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Characterization of tracks from alpha-particles in PADC detectors for passive environmental monitoring of fast neutron radiation field

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Abstract
Fast neutron dosimetry is particularly crucial in the realm of radioprotection, as it significantly contributes to evaluating the radiation protection measures. This assessment aims to ensure and enhance the effectiveness of implemented safeguards and precautions, thereby minimizing the risks and potential harm associated with radiation exposure. Given the substantial biological damage that fast neutrons can inflict when interacting with living tissues, accurate dosimetry is indispensable for ensuring the safety of personnel in these environments and for optimizing radiation therapy treatments. Employing an indirect detector technique, this research focuses on characterizing the geometrical and optical properties of tracks produced by alpha particles resulting from boron-neutron reactions and distinguish them from the background made up of tracks generated from radon decay and impurities on the surface of the detector. This methodology holds potential utility particularly in situations where dosimeters are not adequately stored: while the idea of employing plastic or alternative materials to shield dosimeters from radon may appear straightforward, there are numerous factors complicating its effectiveness and universal applicability, for example when the radon proof encapsulations are not sealed perfectly. Through the development of a robust protocol for fast neutron dosimetry, we can not only differentiate between tracks produced by alpha particles of varying energy levels but also quantify the dose resulting from exposure to the neutron field. A solid-state nuclear track detector system (Politrack, Mi.am, Italy) was used to address the critical need for measuring exposure to fast neutrons, thermalized by polyethylene spheres. This advancement

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facilitates the implementation of more effective radioprotection strategies and contributes to the overall safety of radiation therapy procedures.

Keywords: fast neutron dosimetry, CR-39, PADC, solid state nuclear track detector, automatic image analyzer, dose correlation factor, microphotography

1. Introduction

Heavily ionizing particles leave trails (tracks) of damage on most insulating materials [1]. Chemical and electrochemical etching is performed to enlarge latent tracks caused by nuclear reactions into visible etch-pits, making them observable with microscopes. In dosimetry field, studying track density on dosimeter surfaces is a valuable tool for evaluating the dose received from exposure to radiation fields. Indeed, the variation in track density is proportional to the absorbed dose. Detectors operating on this principle are called 'Solid State Nuclear Track Detectors'—SSNTD.

SSNTD have attractive characteristics, such as non-fading of latent tracks and insensitivity to visible, UV, X, β and γ rays. In particular, the thermosetting poly-allyl diglycol carbonate plastic (PADC), commercially known as Columbian Resins #39 (CR-39), is remarkable as a detection material due to its high sensitivity to both fast and slow neutrons and low threshold energy for track formation [2]. In this study, we employ PADC detectors for slow neutrons through an n-alpha process. PADC is widely used for personal neutron dosimetry [3–6].

The Politrack automatic scanning system performs the morphological tracks analysis of SSNTD to classify tracks coming from different particles with varying energies and remove noise signals. One of the challenging tasks in the application of SSNTDs is the automation in differentiating among recorded tracks due to alpha particles with different incidence energies. This will be very useful in, for example, the studies of airborne radon progeny [7].

Output data files contain all information related to readings, and the operator can apply and/or modify morphological filters and visualize the results in real-time using a graphical tool.

In this study, a filtering protocol was developed to discriminate the signal from alpha particle tracks resulting from boron-neutron reactions (^{10}B(n,α)^{7}Li reaction yields an alpha particle of energy 1.47 MeV and a Lithium ion of energy 0.84 MeV) from background noise.

Background is made of dust, scratches and alpha particles tracks from Radon decays. Initially, the geometry and optical properties of 1.47 MeV alpha particles were studied with a dedicated experimental set-up. Subsequently, the filtering protocol was developed and used for discriminate background from alpha particles signal, and distinguish between 1.47 MeV alpha particles tracks and radon decay alpha particles tracks. Then, the dose-track density factor was evaluated for neutron radiation fields.

Figure 1. Set-up scheme of CR-39 samples and boron deposit on aluminium backing.

