Iran. J. Radiat. Res., 2011; 9(2): 95-102

Experimental and Monte Carlo studies on the response of CR-39 detectors to Am-Be neutron spectrum

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Background: The Poly-Allyl Diglycol Carbonate (PADC) detector is of particular interest for development of a fast neutron dosimeter. Fast neutrons interact with the constituents of the CR-39 detector and produce H, C and O recoils, as well as (n, α) reaction. These neutron- induced charged particles contribute towards the response of CR-39 detectors. *Material and Methods:* Electrochemical etching was used to enlarge track diameter which was made by low energy recoil protons. Before electrochemical etching, a chemical etching was performed for 1 hour. The responses were also calculated by Monte Carlo simulations, using MCNPX code in different energy bins considering H, C and O recoils. The total registered efficiency and partial contributions of the efficiency, due to interactions with each constituent of CR-39, were calculated. *Results:* The optimized condition of etchant was obtained to be 6N KOH 15kV.cm^{-1}, and 6 hours etching time. The obtained results show that track efficiency of CR-39 was a function of incident neutron energy. The tracks caused by O and C recoil nuclei were negligible for neutron energies lower than 1 MeV. At neutron energies lower than 1 MeV, only recoil protons would have sufficient energy to leave visible tracks. But, O and C recoils had important contributions in overall response of PADC at neutron energies of few MeV. *Conclusion:* The efficiency of a CR-39 based dosemeter could be calculated by MCNPX code and the results were in a good agreement with experimental results in energy range of ^{241}Am – Be bare source and ^{241}Am-Be was softened with a spherical polyethylene moderator of radius of 20 cm. **Iran. J. Radiat. Res., 2011; 9(2): 95-102**

Keywords: *Neutron dosemetry, electrochemical etching, MCNPX, simulation.*

INTRODUCTION

The Poly-Allyl Diglycol Carbonate (PADC) detector is of particular interest for the development of a fast neutron dosimeter [(1)]. Neutron elastic interactions with constituent atoms of PADC plastic leave latent recoil charged particle tracks which could be brought out by chemical or electro-chemical etching. Above a threshold of ~7.9 MeV, inelastic alpha particle breakup of ^{12}C was also possible with the energy lost by the neutron partitioned among the three alpha particles [(2)]. Fast neutrons interact with the constituents of the CR-39 detector and produce H, C and O recoils, as well as (n, α) reaction. These neutron- induced charged particles contribute towards the response of CR-39 detectors [(3)]. The majority of tracks produced by neutrons in CR-39 were from recoil protons, generated partly in the surface layer of the CR-39 which was removed by chemical or electrochemical etching. The size and characteristics of the tracks depended upon the charged particle mass, energy and direction of motion. Energy dependence has been reported previously on the distribution of track sizes for monoenergetic neutrons [(4)] and for broad spectrum neutrons [(5)]. Phillips *et al.* have investigated the dependence of the track density with energy, fluence and with direction [(6)].

Calculation of the number of recoils and charged particles that enter the etched layer were generated in it depends on their

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distributions in energy and angle [7]. The track density and the geometrical parameters of tracks depended on the track formation process which was strongly determined by the kind of particles and the etching conditions applied to develop the latent tracks. Therefore, the calculation of the neutron response of CR-39 also had to include a realistic simulation of the etched track formation mechanisms. In order to mimic the actual situation, all significant fast neutron interactions with other constituents of the detector had been taken into account. So, it would have been feasible to determine the contribution of H, C and O recoils and (n, α), (n, p) and (n, d) reactions in O as well as C, which would be called nuclear reactions here after. A theoretical and practical works concerning the response of PADC detector to fast neutrons had been done. This works had considered the contribution of the H, C and O recoils [8]. El-Sersy *et al.* have shown a good agreement between the calculated and the measured values of efficiency for cylindrical source configuration at small source-to-detector distances [9]. The elastic scattering of light nuclei, especially hydrogen, is widely used for detection of fast neutrons. For increasing of efficiency of the CR-39 detector to Am-Be neutron source, the effect of different radiators layer on the response had been studied with MCNP [10]. MCNP method had been developed for calculating the energy response of a solid state nuclear track detector (SSNTD) with polyethylene radiator to neutrons in the energy range from 100 keV to 20 MeV [11]. Phillips *et al.* have investigated the dependence of the track density with energy, fluence and with direction [6]. The CR-39 response to Am-Be neutron source has been studied by H. Zaki-Dizaji *et al.* They have only considered elastic scattering of hydrogen [12].

