Radioisotope implantation with a new facility at the Australian Defence Force Academy

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A 50 - 155 keV ion implanter is being developed on the campus of the University of New South Wales at the Australian Defence Force Academy for the implantation of radioisotopes as part of a UNSW/ANU collaboration. The facility employs a versatile SNICS II negative ion source. Commissioning tests have shown the facility to efficiently produce, mass-select, and transport negative ion beams of various stable isotopes. The mass resolution has been demonstrated to be better than 1 amu and the implantation of stable isotopes was verified with Elastic Recoil Detection (ERD) analysis. Recently, the first implantation of radioactive ^{111}In has been performed successfully. The routine implantation of this and other radioisotopes is envisaged to support analytical techniques in the material sciences, such as Perturbed Angular Correlation (PAC) spectroscopy and Nuclear Magnetic Resonance of Oriented Nuclei (NMRON), and to possibly study the controlled activation of medical implants and the diffusion of radioisotopes in materials.

Introduction

Radioisotopes have many important applications in areas such as nuclear medicine, environmental studies, material science, and solid state physics. Nuclear medicine uses radioisotopes to provide diagnostic information and also for therapy to treat medical conditions such as cancerous tumours, with the ionising radiation weakening or destroying affected cells. Storing and isolating nuclear waste is an environmental challenge due to the possible leakage of radioisotopes. The diffusion of radionuclides in materials can be studied following controlled ion implantation.

In solid state physics, Perturbed Angular Correlation (PAC) spectroscopy and Nuclear Magnetic Resonance on Oriented Nuclei (NMRON) are well established techniques for the study of structural, electrical and magnetic properties of crystalline materials. In both techniques radioisotopes are introduced into the material as probes. The directional distribution of emitted ionising radiation can then be detected providing information about local electromagnetic fields in the immediate vicinity of the probing nuclide. This enables, for example, studies of the structural properties and the annealing behaviour of the host material. While thermal diffusion is a common method to introduce radioisotopes into host materials, this is often not possible for many advanced materials which would be seriously affected by such processing. In these cases radioisotopes need to be introduced by ion implantation where the number of introduced radioactive nuclei can be controlled explicitly and precisely. With high energy (> 1 MeV) ion implantation structural damage to crystals can be severe. The implantation at lower energies (< 1 MeV), potentially minimizes crystal modifications and thus improves the reliability of PAC and NMRON studies.

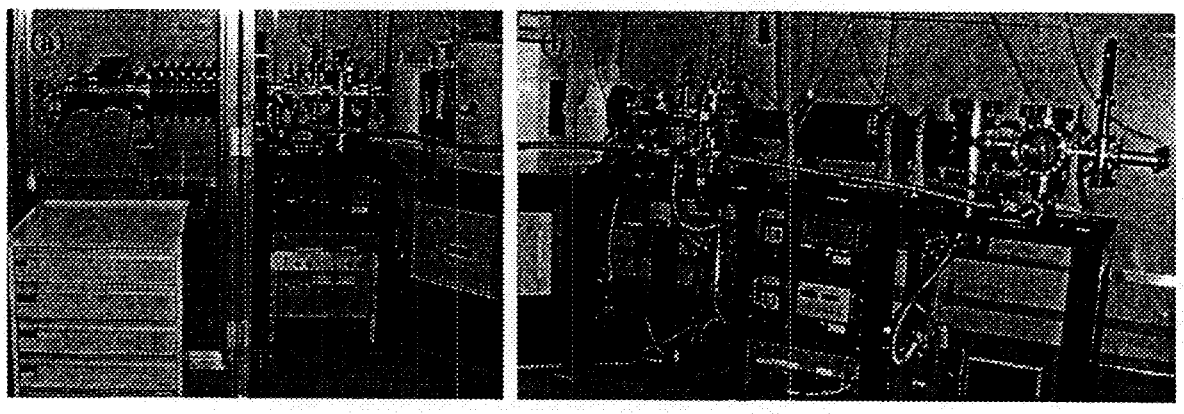

Figure 1: The radioisotope ion implanter at the Australian Defence Force Academy. (a) From left to right, the negative ion source, the acceleration tube, and the input side of the analysing dipole magnet. (b) From left to right, the exit side of the analysing magnet, the electrostatic Einzel lens, and the implantation chamber of the facility.

