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Impact of x-ray dose on the response of CR-39 to 1–5.5 MeV alphas

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The CR-39 nuclear track detector is used in many nuclear diagnostics fielded at inertial confinement fusion (ICF) facilities, large x-ray fluences generated by ICF experiments may impact the CR-39 response to incident charged particles. To determine the impact of x-ray exposure on the CR-39 response to alpha particles, a thick-target bremsstrahlung x-ray generator was used to expose CR-39 to various doses of 8 keV Cu K- α x-rays. The CR-39 detectors were then exposed to 1–5.5 MeV alphas from an Am-241 source. The regions of the CR-39 exposed to x-rays showed a smaller track diameter than those not exposed to x-rays: for example, a dose of 3.0 ± 0.1 Gy causes a decrease of $(19 \pm 2)\%$ in the track diameter of a 5.5 MeV alpha particle, while a dose of 60.0 ± 1.3 Gy results in a decrease of $(45 \pm 5)\%$ in the track diameter. The resulting data were used to evaluate how x-ray doses received by CR-39 in OMEGA and NIF experiments affect the recorded charged-particle data.

I. INTRODUCTION

In many scientific disciplines, including Inertial Confinement Fusion (ICF) research, charged particles and neutrons are often studied with solid-state nuclear track detectors (SSNTD)^{1–12}. These are crystal or polymer materials in which ionizing nuclear particles leave detectable *tracks* of molecular damage sites^{3–5}. CR-39, a transparent SSNTD with chemical composition $C_{12}H_{18}O_7$, is particularly well suited for detecting nuclear particles because its induced tracks are revealed optically by etching with sodium hydroxide. These tracks can then be identified with an optical microscope with 100% efficiency for many charged particles that are normally incident to its surface (such as protons in the energy range 0.5 to 8 MeV^{4,6}). The uniquely high sensitivity of CR-39 has made it the detector of choice for light ion studies because the ionization levels of protons and deuterons are typically below the response thresholds of other detectors⁹.

Previous work with various types of CR-39 has studied the effectiveness of track etchants¹⁰ and has looked at CR-39 response to electrons³, neutrons¹¹, protons⁷, deuterons¹³, tritons¹³, and alpha particles⁹. One thing that has not been quantitatively studied is how the CR-39 response to these particles is modified when the CR-39 is exposed to high fluence of x-rays, as it is when used in experiments at ICF facilities like OMEGA¹⁴ and the NIF¹⁵, where doses of close to 5 Gy are typical.

In this paper, we present a comparison of the response to alpha particles of TasTrak[®] CR-39¹⁶ with and without exposure to x-rays. The theory of track formation in CR-39 is discussed in Section II. Section III presents the methodology and results. Finally, the effect of x-ray dose on the response to alpha particles as a function of both energy and x-ray dose are presented in Section IV, and summarized in Section V.

II. THEORY

Charged particles traveling through the CR-39 material deposit energy in the plastic through Coulomb scattering with electrons, leaving trails of damaged polymer chains¹⁸. When the plastic undergoes chemical etching, the broken molecular chains and free radicals become the seeds for the formation of conical pits or 'tracks'. The geometry of these tracks is dictated by the simultaneous action of two etching processes: chemical dissolution along the track (track etch rate v_t) and general dissolution on the etched surface (bulk etch rate v_b)². The sensitivity of CR-39 is defined by the ratio of track to bulk etch rates as described by

$$\frac{v_t}{v_b} = 1 + k \left(\frac{dE}{dx} \right)_{elec}^n \quad (1)$$

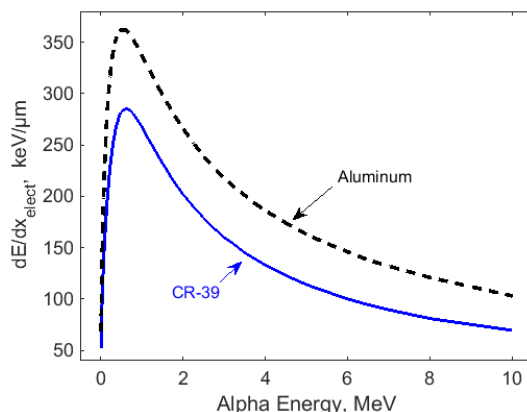


FIG. 1. Stopping power for alphas with energies in the range of 0-10 MeV in CR-39. This function is used to calculate the v_t/v_b ratio given by Eq. 1. As a reference, the stopping power for alphas in aluminum is also plotted for the same energy range. Values obtained from TRIM⁶.

