Experimental determination of electronic stopping for ions in silicon dioxide

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The electronic energy loss for ^{4}He, ^{7}Li, ^{9}Be, ^{12}C, ^{16}O, ^{19}F, and ^{28}Si ions in self-supporting silicon dioxide foils has been measured over a continuous range of energies. The measured He stopping powers are in good agreement with the SRIM-2003 (Stopping and Range of Ions in Matter) prediction. In the case of Li and C ions, the measured stopping powers exhibit some deviation from the SRIM-2003 predictions only around the Bragg peak; however, for Be, O, F, and Si ions, the measure stopping powers exhibit up to 10% deviation from the SRIM-2003 predictions over the entire energy range. The results indicate that the modified Bohr formula is suitable for scaling the stopping number for C and heavier ions in the classical interaction regime. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2041828]

For over a century, the stopping of energetic ions in matter has been a subject of great experimental and theoretical interests. In spite of a long history of studies,[1–13] the electronic stopping force is not adequately described by existing models, particularly for the case of heavy ions in compound targets. Furthermore, current theoretical models predict stopping powers that are not always in good agreement with each other.[5–9] Due to experimental difficulties in preparing and handling compound targets for energy-loss measurements, heavy-ion stopping data in compounds are very limited.

Silicon dioxide (SiO_2) is an important dielectric material in semiconductor processing, and other silicates systems, such as zirconium silicates,[14] are considered as next generation dielectrics for semiconductors. Ion implantation processing is an important tool for the production of nanoclusters in SiO_2 and other silicates for many optical applications.[15] Accurate knowledge of electronic stopping power is, therefore, essential to precise control of dopant concentrations over well-defined depth distributions. Silicates and SiO_2 are also widely used in nuclear[16] and space applications,[17] where predictive performance models depend on accurate predictions of electronic energy loss for radioactive decay particles and galactic and solar particles. Furthermore, SiO_2 and various silicate compositions make up about 60% of the earth's crust. The tetrahedral coordination of SiO_2 is almost invariant in silicate structures, which makes understanding the properties of SiO_2 important to the whole class of silicates.

The present letter employs a recently developed approach[4] to determine the energy loss of ions in SiO_2 over a continuous range of energies, where experimental data are unavailable or limited in the literature. The measured stopping powers are compared with theoretical predictions based on the SRIM-2003 (The Stopping and Range of Ions in Matter) code,[6] and the differences in measured and predicted behavior are discussed.

In this study, the self-supporting SiO_2 foils were produced using physical vapor deposition (Luxel Corporation, Washington). The thickness, composition and roughness in the self-supporting foils have been investigated using a Veeco DekTak3ST surface profiler, x-ray diffraction analysis, Rutherford backscattering spectrometry, conventional elastic recoil detection analysis, and by measuring the energy loss of He ions through the foils, as shown in Fig. 1. The results indicate that the foils are amorphous SiO_2 with a thickness of $89\pm1\ \mu g\ cm^{-2}$ (416 ± 5 nm) and a density of $2.14\ g\ cm^{-3}$. No impurities heavier than Si are detected; and the H observed at the surface is negligible.

Energetic ion beams of ^{4}He, ^{7}Li, ^{9}Be, ^{12}C, ^{16}O, ^{19}F, and ^{28}Si were produced by a 5 MV NEC tandem accelerator at

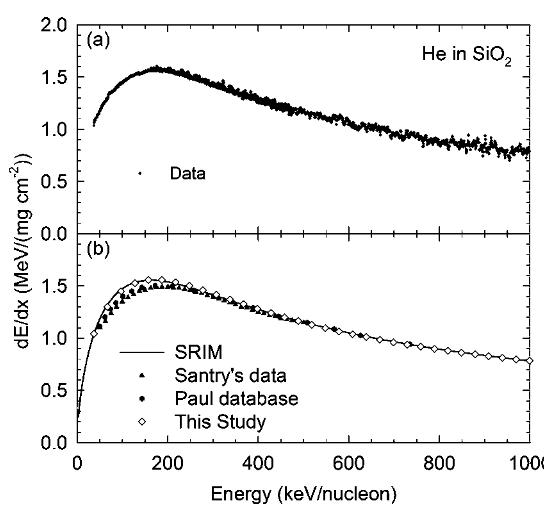

FIG. 1. (a) Experimentally determined He stopping power data from both measurements, only 20% of the data points are actually shown. (b) Comparison of the fitted stopping powers from all the data obtained in this study with literature values and with SRIM-2003 predictions.

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TABLE I. Fitting coefficients [Eq. (1)] to the stopping data for the corresponding validating energy (keV/nucleon) region.

