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Nuclear Instruments and Methods in Physics Research B 219–220 (2004) 430–434
NIM B
Beam Interactions with Materials & Atoms
www.elsevier.com/locate/nimb

ERD, ^{15}N external beam for NRRA in air, HIRBS: ion beam analysis developments on the HVEC EN-1 Tandem

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Abstract

In the last year, EN-1, the first HVEC Tandem accelerator, has gone through a major upgrade in which the injector, charging system and tubes were replaced. In addition, the ion beam analysis facilities have been upgraded and expanded. The gas-counter ERD set-up, previously installed on the former TASCC accelerator at Chalk River, is now operating with a new data-acquisition system. This system also interfaces with the surface barrier detector and TOF based ERD facilities (both of which were developed at the University of Montréal). Gas-counter and ERD-TOF are compared in terms of sensitivity, pileup, depth and mass resolution, and efficiency in the case of hydrogen detection. The ^{15}N NRRA technique has been extended to include an external beam capability. This allows, for example, in situ profiling of hydrogen in metallic hydrides exposed to different partial pressures of hydrogen.

Keywords: Elastic recoil detection; Time-of-flight; Gas-counter; Nuclear resonance reaction analysis; Heavy ion Rutherford backscattering spectrometry

1. Introduction: a bit of history

The Nuclear Physics Laboratory of the Université de Montréal started operation in the mid-1960s with the transfer from Chalk River, Ontario, of the HVEC Tandem accelerator prototype EN-1. The laboratory has become known for a number of groundbreaking innovations in the domain of ion beam analysis. In the mid-1970s, the laboratory was the birthplace of the elastic recoil detection analysis method (ERDA) by L'Écuyer et al. [1], using a 30 MeV Cl beam. In the mid-1980s, the laboratory was the home of the first incarnation of a time-of-flight (TOF) detector applied to the ERD technique [2], which offered a much greater mass separation than a stopping foil and considerably improved depth resolution. The IBA techniques available were used intensively, mainly as a facility to provide depth profiling for the local Thin Layer Group and for international collaborators. With the shutdown of the TASCC accelerator at Chalk River, a gas-counter ERD system has been transferred to the University and further improved.

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doi:10.1016/j.nimb.2004.01.096

In this paper, after describing the recent upgrades made to EN-1, the ERD setups will be presented and compared in terms of depth and mass resolution, depth range, pileup and sensitivity. We will finally show some of the results obtained with the newly developed extracted-beam ^{15}N-NRRA for in situ profiling of hydrogen at high resolution.

2. EN-1 upgrade

During the year 2002, the EN-1 Tandem accelerator went through a major upgrade in which most of its internal parts other than the tank and the column were replaced, as was the injector. The new injector, computer controlled and remotely operated by a fibre optic interface, includes a dual source system: a cathode sputtering source and a duoplasmatron source with a lithium exchange canal. The system features beam optics integrated with the tube (Q-Snout). The accelerator tubes were replaced by new inclined-field tubes. The new configuration translates into a beam intensity improvement by a factor of 3–20. The stripping channel has been replaced by a turbo-pumped chamber that recycles the out-flowing gas in the stripper resulting in a better base pressure in the terminal and better stripping efficiency. Finally, the original Van de Graaff charging system has been replaced by a dual chain Pelletron^{TM} system. Voltage stability of the upgraded system shows an improvement of a factor of $\sim$10.

3. Elastic recoil detection

The laboratory hosts two ERD systems: a time-of-flight system and a gas-counter. The ERD-TOF detector is at a fixed scattering angle of 30° to the beam direction. The timing foil, currently a 20 μg/cm^2 carbon foil, is located at 12.5 cm from the target. The secondary electrons generated from the carbon foil are collected by a microchannel plate detector (MCP) giving the first timing signal. To measure the energy and the second timing signal, we use a surface-barrier detector (SBD), which is located at 62 cm from the carbon foil, subtending a solid angle of 0.18 msr. Both pulses (energy and time) are encoded in an analog-digital converter (ADC, Ortec AD413A). The dead time of the ADC system does not vary with energy.

