Light-ion production in the interaction of 96 MeV neutrons with silicon

U. Tippawan\textsuperscript{1,2}, S. Pomp\textsuperscript{1,}\textsuperscript{*}, A. Ataca\textsuperscript{1}, B. Bergenwall\textsuperscript{1}, J. Blomgren\textsuperscript{1}, S. Dangtip\textsuperscript{1,2}, A. Hildebrand\textsuperscript{1}, C. Johansson\textsuperscript{1}, J. Klug\textsuperscript{1}, P. Mermod\textsuperscript{1}, L. Nilsson\textsuperscript{1,4}, M. Österlund\textsuperscript{1}, N. Olsson\textsuperscript{1,3}, K. Elmgren\textsuperscript{3}, O. Jonsson\textsuperscript{4}, A.V. Prokofiev\textsuperscript{4}, P.-U. Renberg\textsuperscript{4}, P. Nadel-Turonski\textsuperscript{5}, V. Corcalciuc\textsuperscript{6}, Y. Watanabe\textsuperscript{7}, A. Koning\textsuperscript{8}

\textsuperscript{1}Department of Neutron Research, Uppsala University, Sweden
\textsuperscript{2}Fast Neutron Research Facility, Chiang Mai University, Thailand
\textsuperscript{3}Swedish Defence Research Agency, Stockholm, Sweden
\textsuperscript{4}The Svedberg Laboratory, Uppsala University, Sweden
\textsuperscript{5}Department of Radiation Sciences, Uppsala University, Sweden
\textsuperscript{6}Institute of Atomic Physics, Heavy Ion Department, Bucharest, Romania
\textsuperscript{7}Department of Advanced Energy Engineering Science, Kyushu University, Japan
\textsuperscript{8}Nuclear Research and Consultancy Group, Petten, The Netherlands

Abstract

Double-differential cross sections for light-ion (p, d, t, \textsuperscript{3}He and \textalpha) production in silicon, induced by 96 MeV neutrons are reported. Energy spectra are measured at eight laboratory angles from 20° to 160° in steps of 20°. Procedures for data taking and data reduction are presented. Deduced energy-differential, angle-differential and production cross sections are reported. Experimental cross sections are compared to theoretical reaction model calculations and experimental data in the literature.

PACS numbers: 25.40.-h, 25.40.Hs, 25.40.Kv, 28.20.-v

\textsuperscript{*}Corresponding author, Tel. +46 18 471 6850, Fax. +46 18 471 3853, E-mail: stephan.pomp@tsl.uu.se
1 Introduction

In the last years, there has been an increasing request for experimental studies of fast-neutron induced reactions, especially at higher incident neutron energies. For basic physics, nucleon-induced reactions provide useful means to investigate nuclear structure, to characterize reaction mechanisms and to impose stringent constraints on nuclear model calculations. The silicon nucleus is sufficiently heavy for many of the statistical assumptions to hold (high density of excited states), yet not so heavy to give a strong suppression of charged particle emission due to Coulomb barrier effects. Therefore, nuclear reaction models for equilibrium and pre-equilibrium decay can be tested and benchmarked. Experimental data in the literature at incident neutron energies from reaction thresholds up to 60 MeV [1] and between 25 and 65 MeV [2] offer possibilities to test the predictions of reaction models.

In recent years, an increasing number of applications involving fast neutrons have been developed or are under consideration, e.g., radiation treatment of cancer [3], soft-error effects in computer memories [6], accelerator-driven transmutation of nuclear waste and energy production [7], and determination of the response of neutron detectors [8]. Silicon data are particularly important for detailed soft-error simulation in electronic devices [6, 9].

In this paper, we present experimental double-differential cross sections (inclusive yields) for protons, deuterons, tritons, $^3$He and alpha particles produced by 96 MeV neutrons incident on silicon. Measurements have been performed at the cyclotron of The Svedberg Laboratory (TSL), Uppsala, using the dedicated MEDLEY experimental setup [10]. Spectra have been measured at 8 laboratory angles, ranging from $20^\circ$ to $160^\circ$ in $20^\circ$ steps. Extrapolation procedures are used to obtain coverage of the full angular distribution and consequently energy-differential and production cross sections are deduced, the latter by integrating over energy and angle. The experimental data are compared to results of calculations with nuclear reaction codes and to existing experimental data.

The experimental methods are briefly discussed in sect. 2 and data reduction and correction procedures are presented in sect. 3 and 4, respectively. The theoretical framework is presented in sect. 5. In sect. 6 experimental results are reported and compared with theoretical and previous experimental data. Conclusions and an outlook are given in sect. 7.
2 Experimental setup and methods

The neutron beam facility at TSL uses the $^7\text{Li}(p,n)^7\text{Be}$ reaction ($Q = -1.64 \text{ MeV}$) to produce a quasi-monoenergetic neutron beam [11]. The lithium target was 26 mm in diameter and 8 mm thick in the present experiment and enriched to 99.98 % in $^7\text{Li}$. The 98.5±0.3 MeV protons from the cyclotron impinge on the lithium target, producing a full-energy peak of neutrons at 95.6±0.5 MeV with a width of 1.6 MeV (FWHM) and containing 40 % of the neutrons, and an almost constant low-energy tail containing 60 % of the neutrons. The neutron beam is shaped by a collimator system, and delivered to the experimental area. After passage of the target, the proton beam is deflected by two magnets into a well-shielded beam dump, where the beam current is integrated in a Faraday cup. The integrated charge serves as one neutron beam monitor. With a beam intensity of about 5 $\mu\text{A}$, the neutron flux at the target location is about $5 \cdot 10^4$ neutrons/(s·cm²). The collimated neutron beam has a diameter of 80 mm at the location of the target. A thin film breakdown counter (TFBC) [12] installed after the reaction chamber is used as another beam monitor. The two beam monitor readings were in agreement during the measurements.

