Nuclear Instruments and Methods in Physics Research B xxx (2014) xxx–xxx

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

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

New experimental molecular stopping cross section data of Al_2O_3, for heavy ions

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ARTICLE INFO

Article history:
Available online xxxx

Keywords:
Stopping cross section
Stopping power
ERDA
Heavy ion IBA
Light element analysis

ABSTRACT

Molecular stopping cross section data of Al_2O_3, for heavy ions of ^{12}C, ^{16}O, ^{28}Si, ^{35}Cl, ^{79}Br within the energy range of 0.01–1.0 MeV/nucleon were measured. Both direct transmission and bulk analysis methods were applied. Stopping cross sections were calculated both with the SRIM and MSTAR codes. Evaluation and intercomparison of the new data with the calculated and previously measured ones are reported in this paper.

1. Introduction

The successful development and deployment of advanced technologies requires solutions for several materials related problems and challenges. These can range from the ability to design, engineer, and manufacture suitable candidate materials, to reliably predict their life-time and performance from accelerated testing conditions, to the provision of suitable analytical tools and instruments for materials analysis.

Many advanced materials derive their functional properties from compounds containing light elements. Voluminous applications abound for thin films tailored for electrical, electronic, optical, and magnetic properties in which nuclear analytical techniques are being applied. High temperatures, high stresses, corrosive and oxidizing environments, all serve to place high demands on material properties and performances, spawning the development of carbon and oxide ceramic materials. Carbon based materials (e.g. tungsten carbide, titanium carbide, silicon carbide) have very high resistance against high heat loads and can be extremely hard, making them useful for high temperature heat pipes, super hard cutting tools, and plasma facing materials. Alumina, zirconia, and magnesium alloys are being developed for oxidation and corrosion resistant protective coatings [1–3].

Ion Beam Analysis (IBA) methods provide very efficient investigative tools for evaluations, with heavy ion beams from accelerators being applied to analyze surface alterations in a wide range of different matrices. Although, the quality of results highly depends on the existing database for fundamental parameters such as cross sections and stopping powers as well as on the availability of suitable analytical software and its ability to provide reliable and correct results. Erroneous results or misinterpretations of a material's structure and composition can result from inadequate science in the analysis software, insufficient accuracy in basic ion beam data, or inadequate documentation and guidance for people to extract the correct information.

For compounds, the accuracy is often worse than for elemental materials, not least because the Bragg's rule is often assumed in the calculation of stopping powers, that is, a weighted average of the bulk elemental stopping powers is used. Large deviations (up to 10–20% [4]) near the stopping power maximum are often observed in insulating compounds, particularly in oxides and nitrides of heavy elements. The "Stopping Range of Ions in Matter, SRIM code" – which is mostly used for stopping cross section data – [5,6] has a

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http://dx.doi.org/10.1016/j.nimb.2014.02.092
0168-583X/

compound correction feature that leads to improved calculations, but not all materials are yet included, particularly novel ones. Also, most analysis codes do not support molecules and therefore are unable to include SRIM's compound correction, with NDF [7] being one of the few codes that does support it.

The motivation of this work was to deliver demonstrable improvements for the heavy ion ERDA technique to analyze light elements with a higher degree of reliability, accuracy, and user confidence. The strongest constraint identified was the lack of basic stopping power data for heavy ions, particularly in compounds. Therefore, experimental and simulation activities were undertaken for Al_2O_3 samples to acquire data and information to alleviate deficiencies and inadequacies in the analytical methodology, and to improve the reliability and accuracy of heavy ion stopping cross sections. The intercomparison of experimental stopping cross section data for this compound material is reported.

The work reported here is part of a wider project, i.e. the International Atomic Energy Agency Coordinated Research Project: "Improvement of the Reliability and Accuracy of Heavy Ion Beam Nuclear Analytical Techniques" in which the stopping cross sections of a large number of materials were measured for different heavier ions [8].

2. Software and databases of stopping power data

Experimental stopping power data is dispersed in countless articles over the decades. Very often the data is given in graphical form without numerical values. Researchers interested in the experimental stopping power for a given ion-material combination need to search through the literature.

A very strong effort is required to collect existing data into a database. This effort has been made by Ziegler (see [5] and references within), and all the data is shown in the SRIM website [6]. This code is based on theoretical formulations and semi-empirical models, and it includes a host of adjustable parameters that come from adjusting the models to existing experimental data. The data sets are not given numerically, but the references where they were extracted from are given, which makes searching much easier.