To facilitate such work, a low energy (50-155 keV) radioisotope ion implanter is under development on the campus of the University of New South Wales at the Australian Defence Force Academy for shared use with the Australian National University. In contrast to similar facilities overseas [1-2], this is the first time that a negative ion source is used. This offers the advantage of more versatility since negative ions can be produced abundantly for many elements [3-7].

This paper reports on the ion implantation of stable isotopes and the first successful implantation of the radioisotope ^{111}In with the new facility. The latter has been chosen since it is a standard PAC probe.

Description of the facility

A photograph of the facility is shown in Fig. 1. The implanter uses a SNICS II (Source of Negative Ions by Cesium Sputtering) ion source. In this ion source, Cs^+ ions are produced by a tantalum ionizer immersed in Cs vapour [4-7]. The Cs^+ ions strike material inside a cold cathode. Sputtered material partially forms negative ions by accepting electrons from neutral cesium atoms. The negative ions are then accelerated by passing through a potential difference of up to 150 kV. Since

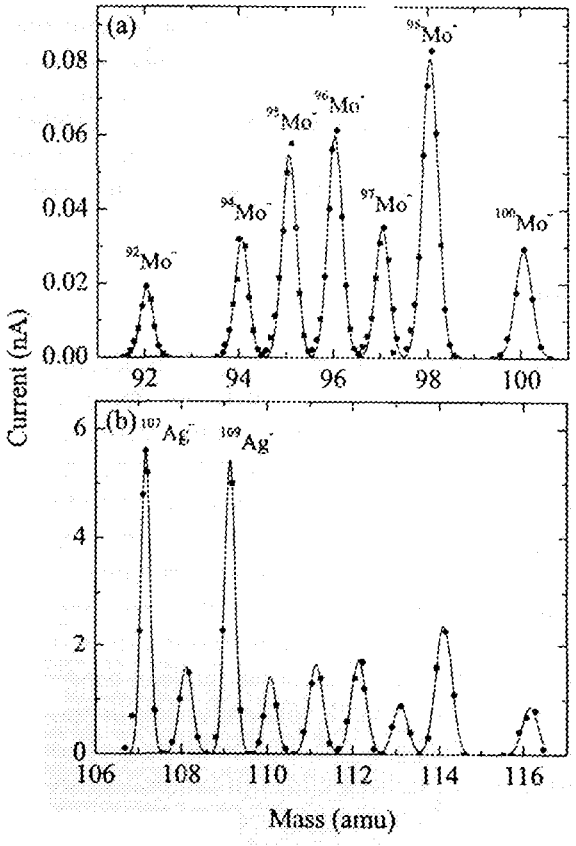

Figure 2: Partial mass spectra for a molybdenum oxide cathode with silver sinter. The solid curves are gaussian fits to the data. (a) Peaks corresponding to negative ions of different Mo isotopes are well separated. (b) The two Ag isotopes and different molecular MoO^- ions can be distinguished.

the cold cathode has an additional negative bias of 5 kV, the maximum energy achievable is 155 keV. Following acceleration, the ion beam is mass-selected using a single-focussing dipole magnet. Double focusing characteristic are provided by two quadrupole magnet singlets before and after the dipole magnet. Mass-selected ions are focussed onto the sample with an electrostatic Einzel lens.

The total ion current from the SNICS II source and the mass selected ion current can be measured with Faraday cups before and after the dipole magnet. The profile of the mass-selected beam can be observed using a beam-profile monitor located between the dipole magnet and the Einzel lens. The beam-profile can be controlled by suitably adjusting collimators before and after the dipole magnets. These collimators limit the beam both in the horizontal and the vertical direction. Rectangular beam spots with dimensions of 2 mm × 3 mm are routinely achieved. Transmission of the mass-selected beam is near 100%.

Figure 2 shows partial mass spectra obtained for a molybdenum oxide source cathode with a silver sinter. This cathode was chosen because of the large number of molybdenum isotopes and the associated oxide molecules in the interesting mass region 90-120 amu, which includes the mass of radioactive ^{111}In. The mass spectra obtained are similar to those measured by Middleton [7]. All the isotopes of molybdenum and the various molybdenum oxide molecules are clearly separated. The two stable silver isotopes, ^{107}Ag and ^{109}Ag, are also seen. It is evident that the mass resolution is better than 1 amu.

