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The stopping power and energy straggling of heavy ions in silicon nitride and polypropylene

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ABSTRACT

The stopping power and energy straggling of ${}^{12}C^{3+}$ and ${}^{16}O^{3+}$ ions with energies between 4.5 and 7.8 MeV in a 0.166-µm-thin silicon nitride and in 4-µm-thin polypropylene foils were measured by means of an indirect transmission method using a half-covered PIPS detector. Ions scattered from a thin gold layer under a scattering angle of 150° were used. The energy spectra of back-scattered and decelerated ions were registered and evaluated simultaneously. The measured stopping powers were compared with the theoretical predictions simulated by SRIM-2008 and MSTAR codes. SRIM prediction of energy stopping is reasonably close to the experimentally obtained values comparing to MSTAR values. Better agreement between experimental and predicted data was observed for C^{3+} ion energy losses comparing to O^{3+} ions. The experimental data from Paul's database and our previous experimental data were also discussed. The obtained experimental energy-straggling data were compared to those calculated by using Bohr's, Yang's models etc. The predictions by Yang are in good agreement with our experiment within a frame of uncertainty of 25%.

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BEAM INTERACTIONS WITH MATERIALS AND ATOMS

1. Introduction

The stopping power and energy straggling of energetic ions in matter have been studied both theoretically and experimentally; various combinations of ions and matter can be seen for example in [1–10]. The knowledge of the ion stopping power in matter is important to many applications dependent on the transport of ions in matter such as elemental depth profiling in polymers and ion beam analytical methods application [11–14], dosimetry and the application of metal composites prepared by ion implantation [15,16]. Significant interest in the stopping power of polymers arises from their similarity to human tissue, which is applicable in radiation safety and radiology. Silicon nitride windows have found application as vacuum exit windows for ion beams and have led to a considerable improvement of gas-detector energy resolution for low-energy ions because of their very good mechanical properties and chemical stability at high temperatures [17,18].

Experimental data on the stopping power and energy straggling of ions in different materials are essential for checking the reliability and validity of existing theoretical and semi-empirical models,

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http://dx.doi.org/10.1016/j.nimb.2014.11.014 0168-583X/© 2014 Elsevier B.V. All rights reserved. describing the passage of ions through matter, and for their improvement. There are several theoretical and semi-empirical models providing stopping-power estimates for different combinations of ions and materials. The MSTAR program [19] uses as input data the alpha-particle stopping powers of various materials and compounds from the ICRU Report 49 [20]. Bragg's formula [21], is used for compounds when input data are missing. The program subsequently performs the fitting of these input data to obtain the stopping powers of heavy ions in diverse materials [22,23]. In the computer code SRIM-2008, electronic stopping powers are deduced from proton-particle stopping powers. For a compound this code takes into account a small Core-and-Bond correction [24] consist of the superposition of the stopping by atomic cores and the stopping corresponding to the bonding electrons.

Simple theoretical models for energy straggling are applicable for fully stripped ions and thin targets only. Bohr's theory [25] is based on the assumption that the energy loss of the incident ions in the target material is very small as compared to incident-ion energy. The Bethe–Livingston theory [26] of energy-straggling estimation assumes that the velocity of the incident ion remains essentially the same during its passage through matter. For partially stripped ions, a charge-exchange straggling component also comes into picture due to fluctuations in the charge-state of the ion during its passage through the target foil [27]. Yang [28]

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developed a semi-empirical approach, which also includes the contribution arising from charge-exchange straggling. This approach is based on Chu's theory [29], which takes into account deviations from Bohr straggling caused by electron binding in the target atoms and additionally includes charge-state fluctuations of the ions.

In the present work, the stopping powers and energy straggling of C and O ions in polypropylene (PP, C_3H_6) and silicon nitride (Si₃N₄) were systematically investigated in the reference energy range of 4.5–7.8 MeV, as the energy stopping power data are still missing and the testing of semi-empirical models is highly desirable.

2. Experiment and data evaluation

2.1. The samples and foil-thickness measurement

The present experiment was carried out with the PP foil purchased from Goodfellow [30], with a thickness of 4.0 μ m, declared by the manufacturer, and the silicon nitride window from Norcada [31], with a declared thickness of 0.2 µm. The thickness of the polypropylene absorber foil was verified by using a microbalance: a square sample of $1\times 1\ \text{cm}^2$ in area was cut from the PP foil and its area was measured by a digital microscope. Then the sample was weighed several times by means of a Mettler Toledo Micro-Balance with a $\pm 1 \mu g$ absolute accuracy. The mean areal mass density of the foil was $0.36 \pm 0.02 \text{ mg/cm}^2$. The stoichiometry and thickness of the silicon nitride window were controlled by 1.5 MeV He⁺ Rutherford Backscattering (RBS). No deviations from the nominal stoichiometry have been found. The real thickness of the silicon nitride window was 166 ± 8.3 nm (the mean areal mass density of the silicon nitride window was 0.05 ± 0.003 mg/cm²). The higher differences between the thickness given by Norcada and found by our group can be caused by the fact that the declared thickness is taken as a mean value over the whole area of the silicon nitride window, whose size was 1.5 \times 1.5 mm^2 .