Ion	He	Li	Be	C	O	F	Si
E_{min}	37	98	56	58	98	46	35
E_{max}	1000	1900	1644	1642	1282	1332	860
A_1	0.2128	0.2427	0.3844	0.5424	0.4357	0.4932	0.5300
A_2	0.4474	0.4998	0.4499	0.4699	0.5661	0.5569	0.6068
A_3	0.3290	0.4502	1.0663	4.0859	6.1068	13.2368	1.6570
A_4	3.1039	16.8266	7.8032	0.7173	0.7456	0.4207	6.877×10^6
A_5	13.3530	126.2888	49.1785	4.8126	5.0707	1.7127	1.1036×10^8

Uppsala University. A bulk Au target was used to forward scatter the primary beams. The experimental arrangement[4,11] consists of a time of flight (TOF) spectrometer that is followed by an ORTEC Si charged-particle detector. The SiO_2 foils were mounted on a push-rod that can be reproducibly moved into and out of the ion path between the TOF spectrometer and the Si detector. The scattered primary ions were detected in a forward direction at an angle of 46° for ^{4}He, ^{7}Li, ^{9}Be, and ^{16}O ions, and an angle of 43.5° for ^{4}He, ^{9}Be, ^{12}C, ^{19}F, and ^{28}Si ions, relative to the primary beam direction. The ^{4}He was measured at both angles as an experiment standard. The energy of individual ions prior to impingement on the foil, E_1, is determined from its TOF data; the exit energy after the stopping foil, E_2, is essentially measured using the Si detector, for which every channel has been calibrated using TOF data without the stopping foil present.[4,11] The corresponding TOF data for the same pulse height in the Si detector are determined from the peak position of the time spectra.[11] This approach takes advantage of the continuous energy spectra to calibrate the Si detector over the whole measured energy region. By eliminating the normal calibration problem for Si detectors associated with heavy ions[18] and obtaining good counting statistics (~three million ion events for each measurement), an uncertainty of ~3% is achieved, where the dominant contribution to the total uncertainty arises from the uncertainty in the foil thickness. Ziegler has shown that the stopping power, $-\langle dE/dx\rangle$(MeV mg^{-1} cm^2), is described well over a range of energies (keV/nucleon) by the empirical expression[6]

$$-\left\langle \frac{dE}{dx} \right\rangle = 1/\left\{ 1/(A_1E^{A_2}) + 1/\left[\frac{1000\times A_3}{E} \ln\left(1+\frac{1000\times A_4}{E}+\frac{A_5E}{1000}\right)\right]\right\}, \quad (1)$$

where A_1–A_5 are fitting coefficients and are provided for each elements in Table I that enables easy implementation into other applications.

Both He stopping data sets determined for the two forward scattering angles are plotted in Fig. 1(a), and the overlapping stopping powers from the two measurements indicate excellent repeatability. For purpose of clarity, the fitted stopping power data using Eq. (1) are compared with the SRIM-2003 predictions[6] and existing literature data[19,20] in Fig. 1(b). The results indicate that the current data for ^{4}He ions are in excellent agreement with the SRIM-2003 predictions. The stopping powers determined from this study are higher than values in the literature around the stopping maximum,[19,20] while good agreement is observed at higher energies.

The stopping data for Li, Be, C, O, F, and Si ions in SiO_2 are shown in Fig. 2 over a wide energy range, together with the SRIM-2003 predictions. Good agreement is observed for Li ions at energies above the stopping peak. The SRIM-2003 predictions, which rely on the Bragg's rule that compounds can be treated as mixtures of pure elements with their stopping power being a sum of weighted atomic stopping cross sections, underestimate the measured stopping powers by up to 4% at the stopping maximum. It has been suggested[21–25] that the stopping of light ions in compounds exhibits greatest deviations from Bragg's rule[1] at the stopping peak,[21] and this deviation has been primarily attributed to strong chemical effects. Since the maximum ion-target interaction occurs at the stopping peak, where the valence electrons of atoms and the plasma electrons of the solid dominate the stopping process, the effect of chemical binding is expected to be more significant at such energies.[24] It is worth noting that semiempirical SRIM predictions may not provide accurate stopping for ions in elemental targets,[11] limited accuracy for predictions in compound targets based on the Bragg's rule is expected. The observed difference among the measured He data in Fig. 1 and between the Li data and the SRIM-2003 predictions around the stopping maximum may be attributed to the chemical effect. The deviations are also observed at the Bragg peak for Be and C ions, but to a smaller extent. At higher energies, the stopping values from SRIM-2003 underestimate the stopping power for Be ions, but provide good predictions for C ions, as shown in Fig. 2(a), respectively.

