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Experimental stopping powers of Al, Mg, F and O ions in ZrO_2 in the 0.1–0.6 MeV/u energy range

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ABSTRACT

The study of the stopping and range of heavy ions ($Z > 3$) in matter isdsc cfddc of both fundamental and practical interest as heavy ions find increasing usage in a wide range of ion beam based techniques. While semi-empirical formulations like the widely used SRIM code can predict the stopping powers of hydrogen and helium in many elemental and compound targets over a wide range of energies quite well, their predictive accuracy for heavy ions is not as good. This is mainly due to the lack of heavy ion experimental stopping power data on which such codes are based. We present results of measurements performed using a Time of Flight–Energy spectrometer to determine the stopping powers of O, F, Mg and Al ions in the oxide ceramic ZrO_2 within the 0.1–0.6 MeV/u energy range. Possible SRIM correction (or correlation) factors for F, Mg and Al ions were extracted from quantitative comparisons of experimental and predicted stopping power values.

1. Introduction

The study of the passage of ion beams in matter is a well-established field in the nuclear sciences spanning over a century of theoretical and experimental research work [1]. Its continual advancement lends itself to the need to understand the nature of the interactions between energetic ion beams and matter, and equally important to the widespread technological and medical applications of the passage of ion beams through matter.

In materials research for example, measurement of the energy loss factor (or stopping power) and range of ion beams in matter is crucial in applications such as ion beam analysis, materials modification and ion implantation. Experimental stopping power data is also needed for the validation of theoretical and semi-empirical models. The latter are invaluable in practical applications as it is impossible to measure the stopping powers of all ions in all kinds of matter for every possible energy range. It is therefore important that the predictive accuracy of such codes be tested whenever possible. We report here on measurements performed to determine the stopping powers of O, F, Mg and Al ions in the oxide ceramic ZrO_2 over the energy range 0.1–0.6 MeV/u.

2. Experimental

In our previous work we reported on a newly built Time of Flight–Energy (ToF-E) spectrometer, set up for Heavy Ion–ERD analyses and adapted for energy loss measurements [2]. The basic experimental arrangement is shown in the sketch in Fig. 1. Further details of the spectrometer can be found in Ref. [2].

The measurement procedure basically entails a coincidence measurement of the time of flight of recoil ions before they hit a stopper foil and their energy after they pass through the foil. The energy of the incident ion before hitting the stopper foil is determined from the *ToF* measurement and the exit energy is determined from a *ToF-E measurement without the stopper foil* between the second timing detector (see Fig. 1) and the energy detector. The energy signal from the surface barrier detector (SBD) is used to tag events of a similar energy on two *ToF-E* curves; from runs with and without the stopper foil, and the corresponding *ToF* then used to calculate the energy in each instance using the known atomic mass and flight length [3]. The difference between the two energies, normalized by the foil thickness, gives the stopping power of the recoil ion in the stopper foil.

A 27.5 MeV Kr^{15+} beam was used to forward scatter desired recoil ions from appropriate target samples in to the detector system. Aluminium and magnesium recoils were obtained from thick ($\sim$1.0 μm) layers of Al_2O_3 and MgO on silicon substrates, respectively. Fluorine ions were recoiled off a thick LiF crystal and oxygen ions were obtained simultaneously with the aluminium recoils

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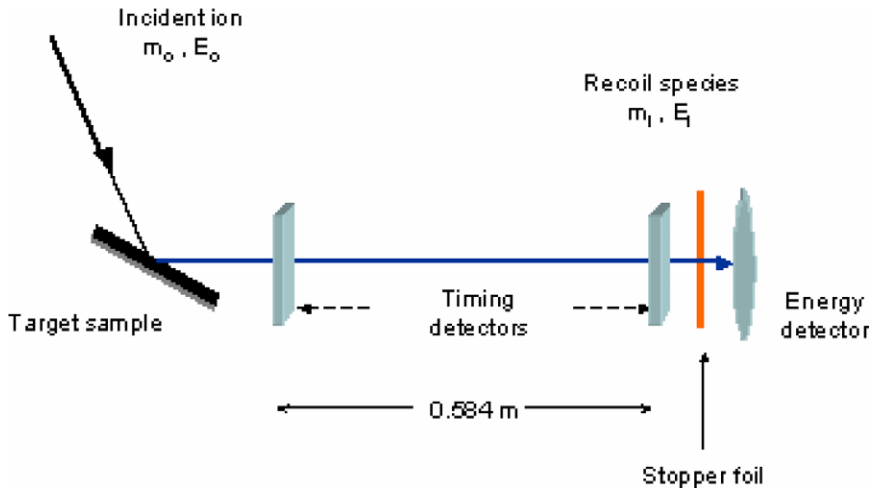

Fig. 1. A schematic of the ToF-E spectrometer set up for the measurement of the energy loss of recoil ions through a stopper foil.

