Sensitization of Solid State Nuclear Track Detector in Carbon Dioxide for Improved Fast Neutron Dosimeter

Monte Carlo Simulation and Experimental Results for Energy Response of Neutron Dosimeter and Quantitative Estimation of Sensitization by CO₂ Pre-etching

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(Received on January 13, 2011)

(Accepted on May 17, 2011)

A pre-etching step is applied with carbon dioxide (hereafter, CO₂ pre-etching) of a solid state nuclear track detector (SSNTD) to improve the sensitivity for protons detection. In this study, we quantitatively estimated the sensitization of SSNTD with CO₂ pre-etching to improve the sensitivity of a personal neutron dosimeter. We measured the etch pit size and the critical angle using a 1.7 MV tandem accelerator at the University of Tokyo to evaluate the proton sensitivity. In addition, we calculated the neutron energy response of SSNTD with a polyethylene radiator with the particle and heavy ion transport system multi-purpose three-dimensional Monte Carlo code. To evaluate the accuracy of these calculations, neutron energy responses of SSNTD with and without CO₂ pre-etching were measured at the Facility of Radiation Standards of the Japan Atomic Energy Agency. The neutron energy responses calculated by the above Monte Carlo code agreed with the experimental results to within 35%. The results obtained in this research indicate that CO₂ pre-etching at a pressure of 0.6 MPa for 3 days increases the radiator effect by ~250% for the 5 and 14.8 MeV neutrons.

KEY WORDS: solid state nuclear track detector, neutron dosimeter, Monte Carlo simulation code, PHITS, carbon dioxide pre-etching, proton irradiation.

I INTRODUCTION

Passive personal neutron dosimeters based on a solid state nuclear track detector (SSNTD) have been widely employed to measure the dose of external neutron exposure at various nuclear facilities. The commonly used dosimeters of this type have a simple, cheap, and robust design, which is comprised of a 1-mm thick polyethylene (PE1) radiator and a Columbia Resin #39 plastic (CR-39 plastic) SSNTD.⁽¹⁾ The proton sensitization effects on SSNTD with CO₂ at several atmospheres (CO₂ pre-etching) were reported in recent years.⁽²⁻⁴⁾ However, quantitative estimates of the sensitization effect caused by CO₂ pre-etching have not yet been performed for proton irradiation.

In this paper, we present quantitative results on the

sensitization of a SSNTD to protons with several incident energies caused by CO₂ pre-etching. Then, we calculate the energy response of a personal neutron dosimeter with and without CO₂ pre-etching using the particle and heavy ion transport system (PHITS) multi-purpose three-dimensional Monte Carlo code.⁽⁵⁾ The accuracy of the PHITS prediction for the energy dependence of the detector response for the PE1 radiator was verified by comparing the simulated and measured radiator effects in mono-energy neutron fields (144 keV, 5.0 MeV, and 14.8 MeV).

II EXPERIMENT

1. Proton Irradiation

The SSNTD used for this experiment was BARYOTRAK (C₁₂H₁₈O₇, 1.32 g cm⁻³, Fukuvi Chemical Industry, Japan), and exposures were performed with a 1.7 MV tandem accelerator, the Rutherford backscattering spectroscopic analyzer with particle induced X-ray emission and ion implantation devices (RAPID) produced by High Voltage Engineering Europe (HVEE), capable of accelerating protons to energies of 3.4 MeV. RAPID is installed at the Department of Nuclear Engineering and Management at the University of Tokyo.

The optimum etch-pit density recovered by the SSNTD and subsequently counted by an optical microscope is 2,000~3,000 tracks per square centimeter.⁽⁶⁾ However, RAPID's minimum

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Fig. 1 Schematic diagram of the small amount of ion irradiation system in ion implantation devices.

beam current is 0.01 nA, and the minimum fluence of ions into the ion implantation devices cause tracks to overlap. To address this issue, a minute amount of ion irradiation was introduced in each ion implantation device (**Fig. 1**). The proton beam passed through a collimator having a diameter hole of 3 mm and it irradiated a 2.5- μm -thick gold foil target. The SSNTD and silicon surface barrier detector (SSB) were indirectly hit by backscatter protons at an angle of 135° from the gold target. The SSNTD was set on the surface of an aluminum plate. The SSB measured the fluence and energy spectrum of the backscatter ions, and an 8-mm aperture slit in the center of the aluminum plate allowed the SSB to detect the ions. The energies used to irradiate the SSNTD were 1.0, 2.0, and 3.0 MeV. Note that these irradiation times are approximately 10 seconds. **Figure 2** shows the energy spectrum of backscatter protons for each irradiation condition. Under these conditions, the energy degradation with a full width half maximum was approximately 300, 250, and 200 keV, respectively. The irradiation provided 2,000~3,000

tracks on the detector surface.

