The development of a time of flight spectrometer for LARN

Mathot, Gilles; Terwagne, Guy

Published in:
Nuclear Instruments and Methods in Physics Research, Section B: Beam Interactions with Materials and Atoms

DOI:
10.1016/S0168-583X(02)00453-6

Publication date:
2002

Document Version
Peer reviewed version

ELSEVIER

Nuclear Instruments and Methods in Physics Research B 190 (2002) 190–194

NIM B
Beam Interactions with Materials & Atoms

www.elsevier.com/locate/nimb

The development of a time of flight spectrometer for LARN

G. Mathot *, G. Terwagne

Laboratoire d'Analyses par Réactions Nucléaires, Facultés Universitaires Notre-Dame de la Paix, rue de Bruxelles 61, B-5000 Namur, Belgium

Abstract

An elastic recoil detection time of flight system to depth profile light elements has been developed on ALTAÏS, the new Tandetron accelerator at LARN. The detector mounted at 45° from the beam axis consists of two isochronous electron detectors for the timing signal (START and STOP) and a 450 mm^2 heavy ion PIPS detector that detects the energy of the recoil atoms. The 2MV Tandem accelerator provides heavy ion beams with a maximum energy of 16 MeV depending on the charge transfer efficiency of the gas exchange canal located in the middle of the machine. A large variety of primary ion beams like ^{28}Si, ^{35}Cl, ^{63}Cu, ^{127}I or ^{197}Au can be produced with the SINIX heavy ion source and accelerated on the target. Typical current around 1 nA can be obtained. The energy transfer to the recoil atoms is typically in the MeV range and depends on the mass and the energy of the projectile. Some secondary effects like the energy loss in the carbon foils of the timing detector but also in the entrance window of the energy detector should not be neglected if we try to depth profile light elements with this technique. Time resolution of about 1 ns for the electrons detectors is suitable to obtain 1 amu mass resolution. Some examples of applications will be developed in this paper.

PACS: 25.70.Bc
Keywords: ERD; TOF; Depth profiling; Light elements

1. Introduction

Elastic recoil detection time of flight system (ERD-TOF) is a simple and fast method for quantitative multi light elements ($Z < 10$) depth concentration profiling [1]. It can be use to analyse the first 200 nm from the surface of metals, semiconductors, glass or polymers. The results obtained by this technique give information on light elements which are difficult to measure with RBS. This technique can be developed on small accelerators which provide heavy ions of about 10 MeV [2]. In this case, the reaction cross-sections are high and a depth resolution of 2 nm and a mass resolution of 1 amu can be achieved.

An ERD-TOF system to depth profile light elements has been developed on ALTAÏS, the new 2MV Tandetron accelerator at LARN, which provides heavy ion beams with energy up to 16 MeV. A large variety of primary ion beams like ^{28}Si, ^{35}Cl, ^{63}Cu, ^{127}I or ^{197}Au can be produced with the SINIX heavy ion source and accelerated on the target. The resolution of the detection system has

* Corresponding author. Fax: +32-81-725474.
E-mail address: gilles.mathot@fundp.ac.be (G. Mathot).

0168-583X/02/$ - see front matter
PII: S0168-583X(02)00453-6

been improved to obtain 1 amu mass resolution for recoil atoms of $Z < 10$. Quantitative analysis of light elements is performed using the interactive spectrum synthesis method.

2. LARN ERD-TOF system developed on ALTAÏS

The TOF detector mounted at 45° from the incident beam axis (Fig. 1) is composed of two isochronous electrons detectors [3] separated 76 cm for the timing signal and a 450 mm^2 heavy ion PIPS detector which detects the energy of the recoil atoms, placed at 96.5 cm from the beam spot of the specimen of interest. The solid angle of the energy detector is 0.48 msr. Circular collimators of 1.5 and 18.0 mm of diameters are placed in front of the T_1 and T_2 timing detectors showing a solid angle of 0.28 and 0.36 msr respectively. It is quite clear that the collimator placed in front of the T_1 timing detector limit the solid angle of the whole system, which is aligned very carefully.