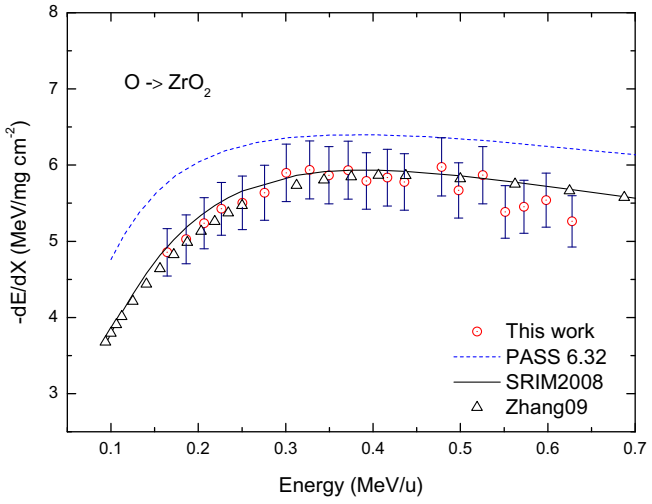

Fig. 2. Experimental stopping power data of ^{16}O ions in ZrO_2 compared to predictions from SRIM [5] and PASS [6] codes and experimental data from Zhang and Weber [7].

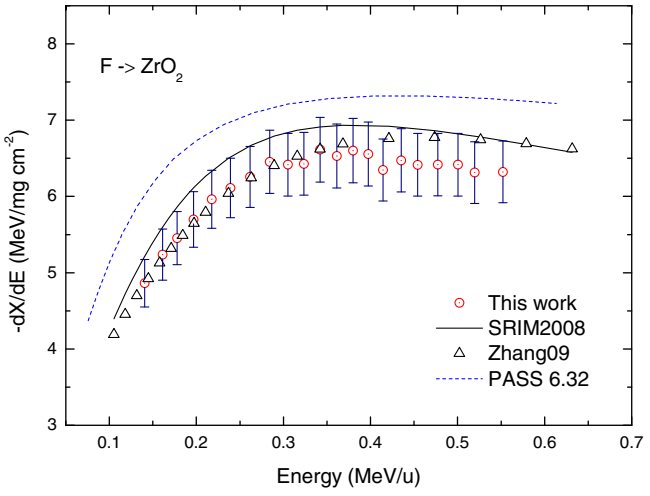

Fig. 3. Experimental stopping power data of ^{19}F ions in ZrO_2 compared to predictions from SRIM [5] and PASS [6] codes and experimental data from Zhang and Weber [7].

from the Al_2O_3/Si sample. The uncertainty in the measurement of the energy loss of the recoil ions through the stopper foil, resulting from uncertainties associated with time calibration and time of flight measurement, was about 4.8%.

The ZrO_2 foil, produced by physical vapour deposition, was purchased from ACF-Metals. The foil topography was scanned using an Atomic Force Microscope and found to have an average roughness of about 3.0 nm. The foil thickness determined both by Rutherford Backscattering (RBS) analysis and energy loss measurement of 5.48 MeV ^{241}Am alphas was found to be 331 μg cm^{-2}. The stoichiometry was confirmed by RBS to be roughly Zr: 1 O: 2. The uncertainty in the foil thickness, determined from consideration of the contributions due to roughness (0.36%), lateral variation (1.0%), energy straggling of the measuring alphas (0.68%), detector energy resolution (0.3%) and uncertainty in the stopping power of helium ions in ZrO_2 (4.1%) [4], worked out to be 4.3%.

