

## Applications of MeV Ion Beams, Nuclear Techniques and Computer Simulation to Surface Analysis of Materials

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**Abstract.** *This work involves surface analysis by nuclear techniques, which are essentially non-destructive, and computer simulation. The energy analysis method for nuclear reaction analysis is used. Energy spectra are computer simulated and compared to experimental data, giving target composition and concentration profile information. The simulations use, mainly, target parameterization and available nuclear data. The method is successfully applied to determination of a diffusion concentration profile of  $^{18}\text{O}$  in a thick oxide target. A uniform concentration profile of  $^{12}\text{C}$  is obtained for a very thin film. Uniform concentration profiles of  $^{16}\text{O}$  are also obtained from (d,p) and (d, $\alpha$ ) reactions along large depths. Elastic scattering is used for depth profiling of Al and Ag and Au thin films.*

**Keywords:** *Surface Analysis, Nuclear Reaction Analysis, Carbon, Oxygen, Elastic Scattering, Computer Simulation*

### 1. Introduction

A broad range of surface analysis techniques has been developed, involving e.g. ion, electron and photon beams interacting with a solid target. The techniques are, generally, complementary and provide target information for depths near the surface. Both nuclear and non-nuclear techniques have been available. Nuclear techniques, which are essentially non-destructive, provide for analysis over a few microns close to the surface giving absolute values of concentrations of isotopes and elements. Their main applications have been in areas such as scientific, technologic, industry, arts and medicine, using MeV ion beams [1-7]. Nuclear reactions permit tracing of isotopes with high sensitivities. We use ion-ion reactions and the energy analysis method where, at a conveniently chosen energy of the incident ion beam, an energy spectrum is recorded of ions from the reaction, coming from several depths in the target.  $\Theta_L$  is the laboratory detection angle and  $\Theta_R$  is the target rotation angle. Such spectra are computationally predicted, giving target composition and concentration profile information [4-8]. Elastic scattering is a particular and important case. A computer program has been developed in this context, mainly for flat targets [4-7]. The non-flat target situation arises as an extension [9]. Imaging techniques are important for checking target surface topography. Applications of the method are made to depth profiling of light nuclei e.g.  $^{18}\text{O}$  and  $^{12}\text{C}$ , for a thick target and a very thin film, using  $^{18}\text{O}(p,\alpha_0)^{15}\text{N}$  and  $^{12}\text{C}(d,p_0)^{13}\text{C}$  reactions, respectively.  $^{16}\text{O}$  nuclei are profiled by (d,p) and (d, $\alpha$ ) reactions for a thick target. The usefulness of elastic scattering is also shown. Scanning electron microscopy is used as an imaging technique to check target surface topography.

The rest of the paper is structured as follows: Section 2 is about the experimental details i.e. the measurement setup and procedure. Results and discussion are given in Section 3. Conclusions are drawn in Section 4.

## 2. Subject and Method

The experimental arrangement has been mentioned [5]. For an incident beam of accelerated ions, and an ion-ion nuclear reaction experiment, ion detection from the sample at laboratory angles  $\Theta_L$  of  $135^\circ$  and  $165^\circ$  used silicon surface barrier detectors, chosen as suitable for the reaction and energy ranges involved. Spectral data were acquired as counts per channel versus channel number, usually for ion beam perpendicular incidence (rotation angle  $\Theta_R=0^\circ$ ), through charge preamplifiers, amplifiers and analogue to digital converters providing for pulse pile-up rejection, data interfaces and an on-line computer equipped with data acquisition software. Following energy calibration of the spectra, spectral yields as counts per unit energy versus energy were obtained. The following samples were used as targets for acquisition of charged particle spectra: (1) an austenitic steel (20/25/Nb steel) sample, labelled S1, which was obtained by high temperature sequential oxidation first in  $C^{16}O_2$  gas and then in  $C^{18}O_2$  gas for 40 hours; an  $^{18}O$  diffusion erfc concentration profile [4] was expected; scanning electron microscopy has shown a reasonably flat oxide; (2) a thick flat sample of quartz ( $SiO_2$ ), labelled S2; it had a very thin surface film of carbon with thickness  $X_1=0.062 \mu m$  [7]; (3) a flat sample, labelled S3, obtained by sequential vacuum deposition of Ag and Au onto an Al thick flat substrate (Al/Ag/Au); film thicknesses of  $X_1=0.0648 \mu m$  and  $X_2=0.1333 \mu m$  were initially expected for Au and Ag, respectively.

