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ScienceDirect
Journal of Nuclear Materials 367–370 (2007) 1596–1600
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Neutron elastic recoil detection for hydrogen isotope analysis in fusion materials

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Abstract

The neutron elastic recoil detection analysis (NERDA) using a 14.2 MeV neutron beam has been developed to measure depth profiles of hydrogen isotopes up to several hundred micrometers under low background conditions. Results of a proof-of-principle experiment using polyethylene and deuterated polyethylene samples were presented. The detected energy spread due to the geometrical configuration was corrected to estimate depth profiles by an unfolding technique based on the iterative Bayes' estimation. Hydrogen isotope depth profiles in the inner baffle plate of JT-60U were also measured as an application of NERDA. The hydrogen profile from the surface to 800 μm had three components, while a few counts of deuterium were detected. The detection limit of about 60 ppm was deduced for deuterium.

1. Introduction

Hydrogen isotopes show complicated behavior on the surface of Plasma Facing Components (PFCs) in fusion devices. The study of the behavior is important for the design of the fuel recycling, plasma control, safe management of tritium inventory, etc. If carbon-based materials as PFC, co-deposited layers of carbon compounds are formed with the thickness of several tens of micrometers on PFC surfaces due to plasma–wall interactions [1–4]. Therefore, it is necessary to measure hydrogen isotope depth profiles in these layers. Conventional non-destructive Ion Beam Analysis (IBA) has been applied to the measurement of deuterium and tritium retention on PFCs [5–7]. However, the range of IBA is limited because of the shallow probing depth.

A powerful and unique method employing isotropic 14 MeV neutrons instead of an ion beam, namely, neutron elastic recoil detection analysis (NERDA) has been developed. The advantages of neutron usage for the material analysis are the ability to probe the deeper depth with the same sensitivity as the conventional ion beam analysis due to large scattering cross sections for hydrogen isotopes. In addition, beam-irradiation induced degradation of the hydrogen isotope profile could be minimized. This method was applied to measure hydrogen isotopes in divertor tiles of the DIII-D Tokamak [8] and limiter tiles of the JET Tokamak [9].

In this study, we propose NERDA using the 14.2 MeV neutron beam to extend the analyzing

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doi:10.1016/j.jnucmat.2007.04.025

depth of hydrogen isotopes up to several hundred micrometers under low background conditions. A proof-of-principle experiment was made using standard samples of polyethylene $(C_2H_4)_n$ and deuterated polyethylene $(C_2D_4)_n$ films containing a known concentration of hydrogen isotopes. The application of NERDA was also demonstrated using a PFC sample of JT-60U.

2. Principle of NERDA

A depth profile of target atoms in a sample is estimated from the detection energy and yield of recoil particles elastically scattered by the probing neutrons. The detection energy E_d of the recoil particle is expressed as:

$$E_d = E_r - dE_2(x), \tag{1}$$

where E_r and $dE_2(x)$ are the recoil energy and the energy loss of the recoil particle before reaching the sample surface. The energy E_r is set by the energy and momentum conservation laws for the neutron and recoil particle. The energy loss term is expressed using stopping power for the recoil particle in the sample.

The yield dY of the recoil particles originating in the region between a depth x and $x + dx$ is expressed by:

$$dY = Q\frac{d\sigma_r(E)}{d\Omega}N\,dx\,d\Omega, \tag{2}$$

where Q is the number of incident neutrons, $d\sigma_r(E)/d\Omega$ is the angular differential recoil cross section in the laboratory system [10,11], N is a density of the target atom, $d\Omega$ is a solid angle of the detector.

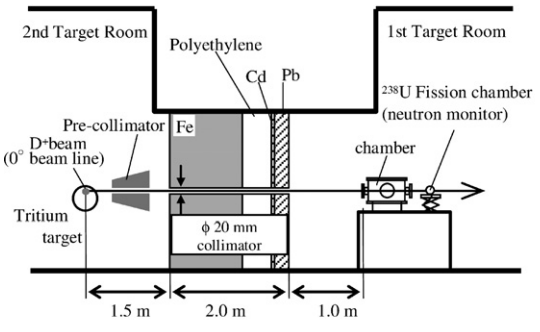
In fact, the profile has uncertainty based on the energy spread due to the geometrical spread of the detection system, the energy straggling of the recoil particle in the sample, the intrinsic resolution of the detector, etc. An unfolding technique based on the iterative Bayes' estimation [12] is used to correct the uncertainty. Using a yield $Y_i(E)$ for the ith energy channel of a measured spectrum ($i = 1,2,\ldots,m$), a yield $Y_j^{(\ell)}(x)$ for the jth depth channel of an estimated spectrum ($j = 1,2,\ldots,n$) revised by ℓth iteration is calculated as follows:

$$Y_j^{(\ell)}(x) = \sum_{i=1}^{m}\left(Y_i(E)\times\frac{Y_j^{(\ell-1)}(x)r_{ij}}{\sum_{j=1}^{n}Y_j^{(\ell-1)}(x)r_{ij}}\right), \tag{3}$$

where r_{ij} is a matrix element of a normalized response function $\boldsymbol{R}(E,x)$. The response function is calculated using the Monte Carlo particle transport code SRIM2003 [13], as discussed in Section 4. In Eq. (2), the estimated spectrum is used as each dY.

