

DEPARTMENT OF PHYSICS
UNIVERSITY OF JYVÄSKYLÄ
RESEARCH REPORT No. 5/2002

HEAVY ION STOPPING POWER MEASUREMENTS WITH TRANSMISSION METHODS

BY
TOMMI ALANKO

Academic Dissertation
for the Degree of
Doctor of Philosophy



Jyväskylä, Finland
December 2002

DEPARTMENT OF PHYSICS,
UNIVERSITY OF JYVÄSKYLÄ
RESEARCH REPORT No. 5/2002

HEAVY ION STOPPING POWER MEASUREMENTS WITH TRANSMISSION METHODS

BY
TOMMI ALANKO

Academic Dissertation
for the Degree of
Doctor of Philosophy

*To be presented, by permission of the
Faculty of Mathematics and Natural Sciences
of the University of Jyväskylä,
for public examination in Auditorium FYS-1 of the
University of Jyväskylä on December 13, 2002
at 12 o'clock noon*



Jyväskylä, Finland
December 2002

URN:ISBN:978-951-39-9441-9
ISBN 978-951-39-9441-9 (PDF)
ISSN 0075-465X

Jyväskylän yliopisto, 2022

ISBN 951-39-1383-X
ISSN 0075-465X

PREFACE

The experimental studies of this thesis have been carried out during the years 1999-2002 at the Department of Physics, University of Jyväskylä.

I wish to express my sincerest gratitude to my supervisor, Professor Jyrki Räisänen for his guidance, teaching and encouragement during the course of this thesis. I want also to express my gratitude to Dr Ari Virtanen for all the help, encouragement and guidance.

I would like to thank all the members, former and present, of the Ion beam based materials physics and applications groups. Especially Mr Vesa Kyllönen, Mr Jani Hyvönen, Mr Pauli Laitinen and Mr Iiro Riihimäki, thanks for the fruitful and pleasant working atmosphere and stimulating conversations related to both physics and especially non-physics topics.

I am grateful for Dr Wladyslaw Trzaska for the guidance in the time-of-flight measurements. My warmest thanks go to all of the co-authors, collaborators and the whole staff of the Department for the friendly working atmosphere and pleasant cooperation.

Finally, I wish to thank my family, especially my wife Laura, for all the love and support.

Vantaa, November 2002

Tommi Alanko

ABSTRACT

In this thesis experimental stopping power values have been determined at the Department of Physics, University of Jyväskylä. The studies have been performed with three variations of the transmission techniques, two conventional transmission methods and the novel time-of-flight (TOF) based method that provides continuous stopping power curves. Time-of-flight based method presented in this thesis extends the point-by-point measurements traditionally carried out by conventional transmission methods to continuous stopping power curves. Also, a TOF based measurement setup for simultaneous determination of stopping powers for several different ions at wide energy range is presented.

The experimental measurements have been performed for several compound materials: havar, polyethylene terephthalate (mylar) and polycarbonate (lexan). Havar is a high strength metal alloy whereas mylar and polycarbonate are polymers. Materials have been selected because of their wide use in various applications, such as absorbers, windows, particle identification and radiation detection. Ions used in this study range from oxygen (O^{16}) up to iodine (I^{127}) with energies ranging from 0.35 to 5.0 MeV/u. The results acquired with different methods are compared with each other and previous literature and theoretical values. The results obtained with the different techniques agree well with each other and previous experimental values. Reasonable agreement can be seen also when compared with values obtained by various parameterisations.

CONTENTS

1	INTRODUCTION	1
2	PURPOSE AND STRUCTURE OF THIS STUDY	4
3	ENERGY LOSS OF IONS IN MATTER	6
3.1	Nuclear stopping power.....	7
3.2	Electronic stopping power	8
4	STOPPING POWER MODELS AND PARAMETERISATIONS...	10
5	EXPERIMENTAL METHODS FOR THE DETERMINATION OF STOPPING POWERS	16
6	TRANSMISSION METHODS IN STOPPING POWER MEASUREMENTS	19
6.1	The conventional transmission method.....	19
6.2	The Time-Of-Flight-Energy method.....	21
6.3	The foil thickness determination.....	25
6.4	Effective energy correction.....	27
7	SUMMARY	29
	REFERENCES	30

1 INTRODUCTION

The slowing down of energetic ions in matter has been a subject of interest already for a hundred years. The use of charged particles in a study of materials was first suggested shortly after its discovery at the beginning of the last century [Cur1900]. Later in 1909 Geiger and Marsden studied the spreading of alpha particle trajectories after passage through a thin foil [Gei09]. They concluded that 0.01% of the alpha particles were scattered backwards from the target, and such events had to be due to isolated single collisions from a heavy scatterer. Two years later in 1911 E. Rutherford was able to show theoretically that backscattering is really a single event [Rut11]. Since then energetic alpha particles have provided a new way to study materials. Before the availability of charged particle accelerators radioactive sources were used to provide the probing particles. Since 1930's accelerators have offered a wide variety of ions and energies to be used in ion beam applications.

The research of charged particle stopping power, i.e. energy loss in different materials has been shifting its focus during decades. From the thirties to the forties the interest was in the range of fission products in fission material. In the fifties the use of charged particle accelerators in nuclear physics needed information about the stopping power behaviour of ions. In the next decade the stopping power determination was a central problem in many nuclear and materials physics analysis methods. Also in many applications the accurate knowledge of stopping powers was needed. Use of ion beams in manufacturing the integrated circuits in the seventies brought again a new aspect to the research of stopping powers. Stopping powers were also needed in the

ion-solid interaction applications in fusion research. In the same decade research on the medical applications was begun and the ion beam influence on human tissue was again a new branch to stopping power research. The first accelerator for purely medical purposes was built in eighties for the Loma Linda University Proton Accelerator Facility, USA.

Table 1. Stopping power data application areas [Sig98].

Materials science and technology	Analysis, modification, radiation damage
Fusion technology	High energy density
Microelectronics devices	Development, fabrication, control
Radiation medicine	Diagnostics, therapy, radiation damage
Bio-, geo-, environmental science	Mass spectroscopy
Physics and chemistry	Cosmic radiation, radioactivity, accelerators, detectors

Nowadays accurate stopping power data are needed in many fields of research and applications (Table 1.), especially in materials characterisation and analysis. The largest uncertainty in many analysis methods is attributable to poorly known stopping power values. For example, depth information of analysed samples based on ion beam techniques is solely based on the slowing down, that is, stopping power of the material for used ion. One of the most interesting applications of ion stopping powers is the cancer treatment with ion beams. In this treatment the cancer tissue is bombarded with ion beam and ions lose the majority of their destructive energy only at a specific depth (Bragg Peak). If the stopping power of the used ion in tissue is well known the destructive effect of the slowing down process can be confined to the sick

tissue without harming the healthy tissue. Ion beam therapy is particularly appropriate if conventional radiotherapy presents an unacceptable risk, for example, in cases of cancer of the eye, the brain or the prostate.