3. Results

Figs. 2–4 and 6 show the results of measurements of the stopping powers of the four ions in ZrO_2. Also included in the figures are predictions from the latest version of SRIM (SRIM2008) [5]

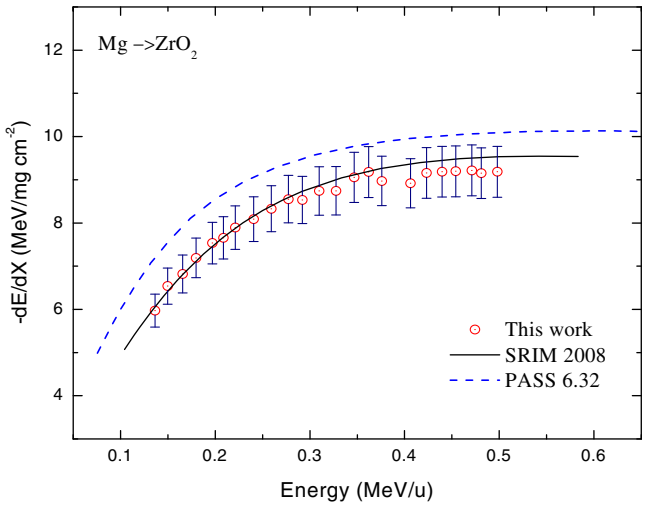

Fig. 4. Experimental stopping power data of ^{24}Mg ions in ZrO_2 compared to predictions from SRIM [5] and PASS [6] codes.

and from Sigmund and Schinner's theoretical code PASS [6]. In addition, Figs. 2 and 3 show results by Zhang and Weber [7], the

only other available experimental stopping power data for O and F ions in ZrO_2. Their data was reproduced here using the following fit equation to their experimental data plots:

$$-\left\langle \frac{dE}{dX} \right\rangle = \left\{ \frac{1}{A_1 E^{A_2}} + \frac{1}{\left[\frac{1000xA_3}{E} \times \ln\left(1 + \frac{1000xA_4}{E} + \frac{A_5 E}{1000}\right)\right]} \right\}^{-1} \quad (1)$$

where E is the energy in keV/u and A_1–A_5 are the fitting coefficients, given in Ref. [7]. For Mg and Al ions, ours are apparently the first experimental results on the stopping of these ions in ZrO_2, over the 0.1–0.6 MeV/u energy range. The relative uncertainty in the measured stopping powers, a resultant of the error terms in the measurement of the energy loss (4.8%) and the foil thickness (4.3%), is estimated to be 6.4%.

At first glance it can be readily seen that PASS consistently overestimates experimental data in all the four cases. The degree of overestimation exceeds the experimental uncertainty in all instances as well. Fig. 2 shows the results for O ions. Good agreement is observed between the two experimental data sets up to 0.5 MeV/u. Compared to SRIM however, it can be noted that prediction slightly (<3%) overestimates the stopping of oxygen ions in ZrO_2 over ~0.10–0.30 MeV/u, the energy range of the stopping power maximum. For F ions in Fig. 3, the two experimental data sets agree quite well between 0.1 and 0.4 MeV/u, after which deviations in the order of the experimental uncertainty occur. In the region of agreement SRIM consistently overestimates the experimental data by over 6%. Within that energy range, a plot of the ratio of experimental to SRIM stopping power values (S_{EXP}/S_{SRIM}) versus energy yields a correction factor (f_C) of 0.94 that may be used on SRIM values to describe stopping more accurately.

Fig. 4 shows the results of measurements for Mg ions. Experimental data and SRIM prediction generally agree over the measured energy range. However there is a tendency by SRIM to overestimate experimental data at low energies and underestimate it at higher energies. This is shown more clearly in Fig. 5, which is a plot of the ratio of experimental to SRIM stopping power values versus energy.

Because of the relatively minor discrepancy between SRIM and experiment, we define instead, an energy dependent SRIM–EXPT *correlation* factor f_{SE} given by a trend line fit to the data in Fig. 5; $f_{SE} = 1.026 - 0.128E$. Experimental and theoretical stopping power data for Al ions are compared in Fig. 6. A trend quite like that for Mg ions is observed; SRIM underestimates experimental data at low energies and overestimates it at higher energies. Similarly from an S_{EXP}/S_{SRIM} versus energy plot the following energy dependent SRIM correlation factor was obtained for aluminium ions;

