Determination of molecular stopping cross section of $^{12}$C, $^{16}$O, $^{28}$Si, $^{35}$Cl, $^{58}$Ni, $^{79}$Br, and $^{127}$I in silicon nitride

N.P. Barradas a,*, A. Bergmaier b, K. Mizohata c, M. Msimanga d,e, J. Räisänen c, T. Sajavaara f, A. Simon g,h

a Centro de Ciências e Tecnologias Nucleares, Instituto Superior Técnico, Universidade de Lisboa, Estrada Nacional 10 ao km 139.7, 2695-066 Bobadela LRS, Portugal
b Institut für Angewandte Physik und Messtechnik, Fakultät für Luft und Raumfahrttechnik, Werner-Heisenberg-Weg 39, D-85577 Neubiberg, Germany
c Department of Physics, University of Helsinki, P.O. Box 43, FI-00014 University of Helsinki, Finland
diThemba LABS Gauteng, National Research Foundation, Private Bag 11, WITS 2050, Johannesburg, South Africa
e Department of Physics, Tshwane University of Technology, Private Bag X680, Pretoria 0001, South Africa
f Department of Physics, University of Jyväskylä, Surventie 9, 40014 Jyväskylä, Finland
g International Atomic Energy Agency, Division of Physical and Chemical Sciences, Vienna International Centre, P.O. Box 100, A-1400 Vienna, Austria
h Institute of Nuclear Research of the Hungarian Academy of Sciences, (ATOMKI), P.O. Box 51, H-4001 Debrecen, Hungary

ARTICLE INFO

Article history:
Received 30 March 2015
Received in revised form 30 July 2015
Accepted 18 August 2015
Available online 27 August 2015

Keywords:
Stopping power
Silicon nitride
Heavy ions
Ion beam analysis

ABSTRACT

Silicon nitride is a technologically important material in a range of applications due to a combination of important properties. Very good thermal shock resistance, high fracture toughness, temperature strength, creep resistance and oxidation resistance, together with a low thermal expansion coefficient and low density, make it a widely used material in a variety of products such as high performance bearings, cutting tools, and engine components [1,2]. On the other hand, silicon nitride is also used in the microelectronics industry as a high permittivity dielectric, as a passivation or protective barrier due to its low permeation to alkali and small ions, and as an etch mask in micromachining [3–5].

Ion beam analysis (IBA) techniques play an important role in developing and optimising systems that include silicon nitride [6]. In particular, heavy ion elastic recoil detection analysis (HI-ERDA) can be used to determine the stoichiometry of silicon nitride films, which often deviates from the ideal Si$_3$N$_4$, as well as the content of impurities such as hydrogen, even in the presence of other materials or in a matrix containing heavier elements. Accurate quantification of IBA results depends on the basic data used in the data analysis. Quantitative depth profiling relies on the knowledge of the stopping power cross sections of the materials studied for the ions involved, which in the case of HI-ERDA is both the primary beam, and the recoiled species. We measured the stopping cross section of $^{12}$C, $^{16}$O, $^{28}$Si, $^{35}$Cl, $^{58}$Ni, $^{79}$Br, and $^{127}$I in a well-characterised silicon nitride membrane. The measurements were made by independent groups utilising different experimental setups and methods. In some cases there is extensive overlap of the energy range in different experiments, allowing a comparison of the different results. The four independent data sets reported in this work are in excellent agreement with each other, in the cases where similar energy ranges were measured. On the other hand, the data are in most cases higher than calculations made with the interpolative schemes SRIM and MSTAR together with the Bragg rule. Better agreement is found with MSTAR in some of the cases studied. This work is a significant extension of the heavy ion stopping power data base for silicon nitride.

© 2015 Elsevier B.V. All rights reserved.

1. Introduction

Silicon nitride is a technologically important material in a range of applications due to a combination of important properties. Very good thermal shock resistance, high fracture toughness, temperature strength, creep resistance and oxidation resistance, together with a low thermal expansion coefficient and low density, make it a widely used material in a variety of products such as high performance bearings, cutting tools, and engine components [1,2]. On the other hand, silicon nitride is also used in the microelectronics industry as a high permittivity dielectric, as a passivation or protective barrier due to its low permeation to alkali and small ions, and as an etch mask in micromachining [3–5].
calculate the molecular stopping power based on the elemental values [12]. For heavy ions, the accuracy of this procedure is variable, with experimental values agreeing well with the SRIM prediction in some cases, and less well in other cases. MSTAR is another interpolative scheme [13], based on a different set of experimental data, and also geared mainly towards elemental stopping. MSTAR does not include data for all ions.

