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Tritium Depth Profiling by Neutron Time-of-Flight*

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Abstract

A method has been developed to measure the depth profile of tritium implanted or absorbed in materials. The sample is bombarded with a pulsed proton beam. Neutron energies from $T(p,n)$ reactions occurring in the target are measured by time-of-flight over a 10.8 m flight path. From the neutron energy the proton energy and thus the tritium depth in the target may be inferred. The measurements were initiated to determine the depletion of tritium absorbed in titanium layers. These layers had served as targets on a 400 keV $T(d,n)$ neutron generator. A depth resolution of 0.4 mg/cm^2 of titanium ($0.9 \text{ }\mu\text{m}$) was obtained. Unlike proton backscattering methods, the technique can be used with thick samples. We estimate that a sensitivity of 0.1 at.% tritium in host materials can be obtained. Extension of the method to deuterium depth profiling is possible with the use of the $D(d,n)$ reaction.

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Introduction

The use of ion backscattering to obtain depth profiles of one atomic species implanted in a matrix of another species is a well developed technique. However, the measurement of the distribution of a light species implanted in a heavy one is difficult because of the unfavorable ratio of the Rutherford cross sections for backscattering for the two species. Blewer¹⁾ has developed a proton backscattering method which overcomes this limitation by using an incident proton energy large enough that nuclear (rather than Coulomb) elastic scattering is the dominant contribution to the backscattering cross section for the light species. An enhancement of two orders of magnitude in backscattering from the light species is observed and a detection sensitivity of 1 at. % of He in Cu is possible. A major limitation of this technique is that high sensitivity can be obtained only if the host material is sufficiently thin that protons scattered from the implanted atoms may be kinematically separated from those scattered from the host material.

We have developed a method for determining the depth profile of tritium in materials by measuring the energy of neutrons produced by the $T(p,n)^3\text{He}$ reaction. From the neutron energy the energy of the reacting proton can be determined. The depth of the tritium in the host material can then be inferred from the proton energy. This technique can be used with any 3 MV accelerator equipped for nanosecond pulsing. The method has several advantages over proton backscatter techniques. The sample thickness which may be surveyed depends upon the difference in the (p,n) thresholds of tritium and the constituents of the host material and the proton energy available but is $10\text{--}20 \text{ mg/cm}^2$ for tritium in heavy ($A > 50$) materials. Sensitivity depends only upon the cross section for the

T(p,n) reaction and the depth resolution desired in the measurement, not upon a ratio of cross sections. Qualitative accuracy (profile shape) may be easily obtained because the energy dependence of the T(p,n) cross section is well known. The technique is nondestructive: no modifications or special conditions are necessary to assay the sample. The present measurements have been performed with samples containing at least one atom of tritium per atom of titanium but we estimate that a sensitivity of 0.1 at. % can be obtained. Using a different nuclear reaction, i.e. D(d,n), this technique can be extended to provide depth profiling of deuterium.

This work was undertaken to investigate the depth profile of tritium in targets used on the Livermore rotating target 14-MeV neutron source.²⁻⁴⁾ The tritium is absorbed in a layer of titanium approximately 5 mg/cm² thick (11 μ m) evaporated on a 1 mm thick backing of Amzirc, a Cu-Zr alloy. We wished to obtain information about the distribution and mobility of the tritium as a function of the bombardment history of the target and, if possible, similar information about the deuterium implanted in the target by the accelerator.

The rotating targets are bombarded by a 400-keV deuteron beam of 20 mA. The neutron yield from the targets decreases with use. Targets are usually changed after the yield has decreased to 70-80% of its initial value. The amount of deuterium injected into the target during a 100 hour bombardment is 2-3 times the total initial tritium content of the target. How much of this deuterium is retained by the target and its location within the titanium layer may greatly affect the transport of the tritium.

Experimental Method

The arrangement for the initial measurements is shown in Fig. 1. A 2.5 MeV proton beam from the EN tandem of the Livermore Cyclograaff⁵⁾ passes through a 1.2 μm Ni foil separating the He filled glovebox containing the target from the accelerator vacuum system. In later measurements small pieces were cut from the large target and placed directly in the vacuum system. The proton beam is bunched and chopped to produce pulses a few ns long at a 2.5 MHz repetition rate. A neutron detector is placed 10.8 m from the target at an angle of 24° with respect to the direction of the proton beam. The detector is behind 4 m of concrete and earth shielding and has a 2 m long water collimator which allows only neutrons originating in the target to be detected. The detector is an 11.4 cm diameter by 5.1 cm thick cell filled with NE 213 liquid scintillator. An RCA 8575 phototube views the cell. Both anode and dynode signals from the phototube are used to obtain fast timing and pulse height information. The difference in time between the arrival of a proton burst at the target (inferred from the arrival at the detector of gamma rays produced by the beam) and the arrival of a neutron at the detector is measured with standard instrumentation and used to calculate the neutron energy.

The depth profile is obtained by taking the spectrum of neutron counts versus time-of-flight, calculating neutron energy and then proton energy from the reaction kinematics. The effects of variation in detector efficiency and $T(p,n)$ cross section with energy must then be removed. From a plot of yield versus proton energy the yield versus depth plot may be obtained by using the stopping cross sections for protons in the

host material. The resolution obtained depends upon several factors: proton burst length (2-4 ns), detector resolving time (1-2 ns), proton energy straggling, and the length of the flight path. The best resolution obtained to date is equivalent to 0.4 mg/cm^2 of Ti ($0.9 \text{ }\mu\text{m}$) at a depth of 1.6 mg/cm^2 ($3.5 \text{ }\mu\text{m}$) in the target. This value was determined by measuring the burst width of neutrons from a thin TiT target ($160 \text{ }\mu\text{g/cm}^2$) in vacuum, converting this to an equivalent depth, and adding straggling effects.