The spectra are building up by measuring in coincidence the total energy E of the detected particles and the time needed by particles to pass between two timing detectors (T_1 and T_2) separated by a length L. The time signal is due to electrons emission when the recoil atom passes through a thin carbon foil (5 μg/cm^2). The timing signal from the T_1 detector is delayed in order to start the coincidence when an event is observed in the energy detector. With this configuration, the faster particles appear in the higher time channels.

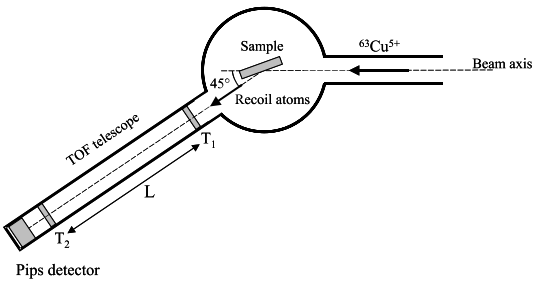

Fig. 1. Schematic of ERD-TOF system. The detector mounted at 45° from the incident beam axis consists of two isochronous electrons detectors separated 76 cm for the timing signal and a 450 mm^2 heavy ion PIPS detector which detects the energy of the recoil atoms, placed at 96.5 cm from the beam spot of the specimen of interest.

In a typical coincidence spectrum, the recoil atom signals are well separated. This effect is due to the mass dependant velocity of the same energy detected recoil atoms:

$$v = \sqrt{\frac{2E}{M}}$$

where v is the velocity, M the mass and E the energy of the recoil atom.

The mass of the detected particle can be calculated using:

$$M = 2E\left(\frac{T}{L}\right)^2$$

where T is the time required by the particle to pass between two time detectors separated by a length L and E is the measured energy of the particle.

Time resolution has been measured using the time spectrum from a Silicon target analysed with 16 MeV $^{63}Cu^{8+}$. Time resolution of 0.700 ns was obtained by fitting the leading edge with a gaussian cumulative distribution function. The detection efficiency has not been measured yet. For 5.486 MeV α-particles less than 50% of the particles are detected. We are still working for high detection efficiency and better time resolution.

3. Mass resolution

3.1. Coating preparation: mixture of $Ti^{14}N$ and $Ti^{15}N$

In order to verify the mass resolution of ERD-TOF system, we prepared coating of TiN which contains an equal mixture of ^{14}N and ^{15}N onto polished and cleaned low carbon steel. The coating was produce by DC-magnetron sputtering [4]. In the Ar sputtering gas, we have introduced the reactive gas, which consists of an equal mixture of $^{14}N_2$ and $^{15}N_2$. The DC-magnetron sputtering vacuum chamber was equipped with a titanium target disc of 19.6 cm^2 area and a 400 l/min turbomolecular pump. The pressure during deposition

and plasma power were maintained at 0.29 Pa and 230 W respectively. The deposition time was adjusted to obtain coating thickness of 210 nm. The deposited film should contain the same amount of ^{14}N and ^{15}N and was used to improve the mass resolution of our ERD-TOF system.

3.2. ERD spectrum

Fig. 2 shows the coincidence between time and energy detectors for the TiN coating deposited on low carbon steel. The incident beam was 12 MeV $^{63}Cu^{5+}$. The recoil spectra of ^{14}N and ^{15}N are mass separated, which means that we have achieved 1 amu mass resolution ($Z < 10$) and that the time resolution of the ERD-TOF system is about 1 ns for the electrons detectors. If we compare the experimental mass curves with the theoretical one (solid lines in Fig. 2), we can observe a difference due to the energy loss in the two carbon foils of the timing detector and also in the entrance window of the energy detector. As these phenomena increase with the depth in the sample and as they are not very well known, we statistically analyse the spectra to obtain mass energy spectra (Fig. 3) or mass time of flight spectra. These mass separated spectra leads to quantitative results by using interactive spectrum synthesis method [5].

