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Abstract

CR-39 was obtained from American Acrylics and Plastics, Inc. (A.A.), N. E. Technology, Ltd. (N.E.), and Tech/Ops Landauer, Inc. (LT). The dosimeters were exposed to radioisotopic neutron sources at SLAC, and moderated ²⁵²Cf at ORNL. The A.A. and N.E. dosimeters were electrochemically etched (pre-etch in 6.5 N KOH at 60° C for 1 hour and 45 minutes, a 5 hour etch at 3000 V and 60 Hz, a 23 minute blow-up step at 2 kHz and a post-etch for 15 minutes). Track densities were determined with the Homann Track Size Image Analyzer.

The LT dosimeters were chemically etched in 5.5 N NaOH at 70° C for 15.5 hours. Some A.A., N.E., and LT dosimeters were etched in 6.25 N NaOH at 70° C for 6 hours. A pre-etch step in 60% methanol and 40% NaOH at 70° C for 1 hour was added for some N.E. dosimeters.

The results of these studies are reported in this paper.

Short Title:
NEUTRON RESPONSE OF CR-39

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Introduction

CR-39 is a polymeric solid-state nuclear track detector which is widely used for neutron dosimetry. CR-39 detects neutrons mainly by the damage trails from the nuclei of its constituent atoms, namely, hydrogen, carbon, and oxygen. The damage trails or tracks can subsequently be revealed by a suitable etching process: either chemical etching (CE) or electrochemical etching (ECE). The track density can then be related to the neutron dose equivalent.

The use of a hydrogenous radiator in contact with CR-39 can enhance its response, because of the additional proton recoils generated within the radiator. CR-39 can detect neutrons over a fairly wide energy range from about 100 keV to 20 MeV. The energy range, sensitivity (tracks $\text{cm}^{-2} \text{ mSv}^{-1}$), and lower limit of detection are dependent on the type of CR-39 material, the presence or absence of a radiator, and the particular etching process used.

In this paper, a comparison is made of the neutron response of CR-39 made by different manufacturers (when exposed under identical irradiation conditions) for both chemical and electrochemical etching.

Experimental Methods

CR-39 was obtained from three different manufacturers: American Acrylics and Plastics, Inc., USA (A.A.); N. E. Technology, Ltd., UK (N.E.); and Tech/Ops, Landauer, Inc., USA (LT).

The A.A. CR-39 (dosimetry grade) was about 0.063 cm thick and covered on both sides with polyethylene 0.01 cm thick.

The N.E. CR-39 materials PN-4 and PN-3 were prototype materials. Two batches of PN-4 (N.E. B1, N.E. B2) were obtained. The PN-4 was about 0.06 cm thick and recommended by the manufacturer for ECE. The PN-3 (N.E.) was about

0.13 cm thick and recommended for CE. Both the PN-3 and PN-4 were covered on both sides with Clingsol.

The LT material "Lantrak," recommended for CE, was about 0.08 cm thick and also covered on both sides with a protective layer of polyethylene.

The A.A. and N.E. CR-39 were laser-cut into pieces of dimensions 1.587×2.85 cm. The protective layers were removed from the detectors prior to exposure. All the detectors except Lantrak were held in a plastic badge whose front surface was about 0.05 cm thick. The Lantrak dosimeters had a polyethylene radiator of thickness 0.1 cm in front, and were held in the Landauer badge.

The neutron irradiations were performed outdoors in a low scatter environment at the Stanford Linear Accelerator Center (SLAC). Corrections for anisotropy of the neutron sources were made in determining the fluences. The fluences were converted to dose equivalents using the methods outlined in NCRP 79⁽¹⁾. Corrections for neutron scattering were also made.