The charged particles are detected by the MEDLEY setup [10]. It consists of eight three-element telescopes mounted inside a 100 cm diameter evacuated reaction chamber. Each telescope consists of two fully depleted $\Delta E$ silicon surface barrier detectors and a CsI(Tl) crystal. The thickness of the first $\Delta E$ detector ($\Delta E_1$) is either 50 or 60 $\mu\text{m}$, while the second one ($\Delta E_2$) is either 400 or 500 $\mu\text{m}$. They are all 23.9 mm in diameter (nominal). The cylindrical CsI(Tl) crystal, 50 mm long and 40 mm in diameter, serves as the $E$ detector. The back-end part of the crystal, 20 mm long, has a conical shape, tapered off to 18 mm diameter, to fit the size of a read-out diode.

To obtain a well-defined acceptance, a plastic scintillator collimator is placed in front of each telescope. The active collimators have an opening of 19 mm diameter and a thickness of 1 mm.

A Passivated Implanted Planar Silicon (PIPS) detector is used as an active target. It has a 32·32 mm² quadratic shape and a thickness of 303 $\mu\text{m}$. It is suspended in a thin aluminium frame using threads and small springs. The dimensions of the frame have been chosen in such a way that it does not interfere with the incident neutron beam. Besides the energy deposited by the detected light ion, the active target recorded the energy deposition due to other products, like recoils, of the same event. This information was, however, not used in the present analysis.

For absolute cross section normalization, a 25 mm diameter and 1.0 mm thick polyethylene $(\text{CH}_2)_n$ target is used. The $np$ cross section at 20° labo-
ratory angle provides the reference cross section [13].

Background is measured by removing the target from the neutron beam. It is dominated by protons produced by neutron beam interaction with the beam tube and reaction chamber material, especially at the entrance and exit of the reaction chamber and in the telescope housings. Therefore, the telescopes at 20° and 160° are most affected. Since the protons in the background originated not from the target but came from different directions, they can be misidentified leading to a large background even for the other particles. For the 160° telescope, i.e., the worst case, the signal-to-background ratios are 2.5, 1 and 0.1 for protons, deuterons and tritons, respectively, whereas the corresponding numbers for the 40° telescope, i.e., the best case, are 8, 12 and 5. In the case of ³He and alpha particles, the background is negligible.

The time-of-flight (TOF) obtained from the radio frequency of the cyclotron (stop signal for the TDC) and the timing signal from each of the eight telescopes (start signal), is measured for each charged-particle event.

The raw data are stored event by event for on-line monitoring and subsequent off-line analysis. Typical count rates for target-in and target-out runs were 10 and 2 Hz, respectively. The dead time of the system was typically 1-2 % and never exceeding 10 %.

3 Data reduction procedures

3.1 Particle identification and energy calibration

The ΔE − E technique is used to identify light charged particles ranging from protons to lithium ions, which is illustrated in Fig. 1a. Good separation of all particles is obtained over their entire energy range. Since the energy resolution of each individual detector varies with the particle type, the particle identification cuts are defined to cover 3σ, where σ is the standard deviation of the energy resolution of each particle type. Typical energy resolutions of the thin ΔE detectors are between 40 and 80 keV, increasing with particle mass. The corresponding values are between 150 and 550 keV for the thick ΔE detectors and between 900 and 1200 keV for the E detectors. For energy depositions in the E detector above 70 MeV in Fig. 1b, the two-dimensional cuts for protons, deuterons and tritons overlap slightly since the energy loss of the hydrogen isotopes in the ΔE₂ detector is rather small. This ambiguity is resolved by a two-dimensional plot (inset of Fig. 1b) of the deviations of the ΔE₁ and ΔE₂ signals from tabulated energy loss values in silicon [14] (solid lines in Fig. 1b). Particle identification is done by cutting along the minimum contour line, and thus possible misidentification should even out.
This technique is also used to improve the separation between $^3$He and alpha particles in some telescopes where the energy resolution is poor.

Energy calibration of all detectors is obtained from the data itself [15]. Events in the $\Delta E - E$ bands are fitted with respect to the energy deposited in the three detectors (solid lines in Fig. 1). This energy is determined from the detector thicknesses and tabulated energy loss values in silicon [14]. The $\Delta E_1$ detectors are further calibrated and checked using a 5.48 MeV alpha source. For the energy calibration of the CsI(Tl) detectors, two parameterizations of the light output versus energy of the detected particle [10] are used, one for hydrogen isotopes and another one for helium isotopes. Supplementary calibration points are provided by the H(n,p) reaction, as well as transitions to the ground state and low-lying states in the $^{12}$C(n,d)$^{11}$B and $^{28}$Si (n,d)$^{27}$Al reactions. The energy of each particle type is obtained by adding the energy deposited in each element of the telescope.

Low-energy charged particles are stopped in the $\Delta E_1$ detector leading to a low-energy cutoff for particle identification of about 3 MeV for hydrogen isotopes and about 8 MeV for helium isotopes (see Fig. 1a). The helium isotopes stopped in the $\Delta E_1$ detector are nevertheless analyzed and a remarkably low cutoff, about 4 MeV, can be achieved for the experimental alpha-particle spectra. These alpha-particle events could obviously not be separated from $^3$He events in the same energy region, but the yield of $^3$He is much smaller than the alpha-particle yield in the region just above 8 MeV, where the particle identification works properly. That the relative yield of $^3$He is small is also supported by the theoretical calculations in the evaporation peak region. In conclusion, the $^3$He yield is within the statistical uncertainties of the alpha-particle yield for alpha energies between 4 and 8 MeV. A consequence of this procedure is that the $^3$He spectra have a low-energy cutoff of about 8 MeV.