Development of accurate theoretical stopping power models has been a subject of interest for decades. Nowadays stopping powers of light ions, mainly protons and alpha particles, can be predicted quite accurately (for example with Bethe-Bloch theory). This is due to many extensive experimental measurements that have enabled the progress in theoretical models. Prediction of stopping power for heavier ions and especially in non-elemental materials continues to be problematic. Most of the commonly used stopping power theories are actually parameterisations or semi-empirical calculations based on experimental data [Pau01]. None of the present theoretical procedures predict the stopping powers as well as well-conducted experimental methods do. Thus, there still clearly exists a need for experimental stopping power measurements.

In this study new experimental stopping power data are measured. Three transmission type approaches are used. Conventional setups employing direct and scattered beams, and a novel *time of flight* -based method that provides continuous stopping power curves, are employed. Also, a combination of this new method together with the versatile capability of the JYFL K130 cyclotron and ECR ion-source to produce so-called ion cocktails is used. By this way the continuous stopping power curves for several ions can be obtained simultaneously.

2 PURPOSE AND STRUCTURE OF THIS STUDY

The purpose of this study is two fold. Firstly, determine experimental stopping power data of elemental and compound materials for heavy ions. Secondly, develop and improve methods for accurate energy loss measurements.

This thesis is based on the following publications. The articles are referred with the Roman numerals in the text.

- I. T. Alanko, J. Hyvönen, V. Kyllönen, J. Räisänen, A. Virtanen, *Stopping powers of havar for 1.6, 2.3 and 3.2 MeV/u heavy ions*, Nuclear Instruments and Methods in Physics Research **B 161-163** (2000) 164-167.
[https://doi.org/10.1016/S0168-583X\(99\)00856-3](https://doi.org/10.1016/S0168-583X(99)00856-3)
- II. T. Alanko, J. Hyvönen, V. Kyllönen, J. Räisänen, A. Virtanen, *Stopping powers of havar and effective charge for 1.4-3.2 MeV/u ¹²⁷I-ions*, Nuclear Instruments and Methods in Physics Research **B 168** (2000) 139-143.
[https://doi.org/10.1016/S0168-583X\(99\)00832-0](https://doi.org/10.1016/S0168-583X(99)00832-0)
- III. T. Alanko, J. Hyvönen, V. Kyllönen, P. Laitinen, A. Matilainen, J. Räisänen, A. Virtanen, *Polycarbonate, Mylar and Havar stopping powers for 1.0-3.25 MeV/nucleon ⁴⁰Ar-ions*, Journal of Physics: Condensed Matter **13** (2001) 10777-10784.
<https://iopscience.iop.org/article/10.1088/0953-8984/13/48/304>

- IV. W.H. Trzaska, T. Alanko, V. Lyapin, J. Räsänen, *A novel method for obtaining continuous stopping power curves*, Nuclear Instruments and Methods in Physics Research **B 183** (2001) 203-211.
[https://doi.org/10.1016/S0168-583X\(01\)00728-5](https://doi.org/10.1016/S0168-583X(01)00728-5)
- V. T. Alanko, W.H. Trzaska, V. Lyapin, J. Räsänen, G. Tiourine, A. Virtanen, *Simultaneous wide-range stopping power determination for several ions*, Nuclear Instruments and Methods in Physics Research **B 190** (2002) 60-63.
[https://doi.org/10.1016/S0168-583X\(01\)01289-7](https://doi.org/10.1016/S0168-583X(01)01289-7)

Author's contribution in the articles

The author has planned and realized the measurements in articles I - III and participated in the manuscript preparation. The author is responsible for most of the data analysis of articles I and II and all of that in article III as well as most of the theoretical calculations in articles I - III. The author participated in the planning, measurements and writing of article IV and planned, participated in the measurements and carried out the data analysis and most of the writing of article V.

3 ENERGY LOSS OF IONS IN MATTER

When an energetic ion penetrates into a target it experiences a slowing-down force. The kinetic energy of the ion decreases and finally the ion stops. The resistive force causing this behaviour results almost completely from the electromagnetic interactions between the ion and the target atoms. The length of the ions path (*range*) and energy loss per unit length (*specific energy loss, stopping power*) is determined by the species of the ion, by the density and composition of the target, and by the energy of the ion itself. Even though the electromagnetic interaction processes have been well known for decades there is still no precise theory for stopping power. This is due to complicated electronic structure of the target ions, their possible bonds to other atoms and the lack of knowledge of the charge state of the ion inside the target i.e. its effective charge during interactions with target atoms.

The energy loss in the target material can be separated into two different processes. One process is the energy loss to bound or free electrons in the target, i.e. the electronic energy loss that is often called inelastic energy loss. The other process called nuclear energy loss is the energy loss in the interaction with screened or unscreened nuclei of the target atoms. This is often called elastic energy loss since it involves elastic collisions between the moving ion and the target atoms. The significant energy regions of electronic and nuclear energy losses in case of Ar-ions in gold target are presented in figure 1.

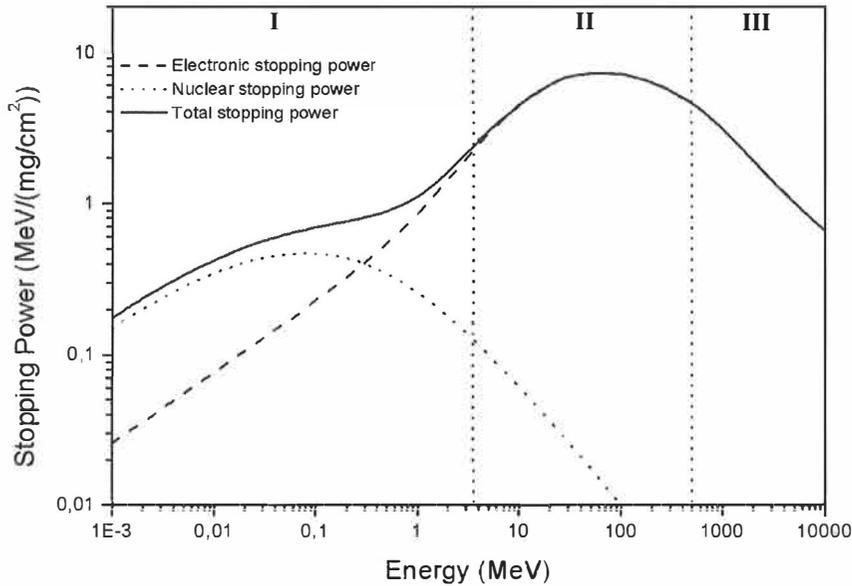


FIGURE 1. Stopping power of gold for ^{40}Ar -ions. The solid line presents the total stopping power that is the sum of the electronic stopping power (dashed line) and the nuclear stopping power (dotted line). The curves have been calculated by using the SRIM-2000 program [Zie01].

