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A performance test of a new high-surface-quality and high-sensitivity CR-39 plastic nuclear track detector – TechnoTrak

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ABSTRACT

We have studied the performance of a newly-commercialized CR-39 plastic nuclear track detector (PNTD), "TechnoTrak", in energetic heavy ion measurements. The advantages of TechnoTrak are derived from its use of a purified CR-39 monomer to improve surface quality combined with an antioxidant to improve sensitivity to low-linear-energy-transfer (LET) particles. We irradiated these detectors with various heavy ions (from protons to krypton) with various energies (30–500 MeV/u) at the heavy ion accelerator facilities in the National Institute of Radiological Sciences (NIRS). The surface roughness after chemical etching was improved to be 59% of that of the conventional high-sensitivity CR-39 detector (HARZLAS/TD-1). The detectable dynamic range of LET was found to be 3.5–600 keV/μm. The LET and charge resolutions for three ions tested ranged from 5.1% to 1.5% and 0.14 to 0.22 c.u. (charge unit), respectively, in the LET range of 17–230 keV/μm, which represents an improvement over conventional products (HARZLAS/TD-1 and BARYOTRAK). A correction factor for the angular dependence was determined for correcting the LET spectrum in an isotropic radiation field. We have demonstrated the potential of TechnoTrak, with its two key features of high surface quality and high sensitivity to low-LET particles, to improve automatic analysis protocols in radiation dosimetry and various other radiological applications.

1. Introduction

It is well known that CR-39 plastic is the most sensitive nuclear track detector material for energetic heavy ions [1]. The CR-39 plastic nuclear track detector (PNTD) is employed widely for radiation dosimetry purposes, for example, in personal neutron dosimetry [2], natural environment radiation (radon) dosimetry [3], space radiation dosimetry [4], and alpha-autoradiography [5]. However, the CR-39 PNTD also holds much promise for non-dosimetric applications, including in the fields of laser-driven ion beam spectroscopy [6], high-energy cosmic ray physics, and nuclear physics [7]. This is because of specific features of the CR-39 PNTD, specifically (1) its spectroscopic resolution, (2) its spatial resolution, and (3) its detection threshold. (1) CR-39 allows measurement of the adjacent nuclear charge and mass of heavy ions, as well as the linear energy transfer (LET) of individual particles, with excellent resolution [8–10]. (2) Since the nuclear tracks recorded by the CR-39 are observed by microscope, the track position and its trajectory in the detector can be reconstructed with sub-micron resolution [11,12]. (3) CR-39 captures only tracks with ionization energy loss rates greater than its detection threshold [13]. Any tracks with ionization energy loss rates below this threshold do not appear, and this allows detection of only the heavy ions in mixed radiation fields that contain photons and low-LET particles. For example, this feature is applicable for measuring only secondary high-LET particles in the therapeutic proton beam [14].

Several types of CR-39 materials are provided by different companies and under different trade names around the world. For example, USF-4 (United States) [15], TASTRAK (United Kingdom) [16], and HARZLAS/TD-1 (Japan) [17] are commonly used for space radiation dosimetry. These, along with INTERCAST (Italy) [18] and PAGE (United Kingdom) [19], are often used for nuclear fragmentation measurements. BARYOTRAK (Japan) [20] is provided as a neutron personal dosimetry product. The specifications of these materials, such as detection threshold, track registration sensitivity for particles with various LET, and surface quality after chemical

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etching, strongly depend on the production procedure (the curing cycle and conditions), the quality of the monomer used, and the addition of antioxidants. For example, BARYOTRAK is made from 99.7% purified monomer and provides a uniform and clean surface without any distortions or bubble-like noise caused by etching damage [21]. This makes automatic analysis possible for personal neutron dosimetry. The track registration sensitivity is, however, lower than that of other CR-39 materials, giving a high-LET threshold of $\sim$15 keV/μm in water [20]. Conversely, the addition of small amounts of the antioxidant Naugard-445 (in the case of HARZLAS /TD-1) and copolymerization with NIPPAm (HARZLAS/TNF-1) increases sensitivity to low-LET particles, giving low LET thresholds of 3.5 keV/μm and 2 keV/μm, respectively [22]. However, this results in poorer surface quality after etching, resulting in distortion and noise caused by chemical damage. The capabilities of automatic analysis are then limited by track-like noise caused by surface roughness. At present, high- (low-) sensitivity materials tend to have low (high) surface quality, resulting in a trade-off to obtain either property.

Recently, materials made from 99.7% purified monomer, as well as BARYOTRAK with the addition of a small amount (0.01 wt%) of antioxidant (IRGANOX1010), have been commercially provided as "TechnoTrak" by Chiyoda Technol Corporation, Japan. TechnoTrak is provided as a personal neutron dosimetry service like BARYOTRAK. The performance of fast neutron dosimetry has previously been analyzed elsewhere [23]. This approach of combining a purified monomer and antioxidant should hopefully lead to a detector with both a high-quality surface and high sensitivity to low-LET particles. In this paper, we verify the systematic performance of TechnoTrak in terms of surface quality, track registration sensitivity for various heavy ions, detection threshold, angular dependence of the track sensitivity, and spectroscopic resolution, and we also provide a comparison with the conventional products BARYOTRAK and HARZLAS/TD-1.