Another strong effort has been made by Paul, given in the stopping power compilation of Paul and Schinner [9,10]. This database may have slightly fewer data sets than SRIM, particularly for protons and alpha particles, but it has the advantage that all the data is given numerically. The data is available at the website created by Paul [11], and also at the IAEA Nuclear Data Section website [12].

The ICRU Report 73 [13] includes a compilation of stopping powers of many targets, including compounds, for ions from Li to Ar, and for Fe.

There are many computer programs that calculate stopping powers in the energy range useful for IBA. The most widely used software is SRIM. Its calculations and models have changed with time, and are regularly updated. SRIM is used by new generation data analysis codes such as SIMNRA [14] and NDF [7]. NDF can also use other software, such as MSTAR [9], or experimental tabulated data [15] for elements and compounds.

3. Methodology

3.1. Materials

As for substrate, commercially available silicon nitride membranes were acquired [16]. Membrane thicknesses of 30 and 100 nm were selected, both having surface areas of 4×4 and 5×5 mm^2. The Al_2O_3 films were grown on the Si_3N_4 membranes by the atomic layer deposition (ALD) technique at Helsinki University, Laboratory of Inorganic Chemistry. The growth temperature was 250 °C and the employed facility was a Picosun SUNALE R-150 reactor. In the preparation process, special attention was paid to eliminate growth of ALD layer on the membrane backside, which would have triggered complications in the analysis of the measurement results. The best combination of the size and substrate thickness was 4×4 mm^2 and 100 nm, respectively.

Al_2O_3 films were also produced by electron beam deposition onto Si_3N_4 foils at the Materials Department of iThemba LABS. The surface topography of the foils was mapped using an Atomic Force Microscope (AFM), scanning over areas of up to 20×20 μm^2 at a time. The film thickness was 38.4 μg cm^{-2} with 0.5 nm roughness.

3.2. Experimental details

The composition, homogeneity and thickness of used samples were characterized by IBA methods. Stopping cross section measurements were conducted in each participating laboratory either with the direct transmission technique or with the bulk method [17–20]. For particle detection ToF telescope, magnetic spectrometer and particle detectors were applied. ^{12}C, ^{16}O, ^{28}Si, ^{35}Cl and ^{79}Br ions were used for the stopping cross section measurements. The energy range was different for different ions and targets, but always in the range useful for IBA. Full details on the experimental conditions are given in [21–23].

The main difficulty with the transmission methods is that foil on which film is deposited should be characterized with high accuracy. Thickness, thickness inhomogeneity, pinholes, and density are all factors that need to be well known. Furthermore, the foils can be tens or hundreds of nm thick, and their physical stability in vacuum and under the beam irradiation is also an issue.

One alternative, which is experimentally much simpler, is to use a bulk sample of the material under study. The resulting experimental RBS spectrum depends on the experimental conditions and on fundamental quantities such as the scattering cross section and the stopping power. A theoretical RBS spectrum can be simulated, assuming given experimental parameters and given fundamental quantities, including a given stopping power, that can be treated as a fit parameter. A physical model for the calculation of the theoretical spectrum is also required.

3.3. Data evaluation and intercomparison

In order to compare results obtained in different labs for the same system, often using independent samples and methods, the precision of each measurement needs to be known. For each measurement the uncertainty budget was calculated using the concept described with special reference to IBA by Wätjen and co-workers [24], and in the Pitfalls chapter of the Handbook of Modern IBA [25]. All uncertainties in this work are given with a coverage factor $k = 1$, that is, they are one standard deviation, except where otherwise stated.

A very strong and thorough effort has been made to explicitly include all sources of error in the uncertainties reported in this work. In almost all cases, the statistical error is a small part of the final uncertainty, which is dominated by other factors, such as the areal density of the samples or the energy loss in the substrates.

This means that deviations between the reported values and, for instance, values calculated with the interpolative schemes such as SRIM, that are outside the uncertainties calculated, should mean that the interpolative scheme is inaccurate in the considered energy range for the particular ion/target combination.

The same reasoning means that, if two different experiments differ by more than their stated accuracies, it should be considered that there is something wrong with one or all of the experiments reported. One should note however, that a coverage factor $k = 1$ (one standard deviation) corresponds to a level of confidence of

Please cite this article in press as: A. Simon et al., New experimental molecular stopping cross section data of Al_2O_3, for heavy ions, Nucl. Instr. Meth. B (2014), http://dx.doi.org/10.1016/j.nimb.2014.02.092