3. Experimental procedure

The experimental setup is shown in Fig. 1. Intense 14.2 MeV neutrons from T(d,n)^{4}He reactions were produced using the Fusion Neutronics



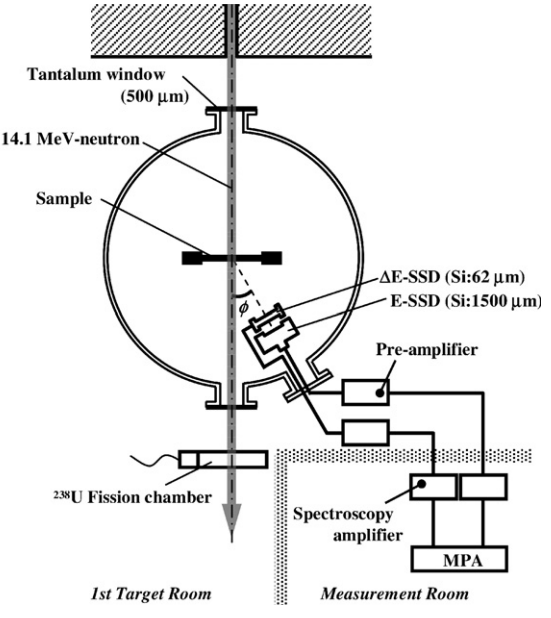


Fig. 1. Schematic of the experimental setup for NERDA.

Source facility of the Japan Atomic Energy Agency. A collimated neutron beam was produced from a fraction of the total neutrons using two collimators each with a hole 20 mm in diameter. The neutron beam was incident on the sample from the direction at an angle of 0°. Particles emitted from the sample were measured using a ΔE–E telescope system which consisted of a pair of Solid State Detectors (SSDs) and a Multi-Parameter Analyzer (MPA). The ΔE–E detector was positioned at the detection angle of 20° with the solid angle of 2.0×10^{-2} sr. Output signals from each SSD were analyzed with the MPA to estimate the particle mass and its energy simultaneously. A typical fluence of incident neutrons was 3.0×10^{13} neutrons/m^2, which was monitored with a ^{238}U fission chamber located behind the target chamber.

A proof-of-principle experiment was performed using standard samples of 50 μm, 500 μm-thick $(C_2H_4)_n$ and 100 μm-thick $(C_2D_4)_n$ films. The $(C_2D_4)_n$ film was fabricated using a solvent-cast method. The deuterium density of the sample surface was 4.6×10^{28}/m^3, which was measured using 4.5 MeV-α ERDA.

The PFC sample of JT-60U was also prepared from the Inner Baffle Plate (IBP) placed at the W-shaped divertor region. The sample was exposed to deuterium plasmas between June 1997 and October 1998. During this period, boronization treatments were preformed twice. The hydrogen discharges were done to clean the PFC surface after the deuterium plasma experiments [7].

4. Results and discussion

4.1. Hydrogen isotope profiles in polyethylene films

A typical ΔE–E contour map for the $(C_2D_4)_n$ sample analysis is shown in Fig. 2. The horizontal and vertical axes show the energy measured on the ΔE-SSD and the E-SSD, respectively. The D(n,d)n recoil deuterons were recorded around $(\Delta E, E) = (120, 680)$ ch. Protons originating in X(n,p) reactions with hydrogen, deuterium and other impurity on/in the target were also distinguishable. For example, D(n,p)2n reaction formed a continuum spectrum from (70, 640) ch to (300, 20) ch.

A deuterium depth profile was estimated using a $\Delta E + E$ total energy spectrum of recoil deuteron shown in Fig. 3. Since the spectral shape became rather broad, a detection probability of deuterons