3.1 Nuclear stopping power

At lower energies (region I in fig. 1) the nuclear stopping power is dominant. This is because of the high probability of elastic collisions between the ion and target nucleus. When the energy is high enough, i.e. there are no screening effects with ion or target nucleus, the collisions can be regarded as scatterings from the Coulomb potential. The scattering cross section is proportional to the product of squares of atomic numbers of ion and target nucleus and inversely proportional to the square of the ion energy. Therefore nuclear stopping power decreases fast with the increasing energy (region I \rightarrow II, fig. 1). However, at very low energies (below 100 keV in case of Ar-ions in Au, fig. 1) the ion and target nucleus are screened by

their electrons and therefore the probability of scattering decreases. The screening effect increases with decreasing ion energy, as more electrons are able to follow the ion. The target nucleus screening also increases since with increasing interaction time the ion also sees the electron cloud surrounding the nucleus.

With the nuclear energy loss processes there is, unlike in the electronic processes, large changes in the direction of ion trajectories. Since the ion directions can vary much in individual collisions also the overall angular straggling is relatively more significant than in the electronic energy loss region at higher energies. In the nuclear energy loss processes also the target nucleus can move in crystalline materials from its lattice location if it gets enough recoil energy from the ion. These events change the structure of the material, i.e. produce damage. The damage profile caused by implanted ions follows very closely the nuclear stopping power curve.

3.2 Electronic stopping power

In the case of electronic stopping power at low ion energies the target atom electrons move much faster than the ion and the ions charge is almost neutral due to electronic screening. The collisions with electrons are mostly adiabatic without energy loss. It can be considered that target atom electrons have low drift velocities relative to the ion. During a collision a net energy is transferred which is proportional to the ion velocity [Lin61, Lin63]. Electronic energy loss is therefore proportional to ion velocity at low energies (region I in fig. 1).

At high energies (region III in fig. 1) when the ion velocity is higher than the velocity of its electrons in its innermost orbit, the ion is fully stripped and can then be considered as a point charge. In this region the energy loss is inversely proportional to the square of ion velocity. When the energy decreases the interaction time with target atoms increases and therefore also the energy loss increases. When the energy is low enough the ions start to pick up electrons and their effective charge becomes lower resulting in a decreasing energy loss (region II in Fig 1). It should be noted that in Fig. 1 there is a peculiar plateau in the total stopping power around 1 MeV where the nuclear stopping power decreases significantly and the electronic stopping power increases. In reality there is no such effect, it is due to the manner in which theoretical energy loss calculations treat nuclear and electronic energy loss processes independently from each other, which is not strictly speaking correct.

Since the electronic stopping power depends on the electronic structure of the target, different physical phases (gaseous, liquid and solid) and chemical bindings should and are known to contribute to stopping powers [Mat77, Zie88]. For water targets the difference between gaseous and solid phases can reach 30% for low energy light ions, <50 keV/u. The chemical binding effect is most significant around the stopping power maximum, usually contributing to few percent changes. For ion energies above 10 MeV/u these effects are estimated to have less than 1% impact on the stopping powers. For energies between 1-10 MeV/u both effects contribute with a few percent change to the stopping power, especially for light target atoms, i.e. $Z < 10$.

4 STOPPING POWER MODELS AND PARAMETERISATIONS

Niels Bohr presented the first attempt for a unified theory of stopping power [Boh13, Boh15] based on the theory of a nuclear atom with a heavy positively charged core. Bohr's original conclusion was that the energy loss in matter could be divided into two components: nuclear stopping and electronic stopping. He also correctly deduced that electronic stopping would be much higher than nuclear for energetic light ions. Twenty years later Bohr's model was greatly improved by Bethe [Bet30, Bet32] and Bloch [Blo33a, Blo33b]. They restated the problem of stopping power in the perspective of quantum mechanics on the basis of Bohr's concepts and Born's perturbation theory. The original Bethe-Bloch relativistic stopping formula can be written as:

$$-\frac{dE}{dx} = \frac{4\pi e^4 Z_2}{m_e v^2} Z_1^2 \left[\ln \frac{2mv^2}{\langle I \rangle} - \ln(1 - \beta^2) - \beta^2 + \Psi(Z_1) \right], \quad (1)$$

where Z_1 is the atomic number of the ion, Z_2 is the atomic number of the target atoms, m_e is the mass of the electron and m is the mass of the ion. Further, v corresponds to the velocity of the ion, β is the relative ion velocity (v/c), and $\langle I \rangle$ stands for the averaged excitation potential per electron. The Bethe-Bloch theory is valid when ions are completely stripped. This theoretical approach is still the basic method for evaluating the energy loss of light ions with energies above 1 MeV/u.

Detailed information and modern variations of the theory can be found in a review article by Ziegler [Zie99].

In 1963 Lindhard, Scharff and Schiøtt made the first unified approach to stopping and range theory. In this theory commonly called LSS-theory [Lin61, Lin63, Sig83] the electronic stopping cross section for a slow heavy ion is obtained as:

$$\varepsilon_e = 8\pi e^2 a_0 \xi_e \frac{Z_1 Z_2}{(Z_1^{2/3} + Z_2^{2/3})^{3/2}} \frac{v}{v_0}, \quad v < v_0 Z_1^{2/3}, \quad (2)$$

where the constant ξ_e is of the order of $Z_1^{1/6}$, and Z_1 and Z_2 are the atomic numbers of the projectile and the target atom, respectively, v is the velocity of the incident ion, a_0 the Bohr radius ($\approx 0.529 \times 10^{-10} m$) and v_0 the Bohr velocity ($\approx c/137$).

