CALIBRATION OF THE CROSS SECTION OF THE $^{18}O(p,\alpha)^{15}N$ NUCLEAR REACTION AT $E_p = 1700-1775$ keV

P.F.A. ALKEMADE *, C.A.M. STAP, F.H.P.M. HABRAKEN and W.F. VAN DER WEG

Department of Atomic and Interface Physics, University of Utrecht, P.O. Box 80.000, 3508 TA Utrecht, The Netherlands

The differential cross section of the $^{18}O(p,\alpha)^{15}N$ nuclear reaction has been calibrated at proton energies between 1700 and 1775 keV and at a detection angle of 155°. For the calibration, two silicon reference samples and a nickel sample were partially oxidized in enriched ^{18}O. The absolute ^{18}O-content in the grown oxide films is measured with RBS by direct comparison with a Bi-implanted silicon calibration standard. The measured resonance curve is fitted with a split Breit–Wigner function. The maximal cross section of the resonance at 1766 keV is found to be 135 ± 5 mb/sr. Its width is 4.5 ± 0.3 keV.

1. Introduction

Calibration of nuclear reaction cross sections has originally received attention mainly by nuclear physicists. Also astrophysicists have shown their interest, especially in relation to the carbon–nitrogen–oxygen cycle within the core of stars [1]. Furthermore, nuclear resonances are applied by materials scientists and chemists to measure quantitatively or qualitatively low-Z, trace elements or to study chemical reactions in solids [2,3]. In order to measure absolute coverages of solid surfaces with low-Z elements, nuclear reaction analysis (NRA) has gained also much interest among surface scientists [4,5]. For these elements, NRA is one of the very few quantitative techniques with submonolayer sensitivities.

In 1980 Davies et al. [4] have calibrated resonances of the $^{16}O(d,p)^{17}O$ reaction with an accuracy of ±2%. Since then many low-Z, particle nuclear reactions have been calibrated accurately [5,6] against this reaction, mainly by use of the so-called frozen-gas technique. Apart from the $^{16}O(d,p)^{17}O$ reaction, also the $^{18}O(p,\alpha)^{15}N$ nuclear reaction is useful to study oxidation processes [8]. The cross section of the latter reaction at proton energies between 300 keV and 3000 keV, has been measured by Carlson already in 1961 [9]. He found a strong and narrow resonance at a proton energy (E_p) of 1766 keV and at 180° detection angle. Amsel has calibrated many resonances of the $^{18}O(p,\alpha)^{15}N$ reaction [10] and has applied the results in many studies of oxidation of solids [11]. Recently, the

broad resonance at $E_p = 700$ to 950 keV has been calibrated with an accuracy of 8% by Cohen et al. [12].

Because of the high value of its cross section, the resonance at $E_p = 1766$ keV is very suitable when very low amounts of oxygen have to be measured quantitatively [8]. However, because of its small width, the resonance can only be applied accurately if the energy distribution of the reacting protons is limited to less than a few keV. On the other hand, the small width and the low cross section at adjacent energies make this resonance very useful to measure depth profiles of oxygen in solids, as is done, e.g., in many studies by Calvert and coworkers [13].

This paper presents the result of a precise calibration of the cross section at $E_p = 1700$ keV to 1775 keV and at a detection angle of 155°. For this purpose, two silicon samples and a nickel sample, partially oxidized in ^{18}O-enriched oxygen are used. The amount of ^{18}O in the grown silicon oxide films is determined by Rutherford backscattering spectroscopy. Silicon samples are used because their RBS spectra can be compared directly with RBS spectra of a bismuth-implanted silicon calibration standard.

2. Experimental

The calibration is performed in a apparatus which consists of a UHV scattering chamber, a load lock and a transport mechanism. The scattering chamber is coupled to the 3 MV Van de Graaff accelerator at the University of Utrecht. The energy spread (standard deviation) of the incoming beam is less than 100 eV for 1.76 MeV protons [14]. Backscattered particles and reaction products are energy analyzed by a movable Si

---

* Present address: Department of Physics, University of Western Ontario, London, Ontario, Canada, N6A 3K7.

surface-barrier detector with an acceptance angle of about 0.0045 sr. The apparatus is described in detail elsewhere [15].

