Nuclear Instruments and Methods in Physics Research B 300 (2013) 1–5

Contents lists available at SciVerse ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Stopping cross sections of atomic layer deposited Al_2O_3 and Ta_2O_5 and of Si_3N_4 for ^{12}C, ^{16}O, ^{35}Cl, ^{79}Br and ^{127}I ions

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ARTICLE INFO

Article history:
Received 16 July 2012
Received in revised form 12 February 2013
Available online 22 February 2013

Keywords:
Stopping cross section
Al_2O_3
Ta_2O_5
Si_3N_4
Heavy-ions
^{12}C
^{16}O
^{35}Cl
^{79}Br
^{127}I
ALD
TOF-ERDA

ABSTRACT

Stopping cross sections of atomic layer deposited Al_2O_3 and Ta_2O_5 and of Si_3N_4 for 0.03–1 MeV/u ^{12}C, ^{16}O, ^{35}Cl, ^{79}Br and ^{127}I ions have been measured by the transmission technique employing a Time of Flight-Elastic Recoil Detection Analysis (ToF-ERDA) setup. Simulation of experimental slowing down energy spectra was employed to obtain stopping cross section curves over a continuous energy range. The experimental results are compared with the predictions of the SRIM2012 parameterization and literature data.

1. Introduction

Oxides like aluminum oxide and tantalum oxide have technological interest, especially due to their optical and electrical properties. Thin oxide films can be applied as optical coatings, ion-sensitive membranes in solid-state sensors and as high-κ dielectric materials in gate and storage capacitor structures [1–4]. Atomic Layer Deposition (ALD) method providing excellent uniformity and thickness control is increasingly used to produce thin oxide films, e.g. Ref. [3].

The characterization of such films by ion beams often requires the knowledge of the stopping cross sections for heavy ions. To our knowledge such stopping cross section measurements of ALD grown oxide films have not been reported in the literature.

In general, stopping cross section data for heavy ions in solids, especially in compounds, is highly desired. Partly due to the experimental difficulties in the preparation and handling of the compound targets needed for the energy-loss measurements, heavy-ion stopping cross section data of compounds is very scarce. Most of the experimental heavy ion stopping studies deal with elemental targets and usually data only within a limited energy range is provided. The available semi-empirical parameterization approaches [5] predict stopping cross section values in varying agreement with the scarce experimental data. Improvement of theories and predictive models require accurate experimental energy loss and stopping cross section data.

ALD provides a new method to produce films for stopping cross section measurements. The advantages of using ALD processed films are accurate control of sample thickness, good sample quality (stoichiometry and homogeneity) and low level of impurities.

In the current work stopping cross sections of ALD films Al_2O_3 and Ta_2O_5 for ^{12}C, ^{16}O, ^{35}Cl, ^{79}Br and ^{127}I ions have been obtained over continuous ranges of energies by transmission technique employing a Time of Flight-Elastic Recoil Detection Analysis (ToF-ERDA) setup. Stopping cross section of Si_3N_4 used as substrate material for the oxide films was measured simultaneously with the oxides. The measured stopping cross sections are compared with the predictions of the SRIM2012 [5] code and the literature data.

2. Experimental

In transmission geometry utilized in this work, thin self-supporting sample foils are required. This requires that the ALD films

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http://dx.doi.org/10.1016/j.nimb.2013.02.010

Table 1
Measured compositions and areal densities of the supporting Si_3N_4 substrates and the sample oxide films. In the areal densities of oxides the areal densities of atoms in the nitride substrate have been taken into account. The error limits are given in the parenthesis.

Sample	Areal density (10^{15} at/cm^2)						
	H	C	N	O	Al	Si	Ta
Si_3N_4	13.4(8)	1.6(5)	360(8)	8.4(12)		302(7)	
Al_2O_3	15.2(12)	3.9(12)	<9	860(20)	548(12)	<7	
Ta_2O_5	77(2)	5.4(12)	<8	420(9)		<7	145(3)

are deposited on thin substrate foils. This sets clear constraints to the available substrate materials. In the current study, thermodynamically very stable Si_3N_4 material was used. This nitride is widely used in high-endurance and high-temperature applications. The Si_3N_4 substrates were purchased from Silson Ltd. Substrates had window areas of 5×5 mm^2 and nominal thicknesses of 100 nm. Detailed technical material property information, such as homogeneity (thickness variations less than 1%) and roughness (less than 0.5 nm), was received from the manufacturer. Information on the stoichiometry and impurity was measured in this work. The amorphous and homogeneous [1,6,7] Al_2O_3 and Ta_2O_5 samples for the stopping cross section measurements were grown on the Si_3N_4 substrates by the ALD technique. The depositions were made in Picosun SUNALE R-150 ALD reactor at 250 °C. The metal precursors were trimethyl aluminum (Al $(CH_3)_3$) and tantalum pentaethoxide (Ta $(OC_2H_5)_5$) for Al_2O_3 and Ta_2O_5, respectively. The oxygen precursor was deionized water.