2. Materials and methods

The PADC plates used in this study were transparent, 2 mm thick, surface area of 25 $\times$ 25 mm^2, and 1.31 g cm^{-3} density. To assess the geometric and optical properties of alpha particles resulting from boron-neutron nuclear reactions, dosimeters were irradiated with 1.47 MeV alpha particles microbeam at the AN2000 facility of INFN-LNL [8, 9], with a particle flux lower than 10^3 alpha cm^{-2} for a duration of 10 s, at different incidence angles ranging from 0° to 75° in 15° increments. The energy of alpha particles impinging upon the detector surface was attained following their passage through a gold foil with a density of 500 μg cm^{-2}. In the second phase of the study, aimed at determining the dose-track density conversion factor for neutron radiation fields, the dosimeters were put in contact with a 0.53 μm thick boron film that was previously deposit on an aluminum backing, as shown in figure 1.

This choice was motivated by boron's well-established high neutron capture cross-section for thermal neutrons, typically in the range of kinetic energies of hundredth eV. The samples were exposed to an Am–Be neutron source with dose rate of 4.4 μSv h^{-1}.

Fast neutrons were thermalized by placing the dosimeters within spherical polyethylene (PE) enclosures with a diameter of 32 cm and a section of 1.2 cm thickness of Lead, placed 8 cm from the external surface of the sphere. The detectors were positioned in a configuration such that neutrons impinged upon their surfaces traversing successively through layers of PE, aluminum, and boron, where nuclear interactions occurred.

Different exposure levels were achieved by adjusting the exposure times, resulting in doses of 75, 110, 220, and 440 μSv, respectively. After the irradiation, the CR-39 plates were separated from the boron films.

For comparison, calibration dosimeters exposed at the Department of Nuclear Engineering at Politecnico di Milano (CeSNEF) at doses of 27, 45, 62 and 94 μSv were also studied.

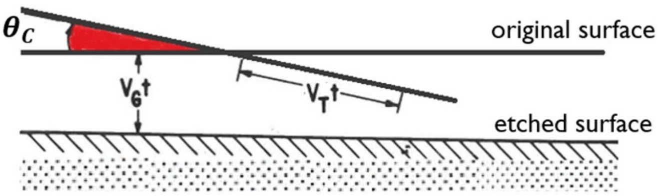

Figure 2. Illustration of the critical angle ϑ_c below which tracks cannot be detected after etching.

The post-exposure etching process was conducted in a 7.25 M NaOH aqueous solution at a temperature of 98 ± 1 °C, controlled using JULABO heating circulators (provided by JULABO GmbH, Germany) for a duration of 40 min. Sodium Hydroxide pellets ($M = 40.00$ g mol^{-1}) were sourced from PanReac AppliChem ITW Reagents. Ultrapure water (resistivity 18.2 MΩ·cm) was obtained via the water purification system located at INFN-LNL. Following a brief rinsing in flowing water, the etching process was halted by immersing the CR-39 dosimeters in a 2% aqueous acetic acid solution for 30 min. Subsequently, the dosimeters underwent another rinsing step in flowing water and were subsequently dried to be scanned and analyzed at the microscope.

In order to determine the amount of bulk etching, plate weights were measured before and after this process by a precision balance, and the bulk etch rate was calculated according to the well-established weight method [10]

$$V_B = \frac{\Delta M}{2\rho A t} \tag{1}$$

where ΔM is the difference between the initial and the after etching weight of the plate *g*, ρ is the density of the detector g cm^{-3}, A is the area of detector cm^2 and t is the etching time *h*.

The critical angle ϑ_c shown in figure 2 represents the minimum angle at which the etched tracks would appear on the detector surface, and can be calculated knowing the bulk etch rate V_B and the track etch rate, that is the etching velocity along the particle trajectory V_T [11]

$$\vartheta_c = \arcsin\left(\frac{V_B}{V_T}\right) \tag{2}$$

Theoretically, for 1.47 MeV alpha particles with a range of 6 μm in PADC calculated with SRIM [12], in the etching condition we mentioned before, ϑ_c ranges from 45° to 50°.

The used microscope is called Politrack, it is a multipurpose solid state nuclear track detector reader provided by Mi.am established in 1994 [13], that deals in the field of equipment for the analysis of radon and natural radioactivity. The system provides morphological information about the tracks. This might include data such as the length, width, shape, and orientation of the tracks, which can be essential for understanding the particles' characteristics and their interactions [13]. It has an intuitive interface that is crucial for researchers to engage with the system, manage the analysis, and comprehend the findings.

Figure 3. Mean grey level of the image obtained by Politrack system from 0 to 255.