Implantation of stable indium

In order to verify whether the implantation of the radioisotope ^{111}In is achievable with the new facility, firstly beams of stable elemental and molecular indium ions have been produced using an ion source cathode filled with pressed indium oxide powder. Figure 3 shows that molecular indium oxide ions are much more abundant than elemental In ions. The $^{115}InO^-$ beam was thus chosen for the implantation into crystalline silicon and implanted at different energies.

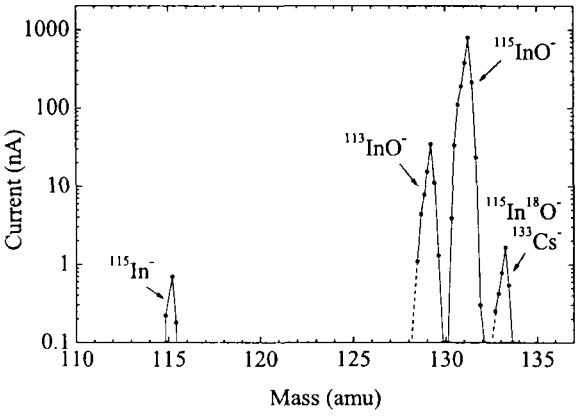

Figure 3: Partial mass spectrum for an In_2O_3 cathode. Peaks corresponding to In and InO ions can be clearly identified and are well separated. A $^{113}In^-$ beam was not detected.

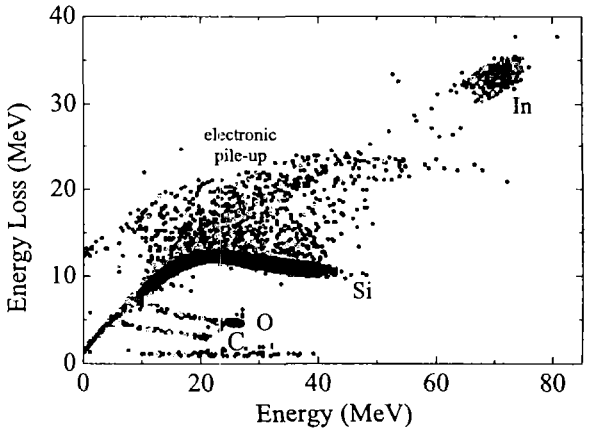

Figure 4: A typical two-dimensional spectrum from the ERD analysis of a silicon sample implanted with $^{115}InO^-$. The labels indicate groups of events associated with a specific element. The grey scale represents the total number of detected events.

Implanted samples were analysed with Elastic Recoil Detection (ERD) using 200 MeV ^{197}Au ions. Details of the ERD technique are given in the Ref. [8]. In this particular case, the recoil ions were detected at an angle of 45° relative to the beam direction using a position-sensitive gas ionization detector with a large solid angle of 3.5 msr. For a 125 keV implantation of $^{115}InO^-$, Fig. 4 shows the measured energy loss signals ΔE of the recoil ions as a function of the detected energy E. In this two-dimensional diagram, groups of events associated with a particular element can be distinguished. The spectrum shows that in addition to In, O and Si, traces of C are also present in the silicon sample. Most of the oxygen is from the implantation and some presumably from surface contamination, while the indium is entirely from the implantation. Analysis of the ERD data has shown that the indium was implanted with a fluence of (5.7 ± 0.7) × 10^{16} ions/cm^2, which is consistent with the fluence deduced from the beam current measurements during the implantation.

