>
</tr>
<tr>
<td>$^3$He</td>
<td>C</td>
<td>C+D</td>
<td>C</td>
<td>C</td>
<td>C</td>
<td>C</td>
</tr>
<tr>
<td>$\alpha$ particle</td>
<td>D</td>
<td>D</td>
<td>D</td>
<td>D</td>
<td>D</td>
<td>C = D</td>
</tr>
</tbody>
</table>

TALYS calculations do not include a coalescence model, but describe PE production of composite particles with direct-like mechanisms, following the Kalbach systematics [15]. TALYS largely overestimates PE production of helium-isotopes from Fe. However, TALYS reproduces high-energy emission of deuterons and tritons at 20 degrees in the laboratory system from Fe. These observations suggest a picture where the best description of the PE emission of composite light-ions from Fe could be given combining direct-like mechanisms and coalescence formation. This result is summarized in Table 5.1, where the production mechanism that best describes the experimental data is indicated for each emission angle, and for each particle. Direct-like mechanisms (D) are needed to describe emission at forward angles, and are more important for deuterons and tritons, whereas coalescence production mechanisms (C) are relevant at larger angles and for $^3$He and $\alpha$ particles.

Light-ion production from Bi at 175 MeV QMN

QMD calculations are not available for production of light-ions from Bi at 175 MeV. TALYS predicts larger PE cross-sections than MCNP6, for all light-ions. However, MCNP6 describes a large cross-section for low-energy emission of deuterons and tritons, in better agreement with data than TALYS at several emission angles. The coalescence production mechanism largely fails to reproduce deuteron production and underestimates also emission of tritons and high-energy emission of $^3$He. In Table 5.2, direct-like and coalescence description of the emission of composite light-ions from Bi are compared with experimental results. Production of deuterons, tritons and $^3$He seems to confirm the pattern observed in the reactions from Fe, presented in Table 5.1. However, $\alpha$ particle production is underestimated by MCNP6 over all the emission spectrum; this may be an indication of some other mechanism and does not reproduce the pattern of a “direct-like versus coalescence” description.
The wide low-energy emission peak observed for most emission angles, except for $^3$He production, is generally not described by TALYS, whereas MCNP6 predicts it for deuteron and triton emission. This peak may be explained with multiple PE emission, i.e. with emission of a low-energy composite light-ion following the emission of first a particle (neutron, proton, deuteron, triton, $^3$He or $\alpha$ particle); the first emitted particle carried most of the energy, however the residual energy is sufficient to allow emission of a composite particle before the system reaches equilibrium.
Part III:
Discussion

In Chapter 6, I compare TALYS calculations with proton-induced light-ion production data in the 120 to 200 MeV energy region. These comparisons provide information on the way TALYS describes the PE emission of composite light-ions. I found an energy dependency in the Kalbach contribution to the EM, as included in TALYS. This discussion is included in Paper III.

In Chapter 7, I use a new parametrization for TALYS, to improve its predictive power for PE emission of composite particles, in the 100 to 200 MeV energy region. Here, 175 MeV QMN data from Fe and Bi are compared with TALYS calculations obtained with the proposed parametrization. Paper III and IV report on these findings.

In Chapter 8, I summarize the findings of this work and conclude.
6. Pre-equilibrium emission of composite light-ions in TALYS

*Every phase of evolution commences by being in a state of unstable force and proceeds through organization to equilibrium.*

(Kabbalah)

Neutron-induced data presented in Chapter 5 are the first available in the 100 to 200 MeV region. Calculations with TALYS show a general overestimation of the experimental results for Fe. When considering Bi data, TALYS shows overestimation of deuterons and $^3\text{He}$ at forward angles, whereas tritons and $\alpha$ particles are underestimated at low emission-energies.

To investigate these discrepancies, I have used the TALYS code to compile cross-sections for proton-induced data available in literature in the 120 to 200 MeV energy region, and for target-nuclei with masses close to Fe and Bi. In the present work I have considered proton-induced light-ion production from Ni at 175 MeV by Piskor-Ignatowicz [11, 57], and proton-induced $^3\text{He}$ and $\alpha$ particle production from Co and Au at 120, 160 and 200 MeV by Cowley et al. [9, 10].