The ALD film and Si_3N_4 substrate compositions and areal densities were studied by the ERDA technique in transmission geometry at the Institute for Applied Physics and Metrology, Universität der Bundeswehr München, using 150 MeV ^{127}I ions. The results of the characterizations are presented in Table 1. A low concentration of impurities may be located at the interfaces and at the sample surfaces, in particular because of adsorption from the ambient air. However, such layers are hardly thicker than few molecular layers and therefore their effect on the measured stopping cross sections are insignificant. Total areal densities of the oxide films used in deduction of the stopping cross section values were $(14.3 \pm 0.3) \times 10^{17}$ at/cm^2 and $(6.5 \pm 0.2) \times 10^{17}$ at/cm^2 for aluminum and tantalum oxide samples, respectively. The oxide stoichiometries as obtained in the characterizations were $Al_2O_{3.1\pm0.2}$ and $Ta_2O_{5.8\pm0.7}$. The areal density of the nitride substrate was $(6.9 \pm 0.2) \times 10^{17}$ at/cm^2 and stoichiometry $Si_3N_{3.58\pm0.11}$.

3. Measurements

In the transmission measurements the energy loss of the ions in the sample foils was determined by employing the ToF-ERDA setup described in detail in Ref [8]. The 5 MV tandem accelerator of the University of Helsinki was used to produce the energetic heavy ion beams of ^{12}C, ^{16}O, ^{35}Cl, ^{79}Br, and ^{127}I. Bulk tantalum target was used to forward scatter the primary beams into the ToF-E telescope. The sample foils were placed between the ToF spectrometer and the Si energy detector as described in Ref. [9]. Each measured spectrum consisted of the ToF and energy data of the scattered primary ions penetrating the sample oxide film on the supporting Si_3N_4 substrate, the plain reference silicon nitride foil, and of those hitting the energy detector without penetrating a slowing down material. These three spectra were measured in a single run. The energy histograms summed over 15 time channels from the data of 10 MeV ^{12}C ions for the Al_2O_3 and Ta_2O_5 samples are shown in Fig. 1.

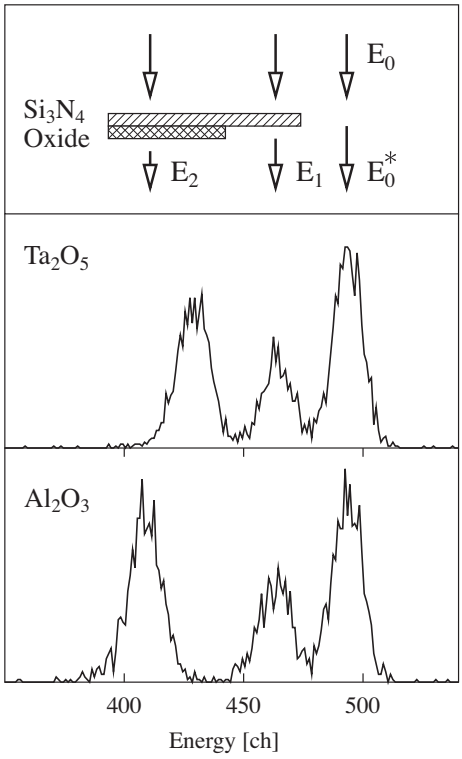

Fig. 1. Schematic illustration of the measurement principle and the energy spectra of 10 MeV ^{12}C ions for Ta_2O_5 and Al_2O_3. Data obtained without sample foil was used to calibrate the energy detector. The calibrated value is described by E_0^*. The ion energy loss in the sample oxide was calculated by subtracting energy E_2 from energy E_1. The energies E_0^*, E_1, and E_2 were obtained from the mean value of the energy peaks in the spectra.

The energy of the individual ions before the sample foil was determined from their ToF data. The ion exit energy after the sample foil was measured using a Si particle detector (Ortec ULTRA series detector with an active area of 300 mm^2), for which each energy channel was calibrated using the ToF data obtained without the sample foil. This approach takes advantage of the continuous energy spectra in calibrating the Si detector by the ToF spectrometer for each channel over the whole measured energy region. It also eliminates the uncertainty resulting from the nominal energy calibration of the Si detector, where parameterization of a simple function is generally used to convert the measured channel number to energy. By eliminating the common Si particle detector calibration problems associated with heavy ions, the corresponding energy value uncertainties in the current study are less than 5%. The effect of the energy loss in the supporting silicon nitride substrate was taken into account by comparing the energy losses in the silicon nitride substrate and in the sample foil with the same ToF and thus the same initial energy. A schematic presentation of the principle is provided in Fig. 1. The energy E_0 of the ions impinging on the sample foils was determined by using the ToF data and tagging the exiting energy E_2 by the Si detector. The exit energy was determined from the corresponding ToF data obtained without the sample foil, based on ions tagged so that they had the same signal response in the Si detector. The ion energy prior to the oxide sample layer is the same as the energy E_1 of the ions exiting the supporting silicon nitride substrate.

4. Results and discussion

The stopping cross section values deduced for pure stoichiometric Al_2O_3, Ta_2O_5 and Si_3N_4 are shown in Figs. 2–4, along with the