Two Si(111) samples (size 10×5 mm^2) are oxidized during four hours at about 1050 and 1150 K, respectively, in oxygen at a pressure of 10 Torr, containing about 50% ^{18}O, 50% ^{16}O and less then 5% ^{17}O. The thickness of the grown oxide films is about 50 nm. The absolute number of ^{18}O atoms per cm^2 is determined by Rutherford backscattering spectroscopy. Measured RBS spectra of both oxidized (Si/SiO$_2$-) samples are compared with spectra, measured under identical conditions, of a Bi-implanted silicon (Si(Bi)-) calibration standard [16]. Relative beam doses are determined by evaluation of the random spectrum height of silicon. The standard was puchased from the Joint European Research Center in Mol, Belgium.

The detector is placed at an angle of $155° \pm 0.25°$ with respect to the incoming beam. In the RBS measurement four different primary energies between 1.25 McV and 2.0 MeV are chosen.

The amount C_O of ^{18}O in a thin SiO$_2$ film is, expressed in known or measurable quantities:

$$\frac{C_O}{C_{Bi}} = \frac{A_O}{A_{Bi}} \frac{H_{Bi}}{H_O} \frac{\sigma_{Bi}}{\sigma_O}, \tag{1}$$

in which C_{Bi} is the number of Bi atoms per cm^2 in the Si(Bi)-standard: $(4.75 \pm 0.10) \times 10^{15}$/cm^2, A_O and A_{Bi} are the areas of the ^{18}O- and of the Bi-peak in a Si/SiO$_2$- and a Si(Bi)-spectrum, respectively, H_O and H_{Bi} are the random substrate heights (of Si!) of the Si/SiO$_2$- and the Si(Bi)-spectrum, respectively, and σ_O and σ_{Bi} are the oxygen and bismuth cross section, including screening by inner-shell electrons [17], for helium scattering.

Since the amount of bismuth in the Si(Bi)-standard is known, the relative RBS measurement enables us to determine the absolute amount of ^{18}O in the Si/SiO$_2$-samples without the use of the stopping power of silicon for He and without an accurate measurement of the absolute beam dose. Note that the determination of the ^{18}O-content is independent of the ^{16}O-content. Thus, knowledge of neither the thickness of the native oxide layer of the silicon sample, nor of the original isotope ratio in the oxidizing gas is necessary for the calibration.

In order to avoid reduction in the spectrum yield by channeling, the angle between the incident beam and the sample normal is $10°$. Furthermore, spectra are measured at many different azimuthal angles of the sample and averaged afterwards. The He$^+$ dose per Si/SiO$_2$-spectrum is about 80 μC and per Si(Bi)-spectrum about 20 μC.

Under identical geometrical conditions as described above, energy spectra of protons backscattered from the Si/SiO$_2$-samples and from the Si(Bi)-sample, together with the reaction products, i.e. α-particles, are measured. No use is made of a foil in front of the detector to stop the (elastically) backscattered protons. Moreover, backscattered protons are used to determine, once again, relative beam doses. It must be noted that the energy of the backscattered protons is too high to stop selectively protons and no α-particles in a foil.

The value for the nuclear reaction cross section σ_0 follows from the same eq. (1) when we substitute for A_O, A_{Bi}, H_O and H_{Bi} the values found in the proton scattering spectra. The bismuth cross section for proton backscattering [17], and, of course, the amount of bismuth and ^{18}O in the samples are known. Note that, since we apply the same equation, a possible systematical error in the amount of bismuth in the calibration standard will cancel in the calculation of the $^{18}O(p,\alpha)$-cross section. As a consequence of the energy loss of the protons in the (thin) oxide film, the cross section is averaged over all energies between the primary energy and the primary energy minus the energy loss in the oxide film.

The cross section of the $^{18}O(p,\alpha)^{15}N$ reaction is not calibrated at the position of the sharp peak but at two energies, belonging to the broad plateau slightly below the peak, i.e. at $E_p = 1746$ keV and 1756 keV. Furthermore, the resonance curve between $E_p = 1700$ keV and 1775 keV is measured qualitatively with one Si/SiO$_2$-sample as well as with a Ni(111)-sample, oxidized in 99% enriched ^{18}O. The nickel oxide layer has a thickness of 2 nm only and is used to minimize the effect of the energy loss in the oxide layer.