After proton irradiation, the samples were separated into two groups. The first group was kept in a pressure vessel filled with CO_2 at a pressure of 0.6 MPa for 3 days (CO_2 pre-etching). The second group was kept in air at atmospheric air pressure for 3 days. Both these groups were chemically etched for 3.0 h in a circulating bath of 6.25 mol/l NaOH solution at a temperature of $90 \pm 0.4^\circ\text{C}$. Then, the etch pits were observed under an optical microscope (the Radometer 2000 made by Radosys, Ltd.) and were measured on PC using the PIXS 2000 Pro ver. 3.0.1 software (Inotech Co. Ltd., Japan).

2. Neutron Irradiation

To measure the sensitivity of a personal neutron dosimeter for mono-energy neutrons with and without applying CO_2 pre-etching, neutron exposures with a PE1 radiator were performed at the Facility of Radiation Standard (FRS) of the Japan Atomic Energy Agency (JAEA). To measure the energy response, the SSNTD with a PE1 radiator was irradiated with mono-energy neutrons of 144 keV, 5.0 MeV, and 14.8 MeV. After neutron irradiation, the etch pits density of these samples were measured by the same method described in section II-1 for the proton irradiation experiment.

III RESULTS

To determine the size of etch pits created by the impact of protons at different energies, the distribution of etch pit diameters for each experimental condition were measured (**Fig. 3**). Note that the mean etch pit diameter increases with decreasing energy, and that it is higher for the CO_2 pre-etched samples than for those that did not undergo the CO_2 pre-etching.

It is known that SSNTD can only detect charged particles within its critical angle. A critical angle is the maximum angles of incidence for a charged particle from countable etch pits on a SSNTD and it depends on the particle type and the incident energy. In this study, the critical angle was calculated from the experimental data. The critical angle φ can be determined from

Fig. 2 Energy spectrum of backscatter protons for each irradiation condition.

Fig. 3 Comparison of etch pit size distributions: (a) 1 MeV proton experiments without CO₂ pre-etching, (b) 1 MeV proton experiments with CO₂ pre-etching, (c) 2 MeV proton experiments without CO₂ pre-etching, (d) 2 MeV proton experiments with CO₂ pre-etching, (e) 3 MeV proton experiments without CO₂ pre-etching, and (f) 3 MeV proton experiments with CO₂ pre-etching.

the relationship

$$\cos\varphi = \frac{G^2 - r^2}{G^2 + r^2} \quad (1)$$

where φ is the critical angle [degree], G is the thickness of the layer removed during etching [μm], and r is the radius of the etch pit [μm]. We determined G by measuring the mean radius of etch pits made by fission fragment irradiations from ²⁵²Cf source in vacuum.⁷⁾ After fission fragment irradiation, the mean radius of etch pits made by fission fragment were measured by the same method described in section II-1 for the proton irradiation experiment with and without CO₂ pre-

etching. (Table 1) In addition, we determined the r values of each experiment from the mode of each etch pit size distributions (Fig. 3).

Figure 4 shows the critical angle of a proton as a function of the linear energy transfer (LET) in CR-39 calculated by equation (1). Note that the critical angle increases approximately as a quadratic function of the LET.

The quadratic approximation equations in Fig. 4 were calculated from Air and CO₂ data by the least square method:

$$\varphi_{\text{Air}} = 0.0323x^2 + 0.1788x - 8.9881, \quad (2)$$

$$\varphi_{\text{CO}_2} = 0.0149x^2 + 1.2659x - 10.732, \quad (3)$$

Table 1 The mean radius of etch pits made by fission fragment irradiations from ²⁵²Cf source in vacuum with and without CO₂ pre-etching.