All dosimeters were exposed on a water phantom under identical irradiation conditions to radiosotopic neutron sources ($^{238}\text{PuBe}$ $E_{av} = 4.2$ MeV, ^{238}PuB $E_{av} = 2.1$ MeV, $^{238}\text{PuF}_4$ $E_{av} = 1.5$ MeV, $^{238}\text{PuLi}$ $E_{av} = 0.5$ MeV, and ^{252}Cf $E_{av} = 2.2$ MeV). The source strengths are all traceable indirectly back to standards at the National Institute of Standards and Technology (NIST). The uncertainties in source strengths are as follows: PuBe 10%, PuB 15%, PuF₄ 20%, PuLi 15%, and ^{252}Cf 3%.

The dosimeters were also exposed on a Lucite slab phantom at Oak Ridge National Laboratory (ORNL) as part of the Fifteenth Personnel Dosimetry Intercomparison Study (PDIS 15) to $^{238}\text{PuBe}$ and ^{252}Cf (with a variety of moderators). The D₂O moderators were 15 cm in radius.

The A.A., N.E. B1 and N.E. B2 dosimeters were electrochemically etched using the following procedure:

A pre-etch in 6.5 N KOH at 60° C for 1 hour and 45 minutes, followed by a 5 hour etch at 3000 V and 60 Hz, then a 23 minute blow-up step at 2 kHz and, finally, a post-etch for 15 minutes. The dosimeters were etched in the standard Homann type etch chamber which can handle up to 24 foils simultaneously⁽²⁾.

This etch differs from Hankins' latest etch process in that it lasts for 5 hours instead of 3 hours, and the blow-up step lasts for only 23 minutes instead of 30 minutes⁽²⁾. The pre-etch reduces the background on the foils. The blow-up step further amplifies the track size after the etch, thus improving the precision that can be attained with the image analyzer which is used for track counting.

The Homann Track Size Image Analyzing System was used to count the tracks⁽²⁾. The system consists of a computer and a camera connected to a standard microscope. A computer-controlled automatic stager moves the foil. The image analyzer displays the track size distribution and the number of tracks. Three fields, each of an area of about 0.6 cm², were counted for each detector.

The electrochemically etched A.A. dosimeters exposed at PDIS 15 were counted using a Zeiss optical microscope at a magnification of 210. A total of 10 fields of total area 0.11 cm² were counted.

The N.E. dosimeters were etched using the procedure recommended by the manufacturer: Pre-etch in 60% methanol and 40% 6.25 N NaOH at 70° C for 1 hour, followed by an etch in 6.25 N NaOH at 70° C for 6 hours. These dosimeters are designated as N.E. (pre-etch) in the text. About 25 μm were removed from one side during the pre-etch and 12 μm during the etch. The pre-etch was recommended for reduction in background caused by defects and radon.

Some N.E., A.A., and LT dosimeters were chemically etched, using the process mentioned above, but with the elimination of the pre-etch step. These dosimeters are designated as N.E., A.A. (CE), and LT (6 h) in the text. The LT detectors were chemically etched in 5.5 N NaOH at 70° C for 15.5 hours as recommended by the manufacturer. These dosimeters are designated as LT (15.5 h).

Track densities were determined with a microscope at a magnification of 300. Nine fields of total area of 3.68 mm² were counted for each dosimeter.

Results and Discussion

Comparison of Response of Different Materials

Table 1 shows a comparison of the response of different materials for both electrochemical and chemical etching. The average sensitivity in tracks cm⁻² mSv⁻¹ (to ²⁵²Cf neutrons) and its standard error are shown in Column 3. The background track density and its standard error are shown in the last column. Three dosimeters were used in each case except as otherwise indicated in Columns 3 and 4 (in brackets). The standard error includes the variation between the dosimeters and the uncertainty in the source strength.

LT (15.5 h) has the highest sensitivity and the lowest background. The sensitivity of LT (15.5 h) is about 2.5 times greater than that of A.A. (ECE). The background of LT (15.5 h) is about 2.5 times lower than that of A.A. (ECE).