### 3.2 Low-energy neutron rejection and background subtraction

Knowing the energy calibration and the flight distances, the flight time for each charged particle from target to detector can be calculated and subtracted from the registered total TOF. The resulting neutron TOF is used for selection of charged-particle events induced by neutrons in the main peak of the incident neutron spectrum. The TOF cut reduces the background of charged particles produced by peak neutrons hitting the chamber and telescope housing since the flight paths are different, especially for the backward telescopes. The widths of the TOF cuts in all detectors are fixed to $3\sigma$ where
\( \sigma \) is the standard deviation of the \( H(n,p) \) peak in the 20° telescope. Fig. 2a illustrates the selection procedure for deuterons at 20° laboratory angle. The solid line is a kinematic calculation of the ground state peak in the deuteron spectra for each corresponding neutron energy. It provides a cross check of the energy and time calibration of the whole energy spectrum.

Background events, measured in target-out runs and analyzed in the same way as target-in events, are subtracted from the corresponding target-in runs after normalization to the same neutron fluence. Fig. 2b shows the resulting spectrum of deuteron events at 20° induced by the main neutron peak. For comparison, the same spectrum without TOF cut is presented. Finally, the target-out background, obtained with the same TOF cut is shown. The signal-to-background ratio is about 4.

### 3.3 Absolute cross section normalization

Absolute double-differential cross sections are obtained by normalizing the silicon data to the number of recoil protons emerging from the \( \text{CH}_2 \) target. After selection of events in the main neutron peak and proper subtraction of the target-out and \( ^{12}\text{C}(n,p) \) background contributions, the latter taken from a previous experiment, the cross section can be determined from the recoil proton peak, using \( np \) scattering data [13]. All data have been normalized using the \( np \) scattering peak in the 20° telescope. As a cross check, \( \text{Si}(n,px) \) spectra have also been normalized using the \( np \) scattering peak in the 40° and 60° telescopes, resulting in spectra in agreement with those normalized to the 20° telescope.

### 4 Corrections

#### 4.1 Thick target correction

Due to the thickness of the target and to the low-energy cutoffs in the particle identification, the measured low-energy charged particles are produced in fractions of the entire thickness of the target. Therefore, not only energy-loss corrections are needed but also particle-loss corrections. Charged particles with the initial kinetic energy \( E_{\text{init}} \) have a well-defined range \( R \) in the target material. If \( R(E_{\text{init}}) \) is equal to or larger than the target thickness, all produced particles can escape from the target and no particle loss correction is required. If, on the other hand, \( R(E_{\text{init}}) \) is smaller than the target thickness, a correction for particles stopped inside the target is needed.
The adopted correction method employs an initial energy \( (E_{\text{init}}) \) distribution called the inverse response function for each measured energy. For the 303 \( \mu \text{m} \) silicon target used in the present experiment, a measured alpha particle of 4 MeV could either be due to a 4 MeV particle from the front surface of the target, a 27 MeV particle from the back surface of the target, or anything in between. Therefore, the content of the measured energy bin should be redistributed over the initial energy region from 4 to 27 MeV. A FORTRAN program, TCORR \[16\], has been developed which calculates the inverse response functions, initially assuming an energy-independent cross section. These inverse response functions are normalized to the corresponding bin content in the measured spectrum and summed to get the true initial energy spectrum. Finally, the particle loss correction is applied. The resulting spectrum is folded with the primary inverse response functions to get improved inverse response functions and the procedure is repeated. Resulting spectra from two successive iterations are compared by a Kolmogorov test \[17\] to judge the convergence.

Results from the correction method have been verified with an independent Monte-Carlo program called TARGSIM, based on the GEANT code \[18\]. This program simulates the measured spectra using the corrected spectra and the MEDLEY geometry as input. The simulation results are in agreement with the experimental data within the statistical errors over the whole energy region.

Obviously, the simulated spectra have much better statistics than the original experimental spectra and, therefore, the statistical fluctuations between neighboring energy bins are much smaller. In a sense, they are fits to the experimental spectra. In order to estimate the systematic uncertainty introduced by the thick target correction, these simulated spectra are corrected with TCORR again and compared with the result of the first correction. The observed differences for individual energy bins are typically 5 % and less than 10 % in general. An extreme value of 40 % was found in the lowest bin of the alpha spectrum at 1400. However, in all cases, except for protons where the statistical errors are very small, the deviations are within the statistical uncertainties of the original corrected data.

In conclusion, the systematic error of the target correction comes essentially from the statistical uncertainties. For protons and deuterons we estimate that this error is about 10 % in the lowest two energy bins decreasing to a few percent from 15 MeV and upwards. Due to less statistics and the increasing width of the inverse response functions, the uncertainty is larger for tritons, \(^3\)He and alpha particles, where it is 20 % in the lowest two bins, decreasing to 10 % above 25 MeV.

In addition, evaluated data \[19\] were used as input to check the reliability
of our programs, obviously because validation with known realistic data is desirable. The latter have been simulated with the TARGSIM program to get pseudo-experimental data and have subsequently been corrected with the TCORR program using the same conditions as in the experiment. The corrected results appear to reproduce the known realistic data well.