The gas-counter ERD system, oriented at 45° to the beam axis, consists of a 30 cm long counter containing low-pressure isobutane (5–65 Torr), isolated from the interaction chamber by a 90 μg/cm^2 stretched polypropylene foil. The prototype of this detector was described earlier [3]. The anode is divided into four sections, which measure four fractions of the total energy (ΔE_1, $\Delta E_{2,\mathrm{right}}$, $\Delta E_{2,\mathrm{left}}$ and E_{rest}). The ΔE_2 electrodes are split into two "backgammon" sections along the length of the detector providing scattering/recoil angle information to make kinematic corrections. The detector subtends a solid angle of $\sim$5 msr. For very light recoils such as H and He a surface barrier detector mounted at the rear of the detector is used, as the pulse heights from the gas detector are too small. It subtends a solid angle of 0.8 msr. Both ERD chambers use the same data-acquisition system.

3.1. ERD systems comparison

The ERD systems described above, intended to achieve mass separation in almost any compound, both have advantages and trade-offs. The TOF system is often perceived as offering a better mass separation even allowing the resolution of different isotopes, but the flight length and detector size make the solid angle small. In contrast, the gas-counter ERD has a large solid angle but requires kinematic correction. In order to appreciate more quantitatively the differences between these two methods, a comparison is presented.

An SiO_2 layer of nominally 528 nm grown on a silicon substrate has been analysed in both systems: using a 30 MeV Cl^{6+} beam for the ERD-TOF and a 40 MeV Cu^{7+} beam for the gas-counter ERD. Fig. 1(a) shows the events plotted as time-of-flight versus energy for the ERD-TOF (7×10^5 events, including scattered beam events) and as ΔE_1 versus E_{rest} for the gas-counter system (3.5×10^5 events, detection angle above critical angle for scattering: no beam events). In the case of the gas-counter ERD, in order to suppress the

