A COMPARISON OF THE NEUTRON RESPONSE OF CR-39 MADE BY DIFFERENT MANUFACTURERS^{*}

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Short Title:

NEUTRON RESPONSE OF CR-39

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Abstract

CR-39 obtained from American Acrylics and Plastics, Inc. (A.A.), N. E. Technology, Ltd. (N.E.), and Tech/Ops Landauer, Inc. (LT) were exposed to radioisotopic neutron sources at SLAC, and moderated 252 Cf at ORNL. The A.A. and N.E. detectors (0.06 cm thick) were electrochemically etched (a pre-etch for 1 hour and 45 minutes in 6.5 N KOH at 60° C, 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).

The LT detectors were chemically etched in 5.5 N NaOH at 70° C for 15.5 hours and some A.A., N.E., and LT detectors 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. detectors.

The results of the background track density and neutron dose equivalent response are reported in this paper.

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), or both combined. On the basis of calibration exposures, 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. When combined with radiators, CR-39 can detect neutrons over a fairly wide energy range from about 100 keV to 20 MeV. The energy range, dose equivalent response $(cm^{-2} mSv^{-1})$, and the lower limit of detection are dependent on the type of CR-39 material, the type and thickness of the radiator, and the particular etching process used.

This paper compares the neutron dose equivalent response of CR-39 plastics made by different manufacturers for both chemical and electrochemical etching.

Experimental Methods

Plastics

CR-39 was obtained from three different manufacturers: (a) American Acrylics and Plastics, Inc., USA (A.A.); (b) N. E. Technology, Ltd., UK (N.E.); and (c) 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 of thickness 0.01 cm.

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The N.E. CR-39 materials PN-3 and PN-4 were prototype materials. Two batches of PN-4 (N.E. B1, N.E. B2) were obtained. The PN-3 material was about 0.13 cm thick and recommended by the manufacturer for CE. The PN-4 material was about 0.06 cm thick and recommended for ECE. Both types were covered on both sides with Clingsol of thickness 0.002 cm.

The LT CR-39 material "Lantrak," recommended by the manufacturer for CE, was 0.08 cm thick and covered on both sides with a protective layer of polyethylene of thickness 0.006 cm.

The A.A. and N.E. CR-39 were laser-cut into pieces of dimensions 1.59×2.85 cm². After removal of the protective layers, the detectors were placed inside a plastic badge whose front side, which serves as radiator, was about 0.05 cm thick. The LT CR-39 detectors had a polyethylene radiator of thickness 0.1 cm in front, in the "Landauer badge." It is important to note that the radiator thicknesses were not the same for the different plastics.

Neutron Irradiations

At the Stanford Linear Accelerator Center (SLAC), the neutron irradiations were performed outdoors in a low scatter environment. Corrections were made for anisotropy of the neutron sources and for neutron scattering. The fluences were converted to dose equivalents using the methods outlined in NCRP $79^{(1)}$.

All detectors were exposed on a cubic water phantom under identical irradiation conditions to radioisotopic neutron sources⁽²⁾ (²³⁸PuBe $E_{av} = 4.2$ MeV, ²³⁸PuB $E_{av} = 2.1$ MeV, ²³⁸PuF₄ $E_{av} = 1.5$ MeV, ²³⁸PuLi $E_{av} = 0.5$ MeV, and ²⁵²Cf $E_{av} = 2.2$ MeV, where E_{av} is the fluence-averaged energy). 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%.

As part of the Fifteenth Personnel Dosimetry Intercomparison Study (PDIS 15), the detectors were exposed on a Lucite slab phantom at the Oak Ridge National Laboratory (ORNL) to 238 PuBe and 252 Cf (D₂O). The D₂O moderator was 15 cm in radius.

ECE Procedure

The A.A. and N.E. detectors 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 detectors were etched in the standard Homann type etch chamber which can handle up to 24 detectors simultaneously⁽³⁾.