The electronic stopping power of a slow ion can thus be calculated from:

$$-\left(\frac{dE}{dx}\right)_S = N\varepsilon_e = 8\pi N e^2 a_0 \xi_e \frac{Z_1 Z_2}{(Z_1^{2/3} + Z_2^{2/3})^{3/2}} \frac{v}{v_0}, \quad v < v_0 Z_1^{2/3}, \quad (3)$$

where N is the atomic density of the target. Direct proportionality between stopping power and ion velocity can be noted. The formula is valid for all ions but only for low energies, i.e. energies below the stopping power maximum.

Many of the stopping power parameterisations use the so-called heavy ion scaling rule:

$$\frac{S_{HI}}{(Z_{HI} \gamma_{HI})^2} = \frac{S_p}{(Z_p \gamma_p)^2}, \quad (4)$$

where S_{HI} stands for the stopping power of heavy ion and S_p for the stopping power of proton at the same velocity. Z is the atomic number of the ion and γ its fractional effective charge. Since for protons the atomic number and effective charge is 1, the scaling rule can be simplified to:

$$S_{HI} = S_p Z_{HI}^2 \gamma_{HI}^2. \quad (5)$$

Northcliffe and Schilling simplified this scaling rule even more in their stopping power tabulation [Nor70]. They assumed that the effective charge of the ion is completely independent of the target material,

$$\frac{S_{l,A}}{S_{l,B}} = \frac{S_{q,A}}{S_{q,B}}, \quad (6)$$

where l and q are different ions with the same velocity and A and B are different target materials. In their tabulations Northcliffe and Schilling determined the stopping power of aluminium for twelve ions by using experimental stopping powers. They then compared the stopping powers of 16 different materials to those of aluminium and by using the scaling law they were able to construct the stopping powers for all ions in any elemental target. Accuracy of the scaling rule is

reasonable when taking into account the simplicity of the method, but not as good as with the newer parameterisations.

The ZBL-parameterisation by Ziegler, Biersack and Littmark [Zie85] treats the electronic and nuclear stopping powers separately. General expressions have been constructed for both components and they have an uncertainty of 5-20 % in most cases. This parameterisation has been combined with a commonly available computer program (TRIM, SRIM2000, SRIM2003) [Zie01, Zie02]. The ZBL-parameterisation is the most commonly used stopping power parameterisation in solid state physics. The high velocity heavy ion part of the parameterisation is based on the theory of Brandt and Kitagawa [Bra82] where the effective charge is defined as

$$\gamma = q + (1 - q) \frac{(v_0 / v_F)^2}{2} \ln \left[1 + \left(\frac{2\Lambda v_F}{a_0 v_0} \right)^2 \right], \quad (7)$$

where q is the degree of ionisation, γ is fractional effective charge of the ion, v_F is the Fermi velocity and Λ is the screening length of the ion which describes the dimensional changes to the electronic distribution as the degree of the ionisation changes. The degree of ionisation q is an empirical quantity and it is expressed as

$$q = 1 - \exp(0.803y_r^{0.3} - 1.3167y_r^{0.6} - 0.38157y_r - 0.008983y_r^2), \quad (8)$$

where y_r is effective ion velocity defined as

$$y_r \equiv \frac{v_r}{v_0 Z_1^{2/3}}, \quad (9)$$

where v_r is the ion velocity relative to the target electron velocity and Z_i is the ion's atomic number. The final stopping power is then calculated with equation (5), where S_p is the proton equivalent stopping.

The Hubert et al. parameterisation [Hub90] is based on the scaling of fully stripped He ion stopping powers instead of the proton ones. Heavy ion stopping power S_{HI} for a fixed medium and ion velocity combination is calculated by means of the scaling law in equation (5). The effective charge of the heavy ion is obtained by the parameterisation

$$\gamma_{HI} = 1 - X_1 \exp(-X_2 (E/A)^{X_3} Z_{HI}^{-X_4}), \quad (10)$$

where (E/A) is the energy per nucleon in MeV/u and X_1, \dots, X_4 are the best fit parameters given by Hubert et al. [Hub90]. The parameterisation for the heavy ion effective charge was deduced from a set of about 600 experimental stopping power values covering the energy range from 3 to 90 MeV/u for 15 incident heavy ions and 18 solid materials. Stopping powers and ranges are tabulated for all ions of atomic number $2 \leq Z \leq 103$ in the energy region of $2.5 \leq E/A \leq 500$ MeV/u for 36 solid materials. The parameterisation also presents a procedure for conversion to different ion and target isotope compositions. Other target media not listed in the tabulations can be obtained by interpolating in Z_2 .

Of the stopping models and parameterisations presented in this chapter, the ZBL-parameterisation provides the best accuracy over the entire energy range and different ion-target combinations. The authors of SRIM2003 report that for the latest version of program stopping power curves have overall uncertainty of 4.8% [Zie02]. The uncertainty of heavy ion stopping powers (Be-U) is 6.1% when in the earlier versions of the program (SRIM98, SRIM2000) it was 8.1%.

5 EXPERIMENTAL METHODS FOR THE DETERMINATION OF STOPPING POWERS

Stopping power values can be measured with several different manners that can be divided into categories according to the experimental method, see [ICR93] for detailed information about the methods. In the first category are the transmission methods in which thin foils are placed in the analyzed primary beam or in a scattered beam and the stopping power is obtained by measuring the energy loss of ions in the foil [Articles I - III]. In case of the scattered beam the intensity and the energy of the primary beam is first reduced by scattering either from a thick high-Z foil or by allowing the beam to be transmitted through a thin scattering foil and then placing the absorber foil in the transmitted beam at specific angle with respect to the incident beam. Also combinations of these methods are used. A variation of the transmission method used in this thesis, the TOF-E method [Article IV], belongs into this category (more in chapter 6). The purity, structure and homogeneity of the foil affect the measured stopping powers and the effect is more pronounced when thin foils are used. Even the possibility that the use of different experimental methods could lead to different stopping power values has been studied [Sem83].

