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Stopping force of Ti for $6 \leqslant Z \leqslant 29$ ions measured by time of flight spectrometry

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ABSTRACT

Accurate stopping force data is of vital importance in ion beam materials analysis using heavy ion beams. The predictive accuracy of theoretical and semi-empirical formulations can only be improved through continual validation of these codes by making available experimental data from a wide range of projectile-target combinations. In this work stopping force measurements of Ti for ^{12}C, ^{16}O, ^{24}Mg, ^{27}Al and ^{63}Cu ions were carried out by heavy ion Time-of-Flight Elastic Recoil Detection (ToF-ERD) spectrometry and the results are compared with semi-empirical calculations by Ziegler's Stopping and Range of Ions in Matter (SRIM-2010) code, and *ab initio* calculations by Grande and Schiewietz's Convolution approximation for swift Particles (CasP 5.2) code. Both SRIM and CasP underestimate stopping in the energy range studied. SRIM predictions average within 10% of data and CasP calculations range within 5–25% of data.

1. Introduction

The study of the passage of energetic ions in matter continues to be an integral part of ion beam physics due to the ever increasing number of ion beam-matter interaction applications [1]. Advances in ion implantation, ion beam materials modification and nuclear analytical techniques [1] all depend on the availability of accurate description and prediction of ion beam passage through matter. The stopping force for ions through matter is one of the key parameters used to characterise this interaction. The accuracy of different theoretical formulations that predict stopping force is restricted to specific energy ranges [2], with pioneering Bethe-Bloch based theories applicable in the high to relativistic energy region and Lindhard, Scharff and Schiott (LSS) based formulations more appropriate for the low energy range. For the intermediate region, where the stopping force maximum lies, a satisfactory theory remains to be devised [2,3]. A number of computer codes based on later developments in stopping theories have been written for the calculation of stopping force for ions in matter. The most common include CasP (Convolution approximation for swift Particles) [4,5], CKLT (Convergent Kinetic Lindhard Theory) [6], PASS (Binary theory) [7], TCS (Transport Cross Section) [8], and calculations by Heredia-Avalos et al. [9]. Of these CasP 5.2 is the most readily available to users (https://www.helmholtz-berlin.de/people/gregor-schiwietz/casp_en.html).

Experimentalists generally rely on semi-empirical formulations – with Ziegler's Stopping and Range of Ions in Matter (SRIM) [10] the most widely used – for stopping force values for ions in matter. In ion beam analysis any inaccuracy in the stopping force, however, is directly reflected in the depth scales determined. While fairly good at energies below and above the Bragg peak for light projectiles traversing elemental targets, SRIM predictions can be off by large margins in the Bragg peak region in compound/complex targets, especially for heavy ions [11–13]. This presents a particularly pertinent problem for heavy ion Elastic Recoil Detection Analysis (ERDA) work, where beam energies are in the 0.1–1.0 MeV/u energy range. The provision of experimental stopping force data to validate theoretical and semi-empirical formulations is thus a vital component of ongoing studies of the passage of ion beams through matter. This article adds on to earlier contributions made by our group to the global database of heavy ion stopping forces of different target materials. We report here on measurements carried out to determine the stopping force of Ti for ^{12}C, ^{16}O, ^{24}Mg, ^{27}Al and ^{63}Cu ions at projectile energies ranging from ~0.1 MeV/u to just below 1.0 MeV/u.

2. Experimental details and measurement

2.1. Target foil

Free standing titanium foils were sourced from Lebow Company (www.lebowcompany.com). The thickness of the foil used for this

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particular study was measured by Rutherford Backscattering Spectrometry (RBS) using 2 MeV He^+ ions at the iThemba LABS 6 MV Van de Graff accelerator. The backscattering angle was set at 165° and the foil surface normal tilted off the beam axis by 10°, away from the detector, to increase the beam path length for better depth resolution. The RBS spectrum is shown in Fig. 1, together with a SIMNRA [14] simulation. The best fit to the experimental data was obtained from a layer structure consisting of very similar thin (Ti_x–O_y–C_{1-x-y}) contamination layers on either side of the main/bulk part of the foil. It could only be expected that these impurity layers be the same since both sides of the foil are, in principle, the same. The front layer was $Ti_{0.54}O_{0.21}C_{0.25}$ of thickness 140×10^{15} at/cm^2 and the underside layer was $Ti_{0.53}O_{0.22}C_{0.25}$, measuring 145×10^{15} at/cm^2 thick. The titanium bulk layer was found to be 2822×10^{15} at/cm^2. The expected low energy peak due to the carbon impurity on the underside of the foil does not show in the experimental spectrum in Fig. 1 because the low energy threshold of the data acquisition electronics was inadvertently set higher than this carbon peak energy. It is for this reason that only the simulated peak is shown. The uncertainty in the thickness value determined this way is a convolution several contributions. The main contributors are the uncertainty in the SRIM stopping force for He ions through the foil 3.5% [10] and the foil thickness non-uniformity, estimated to be 1% by the manufacturer. Other (minor) factors include the RBS detector energy resolution (quoted at 20 keV at 5.46 MeV), 0.5% in the beam energy spread, 0.16% uncertainty in the counting statistics estimated from the Ti peak, and 0.21% uncertainty in the SIMNRA code [15]. All these factors add up to an effective 3.7% uncertainty in the film thickness.