As discussed in Chapter 4, TALYS computes PE emission of composite light-ions complementing the two-component EM with the contribution of direct-like production mechanisms (NT, KO), following the systematics by Kalbach [15]. In single-particle induced reactions, the NT mechanism is described as pick-up of one or more nucleons by an incident particle; the KO mechanism is relevant only for production of high-energy $\alpha$ particles, at forward emission angles.

TALYS calculations presented in this Chapter were performed scaling down the NT contribution, via the `cstrip` parameter. When the best fitting `cstrip` parameter was determined for each target-nucleus and for each incident energy, I have performed calculations scaling down the KO contribution via the `cknock` parameter, keeping `cstrip` fixed.

6.1 Proton-induced data from Ni at 175 MeV

Proton production is fairly well described by default TALYS calculations (Fig. 6.1). The reduction of direct-like production mechanisms for composite particles increases the overall cross-section for emission of protons. This is due to the fact that the TALYS code conserves the total
reaction cross-section. This increase slightly improves the agreement between data and calculations. However, a large difference emerges in the description of composite light-ions, when reducing the NT contribution to PE emission.

Deuteron, triton and $^3$He emission from Co are plotted respectively in Fig. 6.2, Fig. 6.3 and Fig. 6.4. In all three cases, default TALYS calculations overestimate PE emission, whereas they reproduce evaporation and PE low-energy emission.

Table 6.1 summarizes the values for the $cstrip$ parameter providing the best fit of PE emission at all emission angles measured by Piskor-Ignatowicz [11, 57]. Applying these values to TALYS calculations, intermediate- and high-energy PE production is in agreement with experimental data, whereas an underestimation appears at low emission-energies. This underestimation may be explained as multiple PE emission of composite light-ions, as observed in the discussion of neutron-induced results.

The KO mechanism contributes to the production of high-energy $\alpha$ particles at the most forward emission angles. Experimental PE $\alpha$ particle production is overestimated by default TALYS calculations at intermediate and high emission energies, whereas it is underestimated at low emission-energies. Reducing NT and KO contribution improves the agreement at high energies, but increases the discrepancy at low-energy, as observed for production of other composite light-ions. Results for $\alpha$ particle are presented in Fig. 6.5.

**Table 6.1: Proton-induced PE emission of composite light-ions from Ni at 175 MeV: $cstrip$ parameter providing the best agreement with PE experimental data**

<table>
<thead>
<tr>
<th>Composite light-ion</th>
<th>$cstrip$ parameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>deuteron</td>
<td>0.25 to 0.5</td>
</tr>
<tr>
<td>triton</td>
<td>0.10 to 0.25</td>
</tr>
<tr>
<td>$^3$He</td>
<td>0.25</td>
</tr>
</tbody>
</table>

**Figure 6.1:** Experimental $\sigma(E,\theta)$ for the Ni($p,p\alpha$) reaction at 175 MeV incident protons by Piskor-Ignatowicz [11, 57]. Default TALYS calculations (black solid line) are compared with results obtained scaling the NT contribution via the $cstrip$ parameter.
Figure 6.2: Experimental $\sigma(E,\theta)$ for the Ni(p,3dx) reaction at 175 MeV incident protons by Piskor-Ignatowicz [11, 57]. Default TALYS calculations (black solid line) are compared with results obtained scaling the NT contribution via the cstrip parameter.

Figure 6.3: Same as Fig. 6.2, but for the Ni(p,tx) reaction.

Figure 6.4: Same as Fig. 6.2, but for the Ni(p,^3He) reaction.
Figure 6.5: Experimental $\sigma(E,\theta)$ for the Ni(p,α) reaction at 175 MeV incident protons by Piskor-Ignatowicz [11, 57]. TALYS calculations are obtained scaling the KO contribution via the $cknock$ parameter, keeping $cstrip = 0.25$. Default TALYS calculations (black solid line) are reported for comparison: $cknock = 1.0$ and $cstrip = 1.0$. Calculations with $cknock = 0.0$ and $cstrip = 0.0$ (red dashed line) correspond to $preeqcomplex = n$. 