A method based on the observation of the shift in the energy signal of elastically scattered particles is also popular for stopping power [Epp88] and foil thickness determinations. Particles are scattered from a smooth interface on top of which the actual stopping element is evaporated or from the front and back of the stopping

material itself. By observing the energy signal of scattered particles with and without the stopping layer or from the front and back of the stopping layer, respectively, the stopping power can be determined. In this method the problems with surface impurities are reduced since only the signals from the stopping layer are taken into account. Also, since the absorbing material is evaporated on a thick substrate, problems like cracking are much smaller than with self-supporting foils. One other way to determine the stopping power is to observe the intensity of the elastically back scattered particles from a thick target [All53]. In order to have reliable results with this method the total ion current to the target and detector solid angle must be known. The target surface must be very smooth and the elastic-scattering cross sections, including screening effects, must be well known. An advantage of this method is, that it does not need thin self-supporting foils, which may be difficult to fabricate (e.g. ice or chemically active substance).

The second category includes methods where the stopping power is not determined directly from the measured values. In the Inverted Doppler Shift Attenuation (IDSA) method [Ars98] the stopping power is determined by observing the Doppler shift of the gamma rays emitted by the slowing down ion in the decay of the excited state. Without a stopping material, the gamma-ray spectrum would be uniform over the energy limits of the Doppler shift. In the stopping material, the velocity of the moving nuclei at the time of gamma emission will be smaller, and the gamma-ray line shape will be narrower. How narrow the line shape is depends on the ratio of the slowing down time to the lifetime of the excited state [ICR93]. If the half-life of the excited state is known the corresponding Doppler shifted gamma ray peak can be reproduced with Monte Carlo simulations. When the

agreement between experimental and simulated peaks is good, the stopping power curve used in the simulations also agrees well with the real stopping power curve. This method enables the use of materials with arbitrary shape and structure. It is probably the most accurate method to study the chemical binding effects in solids and it can also be successfully used to measure stopping powers of liquids. The accuracy of the method is limited by the accuracy of the lifetime of the excited state. Furthermore, the method is best applicable under conditions where the ion loses a significant part of its energy within a period corresponding to the lifetime of the excited state. Disadvantage of the method is that due to this requirement not all nuclei can be used.

The use of nuclear reaction resonances [Sem83] also belongs into the second category and it is a good technique for obtaining stopping powers of such materials as thin foils and gases [Chi54]. In this method the gamma ray yield of a proton-induced reaction from a light backing material is first measured without the sample film and then with the film on the backing. The ion beam energy is fixed in both cases so that it corresponds to the reaction resonance energy at the backing surface. The difference in initial ion beam energies is the energy loss due to the interposing film. The technique is limited mostly to protons and alpha particles and naturally to the discrete resonance energies of the nuclear reactions.

6 TRANSMISSION METHODS IN STOPPING POWER MEASUREMENTS

6.1 The conventional transmission method

In the direct transmission type measurement the sample foil is exposed to the direct ion beam (figure 2). The detector is placed at 0° with respect to the beam direction, and the sample is placed perpendicular to the ion beam in front of the detector. The

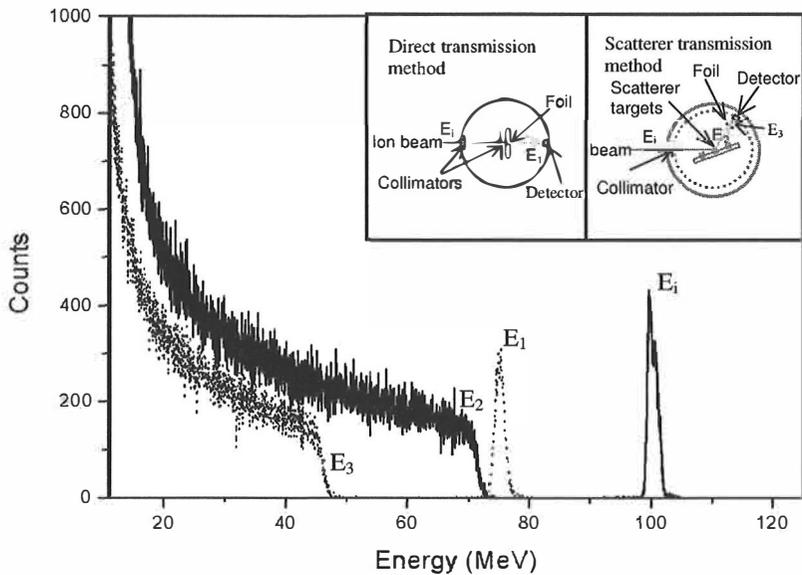


FIGURE 2. Typical direct and scatterer transmission method spectra obtained with 100 MeV ^{40}Ar ions for havar (measured separately). In the direct transmission method E_i corresponds to the initial ion energy and E_1 to the energy after the foil ($\Delta E = E_i - E_1$). In the transmission method, employing a scattering target, E_2 is the ion energy after scattering to an angle of 75° (relative to the beam direction) and E_3 the corresponding energy after the foil ($\Delta E = E_2 - E_3$). The insets show the experimental arrangements together with the appropriate energy labels [ARTICLE III].

energy loss is deduced from measurements of the beam energy before and after the foil [Articles I - III]. In the scatterer method thick targets of elemental materials are used first to scatter the particle beam (figure 2). Several different scattering angles and targets can be used. The combined change of the angle and scattering material enables measurements with several ion energies without tuning the accelerator. As with the direct method, the energy loss is deduced from the measurements of the beam energy before and after the foil [Articles II and III]. If the areal density of the foil is known (section 6.3), the transmission method allows the stopping power determination by simply dividing the energy loss by the areal density.

The main contributors in the uncertainty of the results are the foil thickness determination (section 6.3) and energy calibration. The simplest way to do the energy calibration is to use several mono-energetic ion beams of known energy and make the energy calibration accordingly. If one ion energy is used the detector can be calibrated by using scattered beams. When the ion beam is scattered from a known target into a known angle the energy corresponding to the backscattering edge can be calculated and energy calibration accomplished. When using low ion energies the pulse height defect (PHD) should be taken into account (section 6.2). Even in the high energy region the calibration should be done with care. In Article I the results were obtained using the same energy calibration for the different ions. This leads to increased uncertainty in the stopping power results. Applying an appropriate energy calibration (independent for each ion) to the data obtained in the study of article I, more accurate stopping power values are obtained (table 2) [Ala02].

Table 2. The effect of the energy calibration to the stopping power results of havar [Article I, Ala02].