In this study, the responses of CR-39 to the energy of ^{241}Am – Be bare source and ^{241}Am-Be softened with a spherical polyethylene moderator of radius of 20 cm were evaluated by experiment, and the simulation with MCNPX. The optimized etching condition and computer code simulation based on Monte Carlo technique for ^{241}Am – Be neutron spectrum has been presented.

MATERIALS AND METHODS

Detector and source specification

The detector was a segment of a sheet grade (93% monomer purity) CR-39 manufactured by Track Analysis System Limited. Detectors were of 1 mm thickness with 1.8×1.8 cm^2 dimension. These cards were irradiated for ^{241}Am – Be bare source with 30 mm×60 mm dimension and 370 GBq activity and ^{241}Am-Be softened with a spherical polyethylene moderator of radius of 20 cm. Irradiations were performed with the dosemeters mounted on water-filled phantoms of 30×30×15 cm^3 at normal neutron incidence.

Process and assessment

Before electrochemical etching, the CR-39 was chemically etched for 1 hour in 6 N KOH at 60°C to reduce the background. This was followed by a 6 hour electrochemical etching in 6 N of KOH solutions at the same temperature. In optimizing the etching conditions, two parameters were mainly investigated: first, the background track density, which should have been low, and, second, the neutron dose equivalent response which should have been constant for different electric field strengths [13]. Unirradiated detectors and detectors irradiated with ^{241}Am – Be bare source and ^{241}Am-Be softened with a spherical polyethylene moderator of radius of 20 cm were electrochemically etched in different electric field strength, time and concentration of etchant.

The etched CR-39 detectors were scanned using a high resolution scanner (2000 dpi) with transparency adaptor (UMAX scanner power look III), which was connected to a PC-based image analyzer and counting program. The program was developed in MATLAB 6.5.1 environment [14]. After implementing the image processing, the tracks were automatically counted inside the region of interest (ROI) and the track density was calculated. The ROI included all the tracks inside the scanning area (3 cm^2) whose diameters were greater than 20 µm. The background track density of 18 samples was measured and the average value was obtained equal to 40±5 cm^{-2}.

Assumptions for CR-39 simulation

It was assumed that CR-39 detector was a flat sheet and a neutron source with energy range from 200 keV to 6 MeV was placed at a distance of 100 cm from its surface. The whole set up was placed on a water phantom with 30cm×30cm×15cm dimension as shown in figure 1. In order to assess the interaction probabilities of the neutrons inside the detector, it was assumed that all the heavy charged particles to have elastic interactions produced by H, C and O recoils. The resulting reaction or some products would have carried some energy and would have created latent tracks in the detector along the direction of their motion. In this work the critical angle and the energy for H, O and C recoils, which might have produced visible tracks, were assumed as follows:

a) the incidence angle q_c less than 70°,

b) the proton energy greater than 0.01 MeV and less than 5 MeV [13],

c) the O and C recoils with energy greater than 0.2 MeV [15].

The response of CR-39 for different neutron energy bins was calculated with Monte Carlo simulations using MCNPX 2.4.0 code [16]. MCNPX was Fortran90 Monte Carlo radiation transport computer codes that transport nearly all particles at nearly all energies. The code, allows the user to perform particle transport from GeV energies down to minimum cut-off energy, without having to run numerous codes to obtain the results. The response function was calculated with MCNPX code [15]. MCNPX simulation for ^{241}Am – Be bare source and ^{241}Am-Be softened with a spherical polyethylene moderator of radius of 20 cm was performed with the above considerations in 0.1 mm thickness of the PADC which was mounted on a water phantom. In the simulation also the effect of back scatter of room dimension, as well as room building material was concluded. The simulated source was a broad beam of mono -energetic neutrons normal to the detectors surface and energy ranging from 200 KeV to 6 MeV. The ENDF60 cross- section library was used for ^{241}Am – Be energy ranges. For this purpose, the Particle Track Output Card (PTRAC) was used for calculating H, C (Natural Carbon) and O recoils for the whole history [15]. The PTRAC generated an output file of user-filtered particle events. This option of MCNPX code registered the whole history, such as interaction type, energy, direction and position of particles. For all calculations, the code was run for 10^7 histories for different neutron energies.