6.2 Proton-induced data at 120, 160 and 200 MeV

Comparison between TALYS calculations and experimental $^3$He production from Co at 120, 160 and 200 MeV shows a dependence in energy, when considering the overestimation of PE emission by TALYS (Fig. 6.6). At 120 MeV incident-protons, TALYS is in agreement with backward PE emission, whereas overestimates forward $^3$He emission. At higher incident proton energies, TALYS overestimates experimental data at all emission angles. This overestimation is larger at 200 MeV than at 160 MeV incident protons.

A similar pattern is observed when comparing TALYS calculations with $^3$He production from Au (Fig. 6.6). PE emission is overestimated by TALYS and the discrepancy with experimental data increases with energy. At 120 and 160 MeV incident protons, backward emission of $^3$He is described better than forward emission.

Table 6.2 summarizes the values of $c_{strip}$ providing the best agreement with experimental PE emission at all emission angles, from Co and from Au. These results are consistent with the value of $c_{strip} \approx 0.25$ identified when discussing $^3$He production in 175 MeV proton-induced data from Ni.

However, as observed for other reactions, low-energy PE emission is underestimated by default TALYS calculations and this discrepancy increases when reducing the contribution of the NT mechanism.

Production of $\alpha$ particles from Co, presented in Fig. 6.7, shows the same pattern observed in the reaction from Ni at 175 MeV (Fig. 6.5); whereas production of $\alpha$ particle via the KO mechanism is negligible at both 160 and 200 MeV incident proton-energy, emission is dominated by low-energy PE production.

In Fig. 6.9, it is possible to observe that reducing the NT contribution to the production of $\alpha$ particles from Au, TALYS predicts a larger evaporation peak than in default calculations. At forward angles compound emission is in agreement with $c_{strip} \leq 0.1$, whereas at backward angles (not shown) calculations with $c_{strip} \leq 0.25$ gives the best agreement with measured data.

Table 6.2: Proton-induced PE emission of $^3$He from Co and Au, at 120, 160 and 200 MeV: $c_{strip}$ parameter providing the best agreement with PE experimental data.

<table>
<thead>
<tr>
<th>Incident proton energy (MeV)</th>
<th>Co(p,$^3$Hex) $c_{strip}$ parameter</th>
<th>Au(p,$^3$Hex) $c_{strip}$ parameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>120</td>
<td>0.5 to 0.75</td>
<td>0.75 to 1.0</td>
</tr>
<tr>
<td>160</td>
<td>0.25</td>
<td>0.25 to 0.5</td>
</tr>
<tr>
<td>200</td>
<td>0.1 to 0.25</td>
<td>0.1 to 0.25</td>
</tr>
</tbody>
</table>
Figure 6.6: Experimental $\sigma(E,\theta)$ for the Co(p,$^3$He) reaction at 120, 160 and 200 MeV incident protons by Cowley et al. [9]. Default TALYS calculations (black solid line) are compared with results obtained scaling the NT contribution via the cstrip parameter.

Figure 6.7: Experimental $\sigma(E,\theta)$ for the Co(p,$\alpha$) reaction at 160 and 200 MeV incident protons by Cowley et al. [10]. TALYS calculations are obtained scaling the KO contribution via the cknock parameter; at 160 MeV cstrip = 0.25, at 200 MeV cstrip = 0.15. Default TALYS calculations (black solid line) are reported for comparison: cknock = 1.0 and cstrip = 1.0.
Figure 6.8: Experimental $\sigma(E,\theta)$ for the $^{208}\text{Au}(p, ^3\text{He})$ reaction at 120, 160 and 200 MeV incident protons by Cowley et al. [9]. Default TALYS calculations (black solid line) are compared with results obtained scaling the NT contribution via the $cstrip$ parameter.

Figure 6.9: Experimental $\sigma(E,\theta)$ for the $^{208}\text{Au}(p,\alpha\alpha)$ reaction at 160 and 200 MeV incident protons by Cowley et al. [10]. TALYS calculations are obtained scaling the KO contribution via the $cknock$ parameter; at 160 MeV $cstrip = 0.25$, at 200 MeV $cstrip = 0.15$. Default TALYS calculations (black solid line) are reported for comparison: $cknock = 1.0$ and $cstrip = 1.0$. 