## 3. Results and Discussion

Computer simulated spectra mainly take into account: target parameters, such as composition and concentration profiles; energy spread of the incident ion beam; geometric factors and target rotation; stopping power; differential cross section; energy straggling; detector resolution [5]. An option permits calculation of effects from: small forward angle multiple scattering; incident beam size and angular divergence; detector angular aperture. Gaussian straggling is considered, based on Bohr theory [10]. Lindhard-Scharff theory [11] is the main option. In building a basic spectrum, with ingoing ions penetrating the target and outcoming ions travelling towards the detector, we calculate mainly: ingoing and outcoming energy distributions, dependence of the yield on detection system resolution and final yield versus energy [6]. By varying target parameters, for visually fitting simulated spectra to experimental data, target composition and concentration profiles are obtained. In simulating spectra for nuclear reactions we used published stopping power [12-15] and differential cross-section data [5, 16-17]. Similarly for elastic scattering, where Rutherford differential cross-sections applied.

The oxidized steel sample containing  $^{18}O$ , S1, was analysed through a proton beam at a bombarding energy  $E_p=1.78$  MeV, an energy slightly above the resonance energy at 1.766 MeV of the  $^{18}O(p,\alpha_0)^{15}N$  reaction, and  $165^\circ$ . A good computer fit to data was obtained, as shown in Fig. 1. An  $^{18}O$  distribution due to diffusion was found, described by a complementary error function [4] with a diffusion coefficient  $D=2.7 \times 10^{-14} \text{ cm}^2 \text{ s}^{-1}$ . This value was also confirmed from analysis of the spectrum obtained at  $165^\circ$  in another detector, symmetrically located with respect to the incident beam. This diffusion coefficient is higher than the value determined from the resonance method of analysis using the 1.766 MeV resonance [18], as the method of the present work permits higher depth resolution.

The quartz sample, S2, was analysed with a deuteron beam at  $E_d=0.993$  MeV and  $135^\circ$ . The bombarding energy was chosen to obtain insignificant yields of deuteron induced reactions in  $^{28}Si$ . For the  $(d,p_0)$  reaction in  $^{12}C$ , published differential cross-section data were used [5]. A good computed fit to data was obtained. A very thin surface film of  $^{12}C$  was found with

uniform concentration and thickness  $X_1=0.061 \mu\text{m}$ . A uniform step concentration profile distribution of  $^{16}\text{O}$  was found in the quartz substrate. The corresponding thickness parameters  $X_2$  used in the predictions were, by descending order,  $5.49 \mu\text{m}$  for  $(d,p_0)$ ,  $5.20 \mu\text{m}$  for  $(d,p_1)$  and  $3.39 \mu\text{m}$  for  $(d,\alpha_0)$ . Details of the fit are shown in Fig. 2, for the spectral shape of the  $^{16}\text{O}(d,p_1)^{17}\text{O}^*$  reaction peak.

The sample Al/Ag/Au, S3, was analysed with a  $(^4\text{He})^+$  ion beam at  $E_\alpha=1.5 \text{ MeV}$ , and  $165^\circ$ . This initial fit suggested that a small percentage of Ag might be incorporated in the Au film. A further analysis at a higher bombarding energy,  $E_\alpha=2.9 \text{ MeV}$ , permitted to conclude that the sample is better described by a structure Al/Ag/(Au,Ag) where the surface film consists of a mixture of Au and Ag as a result of the sequential vacuum evaporation. An improved computer fit to the new data was obtained as shown in Fig. 3. Step concentration profiles were used for Au and Ag in the surface layer with  $X_1=0.060 \mu\text{m}$  and for Ag in the middle layer with  $X_2=0.126 \mu\text{m}$ . These thickness values are close to the nominal values initially estimated. A relative atomic density  $C_{\text{Ag}}/C_{\text{Au}}$  of 5.6% in the mixture was used. Details of the new fit are shown in Fig. 4 for the Au and Ag shapes.

