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Plastics as nuclear track detectors for thermal neutron dosimetry

S M FARID, A P SHARMA and S A DURRANI*
Department of Physics, Kurukshetra University, Kurukshetra 132 119, India
*Department of Physics, Birmingham University, England (UK)

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Abstract. An attempt is made to determine the response of CR-39 and cellulose nitrate plastic track detectors subjected to thermal neutrons. The α-particles are produced from (n, α) reactions in lithium tetraborate convertor placed in contact with different plastics and are recorded in the detectors. The corrected track density gives a fluence sensitivity and dose sensitivity of the order of 10^{-4} tracks per neutron and 10^2 tracks/cm^2 mrem respectively. A linear relationship is observed between track density and neutron fluence.

Keywords. Solid state nuclear track detectors; neutron dosimeter; fluence sensitivity; dose sensitivity; spark counter; electrochemical etching.

1. Introduction

Solid state nuclear track detectors (SSNTDs) are increasingly being used for neutron dosimetry. Neutrons can be detected either intrinsically on some detectors or extrinsically. Extrinsic detection normally requires an external radiator (convertor foil) placed in contact with the detector to produce charged particles. Nuclides having a high (n, α) cross-section, *e.g.* ^{6}Li and ^{10}B have been employed as external radiators for thermal neutron dosimetry. In this paper we report the fluence and dose sensitivities of CR-39, CA 80-15 and LR-115 subjected to thermal neutrons. To avoid slow and tedious evaluation with optical microscope, the spark counting technique for LR-115 and electrochemical etching technique for CR-39 are also attempted.

2. Production of α-particles in lithium tetraborate

The α-particles produced as a result of the reaction in the radiator will leave damaged tracks in the detector placed in contact with the target. The number of tracks in the detector will be reduced as soon as the source is sufficiently thick to stop the most obliquely moving α-particles. The question now arises as to how many of α-particles reach the detector from a thick source.

Let us consider that there are n disintegrations per sec/unit volume in the material of the source (radiator) and that the range of each α-particle in this material is r. Let us assume a thin layer of thickness dx at a distance x below the surface (figure 1.) Only those particles reach the surface which are emitted within an angle θ to the normal to the surface, where θ is given by $\cos \theta = x/r$. This limits

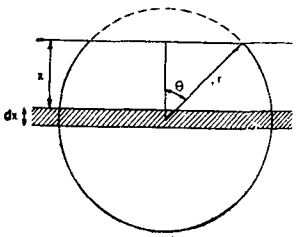

Figure 1. Calculation of α-particle emission from a thick source.

them to a cone of semivertical angle θ, which contains a solid angle $2\pi(1 - \cos\theta)$.

The number of disintegrations/sec within the layer of thickness dx is ndx per unit area. Since the α-particles from these will be distributed over a solid angle of 4π, the number from this layer which reach the surface is given by

$$\frac{2\pi(1-\cos\theta)}{4\pi} n\,dx/\text{unit area/sec.}$$

If t is the thickness of the source, then the total number of α-particles reaching the surface per second n_t, is given by

$$n_t = \frac{1}{2} n \int_0^t \left(1 - \frac{x}{r}\right) dx$$

When $t = r$, $n_t = \frac{1}{4} nr$/sec/unit area.

No particles reach the surface from layers below $t = r$. Thus the number of α-particles which reach the surface of a thick source is just a quarter of the number of disintegrations which take place within a layer of the source one α-range thick.

Following, the procedure described by Khan *et al* (1976) we have obtained the relationship between track density, ρ_T and neutron fluence, $F = \phi t$ as,

$$\rho_T = 6{\cdot}65 \times 10^{-4}\ \text{F/cm}^2.$$

The theoretical and experimental values of track density for different neutron fluencse show good agreement. It may be mentioned that the triton ejected as (n, α) reaction product does not produce any etchable damage trail (Khan *et al* 1976). But ^{7}Li which is also ejected as (n, α) reaction products produce etchable damage trail (Palfalvi 1982). Since the range of ^{7}Li nuclei in the detectors is very small ($\sim 2\mu$m), the tracks produced by ^{7}Li nuclei will be completely removed due to bulk etching of the detectors under specified etching conditions. Thus their contribution is neglected in determining the track density.