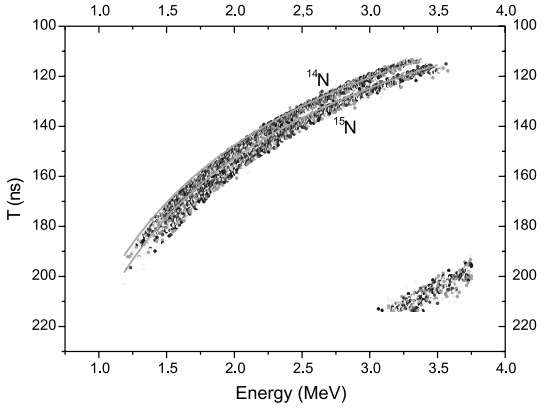

Fig. 2. Coincidence between time and energy spectra for a TiN coating analysed with 12 MeV $^{63}Cu^{5+}$ beam. The ^{14}N and ^{15}N are well separated. Comparison between experimental data (···) and theoretical curves (—).

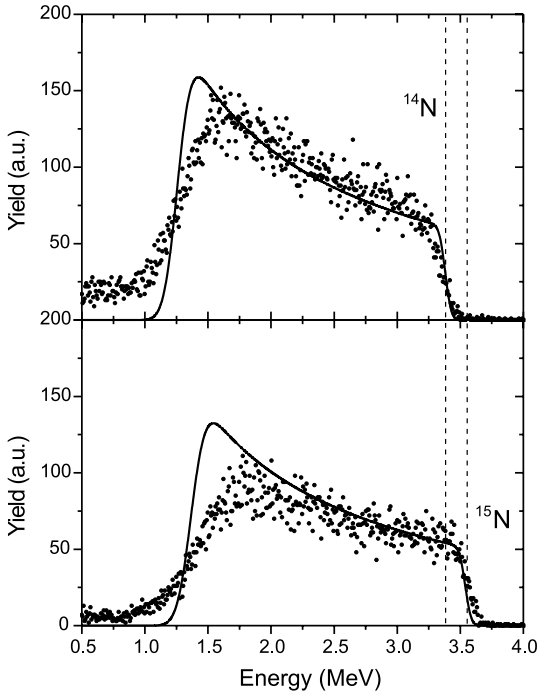

Fig. 3. Converted energy spectra for ^{14}N and ^{15}N. Comparison between experimental (···) and interactive spectrum synthesis method (—). The energy at which the front edge of ^{14}N and ^{15}N is marked with the dashed lines. The simulations (—) are obtained for equal concentration of ^{14}N and ^{15}N.

4. Quantitative results

4.1. Interactive spectrum synthesis method

In this method, theoretical ERD spectra of the sample assumption are calculated and compared with the experimental data. The calculation takes into account the straggling, the energy loss in the carbon foils of the start and the stop detectors and the entry window of the particle detector. The first step is to describe the sample as a stack of sub-layers. Each one is characterised by its thickness and its uniform composition. The concentration depth profile is determined by trial and error, changing sub-layers specifications. The simulated spectra are made up of contributions from each sub-layer in the sample [6]. To determine the energy location of each contribution, we compute the

energy lost by the beam and the recoil particle in the inward path and the outward path respectively. Using the Bragg rule for compounds, energy loss can be calculated for any particle target combination and beam energy between 100 keV and 15 MeV. Also the energy transfer in the scattering event has to be calculated. It depends on the masses of the incident and recoil particle and the scattering angle. The heights of the spectra are evaluated mainly from the incident projectile fluence, the atomic number density of the recoil atom, the detector solid angle, the recoil differential cross-section in laboratory system [7]. When the incident particles penetrate the material, statistical collisions slow down the ions. Straggling effect [8] leads to an energy spread for the incident and recoils atoms, which can be a limiting factor in the depth resolution of ERD-TOF depth profile technique.