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6.3 Summary

Proton production from Ni at 175 MeV is well reproduced by TALYS. PE emission of composite light-ions is generally overestimated from Ni at 175 MeV, from Co at 160 and 200 MeV, and from Au at 160 and 200 MeV; however, low-energy emission shows a broader peak not described by TALYS. At 120 MeV, TALYS calculations show a better agreement with experimental results: \(^3\)He production from Co and Au is overestimated at forward emission-angles, but is well described at backward angles; \(\alpha\) particle PE production is overestimated at high-emission energies and underestimated at low-emission energies.

Calculations with reduced NT (and eventually KO) contribution to PE emission of composite light-ion show better agreement with experimental data at 160, 175 and 200 MeV than default TALYS calculations. Results are summarized in Tables 6.1 and 6.2.

The underestimation of the low-energy PE emission of composite light-ions, observed also in default calculations, is enhanced when the contribution of the NT mechanism is reduced. This underestimation is an indication of multiple PE emission of composite particles: this mechanism is included in the present version of TALYS only for protons and neutrons.

The TALYS code conserves the total production cross-section, thus decreasing the PE production of composite particles favors other production mechanisms and/or emission of other particles. In Fig. 6.9 it is possible to observe that scaling down the NT contribution for \(\alpha\) particle production from Au, increases the differential cross-section for evaporation emission. In Fig. 6.1, proton production is enhanced when the cstrip parameter is applied to composite light-ions to reduce the NT contribution to PE emission.
7. Energy dependence of direct-like mechanisms in TALYS

*And now for something completely different.*

*(Monty Python’s Flying Circus)*

In Chapter 5, I have shown that TALYS calculations overestimate 175 MeV QMN-induced PE emission of composite light-ions from Fe, as well as production of deuterons and $^3$He from Bi.

In Chapter 6, proton-induced data at 120, 160, 175 and 200 MeV were compared with TALYS calculations; TALYS overestimates PE production of intermediate- and high-energy composite light-ions. By reducing the contribution of NT and KO mechanisms as described by Kalbach systematics in TALYS, it is possible to obtain a good agreement between calculations and PE emission of composite particles in proton-induced data. The scaling factor for the NT mechanism ($cstrip$ parameter) shows an energy dependence (Table 6.2): $cstrip$ decreases when incident energy increases. TALYS shows a better agreement with data at backward emission angles than at forward emission angles. For production of $\alpha$ particles, a contribution via the KO mechanism seems negligible.

However, when considering QMN-induced data, it is not possible to simply apply a common $cstrip$ (and eventually $cknock$) scaling factor to TALYS. Calculations need to be folded with the incident quasi-monoenergetic spectrum, and the study of proton-induced data showed that the $cstrip$ parameter giving the best agreement with the data is dependent on the incident energy.

Therefore, an energy dependence for $cstrip$ has been used in the following analysis, based on the results from proton-induced data:

$$\begin{align*}
cstrip = \begin{cases} 
1.0 & \text{if } E \leq 90 \text{ MeV} \\
1.9 - \frac{E}{100 \text{ MeV}} & \text{if } 90 \text{ MeV} \leq E \leq 180 \text{ MeV} \\
0.1 & \text{if } 180 \text{ MeV} \leq E
\end{cases} \quad (7.1)
\end{align*}$$

TALYS does not allow to introduce an angular dependence in the NT (and KO) scaling factors, thus Eq. 7.1 has been derived from average values for $cstrip$ in Tables 6.1 and 6.2. Even though the proton-induced data from Co and Au, as well as neutron-induced data from Fe and Bi seem to suggest a target mass dependence in the overestimation of PE emission by TALYS, the same dependence has been used for all calculations.
Comparison of the TALYS calculations, obtained by reducing the $cstrip$ parameter, with experimental 175 MeV QMN-induced data is presented in Figs. 7.1 to 7.7.

As observed in the Ni(p,px) reaction at 175 MeV (Fig. 6.1), reducing the NT contribution for production of composite particles, increases the overall cross-section for emission of protons in the Fe(n,px) reaction (Fig. 7.1); this effect improves the agreement between experimental data and calculations. We observe this effect also in the Bi(n,px) reaction (Fig. 7.5).