3. Experimental procedure

3.1 *Irradiation technique*

The plastic foils, CA 80-15 and LR-115, (M/s Kodak Pathe, France) are cellulose nitrates having chemical composition $C_6H_8O_9N_2$. The other plastic is CR-39 (M/s Pershore Mouldings, UK) having chemical composition $C_{12}H_{18}O_7$. Thin sheets are cast from allyl diglycol carbonate monomer (M/s Arinor, Paris). The thermal neutron irradiation facility of Reactor group in Birmingham University (Benitez 1978) is used for the present study. Cellulose nitrate plastic (LR-115) coated with lithium tetraborate ($Li_2B_4O_7$) layer of 15 μm thick has been used as convertor foil.

To prevent any scattering of α-particles or decrease in its energy, detector samples are tightly packed together with the external radiator. The samples are then exposed to thermal neutron flux of $1{\cdot}4 \times 10^3$ n/cm^2/sec and to the total fluence ranging from $2{\cdot}52 \times 10^6$ to $4{\cdot}94 \times 10^8$ n/cm^2. After irradiation the samples are stored in a refrigerator. The low temperature reduces the probability of track fading, and the enclosed atmosphere keep the background level low.

3.2 *Etching technique*

CR-39 ($\sim$ 50 μm thick), CA 80-15 ($\sim$ 100 μm thick) and LR-115 having a thin ($\sim$ 13 μm) layer of intensely red-dyedcellulose nitrate on a thick (100 μm) polyester base are used in the present study. Table 1 shows the methods by which a thermal neutron incident on lithium tetraborate can produce (n, α) interaction. It is observed that the energy of α-particles produced ranges from 1·5 to 2·05 MeV. It is therefore necessary to determine the optimum etching condition beforehand for different plastics irradiated with α-particles of different energies. A gold-covered ^{241}Am source is used to irradiate the plastics. The α-particle energy is varied by covering the source with Mylar foils of different thickness. The energy of α-particle is always determined by the surface barrier detector. Plastic samples are irradiated with α-particles in the range 1·5 to 4·1 MeV. Track density is determined by etching the sample in 6 M NaOH solution at 60°C for different intervals of time. The plots of track density versus etching time for α-particles of different energies reveal that almost 100% of the tracks are etched in different plastics under the following etching conditions: CR-39: 3 hr etching in 6 M NaOH at 60°C. CA-80-15: 15 min etching in 6 M NaOH at 60°C. LR-115: 100 min etching in 6 M NaOH at 60°C.

Table 1. The possible (n, α) interactions in lithium tetraborate.

Reaction	Energy of α-particles (MeV)	Relative probability	Cross-section (barns)
^{6}Li(n, α)^{3}H	2·05	1·00	940 ± 4
^{10}B(n, α)^{7}Li	1·78	0·064	3837 ± 9
^{10}B(n, α)^{7}Li*	1·50	0·936	

4. Results and discussion

4.1 *Track density for different neutron fluences*

Samples of CR-39 irradiated with thermal neutrons to various fluences are etched in 6 M NaOH for 3 hrs and then washed and dried. The track density is measured with an optical microscope for about ten samples and the average value is noted. Similar procedure is adopted for track density measurement in CA 80-15 and LR-115 exposed to neutrons. The average value of track density corresponding to each neutron fluence is noted for different plastics.

4.2 *Track density due to epithermal neutrons*

To find the contribution from epithermal neutrons, the plastic samples covered with 0·75 mm thick cadmium foil are exposed to thermal neutrons for 98 hrs. After irradiation the samples are etched in 6 M NaOH for the required time. The track density for different plastics is measured with optical microscope. The epithermal neutron contribution for other fluences are calculated from this value.

4.3 *Background*

Unirradiated plastic samples are etched in 6 M NaOH at 60°C. The track density for each plastic is determined. These values agree with those obtained by others (Dutrannois and Tuyn 1976; Baroni 1976; Benton 1979).

4.4 *Conversion factors from fluence to dose equivalent*

The conversion factors from fluence to dose equivalent are given in NCRP Report No. 38 for specific neutron energies. A table of the dose equivalent values is given by Hankins (1977), and these values are used in our calculations.