2.1. Protocol

The developed protocol is written in Python, and for data analysis, libraries such as NumPy, Pandas, and SciPy were used. The code enables the fast and simultaneous analysis of multiple output files from the Politrack reading system. This reduces the analysis time for individual samples and allows for cross-data analysis. The protocol requires input from Politrack system files, which consist of raw data (unprocessed readings without track distinction filters). In the initial phase, the protocol provides statistical information on this data, particularly regarding outliers in each distribution (*e.g.* track area distribution, perimeter, etc), average values, associated errors, and allows for their removal from the track count.

Additionally, it allows for the visualization of results and their rapid storage, typically within seconds for hundreds of files with a track count of less than 10.000, which means hundreds of dosimeters with a number of tracks (before the filtering process) of the order of $3{\cdot}10^3$ tr cm^{-2}.

3. Results and discussions

The mass decrement method was employed to determine V_B and its associated error for seven dosimeters, finding a value of 12.224 ± 0.001 μm h^{-1}. This result aligns with the values reported by Nikezic and colleagues [7], indicating the alpha particle range remains partially unaffected by the chemical attack, allowing them to be visible under microscope observation after etching. While, in the second part of the study, the tracks of Lithium ions are completely eroded by this etching process.

3.1. Geometrical characterization of tracks

To distinguish the signal generated by tracks from alpha particles given by the boron-neutron nuclear reaction, first, CR-39 dosimeters were irradiated with alpha particles of 1.47 MeV energy. The irradiation was performed at different angles.

The critical angle observed for 1.47 MeV alpha particles under the specified etching conditions approximates 45°. Consequently, in dosimeters exposed to incident angles below this threshold, the signal is prominently discernible. Conversely, when exposed at angles beyond this value, the signal tends to become indistinguishable from the background. The mean grey level of tracks, as determined through microscopic analysis post-etching shown in figure 3, serves as one of the most effective parameters for distinguishing the signal from background noise. These signals exhibit a grayscale range spanning from 50 to 150.

Table 1. Mean track radius for dosimeters irradiated with 1.47 MeV alpha particle after filters on grey scale, axis dimension and axis ratio were applied.