RESULTS

Experimental measurement for optimization of etching condition

In order to optimize etching condition,

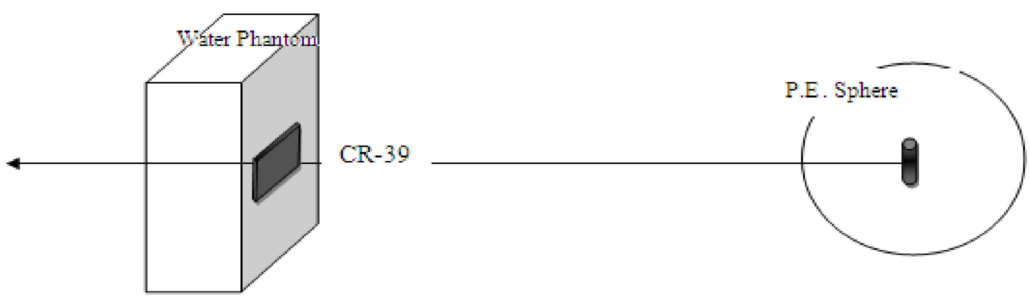

Figure 1. Irradiation geometry of Am-Be neutron source.

the field strength, concentration of etchant and etching time were studied. Both un-irradiated detectors, and the detectors irradiated with Am-Be neutrons, were etched chemically for 1 hour in 6 N of the KOH at 60°C as the pre-etching process. Subsequently, all the detectors were electro-chemically etched in different electric field strength which varied from 10 kV.cm^{-1} to 30 kV.cm^{-1}, etching time and from 3 hours to 8 hours with concentration of etchant at 3 N KOH to 15 N KOH.

The optimized condition found at 6 N of the KOH at 15 kV.cm^{-1} for 6 hours etching time. The result of background assessment is shown in figure 2 and figure 3. These figures show the effect of different etching condition such as field strength, time of etching and concentration of KOH on the CR-39 plastic, which show the relation between background tracks in different etching condition. The results depicted in figure 2 and figure 3 show that, the background increases with the field strength and concentration of KOH. The background had varied from 30 tracks.cm^{-2} at10 kV.cm $^{-1}$ to about 160 tracks.cm^{-2} at 30 kV.cm^{-1}. The obtained results on optimization condition were comparable with the results by different laboratories [14].The average background track density was measured under the optimized condition inside the region of interest (ROI) with 20 unexposed detectors at about 40±5 cm^{-2}. The electric field strength was mainly responsible for the lowest detectable proton energy which was significantly affected by the response curve in the energy range of 1 MeV. The field strength and the etching conditions such as the temperature, etching time and etching solution might affect the maximum value of the neutron energy response. The average standard deviation of the response for different etching condition is presented in table 1. From the table 1, it is implied that the minimum mean standard deviation has been obtained for 15 kV.cm^{-1} field strength at 6 N of KOH concentration for 6 hours etching time. It means that in such condition the minimum error could be achieved and the, minimum distortion on etched CR-39 plastic and the size of the tracks had been big enough for counting system.

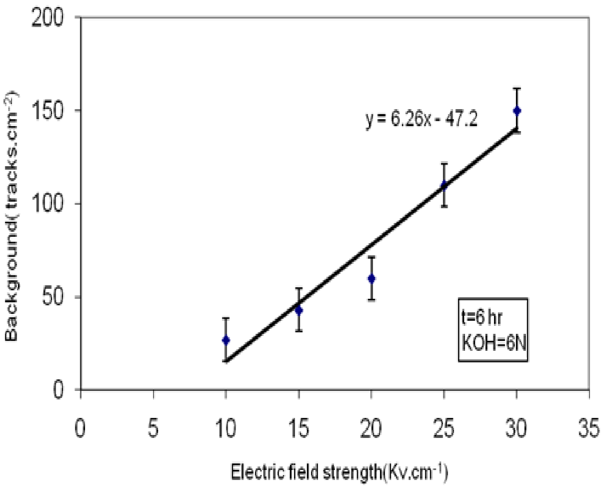

Figure 2. Background track density plotted against field strength.

