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Stopping cross section of vanadium for H⁺ and He⁺ ions in a large energy interval deduced from backscattering spectra



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ABSTRACT

We have experimentally determined electronic stopping cross sections of vanadium for 50–2750 keV protons and for 250–6000 keV He ions by relative measurements in backscattering geometry. To check the consistency of the employed procedure we investigate how to define adequate reference stopping cross section data and chose different reference materials. To proof consistency of different reference data sets, an intercomparison is performed to test the reliability of the evaluation procedure for a wide range of energies. This process yielded consistent results. The resulting stopping cross section data for V are compared to values from the IAEA database, to the most commonly employed semi-empirical program SRIM, and to calculations according to CasP. For helium, our results show a significant deviation of up to 10% with respect to literature and to SRIM, but are in very good agreement with the CasP predictions, in particular when charge-exchange processes are included in the model.

1. Introduction

When an energetic particle penetrates matter, the projectile will experience a retarding force F = -dE/dx commonly referred to as stopping power S. Energy loss due to interactions with the nuclei and electrons of the target atoms is denoted as nuclear and electronic stopping, respectively [1]. In many applications, an alternative and more convenient expression is obtained by normalizing the stopping power by the density of the material -(1/n) dE/dx, i.e. the stopping cross section, ε . Stopping cross section has the main advantage to relate the energy loss to a certain number of target atoms rather than to path length. For H⁺ and He⁺ ions in the Ion Beam Analysis (IBA) energy range, the dominant contribution to the total stopping power is due to electronic stopping; nuclear stopping is negligible for proton equivalent energies $E/A \gtrsim 100 \text{ keV/u}$, with A being the relative atomic mass of the projectiles. In this regime, backscattering spectra can be analyzed within the single scattering model. Thus, analytical modeling of the energy spectra of backscattered ions represents an accurate description of the experimental spectra. At lower energies, multiple scattering becomes more important, and Monte Carlo simulations are required to disentangle electronic and nuclear losses, due to the lack of an analytical model.

These quantities and the underlying processes, although under experimental and theoretical investigation over more than one century [2], still provide remarkable insights into ion-solid theory [3] and complex non-equilibrium atomic physics [4,5]. Apart from fundamental science, accurate knowledge on energy deposition in materials plays also a crucial role in many fields of modern science and technological applications. It is a fundamental pre-requisite for almost all IBA techniques to obtain accurate sample composition profiles [6]. Stopping powers also play a prominent role in material modifications (such as ion implantation and irradiations) [7], plasma-wall interactions (fusion research) [8], and beam-patterning studies (nanotechnology) [9].

A remarkable application of energy deposition by ions is in radiotherapy by use of protons or heavier ions (ion-therapy) [10]. For deep lying tumors, ions have advantage over conventional photon-therapy that less energy is deposited along the path to the tumor, while confining most of the energy around a well-defined region – the Bragg peak – sparing the healthy tissues around the tumor. Hence, accurate predictions of the stopping power are of high relevance for a better accuracy of therapeutical approaches based on energetic ions [11].

Modern applications are increasingly pushing the ion physics community for more accurate data. In this aspect, recent experimental efforts have aimed at improving the accuracy and traceability of stopping

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Fig. 1. Experimental (open circles) and simulated (solid lines) energy spectra of 200 keV D + ions backscattered from Cu (black data) and V (red data) are shown. Simulation and experiment coincide perfectly energy interval used in the evaluation (dashed blue lines, see Evaluation Procedure in Section 3). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

power measurements, by employing a fully traceable protocol of energy loss measurements, achieving results within overall uncertainties of $\approx 1\%$ using transmission method [12]. An up-to-date list of experimental ε is maintained by the IAEA in an online database [13], and contains data sets from more than 850 references [14]. Examining this database, one can easily identify weaknesses for many ion-target combinations: either there are no experimental data published or, in the worst case, available data are widely spread and different data sets exhibit discrepancies well beyond stated uncertainties. Nevertheless, for elements which are commonly available with high purity (such as C, Al, Si, Cu, Ag and Au), there are numerous data sets in mutual agreement, especially for more recent data. In contrast, for transition and rare earth metals experimental data are scarce, with worse overall agreement between the different datasets [15].