Incidence angle (°)	Mean radius (μm)
0	6.30 ± 0.01
15	6.22 ± 0.02
30	5.85 ± 0.05

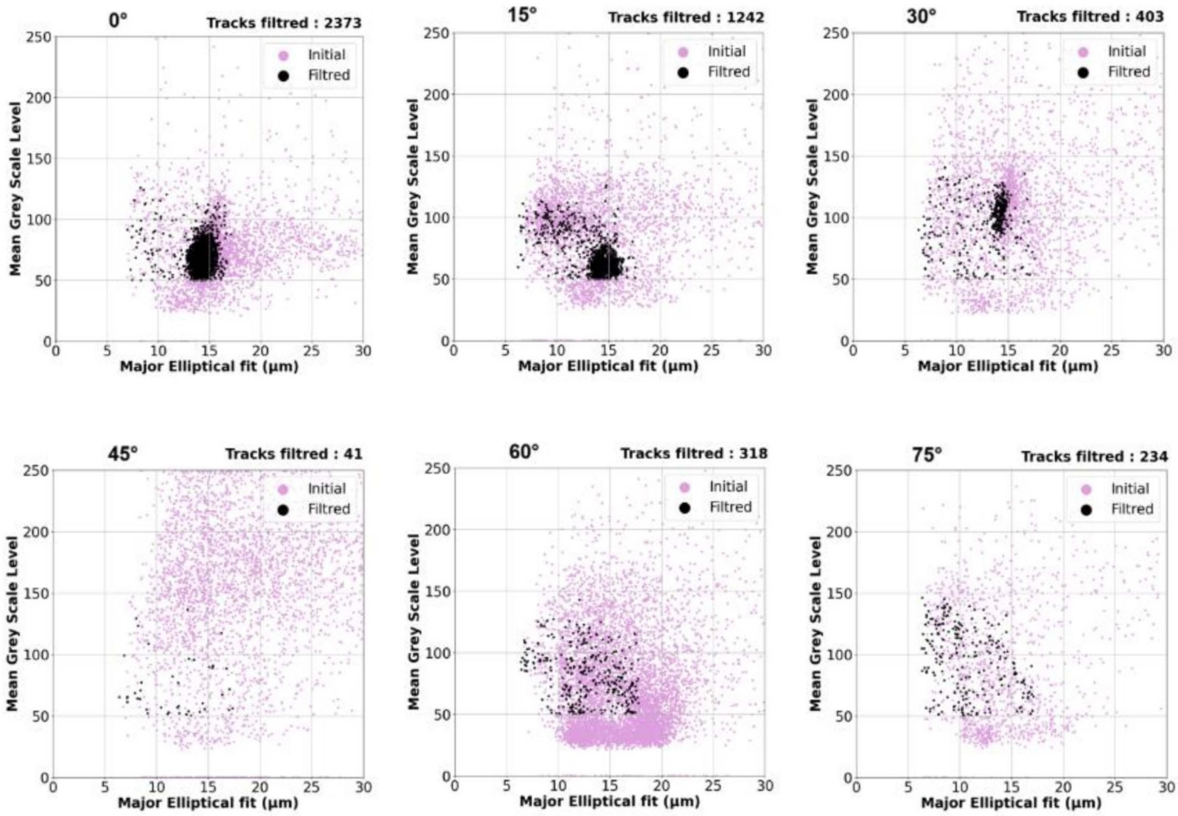

Figure 4. The 1.47 MeV alpha particles' signal on CR-39, when exposed to varying incidence angles, was analyzed after applying filters on the following parameters: axis dimensions, axis ratio, mean grey level, upper area limit, upper convex hull (C. H.) area limit, and residual of the fit.

Additionally, the axis ratio (major over minor) was considered between 1.0 and 1.5 (where 1.0 identify perfectly circular tracks). Furthermore, the observed tracks exhibited axis dimensions spanning from 4 to 18 μm and this range was used as additional tracks filter.

After applying the previously discussed filters, the mean track radius decreases increasing the incidence angle, as shown in table 1.

For perpendicular incidence, light rays within the CR-39 detector undergo total internal reflection at all points on the track surface, preventing them from reaching the detection hemisphere of the optical microscope. As a result, tracks appear as completely dark circles. However, as the incidence angle increases, the trajectory of the track becomes more superficial with respect to the CR-39 detector, leading to a transition phase where the track profile becomes nearly parallel to the detector surface. In this configuration, the track appears as an elliptical form, with shorter parallel side inside the CR-39 and parallel side on the surface [11]. This enables the assessment of the maximum area boundary and the maximum convex hull (the area of the largest polygon that can be enclosed within the track's perimeter) in the dosimeter exposed to perpendicular incidence, resulting in a value of 190 μm^2.

Finally, plotting the Q–Q plots, the mean residual value was evaluated, resulting in a value of 12. The filtered signals are shown in figure 4.

Seven unexposed dosimeters were stored for two months in a room with a mean annual radon concentration equal to the value of 100 ± 9 Bq m^{-3}, measured with ENEA's CR-39 radon detectors, enclosed in a closed diffusion chamber during two different periods in a year. The mean background number of tracks was evaluated, and a number of 300 ± 100 tracks was found, as shown in table 2. So, as expected, the signals of dosimeters irradiated at incidence angles close to and higher than the critical value, after the filtering process, are comparable with noise signal since these tracks are eroded during the chemical attack.

Table 2. Table of background number of tracks for 7 dosimeter not exposed and mean value.

Background	Track count
Dosimeter 1	296
Dosimeter 2	153
Dosimeter 3	178
Dosimeter 4	195
Dosimeter 5	364
Dosimeter 6	443
Dosimeter 7	433
Mean	300 ± 100

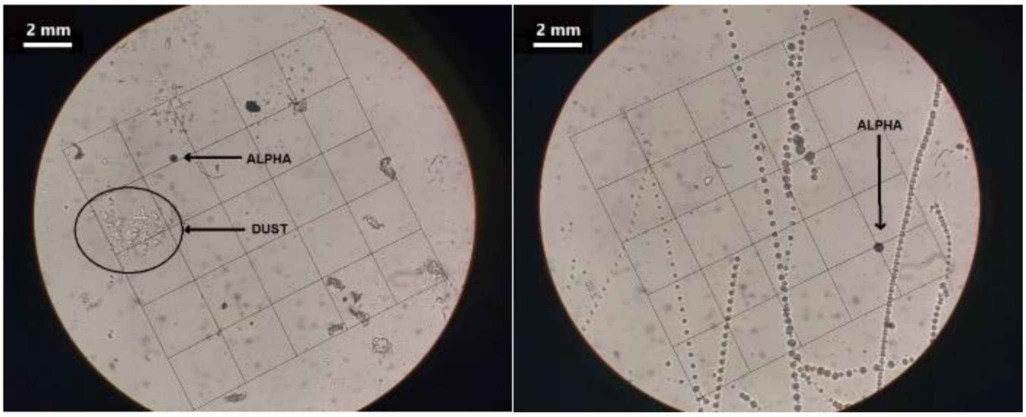

Figure 5. Photomicrograph of recoil tracks in CR-39 plastic detector, etched in 7.25 M NaOH at 98 °C for 0.67 h.