2.2. The measurement of stopping power and energy straggling

The measurements were performed on a 3 MV Tandetron MC 4130 accelerator at the Nuclear Physics Institute in Rez near Prague. The ion beams of C^{3+} and O^{3+} were scattered from a thin Au layer prepared by vacuum evaporation onto a glassy carbon substrate. The thickness of the Au layer was determined by Atomic Force Microscopy analysis (AFM) as 18.7 ± 1.8 nm. The ions scattered at a scattering angle of 150° were detected by a partially depleted PIPS detector. An aluminium collimator with two holes 1 mm in diameter was placed in front of the detector and one of the holes was covered with PP foil or a silicon nitride window. This made it possible to register both the primary and decelerated ions simultaneously. All of the measured peaks were of Gaussian shapes. A typical system resolution for 5.486 MeV C and O ions was FWHM ~55 keV and ~78 keV, respectively [32]. This technique had been applied successfully before, see e.g. in [33–35].

The energy spectra were analysed by the ITAP code, which had been developed in our laboratory. This code, written in FORTRAN 90, uses the Levenberg–Marquardt algorithm for nonlinear leastsquares Gaussian fit of the peaks corresponding to the direct and decelerated ions. This code is used to determine the positions and FWHM of both peaks. The experimental values of energy loss (ΔE) were deduced from the positions of energy peaks from primary and decelerated ions. Energy gain (keV/channel) was determined in a separate experiment. The stopping power in MeV mg⁻¹ cm² was calculated as $S(E_{av}) = \Delta E/\rho \Delta x$, where ρ is the mass density and Δx is the thickness of the foil [36]. We define



 $R_a = 1.9 \text{ mm}, \text{RWIS} = 2.3 \text{ mm}$

Fig. 1. AFM image of the surface of the $4\,\mu m$ thick PP foil used in the present experiment.

reference energy as $E_{av} = E_1 - \Delta E/2$, where E_1 is the energy of the back-scattered ions coming from the Au target. The uncertainty in the measured stopping power is composed of energy-loss error and foil-thickness error. Moreover, the energy-loss uncertainty is affected by the energy resolution of the used energy detector and the peak-fitting procedure. The error in the foil thickness was determined from repeated foil weighing. The total uncertainties in the measured stopping power for C and O ions were estimated finally as 2.8% and 3.3% for C and O ions in PP and 5.5% and 6.6% for C and O in silicon nitride, respectively.

The energy straggling Ω_{exp} was determined from the FWHMs of primary and decelerated peaks, using the ITAP code, as $\Omega_{exp}^2 = (FWHM_{foil}^2 - FWHM_{dir}^2)/(8 \times (ln2)^2)$, where FWHM_{foil} and FWHM_{dir} are the peak widths of decelerated and primary ions, respectively. The measured energy-loss straggling value in PP foil was corrected for foil roughness and thickness inhomogeneity by Besenbacher's formula [37], i.e. the value of the final energy straggling Ω is given as following

$$\Omega^2 = \Omega_{\rm exp}^2 - \delta^2 \cdot \Delta E^2 \tag{1}$$

where $\delta = \sigma/\Delta x$ is the standard deviation of the foil thickness relative to its mean thickness. The mean surface roughness σ was determined from AFM. The parameter δ was determined as $\delta = 6.2 \times 10^{-4}$. A typical AFM image of the PP foil is shown in Fig. 1. The measured energy-straggling values in silicon nitride were not corrected for the correlation effects because of the smoothness of the samples. The mean surface roughness of the silicon nitride window has been determined as ~0.46 nm.

3. Results and discussions

3.1. Stopping power

The measured values of stopping power in the PP foil for C and O as a function of ion reference energy are presented in Fig. 2(a) and (b), respectively. The values calculated using the SRIM-2008 and MSTAR codes and our previous experimental data [33] are also presented in Fig. 2. For C ions in the reference energy range of 5.7–7.8 MeV, better agreement is seen with the SRIM calculation than with the MSTAR simulation, the differences between the measured and calculated values being below 3%. The experimental stopping powers are significantly higher than those calculated by the MSTAR. A small difference between our present and earlier data in the case of C ions (Fig. 2a) arises from the better experimental arrangement and data evaluation used in the present study. The measured stopping powers of 4.9–6.0 MeV O ions in PP foil lie between those calculated by the SRIM-2008 and MSTAR codes. In

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