4.2 Collimator correction

As mentioned in sect. 2, active collimators have been placed in front of the telescope in order to define the solid angle. However, due to malfunctioning in the present experiment, the signal from these collimators could not be used to suppress events hitting them. Therefore, although the collimators actually work as passive collimators for helium particles below 35 MeV, their effect when particles punch through them has to be corrected for. To this end, a FORTRAN program has been developed that, based on the measured spectrum of particles and an iteration procedure, estimates the shape and fraction of the energy spectrum of particles hitting the collimator. It has been found that the corrections in shape are rather small and under control in all cases. The systematic error related to this correction comes from the uncertainty in the solid angle subtended by the silicon detectors for high-energy protons relative to the solid angle subtended by the collimator opening. This uncertainty is estimated to be 5 % and, due to the normalization procedure, only affects the helium spectra and the low-energy part of the hydrogen spectra.

4.3 Other corrections

The 17 MHz repetition rate of the cyclotron beam pulse, which limits the TOF window to 58 ns, causes wrap-around problems. Thus, it is not possible to distinguish 96 MeV neutrons from those of 26 MeV created by the previous beam burst, since the latter have the same apparent TOF. This can be seen in Fig. 2a, where the bent band from the low-energy neutron tail crosses the straight band of the full-energy neutrons. Since the Q-value for the $^{28}\text{Si}(n,d)$ reaction is $-9.4$ MeV, this interference shows up as a bump below 20 MeV in Fig. 2b. A correction for this effect is applied, using tabulated values from Ref. 23 and a ratio of the neutron fluence in the wrap-around region and at 96 MeV of 6.3 %. For the neutron-energy spectrum in the wrap-around region, a square distribution ranging from 24 to 29 MeV is assumed. Thus, the cross sections at 24, 26 and 28 MeV as given in Ref. 23 are used, with the 24 MeV values entering at half weight. The data for 80, 100, 120, 140 and 160 degrees are obtained by linear interpolation. Only the spectra for proton, deuteron and alpha particles are corrected. The effect of this correction is a
reduction of about 5% in the production cross section. In the (n,d) spectra presented in Fig. 4, some structure at 20° and 40° around 15 MeV might be attributed to deficiencies in the correction. The triton production cross sections given in Ref. [23] (Q-value = -16.2 MeV) indicate that the correction would be about an order of magnitude lower and is therefore negligible. For 3He production (Q-value = -12.1 MeV), the correction is also negligible due to the high-energy cutoff of 8 MeV.

There is a TOF shift problem, seen as a band parallel with the main band in Fig. 2a. The reason for this is probably that the electronic timing module has not worked properly. This is corrected by extending the TOF cut with the dotted rectangle in the same figure, to include these events. This method could be applied only in the energy region where there is no interference from the low energy neutron tail. Therefore, the ratio of the number of events between the parallel and the main band is determined and then applied to the low-energy region as well. This ratio is 1.3% in the worst case.

Albeit a majority of the neutrons appears in the narrow full-energy peak at 95.6 MeV, a significant fraction (about 25%) belongs to a tail extending towards lower energies, remaining also after the TOF cut. The average neutron energy with these tail neutrons included is 92.4 MeV. This effect has been taken into account in the normalization of the data.

Minor corrections of a few percent are applied to the experimental spectra for the CsI(Tl) intrinsic efficiency [11] and for the dead time in the data acquisition system.

5 Theoretical models

Data have been compared with nuclear theory predictions, computed with the two nuclear reaction codes GNASH [20, 21] and TALYS [22]. While GNASH has been widely used during the last years, TALYS is a new code still under development. Two sets of GNASH calculations are presented, one with parameters as presented in a recent evaluation [23], and another set with modified parameters [19] as described in sect. 5.2. The latter parameter set is developed as part of another data evaluation [24]. Since the latter work and TALYS are not published, they are described in some detail below.

Both GNASH and TALYS integrate direct, pre-equilibrium, and statistical nuclear reaction models into one calculation scheme and thereby give predictions for all the open reaction channels. Both codes use the Hauser-Feshbach model for sequential equilibrium decay and the exciton model for pre-equilibrium emission. The angular distributions are obtained using the
5.1 TALYS calculations

The purpose of TALYS is to simulate nuclear reactions that involve neutrons, photons, protons, deuterons, tritons, $^3$He and alpha particles, in the 1 keV – 200 MeV energy range. Predicted quantities include integrated, single- and double-differential cross sections, for both the continuum and discrete states, residue production and fission cross sections, gamma-ray production cross sections, etc. For the present work, single- and double-differential cross sections are of interest. To predict these, a calculation scheme is invoked which consists of a direct + pre-equilibrium reaction calculation followed by subsequent compound nucleus decay of all possible residual nuclides calculated by means of the Hauser-Feshbach model.

First, dedicated optical model potentials (OMP) were developed for both neutrons and protons on $^{28}$Si up to 200 MeV. The used parameters are from the OMP collection of Ref. [26]. These potentials provide the necessary reaction cross sections and transmission coefficients for the statistical model calculations. For complex particles, the optical potentials were directly derived from the nucleon potentials using the folding approach of Watanabe [27].