Table 3. Mean number of tracks vs. dose and mean number of tracks per dose vs. dose for dosimeter exposed to Am–Be neutron source for different dose.

Dose (μSv)	75	110	220	440
Mean track number (tr)	911 ± 140	1511 ± 215	3491 ± 372	5501 ± 186
Mean track number per dose (tr μSv^{-1})	12.15 ± 1.87	13.74 ± 1.95	13.55 ± 1.69	12.50 ± 0.43

From the figure 5 it is possible to notice the differences in optical and geometrical properties between alpha particles tracks and noise.

3.2. Boron-neutron alpha particles dose evaluation

The second part of the study, aimed to the assess of the dose-track density conversion factor for a neutron radiation field. Dosimeters coupled with boron films were exposed to 75, 110, 220 and 400 μSv from Am–Be neutron source. Five dosimeters were exposed to every dose. The mean number of tracks, as depicted in table 3, is observed to increase with the dose after the application of the developed filtering protocol, consistent with previous knowledge. Also, as shown in figure 6, the mean track number per dose vs. dose linear fit gives the equation $y = +13.30 \pm 0.80$ and R-square equal to 0.999 indicating the goodness of the fit.

Furthermore, the use of the grayscale filter proves instrumental in the identification of particles, as depicted in figure 7, showing the graph of mean grey level vs. hydraulic radius (that is the ratio between pit area and pit perimeter).

The filtered signal is derived subsequent to the application of the designed filters, excluding the one pertaining to grayscale levels. Notably, two discernible clusters manifest: the lower cluster corresponds to alpha particles resulting from Radon decay, characterized by higher energy levels than alpha particles generated by boron-neutron interactions, enabling them to penetrate the detector surface more deeply and exhibit darker appearances (mean grey level lower than 50). Meanwhile, the upper cluster is attributed to alpha particles originating from boron-neutron reactions (mean grey level higher than 50).

Then, we evaluated the mean background track density. The average background tracks density was calculated to be 120 ± 40 tr cm^{-2} for dosimeters irradiated at INFN-LNL, this value is due to ageing effects and non-compliance with proper dosimeter storage. Also, a separate set of dosimeters exposed to doses of 27, 45, 62 and 94 μSv at CeSNEF (five dosimeters at every dose) with lower mean background track density of 6 ± 3 tr cm^{-2} was studied. The track density per dose against dose graph of both samples is illustrated in figure 8. The linear regression equations for the samples are shown in table 4. The comparison indicates

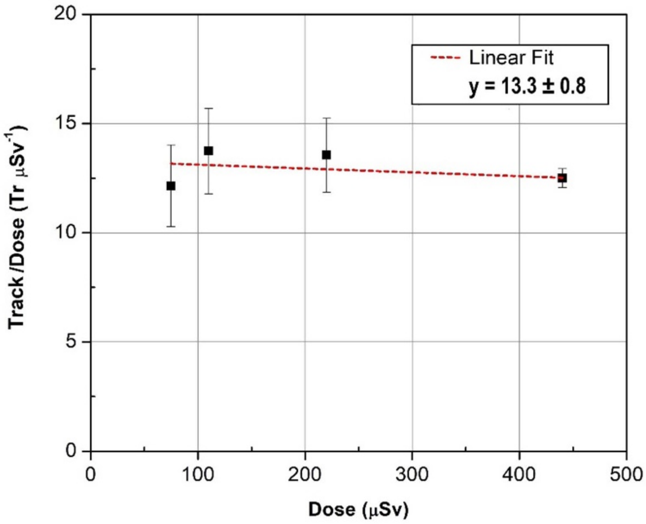

Figure 6. Linear fit of mean track number per dose vs. dose for dosimeters exposed to Am–Be neutron source at different exposure levels (75, 110, 220 and 440 μSv), without subtracting the background.