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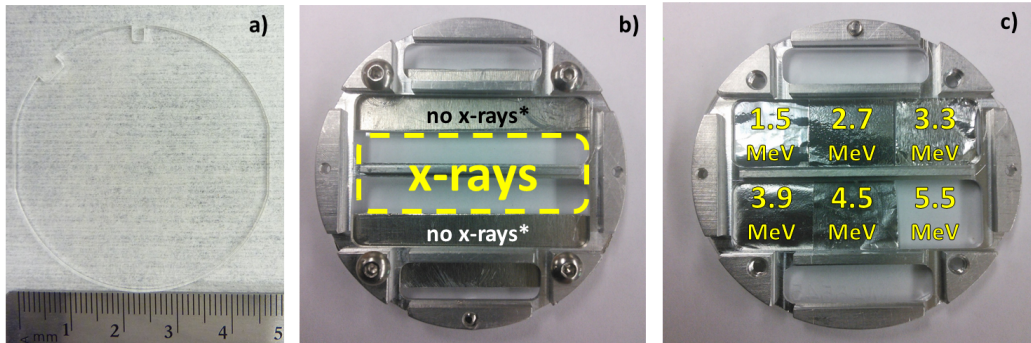


FIG. 2. a) A typical sample of 5-cm diameter, 1.5-mm-thick CR-39 used in this study. b) The x-ray shielding mask used in this study: two 1mm thick aluminum plates cover half of each mask window (labeled with an asterisk in the figure). The aluminum shield effectively blocks the 8 keV x-rays, transmitting only 10^{-6} of the incident intensity. c) Alpha particle range filters used in the study. Six different alpha energies were obtained on the CR-39 by ranging down 5.5 MeV alphas from an Am-241 source using different Aluminum filters. With this combination of x-ray and alpha filters, 12 regions of data were obtained on each CR-39 sample: x-ray-exposed and unexposed regions for each of 6 alpha particle energies. The area covered with aluminum between the two main windows provides a small background region on the CR-39. Each piece was then etched for 2 hr in an 80°C solution of 6 N NaOH.

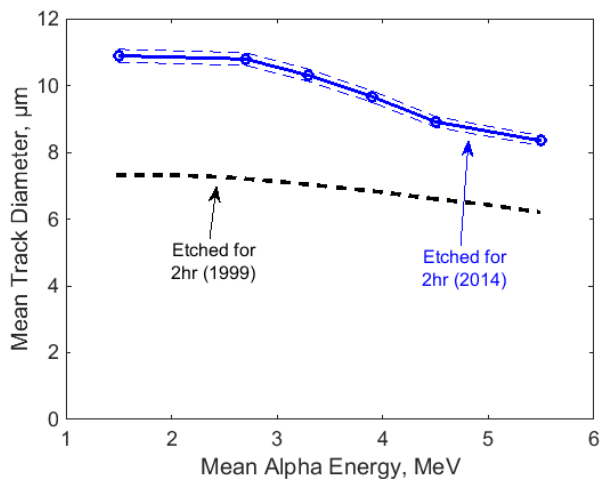


FIG. 3. Alpha track diameter and standard deviation as a function of mean energy for TasTrak[®] CR-39. Blue data were obtained from these experiments; for comparison, the measured data from the same experiment performed in 1999 by Hicks is also shown (black)¹⁷. As will be discussed, this $\sim 33\%$ difference is due to the change in the chemistry of the material (and therefore its etch rate) over the years.

where k and n are constants¹⁹. Typical values of k and n are¹⁷ 0.002 and 1.9, respectively. Here and throughout the paper, calculations involving the stopping power dE/dx are in units of $\text{keV}/\mu\text{m}$. Electronic stopping power is used, rather than the total stopping power, since the collisions with the electrons produces most of the ionization responsible for the molecular damage. $(dE/dx)_{elec}$ is determined from the ion stopping code SRIM⁶ (Fig. 1). The range of 5.5 MeV alpha particles in CR-39 is approximately $29 \mu\text{m}$.