Ion	<i>Same energy calibration for different ions</i>		<i>Separate energy calibration for different ions</i>	
	E (MeV/u)	S (MeV cm ² /mg)	E (MeV/u)	S (MeV cm ² /mg)
⁵⁶ Fe	1.62	23.9	1.69	20.1
⁸⁰ Kr	1.71	34.8	1.68	29.3
⁵⁶ Fe	2.24	34.0	2.17	28.3
⁸⁴ Kr	2.25	35.0	2.32	29.5
²⁸ Si	2.27	11.3	2.33	9.5
⁵⁴ Fe	3.21	24.0	3.27	20.2
³⁶ Ar	3.21	14.8	3.27	12.5
⁸⁴ Kr	3.24	34.6	3.29	29.1
³⁰ Si	3.31	10.0	3.36	8.4

6.2 The Time-Of-Flight-Energy method

The basic idea behind the Time-of-Flight-Energy (TOF-E) method [Article IV] is to use scattered particles with broad energy distribution and to perform the needed energy selection by using a time-of-flight (TOF) spectrometer placed before the sample foil while relying on energy measurements directly from a semiconductor detector. Figure 3 shows the schematic outline of the experimental setup. The ion beam is focused on the scattering target and part of the scattered beam, with the intensity adjusted to the counting rate capability of the setup, goes first through the TOF spectrometer and then through the sample foil. The foil is placed in a rotating target wheel where one slot is empty thus providing a curve without foil. The ions are finally stopped in the energy detector. Particles that traverse the sample foils have lower energy than ions reaching the energy detector without passing through the foil. Nevertheless, the TOF values are the same because the TOF spectrometer is placed before the foils. In this way it is possible to select from two continuous energy spectra the related events, i.e. with and without the foil, and calculate the

energy loss in the foil. Since the TOF-E spectrometer is able to separate different ions with the same energy, even ion beam cocktails [Cla90, Koi94] can be used if the energy loss is smaller than the energy difference between individual beam components, as is the case of article V. In a variant of this procedure the initial ion beam is used to generate secondary recoil beams from elements of compound targets for energy loss measurement [Zha99, Zha01]. In this manner stopping powers for several ions can be determined simultaneously.

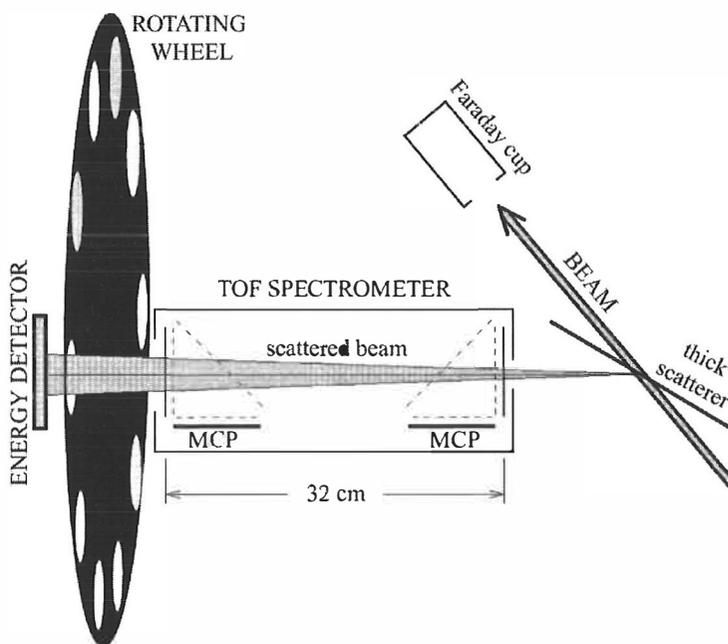


Figure 3. Schematic view of the TOF-E setup [ARTICLE IV].

Figure 4 shows typical TOF-E spectra obtained with and without the absorber. Thin slices are taken from the TOF axis and a projection to the energy axis is made. The projection represents the energy information of that particular TOF slice. When the projection is made for data obtained with and without the absorber,

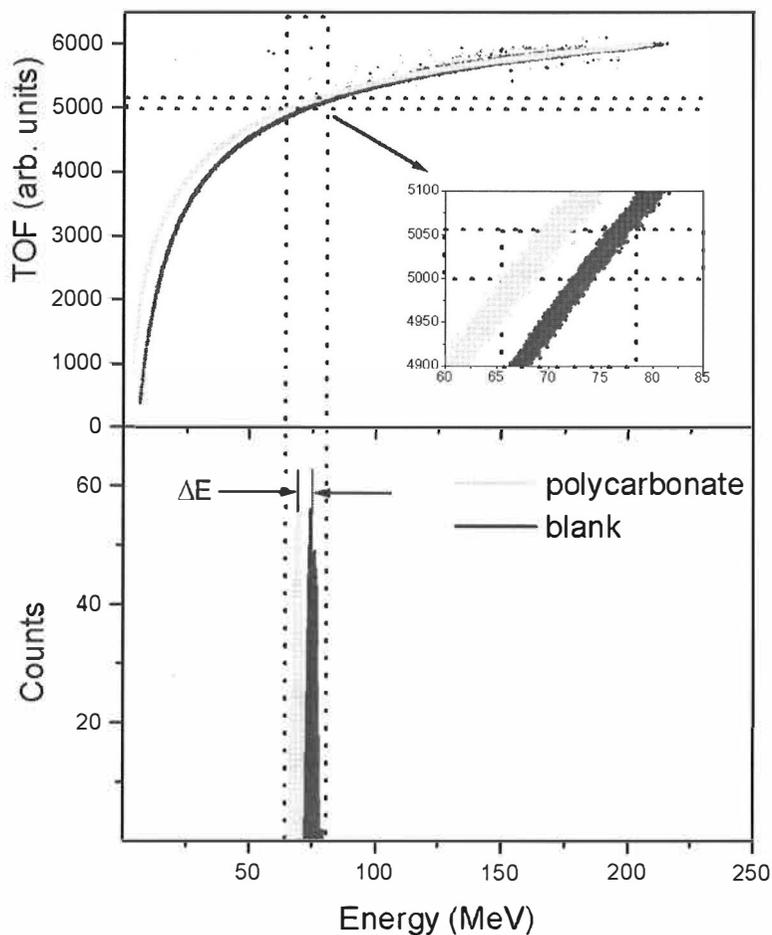


Figure 4. TOF-E spectra obtained with and without the absorber. By projecting thin slices from the TOF axis to energy axis, energy loss information can be obtained [ARTICLE V].

conventional transmission spectra with two peaks (E_i and E_f) are acquired (as in figure 2). The energy loss in the absorber is then obtained from the energy difference ($\Delta E = E_i - E_f$) of these two peaks. When the slices are taken throughout the whole TOF region by small steps, a continuous energy loss curve can be constructed [Articles IV and V].