Pre-equilibrium emission takes place after the first stage of the reaction but long before statistical equilibrium of the compound nucleus is attained. It is imagined that the incident particle step-by-step creates more complex states in the compound system and gradually loses its memory of the initial energy and direction. The default pre-equilibrium model of TALYS is the two-component exciton model of Kalbach [28]. In the exciton model (see Refs. [29] and [30] for extensive reviews), at any moment during the reaction, the nuclear state is characterized by the total energy $E_{\text{tot}}$ and the total number of particles above and holes below the Fermi surface. Particles ($p$) and holes ($h$) are indiscriminately referred to as excitons. Furthermore, it is assumed that all possible ways of sharing the excitation energy between different particle-hole configurations with the same exciton number $n = p + h$ have equal probability. To keep track of the evolution of the scattering process, one merely traces the development of the exciton number, which changes in time as a result of intranuclear two-body collisions.

The basic starting point of the exciton model is a time-dependent master equation, which describes the probability of transitions to more and less complex particle-hole states as well as transitions to the continuum, i.e., emission. Upon integration over time, the energy-averaged emission spectrum is obtained. The assumptions above make the exciton model amenable to practical calculations. This, however, requires the introduction of a free
parameter, namely the average matrix element of the residual two-body interaction, occurring in the transition rates between two exciton states. Without going into details, the basic formulae are given for the two-component exciton model. The created particles and holes of proton and neutron type are explicitly followed throughout the reaction. A notation is used in which \( p_\pi(p_\nu) \) is the proton (neutron) particle number and \( h_\pi(h_\nu) \) the proton (neutron) hole number. Following Kalbach \[28\], the exciton model cross section is now given by

\[
\frac{d\sigma^{EM}}{dE_k} = \sigma^{CF} \sum_{p_\pi=p_{\pi}^0} \sum_{p_\nu=p_{\nu}^0} w_k(p_\pi, h_\pi, p_\nu, h_\nu, E_k) S_{pre}(p_\pi, h_\pi, p_\nu, h_\nu),
\]

where \( \sigma^{CF} \) is the compound formation cross section and \( S_{pre} \) the time-integrated strength which determines how long the system remains in a certain exciton configuration \[28\]. The initial proton and neutron particle numbers are denoted \( p_{\pi}^0 = Z_p \) and \( p_{\nu}^0 = N_p \) with \( Z_p(N_p) \) being the proton (neutron) number of the projectile. In general, \( h_\pi = p_\pi - p_{\pi}^0 \) and \( h_\nu = p_\nu - p_{\nu}^0 \), so that the initial hole numbers are zero, i.e. \( h_{\pi}^0 = h_{\nu}^0 = 0 \), for primary pre-equilibrium emission. The pre-equilibrium part is calculated by Eq. 1 using \( p_{\pi}^{eq} = p_{\nu}^{eq} = 6 \), whereas the remainder of the reaction flux is distributed through the Hauser-Feshbach model. In addition, the never-come-back approximation is adopted.

The emission rate \( w_k \) for ejectile \( k \) with spin \( s_k \) is given by

\[
w_k(p_\pi, h_\pi, p_\nu, h_\nu, E_k) = \frac{2s_k + 1}{\pi^2\hbar^3} \mu_k E_k \sigma_{k,inv}(E_k) \frac{\omega(p_\pi - Z_k, h_\pi, p_\nu - N_k, h_\nu, E_x)}{\omega(p_\pi, h_\pi, p_\nu, h_\nu, E_{tot})},
\]

where \( \sigma_{k,inv}(E_k) \) is the inverse reaction cross section as calculated from the optical model and \( \omega \) is the two-component particle-hole state density. The expression for \( S_{pre} \) contains the adjustable transition matrix element \( M^2 \) for each possible transition between neutron-proton exciton configurations. A proton-neutron ratio of 1.6 for the squared internal transition matrix elements was adopted to give the best overall agreement with experiment, i.e., \( M_{\pi\nu}^2 = M_{\nu\pi}^2 = 1.6M_{\pi\pi}^2 = 1.6M_{\nu\nu}^2 \). Partial level density parameters \( g_\pi = Z/15 \) and \( g_\nu = N/15 \) were used in the equidistant spacing model for the partial level densities.

At incident energies above several tens of MeV, the residual nuclides formed after binary emission may have so large excitation energy that the presence of additional fast particles inside the nucleus becomes possible. The latter can be imagined as strongly excited particle-hole pairs resulting from the first binary interaction with the projectile. The residual system is then
clearly non-equilibrated and the excited particle that is high in the continuum may, in addition to the first emitted particle, also be emitted on a short time scale. This so-called multiple pre-equilibrium emission forms an alternative theoretical picture of the intra-nuclear cascade process, whereby the exact location and momentum of the particles are not followed, but instead the total energy of the system and the number of particle-hole excitations (exciton number).

In actual calculations, the particle-hole configuration of the residual nucleus after emission of the ejectile, is re-entered as initial condition in Eq. 1. When looping over all possible residual configurations, the multiple pre-equilibrium contribution is obtained. In TALYS, multiple pre-equilibrium emission is followed up to arbitrary order, though for 96 MeV only secondary pre-equilibrium emission is significant.

It is well-known that semi-classical models, such as the exciton model, have always had some problems to describe angular distributions (essentially because it is based on a compound-like concept instead of a direct one). Therefore, as mentioned previously, the double-differential cross sections are obtained from the calculated energy spectra using the Kalbach systematics [25].

To account for the evaporation peaks in the charged-particle spectra, multiple compound emission was treated with the Hauser-Feshbach model. In this scheme, all reaction chains are followed until all emission channels are closed. The Ignatyuk model [31] has been adopted for the total level density to account for the damping of shell effects at high excitation energies.