3. Results

Fig. 1 shows the sum of sixteen RBS spectra measured of one Si/SiO$_2$-sample together with the sum of sixteen Si(Bi)-spectra. The primary beam is 2 MeV He$^+$. For clarity, the substrate height of the Si(Bi)-spectrum is scaled with respect to that of the Si/SiO$_2$-spectrum.

Table 1 summarizes the results of the RBS measurement of the oxygen content of both samples. The area of the oxygen peaks is calculated by subtracting the silicon background from the spectrum. The height of the background is estimated by a quadratic interpolation. Consequently, the statistical error in the net oxygen peak is about $\pm 3\frac{1}{2}\%$. The ^{18}O-content, the $^{18}O/^{16}O$-ratio and the corresponding oxide film thickness, as measured using different primary energies, do not scatter more than the statistical errors. It is therefore concluded that nuclear $^{18}O(\alpha,\alpha)^{18}O$ resonances with cross section larger than about 6% of the Rutherford cross section, are not present below 2.0 MeV. It is noted that for other light elements, no deviation of more than 2% from the cross section for the scattering below 2 MeV has been observed [18].
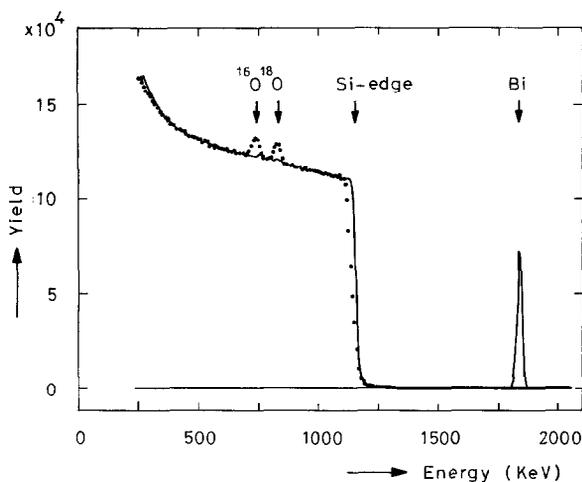
Fig. 1. RBS-spectra of a Si/SiO_2-sample and of a bismuth implanted silicon calibration standard, measured under identical experimental conditions. The ^{18}O-content of the Si/SiO_2-sample is calculated from the $^{18}O/Bi$ peak area ratio, the backscattering cross section ratio, and the known bismuth content of the calibration standard.

Table 1
Measured ^{16}O- and ^{18}O-content of both Si/SiO_2-samples. Film thicknesses are calculated using the density of bulk SiO_2. The quoted errors are only statistical.

Sample	Helium energy (MeV)	^{16}O-content (10^{15} cm^{-2})	^{18}O-content (10^{15} cm^{-2})	Ratio $^{18}O/^{16}O$	Film thickness (nm)
No. 1	1.25	115 ± 5	95 ± 5	0.83 ± 0.05	40
	1.5	116 ± 4	105 ± 4	0.91 ± 0.04	40
	1.75	123 ± 4	107 ± 4	0.87 ± 0.04	40
	2.0	111 ± 4	101 ± 4	0.91 ± 0.04	40
No. 2	2.0	231 ± 7	211 ± 7	0.91 ± 0.04	80

Table 2
Measured cross sections of the $^{18}O(p,\alpha)^{15}N$ reaction at $E_p = 1746$ and 1756 keV. The quoted errors are only statistical.

Sample	Proton energy (keV)	Cross section (mb/sr)
No. 1	1746	43.0 ± 1.5
No. 1	1756	48.0 ± 1.5
No. 2	1756	46.0 ± 1.8

The fluctuation in the substrate Si-height of individual spectra is about $\pm5\%$. This is mainly caused by channeling and partially by fluctuations in the measurement of the absolute beam dose, caused by irregularities in the secondary electron emission. It is concluded that the ratio in substrate height of the total Si/SiO_2-spectrum and that of the total $Si(Bi)$-spectrum, is within $\pm2\%$ equal to the ratio in the He beam dose. Neglecting the uncertainties in the $^{18}O(\alpha,\alpha)^{18}O$ cross section and in the bismuth content of the calibration standard, the oxygen content of sample no. 1 is determined with a precision of $\pm2.5\%$, and that of sample no. 2 with a precision of $\pm3.5\%$.