The TOF-E method has several properties that make it better than conventional point-by-point transmission methods. The first and the most obvious is the obtaining of continuous stopping power curves. Virtually hundreds of data points can be acquired simultaneously over a wide energy range. If the statistics are good the differences in close lying data points are very small providing smooth stopping power curves [articles IV and V]. When beam cocktails are used, as in article V, the relative stopping power values for different ions are very accurate (0.2% uncertainty) even though absolute uncertainty values are larger (2.3%). If very thin foils are used, the uncertainty in areal density determination (section 6.3) is larger than with thicker foils thus leading to larger uncertainty in the stopping powers. Nevertheless, very thin foils are needed when the low energy part of the stopping power curve is measured. With the TOF-E method foils of the same material but with different thickness can be placed in the rotating target wheel. The energy of the ions from the accelerator is then chosen so that all measured energy loss curves overlap at some energy interval. The thickest foil gives best absolute stopping power values and the curves measured with the thinner foils can be normalised to the values obtained by the thickest foil thus providing stopping power information at the same relative uncertainty as with the best foil.

A well-known problem with semiconductor detectors is the so called *pulse height defect* (PHD) [Cli90, Com92, Mou78]. The energy response characteristics of a semiconductor detector for heavy ions are both non-linear and mass dependent because of the PHD. It is conventionally defined as the difference between the true energy of the heavy ion and that of an alpha particle required to produce the same

pulse height in the detector. The pulse height defect is more significant for heavier ions and lower energies.

In the TOF-E spectrometer there is the advantage that one can make a proper correction for the PHD without the need of knowing all the particular details for each phenomena [Trz02]. To accomplish this the time of flight spectrometer is first calibrated with a high precision time calibrator. Then the calibration offset can be acquired by comparing the calculated TOF value with the time peak position produced by the scattered high-energy particles. Ions lose some energy in the TOF stop detector and that lost energy must be taken into account in the energy detector calibration. To do that, a foil similar to the stop detector foil is placed in the rotating target wheel among the other foils. When measurements are done with a single foil (just the stop detector) and double foils (stop detector and foil in the target wheel), reliable corrections for the energy loss in the stop detector can be extracted. With this correction a direct relationship with the TOF and the semiconductor detector pulse height is obtained. The energy detector can then be calibrated with time of flight for each ion species separately. Detailed information about the measurement equipment and analysis can be found in ref. [Trz02].

6.3 The foil thickness determination

The main contributor to the stopping power uncertainty in the transmission methods is the error in foil thickness along with energy calibration inaccuracy [Articles I-V]. Perhaps the most common method to obtain the foil thickness is weighing of the foil with a known area (circular piece). Since the microbalances

have constant uncertainty the heavier, i.e. the thicker foils, can be weighed more accurately. Using a microscope with a lateral resolution of 0.1 mm, an ultimate resolution of 0.05 mm can be achieved for the diameter measurement [Bau87]. When uncertainties in the foil area and weighing procedures are combined, an uncertainty for the foil areal density is obtained. In the literature slightly different uncertainty values for the foil thickness have been reported, but they are generally of the order of 2-3 %. The foil thickness is usually expressed as an areal density (mg/cm^2) since often the exact density of the foil material is unknown. The areal density is sufficient if the experimental stopping powers are to be obtained in units of $\text{MeV}/(\text{mg}/\text{cm}^2)$.

Another commonly applied method is to measure the energy loss of alpha particles in the foil and then calculate the areal density by using literature stopping power values. The stopping power values thus obtained are relative values to the literature or calculated alpha particle stopping power data.

Sometimes foils used in measurements are not homogeneous and ions penetrating in different foil locations lose different amounts of energy. This leads to worse resolution and a growing uncertainty in the stopping power values. Solution to this problem is to use alpha particle energy loss for foil homogeneity measurements. Since the beam spot size employed in actual energy loss measurements is usually much smaller than the foil size convenient to weigh, the actual foil homogeneity can be mapped with alpha particle energy loss. The obtained average alpha particle energy loss corresponds to the measured areal density value.

6.4 Effective energy correction

The stopping power values, $-dE/dx$, are usually approximated with measured quantities $\Delta E/\Delta x$. This value of stopping power is then said to be valid at average ion energy $E_{av}=(E_i+E_f)/2$, where E_i and E_f are the initial and final energy of the ion. This approximation assumes that the stopping power is changing linearly with the energy and it is true as long as ion energy loss is small and the energy of the ion is well beyond the stopping power maximum. However, if the variation of $-dE/dx$ with energy is rapid or the used foil is thick ($\Delta E \ll E_i$ is not valid), the stopping power values should be assigned to a corrected intermediate value E_{eff} that depends on the shape of stopping power curve. In the literature it can be noted that the measured stopping power values are often assigned to E_{av} and the corrections are neglected even if the requirement $\Delta E \ll E_i$ is not fulfilled [Com66].

In the following a general treatment of the effective energy correction is presented. If the analytical expression of the stopping power curve is known (measured) or assumed (stopping power models and parameterisations), the correction for average energy E_{av} can be calculated [Chi54, All53]. Let us assume, that the stopping power curve has following expression:

$$-\frac{dE}{dx} = f(E). \quad (11)$$

When the initial and final energies of the ion are known the thickness of the foil can be integrated

$$\Delta x = \int_{E_i}^{E_f} \frac{dE}{f(E)}. \quad (12)$$

If E_{eff} is the true energy for which $(E_i - E_f)/\Delta x = -dE/dx$, and by using the notation

$$-\left. \frac{dE}{dx} \right|_{E_{eff}} = \frac{\Delta E}{\Delta x}, \quad (13)$$

we obtain from the equation (11)

$$\Delta x = \frac{\Delta E}{f(E_{eff})}. \quad (14)$$

If Δx in equations (12) and (14) is equal, we have

$$f(E_{eff}) = \frac{\Delta E}{\int_{E_i}^{E_f} \frac{dE}{f(E)}}. \quad (15)$$

From equation (15) the effective energy can be solved either analytically or numerically depending on the complexity of the stopping power function $f(E)$. The correction to the average energy is small (0.05%) when the energy loss is less than 20 % of the initial ion energy [And66]. When the energy loss increases the correction also increases [Lho84]. In article II the correction was 2.5 % on average for ^{127}I -ions losing 85-96 MeV (23-37 %) of their initial energy in a 1.845 mg/cm² thick havar foil.