We have measured the stopping cross section of $^{12}$C, $^{16}$O, $^{28}$Si, $^{35}$Cl, $^{58}$Ni, $^{79}$Br, and $^{127}$I in a well-characterised silicon nitride membrane, in the context of a Coordinated Research Project of the International Atomic Energy Agency, where many other systems were measured [14–16]. The measurements were made by independent groups utilising different experimental setups and methods. In some cases there is extensive overlap of the energy range in different experiments, allowing a comparison of the different results.

2. Methodology

2.1. Experimental details

The stopping cross section measurements employed in all cases the direct transmission technique [17,18]. For particle detection, each participating laboratory used its own detection system. Helsinki employed a ToF telescope, consisting of two carbon-foil timing gates separated by a 684 nm flight length and an Ortec Ultra series ion implanted detector [19]. The sample foils were placed between the second timing gate and the energy detector.

Elemental bulk targets of Ge and Re were used to forward scatter the projectile ions. The ions are scattered from a range of depths in the targets, and therefore have a continuous range of energies, from a maximum slightly below the initial beam energy, down to zero [20]. Measurements are made with and without the silicon nitride membrane in position. The energy before the foil is determined with the ToF detector and the energy after the foil by the particle detector, leading to the determination of a continuous stopping power curve.

Jyväskylä used a ToF-E telescope at a 1.7 MV Pelletron, with good detection efficiency for H, higher than 90% for He, and higher than 99.5% for C. The timing resolution was 155 ps measured for He (FWHM). A gas ionisation detector was used for the energy measurement. The data acquisition was realised in list-mode, and a data stamp with an accuracy of 25 ns was given for each event. Coincident events are determined off-line. The primary beam was scattered from a 1 nm Au layer on Si substrate and this scattered beam either went through the silicon nitride membrane or it was scattered directly to the ToF-E telescope.

At iThemba LABS [20], a mass dispersive Time of Flight (ToF) spectrometer was used, consisting of two carbon foil based Microchannel Plate (MCP) timing detectors 0.6 m apart, and a passivated implanted planar silicon (PIPS) semiconductor energy detector at the end of the flight path just behind the second time detector. The ToF telescope sits at 30° to the incident beam direction. The ions whose energy loss is measured could either be incident projectile ions from the accelerator, scattered by a suitable heavy target element, or recoil ions ejected from the target by the incident beam. In either case the beam of ions incident on the stopper foil has a continuous range of energies. Measurements were done with and without the foil, positioned between the ToF and the PIPS detector [20]. In this case, the data was divided in energy bins and the stopping power was calculated for selected points.

Munich used the Q3D magnetic spectrometer at the Munich tandem accelerator [21]. The main features of the Q3D magnetic spectrometer are its large dispersion ($\Delta E/E \approx 2 \times 10^{-4}/\text{mm}$), the high intrinsic resolution ($\Delta E/E = 2 \times 10^{-3}$), the large solid angle of detection (up to 14.3 mrs) and, most importantly, the possibility to correct for the kinematical shift up to the fourth order, by means of a magnetic multipole element. Routinely, the kinematical shift is corrected up to the third order, leading to an overall energy resolution of $7 \times 10^{-4}$ even when a large solid angle of detection of 5 mrs is used. Without correction the kinematical shift would be larger than 6% energy spread at the usually used mean scattering angle of 15°. At the end the multipole element is adjusted in such a way that the recoil ions scattered from a certain depth are focused to a certain position of the focal plane of the Q3D where the ions are identified and their position is measured. Thin foil targets were mounted for the stopping measurements perpendicular to the incident beam. After passing the thin foils, the energy loss of the ion beam is analysed with the Q3D spectrograph at 0° scattering angle.

All taken together, experiments were made for $^{12}$C, $^{16}$O, $^{28}$Si, $^{35}$Cl, $^{58}$Ni, $^{79}$Br, and $^{127}$I. The energy range was different for different ions, but always in the range useful for IBA (see Table 1).