Tritium Depth Profiles

The depth profile of tritium in an unused portion of a target is shown in Fig. 2. This profile was obtained in a 15 minute run with a 100 nA beam. Statistical accuracy is better than 3% for points at the top of the distribution. The multiplicative factor applied to the raw data to correct for cross section and detector efficiency variation ranged from 1.00 at 1.5 MeV neutron energy (the normalization point) to 1.80 at 1.03 MeV. As the cross section and detector efficiency vary smoothly with energy the relative value of these corrections is quite good. Resonances in the total cross sections of nitrogen and oxygen result in differential transmission of neutrons through the air along the 10.8 m flight path. Point scatter of 5-10% can result from this effect. No corrections for the variation in transmission have been applied. If all corrections are applied to obtain a quantitative depth profile an absolute accuracy of 5-10% is possible, limited primarily by the absolute value of detector efficiency.

A depth profile from a used portion of the same target is shown in Fig. 3. The solid line is a smooth curve drawn through the distribution shown in Fig. 2. Depletion of tritium is most severe near the end of the 1.6 mg/cm^2 range of the 400 keV deuterons. The difference in

maximum concentration between the two profiles is consistent with the yield variation observed in different portions of the large targets and is not significant. A difference in the thickness of $\sim 0.5 \text{ mg/cm}^2$ in the titanium layer is readily apparent, however. Extrapolation of the concentration at the rear of the target to the front surface and comparison of areas indicates that 15% of the tritium originally contained in the target has been evolved. There is little migration of tritium from the rear of the target to the depleted region. Integration of the $T(d,n)$ cross section over the depleted profile gives a predicted yield of 80% of that obtained by integration over the assumed undepleted profile. This calculated yield is consistent with the observed decrease in yield of the target in operation.

There are two sources of uncertainty in the depth scale of the profiles. The origin has been placed at the neutron energy corresponding to the incident proton energy. The displacement of the depth profile from the origin suggests the possibility of a dead layer or contaminant layer on the front surface. Secondly, the tritium and injected deuterium have been ignored in the calculation of proton energy loss. For a $TiTi_{1.5}$ target this neglect causes an error of 7-14% depending upon the value assumed for the density of the hydrated layer. Correction for this error was omitted as it is less than the resolution at the depth of interest.

Deuterium Content

We have determined the residual deuterium content of the target by using a 2.5 MeV pulsed deuteron beam in place of a proton beam. Because of the large difference in reaction Q values (17.568 MeV vs 3.266 MeV) the neutrons from the $T(d,n)$ reaction are separated from those from the

D(d,n) reaction. The time-of-flight spectrum obtained is shown in Fig. 4. The ratio of the D(d,n) to T(d,n) yields indicates that the deuterium content of the target after bombardment is approximately 35% of the initial tritium content. While only 10-20% of the implanted deuterium remains in the target, the excess of deuterium implanted over tritium evolved has increased the net hydrogen content of the target.

This measurement was repeated at 1 MeV deuteron energy (lower deuteron energy yields lower neutron energies, hence better resolution for a fixed resolving time) to provide information about the depth profile of the implanted deuterium. The resolution obtained was marginal ($\sim 1.5 \text{ mg/cm}^2$) but comparison of the shape of D(d,n) yields from this target and one uniformly loaded with deuterium suggests that the distribution of the implanted deuterium is peaked at $\sim 1.2 \text{ mg/cm}^2$ into the target and is not uniform.

Conclusion

We have demonstrated a technique for tritium depth profiling in host materials. Extension of the technique to deuterium depth profiling is straight forward. We estimate that with a 1 ns burst width and detector resolving times a depth resolution of 0.1 mg/cm^2 of titanium could be achieved for tritium and deuterium profiling. At that level, straggling in the host material would be the main resolution limitation. Sensitivity of 0.1 at. % with 10% accuracy can be obtained for tritium or deuterium. The technique may be used for sample thicknesses up to 20 mg/cm^2 . The depth profiling of other light nuclei, e.g. ^7Li and ^9Be , for which the ground state-first excited state separation is large is also possible.

This technique serves as a useful complement to the proton recoil method as a depth profiling method for some light ions in heavy metal hosts. The major advantages of the technique are that one may use thick or backed samples, which may be radioactive, that no elaborate sample preparation is required and that it is nondestructive. The disadvantages and limitations are that more sophisticated accelerator and detector systems are required and that the data reduction is more involved if absolute concentration values are desired.

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Figure Captions

- Fig. 1 Schematic layout of the experimental equipment.
- Fig. 2 Tritium depth profile from an unbombarded portion of a target.
- Fig. 3 Tritium depth profile from a bombarded portion of the same target. The solid line is a smooth curve drawn through the profile in Fig. 2. Differences in the maximum height and width of the profiles are discussed in the text.
- Fig. 4 Time-of-flight spectrum from 2.5 MeV deuterons on a bombarded portion of the target. Only even numbered channels have been plotted except on the peaks. Neutrons from the $^{12}\text{C}(\text{d},\text{n})$ reaction require more than 400 ns (the interval between current bursts) to reach the detector--thus the double energy scale.

Tritium Concentration