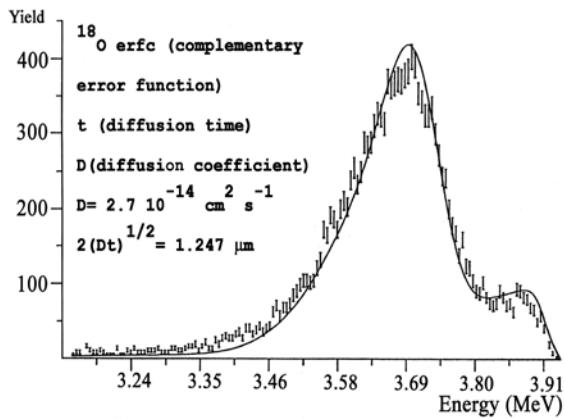


Fig. 1. Computed fit to the  $^{18}\text{O}(p,\alpha)^{15}\text{N}$  reaction data from the oxidized steel target, S1, for  $E_p=1.78 \text{ MeV}$ ,  $\Theta_L=165^\circ$ .

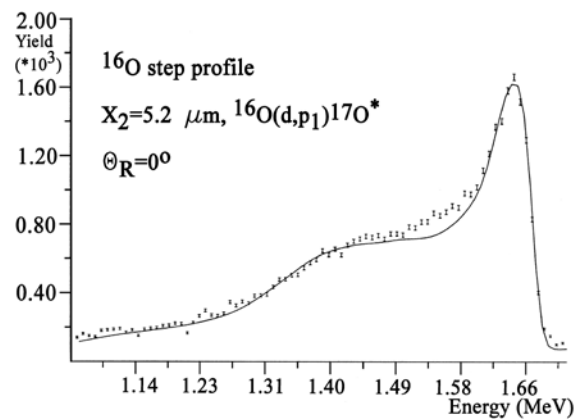


Fig. 2. Computed fit to data of the  $^{16}\text{O}(d,p)^{17}\text{O}^*$  reaction peak from the quartz target, S2, for  $E_d=0.993 \text{ MeV}$ ,  $\Theta_L=135^\circ$ .

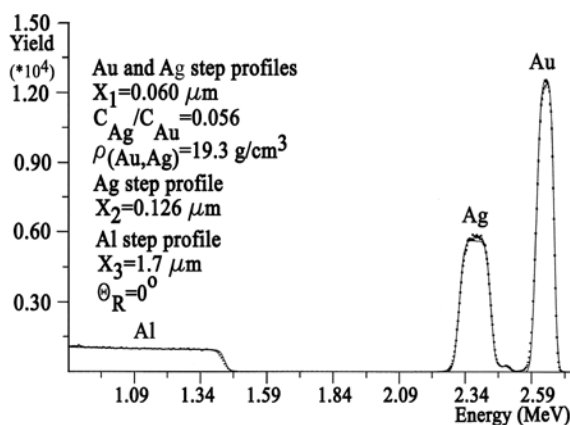


Fig. 3. Computed fit to the elastic scattering data from the Al/Ag/Au sample, S3, for  $E_\alpha=2.9 \text{ MeV}$ ,  $\Theta_L=165^\circ$ .

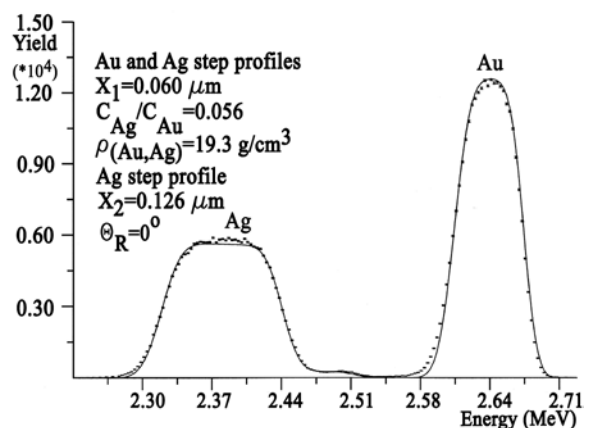


Fig. 4. Computed fit to the elastic scattering shapes from Ag and Au in the Al/Ag/Au sample, S3, for  $E_\alpha=2.9 \text{ MeV}$ ,  $\Theta_L=165^\circ$ .