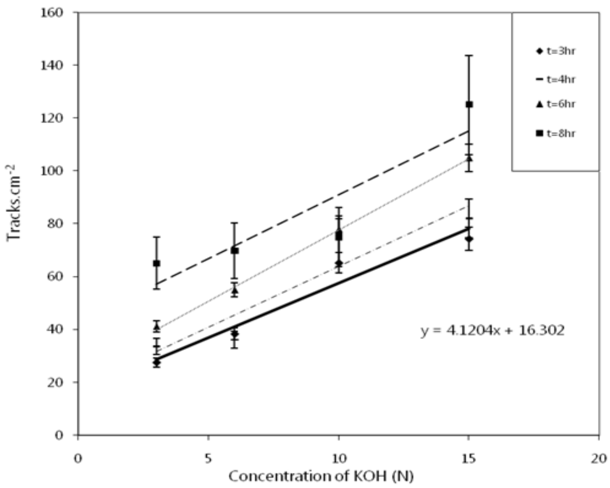

Figure 3. Background track density plotted against different concentration of KOH.

Table 1. Calculated standard deviation of etched tracks in different concentration of etching solution and different etching time. These values display the uncertainties corresponding to one standard deviation.

Concentration of KOH (N)	Standard deviation			
	3 hr	4 hr	6 hr	8 hr
3	13%	14%	16%	25%
6	15%	7%	5%	32%
10	17%	24%	39%	42%
15	32%	48%	62%	74%

The relevant neutron dose equivalent response was obtained, and it showed linear response for ^{241}Am – Be bare source and ^{241}Am-Be softened with a spherical polyethylene moderator of radius of 20 cm at a field strength of 15 kv.cm^{-1} at 6 hours etching time and concentration of 6 N of KOH. The response of the detector based on different concentration of KOH in the different etching time for 1 mSv is shown in figure 4. Figure 4 shows that the optimized condition has been obtained at 15 kV.cm^{-1} field strength at 6 N of KOH concentration for 6 hours etching time. This condition had minimum average standard deviation, which was equal to ±5, and it had produced the lowest uncertainty for experiment, which was the combination of a number of systematic and random errors.

The diameter of neutron track on CR-39 detector was calculated from 50 µm to 625 µm corresponding to the concentration of KOH and etching time. Figure 5 shows the diameters of the majority of tracks versus different etching time for different KOH concentration at 15 kV.cm^{-1} field strength. The minimum average standard deviation of track diameters at optimized condition was less than ±4. From figure 5, it is clear that with increasing the etching time and for higher concentration of the KOH, the size of the tracks have increased, and it was resulted in large counting error. For more clarification, the shape of tracks in different etching condition is presented in figure 6.

The experimental average response was evaluated from the net measured track densities (measured track density minus average background) with the corresponding reference values of each source which have been corrected for the effect of source-to-detector distance. The experimental and simulated average response at normal incidence and for the calibration sources are shown in table 2, which also displays the uncertainties corresponding to one standard deviation. The uncertainty for experiment was almost %10, which has been resulted by the combination effects of a number of systematic and random errors. The experimental uncertainty such as uncertainty of calibration exposure, the counting technique and variation in local background were considered.

Simulation of the detector

The calculation of track density in the etched layer of CR-39 was carried out with the assumption of the cross sections, the energy, angular distribution of recoils, and alpha particles expected in the etched layer of the CR-39 detector; however, tracks produced by neutrons via charged particles become visible only if the angle between the direction of the charged particle and the surface normal was less than a critical angle which varies significantly with particle energy. The registered track efficiency was the density of tracks, which were produced in the CR-39 per incident neutron. The total

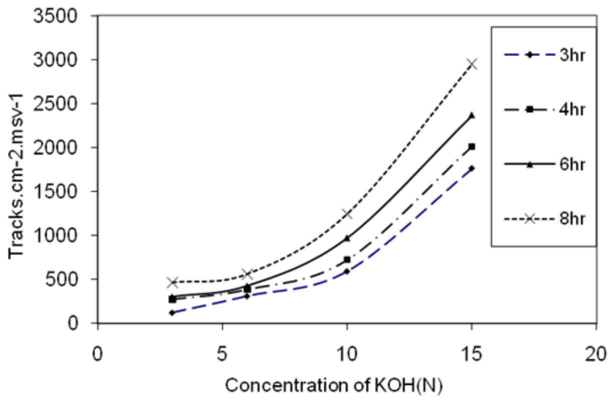

Figure 4. Neutron response against different concentration of KOH in the different time of etching for 1 mSv.