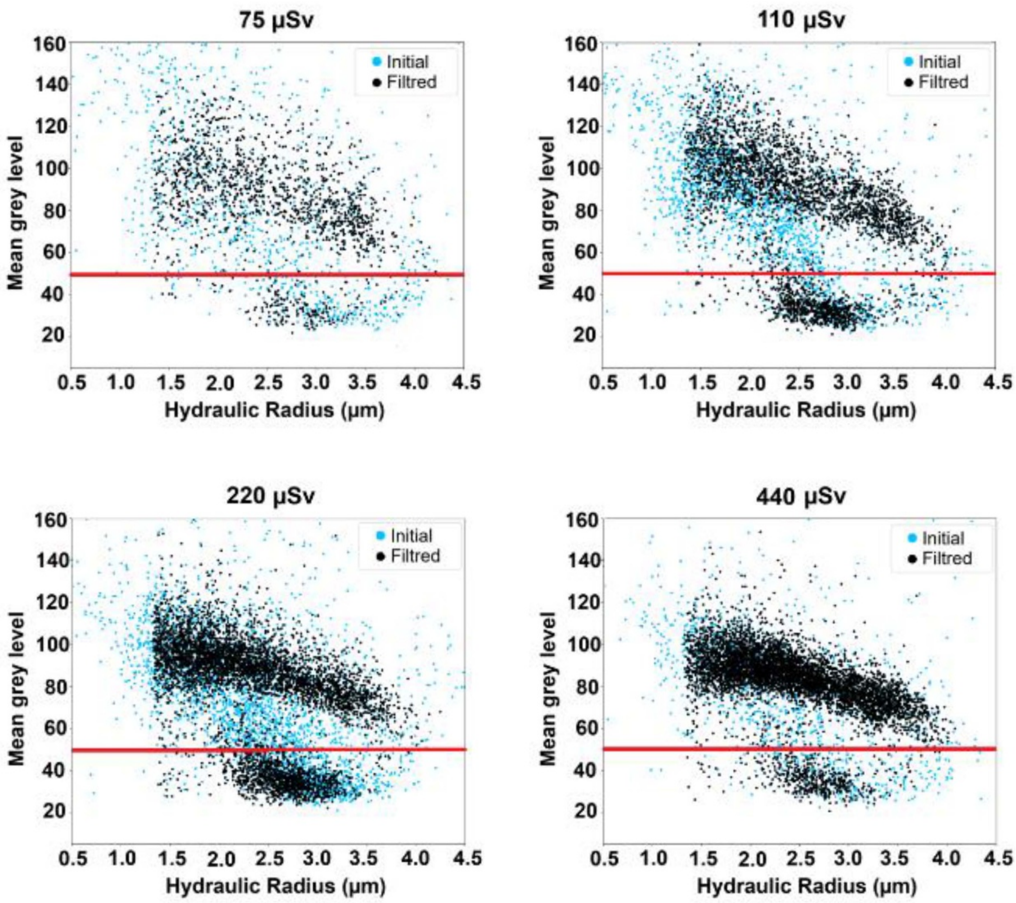

Figure 7. The signal generated by 1.47 MeV alpha particles on CR-39 coupled with boron films and exposed to an Am–Be neutron source was analyzed after applying filters (with the exclusion of the one related to grey scale). The red line is indicative of the grey scale level of 50.

that the protocol effectively eliminates significant background interference.

Finally, to evaluate the conversion factor we assessed the slopes of track density vs. dose graph. The results are illustrated in figure 9. The t-test revealed that the conversion factors obtained from these two sets of experiments, shown in table 5, are within the 55% confidence interval.

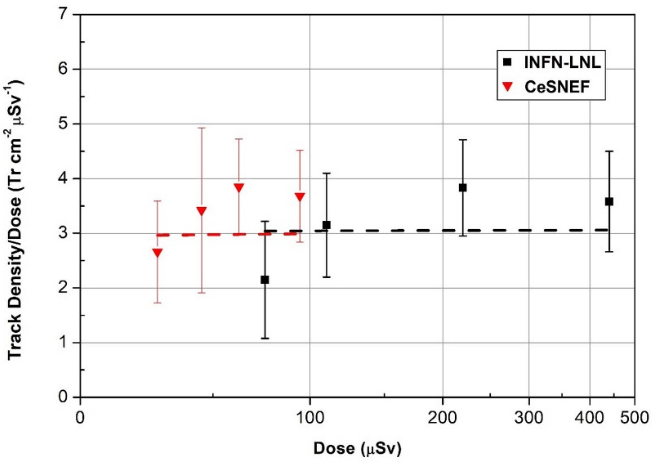

Figure 8. Linear regression of data for samples exposed at INFN-LNL at different doses (75, 110, 220 and 440 μSv) in black squares, and linear regression for samples irradiated at CeSNEF at different doses (27, 45, 62 and 94 μSv) in red triangles. Every point in the graph is given by the difference between the mean of five values and the mean background track density.