4.5 *Response of the detectors*

The corrected track density is determined by subtracting the background and epithermal neutron contribution from the average track density. Figure 2 shows the

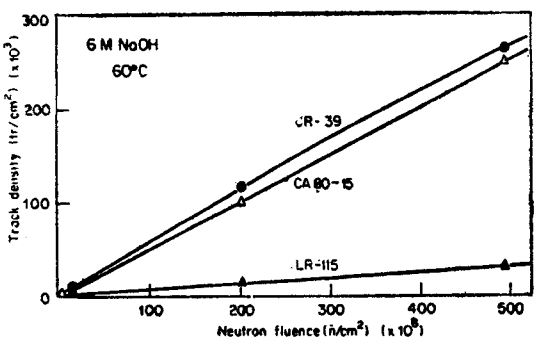

Figure 2. The relation between the track density and the neutron fluence.

relation between track density as a function of neutron fluence for CR-39, CA 80-15 and LR-115 respectively. This linear relationship can be used as calibration curve for neutron fluence ranging from 10^6 to 10^8 n/cm^2 for CR-39 and CA 80-15. However, in LR-115 this linearity starts from a fluence of 10^7 n/cm^2. Due to difficulties associated with the neutron irradiation arrangement it is not possible to irradiate the samples with neutron fluences higher than 4.94×10^8 n/cm^2. These detectors are not used at higher fluence so as to avoid overlapping of etched tracks.

The fluence (tracks/neutron) and dose sensitivities (tracks/cm^2 mrem) are also calculated for each detector to compare the neutron sensitivity of SSNTDs. These sensitivities of different plastics for neutron fluence of 4.94×10^8 n/cm^2 is tabulated in table 2. The relation between neutron fluence and sensitivity is shown in figure 3. The fluence sensitivity varies at low neutron fluences but remains approximately constant after the neutron fluence of about 2.02×10^8 n/cm^2. But one would expect a constant value of sensitivity for all fluences. It will be interesting to observe whether the sensitivity actually varies at low fluences or whether this variation is due to experimental errors.

Table 2. Fluence sensitivity and dose sensitivity of different detectors.

Detector	CR-39	CA 80-15	LR-115
Average track density (tr/cm^2)	$(2{\cdot}8 \pm 0{\cdot}04) \times 10^5$	$(2{\cdot}67 \pm 0{\cdot}03) \times 10^5$	$(3{\cdot}43) \pm (0{\cdot}08) \times 10^4$
Background track density (tr/cm^2)	150 ± 7	100 ± 6	55 ± 4
Track density due to epithermal neutrons (tr/cm^2)	$(1{\cdot}6 \pm 0{\cdot}04) \times 10^4$	$(1{\cdot}46) \pm (0{\cdot}02) \times 10^4$	$(2{\cdot}83) \pm (0{\cdot}08) \times 10^3$
Corrected track density (tr/cm^2)	$(2{\cdot}64 \pm 0{\cdot}05) \times 10^5$	$(2{\cdot}53) \pm (0{\cdot}05) \times 10^5$	$(3{\cdot}15) \pm (0{\cdot}08) \times 10^4$
Fluence sensitivity (tr/neutron)	$(5{\cdot}34 \pm 0{\cdot}1) \times 10^{-4}$	$(5{\cdot}12) \pm (0{\cdot}1) \times 10^{-4}$	$(0{\cdot}64) \pm (0{\cdot}02) \times 10^{-4}$
Dose sensitivity (tr/cm^2 mrem)	$(5{\cdot}11) \pm (0{\cdot}1) \times 10^2$	$(4{\cdot}99) \pm (0{\cdot}1) \times 10^2$	$(0{\cdot}60) \pm (0{\cdot}01) \times 10^2$

neutron fluence = $4{\cdot}94 \times 10^8$ n/cm^2

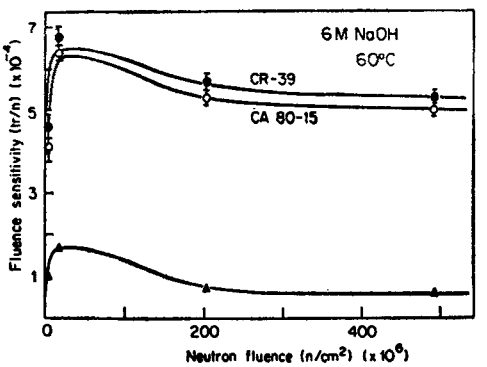

Figure 3. Variation of sensitivity with neutron fluence.