The sensitivity of A.A. (ECE) is about 1.5 to 2 times greater than that of N.E. B2 and N.E. B1. The background of A.A. (ECE) is about the same as that of N.E. B1. The sensitivity and background of N.E. B2 is slightly higher than that of N.E. B1. The reduced sensitivity of N.E. B1 and N.E. B2 could be attributed to the presence of additives such as DOS (dioctyl sebacate)⁽³⁾. The American Acrylics CR-39 does not contain any additives.

Track size distribution studies with the image analyzer indicated that track sizes for A.A. (ECE) are larger than for N.E. (ECE) for both background and the ^{252}Cf exposure.

Independent measurements by Hankins indicate a sensitivity of 467 tracks $\text{cm}^{-2} \text{ mSv}^{-1}$ for ^{252}Cf for A.A. (ECE)⁽²⁾. This is in good agreement with our results.

For the chemically etched CR-39, A.A. (CE) is about 1.5 times more sensitive than N.E. (pre-etch). N.E. (pre-etch) has a higher sensitivity and lower background than N.E. The pre-etch was recommended by the manufacturers for reduction in background. Data provided by the manufacturer for N.E. (pre-etch) indicates a sensitivity of 240 tracks $\text{cm}^{-2} \text{ mSv}^{-1}$ for ^{252}Cf , which is in good agreement with the measured sensitivity.

LT (6 h) has a slightly lower sensitivity than N.E.; however, its background is much lower.

Energy Response of CR-39 (ECE)

Figure 1 shows the sensitivity of electrochemically etched CR-39 as a function of the average neutron energy. For all the dosimeters, the response generally increases as the average neutron energy decreases except for a dip in response to the PuB neutrons. According to studies by Hankins, the response of A.A. to PuBe neutrons is about 30% lower than its response to ^{252}Cf neutrons⁽²⁾. These results are in excellent agreement with Hankins' studies.

A.A. has a higher response than N.E. for all the neutron sources. N.E. B2 has a slightly higher response than N.E. B1 for all the sources.

Energy Response of CR-39 (CE)

Figure 2 shows the sensitivity of chemically etched CR-39 as a function of average neutron energy. LT (15.5 h) shows the highest sensitivity for all the neutron sources, with the response generally increasing as average energy decreases, except for a dip in response to the PuB exposure. The response to PuLi is about 3 times higher than the response to PuBe.

When compared to A.A. (CE), LT (15.5 h) is about 3 times more sensitive to PuBe and about 8 times more sensitive to PuLi. LT (15.5 h) is about 2 to 4 times more sensitive than A.A. (ECE), depending upon the neutron source.

The sensitivity of LT (15.5 h) to AmBe as provided by the manufacturer is also shown in Figure 2. This appears to be in reasonable agreement with the sensitivity for PuBe.

LT (6 h) has a higher response than A.A. (CE) for PuLi, whereas for the other sources, the responses are not significantly different. Overall, the LT (6 h) response increases slightly with decreasing average energy. Clearly, A.A. (CE) does not show this trend, but appears to be fairly independent of average energy.

The chemically etched N.E., with pre-etch and without pre-etch, also appears to have responses fairly independent of average energy. The sensitivity of N.E. is about the same as that of A.A. (CE). N.E. (pre-etch) is more sensitive than N.E.

PDIS 15 Results

The results of PDIS 15, held at ORNL, are shown in Table 2.

The first and second columns show the source and the dose equivalent delivered. The last three columns show the sensitivity and its standard error for A.A. (ECE), LT (15.5 h), and N.E. (pre-etch). The standard error includes only the variations in track density between the dosimeters. Three A.A. dosimeters were used for each

exposure, except in the case of Exposure 1 (only two dosimeters were used). Six dosimeters were used in each case for LT (15.5 h) and three for N.E. (CE) except for Exposure 6 (only two dosimeters were used).

Results for electrochemically etched N.E. are not reported because of the very high background track densities found on the controls.

One would expect the sensitivities to be the same for Exposures 1, 3 and 4. Statistical *t* tests indicate that these sensitivities are the same for each manufacturer type.