2.2. Silicon nitride membranes

A key issue in the experimental determination of stopping power is the availability of adequate targets. These must be very well characterised. In particular, in transmission or thin film experiments, their thickness and areal density must be well known, as it directly influences the results obtained. Surface roughness, the presence of impurities, and the exact stoichiometry in multielemental targets, are also important parameters.

Commercially available silicon nitride membranes were acquired [22], with thicknesses of 30 and 100 nm. The manufacturer information stated that real thickness values can deviate up to 10%, with a batch-to-batch variation up of 3–4%. However, within a batch, adjacent wafers are very similar in thickness, and the variation across a single wafer is better than 1%; membranes for a single order are supplied from a single batch. Across a single membrane, the thickness variation will be much better than 1%. Finally, the membrane roughness should be low, around 5 Å. These parameters are ideally suited for energy loss measurements and to be used as substrates for deposition of thin films of other materials, provided that the actual areal density of the membranes used is measured.

The composition of the membrane was determined with an HI-ERDA experiment made at the Munich Q3D magnetic spectrograph [21] using a 150 MeV $^{12}$C beam, with a AE-E detection system at a 38° scattering angle. The results are given in Table 2, where the statistical uncertainties are also given. However, there are other sources of uncertainty, which need to be taken into account, and we elaborated an uncertainty budget, summarised in Table 3.

<table>
<thead>
<tr>
<th>Ion</th>
<th>Energy range (MeV)</th>
<th>Institute</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{12}$C</td>
<td>1.89–5.05</td>
<td>iThemba</td>
</tr>
<tr>
<td>$^{16}$O</td>
<td>2.11–6.90</td>
<td>iThemba</td>
</tr>
<tr>
<td>$^{28}$Si</td>
<td>3.56–8.71</td>
<td>iThemba</td>
</tr>
<tr>
<td>$^{35}$Cl</td>
<td>34.9</td>
<td>Munich</td>
</tr>
<tr>
<td>$^{58}$Ni</td>
<td>59.7</td>
<td>Munich</td>
</tr>
<tr>
<td>$^{79}$Br</td>
<td>39.7</td>
<td>Munich</td>
</tr>
<tr>
<td>$^{12}$C</td>
<td>0.56–9.92</td>
<td>Helsinki</td>
</tr>
<tr>
<td>$^{16}$O</td>
<td>0.31–7.88</td>
<td>Helsinki</td>
</tr>
<tr>
<td>$^{35}$Cl</td>
<td>0.53–19.72</td>
<td>Helsinki</td>
</tr>
<tr>
<td>$^{79}$Br</td>
<td>2.99–42.49</td>
<td>Helsinki</td>
</tr>
<tr>
<td>$^{127}$I</td>
<td>4.12–77.1</td>
<td>Helsinki</td>
</tr>
<tr>
<td>$^{12}$C</td>
<td>0.20–8.16</td>
<td>Jyväskylä</td>
</tr>
<tr>
<td>$^{16}$O</td>
<td>0.26–8.08</td>
<td>Jyväskylä</td>
</tr>
<tr>
<td>$^{35}$Cl</td>
<td>0.59–10.89</td>
<td>Jyväskylä</td>
</tr>
</tbody>
</table>
We used the analytical code NDF [27] and the Monte Carlo code mined by measuring the energy loss of 60 MeV $^{58}\text{Ni}$ in transmission geometry at normal incidence. The same carbon foil is then measured with ERDA in the same conditions as the sample of interest, and the scattering angle and solid angle do not contribute to the uncertainty. The measurement of the beam fluence does contribute, both when the carbon foil is measured with ERDA, and when the actual sample is measured. The beam current is measured with a rotating wire loop located in the beam line 2 m in front of the scattering chamber. A pico-Ampere metre transforms the measured current to an analogue voltage output. This voltage enters twice in the uncertainty budget, one for the silicon nitride measurement. The 0.8% uncertainty in the areal density of a reference carbon foil can be determined at Munich, for 60 MeV $^{58}\text{Ni}$ ions on 100 nm Si$_3$N$_4$. The uncertainty budget for the silicon nitride membrane nominally 30 nm thick is almost identical. The only contributing term that is different is the counting statistics, which is 4.5%. This leads to a precision of 4.61%, an accuracy of 4.83%, and an accuracy for stopping calculations including the layer thickness inhomogeneity of 4.9%. As for the 100 nm membrane, the counting statistics dominates the total uncertainty.