Table 2 summarizes the results of the calibration of the reaction cross section at $E_p = 1746$ and 1756 keV. The uncertainties (of $\pm3\frac{1}{2}$ and $+4\%$) in the quoted cross sections are caused by the uncertainty (of $\pm2.5\%$ or $\pm3.5\%$) in the oxygen content; by the statistical uncertainty in the area of the $^{18}O(p,\alpha)$-peak ($\pm2\%$); and by the uncertainty in the (relative) proton beam dose ($\pm1\%$). It is noted that no effort has been made to calibrate the exact energy of the resonance. Nevertheless, energies are determined with a relative accuracy better than ±0.5 keV. The 40 nm Si/SiO_2- and the 2 nm Ni/NiO-sample are used to measure qualitatively the resonance curve between 1700 keV and 1775 keV proton energy. The energy loss of 1750 keV protons in a 40 nm thick SiO_2 layer, using stopping power data of
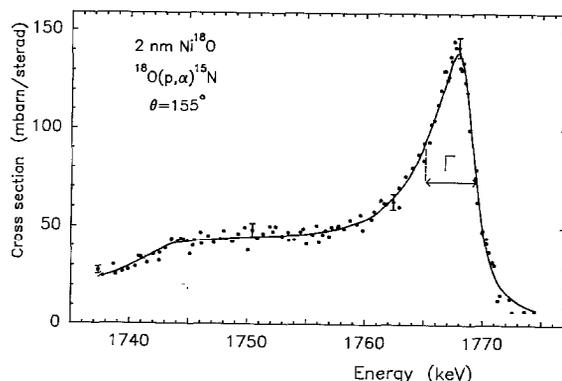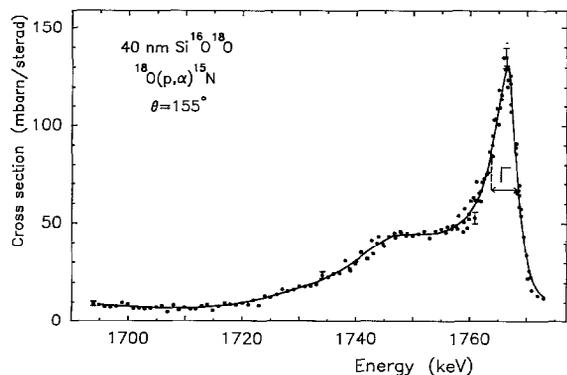

Fig. 2. Resonance curve at $155°$ detection angle, as measured with the Si/SiO_2 (left) and with the Ni/NiO (right) sample. The solid curve is the result of a fitting procedure of the data with a split Breit–Wigner function for the peak shape. Some typical errors are indicated.

silicon and oxygen for protons [19] and Bragg's rule for addition of elemental stopping powers, is 1.7 keV. The energy loss in a 2 nm NiO layer is only 0.3 keV. Therefore, the measured resonance curves are shifted and slightly broadened.

Fig. 2 shows the measured resonance curves. Different runs, but measured with the same sample, are combined. The drawn curve in the figures is the result of a fitting procedure of the data with a split Breit–Wigner function. The split Breit–Wigner function for the shape of a resonance curve, superimposed upon a background, is given by [12]:

$$\sigma(E) = \sigma_0 \left[C_i(E) + \frac{\frac{1}{4}\Gamma_i^2}{\frac{1}{4}\Gamma_i^2 + (E - E_0)^2} \right] \frac{1}{C_i(E) + 1},$$
(2)

in which E is the proton energy, σ_0 is the maximal cross section, E_0 is the energy at the maximum, Γ_i is the width of the peak, C_i is a linear function for the background. Both Γ_i and C_i may be different for the upper and lower energy side of the peak, $\Gamma_i = \Gamma_1$ for $E < E_0$ and $\Gamma_i = \Gamma_h$ for $E > E_0$.

Eq. (2) is convoluted with a block-function which represents the energy distribution of the reacting protons in the oxide. Effects of straggling and the energy spread of the primary beam are small and therefore not included.

The total width $\Gamma (= \frac{1}{2}(\Gamma_1 + \Gamma_h))$ of the fitted split Breit–Wigner functions, is 4.5 ± 0.3 keV in both measurements with the 40 nm Si/SiO$_2$-sample and with the Ni/NiO-sample. This figure is considerably larger than the mean energy loss (of 1.7 keV) of protons in the 40 nm SiO$_2$ film, let alone in the NiO layer (0.3 keV).