Modified TALYS calculations show a good agreement for production of deuterons (Fig. 7.2) and tritons (Fig. 7.3) from Fe; TALYS still overestimates production of $^3$He, however reducing the NT contribution largely reduces the discrepancy with experimental data (Fig. 7.4).

PE $^3$He production from Bi is better reproduced by the modified calculations than by default TALYS, except for high-energy emission at the most forward angles.

The proposed reduction of the NT contribution does not address the discrepancy observed between default TALYS calculations and data for production of deuterons and tritons from Bi in most cases; the large underestimation in the low-energy PE emission observed $e.g.$ in Fig. 7.6 may be explained in terms of multiple PE emission.
Figure 7.2: Experimental $\sigma(E, \theta)$ for the Fe(n,dx) reaction at 175 MeV QMN. Default TALYS calculations (solid line) are compared with results obtained scaling the NT contribution via the cstrip parameter, following Eq. 7.1 (dashed line).

Figure 7.3: Same as Fig. 7.2, but for the Fe(n,tx) reaction.

Figure 7.4: Same as Fig. 7.2, but for the Fe(n,$^3$Hx) reaction.
Figure 7.5: Experimental $\sigma(E,\theta)$ for the Bi(n,px) reaction at 175 MeV QMN. Default TALYS calculations (solid line) are compared with results obtained scaling the NT contribution via the $cstrip$ parameter, following Eq. 7.1 (dashed line).

Figure 7.6: Same as Fig. 7.5, but for the Bi(n,dx) reaction.

Figure 7.7: Same as Fig. 7.5, but for the Bi(n,$^3$He) reaction.
Development of ADS for incineration of spent nuclear fuel requires nuclear data beyond the traditional limits set by data needs for both thermal and fast nuclear reactors. The NEXT project at Uppsala University participates in a European effort to produce nuclear data for ADS.

The Medley spectrometer has been upgraded to measure light ions in neutron-induced reactions with energies up to 175 MeV. Several experimental campaigns have been realized since 2007; activities included the characterization of the 175 MeV QMN beam and the ANITA white neutron spectrum at TSL, and the measurement of inclusive double-differential cross-section for production of protons, deuterons, tritons, $^3$He and $\alpha$ particles from C, O, Si, Fe, Bi and U at 175 MeV QMN.

In this Thesis, I have presented cross-sections for light-ion production from Fe and Bi. These are the first neutron-induced data available for energies above 100 MeV. Results for other target materials are currently under analysis; preliminary results for proton, deuteron and triton production from C [25], proton and deuteron production from Si [26] and proton production from O [Paper V] are presented in the respective references.

Experimental results have been compared with model calculations, realized with the TALYS-1.2 reaction code, the Monte Carlo particle transport code MCNP6, and the JQMD code modified to complement the QMD model with a surface coalescence production mechanism. Comparison with experimental Fe and Bi data shows that no code is able to reproduce all the measured data. The results presented in this work, suggest that surface coalescence models and direct-reaction mechanisms need to be integrated to provide the best description of the data.

Comparison of TALYS calculations with proton-induced data in the 120 to 200 MeV energy range, suggested a new energy dependence for the application of the Kalbach systematics to TALYS calculations. This result improved the predictive power of TALYS for neutron-induced light-ion production from Fe at 175 MeV QMN; this new energy dependence improved also agreement of calculations with $^3$He production from Bi at the same incident energy. This work suggests evidence for the importance of multiple PE emission, to describe low-energy production of all composite light-ions.
The importance of the surface coalescence model in QMD calculations has been confirmed by comparison with the Fe data. Light-ion production from Fe was described by MCNP6 better than production from Bi. Results showed the need to include direct-like mechanisms in the QMD and in the MCNP6 description, to account for high-energy emission of deuterons, tritons and $^3$He.

These experimental data will provide benchmark points for other theoretical models and nuclear reaction codes, helping to produce reliable evaluated data needed for the development of ADS.