If we consider the case of normal alpha incidence on the CR-39, simple geometry gives an analytical expression² for the track diameter (D):

$$D = 2v_b t \sqrt{\frac{v_t/v_b - 1}{v_t/v_b + 1}}, \quad (2)$$

where t is the etch time. Based on Equations (1) and (2), x-ray exposure could modify the sensitivity of the CR-39 through changing the bulk etch rate, track etch rate, or both. Combining Eq. (1) and Eq. (2) gives an equation of D as a function of the etch time, bulk etch rate, and stopping power

$$D = 2v_b t \left(\frac{2}{k \left(\frac{dE}{dx} \right)^n + 1} \right)^{-0.5}. \quad (3)$$

As a reference, Fig. 3 shows the mean track diameter as a function of alpha particle energy after a 2 hr etch when no x-rays are applied to the piece. Eq. 3 correctly explains this behavior for $k = 0.002$ and $n = 1.9$, as suggested in the literature¹⁷.

In the limit where $k(dE/dx)^n \gg 2$ (that is to say $v_t/v_b \gg 1$), the mean track diameter becomes independent of dE/dx and $D \approx 2v_b t$. For particles with large stopping powers, track diameters all saturate at a maximum value set by the bulk etch rate only (assuming that t_{etch} was carefully selected to avoid tracks from etching out). This behavior can be observed in the 1.5 and 2.7 MeV points in Fig. 3, for which the diameter has saturated at a maximum value ($11 \mu\text{m}$). In fact, the stopping power of the alpha particles in CR-39 continues to grow as alpha energy is reduced, until it peaks at around 0.7 MeV (see Fig. 1). The alpha energies studied in this work, therefore, probe the high-stopping-power limit. This feature, and a study of the bulk etch rate directly by measuring

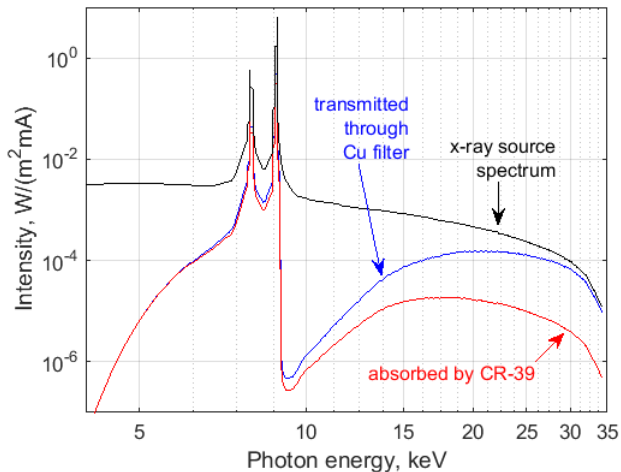


FIG. 4. Source, filtered, and absorbed x-ray spectrum for an 8keV (35kV and 18mA) x-ray line.

the volume and mass of the CR-39 before and after etching, is essential for understanding how x-rays impact the CR-39 sensitivity to particles.

III. EXPERIMENTAL RESULTS

A. Experimental study of track diameter versus x-ray dose

The experimental setup is shown in Fig. 2. CR-39 pieces (1.5 mm thick and 5 cm diameter) were exposed to different x-ray doses defined by several filters positioned as shown in the figure. The x-ray irradiation was performed using a thick-target bremsstrahlung x-ray machine (Philips Electronic Instruments X-ray Generator). This x-ray machine emits Cu K- α line-radiation at 8 keV, with a spectrum as shown in Fig. 4. A 35 micron Cu filter was positioned between the source and the CR-39 to isolate the 8-keV Cu K- α lines: 91% of the energy absorbed by the CR-39 was from this line radiation. Radiochromic film (RCF) was used to measure the incident intensity of x-rays in the sample plane²⁰. An average intensity was found, and was used to calculate the dose per unit time in the CR-39 pieces. Additionally, a surface barrier detector (SBD) was used to account for fluctuations in machine power output during the exposure.