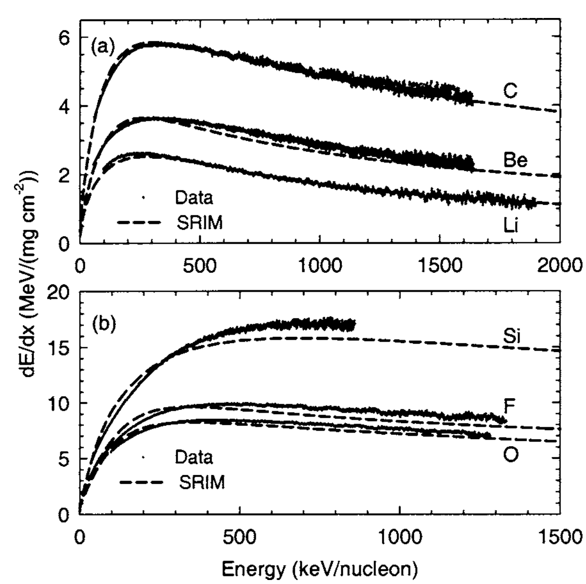

FIG. 2. Experimental stopping data for (a) Li, Be, and C ions, (b) O, F, and Si ions in SiO_2, along with the SRIM-2003 predictions.

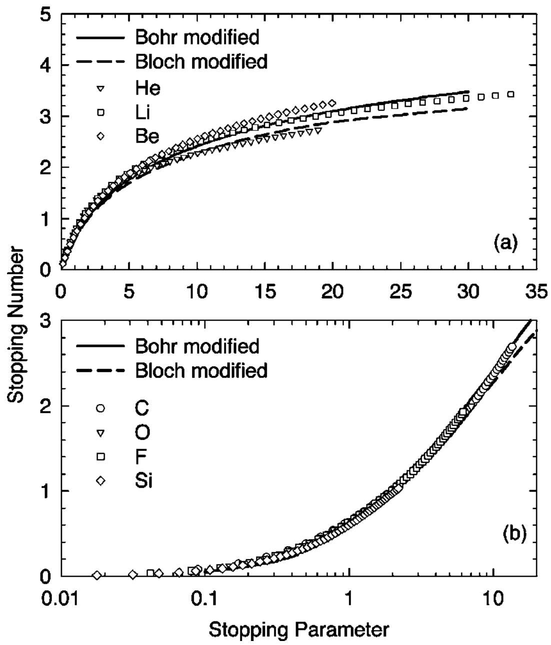

FIG. 3. The experimental stopping number, L_{exp}, against the stopping parameter ξ for (a) He, Li, and Be ions and (b) C, O, F, and Si ions. The stopping numbers from modified Bohr and Bloch formulas are plotted for comparisons.

The measured stopping data for O, F, and Si ions in Fig. 2(b) at low energies up to ~300 keV/nucleon indicate that the SRIM-2003 predictions overestimate the stopping power; however, at higher energies, the SRIM-2003 predictions underestimate the stopping powers by up to 5% for O, 10% for F, and 7% for Si. The results for O and Si ions have some implications for performance modeling of radiation effects in SiO_2 or other silicates in nuclear environments, where much of the energy deposition and radiation damage is caused by energetic self-ion recoils produced radiation.

Figure 3 shows the stopping numbers of L_{exp} derived from the fitting coefficients in Table I as a function of the stopping parameter, ξ.[26,27] The mean excitation energy of 180 eV is used for SiO_2. The modified stopping numbers,[27] L_{Bohr} and L_{Bloch}, where the logarithmic cutoff is removed from the original Bohr and Bethe expressions,[26,27] are also shown in Fig. 3 for comparison. The transition between the Bohr and Bethe theories[26–28] is at an energy of $\sim Z_1^2 \cdot 100$ (keV/nucleon), which indicates that the stopping measurements for He, Li, and Be ions in the current study cover the transition energy region, and the other ions lie within the classical regime. At low energies, the Bloch formula approaches the modified Bohr formula, and both formulas predict the measured L_{exp} well, as shown in Fig. 3(a). The different trends at high energies for He, Li, and Be ions, as compared with the Bohr and Bethe predications, might be associated with a dominant target excitation and/or ionization scattering processes for the fast lighter ions. The data for heavier ions converge into a single trend, and good agreement with both modified formulas at low particle energies is evident, as shown in Fig. 3(b). As the particle velocity increases, the experimental data indicates a trend that is consistent with the Bohr theory, which suggests that the modified Bohr formula is suitable for scaling the stopping number of heavy ions in the classical interaction regime for SiO_2.

The stopping powers for ions in SiO_2 are determined. Good agreement between SRIM-2003 predictions and experimental results are observed for He ions over the entire energy range and for Li and C ions above the stopping maximum. The deviations around the Bragg peak for Li, Be, and C ions may be primarily attributed to strong chemical effects. Different energy dependencies for Be, O, F, and Si ions, as compared with the SRIM-2003 predictions, are observed. The stopping powers for ions in SiO_2 determined in the current study have been parameterized for easy implementation into other applications. The modified Bohr theory is appropriate for scaling the stopping number of heavy ions in SiO_2.

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