In Exposure 2, the phantom was rotated through 45° about the vertical centerline. Hankins' studies indicate that a 45° rotation causes the response of A.A. (ECE) to reduce to 60% of that for normal incidence⁽²⁾. The A.A. (ECE) and LT (15.5 h) responses are about 87% and 60% of that for normal incidence, respectively. Because of the large variation between the dosimeters no conclusive statement can be made about the N.E. response.

About 56% of the contribution to dose equivalent for Exposure 6 is from moderated ²⁵²Cf and 46% from bare ²⁵²Cf. As is evident from Tables 1 and 2, the sensitivity to moderated and bare ²⁵²Cf is about the same for A.A. (ECE). For LT (15.5 h), the sensitivity to moderated ²⁵²Cf is about 20% higher than bare ²⁵²Cf. The response of N.E. to moderated ²⁵²Cf is only 50% of that for bare ²⁵²Cf. Thus, the response of all manufacturer types to Exposure 6 is reasonable.

The sensitivity of A.A. (ECE) to PuBe is about 90% higher than that obtained with the exposures at SLAC. The sensitivity of LT (15.5 h) is about 50% higher than that of the SLAC exposures. We are unable to explain this discrepancy.

Conclusions

The neutron response of CR-39 made by different manufacturers, when exposed under identical conditions, has been compared for both chemical and electrochemical etching.

The Lantrak 15.5 h chemically etched CR-39 has the highest sensitivity for all neutron sources and the lowest background. The response is energy (E_{av}) dependent and generally increases as the average energy decreases. The response to PuLi neutrons is about 3 times higher than the response to PuBe neutrons.

Electrochemically etched American Acrylics has a much lower sensitivity than Lantrak [LT (15.5 h)]. It has a higher background and its energy response is similar to Lantrak, but not as pronounced.

Electrochemically etched N.E. has a lower sensitivity than that of A.A. (ECE). The background and energy response are similar to A.A. (ECE).

Chemically etched N.E. has about the same sensitivity as electrochemically etched N.E., but its background is higher. The response, however, is fairly independent of average energy.

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Table Captions

1. Comparison of Different Materials.
2. PDIS 15 Results.

Figure Captions

1. Sensitivity of CR-39 (ECE) vs Average Neutron Energy.
2. Sensitivity of CR-39 (CE) vs Average Neutron Energy.

Fig. 1

Fig. 2

Table 1

Etch Process	Material	Sensitivity to ²⁵² Cf (tracks cm ⁻² mSv ⁻¹)	Background (tracks cm ⁻²)
ECE	A.A.	471 ± 26	69 ± 22 (2)
	N.E. B1	231 ± 9	65 ± 6 (6)
	N.E. B2	284 ± 11	82 ± 12 (6)
CE	A.A.	333 ± 57	217 ± 270 (2)
	N.E.	204 ± 49	1060 ± 250 (2)
	N.E.(pre-etch)	231 ± 42	571 ± 95
	LT (15.5 h)	1148 ± 48 (2)	27 ± 27 (2)
	LT (6 h)	181 ± 34	63 ± 4

Table 2

Source	Dose Equivalent (mSv)	Sensitivity (tracks cm ⁻² mSv ⁻¹)		
		A.A. (ECE)	LT (15.5 h)	N.E. (pre-etch)
1) ²⁵² Cf (D ₂ O)	1.40	467± 6	1305± 67	136±38
2) ²⁵² Cf (D ₂ O) non-perpendicular	2.05	368±33	829± 60	146±78
3) ²⁵² Cf (D ₂ O) without cadmium	1.42	589±74	1401± 49	115±42
4) ²⁵² Cf (D ₂ O) + ¹³⁷ Cs	0.45	547±28	1444±127	—
5) ²³⁸ PuBe	0.38	631±62	1313±150	—
6) ²⁵² Cf (D ₂ O) + ²⁵² Cf	3.49	567±35	1444± 60	179±21
Controls		Tracks cm ⁻²		
		84± 9	0± 0	598±17

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