The sensitivities of normal LR-115, CA 80-15 and CR-39 plastic detectors (Lotz *et al* 1976; Garcia *et al* 1979; Al-Najjer *et al* 1979) for fast neutron dosimetry using intrinsic detection technique are $\sim 10^{-6}$ tracks/n, $\sim 10^{-5}$ tracks/n and $\sim 10^{-4}$ tracks/n respectively. Typical sensitivity (Spurny and Turek 1976) for thermal neutron dosimetry using (n, α) reaction is $\sim 10^{-5}$ tracks/n. The present study shows that the fluence sensitivity is $\sim 10^{-4}$ tracks/n. The dose sensitivity is found to be $\sim 10^{2}$ tracks/cm^2 mrem which is reasonably good. The results indicate that the fluence and dose sensitivities of CR-39 are higher than other detectors. This makes CR-39 together with lithium tetraborate as external radiator an excellent candidate for thermal neutron dosimetry having excellent sensitivity.

5. Automatic measurement of tracks

To avoid the slow and tedious process of track density measurement with optical microscope, the jumping spark counter proposed by Cross and Tommasino (1970) is used for LR-115. For CR-39, the electrochemical etching (ECE) technique (Tommasino and Armellin 1973) where the tracks in solid dielectrics are enlarged to macroscopic sizes so that there is no need for an optical microscope.

5.1 *The spark counting*

Samples of strippable LR-115 are placed tightly in contact with lithium tetraborate foils and irradiated with thermal neutrons for 3 hrs. After irradiation the samples are etched in 6 M NaOH at 60°C for 100 min, washed and dried.

The spark counter available in the SSNTD laboratory of Birmingham University (Malik and Durrani 1974) is used for track density measurements. The background tracks are determined by etching unirradiated samples of LR-115 for 100 min in 6M NaOH at 60°C. The spark of each sample is then counted. The average background spark density is 42 ± 3. The corrected spark density (spark counts/cm^2), fluence sensitivity and dose sensitivity are tabulated in table 3.

The average track density obtained by optical microscope for normal (non-strippable) LR-115 and strippable LR-115 are $2{\cdot}57 \times 10^3$ and $1{\cdot}13 \times 10^3$ respectively for the same fluence, $F = 1{\cdot}51 \times 10^7$ n/cm^2. The average spark density of strippable LR-115 is $1{\cdot}07 \times 10^3$ giving a spark counting efficiency* of 0·94. This large difference may be due to following reasons. The thickness of sensitive layer is ~ 15 μm in strippable LR-115. This thickness is greater than that of nonstrippable LR-115

Table 3. Response of LR-115 (strippable).

Sample	Corrected track density (tr/cm^2)	Fluence Sensitivity (tr/n)	Dose sensitivity (tr/cm^2 mrem)
Sample 1	$1{\cdot}10 \times 10^3$	$0{\cdot}72 \times 10^{-4}$	72·8
Sample 2	$1{\cdot}05 \times 10^3$	$0{\cdot}69 \times 10^{-4}$	69·5

Neutron fluence = $1{\cdot}51 \times 10^7$ n/cm^2; Counting voltage = 550V

*Spark counting efficiency $\eta = N_s/N_0$ where N_s is the corrected spark counting density and N_0 is track density obtained by optical microscope.

which is ~13 μm. Thus, a greater number of low energy α-particles fails to produce etch through tracks in strippable LR-115. The strippable LR-115 have a lower registration efficiency than nonstrippable LR-115. Somogyi *et al* (1978) have reported that the track counting under optical microscope on normal LR-115 films, exposed to soil radon gas and etched down to a 5 μm residual thickness, gives a typical track density of about 300 tracks/cm^2 day. In strippable films, this counting is about 150 tracks/cm^2 day. Lotz *et al* (1976) also used normal and strippable LR-115 for fast neutron dosimetry. The microscopic observation in normal LR-115 shows a fluence sensitivity of $\sim 10^{-6}$ tracks/n while the spark counting in strippable LR-115 gives a fluence sensitivity of $\sim 10^{-7}$ tracks/n. These experimental results also show the lower registration efficiency of strippable LR-115 films.