In this work, the nominally 100 nm thick membrane was used for the stopping power determination.

### 2.3. Calculation of uncertainties

In this work we compare the results obtained independently by different groups, each with its own method and set of experimental conditions. In order for the comparison to be valid, the uncertainty of each measurement must be carefully evaluated. The Guide to the Expression of Uncertainty in Measurement [29], known as GUM, is a document prepared by the Joint Committee for Guides in Metrology of the Bureau International des Poids et Mesures, that sets the standard for evaluation of uncertainties. Sjöland et al. [30] developed the concepts and methods described in GUM for the particular details of IBA, leading to the construction of an uncertainty budget. All uncertainties in this work follow Sjöland et al., and are given with as one standard deviation.

For each experiment, two uncertainty budgets were made. The first one is to evaluate the uncertainty of the energy at which each stopping power value is determined. We give one example in Table 4 for the determination of the energy at which the stopping is determined at Munich, for 60 MeV Ni ions on 100 nm Si$_3$N$_4$. The first term is the systematic uncertainty in the energy calibration of the magnet, which includes a possible energy drift from experiment to experiment. Without detailed knowledge, a conservative 5 keV value was taken. The beam and energy spread is $4 \times 10^{-4}$ at FWHM, and for 60 MeV the standard deviation is 10 keV. The energy of the transmitted beam is also determined with a given uncertainty, from the mean or mode (most probable value) of the beam energy distribution. This can usually be determined with a much higher accuracy than its width [31], which was 54.5 keV.

### Table 4

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beam and system energy spread $\Delta E/E = 4 \times 10^{-4}$ (FWHM)</td>
<td>10 keV</td>
</tr>
<tr>
<td>Uncertainty in estimation of transmitted beam</td>
<td>5 keV</td>
</tr>
<tr>
<td>Total energy loss in the foil (divided by 2.35 to obtain sigma)</td>
<td>402 keV</td>
</tr>
<tr>
<td>Standard uncertainty (precision)</td>
<td>402 keV</td>
</tr>
<tr>
<td>Beam energy - uncertainty in calibration of magnet</td>
<td>5 keV</td>
</tr>
<tr>
<td>Total combined standard uncertainty (accuracy)</td>
<td>402 keV</td>
</tr>
</tbody>
</table>

The Q3D spectograph ion beam energy spread and detector resolution combine to a FWHM of $\Delta E/E = 4 \times 10^{-4}$. The scattering cross section depends on the energy with $1/E^2$, leading to an uncertainty in the areal density of 0.08%.

Normalisation of the scattering angle, solid angle of the detector, and beam fluence was made relative to a known target. The Munich group has previously determined the stopping power for 60 MeV $^{58}\text{Ni}$ in carbon foils with an accuracy of 0.8% [23]. Therefore, the areal density of a reference carbon foil can be determined by measuring the energy loss of 60 MeV $^{58}\text{Ni}$ in transmission geometry at normal incidence. The same carbon foil is then measured with ERDA in the same conditions as the sample of interest, and the scattering angle and solid angle do not contribute to the uncertainty. The measurement of the beam fluence does contribute, both when the carbon foil is measured with ERDA, and when the actual sample is measured. The beam current is measured with a rotating wire loop located in the beam line 2 m in front of the scattering chamber. A pico-Ampere metre transforms the measured current to an analogue voltage output. This voltage is the input for a separate ADC in the data acquisition, which is triggered with a 50 Hz pulse. The beam current is written together with the other ADC data to disk in list mode. The stability and accuracy of this measurement was around 0.5%. This value enters twice in the uncertainty budget, one for the carbon measurement and one for the silicon nitride measurement. The 0.8% uncertainty in the stopping power of 60 MeV $^{58}\text{Ni}$ in carbon, and a 0.04% contribution from the uncertainty in the beam energy when measuring the carbon reference material with ERDA are also included.

The total counting statistics uncertainty, for the sum of all the elements present, is 2.48%.

Plowup can significantly change the measured yield for high beam currents and high count rates. In this experiment, the beam current was kept at very low values, with dead time nearly 0%.

We include a Type B uncertainty of 0.83% due to the data analysis. The value quoted was derived from the IAEA Intercomparison of IBA softwares [24], derived for $^4\text{He}$-ERDA.