	Without CO ₂ pre-etching	With CO ₂ pre-etching
Mean radius of etch pits (μm)	19.31 $\pm$ 0.20	19.61 $\pm$ 0.23

Fig. 4 Critical angle versus LET. The lines are quadratic fits to the data.

where ϕ_{Air} is the critical angle in the case without CO_2 pre-etching, ϕ_{CO_2} is the critical angle in the case with CO_2 pre-etching, and x is the LET ($keV \mu m^{-1}$) of the incident protons. Using these equations we calculated the critical angle of proton as a function of its energy (MeV) (**Fig. 5**) with the stopping and range of ions in matter code (SRIM).⁸⁾ Note that the critical angle decreases as the energy increases.

IV DISCUSSION

To study the change in the neutron energy response of a personal neutron dosimeter due to CO_2 pre-etching, we calculated the neutron energy response of a PE1 radiator for the above critical angle conditions using PHITS. The radiator effect, i.e., the increase in the detection efficiency, is defined as the number of charged particles per incident neutron that generate in the radiator, traverse it, reach the SSNTD region, and become observable as etch pits. The radiator effect RE is determined by:

$$RE = (T_{with} - T_{without}) / \Phi, \quad (4)$$

where Φ is the neutron fluence into the count area, T_{with} is the number of etch pits per count area with the radiator effect, and $T_{without}$ is the number of etch pits per count area without the radiator effect.

To calculate RE using PHITS, an event generator mode was set to consider nuclear reactions in the PHITS input file, and the cross sections were based on JENDL-3.3⁹⁾; in addition, the SPAR code¹⁰⁾ was applied to calculate the range of charged particles. Calculation of RE with PHITS consists of three steps: (1) setting the irradiating neutron's energy and angle of incidence, (2) tallying the distributions of proton energy and angle of incidence on the surface of the SSNTD, and (3) summing the number of detectable protons (i.e., using **Fig. 5** to determine and sum the detectable protons within a certain angle and energy range both with and without CO_2 pre-etching).

Figure 6 compares the calculated and experimental neutron energy dependences of RE for PE1. The uncertainty bars on the calculation results represent the statistical uncertainty

Fig. 5 Dependence of a proton's critical angle on its energy for SSNTD (BARYOTRAK) samples without (Air) and with (CO_2) pre-etching.

Fig. 6 Calculated and experimental dependence of RE for PE1 for neutrons from 100 keV to 20 MeV with and without CO_2 pre-etching.

(less than 5%). The uncertainty bars on the experimental results represent both the standard deviation of the number of etch pits within the counted area ($46.8 mm^2$) and the counting uncertainty due to the optical microscope. In **Fig. 6**, CO_2 pre-etching increases RE by $\sim 250\%$ for the 5 and 14.8 MeV neutrons, but does not increase it for the 144 keV neutrons; this may be caused by almost same critical angles below few hundred keV (**Fig. 5**). The underestimation of the calculated values of RE for 14.8 MeV neutrons compared to the experimentally measured values is probably due to not including heavy ions (alpha, deuteron, tritium, etc.) in the calculation. However, the agreement between the calculated and experimental data on the fluence response of RE is within 35% and thus, good overall. By determining each critical angle of SSNTD, we can calculate and design of neutron dosimeter using PHITS.

V CONCLUSION

The sensitization of a personal neutron dosimeter with CO_2

pre-etching was successfully achieved and verified by mono-energy neutron measurements. The calculated values of *RE* agreed well (within 35%) with the experimental values, both with and without CO₂ pre-etching, in the neutron energy range from 144 keV to 14.8 MeV. And CO₂ pre-etching increases *RE* by ~250% for the 5 and 14.8 MeV neutrons at a pressure of 0.6 MPa for 3 days. By determining critical angle of SSNTD, we can design neutron dosimeter and calculate neutron energy response using PHITS.

ACKNOWLEDGEMENTS

The authors thank Dr. Y. YAMAUCHI, Dr. K. ODA, Mr. H. FUKAO, and Mr. M. KAJIHARA of Kobe University for the valuable discussions and Mr. S. ITO and Mr. A. MORITA of the University of Tokyo for their help with the experiments at the RAPID.

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