2. The principle of heavy ion detection

A charged particle passing through a nuclear track detector leaves a radiation damage trail (called a latent track) along its trajectory. The etchant is removed preferentially along the latent track at the track etch rate (V_t), while it is also removed from the surface generally at the bulk etch rate (V_b). As a result, a conical etched pit appears in the detector. The track registration sensitivity (S) is defined as the reduced etch rate ($S \equiv V_t/V_b - 1$), and depends on the ionization energy loss including ejected secondary electrons (δ-rays). It is known that high-energy δ-rays carry energy away from the track core region and hence do not contribute to latent track formation. S is thought to be a function of the restricted energy loss (REL) [24,25], which is defined as the energy loss rate along the track core region near the particle trajectory, taking into account the contribution from relatively low-energy δ-rays. In the case of CR-39, a δ-ray cut-off energy (ω_0) of 200 eV has been determined empirically [20,26]. The REL (ω_0 = 200 eV) in CR-39 [MeV cm^2/g] is finally converted into an LET (∞) in water (L) [keV/μm] using a conversion function [27].

S is obtained from the geometrical parameters of the etched pit [28]:

$$S \equiv \frac{V_t}{V_b} - 1 = \sqrt{\frac{16B^2D_A^2}{\left(4B^2 - D_B^2\right)^2} + 1} - 1. \tag{1}$$

Here D_A and D_B denote the major and minor axes of the elliptical opening of the etched pit, respectively, and B denotes the amount of bulk etching ($B = V_b \cdot t$, where t is the etching time).

3. Experiment

TechnoTrak plates (0.9 mm thick), polymerized using 99.7% purified monomer with 0.01wt% of antioxidant IRGANOX1010, were provided by Chiyoda Technol Corporation, Japan. These newly-commercialized detectors were irradiated with various heavy ions from protons (Z = 1), helium (Z = 2), carbon (Z = 6), neon (Z = 10), silicon (Z = 14), argon (Z = 18), iron (Z = 26) and krypton (Z = 36), with various primary energies from 30 MeV/u to 500 MeV/u, in the AVF-Cyclotron and the Heavy Ion Medical Accelerator in Chiba (HIMAC) at the National Institute of Radiological Sciences (NIRS). The range of LET values in water goes from 3.2 keV/μm to 600 keV/μm. Poly Methyl Methacrylate (PMMA) absorbers with water-equivalent thicknesses were set in front of the detectors to act as energy degraders and sources of lighter fragments from primary ions. The detectors were irradiated by beams at different dip angles from $\theta = 90°$ (perpendicular to the detector surface) to 15° with intervals of 15° or 10°. Each irradiated fluence was monitored and controlled to be about 1500 cm^{-2} using a plastic scintillation counter set beside the CR-39. The incident energy and REL were calculated using the range-energy calculation code SRIM [29] and Benton's formula [24,25], respectively.

All of the detectors were etched in a 7 mol/l sodium hydroxide solution at 70 °C for 24 h. For obtaining the bulk etch rate (V_b), the etching time was varied from 2 to 24 h in intervals of 2 h. Several research groups use materials other than HARZLAS, which require different etching conditions. This can include different concentrations, temperatures, and durations, but can also involve etching in a completely different solution such as potassium hydroxide (KOH). It may be possible to improve the surface quality by optimizing the etching conditions. In this work, we compare the performance of several materials (TechnoTrak, BARYOTRAK, and HARZLAS/TD-1) under the same etching conditions, which we have employed over many years.

A microscopic image of an etch pit was automatically taken using a high-speed imaging microscope (HSP-1000, SEIKO Precision Inc., Japan) [30]. The typical scanned image size was 4 mm by 4 mm. A massive scan of size 30 mm by 30 mm was also carried out for determining the charge resolution with projectile fragments. The pixel resolution was 0.35 μm under an objective lens of ×20.

The track size (D_A and D_B) of the etch pit was analyzed using PitFit software [30]. The amount of bulk etching (B) was measured by a direct measurement of the change in the detector thickness before and after etching using a micrometer. The track registration sensitivity (S) was obtained using Eq. (1).

An atomic force microscope (AFM) image was also taken using an AFM (Dimension V; Veeco) to quantitatively compare the surface quality after etching. Images of size 10 μm by 10 μm with 1024-pixel resolution were scanned under the tapping mode of operation.

4. Results and discussion

4.1. Etching properties and surface quality

The variation in the amount of bulk etching (B) as a function of etching time is shown in Fig. 1. The bulk etch rate (V_b) corresponds to 2.1 μm/h, which is similar to that of the other CR-39 materials (BARYOTRAK [31] and HARZLAS/TD-1 [32]) in 7 mol/l NaOH at 70 °C. The microscopic images of the etching pits formed by Fe (421 MeV/u) and the atomic force microscopic (AFM) images after etching for 24 h are compared for BARYOTRAK, HARZLAS/TD-1, and TechnoTrak in Figs. 2 and 3, respectively. The opening mouth of the etch pit is clearly identifiable in each material, while the quality of