The value for the maximal cross section σ_0 is given by the calibrated cross section at $E_p = 1746$ keV and at 1756 keV. It is, according to the fitted split Breit–Wigner function, 132 ± 6 mb/sr in the measurement with the Si/SiO$_2$-sample and 138 ± 6 mb/sr with the Ni/NiO-sample. The final, or mean value, is 135 ± 5 mb/sr.

4. Discussion and conclusion

The resonance curve of the $^{18}O(p,\alpha)^{15}N$ nuclear reaction between $E_p = 1700$ keV and 1775 keV has been measured quantitatively at a detection angle of 155°. Two partially oxidized silicon samples with an oxide film thickness of 40 and 80 nm, respectively, and a nickel sample (with a 2 nm thick oxide) are used. The ^{18}O-content of the SiO$_2$-films is measured by RBS with an absolute accuracy of about ±3.5%, using a bismuth implanted silicon calibration standard.

The $^{18}O(p,\alpha)^{15}N$ reaction has a narrow and strong resonance at $E_p = 1766$ keV. The maximal cross section

is found to be 135 ± 5 mb/sr. The width Γ of the resonance is 4.5 ± 0.3 keV. Furthermore, there is a plateau between $E_p = 1740$ keV and 1760 keV. The cross section at the plateau ($E_p = 1753$ keV) is 45.0 ± 1.5 mb/sr.

The uncertainty of ±4% in the maximal cross section originates from counting statistics (±2.5%, mainly in the ^{18}O-peak in the RBS spectra of the Si/SiO$_2$-samples); the relative beam dose measurements (±1%); the conversion of the calibrated cross section (at the plateau) to the top (±1.5%); and the uncertainty in the elastic $^{18}O(\alpha,\alpha)^{18}O$ scattering cross section at energies below 2 MeV (±3%). It is noted that, at energies below 2 MeV, no deviation from the (screened) Rutherford cross section is expected [18]. The error of ±3% may therefore be too pessimistic.

Carlson [9] found a maximal cross section of 79 ± 7 mb/sr. At the plateau he has found a value of 41 ± 4 mb/sr. Amsel [10] found a maximal cross section of 115 mb/sr at a detection angle of 170°, which corresponds (according to the relative angular dependence as measured by Carlson [9]) to 95 mb/sr at 155°. His value at the plateau corresponds to 36 mb/sr. (No errors are given.) Our value for the plateau is higher than the above mentioned values. The difference is, however, not excessive. The $E_p = 1766$ resonance has been measured also, although only qualitatively, by Neild et al. [20]. They have found a ratio of 2.17 between the maximal and the plateau cross section. Carlson found a ratio of 1.92; Amsel of 2.64; and we have found a ratio of 3.02. The large variation in the ratios is, most likely, due to the poorer accuracy in the measurements by the above mentioned authors (in these cases only a few points are measured in the narrow peak).

The accuracy of ±4% in our calibration of the cross section is comparable to that achieved by Davies et al. in their calibration of the cross section of some (d,p)-, (α,α)- and (p,α)-reactions [5–7]. However, the technique of sample preparation and the method of measurement are quite different. Besides, the cross sections of the above mentioned reactions are generally an order of magnitude lower. A special advantage of using ^{18}O, with a natural abundance of only 0.2%, is the strong reduction of the risk for possible contamination by natural oxygen. Moreover, the $^{18}O(p,\alpha)^{15}N$ reaction in combination with e.g. a $^{16}O(d,p)^{17}O$ reaction or with Auger electron spectroscopy, may be useful to study different chemical surface or bulk reactions.

The $^{18}O(p,\alpha)^{15}N$ resonance at $E_p = 1766$ keV is one of the strongest known nuclear resonances [21]. Therefore, this resonance is very useful for quantitative surface analysis, as we have shown for oxygen adsorption on clean Ni(100) [8]. This resonance can also be applied for oxygen depth profiling, as is e.g. done in many studies by Calvert and coworkers [13]. The high energy of the protons, the relatively small width of the narrow peak,

and the low cross section at energies below 1730 keV and above 1775 keV [21] make this resonance very suitable to measure profiles to a depth of several micrometers.

Because of the relatively high energy of the backscattered protons, no foil can be used to stop selectively the elastically backscattered protons. In order to avoid too much pile up of detector pulses, the beam current and the acceptance angle of the detector must be limited. (Note, however, that the energy of the α-particles, 4 MeV, is so high that only the pile up of three or more pulses may cause a background below the α-peak in the spectra.)