For pre-equilibrium reactions involving deuterons, tritons, $^3$He and alpha particles, a contribution from the exciton model is automatically calculated with the formalism described above. It is, however, well known that for nuclear reactions involving projectiles and ejectiles with different particle numbers, mechanisms like stripping, pick-up and knock-out play an important role and these direct-like reactions are not covered by the exciton model. Therefore, Kalbach developed a phenomenological contribution for these mechanisms [32], which is included in TALYS. It has recently been shown (see Table I of Ref. [33]) that this method gives a considerable improvement over the older methods. The latter seemed to consistently underpredict neutron-induced reaction cross sections.

5.2 GNASH calculations

For the present work, GNASH calculations have been performed with a modified parameter set. The calculation procedure is outlined in Ref. [24]. Transmission coefficients needed for the GNASH input were calculated using the
optical potential parameters by Sun et al. [34] for neutrons and protons, Daehnick et al. [35] for deuterons, Becchetti-Greenlees [36] for tritons and $^3$He particles, and Avrigeanu et al. [37] for alpha particles.

Like in the TALYS case, default level density parameters were used with the Ignatyuk level density formula [31]. The normalization factor used in the pre-equilibrium model calculation was determined by analyses of proton-induced reactions. The calculated result of pre-equilibrium deuteron and alpha emission is different from that of the original GNASH code calculation [23]. In the deuteron emission, the component with the exciton number 3 was ignored. The direct pick-up component was calculated using a phenomenological approach [38] with a normalization that is independent of the incident energy. This normalization was determined from analysis of experimental (n,dx) energy spectra up to 60 MeV [1]. The alpha knockout component given by the same phenomenology [38] was ignored.

The results are given in the laboratory system. Like in the TALYS case, angular distributions are obtained using the Kalbach systematics [25]. The required pre-equilibrium fraction is taken from the GNASH output. The c.m.-to-lab transformation is performed using the kinematics of one-particle emission as described in Refs. [20, 21].

The exciton model implemented in GNASH is a one-component exciton model developed by Kalbach [39], with a parameterisation for the energy dependence of the squared internal transition matrix element which has been validated at relatively low incident energies (below 40 MeV). There are indications that at higher incident energies, this energy dependence is no longer appropriate and that a more general form, covering a wider energy range, is needed. Such a smooth form has been implemented in TALYS, on the basis of a collection of double-differential (nucleon-in,nucleon-out) cross section measurements [22].

6 Results and discussion

Double-differential cross sections at laboratory angles of 20°, 40°, 100° and 140° for protons, deuterons, tritons, $^3$He and alpha particles are shown in Figs. 3 - 7, respectively. All spectra for each particle type are plotted on the same cross section scale to facilitate the comparison of their magnitude. The choice of the energy bin width is a compromise between the energy resolution in the experiment, the width of the inverse response functions and acceptable statistics in each energy bin. The error bars represent statistical uncertainties only.

The overall relative statistical uncertainties of individual points in the
double-differential energy spectra at 20° are typically 3 % for protons, 7 % for deuterons, 20 % for tritons, 20 % for $^3$He and 15 % for alpha particles. As the angular distributions are forward-peaked, these values increase with angle. The systematic uncertainty contributions are due to thick target correction (1 − 20 %), collimated solid angle (1 − 5 %), beam monitoring (2 − 3 %), the number of silicon nuclei (1 %), CsI(Tl) intrinsic efficiency (1 %), particle identification (1 %) and dead time (<0.1 %). The uncertainty in the absolute cross section is about 5 %, which is due to uncertainties in the $np$ scattering angle, the contribution from the low-energy continuum of the $^7$Li(p,n) spectrum to the $np$ scattering proton peak (3 %), the reference $np$ cross sections (2 %) [13], statistics in the $np$ scattering proton peak (2 %), the carbon contribution (0.1 %) and the number of hydrogen nuclei (0.1 %).

From Figs. 3 - 7 it is obvious that the charged-particle emission from 96 MeV neutron irradiation of silicon is dominated by proton, deuteron and alpha particle channels. The spectra of the two other particle types studied in this work (tritons and $^3$He) are more than an order of magnitude weaker. All the spectra have more or less pronounced peaks at low energies (below 10−15 MeV), the angular distributions of which are not too far from isotropy. This low-energy peak is not observed in the $^3$He spectra due to the 8 MeV low-energy cutoff discussed in sect. 3.1.

All the particle spectra at forward angles show relatively large yields at medium-to-high energies. The emission of high-energy particles is strongly forward-peaked and hardly visible in the backward hemisphere. It is a sign of particle emission before statistical equilibrium has been reached in the reaction process. In addition to this broad distribution of emitted particles, the deuteron spectra at forward angles show narrow peaks corresponding to transitions to the ground state and low-lying states in the final nucleus, $^{27}$Al. These transitions are most likely due to pick-up of weakly bound protons in the target nucleus, $^{28}$Si.

### 6.1 Comparison with theoretical model calculations

In Figs. 3-8 the experimental results are presented together with theoretical model calculations. The GNASH calculations of Ref. [23] have been done for protons, deuterons and alpha particles, whereas the other two calculations have been performed for all five particle types.

Fig. 3 shows a comparison between the double-differential (n,px) experimental spectra and the calculations based on the TALYS and GNASH models. For protons above 25 MeV, all calculations give a good description of the spectra. Below this energy, some differences can be observed, e.g., at forward angles TALYS gives a better description of the statistical peak than
the GNASH calculations.

The situation is quite different for the deuteron spectra (Fig. 4). None of the predictions do account for the data. At all angles deviations of a factor of two or more are present. At forward angles the high-energy part is strongly overestimated, indicating problems in the hole-strength treatment. There is a large difference in the spectral shapes calculated with the two versions of GNASH [19, 23]. This difference is due to the fact that emission from the configurations with exciton number 3 is neglected in the present GNASH calculations. This component is taken into account as a direct pickup component calculated with an empirical formula due to Kalbach [38].