2.2. Measurement set up and data analysis

Stopping force measurements were done at the iThemba LABS 6 MV tandem accelerator, using the Heavy Ion ERDA set up [16]. A 26.0 MeV $^{63}Cu^{7+}$ projectile beam was used to forward recoil ^{12}C and ^{27}Al ion species from C-graphite and thick Al_2O_3-on-Si target samples, respectively, towards the target stopper foil. Similarly ^{16}O and ^{24}Mg ions were recoiled off a thick MgO target layer on a silicon substrate. Copper ions incident on the stopper foil were obtained by scattering the incident beam off a thick Au-on-Si layer sample.

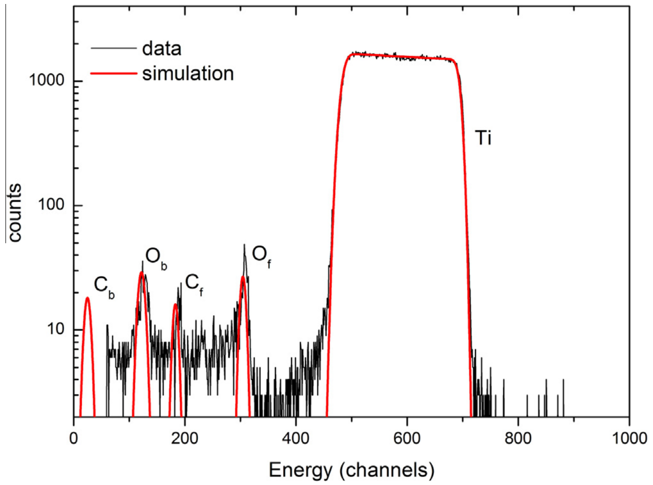

Fig. 1. Measured and simulated RBS spectra from the analysis of the titanium foil. Low energy peaks represent signals from oxygen and carbon impurities from the front (f) and back (b) surfaces of the foil, respectively.

A time-of-flight spectrometer was used to measure the energy (E_1) of the recoil ions before passing through the stopper foil and a Silicon PIPS® detector used to tag the exit energy (E_2) after the foil. Coincidence measurement of the time of flight and energy of the recoils *without* the stopper foil facilitated a one-to-one channel-to-energy calibration of the energy detector and so avoided the non-linear response of Si detectors commonly associated with heavy ions. The titanium foil used here was 'sandwiched' between two thin impurity layers as described in Section 2.1. The measured energy loss ($\Delta E = E_1 - E_2$) was that through the foil and the impurity layers combined and so corrections have to be made to determine the energy loss ΔE_{Ti} through the pure titanium alone.

The recoil ion energy (E_1') after passing through the top contamination layer is given by

$$E_1' = E_1 - x_1 \cdot S(E_1) \tag{1}$$

where x_1 is the thickness of the contamination layer and $S(E_1)$ is the stopping force for the recoil ion through that layer. Similarly, the residual ion energy (E_2') after passing through the pure titanium, and just before entering the bottom contamination layer is given by

$$E_2' = E_2 - x_2 \cdot S(E_2) \tag{2}$$

where x_2 is the thickness of the contamination layer and $S(E_2)$ is the stopping force for that recoil ion through that layer, assumed to be the same as at the measured exit energy E_2. $S(E_1)$ and $S(E_2)$ were estimated by SRIM calculations. The energy loss through the pure titanium foil (i.e. the difference between E_1' and E_2') is then given by

$$\Delta E_{Ti} = E_1 - E_2 - x_1 \cdot S(E_1) - x_2 \cdot S(E_2) \tag{3}$$

The uncertainty in the energy loss as calculated by Eq. (3) is dominated by a 3.4% uncertainty in the determination of ($E_1 - E_2$) from the time of flight spectra [17]. The energy loss correction terms in Eq. (3) are both in the range of 0.1–1.0% for high and low energies, respectively. Combining these uncertainties gives an upper limit of 3.7% in the calculated ΔE_{Ti} values. The experimental stopping force of titanium for the different ions is calculated using

$$S_{exp} = \frac{\Delta E_{Ti}}{x_{Ti}} \tag{4}$$

where x_{Ti} is the thickness of the pure titanium foil. The range of uncertainties in the experimental stopping force values then varies from a minimum up to an estimated maximum of 5.2%, calculated from the uncertainties in ΔE_{Ti} (3.7%) and x_{Ti} (3.7%).

3. Results and discussion

Fig. 2 shows the results of stopping force measurements for ^{12}C ions through Ti, compared with predictions by the semi-empirical SRIM code and Grande and Schwietz's CasP *ab initio* calculations. Also included in the plot is data from Zheng et al. [18]. The results show that data obtained in this work is in agreement with both Zheng and co-workers' data and SRIM prediction. CasP on the other hand underestimates experimental data by over 20% at the Bragg peak region. Results of measurements of the stopping force for ^{16}O ions through Ti are plotted in Fig. 3. Current data is in agreement with the only other data set available in the literature by Lu et al. [19], where the two data sets overlap in energy. SRIM describes data well up to about 0.38 MeV/u but thereafter underestimates experiment. CasP predictions fall below experimental data throughout the whole energy range.

Stopping force measurement results for ^{24}Mg ions in Ti are presented in Fig. 4, and again are compared with SRIM and CasP predictions. There is no other data set available in the literature for this ion-target pair. SRIM underestimates current experimental data by over 10% at the Bragg peak region but tends to agree with data at lower energies. CasP underestimates data by up to 20% at the Bragg peak region. The results for Al stopping in Ti are shown