And ultimately contribute to the sustainability of nuclear energy.
Neutroninducerad lättjonproduktion from järn och vismut vid 175 MeV

Kärnkraft står för en betydande andel av elproduktionen i Sverige och hela världen. Enligt Energimyndighetens statistik från november 2010, stod kärnkraft under åren 2005-2009 för 43% av elproduktionen i Sverige, och vattenkraft för 46%. Därmed tillhör Sverige de OECD länder vars elproduktion genererar allra lägst koldioxidutsläpp per BNP. Globalt producerades år 2008 13,5% av elen i kärnkraftverk, en andel som under de senaste åren långsamt minskat från 17% år 2001. Under samma period har den globala elproduktionen ökat med 3.8% årligen och användningen av kol med i snitt 5%.


EU har inom sin Strategic Energy Technology Plan (SET) identifierat flera teknologier för elproduktion med låg koldioxidproduktion. Kärnkraft har identifierats som ett viktigt bidrag inom SET-planen och en Sustainable Nuclear Energy Technology Platform (www.SNETP.eu) har skapats som bland annat ska lösa avfallsproblematiken. Separation och transmutation är en metod som kan bidra till att minska mängden kärnavfall samtidigt som en betydligt större andel av bränslet kan utnyttjas än vad som är fallet i dagens
reaktorer. Både avfallsängden och lagringstiden för avfallet kan minskas cirka en faktor 100 jämfört med dagens situation. Istället för att behöva vänta i ca. 100 000 år tills aktiviteten i kärnavfallet når den nivå som motsvarar uranmalms skulle 1000 år vara tillräckligt.

En del i detta framtidsscenario för kärnkraft som ska leda till en sluten bränslecykel och hållbar kärnkraft är så kallade acceleratordrivna system (ADS). Här handlar det om underkritiska system som drs genom produktion av neutroner från en extern källa, en intensiv protonstråle med energin 600–1000 MeV. Protonerna träffar på ett strålmål av t ex bly och vismut och producerar på så sätt de neutroner som ska leda till kärnklyvningar i bränslet. Därmed är kontrollen över neutronflödet inte enbart beroende av de neutroner som produceras från själva fissionsreaktionerna. ADS kan användas som dedikerade reaktorer för transmutation, dvs. system som omvandlar klyvbart material som plutonium från det använda bränslet till kortlivade fissionsprodukter. Dessutom kan systemet också utformas för produktion av energi. En fördel med ADS är att nästan alla bränslesammansättningar kan användas i systemet, medan framtida kritiska reaktorer har vissa begränsningar för att garantera en säker drift. Med en framtida reaktorpark bestående av lättvattenreaktorer, snabba reaktorer och ADS kan en sluten bränslecykel och med mer hållbar energiproduktion för lång framtid bli verkligt.

För designen av ADS behövs nya tvärsnittsdata och förbättrade modeller för kärnreaktioner vid höga energier, speciellt neutroner. NEXT (Neutron data EXperiments for Transmutation) projektet vid Uppsala universitet, är ett bidrag till den europeiska insatsen för att förbättra vår kunskap om kärndata som är relevanta för transmutation. Inom projektet mäts de första experimentella data om produktion av lätta joner från neutroner med en energi i intervallet mellan 100 och 200 MeV. NEXT startades 2006 vid Institutionen för neutronforskning (numera avdelningen för tillämpad kärnfysik) vid Uppsala universitet med finansiering från Strålsäkerhetsmyndigheten SSM, SKB och Ringhalsverket AB. Stöd erhölls även från EU inom FP7 projektet ANDES.

Inom ramen för NEXT har vi mätningar utförts av dubbel-differentiella tvärsnitt för produktion av lätta joner från järn och vismut av kvassimonoenergetiska neutroner (QMN) med energier up till 175 MeV. Mätningarna genomfördes under 2009 med Medley uppställningen vid The Svedberg-laboratoriet i Uppsala.

I avhandlingen jämförs de experimentella resultaten med teoretiska modellräkningar. Beräkningar har utförts med den i Europa utvecklade koden TALYS-1.2, en modifierad version av den japanska JQMD koden, och den senaste versionen av den amerikanska koden MCNP6. Dessa koder kan användas för att producera evaluerade kärndata bibliotek, förutsätta resultat för kärnreaktioner som inte har mätts, och förstå reaktionsmekanismer i
växelverkan mellan både neutroner och lätta joner och ett brett spektrum av olika material.