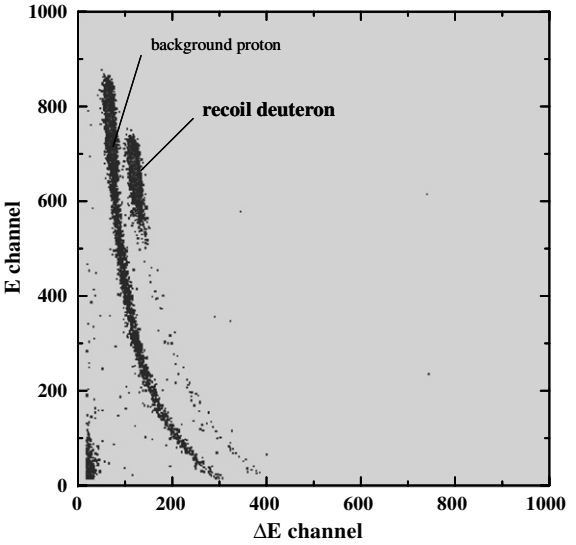


Fig. 2. Typical ΔE–E contour map on MPA obtained from the $(C_2D_4)_n$ sample. The horizontal and vertical axes indicate the detection energy of ΔE-SSD and E-SSD, respectively.

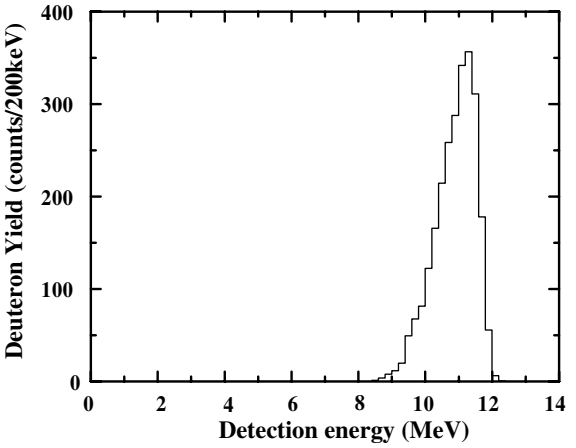


Fig. 3. Total energy $(\Delta E + E)$ spectrum of deuterons obtained from the contour map.

at energy E_i from each depth region $x_j \sim x_j + \Delta x$ were calculated to correct this uncertainty using the SRIM2003 code, as is shown in Fig. 4. The depth profile was estimated with a sum of these probabilities, i.e. the response function in Eq. (3).

The deuterium profile is shown in Fig. 5. The profile indicated a homogeneous distribution with the deuterium density of 4.5×10^{28}/m^3, which was in good agreement with the evaluated density described in Section 3.

The hydrogen profiles of both 50 and 500 μm-thick $(C_2H_4)_n$ samples also indicated homogeneous

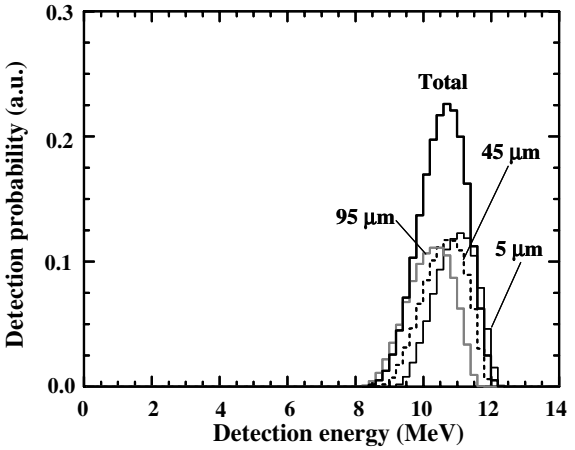


Fig. 4. Distributions of detection probabilities for the depth of 5 (solid line), 45 (broken line) and 95 μm (gray line) in the $(C_2D_4)_n$ sample, for example. The heavy line indicates the normalized total-probability calculated with them.

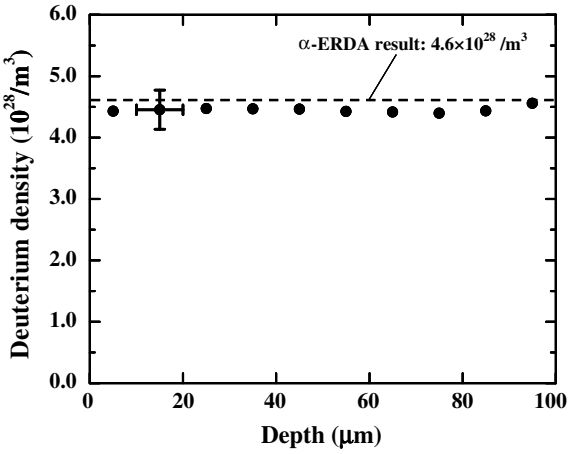


Fig. 5. Depth profile of deuterium in the $(C_2D_4)_n$ sample estimated using the total energy spectrum (Fig. 3) and response function (Fig. 4).

distributions with densities of 7.1 and $7.3 \times 10^{28}/m^3$, respectively, which were in fair agreement with the nominal density $7.9 \times 10^{28}/m^3$ of polyethylene.

