Development of Etched Track Detector System for Low Fluxes of Thermal Neutrons

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ABSTRACT

A new etched track detector system for measuring low fluxes of thermal neutrons was developed. The detector consists of two foils of CR-39 etched track detector in close contact. One of the foils has a thin layer of $^{10}$B implanted just below the surface. The detection of thermal neutrons through products of $^{10}$B(n, $\alpha$)$^7$Li reaction with a pair of CR-39 etched track detectors in close contact during irradiation and chemical etching of latent tracks was studied. After counting, only tracks, produced by the same absorbed neutron, which leaves tracks of alpha particle on surface of one detector and tracks of lithium ions on surface of other detector, were taken into account. In this way, by utilising coincidence detection method, the background due to objects that can not be separated from tracks by automatic counting system was significantly reduced. A lower neutron detection limit was found to be $<10^5$ neutrons/cm$^2$ (for 1 cm$^2$ detector’s area) and is two orders of magnitude lower than that obtained by an increase of the implanted $^{10}$B density or by an increase of the counted detector area.
1 INTRODUCTION

Limited sensitivity and unpredictable background are the major drawbacks of Solid State Nuclear Track Detectors (SSNTD) in the assessment of low fluences of thermal neutrons. The latest arises from impurities in the detector and tracks, which do not originate from neutron induced events. Counting coincidence tracks on two detectors foils, in contact during irradiation, has effectively solved this problem. The emphasis of the present study is given on the evaluation of the response (tracks/neutron) of such a detector system. For this reason the calculation and experimental measurement of the response of the described coincidence detector was carried out. Thus, this work deals with the development of the passive time-integrating dosimeter for environmental monitoring of low fluences of thermal neutrons. The dosimeter is based on the $^{10}$B(n, $\alpha$)$^7$Li reaction and utilizes SSNTD to record alpha particles and lithium ions (Fig. 1). The track parameters (track size, optical properties of tracks) of low energy alpha particles (1.48 MeV) and lithium $^7$Li ions (0.83 MeV) were studied.

Figure 1: Thermal neutron dosimeter – schematic representation of the doped CR-39 by implantation of thin boron layer (first detector) pressed against a CR-39 track detector (second detector). After etching tracks were counted on the surfaces denoted by A and B. The detector layer removed by chemical etching is denoted by h.

The response of dosimeter for detection of fast neutrons with a pair of CR-39 SSNTDs in close contact was already studied (Lengar, 2001). The advance of this method is to eliminate the background by using the combination of CR-39 doped with implanted thin layer of $^{10}$B at energies of 30 keV at $5.0 \times 10^{16}$ ions/cm$^2$ under surface pressed against another CR-39 and evaluation of coincidence tracks – pairs of tracks from alpha particles and from $^7$Li ions. After counting, only the tracks found on the same spot on both surfaces (produced by the same neutron) were taken into account. These tracks are called coincidence tracks. Since the probability of track-similar defects being on both detectors at the same spot is small, they can
be to the large extend excluded from the signal. With the same detector configuration neutrons can be detected in the standard way, namely by deriving the fluence from the measured track density on the single detector foil.

Compared to the dosimeter, where a BN-1 (Kodak Pathe) thick layer was used (Fleischer et al., 1975; Ilić et al., 1995) and dosimeter described three years ago (Izerrouken et al., 2003), where a very thin layer of implanted $^{10}$B in silicon was used, our dosimeter is one step further. Thin neutron converter ~70 nm at the depth of about 0.1 μm offers coincidence tracks observation and evaluation.

2 EXPERIMENTAL

Neutron irradiation was carried out in the thermal column of the TRIGA Mark II reactor of the Jožef Stefan Institute, Ljubljana. For calibration measurement of the neutron fluence the activation analysis method with $^{197}$Au-foils was used. Neutron flux was $3.67 \times 10^5$ cm$^{-2}$ s$^{-1}$ and the Cd ratio was about 10. Detector foils of CR-39 made by Intercast (Parma, Italy) were used. For every measurement first detector with implanted boron was tightly pressed on the second detector, so that there was no air gap between the two surfaces. We can consider the $^{10}$B(n, α)$^7$Li reaction as isotropic for the entire neutron energy region. In all experiments the incident neutron beam was perpendicular to the detector surface.