Jämförelser mellan våra mätdata och beräkningar med TALYS koden tillät oss att hitta ett nytt energiberoende för produktion av sammansatta lätta joner. Detta resultat kommer att förbättra kodens prognosförmåga vilket bidrar till att producera bättre kärndatabibliotek. Våra data har också visat på behovet av att inkludera en mekanism som beskriver emissionen av fler än en sammansatt lättjon innan kärnan når jämvikt i TALYS koden, s k *multiple pre-equilibrium emission*. Denna mekanism tas det nu endast hänsyn till vid produktion av protoner och neutroner.

Mätdata jämförs också med JQMD koden, som utvidgades med en modell för produktion av sammansatta partiklar vid kärnans yta, en s.k. *surface coalescence model*. De experimentella mätdata bekräftar betydelsen av denna mekanism. Utöver detta visas att den nuvarande beskrivningen behöver kompletteras med en mekanism för direkt produktion som bättre beskriver de observerade energispektrumen för sammansatta partiklar i framåtriktningen.

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This was the dedication of my Licentiate Thesis. This is the dedication of my life. Your support and your love contributed to the success of my doctoral studies more than anything else. In the harsh climate of Sweden as well as in the humid climate of Kyushu, you and me walked hand in hand. Nothing is out of our reach. The thesis is finally handed in, now is time to concentrate on our wedding! June 18, 2011. I love you.

Some words on my life in Uppsala
The lack of sun and warmthness in the dry sky of Uppsala would have not been bearable without the support, the friendship and the great heart of my friends. First of all Bea and Lukasz, my second family here in Sweden. I still remember the funny coincidences of our first meeting at Karin’s goodbye party. Bea, do you remember the first coffees in the city and the work together in the nations when Lukasz was in Falun! And then all together, the barbecues in the park, lunches, dinners, and hot teas. You, guys, are the best part of this city! Szczęśliwego nowego roku!

And if I am strolling downtown Uppsala, and someone will see me and yell "Duuuuude!", well, it would be for sure Suzan! The city would be empty without your energy! And I am just grateful that I was in Japan, when you were in Canada! Thank you for everything. You know that we love you!

The circumstances of my first meeting with Izabela and Justyna should be probably left out of a doctoral thesis. But it was Halloween, and the fact that I was dressed up with a red bathing-gown was just part of it. You girls cheered up my days in Uppsala, and I hope we will have again the chance to live in the same city!

If you think, dear reader of this dissertation, that my writing was too lengthy, I have only one explanation: you are the only person in this city who did not meet (yet) Marco Chiaretta! I did my best to write more than he did in his thesis. It was a lost battle. Marco, che dire, sono davvero contento di averti incortrato, e sono sicuro che non ci perderemo di vista! Mi raccomando, continua ad essere quello che sei!

The Beach Boys were singing "I wish they all could be California girls"! This was before Naffe did spend a year there. Now they sing about Linköping girls. You rock girl, and you are probably the only person I know that traveled more than (or at least as much as) me and Niroj did, last year.

I have been an active member at Värmlands Nation, first as wardrobe staff, then as Dörrchef and finally as International Secretary. I have also worked for
three years as waiter at the “2 rum & kök” restaurant and at the Sommarrestaurang at GH Nation. In this quality, I thank all the customers who gave me a tip! (Yes, if you like me, tip me!)

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An exception to this rule are Igor and Nives. I had to give up the regular dinner I could enjoy at their place, at least once a week, when I was living in Trieste, and the long discussions about "the meaning of life, the universe, and everything". But we have been able to keep in touch with mobile telephones and Skype, and to catch up in (almost) all my visits to Italy.