Radioisotope Implantation

Following the successful implantation of stable indium isotopes, implantations of the radioisotope ^{111}In were performed to evaluate the feasibility of the approach. In order to make a cathode filling with radioactive ^{111}In, rhodium metal was pressed into a copper ion source cathode. The rhodium in the cathode was then bombarded with 76 MeV ^{12}C delivered by 14UD Pelletron tandem accelerator at the Australian National University [9,10]. The beam current was typically 2 mA and the ^{12}C bombardment was over 15 hours. The following nuclear fusion evaporation reactions lead to the production of ^{111}In:

$$^{103}Rh(^{12}C, p3n)^{111}Sn \xrightarrow[EC,\beta^+]{35min} {}^{111}In$$

$$^{103}Rh(^{12}C, 4n)^{111}Sb \xrightarrow[EC,\beta^+]{1.3min} {}^{111}Sn \xrightarrow[EC,\beta^+]{35min} {}^{111}In$$

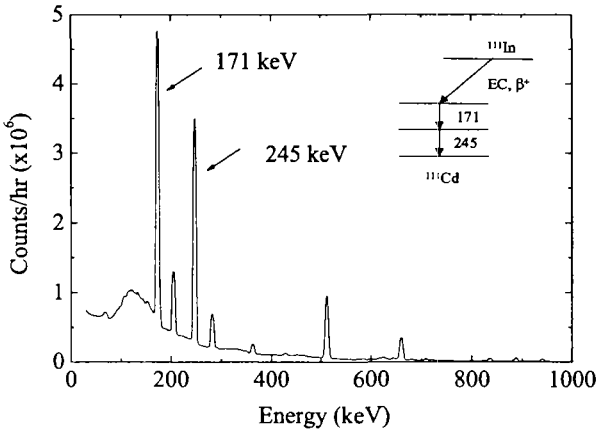

Figure 5: The γ-ray spectrum from the activated rhodium cathode measured with a Ge-detector. The 171 keV and 245 keV γ-rays following the decay of ^{111}In are evident.

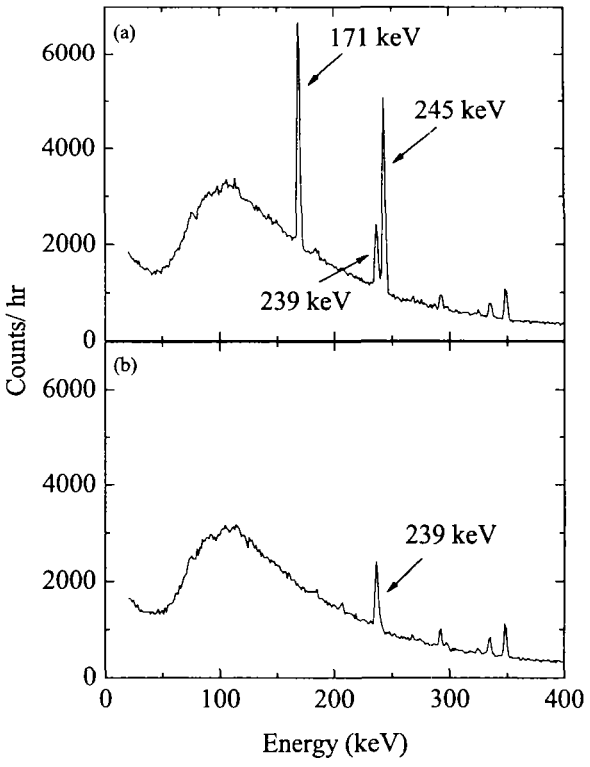

Figure 6: (a) The partial γ-ray spectrum from a silicon sample implanted with $^{111}InO^-$ showing the characteristic 171 keV and 245 keV lines which are not present in the background spectrum displayed in (b). The background line at 239 keV is due to ^{212}Pb.

Figure 5 shows a Ge-detector spectrum for the activated ion source cathode. In this spectrum, the 171 keV and 245 keV γ-rays indicate the presence of ^{111}In in the cathode. The activity of ^{111}In in the cathode was estimated through comparison with a calibrated ^{133}Ba source to be (0.5 ± 0.1) mCi.

The activated ion source cathode was then installed in the SNICS II ion source of the radioisotope implanter. A beam of 125 keV $^{111}InO^-$ beam was produced and transported to the implantation chamber. The samples implanted included steel, crystalline silicon and an indium nitride thin film. A typical γ-ray spectrum for a silicon sample implanted with $^{111}InO^-$ is shown in Fig. 6 and compared with the background spectrum. In addition to the background lines, 171 keV and 245 keV γ-rays are clearly evident in 6(a), which verifies the successful implantation of ^{111}In. Implantation of ^{111}In is further evidenced by the half-life of these decays which was measured to be (2.83 ± 0.03) days and agrees with the half-life of ^{111}In (Figure 7).