For tritons (Fig. 5), the TALYS calculations give a slightly better description of the experimental data, whereas for $^3$He (Fig. 6) some large deviations can be observed. The TALYS calculations seem to account better for the spectrum shapes.

The overall description of the alpha particle spectra (Fig. 7) is fair. The GNASH calculations overpredict the high-energy data at forward angles, whereas the TALYS predictions are too large at backward angles.

The ability of the models to account for the low-energy peak caused by evaporation processes is not impressive. In general, the models tend to overpredict the cross sections. It should, however, be kept in mind that the peak maximum is close to (for $^3$He below) the low-energy cutoff, which complicates the comparison. Another complication in this context is that the c.m.-to-lab transformation of the calculated TALYS spectra could, at least in some cases, make a considerable difference. The GNASH cross sections are given in the lab system, but the c.m.-to-lab transformation is performed using the kinematics of one-particle emission [20, 21], which obviously is an approximation.

### 6.2 Integrated spectra

For each energy bin of the light-ion spectra, the experimental angular distribution is fitted by a simple two-parameter functional form, \( a \exp(b \cos \theta) \) [25].

This allows extrapolation of double-differential cross sections to very forward and very backward angles. In this way coverage of the full angular range is obtained. By integration of the angular distribution, energy-differential cross sections \((d\sigma/dE)\) are obtained for each ejectile. These are shown in Fig. 8 together with the theoretical calculations. All calculations are in good agreement with the proton experimental data over the whole energy range. In the cases of deuterons and alpha particles, the models overpredict the high-energy parts of the spectra.
The production cross sections are deduced by integration of the energy-differential spectra (see Table 1). As explained above, the experimental values in Table 1 have to be corrected for the undetected particles below the low-energy cutoff. This is particularly important for $^3\text{He}$ because of the high cutoff.

The proton and deuteron production cross sections are compared with previous data at lower energies [40] in Figs. 9 and 10. There seems to be general agreement between the trend of the previous data and the present data point. The curves in these figures are based on a GNASH calculation [23].

7 Conclusions and outlook

In the present paper, we report an experimental data set on light-ion production induced by 96 MeV neutrons on silicon. Experimental double-differential cross sections ($d^2\sigma/d\Omega dE$) are measured at eight angles between $20^\circ$ and $160^\circ$. Energy-differential ($d\sigma/dE$) and production cross sections are obtained for the five types of outgoing particles. Theoretical calculations based on nuclear reaction codes including direct, pre-equilibrium and statistical calculations give generally a good account of the magnitude of the experimental cross sections. For proton emission, the shape of the spectra for the double-differential and energy-differential cross sections are well described. The calculated and the experimental alpha-particle spectra are also in fair agreement with the exception of the high energy part, where the theory predicts higher yields than experimentally observed. For the other complex ejectiles (deuteron, triton and $^3\text{He}$) there are important differences between theory and experiment in what concerns the shape of the spectra.

For the further development of the field, data at even higher energies are requested. The results suggest that the MEDLEY facility, which was used for the present work, should be upgraded to work also at 180 MeV, i.e., the maximum energy of the TSL neutron beam facility. At present, a new neutron beam facility is under commissioning at TSL, covering the same energy range, but with a projected intensity increase of a factor five. This will facilitate measurements at higher energies than in the present work.

The setup described in this paper comprises an active target, the information of which was not used in the analysis here but can provide valuable information on the kinetic energy transferred to the residual nucleus. This information might be crucial for soft-error studies and therefore it is of interest to compare this measurement with theoretical calculations. Work along this line is in progress.
Acknowledgments

This work was supported by the Swedish Natural Science Research Council, the Swedish Nuclear Fuel and Waste Management Company, the Swedish Nuclear Power Inspectorate, Ringhals AB, and the Swedish Defence Research Agency. The authors wish to thank the The Svedberg Laboratory for excellent support. One of the authors (U.T.) wishes to express his gratitude to the Thai Ministry of University Affairs and to the International Program in the Physical Sciences at Uppsala University. One of the authors (Y.W.) is grateful to the scientific exchange program between the Japan Society for the Promotion of Science and the Royal Swedish Academy of Sciences.

References


Figure captions

1. (a) Particle identification spectra at 20° for the $\Delta E_1 - \Delta E_2$ (a) and $\Delta E_2 - E$ (b) detector combinations. The solid lines represent tabulated energy loss values in silicon [14]. The insert in (b) illustrates the separation of high-energy protons, deuterons and tritons discussed in sect. 3.1.

2. (a) Neutron TOF spectrum versus deuteron energy for the Si(n,dx) reaction at 20° and the selection of deuterons associated with the full-energy neutron peak. The neutron-energy scale is given to the right. The solid line is a kinematic calculation of the ground-state deuteron energy as a function of the neutron energy. The lower rectangular cut is associated with neutrons in the full-energy peak, whereas the adjacent rectangular cut is used when correcting for the observed timing shift discussed in sect. 4.3. (b) Deuteron energy spectrum at 20° with (solid histogram) and without (dashed histogram) the full-energy neutron cut. The cross-hatched histogram shows the target-out background. The bump below 20 MeV in the solid histogram is due to wrap-around effects discussed in sect. 4.3.