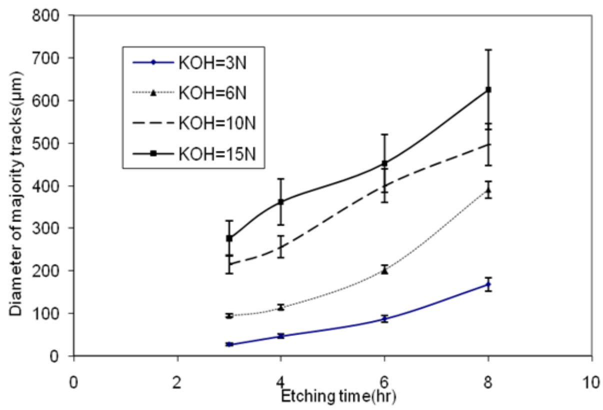

Figure 5. Diameter of the majority track on CR-39 detector in different etching time and different KOH concentration.

and partial contributions of the registered track efficiency of CR-39 from interactions with each constituent are shown in figure 7. This figure shows that registered track efficiency of CR-39 is a function of incident neutron energy. The tracks originating from the O and C recoils were negligible for neutron energy lower than 1 MeV. At low energy, only protons had sufficient energy to leave visible tracks, but O and C recoils made remarkable contributions to the overall response for higher neutron energies. Therefore the registered efficiency of the detector increased with the increase of the neutron energy up to 1 MeV, and later it decreased with the increase of the neutron energy. As the neutron energy increased, the elastic scattering cross-sections with H, C and O decreased. In addition, the ranges of C and O in CR-39 were shorter than the range for protons of the same energy. The model which has been presented in this work showed good improvement of the PADC response to fast neutron. This study added more data related to C and O recoils to the result which has been reported earlier [12], at the sensitivity of the detector for low energies of the incident neutron was considerably low. The response could have been on increased by using different radiators in contact with PADC based on the results presented [10, 12].

Figure 7 shows that C and O recoils contribute to the registered efficiency of CR-39, but it may not be possible to test it by experiment. The uncertainty for simulation