4.2. Hydrogen isotope profiles in PFCs of JT-60U

The hydrogen energy spectrum of the IBP sample is shown in Fig. 6. Recoil protons formed a continuous spectrum from 13.7 to 2.6 MeV as the lower detection limit. There is the possibility that back ground reactions, $^{10}B(n,p)^{10}Be$ and $^{11}B(n,p)^{11}Be$, overlapped with the recoil proton spectrum. Fortunately, the contribution of both reactions could be

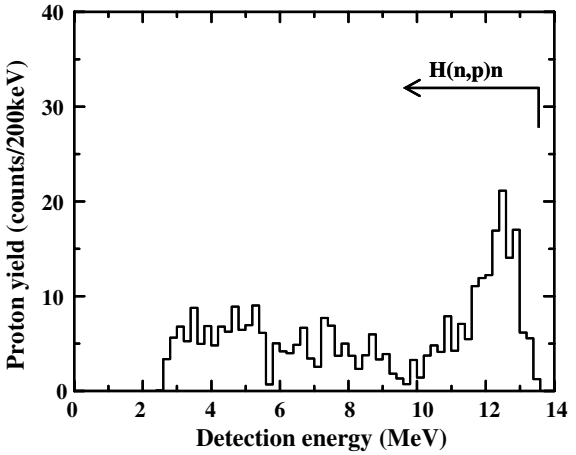


Fig. 6. Total energy spectrum of protons measured for the IBP sample. Protons below the energy of 2.4 MeV cannot be detected using the ΔE–E system.

negligible because deuteron yields of $^{10}B(n,d)^{9}Be$ or $^{11}B(n,d)^{10}Be$ reactions comparable to those proton yields were not observed in the contour map.

In Fig. 7, the hydrogen profile in IBP is shown together with that in a standard graphite sample for comparison. Hydrogen retention for IBP in regions (I) near the surface, (II) 200–400 μm and (III) 400–800 μm in depth was estimated to be 2.8, 0.3 and $3.2 \times 10^{22}/m^2$, respectively. The retention in IBP was 1.3 times as great as that in the graphite in the region (I) due to the hydrogen discharges. Less hydrogen retention in the region (II) might be attributed to the bake out during plasma experi-

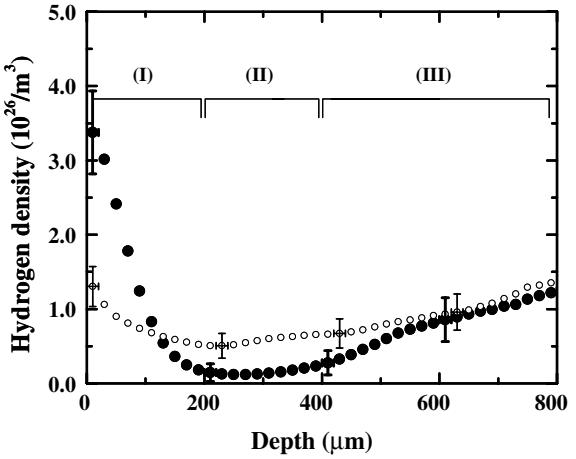


Fig. 7. Hydrogen profile in the IBP sample (closed circle) calculated from the proton spectrum shown in Fig. 6. Hydrogen profile in the graphite sample (open circle) is also indicated for comparison.

ments. In the region (III), IBP retained the same amount of hydrogen as the graphite. The total hydrogen retention of $6.3 \times 10^{22}/m^2$ was consistent for the result of thermal desorption spectroscopy [4].

A deuterium yield of only 7 counts was recorded during the analysis, which was not enough for accurate statistics. The estimated deuterium retention of $3.5 \times 10^{21}/m^2$ corresponded to that of $3.3 \times 10^{21}/m^2$ obtained by the deuteron induced nuclear reaction analysis for the same sample [7]. This implies that the detection limit of deuterium was less than 60 ppm for the IBP sample.

5. Summary

The elastic recoil detection analysis with a neutron beam has been proposed to extend the analyzing depth of hydrogen isotopes up to several hundred micrometers. The proof-of-principle experiment was performed using the standard sample of polyethylene films. With use of the neutron beam and ΔE–E system, proton and deuteron spectra were obtained separately under low background conditions. It is confirmed that the uniform depth profile of hydrogen isotope consistent with the nominal density was obtained by the iterative Bayes' estimation.

The PFC sample from JT-60U was analyzed as an application of NERDA. A hydrogen depth profile from the surface to 800 μm with three components was obtained. In contrast, few counts of deuterium were observed and the deuterium detection limit of 60 ppm was deduced. The NERD method will be applied to measuring depth profiles of hydrogen isotopes at different areas in PFC of JT-60U.

Acknowledgements

The authors appreciate being given the opportunity for the JT-60U sample analysis to be done by Dr N. Miya and the JT-60U team.

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