The screening model of Andersen et al. [25] Rutherford cross section – screening model 0.87% for the stopping power determination. We give one example in Table 4 for the determination of the energy at which the stopping is determined at Munich, for 60 MeV Ni ions on 100 nm Si$_3$N$_4$. The uncertainty budget for the silicon nitride membrane nominally 30 nm thick is almost identical. The only contributing term that is different is the counting statistics, which is 4.5%. This leads to a precision of 4.61%, an accuracy of 4.83%, and an accuracy for stopping calculations including the layer thickness inhomogeneity of 4.9%. As for the 100 nm membrane, the counting statistics dominates the total uncertainty.

In this work, the nominally 100 nm thick membrane was used for the stopping power determination.

### Table 2

Elemental composition of the silicon nitride membranes.

<table>
<thead>
<tr>
<th>Layer</th>
<th>Si</th>
<th>N</th>
<th>H</th>
<th>C</th>
<th>O</th>
</tr>
</thead>
<tbody>
<tr>
<td>100 nm Si$_3$N$_4$</td>
<td>3.021E+17</td>
<td>3.597E+17</td>
<td>1.33E+16</td>
<td>1.596E+15</td>
<td>8.377E+15</td>
</tr>
<tr>
<td>30 nm Si$_3$N$_4$</td>
<td>6.536E+15</td>
<td>7.873E+15</td>
<td>8.365E+14</td>
<td>5.321E+14</td>
<td>1.185E+15</td>
</tr>
</tbody>
</table>

### Table 3

Uncertainty budget for the determination of the 100 nm silicon nitride areal density.

<table>
<thead>
<tr>
<th>Uncertainty Source</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beam energy spread $\Delta E/E = 4 \times 10^{-4}$ (FWHM)</td>
<td>0.08%</td>
</tr>
<tr>
<td>Counting statistics (total, added for all elements present)</td>
<td>2.48%</td>
</tr>
<tr>
<td>Accuracy of current measurement in measurement of sample</td>
<td>0.5%</td>
</tr>
<tr>
<td>Uncertainty in N$_t$ of reference material due to beam energy spread</td>
<td>0.04%</td>
</tr>
<tr>
<td>Accuracy of current measurement in measurement of C reference foil</td>
<td>0.5%</td>
</tr>
<tr>
<td>Pileup uncertainty</td>
<td>0%</td>
</tr>
<tr>
<td>Uncertainty in estimation of energy of transmitted beam</td>
<td>5 keV</td>
</tr>
<tr>
<td>Accuracy of current measurement in measurement of C reference foil</td>
<td>0.8%</td>
</tr>
<tr>
<td>Total combined standard uncertainty (accuracy)</td>
<td>2.96%</td>
</tr>
<tr>
<td>Standard uncertainty (precision)</td>
<td>2.58%</td>
</tr>
<tr>
<td>Scattering angle: relative measurement</td>
<td>0%</td>
</tr>
<tr>
<td>Stopping power of $^{58}\text{Ni}$ in C, used to determine reference material C</td>
<td>0.8%</td>
</tr>
<tr>
<td>Ne code uncertainty</td>
<td>0.83%</td>
</tr>
<tr>
<td>Rutherford cross section – screening model</td>
<td>0.87%</td>
</tr>
<tr>
<td>Layer thickness inhomogeneity (from manufacturer)</td>
<td>1%</td>
</tr>
<tr>
<td>Total combined standard uncertainty for stopping (accuracy)</td>
<td>3.1%</td>
</tr>
</tbody>
</table>

Cortesio [28] to calculate the screened cross section for different screening options, for the conditions of this experiment. The extra uncertainty arising from the screening model is 0.87%.

Finally, when determining stopping from measurements with different silicon nitride membranes, a 1% extra uncertainty due to the layer thickness inhomogeneity must be added as well. All in all, in this experiment it is the counting statistics that dominates the final total uncertainty.

The uncertainty budget for the silicon nitride membrane nominally 30 nm thick is almost identical. The only contributing term that is different is the counting statistics, which is 4.5%. This leads to a precision of 4.61%, an accuracy of 4.83%, and an accuracy for stopping calculations including the layer thickness inhomogeneity of 4.9%. As for the 100 nm membrane, the counting statistics dominates the total uncertainty.