Figure 8 shows Ge-detector and NaI-detector spectra for the steel sample illustrating the advantage of using a Ge-detector to verify the implanted radioactivity. In the case of the NaI-detector the 245 keV γ-ray is not separated from a neighbouring background peak.

Despite the successful first radioisotope implantation, the activity implanted in the samples was too low to successfully perform PAC spectroscopy. The activity implanted in the Si sample was measured as ~0.2 nCi. For reliable PAC studies about 30 μCi are needed. Measurements showed that the residual activity in the ion source cathode following implantation was only 20 μCi, thus the activity was almost entirely sputtered out of the cathode. Based on absorbed dose

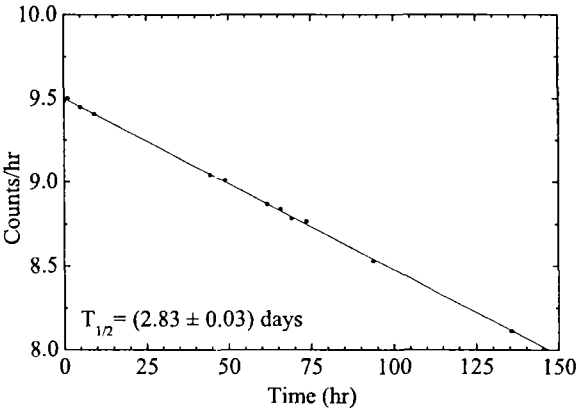

Figure 7: The count rate of the 171 keV γ-ray over time. The solid line is a linear fit to the data corresponding to the indicated half-life $T_{1/2}$.

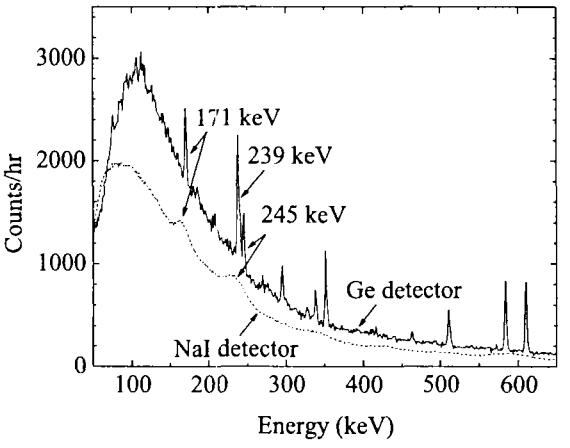

Figure 8: Partial γ-ray spectra for a steel specimen implanted with $^{111}InO^-$. 171 keV and 245 keV peaks are evident in the case of Ge-detector spectrum while there is only an indication of those lines in the spectrum from the NaI-detector.

measurements at the ion source, more than 90% of the ^{111}In-activity remained, however, in the ion source chamber. TRIM [11] calculations indicate that the ^{111}In produced in the source cathode with the recoil implantation of ^{12}C into ^{103}Rh would be distributed within a thin layer about 10 μm below the surface. Based on the depth of the crater in the cathode it has been derived that the activity was most likely sputtered within only 30 minutes of implanter operation. This means almost all

the activity in the cone was already sputtered when the implanter was being set up. This may explain the low activity of ^{111}In implanted in the samples. This problem could be overcome if an ion source cathode is used where the activity is more uniformly distributed over a depth of a few millimetres.

Conclusions

The production, transport and implantation of different ion beams of stable isotopes have been demonstrated for a new ion implanter facility intended for radioisotope implantation. Mass resolution of better than 1 amu has been achieved. The implantation of stable indium was successfully performed and has been independently verified with ERD analysis. Implantation of the radioisotope ^{111}In has been demonstrated using $^{111}InO^-$ ions. Though the ^{111}In activity introduced into the sample is much less than what is required for PAC studies, the result suggests that the implantation of this and other radioactive isotopes is feasible with the new facility. In the next development step it is planned to use commercially available $^{111}InCl_3$ to prepare a uniformly activated ion source cathode and thus ensure prolonged source output of the radioisotope.

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