Each piece was then exposed to alphas from a 0.1 μ Ci Am-241 alpha source (placed 9 cm away from the CR-39), which produced an approximate fluence of 15000 alphas per cm^2 . The 5.48-MeV alphas produced by the source were ranged down by a set of aluminum filters to obtain different energies of particles on the CR-39, as shown in Fig. 2. The x-ray- and alpha- exposed regions were overlapped such that each region with a particular alpha energy contained both x-ray exposed and unexposed portions. In total, 35 different experiments were conducted with various doses of x-rays from 0 to 60 Gy, including

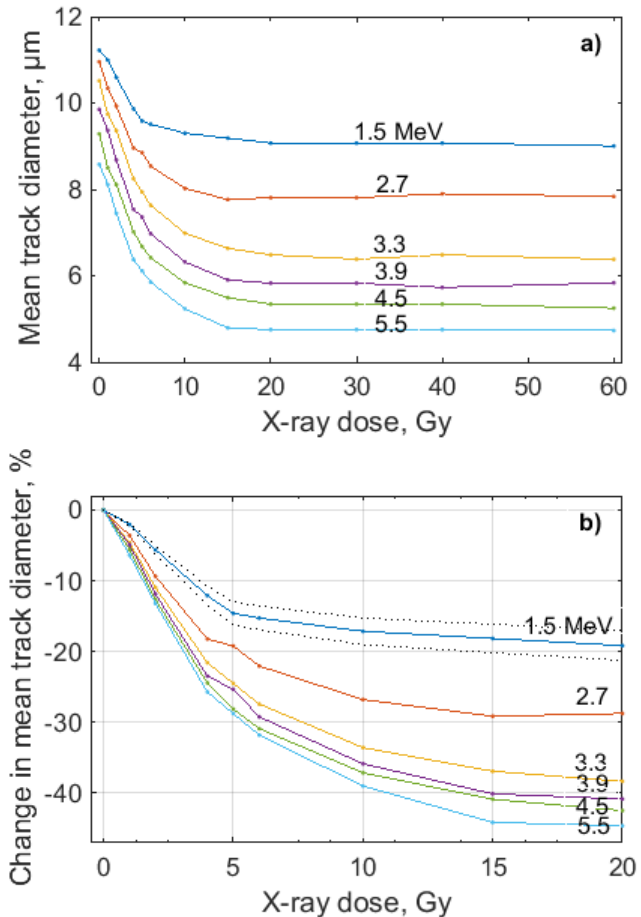


FIG. 5. a) Alpha track diameter as a function of x-ray dose. A characteristic uncertainty (from repeatability experiments) of track diameter of less than 3% (comparable to twice the size of the points) was observed in each case. Before achieving an asymptotic behavior, the greatest change happens below 10Gy. b) Percentage change of mean track diameter for each alpha energy as a function of dose. The standard deviation corresponds to approximately 10% of each value, but it is only plotted for the 1.2 MeV case for clarity. All samples were etched for 2hr in an 80°C solution of 6 N NaOH.

repeatability tests.

After exposure, the samples were all etched for 2 hours in an 80°C solution of 6N NaOH. An automated optical microscope system was used to scan each CR-39 sample and record individual track information (diameter, contrast and uncertainty) for later analysis¹³.

The mean alpha track diameters were observed to decrease with increased x-ray dose, as illustrated in Fig. 5. The mean track diameters are reduced roughly linearly with x-ray dose below 5 Gy, and approach an asymptotic value for doses of 20 Gy and above. The percentage change in the mean track diameter for each alpha energy as a function of dose is shown in Fig. 5b. From this

graph, for example, we can observe that a 1.5-MeV alpha will result in a track diameter smaller than normal by $12\pm 1\%$ if the piece of CR-39 is exposed to 4 Gy.

B. Experimental measurement of the Bulk etch rate versus x-ray dose

Given the track diameter versus x-rays dose behavior shown in Fig. 5a, we assumed that this trend was due to changes in the bulk etch rate as given by Eq. 3. As a result, the following v_b measurements were performed to investigate this hypothesis. The bulk etch rate was determined directly by measuring the change of CR-39 thickness before and after etching. An optical microscope was used to find this thickness by finding the difference in the height of the optical stage between focusing on the front and the back surfaces of the piece (optical thickness or Δz). The optical thickness is related to the true thickness by a function dependent on the index of refraction, $f(n)$. True thickness is defined as $d = \Delta z f(n)$. The bulk etch rate is given by²

$$v_b = \frac{d_f - d_i}{2t_{etch}} = \frac{\Delta z_f - \Delta z_i}{2t_{etch}} f(n), \quad (4)$$

where Δz_f and Δz_i are the final and initial optical thicknesses. A piece of CR-39 was divided into 3 sections, each exposed to a different x-ray dose. An unexposed region was measured as well for background reference. The optical thickness and the X and Y coordinates were recorded at the 18 regions measured in every piece. The piece was then etched for 5 hours, and the optical thickness was determined again in the locations previously recorded. This method allowed us to find a ratio between v_{bX} (bulk etch rate of regions exposed to x-rays) and v_{b0} (bulk etch rate of regions unexposed to x-rays), which is independent of the index of refraction:

$$\frac{v_{bX}}{v_{b0}} = \frac{(\Delta z_f - \Delta z_i)_{x-rays}}{(\Delta z_f - \Delta z_i)_{no-x-rays}}. \quad (5)$$