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⁽³⁾. The pre-etch reduces the background on the detectors. 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.

CE Procedure

The N.E. detectors were chemically etched using the procedure recommended by the manufacturer: a 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 detectors are designated as N.E. (pre-etch) in the text. About 25 μ m were removed from one side during the pre-etch and 12 μ m during the etch. The pre-etch was recommended for reduction in background caused by defects and radon.

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

Track Counting

The Homann Track Size Image Analyzing System was used to count the tracks⁽³⁾ for the electrochemically etched CR-39 detectors. The system consists of a computer and a camera connected to a standard microscope. A computer-controlled automatic stager moves the detector. 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. detectors 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 cm^2 were counted.

Track densities for the chemically etched CR-39 detector were determined with a microscope at a magnification of 300. Nine fields of total area of 3.68 mm^2 were counted for each detector.

Results and Discussion

Comparison of Response of Different Materials

Table 1 compares the background and the ²⁵²Cf neutron response of different materials for both electrochemical and chemical etching. The results are mean values and standard deviations (of the means) of three detectors except as otherwise indicated in brackets. The standard deviation includes the variation between the detectors and the uncertainty in the source strength. Chemically etched LT (15.5 h) has the highest response and the lowest background. This response is about 2.5 times greater and the background is about 2.5 times lower than that of A.A. (ECE).

The response 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 response and background of N.E. B2 is slightly higher than that of N.E. B1. 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 neutron exposure. The lower response of N.E. B1 and N.E. B2 could be attributed to the presence of additives such as DOS (dioctyl sebacate)⁽⁴⁾. The A.A. CR-39 does not contain any additives.

The other etching condition of Hankins for A.A. $(ECE)^{(3)}$ indicates a similar ²⁵²Cf neutron response of 467 cm⁻² mSv⁻¹.

Within the uncertainty of measurement, chemically etched A.A. (6 h) CR-39 is about 1.5 times more sensitive than N.E. (pre-etch) and N.E. (6 h). N.E. (pre-etch) has a slightly higher response and a much lower background than N.E. (6 h). Data provided by the manufacturer for N.E. (pre-etch) indicates a response of 240 cm⁻² mSv⁻¹ for 252 Cf neutrons, which is in good agreement with our measurement.

LT (6 h) has a similar but lower response than N.E. (6 h); however, its background is more than one order of magnitude lower.

Energy Response of CR-39 (ECE)

Figure 1 shows the response of electrochemically etched CR-39 as a function of the average neutron energy which, for all the detectors, generally decreases with increasing neutron energy except for a lower response to the PuB neutrons. The response of A.A. to PuBe neutrons is about 15% lower than its response to 252 Cf neutrons, whereas a 30% reduction is reported by Hankins⁽³⁾.

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

Energy Response of CR-39 (CE)

Figure 2 shows the response of chemically etched CR-39 as a function of average neutron energy. For the information of the reader, it must be pointed out that Figure 2 has a broken scale. The scale in Figure 2 is also different from that in Figure 1. LT (15.5 h) shows the highest response for all the neutron sources. The response, in general, increases as the average neutron energy decreases.

LT (15.5 h) is about 3 to 8 times more sensitive than A.A. (6 h) and about 2 to 4 times more sensitive than A.A. (ECE), depending upon the neutron source.

The AmBe response of LT (15.5 h) as provided by the manufacturer appears to be in reasonable agreement with the response for PuBe results.

As for LT (6 h), the response increases with decreasing average neutron energy by more than a factor of 2. Clearly, A.A. (6 h) does not show this trend, and appears to be fairly independent of the average neutron energy.

The chemically etched N.E., with and without pre-etch, also appears to have responses fairly independent of average neutron energy within a factor of about 2. The response of N.E. (6 h) is about the same as that of A.A. (6 h). N.E. (pre-etch) is more sensitive than N.E. (6 h).