4.2. Example of application: Ti($^{14}N + {}^{15}N$)

The interactive spectrum synthesis method has been applied to the mass separated spectra of the TiN coatings. The comparison between the computed and measured spectra is shown in Fig. 3. The energy at which the front edge of ^{14}N and ^{15}N appears depends of on kinematic factor of the elastic collision between the $^{63}Cu^{5+}$ and the ^{14}N and ^{15}N respectively. The best simulation is obtained for equal concentration of ^{14}N and ^{15}N. This result is in agreement with the TiN deposition process. At low energies, the simulated spectra are often not in agreement with the experimental data. It is due to the stopping power uncertainties and the unknown time detectors efficiency [9]. These phenomena lead to bad yield and concentration calculations.

In order to verify the ERD-TOF results and to measure the concentration of Ti in the sample, we have measured the same sample, tilted at 45° from the beam axis, by RBS at 165° with 2 MeV α-particles. The experimental sample spectra have been compared with simulations done with the SIMNRA code [10] using the Rutherford cross-section. The best simulation was obtained with a thickness for TiN film of 1.760×10^{18} at./cm^2 and a Ti concentration of 42 at.%. The number of Ti atoms was 7.39×10^{17} at./cm^2. Unfortunately, no information about the ^{14}N and ^{15}N concentrations can be obtained in the RBS spectrum.

The concentration for ^{15}N in the TiN coating has been measured by $^{15}N(p, \alpha\gamma)^{12}C$ resonant nuclear reaction at proton energy of 429 keV. This reaction was revealed by the specific 4.43 MeV γ-rays of the ^{12}C detected with a NaI detector. In order to obtain the ^{15}N concentration profile, the incident particle energy was increased by small increments starting from near-resonance energy. The number of ^{15}N was 3.32×10^{17} at./cm^2.

The ^{14}N concentration was measured by $^{14}N(\alpha, p_0)^{17}O$ and $^{14}N(\alpha, p_1)^{17}O$ reactions with 5.5 MeV α-particles. The experimental set-up consists of two detectors. The first one at 165° was used to take the BS spectrum. The second one (NRA) in which $^{14}N(\alpha, p_0)^{17}O$ and $^{14}N(\alpha, p_1)^{17}O$ reactions were detected, was located at 135° relative to the incident beam and a Mylar absorber foil of 24.4 μm was placed in front of the detector to stop the scattered α-particles. The sample was tilted at 45° from the beam axis. On the BS spectrum (Fig. 4(b)), we can observe the elements contained in the sample like Fe, Ti, ^{15}N, ^{14}N and C. The spectrum was compared with a simulation made with SIMNRA code. No reliable information on ^{14}N and ^{15}N could be obtained in this spectrum. The thin carbon peak is observed. It is due to a surface contamination which is caused by the incident beam. The simulation shows that the TiN coating contain a small amount of carbon (about 2 at.%) and the low carbon steel substrate in contamined by 5 at.% of carbon. These results were also observed by ERD-TOF. The NRA spectrum (Fig. 4(a)) consists of $^{14}N(\alpha, p_0)^{17}O$ and $^{14}N(\alpha, p_1)^{17}O$. It was simulated with the SIMNRA code using reactions cross-sections measured by Terwagne [11]. The best simulation is obtained with 4.35×10^{17} at./cm^2 of ^{14}N.

As consequence, the ERD-TOF results which tell that the sample contains the same amount of on ^{14}N and ^{15}N are not in good agreement with the measurement of ^{15}N and ^{14}N by $^{15}N(p, \alpha\gamma)^{12}C$ resonant nuclear reaction and $^{14}N(\alpha, p_0)^{17}O$, $^{14}N(\alpha, p_1)^{17}O$ nuclear reactions respectively. With these techniques the total amount of nitrogen in TiN film should be 57 at.% ^{14}N and 43 at.% of ^{15}N.