Particularly for compounds, there is a lack of ε data and specially for low energies the stopping predictions of compounds are still deduced from elemental standards by applying Bragg's rule [16]. However, due to chemical state effects [17,18], making predictions from elemental standards do not lead to reliable enough results [19,20].

Therefore, the aim of the present work is twofold: firstly, to describe in detail how one can deduce reliable ε data within averaged random uncertainty of $\lesssim 3\%$ for protons and for helium from the heights of backscattering spectra obtained for elemental solid samples of a reference material and the target material of interest. Secondly, this methodology is applied to subsequently deduce electronic ε data for a material system of high technological relevance in thin film applications, i.e. vanadium [21], by comparison to a well characterized reference system (Cu). Consistent data were obtained in three different laboratories (described in the next section) for both proton and helium projectiles in a large energy interval covering also the Bragg peak. The resulting ε data are compared to data from IAEA database [13], to SRIM predictions [22], and to CasP code [23].

2. Experiment

Experiments to deduce stopping cross sections from backscattering spectra measurements were performed on copper (reference) and vanadium (material of interest) bulk samples in three different laboratories. The measurements at low primary energies were carried out at the AN-700 accelerator at the Johannes Kepler University in Linz, Austria, employing atomic beams of D^+ and H^+ in the energy range from 100 to 650 keV, and He⁺ from 250 to 550 keV. Backscattering spectra were recorded with a high-resolution surface barrier detector (SBD) placed at a scattering angle $\theta = 154.6^{\circ}$, with resolution (for He⁺ [24]) of 7 keV FWHM. Intermediate energies were covered using the 2-MV NEC-5SDH tandem accelerator at the University of Sao Paulo, Brazil, using H⁺ ions from 500 to 2000 keV, and He⁺ ions from 700 to 3000 keV primary energy, employing a passivated implanted planar silicon (PIPS) detector located at a scattering angle of $\theta = 170^{\circ}$, with resolution of 15 keV FWHM. The high energy measurements presented in this work were performed at the 5-MV NEC-15SDH-2 tandem accelerator at the Uppsala University, Sweden, employing H⁺ ions from 2000 to 2750 keV, and He $^+$ ions from 2000 to 6000 keV primary energy, with a PIPS detector located at a scattering angle of $\theta = 170^{\circ}$, and with resolution of 17 keV FWHM.

Since transition metals are chemically active, information on possible impurities in the employed vanadium and its chemical state are essential to warrant the accuracy of the results. To this aim, a piece of 25×25 mm with ≈ 1 mm of thickness, and 99.99% of nominal purity has been cut and sent to the laboratories after it has been analyzed by time-of-flight elastic recoil detection analysis ToF-ERDA at Uppsala University (details of the set-up and evaluation can be found in [25] and references therein). As a result, a thin oxide layer (V₂O₅) on the surface was observed, whereas in the bulk no pronounced impurities were found, apart from contaminations of carbon and oxygen, being presents on an almost constant level of 0.5 at.% and 1.5 at.%, respectively, within the depth range investigated by ToF-ERDA (≈ 500 nm). Other impurities could not be detected, i.e., their concentrations are below ≤ 0.1 at.%. The influence of the impurities has been taken into the account in the evaluation procedure, as described in Section 3.

To ensure that both samples were irradiated with the same charge, data acquisition was performed by repeated (5–10 times) alternating measurements of the Cu and V spectra, for an identical exposure time. The reference and interest samples were loaded on a rotatable sample holder wheel in such way that the surfaces of the sample during exposure were at the same distance from the detector, avoiding any problem with the solid angle. After measurements, the number of total counts for each spectrum was compared. From the scatter of the total

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