The $^{18}O(p,\alpha)^{15}N$ reaction has also a broad resonance at 846 keV. The maximal cross section is, as calibrated recently by Cohen et al., [12] 60 ± 5 mb sr at $40°$ and 35 ± 2 mb/sr at $150°$. In this case, a foil can be used to stop selectively the backscattered protons. Consequently, high beam currents and large acceptance angles can be used e.g. for measuring absolute surface coverages. However, because the resonance is rather broad ($\Gamma = 47$ keV), it is not very useful for oxygen depth profiling. The reaction has also a very narrow resonance at 629 keV (width is only 2.0 keV FWHM). The maximum cross section is 100 mb/sr at $150°$ [21]. Because of the small width, the resonance is very suitable for depth profiling. However, the non-zero cross section at adjacent energies (the cross section is on the average about 10 mb/sr at energies between 500 keV and 800 keV [21]) and the broad resonances at $E_p = 846$ keV limit the depth range for which the $E_p = 629$ keV resonance can be used. Furthermore, since the cross section of this resonance varies so strongly with energy, the setting of the energy of the accelerator must be very stable and reproducible if this resonance is used for quantitative surface analysis. In conclusion, it depends on the details of an experiment which of the discussed three resonances can be applied best. Accurate data for the shape of all these resonances are required for accurate, quantitative oxygen related studies.

References

[1] H.-B. Mak, H.C. Evans, G.T. Ewan and J.D. MacArthur, Nucl. Phys. A304 (1978) 210.

[2] G. Amsel and D. Samuel, Anal. Chem. 39 (1967) 1689.

[3] A. Turos, L. Wielunski and J. Jelinska, Acta Phys. Polon. A 43 (1973) 657.

[4] J.A. Davies and P.R. Norton, Nucl. Instr. and Meth. 168 (1980) 611.

[5] G. Amsel and J.A. Davies, Nucl. Instr. and Meth. 218 (1983) 177.

[6] J.A. Davies, T.E. Jackman, H. Plattner and I. Bubb, Nucl. Instr. and Meth. 218 (1983) 141.

[7] J.A. Sawicki, J.A. Davies and T.E. Jackman, Nucl. Instr. and Meth. B15 (1986) 530.

[8] P.F.A. Alkemade, S. Deckers, F.H.P.M. Habraken and W.F. van der Weg, Surf. Sci. 189/190 (1987) 161.

[9] R.R. Carlson, C.C. Kim, J.A. Jacobs and A.C.L. Barnard, Phys. Rev. 122 (1961) 607.

[10] G. Amsel, Ann. Phys. (Paris) 9 (1964) 297.

[11] G. Amsel, J.P. Nadai, C. Ortega and J. Siejka, Nucl. Instr. and Meth. 149 (1978) 713.

[12] D.D. Cohen, A. Katsaros and D. Garton, Nucl. Instr. and Meth. B15 (1986) 555.

[13] See e.g. M.R. Taylor, J.M. Calvert, D.G. Lees and D.B. Meadowcroft, Oxid. Met. 14 (1980) 499.

[14] J.W. Maas, E. Somorjai, H.D. Graber, C.A. van den Wijngaart, C. van der Leun and P.M. Endt, Nucl. Phys. A301 (1978) 213.

[15] P.F.A. Alkemade, Ph.D. Thesis, Utrecht (1987).

[16] I.V. Mitchell, H.L. Eschbach, L. Avaldi and W. Dobma, Nucl. Instr. and Meth. 218 (1983) 91.

[17] J. L'Ecuyer, J.A. Davies and N. Matsunami, Nucl. Instr. and Meth. 160 (1979) 337.

[18] J.R. MacDonald, J.A. Davies, T.E. Jackman and L.C. Feldman, J. Appl. Phys. 54(4) (1983) 1800.

[19] J.F. Ziegler, J.P. Biersack and U. Littmark, The Stopping and Range of Ions in Solids (Pergamon Press, New York, 1985).

[20] D.J. Neild, P.J. Wise and D.G. Barnes, J. Phys. D5 (1972) 2292.

[21] L.C. Feldman and S.T. Picraux, in: Ion Beam Handbook for Material Analysis, eds. J.W. Mayer and E. Rimini (Academic Press, New York, 1977).