7 SUMMARY

This thesis has concentrated on the study of heavy ion stopping powers and development of measurement methods. Experimental results are presented for several ions ($Z=8-53$) in three compound materials in the energy range 0.35-5.0 MeV/u. A new time-of-flight based measurement setup is presented along with the results of continuous stopping power curves, which are measured simultaneously for three different ion species. As seen, the new TOF based method for stopping power measurements is certainly very powerful. It is possible to acquire continuous curves within an energy range of several hundreds of MeV in the same time that it takes to measure a couple of individual points with conventional methods. Also, when using ion cocktails ambiguities connected with setup changes (energy calibration, detectors etc.) are removed and one gets very reliable relative values of stopping powers for different ions that are free from foil thickness determination. The results can be used in many fields of research and applications, especially in materials characterisation and analysis.

One of the future prospects is to study in detail how the pulse height defect influences at the lower energy region. Below the maximum of the stopping power curve the PHD is having more and more effect and it should be taken carefully into account. Co-operation in this and other matters related to the stopping power measurements with the TOF based method will be started with our colleagues in Sweden [Zha01].

REFERENCES

- [Ala02] T. Alanko, T. Sajavaara, W.T. Trzaska, J. Räsänen, to be published.
- [All53] S.K. Allison, S.D. Warshaw, *Rev. Mod. Phys.* **25** (1953) 779.
- [And66] H.H. Andersen, A.F. Garfinkel, C.C. Hanke and H. Sørensen, *Mat. Fys. Medd. Dan. Vid. Selsk.*, **35** no. 4 (1966).
- [Ars98] K. Arstila, Ph.D. Thesis, *Acta Polytech. Scand. Appl. Phys. Series No.* 220, 1998.
- [Bau87] P. Bauer, *Nucl. Instr. And Meth.* **B27** (1987) 301.
- [Bet30] H. Bethe, *Ann. Phys.* **5** (1930) 325.
- [Bet32] H. Bethe, *Z. Phys.* **76** (1932) 293.
- [Blo33a] F. Bloch, *Ann. Phys.* **16** (1933) 285.
- [Blo33b] F. Bloch, *Z. Phys.* **81** (1933) 363.
- [Boh13] N. Bohr, *Philos. Mag.* **25** (1913) 10.
- [Boh15] N. Bohr, *Philos. Mag.* **30** (1915) 581.
- [Bra82] W. Brand and M. Kitagawa, *Phys. Rev.* **B** (1982) 5631.
- [Chi54] A.B. Chilton, J.N. Cooper, J.C. Harris, *Phys. Rev.* **93** (1954) 413.
- [Cla90] D.J. Clark, G.J. Wozniak, *Nucl. Instr. And Meth.* **A295** (1990) 34.
- [Cli90] L. Cliché, S.C. Gujrathi and L.A. Hamel, *Nucl. Instr. And Meth.* **B45** (1990) 270.
- [Com66] J.R. Comfort, J.F. Decker, E.T. Lynk, M.O. Scully, A.R. Quinton, *Phys. Rev.* **150** (1966) 249.
- [Com92] D. Comedi and J. Davies, *Nucl. Instr. And Meth.* **B67** (1992) 93.
- [Cur1900] M. Sklodowska-Curie, *Compt. Rend. Acad. Sci.* 130 (1900) 76.

- [Epp88] C. Eppacher, D. Semrad, Nucl. Instr. And Meth. **B35** (1988) 109.
- [Gei09] H. Geiger and E. Marsden, Proc. R. Soc. **82** (1909) 495.
- [Hub90] F. Hubert, R. Bimbot, H. Gauvin, At. Data and Nucl. Data Tables **46** (1990) 1.
- [ICR93] ICRU (1993). International Commission on Radiation Units and Measurements. ICRU Report 49, Stopping Powers and Ranges for Protons and Alpha Particles.
- [Koi94] H. Koivisto, J. Ärje, M. Nurmi, Nucl. Instr. And Meth. **B94** (1994) 291.
- [Lho84] A. L'Hoir and D. Schmaus, Nucl. Instr. And Meth. **B4** (1984) 1.
- [Lin61] J. Linhard and M. Scharff, Phys. Rev. **124** (1961) 128.
- [Lin63] J. Linhard, M. Scharff and H.E. Schiøtt, Mat. Fys. Medd. Dan. Vid. Selsk., **33** no. 14 (1963).
- [Mou78] J.B. Moulton, J.E. Stephenson, R.P. Schmitt and G.J. Wozniak, Nucl. Instr. And Meth. **157** (1978) 325.
- [Nor70] L.C. Northcliffe, R.F. Schilling, Nucl. Data Tables **A 7** (1970) 233.
- [Pau01] H. Paul and Andreas Schinner, Nucl. Instr. And Meth. **B179** (2001) 299.
- [Rut11] E. Rutherford, Philos. Mag. **21** (1911) 669.
- [Sem83] D. Semrad, P. Bauer, F. Aumayer, P. Hubert, W Obermann, Nucl. Instr. and Meth. **218** (1983) 811.
- [Sig83] P. Sigmund, Phys. Scripta **28** (1983) 257.
- [Sig98] P. Sigmund, Nucl. Instr. And Meth. **B135** (1998) 1.
- [Trz02] W.H.Trzaska, V.Lyapin, T.Alanko, M.Mutterer, J.Räisänen, G.Tjurin and M.Wojdyr, Nucl. Instr. and Meth. **B195** (2002) 147.

- [Zha99] Y. Zhang, T. Winzell, H. Whitlow, Nucl. Instr. and Meth. **B159** (1999) 101.
- [Zha01] Y. Zhang, G. Possnert, H. Whitlow, Nucl. Instr. and Meth. **B183** (2001) 34.
- [Zie85] J.F. Ziegler, J.P. Biersack and U. Littmark, The Stopping and Range of Ions in Solids, Volume 1 of The Stopping and Ranges of ions in Matter, Pergamon Press, New York, (1985).
- [Zie88] J.F. Ziegler and J.M. Manoyan, Nucl. Instr. and Meth. **B35** (1988) 215.
- [Zie99] J.F. Ziegler, App. Phys. Rev. **85** no. 3 (1999) 1249.
- [Zie01] J.F. Ziegler and J.P. Biersack, SRIM-2000.40 computer code, private communication (2001).
- [Zie02] <http://www.srim.org/>