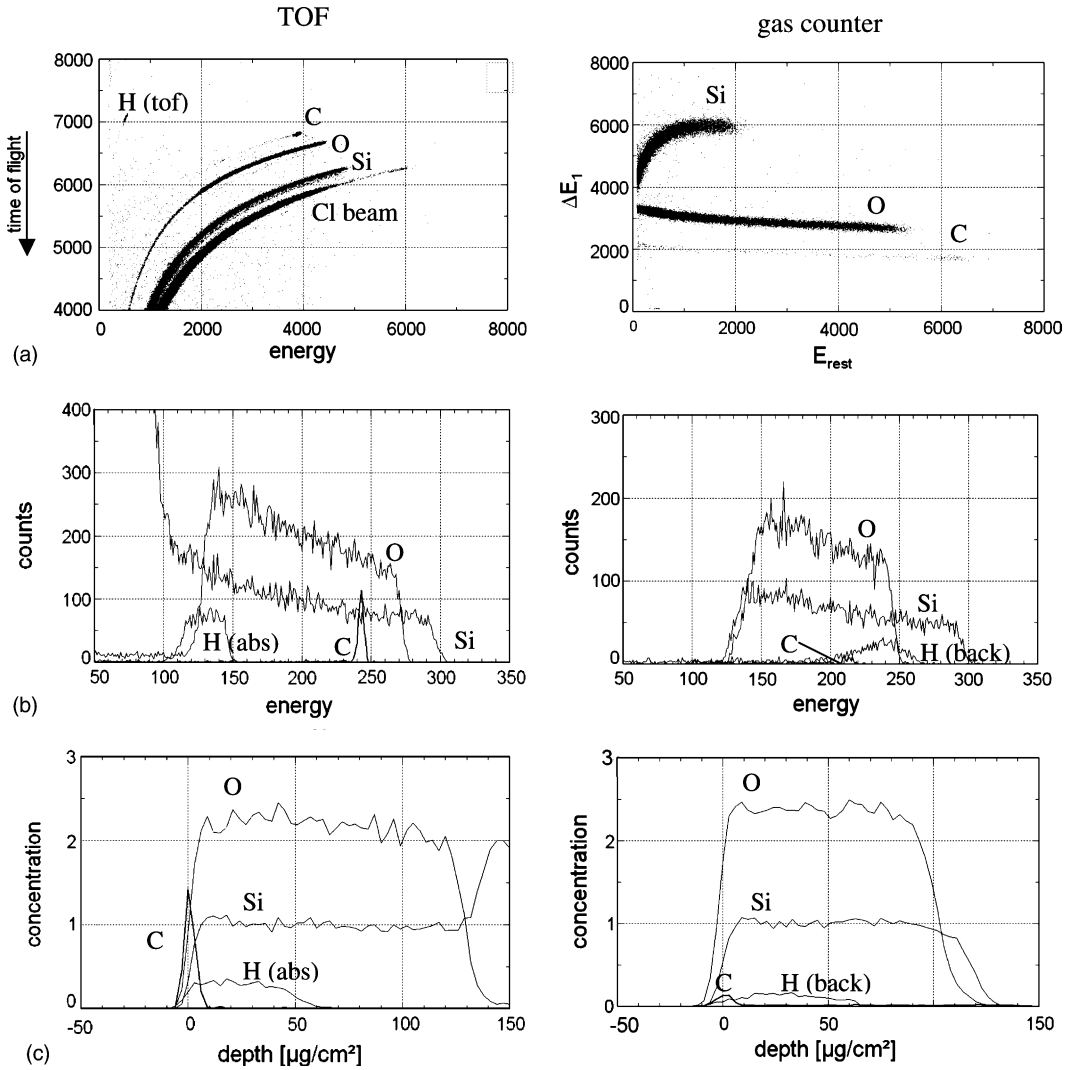

Fig. 1. Comparative analysis of a 528 nm SiO_2/Si layer by ERD-TOF and gas-counter ERD. (a) 2-parameter plot of the events that show the traces of the different elements; (b) corresponding energy spectra; (c) depth profiles of each element. In the case of the gas-counter ERD, only the events entering near the centre of the detector are shown in (a) and (b). Hydrogen measurements are carried out by means of an absorber-based ERD (abs) in the case of ERD-TOF, and by a surface barrier detector located at the back of the gas chamber (back) in the case of the gas-counter ERD.

effect of kinematic broadening on the displayed data, only the events entering near the centre of the detector are shown in Fig. 1(a) and (b), while the depth profiles calculations in (c) are based on all events. The gas-counter ERD shows better mass separation at high energy, which can also be tuned to some extent by optimizing the gas pressure. However, the signals get closer for small values of E_{rest}. The TOF detector shows a more regular mass separation down to fairly low energies. The TOF also features a mass-based separation, potentially providing isotopic resolution especially for light elements, while the gas-counter system is sensitive only to atomic number. In terms of pileup or "misplaced" events, pileup events in the TOF detector are generally caused by more than one

timing signal occurring during an event, generating the wrong time-of-flight value for a given energy. Given the relatively small solid angle, such pileup cannot be avoided in order to stay within a reasonable acquisition time. For the gas-counter detector, the amount of misplaced events is low because the characteristics of gas-counters this size requires that the rates be kept below 1 kHz.

The element-separated spectra are shown on row (b) of Fig. 1. In the case of the gas-counter ERD measurement, the data do not extend to low energies because there is an energy threshold, which is determined by energy loss in the entrance foil and by the requirement that the particle gives a signal in E_{rest}. As a result, the depth range is reduced from that with TOF, barely reaching the thickness of the analysed sample. Finally, it is worth mentioning that the hydrogen signal from the ERD-TOF actually comes from an absorber ERD detector (stopping foil: Al 4600 μg/cm^2), because the TOF detector has considerably reduced efficiency for recoiled hydrogen (3% in this case) [4]. The trade-off is that the depth resolution in this case is significantly reduced. Similar arguments apply to the H detection by the surface barrier detector located at the back of the gas-counter detector.

The spectra were converted iteratively into depth profiles using the computer code Allegria [5]. The profiles are compared in Fig. 1(c). Both analyses show a carbon peak, which can be used to estimate the surface depth resolution assuming that the layer is sufficiently thin for instrumental resolution to dominate. Depth resolution can also be estimated from the step edges of the oxygen and silicon depth profiles. While the ERD-TOF system shows a surface depth resolution of $2.42 \pm .0.05$ μg/cm^2, the gas-counter presents a slightly worse value of 3.1 ± 0.1 μg/cm^2. At the Si/SiO_2 interface, the ERD-TOF resolution is 4.9 ± 0.4 μg/cm^2. For the gas-counter ERD, the interface corresponds to the maximum probing depth and we estimate the resolution to be 5 ± 1 μg/cm^2. The larger increase for the TOF is likely due to the shallower angle of detection and hence longer path length.