The observed ratio of v_{bX} to v_{b0} is shown in Fig. 6 as a function of x-ray dose. As shown, the ratio exhibits a similar behavior to that of the alpha diameters, changing rapidly with low doses and approaching an asymptotic value at ~ 20 Gy. Because the mean track diameter is proportional to the bulk etch rate (Eq. 3), we infer that the observed track dependence on x-ray dose is produced primarily by this trend in the bulk etch rate.

v_{b0} was also found using another method. If the initial (before etch) and final (after etch) masses of the sample are known, then the bulk etch rate is defined by

$$v_{b0} = \frac{\Delta m}{2A\rho t_{etch}}, \quad (6)$$

where the density $\rho = 1.30 \text{ g/cm}^3$ and the area $A = 19.6 \text{ cm}^2$. Using this formula, v_{b0} was found to be $2.66 \pm 0.06 \text{ } \mu\text{m/hr}$, which means that doses greater than

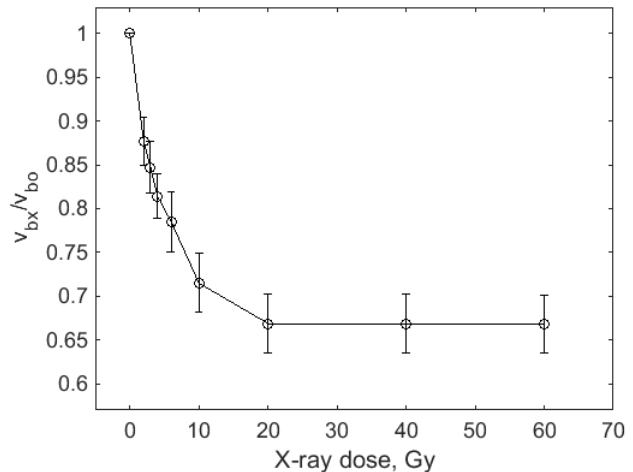


FIG. 6. Dimensionless bulk etch rate as a function of dose. A reduction to an asymptotic value of 0.66 ± 0.03 at approximately 20 Gy is observed. This behavior is consistent with the change in track diameters shown in Fig. 5a.

20 Gy result in an etch rate of approximately $1.80 \text{ } \mu\text{m/hr}$. Other researchers¹⁷ found in 1999 a value of $2.00 \text{ } \mu\text{m/hr}$ for v_{b0} , which suggests a change in the chemistry of CR-39 over the years. In fact, this $\sim 33\%$ difference in v_{b0} explains the $\sim 33\%$ disagreement observed in Fig. 3 between the experiments performed in 1999 and the ones presented in this paper.

IV. DATA ANALYSIS AND INTERPRETATION

As explained below, our interpretation of the data is that x-rays mainly change the bulk etch rate of the CR-39. Physically, this behavior takes place due to the rupture of the bonds in the polymer, which reduces the possibility of OH^- ions (from the NaOH) to react and extract parts of the molecules (etching).

Experiments showed that the bulk etch rate was dependent on the dose. Therefore, the mean track diameter of alphas is a function of both particle energy and applied dose. Because both the mean track diameter and bulk etch rate were measured for different doses, we can evaluate the extent to which the track diameter reduction is explained by the bulk etch-rate change, by calculating:

$$F(E, X) = \frac{D(E, X)}{D(E, 0)} \bigg/ \frac{v_b(X)}{v_b(0)}, \quad (7)$$

where E =energy and X =x-ray dose. This dimensionless parameter $F(E, X)$ turned out to be different than 1, which means that most of the x-ray effect is explained by the bulk etch rate change (Fig. 6), but there is still some residual effect. The explanation is that the track etch rate (v_t , see Eq. 2) also changes with changing x-

ray dose. As a result, an empirical model for Eq. 7 was found to quantify how much v_t is changing:

$$F(E, X) = 1 + \operatorname{erf}(aX) \left(b e^{\frac{-(E-c)^2}{2d^2}} + e \right) \quad (8)$$

Here, a fit to the data gives $a = (95.83 \pm 4.79)10^{-3}$, $b = (37.37 \pm 1.79)10^{-2}$, $c = 1.589 \pm 0.079$, $d = 1.054 \pm 0.053$, and $e = (-16.27 \pm 0.81)10^{-2}$.