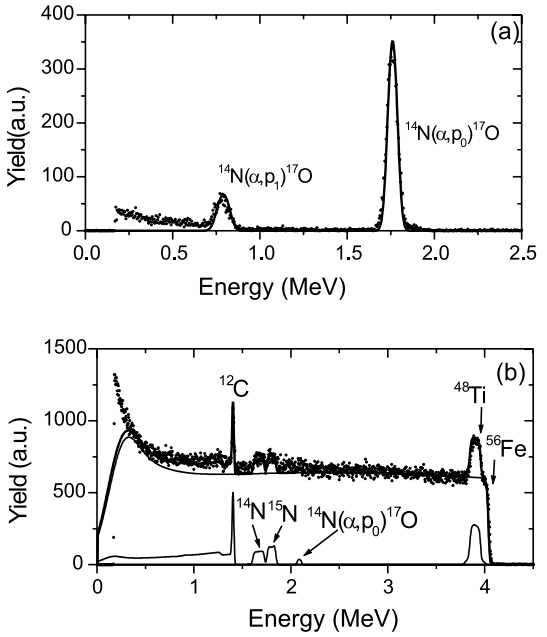

Fig. 4. (a) NRA spectrum obtained with 5.5 MeV α-particles for the Ti (^{14}N, ^{15}N) coating deposited on steel. The p_0 and p_1 peaks have been simulated with the SIMNRA code [10] for 4.35×10^{17} at./cm^2 of ^{14}N. (b) BS spectrum (···) measured simultaneously at backward angle. The spectrum has been simulated with the SIMNRA code [10].

The same concentrations were obtained by $^{14}N(d, \alpha)^{12}C$, $^{14}N(d, p)^{15}N$ and $^{15}N(d, \alpha)^{13}C$ reactions produced by 1.1 MeV deuterons. These measurements show also the presence of carbon in the TiN coating.

5. Conclusions

The ERD-TOF system with heavy ions of maximum energy of 16 MeV is a useful technique to depth profile light elements. A mass resolution of 1 amu for the light elements ($Z < 10$) has been achieved. Quantitative analysis with ERD-TOF is not easy. In the example the ERD-TOF results are not in good agreement with other improved techniques. A better knowledge of the stopping power of heavy ions in the energy range between 1 and 20 MeV will certainly improve the quantitative analysis. The desorption during the measurement and the efficiency of the time detectors have to be measured to calculate elements concentrations. Also the incident beam fluence at grazing incidence should be measured carefully.

As a TOF telescope has a better resolution than the PIPS detector, we would like to convert the time spectra into energy spectra and also to use the detector at backward angles to make conventional RBS with eventually ions heavier than α-particles.

References

[1] A. Climent Font, J.C. Banks, B.L. Doyle, Nucl. Instr. and Meth. B 161–163 (2000) 255.
[2] Y.S. Kim, J.K. Kim, H.W. Choi, G.D. Kim, H.J. Woo, Nucl. Instr. and Meth. B 136–138 (1998) 724.
[3] A.M. Zebelman, W.G. Meyer, K. Halbach, A.M. Poskanzer, R.G. Sextro, G. Gabor, D.A. Landis, Nucl. Instr. and Meth. B 141 (1977) 439.
[4] Ph. Roquiny, G. Mathot, G. Terwagne, F. Bodart, P. van den Brande, Nucl. Instr. and Meth. B 161–163 (2000) 600.
[5] J.R. Tesmer, M. Nastasi, J. Charles Barbour, C.J. Maggiore, J.W. Mayer, Handbook of Modern Ion Beam Materials Analysis, MRS, Pittsburgh, 1995.
[6] L.R. Doolittle, Nucl. Instr. and Meth. B 9 (1985) 344.
[7] J. Tirira, Y. Serruys, P. Trocellier, Forward Recoil Spectroscopy, Plenum Publishing Corporation, New York, 1996.
[8] Q. Yang, D.J. O'Connor, Nucl. Instr. and Meth. B 61 (1991) 149.
[9] Y. Zhang, H.J. Whitlow, T. Winzell, I.F. Bubb, T. Sajavaara, K. Arstila, J. Keinonen, Nucl. Instr. and Meth. B 149 (1999) 477.
[10] M. Mayer, SIMNRA user's Guide, IPP-Report 9/113, Max-Plank-Institute für Plasmaphysik, Garching, 1997.
[11] G. Terwagne, in press.