#### 4. Conclusions

The present work has given positive results in surface analysis by nuclear reactions, for depth profiling of  $^{12}\text{C}$ ,  $^{16}\text{O}$  and  $^{18}\text{O}$  nuclei, and elastic scattering. The predictions obtained by computer simulation have given good descriptions of experimental spectra for relevant profiling of:  $^{12}\text{C}$  for a very thin film;  $^{16}\text{O}$  along large depths in a thick target;  $^{18}\text{O}$  in a thick oxidized steel target; thin films of Ag and Au deposited onto a thick Al substrate. Nuclear techniques have shown to be highly powerful and important analytical tools in this context. SEM was useful for checking surface topography. The results presented would be very difficult to obtain by non-nuclear techniques.

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#### References

- [1] Tesmer JR, Nastasi M. (Eds.) Handbook of Modern Ion Beam Materials Analysis, Materials Research Society, Pittsburgh, PA, 1995.
- [2] Amsel G, Battistig G., Nucl. Instr. and Meth. B 240 (2005) 1.
- [3] Calvert JM, Derry DJ, Lees DG. J. Phys. D: Appl. Phys. 7 (1974) 940.
- [4] Pacheco de Carvalho JAR. Ph. D. Thesis, University of Manchester, England, 1984.
- [5] Pacheco de Carvalho JAR, Reis AD. Nucl. Instr. and Meth. B 266, 10 (2008) 2263.
- [6] Pacheco de Carvalho JAR, Reis AD. Bol. Soc. Esp. Ceram. V. 47, 4 (2008) 252.
- [7] Pacheco de Carvalho JAR, Ribeiro Pacheco CFFP, Reis AD. Nucl. Instr. and Meth. B 269, 24 (2011) 3054.
- [8] Barradas NP, Arstila K, Battistig G, Bianconi M, Dytlewski N, Jeynes C, Kótai E, Lulli G, Mayer M, Rauhala E, Szilágyi E, Thompson M. Nucl. Instr. and Meth. B 262 (2007) 282.
- [9] Pacheco de Carvalho JAR, Pacheco CFFPR, Reis AD. Bol. Soc. Esp. Ceram. V. 49, 1 (2010) 23.
- [10] N. Bohr, Mat. Fys. Medd. Dan. Vid. Selsk. 18 (8) (1948) 1.
- [11] J. Lindhard, M. Scharff, Mat. Fys. Medd. Dan. Vid. Selsk. 27 (15) (1953) 1.
- [12] D. C. Santry, R. D. Werner, Nucl. Instr. and Meth. 188 (1981) 211.
- [13] C. Williamson, J. P. Boujot, J. Picard, C. E. A. Rep. N° 3042, Centre d'Études Nucleaires de Saclay, 1966.
- [14] H. H. Andersen, J. F. Ziegler, in: J. F. Ziegler (Ed.), The stopping and ranges of ions in matter: Hydrogen stopping powers and ranges in all elements, vol.3, Pergamon Press Inc., Oxford. 1977.
- [15] J. F. Ziegler, in: J. F. Ziegler (Ed.), The stopping and ranges of ions in matter: Helium stopping powers and ranges in all elements, Vol.4, Pergamon Press Inc., Oxford, 1977.
- [16] G. Debras, Thesis, Facultés Universitaires de Namur, Belgium, 1977.
- [17] G. Amsel, Thesis, University of Paris, Faculty of Science, Orsay, 1963.
- [18] Reich DF. Ph. D. Thesis, University of Manchester, England, 1982.