The results, shown in Fig. 7, were found by dividing the results shown in Fig. 5a (normalized), by the results shown in Fig. 6. This figure might seem confusing at first sight because of its interesting energy dependence, but it can be mathematically explained as follows. Let us define $D(E, X)/D(E, 0)$ as $D_{x/o}$ and $v_b(X)/v_b(0)$ as $v_{x/o}$. We know that $v_{x/o}$ is independent of the incident particle energy, therefore, $D_{x/o}$ must be the term responsible for the energy dependence of $F(E, X)$. Figure 7 indicates that as the incident particle energy decreases, $D_{x/o}$ increases. Nevertheless, this increase only means that $D_{x/o}$ is closer to unity, and because $v_{x/o}$ behaves as in Fig. 6 then $F(E, X)$ will have values greater than one for low energies ($E < 3$ MeV) and values between zero and one for high energies.

Additional experiments were also performed to confirm the assumption that $(dE/dx)_{elec}$ was not changing with x-ray dose. In these experiments, pieces of CR-39 were exposed to different x-ray doses after alpha particles were already deposited in the sample. These alphas-before-x-rays experiments still show a decrease in the mean track diameter, as expected, but $D_{x/o}$ was 15-20% higher than with the normal (x-rays-before-alphas) methodology. This means that the curves in Fig. 5b scale up by 15-20% in each case. These observations support the fact that most of the effect of x-rays on the response of CR-39 to alphas comes from the change in the chemical structure of the polymer. This behavior is opposite from other observed environmental effects of the CR-39. It has been demonstrated, for example, that vacuum exposure longer than 16 hours *before* proton irradiation on CR-39 result in a decrease in proton sensitivity, whereas no effect is observed for up to 67 hours of vacuum exposure *after* proton irradiation¹⁹.

V. CONCLUSIONS

A comprehensive study of how x-ray exposure affects the response of CR-39 nuclear track detector to 1–5.5 MeV alphas showed that the mean track diameter of alpha particles for regions exposed to x-rays decreased rapidly for doses of less than 20 Gy, but that it reached a constant value for greater doses. This variation in the track diameter is primarily due to a proportional change in the bulk etch rate in the piece. A dimensionless function $F(E, X)$ was found to quantify the residual effect on the tracks due to changes in track etch rate. These results represent the first quantitative measurements of

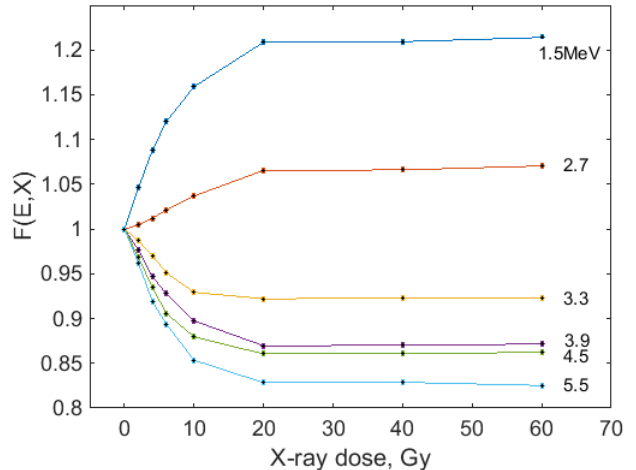


FIG. 7. $F(E, X)$ is a dimensionless parameter given by Equation 7 which describes the residual effect due to changes in the track etch rate when CR-39 samples are exposed to x-rays. All CR-39 pieces were etched for 2 h in an 80°C solution of 6 N NaOH (the model is independent of t_{etch} though.)

x-ray effects on CR-39, which will be of great importance for researchers who make use of this nuclear track detector in environments with high levels of x-rays. Future studies to examine the effect on other charged particles, especially protons and deuterons, will be particularly important for experimental applications that record these particles.

VI. ACKNOWLEDGMENTS

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