In this work, the nominally 100 nm thick membrane was used for the stopping power determination.
For asymmetric distributions the mean and the mode do not coincide, and there is some ambiguity about which value should be taken, but in this case the energy distribution of the transmitted beam was nearly Gaussian-shaped. The fit of a Gaussian to the energy distribution yielded an uncertainty for the mean 5 keV or smaller, and we adopted this value. The largest contribution to the beam energy uncertainty is the actual energy loss in the Si₃N₄ membrane, which was 943.7 keV: the initial beam energy was 60 MeV, and the final beam energy was 59.0563 MeV. The stopping power determined is an average value over this energy range. We take the average energy in the membrane, 59.5281 MeV, as the energy for which the stopping power is determined, and the 943.7 keV energy loss is the FWHM of this energy. The corresponding standard deviation is 402 keV.

The uncertainty in the actual stopping power values is determined with a separate uncertainty budget, given in Table 5 for the same case. The terms to be considered are the beam and system energy spread, and the uncertainty in the estimation of the energy of the transmitted beam, relative to the actual energy loss. The final total uncertainty is obtained by also including the uncertainty in the areal density of the Si₃N₄ membrane, which was calculated in Section 2.2.

### 3. Results and discussion

Sun et al. [32] measured previously silicon nitride stopping power for different ions, including ¹²C and ¹⁶O. However, the sample they used had a stated composition Si₃N₃.1H₀.0₆, quite far from stoichiometric. The membrane used in our experiments was nominally stoichiometric, i.e. Si₃N₄, but the actual composition as measured with heavy ion ERDA was Si₃N₃.5H₀.₁₃C₀.₀₂O₀.₀₈. To ascertain the effect of the composition differences, we used SRIM and MSTAR to calculate the molecular stopping power for the different stoichiometries, based on the Si and N elemental stopping powers together with the Bragg rule. Nuclear stopping power was calculated with SRIM, and included in both SRIM and MSTAR calculations.

The SRIM calculations for the three different silicon nitride compositions are shown in Figs. 1 and 2 for ¹²C and ¹⁶O, respectively, which are the cases measured by Sun et al. The MSTAR calculation for the stoichiometric silicon nitride is also shown. The stopping powers for stoichiometric silicon nitride and for the composition determined at Munich are very similar for both ions, with a difference at the Bragg peak less than 0.7%.

The difference at the Bragg peak for stoichiometric silicon nitride and the Sun et al. sample is between 2.2% and 2.3% for the two ions. This difference is smaller than the stated accuracy of most stopping power measurements published, including all the new results reported by us in this work. For the stoichiometric sample, the difference at the stopping power maximum as calculated with SRIM and MSTAR is 4.6%. The energy at which the maximum is found is also different in the two calculations. This means that the program used to calculate the stopping power leads to a larger difference in the results than the actual sample stoichiometry.

Therefore, the stoichiometry taken for the SRIM and MSTAR calculation is not a determining factor. Nevertheless, for ¹²C and ¹⁶O SRIM results for stoichiometric silicon nitride and for the Sun et al. sample will be shown together with the experimental results.

The Helsinki results will be shown as two lines that represent the 1σ (one standard deviation) confidence limits of the stopping power determined. The actual stopping power is the average of the two curves shown.

The ¹²C stopping cross section is shown in Fig. 3, together with the Sun et al. data [32] and with the SRIM and MSTAR stopping power curves. The nuclear stopping was included in the calculations. At the lowest energy measured, around 0.02 MeV/nucleon, it is 6% of the total calculated value, and it decreases quickly for higher energies.

The Sun et al. data agrees well with the SRIM and MSTAR predictions, except at the highest energy reported, where it is considerably higher. On the other hand, the three new data sets agree with each other but not with the calculations: they are higher than the entire energy range measured, with a deviation that is outside their uncertainty.

The ¹⁶O stopping cross section is shown in Fig. 4. Nuclear stopping is 10% of the contribution of electronic stopping for the lowest energy measured, 0.016 MeV/nucleon. This decreases rapidly for

### Table 5

Uncertainty budget for the stopping power of 60 MeV Ni ions on 100 nm Si₃N₄ as determined at Munich.