3. Experimental double-differential cross sections (filled circles) of the Si(n,px) reaction at 96 MeV at four laboratory angles. The curves indicate theoretical calculations based on GNASH [23] (dashed), TALYS-present work (dotted) and GNASH-present work (solid). The TALYS result is in the c.m. system and the GNASH results are in the lab system.

4. Experimental double-differential cross sections (filled circles) of the Si(n,dx) reaction at 96 MeV at four laboratory angles. The curves indicate theoretical calculations based on GNASH [23] (dashed), TALYS-present work (dotted) and GNASH-present work (solid). The TALYS result is in the c.m. system and the GNASH results are in the lab system.

5. Experimental double-differential cross sections (filled circles) of the Si(n,tx) reaction at 96 MeV at four laboratory angles. The curves indicate theoretical calculations based on TALYS-present work (dotted) and GNASH-present work (solid). The TALYS result is in the c.m. system and the GNASH result is in the lab system.

6. Experimental double-differential cross sections (filled circles) of the Si(n,${}_3^3$Hex) reaction at 96 MeV at four laboratory angles. The curves
indicate theoretical calculations based on TALYS-present work (dotted) and GNASH-present work (solid). The TALYS result is in the c.m. system and the GNASH result is in the lab system.

7. Experimental double-differential cross sections (filled circles) of the Si(n,αx) reaction at 96 MeV at four laboratory angles. The curves indicate theoretical calculations based on GNASH [23] (dashed), TALYS-present work (dotted) and GNASH-present work (solid). The TALYS result is in the c.m. system and the GNASH results are in the lab system. Note the logarithmic scale.

8. Experimental energy-differential cross sections (filled circles) for neutron-induced p, d, t, 3He and α production at 96 MeV. The curves indicate theoretical calculations based on GNASH [23] (dashed), TALYS-present work (dotted) and GNASH-present work (solid). The TALYS result is in the c.m. system and the GNASH results are in the lab system.

9. Neutron-induced proton production cross section as a function of neutron energy. The full circle is from the present work, whereas the open circles are from previous work [2]. The curve is based on a GNASH calculation [23]. The data as well as the calculations correspond to a cutoff energy of 4 MeV. Note that the cutoff energy is different from that in Table 1.

10. Same as Fig. 9 for deuteron production, with a cutoff energy of 8 MeV.
Table 1: Experimental production cross sections for protons, deuterons, tritons, $^3$He and alpha particles from the present work. Theoretical values resulting from GNASH and TALYS calculations are given as well. The experimental data in the second column have been obtained with cutoff energies of 2.5, 3.0, 3.5, 8.0 and 4.0 MeV for p, d, t, $^3$He and alpha particles, respectively. The third column shows data corrected for these cutoffs, using the GNASH calculation of the present work.

<table>
<thead>
<tr>
<th>$\sigma_{prod}$</th>
<th>Experiment (mb)</th>
<th>Experiment (cutoff corr.)</th>
<th>GNASH (Ref. [23])</th>
<th>GNASH (present)</th>
<th>TALYS (present)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(n,px)</td>
<td>436±22</td>
<td>507</td>
<td>670.3</td>
<td>701.9</td>
<td>558.3</td>
</tr>
<tr>
<td>(n,dx)</td>
<td>81±4</td>
<td>89.5</td>
<td>77.0</td>
<td>109.6</td>
<td>107.6</td>
</tr>
<tr>
<td>(n,tx)</td>
<td>15.2±0.8</td>
<td>17.9</td>
<td>–</td>
<td>15.0</td>
<td>13.1</td>
</tr>
<tr>
<td>(n,$^3$Hex)</td>
<td>7.8±0.5</td>
<td>13.0</td>
<td>–</td>
<td>10.6</td>
<td>14.5</td>
</tr>
<tr>
<td>(n,αx)</td>
<td>144±7</td>
<td>183</td>
<td>175.8</td>
<td>202.4</td>
<td>146.8</td>
</tr>
</tbody>
</table>
\( \Delta E_2 \) [MeV] vs. \( E \) [MeV]

- \( \alpha \)
- \(^3\)He
- \( p \)
- \( d \)
- \( t \)
Background

Deuteron Energy [MeV]

Yield [counts]
\[ \frac{d^2\sigma}{d\Omega dE} \text{ [mb/(sr MeV)]} \]

Proton Energy [MeV]

\( \theta = 20^\circ \)
\( \theta = 40^\circ \)
\( \theta = 100^\circ \)
\( \theta = 140^\circ \)
$\frac{d^2\sigma}{d\Omega dE} = \text{mb/(sr}\cdot\text{MeV})$
\[
d\sigma/d\Omega dE \quad \text{[mb/(sr \cdot MeV)]}
\]

- \( \theta = 20^\circ \)
- \( \theta = 40^\circ \)
- \( \theta = 100^\circ \)
- \( \theta = 140^\circ \)

Triton Energy [MeV]
$d^2\sigma/d\Omega dE$ [mb/(sr⋅MeV)]

$\theta = 20^\circ$

$\theta = 40^\circ$

$\theta = 100^\circ$

$\theta = 140^\circ$
\[ \frac{d^2 \sigma}{d \Omega dE} \text{ [mb/(sr \cdot MeV)]} \]

\[ \theta = 20^\circ, 40^\circ, 100^\circ, 140^\circ \]

\[ \alpha \text{ Energy [MeV]} \]
$\sigma_{\text{prod}}(n,p_x)$ [mb] vs. Neutron Energy [MeV]
\[ \sigma_{\text{prod}}(n,dx) \text{ [mb]} \]

Neutron Energy [MeV]

\[ \sigma_{\text{prod}}(n,dx) \text{ [mb]} \]