PDIS 15 Results

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

The first and second columns show the neutron source and the dose equivalent delivered and the last three columns show the mean response and its standard deviation for the CR-39 plastics: A.A. (ECE), LT (15.5 h), and N.E. (pre-etch). The standard deviation includes only the variations in track density between the detectors since the uncertainty in source strength was not reported by ORNL. Three A.A. detectors were used for each exposure, except two for Exposure 1. Six detectors were used in each case for LT (15.5 h) and three for N.E. (6 h) except two for Exposure 6. Results for electrochemically etched N.E. are not reported because of the very high background track densities found on the controls. Since track counting was done over an area of only 0.11 cm^2 , there is a high random uncertainty associated with the A.A. (ECE) results.

As expected, within the uncertainty of measurement, the responses are the same for each plastic for Exposures 1, 3 and 4.

In Exposure 2, the phantom was rotated through 45° about the vertical centerline. The LT (15.5 h) and A.A. (ECE) responses are about 60% and 70% of that for normal incidence, respectively, which is in good agreement with the 60% reported by Hankins for A.A. (ECE). Because of the large random uncertainty of track counting, the A.A. and N.E. results cannot be used for intercomparison.

For Exposure 6, about 56% of the contribution to dose equivalent is from moderated 252 Cf and 46% from bare 252 Cf neutrons. As is evident from Tables 1 and 2, the response to moderated and bare 252 Cf is about the same for A.A. (ECE). For LT (15.5 h), the response to moderated 252 Cf is about 20% higher than bare 252 Cf; for N.E., however, the response is only 60% of that for bare 252 Cf. Thus, based on the sensitivities of the individual detectors to bare and moderated 252 Cf neutrons, these results are reasonable [within 11% for A.A. (ECE), 4% for N.E. (pre-etch) and 11% for LT (15.5 h)].

We are unable to explain the discrepancy that the responses of both A.A. (ECE) and LT (15.5 h) to PuBe are about 60% higher than that obtained with the exposures at SLAC.

Conclusions

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

The Lantrak 15.5 h chemically etched CR-39 has the highest dose equivalent response 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 response than Lantrak [LT (15.5 h)]. It has a higher background and its energy response is similar to Lantrak, but not as pronounced. The less pronounced energy response is desirable for neutron dosimetry.

Electrochemically etched N.E. has a lower response 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 response as electrochemically etched N.E., but its background is higher. The response, however, is fairly independent of average energy.

Acknowledgements

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Table Captions

 Dose equivalent response, background and standard deviations of different CR-39 plastics.

Footnote: Three detectors were used for each plastic except where otherwise indicated in brackets.

2. Dose equivalent response and standard deviations for PDIS 15 intercomparison study.

Figure Captions

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- Response of CR-39 (ECE) as a function of average neutron energy for different radioisotopic neutron sources: (a) PuLi; (b) PuF₄; (c) PuB; (d) ²⁵²Cf; and (e) PuBe.
- Response of CR-39 (CE) as a function of average neutron energy for different radioisotopic neutron sources: (a) PuLi; (b) PuF₄; (c) PuB; (d) ²⁵²Cf; and (e) PuBe.

Etch	Material	Response to ²⁵² Cf	Background
Process		(cm ⁻² mSv ⁻¹)	(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. (6 h) N.E. (6 h) N.E.(pre-etch) LT (6 h) LT (15.5 h)	$333 \pm 57 204 \pm 49 231 \pm 42 181 \pm 34 1148 \pm 48 (2)$	$217 \pm 70 (2)$ $1060 \pm 250 (2)$ 571 ± 95 63 ± 4 $27 \pm 27 (2)$

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Table 1

	Dose	Response (cm ⁻² mSv ⁻¹)		
Source	Equivalent (mSv)	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) ${}^{252}_{45^\circ}$ (D ₂ O) 45^\circ irradiation	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

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Table 2



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Fig. 1



Fig. 2