Table 4. Linear fit parameters' summary for samples irradiated at INFN-LNL and at CeSNEF, with background subtraction.

	Intercept		Slope		Statistics
Sample	Value	St. Er.	Value	St. Er.	R-Square
INFN-LNL	2.92	0.64	0.00	0.00	0.876
CeSNEF	2.21	0.72	0.01	0.01	0.899

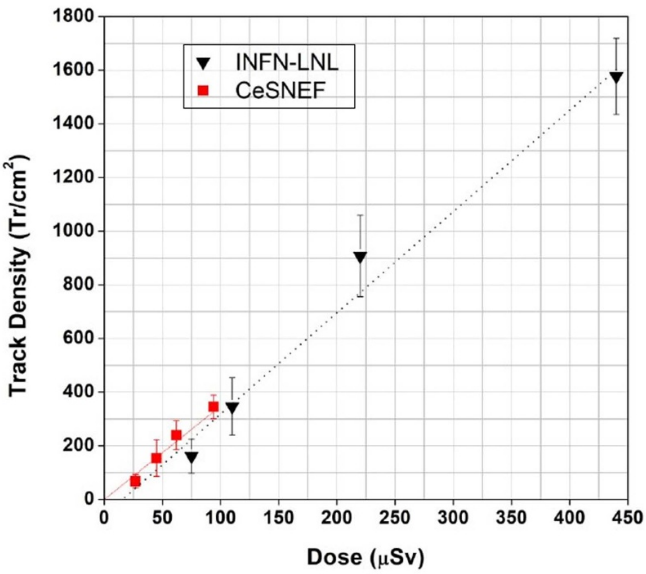

Figure 9. Linear regression of data for samples exposed at INFN-LNL at different doses (75, 110, 220 and 440 μSv) in black triangles, and linear regression for samples irradiated at CeSNEF at different doses (27, 45, 62 and 94 μSv) in red squares. Every point in the graph is given by the difference between the mean of five different samples and the mean background track density.

Table 5. Linear fit parameters' summary for samples irradiated at INFN-LNL and at CeSNEF.

	Intercept		Slope		Statistics
Sample	Value	St. Er.	Value	St. Er.	R-Square
INFN-LNL	−60.50	55.30	3.776	0.356	0.965
CeSNEF	−1.20	4.90	3.488	0.312	0.969

4. Conclusion and remakes

The impact of various elements, such as the angle of incidence and the conditions under which etching occurs, on the intricate nature of track geometry was emphasized in this study. Our investigation has brought to light a correlation between particle incidence angles and etching conditions, thereby affirming the theoretical critical angle through empirical findings. Furthermore, our research has elucidated the influence of the angle at which particles approach a material on the geometric characteristics of tracks, revealing that higher incidence angles lead to decreased axis dimensions of tracks.

The grayscale properties of the tracks serve as quantitative indicators for distinguishing between different energy particles. Variations in the grayscale levels of the tracks facilitate the differentiation of particles based on their energy levels. We observed that alpha particles with higher energy, such as those generated by the decay products of Radon, penetrate deeper into the CR-39 material, resulting in darker tracks with grayscale levels below 50. Conversely, tracks originating from neutron interactions with boron yield lighter tracks. In future investigations, it would be of particular interest to differentiate between tracks caused by alpha particles stemming from Radon decay.

The newly implemented protocol for the Politrack system was utilized to evaluate the dose-track density conversion factor, and the results obtained with a 55% confidence level indicates the reliability of the protocol. This capability remains resilient even under challenging conditions characterized by significant background interference, represented by the presence of thousands of background tracks, thus affirming the efficacy of the protocol. The developed protocol is a starting point for future optimization of tracking parameters and a more precise reading of PADC dosimeters.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Acknowledgments

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