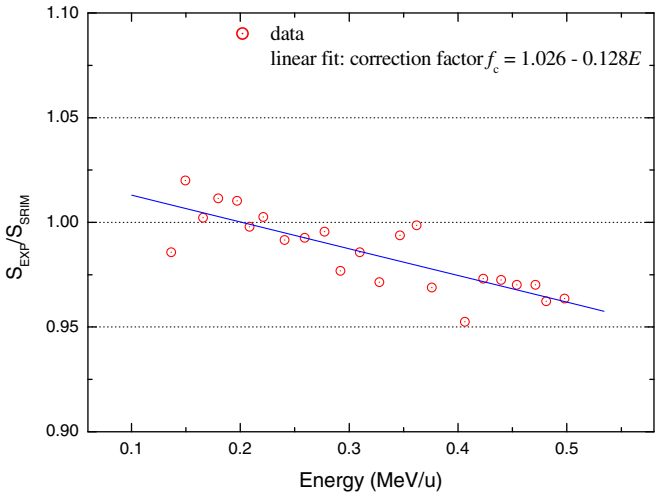

Fig. 5. Plot showing the energy dependency of the ratio of experimental to SRIM stopping power values of ^{24}Mg ions in ZrO_2.

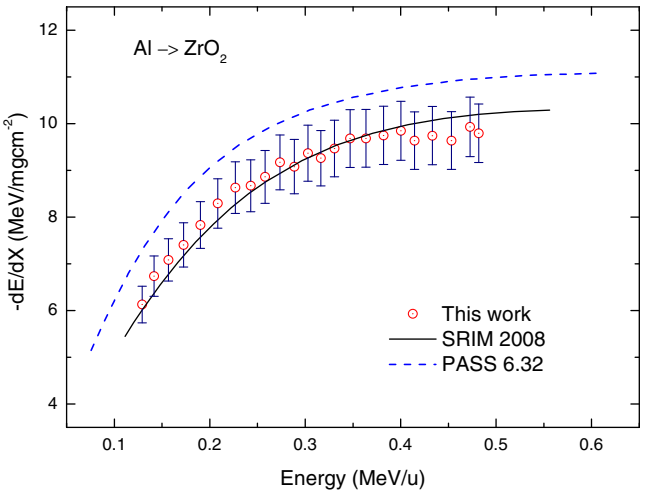

Fig. 6. Experimental stopping power data of ^{27}Al ions in ZrO_2 compared to predictions from SRIM [5] and PASS [6] codes.

$f_{SE} = 1.0814 - 0.251E$. These correlation factors are expected to be valid over the energy range of the measurement; 0.10–0.60 MeV/u.

4. Discussion and conclusion

The relatively slight deviation of <3% between experimental data and SRIM prediction for O ions can be considered to be due to experimental scatter. The stopping powers of Al and Mg ions in ZrO_2 measured here are the first experimental data of the stopping of these ions in this ceramic. Results show that within experimental limits, SRIM predictions adequately describe the stopping of both ions. For F ions however, over the 0.1–0.4 MeV/u energy range, it would be incorrect to attribute the (level of) disagreement between experiment and theory to experimental uncertainties. There are limitations in SRIM calculations that may account for the discrepancy observed.

In the calculation of stopping powers SRIM uses what is known as Bragg's additivity rule, whereby the stopping power of a compound for a given projectile ion is estimated by the linear summation of the stopping powers of its constituent elements for that ion. Correction factors to Bragg's rule, such as those calculated through the cores-and-bonds (CAB) approach [8], are used to provide SRIM correction factors for quite a number of compounds in the SRIM database, mostly hydrocarbons. Compounds like metallic oxides, have not been included because a lot of previous measurements concluded that for such compounds, which contain heavy elements, deviations from Bragg's rule disappear [8]. For most of these measurements however, tests of deviation from Bragg's rule were limited to protons and helium ions [9,10]. The level of disagreement between experiment and SRIM prediction for heavier ions might not be insignificant in some instances. The stopping powers of F ions measured in this work and also from Ref. [7] provide a good example that attests to this assertion.

The experimental correlation factors (f_{SE}) obtained here for Mg and Al ions in ZrO_2, may be used to facilitate quantitative comparison of current data against any other experimental result. For F ions we propose that the 0.94 correction factor be considered the first attempt at obtaining an experimental CAB correction factor for F ions in ZrO_2.

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