<table>
<thead>
<tr>
<th>Uncertainty</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total energy loss in foil</td>
<td>943.7 keV</td>
</tr>
<tr>
<td>Beam and system energy spread</td>
<td>10 keV</td>
</tr>
<tr>
<td>Uncertainty in estimation of energy of transmitted beam</td>
<td>5 keV</td>
</tr>
<tr>
<td>Total energy uncertainty as percentage of energy loss in foil</td>
<td>1.2%</td>
</tr>
<tr>
<td>Standard uncertainty (precision)</td>
<td>1.2%</td>
</tr>
<tr>
<td>Uncertainty in areal density of foil</td>
<td>3.1%</td>
</tr>
<tr>
<td>Total combined standard uncertainty (accuracy)</td>
<td>3.3%</td>
</tr>
</tbody>
</table>

Fig. 1. Stopping cross section of silicon nitride for ¹²C, calculated with different models. The "Munich" and "Sun" curves are for Si₃N₃.5H₀.₁₃C₀.₀₂O₀.₀₈ and Si₃N₃.1H₀.₀₆, respectively.

Fig. 2. Stopping cross section of silicon nitride for ¹⁶O, calculated with different models. The "Munich" and "Sun" curves are for Si₃N₃.5H₀.₁₃C₀.₀₂O₀.₀₈ and Si₃N₃.1H₀.₀₆, respectively.
higher energy values, and at 0.1 MeV/nucleon nuclear stopping is only 1\% of the total.

The same observations as made for \(^{12}\text{C}\) are seen for \(^{16}\text{O}\): the Sun et al. data agree with the SRIM and MSTAR calculations, except for the highest energy point measured, while the three new data sets all agree with each other, and they are consistently above the calculation. However, in the low energy range, MSTAR comes closer to the data than SRIM. We conclude that, for the two ions \(^{12}\text{C}\) and \(^{16}\text{O}\), the SRIM interpolation, based on the Bragg rule, leads to calculated stopping powers lower than the experimental values, which also partially holds true for MSTAR.

The \(^{28}\text{Si}\) stopping cross section is shown in Fig. 5. We were unable to find previous experimental data in the literature. The data now presented are consistently above SRIM, but the difference is within two standard deviations for most data points. On the other hand, the data are in very good agreement with the MSTAR calculation.

The \(^{35}\text{Cl}\) stopping cross section is shown in Fig. 6. No previous data could be found in the literature. The contribution of nuclear stopping in this case is 27\%, 2.4\%, and 0.56\% at 0.017 MeV/nucleon (lowest energy measured), 0.1 MeV/nucleon, and 0.56 MeV/nucleon (highest energy measured). The two points for Munich were measured using the 30 nm and 100 nm Si\(_3\)N\(_4\) membranes. The three data sets reported are all above the curves calculated with both SRIM and MSTAR from the elemental stopping values

The Bragg rule. The difference is often larger than two standard deviations, and therefore it can be concluded that the Bragg rule is not accurate in this case.

The \(^{58}\text{Ni}\) stopping cross section is shown in Fig. 7. For the energies measured, the nuclear stopping power contribution is small, around 0.3\%. To our knowledge, this is the first experimental data

\begin{figure}[h]
\centering
\includegraphics[width=0.45\textwidth]{12C.pdf}
\caption{Stopping cross section of silicon nitride for \(^{12}\text{C}\). Sun is the data from Ref. [32], measured for \(^{13}\text{C}\) ions.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.45\textwidth]{28Si.pdf}
\caption{Stopping cross section of silicon nitride for \(^{28}\text{Si}\).}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.45\textwidth]{35Cl.pdf}
\caption{Stopping cross section of silicon nitride for \(^{35}\text{Cl}\).}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.45\textwidth]{58Ni.pdf}
\caption{Stopping cross sections of silicon nitride for \(^{58}\text{Ni}\).}
\end{figure}
presented for this system. The SRIM calculation underestimates the experimental values, with a difference three to four times larger than the uncertainty of the experiment, and we conclude that SRIM with the Bragg rule does not reproduce the data within the accuracy of the experiment.

The $^{79}$Br stopping cross section is shown in Fig. 8. The contribution of nuclear stopping relative to that of electronic stopping goes from 24% at the lowest energy (0.04 MeV/nucleon) to 0.6% at the highest energy (0.54 MeV/nucleon). The two experimental data sets, from Helsinki and Munich, agree well with each other, and show that the SRIM calculation with the Bragg rule underestimates the experimental values.