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and about my life in Japan

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I will never forget the first sight of sakura, the sounds of cicadas, the rainy season, the typhoon season, the red lanterns in Nagasaki, the festivals in Fukuoka and Maiko-san in Kyoto.
In italiano

Se mi aveste chiesto, all’età di tre anni, "Riccardo, che vuoi fare da grande?", avrei certo risposto "il premio Nobel per la fisica". Mia madre, credetemi, ne ha tutta la responsabilità. Qualche anno dopo ridimensionai le mie aspettative, ed alla stessa domanda rispondevo con più umiltà di voler un futuro da Papa, o in alternativa da Supereroe. Avendo intrapreso con successo entrambe le carriere, prima quella papale in seminario, e poi quella eroica come autista di ambulanza, non mi è rimasta altra scelta che avventurarmi sulla strada della fisica. Ed oggi eccomi qui, a pubblicare la tesi di dottorato.

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A. Complete data set.

“What is the use of a book”, thought Alice, “without pictures or conversations?”
(Lewis Carrol, Alice’s Adventures in Wonderland)

Double-differential cross sections for inclusive light-ion production in the interaction of 175 MeV QMN with Fe and Bi are reported here, in comparison with model predictions. Results are presented at eight angles in the laboratory system, from 20 degrees to 160 degrees. Model calculations are folded with the accepted neutron spectrum presented in Fig. 3.3.
Figure A.1: Experimental $\sigma(E,\theta)$ for the Fe(n,px) reaction at 175 MeV QMN. Calculations: default TALYS-1.2 calculations (solid black line); QMD with surface coalescence model following Watanabe and Kadrev (short-dashed blue line); MCNP6 calculations with the CEM03.02 event generator (long-dashed red line).
Figure A.2: Experimental $\sigma(E,\theta)$ for the Fe(n,dx) reaction at 175 MeV QMN. Calculations: default TALYS-1.2 calculations (solid black line); QMD with surface coalescence model following Watanabe and Kadrev (short-dashed blue line); MCNP6 calculations with the CEM03.02 event generator (long-dashed red line).
Figure A.3: Experimental $\sigma(E,\theta)$ for the Fe(n,tx) reaction at 175 MeV QMN. Calculations: default TALYS-1.2 calculations (solid black line); QMD with surface coalescence model following Watanabe and Kadrev (short-dashed blue line); MCNP6 calculations with the CEM03.02 event generator (long-dashed red line).
Figure A.4: Experimental $\sigma(E, \theta)$ for the Fe(n, $^3$He) reaction at 175 MeV QMN. Calculations: default TALYS-1.2 calculations (solid black line); QMD with surface coalescence model following Watanabe and Kadrev (short-dashed blue line); MCNP6 calculations with the CEM03.02 event generator (long-dashed red line).
Figure A.5: Experimental $\sigma(E,\theta)$ for the Fe(n,\alpha) reaction at 175 MeV QMN. Calculations: default TALYS-1.2 calculations (solid black line); QMD with surface coalescence model following Watanabe and Kadrev (short-dashed blue line); MCNP6 calculations with the CEM03.02 event generator (long-dashed red line).
Figure A.6: Experimental $\sigma(E,\theta)$ for the Bi(n,px) reaction at 175 MeV QMN. Calculations: default TALYS-1.2 calculations (solid black line); MCNP6 calculations with the CEM03.02 event generator (long-dashed red line).
Figure A.7: Experimental $\sigma(E,\theta)$ for the Bi(n,dx) reaction at 175 MeV QMN. Calculations: default TALYS-1.2 calculations (solid black line); MCNP6 calculations with the CEM03.02 event generator (long-dashed red line).
Figure A.8: Experimental $\sigma(E,\theta)$ for the Bi(n,tx) reaction at 175 MeV QMN. Calculations: default TALYS-1.2 calculations (solid black line); MCNP6 calculations with the CEM03.02 event generator (long-dashed red line).
Figure A.9: Experimental $\sigma(E,\theta)$ for the Bi(n,$^3$He) reaction at 175 MeV QMN. Calculations: default TALYS-1.2 calculations (solid black line); MCNP6 calculations with the CEM03.02 event generator (long-dashed red line).
Figure A.10: Experimental $\sigma(E,\theta)$ for the Bi(n,\alpha) reaction at 175 MeV QMN. Calculations: default TALYS-1.2 calculations (solid black line); MCNP6 calculations with the CEM03.02 event generator (long-dashed red line).
Bibliography


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