For strippable LR-115 the track density obtained by spark counting is less than that obtained by microscopic observation. This is simply due to the loss of counts when samples are spark-counted. When high voltage is applied in preliminary run there are several sparks per hole and large areas of Al electrode are evaporated. Some areas in the electrode may become insulated electrically from the voltage source before all the insulator tracks in these areas have been cleaned out, thus subsequently giving a lower total when the tracks are counted at lower counting voltage. The final reason is the loss of counts arising from overlap of holes in Al electrodes around track holes at high track density. Somogyi *et al* have also observed a similar decrease in the spark counting efficiency with the increase of track density in normally incident 2·1 MeV α-particle in strippable LR-115 films.

5.2 *The electrochemical etching*

The ECE is obtained whenever the damage track detectors are stressed by a.c. electric field during chemical etching. The experimental set-up of Birmingham University is used. The description and operation technique of the apparatus is reported by Durrani and Al-Najjar (1980).

Samples of CR-39 in contact with $Li_2B_4O_7$ are irradiated with thermal neutrons for 3 hrs. In the present experiment, the duration of pre-etching in 6 M NaOH at 60°C is 6 hrs followed by ECE for 6 hrs. The following values of the electrical parameters are used in the experiment.

Electric field: 20 kV/cm, frequency: 15 kHz, normal etching: 6 hrs, the etching being carried out at 60°C in 6 M NaOH. The ECE is continued up to 16 hrs in steps of 2 hrs. Each time the number of spots are counted on the screen of the Microfiche reader.

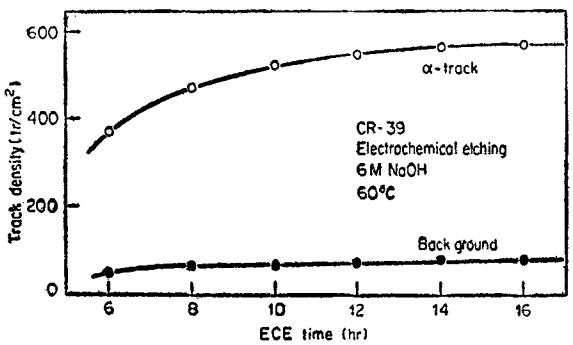

Figure 4. Variation of track density with ECE time for α-particle tracks in CR-39.

Table 4. Response of CR-39.

Thermal neutron fluence (n/cm²)	$1{\cdot}51 \times 10^{7}$
Density of normal etched tracks (tr/cm²), ρ_N	$1{\cdot}14 \times 10^{4}$
Density of ECE tracks (tr/cm²), ρ_E	494·7
ECE sensitivity (spots/n)	$3{\cdot}27 \times 10^{-5}$
Dose sensitivity (spots/cm² mrem)	31·5
ECE efficiency, $\eta = \rho_E/\rho_N$	4·3%

Normal chemical etching = 6 hrs. ECE = 16 hrs.

The variation of track density with ECE time and variation of background are shown in figure 4. A saturation tendency can be clearly recognized. The fluence sensitivity and dose sensitivity are presented in table 4. It is noted that the fluence sensitivity for ECE is $\sim 10^{-5}$ while that for microscopic measurement is $\sim 10^{-4}$. This decrease is due to the fact that when high fluence is used the ECE track density it high and the etched spots become very close to each other and many tracks will nos have a chance to produce treeing and thus to develop etch spots, because they may be lying between nearby tracks where treeing has already started. Tracks in these positions are shielded from the electric-field. Thus the growth of the electric tree number is slowed down. Al-Najjar and Durrani (1980) have shown that only a few % of the normally etched tracks produce spots after ECE. In the present experiment the efficiency is $\sim 4\%$. The ECE track-spot efficiency can be improved by proper choice of pre-etching condition and optimizing the dose. The other factor which affects the fluence sensitivity is the high background spots. This background can be attributed respectively to α-exposure from radon during the detector shelf-life, surface defects and contaminants introduced during manufacturing process (Tommasino 1979). The background can be reduced (i) by using plastics with smooth surface (ii) by applying field strength as low as possible for ECE and (iii) by covering both sides of CR-39 with polyethylene coating.

6. Conclusion

As a final comment the experiments done and results obtained so far give important and encouraging information that will form the basis of future work on this type of neutron dosimeter. More experiments and data are required before it can be realized that a personnel neutron dosimeter can be constructed by extrinsic detection in SSNTDS.

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