The $^{127}$I stopping cross section is shown in Fig. 9. Nuclear stopping power is very large for this system, it. At the lowest energy probed (0.02 MeV/nucleon) it is 63% of the value calculated with SRIM for electronic stopping. At the highest energy (0.29 MeV/nucleon), it is still 2.5%. We were unable to find previous measurements in the literature, and to the best of our knowledge this is the first data reported for this system. The difference between the experimental data and the SRIM calculation is extremely large, by a factor of two at some energy values. Such large deviations are not expected. We also note that the shape of the measured and calculated stopping cross section curves are very different, while for lighter ions where deviations were also observed, the shape of the measured and calculated stopping curves was similar. On the other hand, large deviations are more likely to exist in materials such as this one, made of two elements with extremely different atomic numbers, forming an insulator. Furthermore, taking as a whole the results now reported for all the ions, a trend seems to appear, where the deviation between the data and the SRIM calculation increases as the atomic number of the projectile increases. In any case, further data should be collected for $^{127}$I, to confirm the present results.

All in all, considering that for the other ions used in this work the experimental stopping cross section values are consistently above the SRIM values, there is clearly scope for an improvement of the interpolative calculations made by future versions of SRIM for the molecular stopping cross section of silicon nitride.

The stopping power of heavy ions in matter depends on the charge state of the ions. SRIM calculations are made for the equilibrium charge state, the beam ions may have a different charge state distribution. In particular, the primary beam ions usually have a single well defined charge state. However, the experiments made at iThemba, Helsinki and Jyväskylä have used scattered ions or recoils, and thus the ions on leaving the scatter target no longer have the initial charge state, but instead a charge distribution that approaches the equilibrium within the scatter target. The Helsinki and iThemba experiments have also the timing carbon foils located before the membranes, and one would expect that they reach the equilibrium charge state distribution in carbon when the reach the Si$_3$N$_4$ membranes. Only in Munich the ions enter the Si$_3$N$_4$ with non-equilibrium charge state.

The equilibrium charge state depends on the atomic number of the target material. For the experiments with timing carbon foils, the atomic number of C is not far from the Si$_3$N$_4$ membranes, and the beam should arrive to the Si$_3$N$_4$ membrane with a charge state distribution close to equilibrium. For the velocities used in this work, the equilibrium charge state in Au can be around 10–20 lower than in Si$_3$N$_4$ [33], which affects the stopping power while the equilibrium charge state in the Si$_3$N$_4$ membranes is not reached. However, the thickness necessary to achieve equilibrium is usually a few nm for the velocities under consideration; for 1 MeV/amu Ni ions in C, it is around 6 nm [34], and it decreases for lower velocities [34]. This is much smaller than the 100 nm thickness of the Si$_3$N$_4$ membranes used in almost all the experiments reported here. Therefore, the effect on the results should not be significant.

4. Summary

We determined the stopping cross section of $^{12}$C, $^{16}$O, $^{28}$Si, $^{35}$Cl, $^{58}$Ni, $^{79}$Br, and $^{127}$I using a well-characterised silicon nitride membrane. The measurements were made using different experimental setups and methods by independent groups. In some cases the energy range covered by different experiments for the same ion overlaps extensively, allowing a comparison of the different results.

We compared the results with calculations made with SRIM, and where possible, also with MSTAR, using the Bragg rule to calculate the molecular stopping cross section from the elemental values. It should be noted that SRIM and MSTAR calculations for this system do not agree very well with each other, and hence the comparison with both calculations is useful. For the $^{12}$C and $^{16}$O ions, the new results were also compared with previous experimental data [32].

In the experiments where similar energy ranges were measured, the independent data sets reported in this work are in excellent agreement with each other. On the other hand, the data are in often higher than the SRIM and MSTAR calculations. Better agreement is found with MSTAR in some of the cases studied.

This work is a significant extension of the heavy ion stopping power data base for silicon nitride.
Acknowledgements

We would like to thank Dr. Jim Ziegler and Prof. Helmut Paul for many useful discussions. This work was partially funded by the International Atomic Energy Agency under the Coordinated Research Project F11013 and by Fundação para a Ciência e Tecnologia under grant UID/Multi/04349/2013.

References