After irradiation the detector foils were chemically etched in 6.25 N NaOH at 70 °C for a selected time. The scanning and analysis of the etched detector foils were performed by a TRACOS automatic image analysis system (Skvarč, 1993). The criteria used whether a track should be considered were based on the properties of the TRACOS automatic system, and may be summarised as follows: (i) the major axis of the eliptically shaped track opening must be larger than 2 μm; (ii) the grey level treshold was established empirically by observing tracks from alpha particles and lithium ions, two tracks were decided to be coincident if the distance between them was less than 8 μm; and (iii) criterion for the determination of coincidence tracks is their shape and orientation (Lengar, 2002).

It should be noted that the criteria for deciding whether a track should be registered or not, due to their simplicity, are the largest possible sources of error. This refers more to criterion especially for other particles, i.e. recoil protons or alpha particles from radon and its daughters whose detection efficiency can be slightly higher, since the reference observations were performed for alpha particles and lithium ions emerged from boron only (Izerrouken, 1999). To overcome the above mentioned difficulties with the first detector doped CR-39 with implanted $^{10}$B, a special two step etching and analysis procedure was developed. First the detector was etched for few minutes and $^{10}$B layer was removed and then the analysis procedure was continued at the same conditions with the second detector.

This work was performed to acquire the following: (i) to determine the response of the coincidence detector - this is given as the number of detectable tracks per incident neutron, and (ii) to obtain the best value for the thickness of the detector layer removed during chemical etching and the calculated ratio between the number of the coincidence tracks and between the number of tracks found on each single detector.

3 RESULTS

The major part of reaction products - alpha particles and $^7$Li ions, emerge from boron with the energies 1.48 and 0.83 MeV (ENDF/B-VI, 1991; Ziegler, 1996), hit the first detector with the same energy and hit the second detector with energies of 1.45 and 0.80 MeV, respectively. The results obtained showed that the thermal neutron dosimeter described has the following characteristics:
The use of a very thin layer of \( {^{10}}B \) as converter and adequate etching parameters (Izerrouken, 1999), where the detector layer thickness removed was \( \sim 2 \, \mu m \), allows easy separation between the tracks of alpha particles, lithium ions and background. In Fig. 2 the gray level vs major axis of \( {^{10}}B(n, \alpha)^{7}\text{Li} \) reaction product tracks obtained at a removed thickness of 2 \( \mu m \) is presented. Two kinds of tracks are clearly observed at both detectors. These are tracks of \(^{7}\text{Li} \) ions and those of alpha particles. The energy of Li ions is up to 0.83 MeV and has a smaller range in CR-39. The tracks are smaller and brighter compared to alpha particle tracks which are in major part bigger and darker. Discrimination between alpha particles and Li ions under these conditions is very simple. For selection of coincidence tracks along with sizes also their exact positions relative to markers (coordinates) were stored. From the two lists of measured tracks a match of coincidence tracks was found by an automated procedure and reevaluated by hand, too.

The energy resolution can be calculated by the following equation (Skvarč, 1999):

\[
\frac{\Delta E}{E} = \left[ \frac{(E_1 - E_2)}{(D_1 - D_2)} \right] \times \frac{1}{2} \left[ \frac{1}{2} (E_1 + E_2) \right] \Delta D
\]

where the \( E_1 \) and \( E_2 \) are the energies of incident alpha particles and \(^{7}\text{Li} \) ions, respectively, \( D_1 \) and \( D_2 \) are the mean track diameters corresponding to each energy and \( \Delta D \) is the width of the distribution. \( D_1, D_2 \) and \( \Delta D \) are determined by fitting the track size distributions from the Fig. 2. Taking into account the straggling of alpha particles and \(^{7}\text{Li} \) ions in air as determined by the SRIM program and error due to inaccuracy in the measurement of the source-detector distance a resolution of about few 100 keV is obtained from the equation. It was observed that

![Figure 2: Separation between the tracks of alpha particles, lithium ions and background: (A) grey level vs major axis of \( {^{10}}B(n, \alpha)^{7}\text{Li} \) reaction product tracks in CR-39 doped by implantation of \( {^{10}}B \) of 30 keV at \( 5 \times 10^{16} \) ions/cm\(^2 \) (first detector) and CR-39 (second detector); and (B) tracks of alpha particles lithium ions, background tracks and impurities. The highest resolution is easily obtained when the layer thickness removed from the detector just exceeded the range of \(^{7}\text{Li} \) ions in CR-39.](image)
(ii) The values of the mean track diameter of alpha particles and of $^7$Li ions for a selected layer removed from the detector were investigated. The corresponding distributions of track size (diameter / major axis) are presented in Fig. 3.