Here, we have compared the two types of ERD for a given number of events, but the solid angle of the gas-counter detector is significantly larger than that of the TOF detector, making it ~25 times more sensitive. The large solid angle of the gas-counter and the position information enable detection of blocking patterns giving lattice location information.

4. NRRA in air and HIRBS

Hydrogen profiling by nuclear resonant reaction analysis using ^{15}N is known as one of the methods that offers the best depth resolution [6]. In many cases, however, profiling hydrogen in vacuo is not suitable. A good example is the metallic hydrides used with fuel cells. In order to understand the hydrogen behaviour in such materials, one wants to measure the depth profile at various partial hydrogen pressures. This means that the beam needs to be extracted ex vacuo in order to carry out depth profiling in a hydrogen gas ambient that can reach atmospheric pressure.

In order to carry out such measurements, we have implemented the technique on one of the beam lines by passing the ion beam through an isolating membrane. The main concern is to extract the beam without significantly affecting the beam energy spread induced by straggling. To do so, we used Si_3N_x low-stress membranes about 300 nm thick and a few mm in diameter. Used carefully, they can survive a complete day of operation at atmospheric pressure with beam intensities of 10 nA. While a complete description of the technique and results will be published elsewhere [7], Fig. 2 shows initial measurements obtained with the technique in a thin Pd/Mg layer on Ni. The depth scale is in units of μg/cm^2, which represents a much larger distance in air than in the material. The first peak (at left) is the amount of hydrogen in the membrane, then in ~1.6 mm (200 μg/cm^2) of dry air, and finally in the metallic hydride layer on Ni. Based on the width of the step edges, the resolution is <3 μg/cm^2 just after the membrane and 14 μg/cm^2 at the surface of the sample. According to SRIM simulations, the straggling in the air should contribute to only 2 μg/cm^2, the remainder being due to surface roughness or other effects.

Finally, to complement the ERD setup, a Heavy-Ion RBS system using time-of-flight measurement

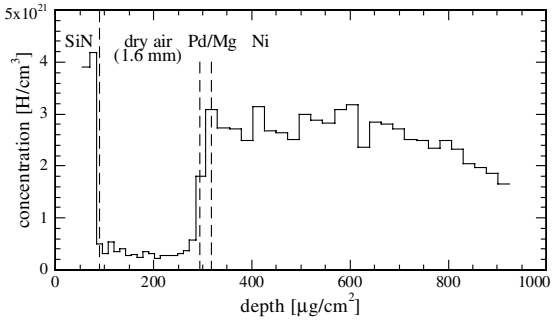

Fig. 2. Example of an ex vacuo NRRA analysis of hydrogen in Pd/Mg/Ni layers using an extracted-beam. Each step of the histogram corresponds to a single acquisition.

for the energy has been developed. Currently, the start signal is provided by a timing foil and a MCP, while the stop signal comes from a surface barrier detector. Improvements to the energy resolution will be achieved by the addition of a second timing foil for the stop signal.

5. Summary

A major upgrade the EN-1 Tandem accelerator has been made. In parallel, several improvements to the ion beam analysis techniques have been made. A comparison of elemental depth profiling using two types of ERD detectors reveal that while the TOF detector achieves slightly better surface depth resolution, better mass separation at low energies and a greater analysis depth, the gas counter offers higher sensitivity, a lower number of pileup events, and potentially lattice location information. Finally, NRRA profiling of hydrogen has been achieved in air at atmospheric pressure allowing measurement of hydrogen in samples exposed to different hydrogen pressures ex vacuo.

Acknowledgements

The authors wish to thank R. Gosselin and L. Godbout who carried out most of the EN-1 Tandem upgrade. This work has been supported by the Natural Science and Engineering Research Council of Canada, the Fonds Québecois de Recherche sur la Nature et les Technologies, NanoQuébec and the Canadian Foundation for Innovation.

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