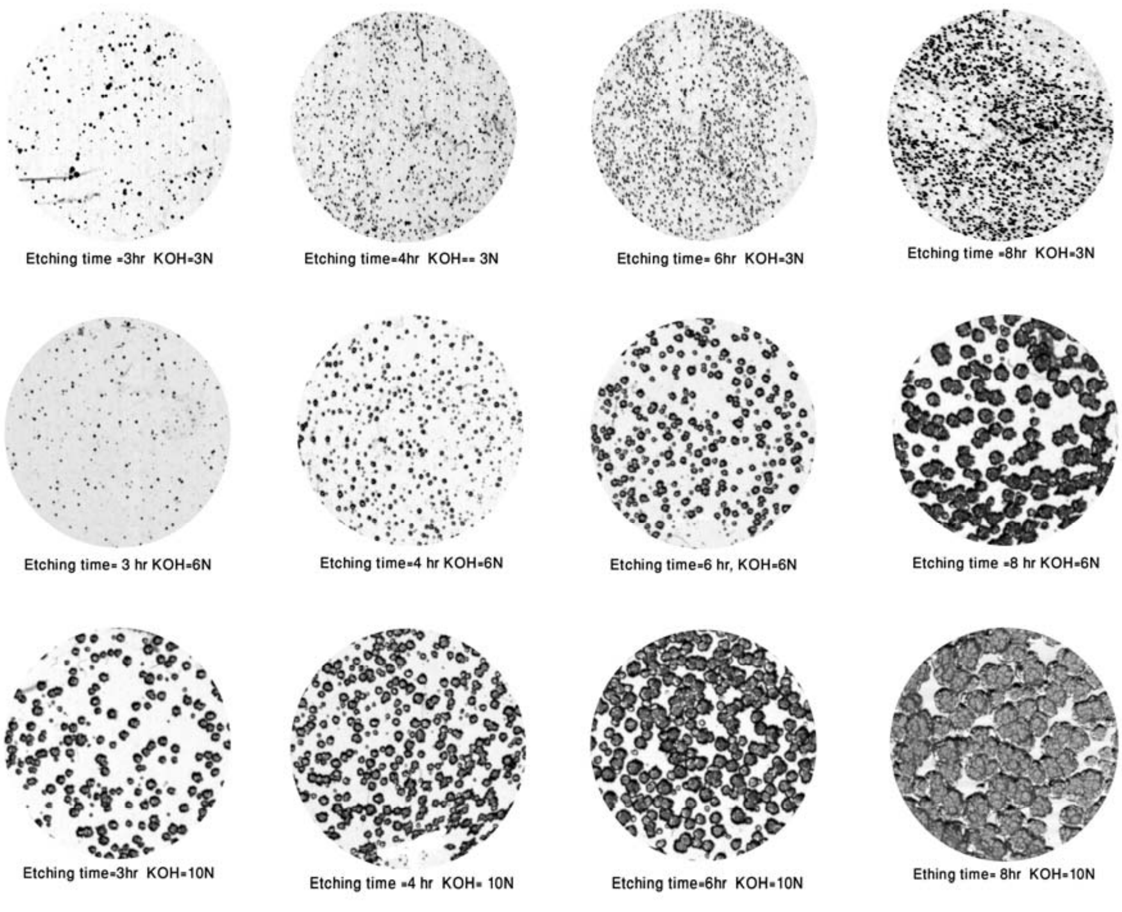

Figure 6. Shape of the track in different etching condition, which is scanned using a high resolution scanner (2000 dpi) with transparency adaptor.

Table 2. Experimental and simulated dose equivalent response of the PADC.

Source	Experimental average response(Tracks.cm^{-2}.mSv^{-1})	Calculated average response (Tracks.cm^{-2}.mSv^{-1})
Am-Be	415 ± 23	463 ± 32
Am-Be with 20 cm Polyethylene	512 ± 42	579 ± 38

was 0.07 relative errors, which was caused by all assumptions considered for simulation and the number of selected histories.

A very good agreement can be seen between the simulated and the experimental results of the registered efficiency of the detector for both sources. The average response increases as energy of neutrons moderated. This fact can be attributed to a detection efficiency increase when the energy of the source decreases.

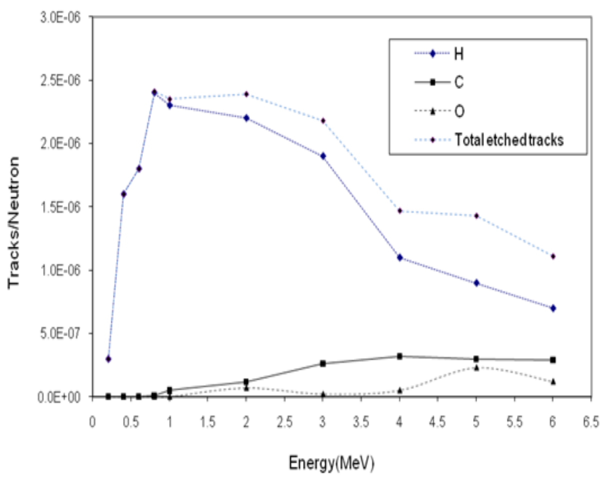

Figure 7. Energy dependence of the response for hydrogen, carbon, oxygen nuclei and total registered efficiency.

CONCLUSION

The registered efficiency of a CR-39 based detector to ^{241}Am – Be bare source could be calculated by MCNPX code, and the results were in a good agreement with the experimental results, if the appropriate critical angles for protons, carbon, oxygen and the optimized condition have been selected. Based on composition of CR-39 detectors, the elastic scatterings of H recoil was a dominating process at neutron energy below 1 MeV.

ACKNOWLEDGMENT

Authors thank Dr. Abbasi, Head of Physics group of Shahid Beheshti University and Mr. M.R. Sadegh Khani for their preparing instruments and laboratory tests on this study.

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