(iii) The calculation (and experimental estimation) of the response for the described coincidence detector was found to be approximately the same as the response to lithium ions on both detectors and the values of the detector responses are shown in Table 1.

![Figure 3: The corresponding distributions of track size (diameter / major axis (MAJ)) in »pixels« (1 pix is app. 0.35 μm) - etched track size distribution of alpha particles and $^7$Li ions after selected layer thickness $h$ are removed from the detector.](image)

Table 1: Response $K_i$ and background density $\rho_i$ for detector. Subscripts (1) and (2) denote tracks in first and second foil and (c) coincidence tracks

<table>
<thead>
<tr>
<th></th>
<th>Response (tracks/neutron)</th>
<th>Background (tracks/cm²)</th>
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<tbody>
<tr>
<td>First detector</td>
<td>$K_{1, \alpha} = 0.79 \times 10^{-4}$ (1 ± 0.10)</td>
<td>$\rho_1 = 45$ (1 ± 0.2)</td>
</tr>
<tr>
<td></td>
<td>$K_{1, Li} = 0.76 \times 10^{-4}$ (1 ± 0.10)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$K_1 = 1.56 \times 10^{-4}$ (1 ± 0.20)</td>
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<tr>
<td>Second detector</td>
<td>$K_{2, \alpha} = 0.79 \times 10^{-4}$ (1 ± 0.10)</td>
<td>$\rho_2 = 45$ (1 ± 0.2)</td>
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<tr>
<td></td>
<td>$K_{2, Li} = 0.76 \times 10^{-4}$ (1 ± 0.10)</td>
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</tr>
<tr>
<td></td>
<td>$K_2 = 1.56 \times 10^{-4}$ (1 ± 0.20)</td>
<td></td>
</tr>
<tr>
<td>Coincidence tracks</td>
<td>$K_c = 1.52 \times 10^{-4}$ (1 ± 0.05)</td>
<td>$\rho_c = 0.1$ ± 0.1</td>
</tr>
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</table>

In the fluence region where the errors due to false coincidences and background are small enough, there exists a linear relation between the track density $\rho_i$ and the neutron fluence $\Phi_i$:

$$\Phi_i = [\rho_i - \rho_{i0}] / K_i$$  \hspace{1cm} (2)$$

where the $\rho_{i0}$ is the background and $K_i$ is the detector response (tracks/neutron). Index $i$ stands for the first detector (1), second detector (2) or for the coincidence (c) tracks, respectively. A calibration curve is shown in Fig. 4.
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Figure 4: Calculation of present characteristics of the advanced detector (for boron density of $5 \times 10^{16}$ ions/cm$^2$ in CR-39) – track density vs neutron fluence for single (A) and coincidence tracks (B).

With an evaluation of coincidence tracks only the background is eliminated. This improves the signal to noise ratio. With an appropriate selection of the implanted $^{10}$B density further increasing of signal to noise ratio is achieved and hence the lower limit of detection is reduced down to $10^4$ neutrons/cm$^2$ or even more $<10^3$ neutrons/cm$^2$ if we increase the detector area. We could also adjust the upper limit of detection to be $>10^{10}$ neutrons/cm$^2$.

4 CONCLUSIONS

The main characteristics of the dosimeter developed are: (i) high sensitivity (very low fluences of thermal neutrons can be measured), (ii) high selectivity (applying the coincidence technique the signal due to background is practically eliminated), and (iii) response stability to meteorological conditions (moisture collected on the detector and track fading are eliminated). The dosimeter has already been applied for measuring of neutron exposure induced by cosmic radiation of air crew who are liable to be subject to exposure to more than 1 mSv per year. It has been found that the dosimeter could be used for dosimetry in working environment, personal dosimetry and also for detection of neutron sources.

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REFERENCES


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