

Registration Properties of LR-115 NTD_s and their use in Radon Level Determination and Equivalent Dose Evaluation

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Abstract: Cellulose Nitrate (LR-115) Nuclear Track Detectors (NTDs) were irradiated systematically with alpha particles from ²⁴¹Am alpha source with an energy range of about 1 to 5 MeV at an incident angle 90°. Etching characteristics and registration properties were studied. The alpha tracks in LR-115 films were measured with an optical microscope attached to an eyepiece micrometer with a precision division of $\pm 0.22 \mu\text{m}$. The main parameter is the bulk etch rate (V_B), which was carried out that controlled during the track formation. The dependence of V_B on etchant concentration and temperature was extensively investigated. The activation energy, E_b , of etching for the studied detectors was calculated, and an average value of $E_b = 0.93 \pm 0.03 \text{ eV}$ was extracted. The registration parameters were performed. The used plastic foils were pre-sparked to clear the alpha track diameter openings. The registration properties of pre-sparked LR-115 NTDs were enhanced up to the value of 5.0 MeV α -energy. The Response function, V , as a function of residual range, R , and restricted energy loss (REL_{350}) was deduced. In addition, the registration parameters and the excess etch-rate ratio ($V-l$) of alpha tracks were achieved. The dependence of response, V , and the excess etch-rate ratio ($V-l$) upon R as well as REL_{350} , was investigated. Also, it is clear that response V was decreased rapidly, and the excess etch-rate ratio ($V-l$) was increased in a power fitting formula. The relation between ($V-l$) and REL_{350} obeys a power law that agrees well with some previous works.

The radon concentration, C_{Rn} , was related to track densities using an AB-5 calibrated monitor through a calibration factor, k , of LR-115 detectors. A radon concentration chamber was designed and attached with an AB-5 monitor to determine the calibration factor, k . The average value of k was found to be $k = 0.0599 \text{ track cm}^{-2}\text{d}^{-1} \text{ per Bq m}^{-3}$. Also, the equilibrium factor, F , was calculated experimentally with the empirical equation of the open and closed cup mode technique for track ratio. The equilibrium factor, F , was calculated as $F = 0.45$. The LR-115 detectors were used in the radon level determination as CR_n and radon equivalent dose, Deq , in some dwellings in the Menoufia zone in Egypt. The minimum detectable value of equivalent dose to the maximum value was 0.4 - 0.6 mSv/y in two different modes of measurement. An average value of radon equivalent dose was deduced as $Deq = 0.5 \text{ mSv/y}$. An average value of radon-annual equivalent dose in houses was found to be $\langle DRAED \rangle = 0.72 \text{ mSv/y}$. Results of this study are discussed within the framework of nuclear track formation theories and the etching concepts mechanism.

Keywords: LR-115 NTDs, Etching, Sparking, α -Spectroscopy, AB-5 radon concentration monitor, Radon Calibration Factor k , Radon Equilibrium Factor F , Equivalent dose Deq .

1 Introduction

Nuclear Track Detectors (NTDs) technique is being used method in different fields of radiation science and technological applications [1-8]. CR-39 and LR-115

cellulose nitrate track detectors are oftentimes used in radon dosimetry and many other applications, such as fast neutron detection and ²³⁸U and ²³²Th determination in rock samples [9-12]. It is utilized in many fields like nuclear fission induced by neutron activation in a nuclear reactor,

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sterilization surgical equipment, treatment of agricultural products and polymer modification as well as in several technological areas of mixed field of radiation, such nuclear track detectors may sometimes be exposed to high intensity of γ -radiation including the external surface of spacecraft and satellites, space environment, exposure to radiation in nuclear power plants and accelerators which can modify polymer characteristics [13-15]. In addition, films, specifically, are commonly used in the fields of radiation detection and nuclear physics, as well as utilized in heavy ion as a source for focused beams [16]. Furthermore, several dosimeters and new kinds of detectors can be used to measure directly in wide doses and are currently available for therapeutic radiation by changing their color. They are EBT2, EBT3, and EBT4 GafChromic® films to detect α -radiation from a ^{241}Am source, as well as X-ray, and γ doses were investigated. The excess response of EBT2; EBT3 GafChromic® films by removing one protective layer from the film, the response to α -irradiation was enhanced, allowing it to measure doses up to 90 Gy. Also, GafChromic EBT4 is considered a superior response in comparison with EBT3 when exposed to 6-18 MV X-rays, as the calibration curves for EBT4 indicated improved signal-to-noise ratios. In radiotherapy case studies, EBT4 provided the best agreement with the planned doses for patients than EBT3 [17-22].

One of the interesting applications of nuclear track detectors is in measuring high response α registration for the activity of ^{222}Rn concentration through alpha track determination. Cellulose nitrate (LR-115 films from Kodak Pathe Company, France) films and Makrofol® TP-244, available as Makrofol detectors (Bayer® AG, Leverkusen, Germany) provided the single-side UV-protected Makrofol® TP 244, have been used as track detectors in which visible tracks can be formed after ion irradiation and suitable chemical etching. The etchability properties of such detectors were determined through the measurements of bulk (V_B) and track (V_T) etch rates. Several authors have studied the etching properties of various plastic materials under different conditions. Tracks were considered as three-dimensional objects, which enabled calculations of the track parameters, including the lengths of the major and minor axes of track openings, and plotting of their profiles [23-27].

In this paper, a detailed study of LR-115 etching properties is carried out to obtain the optimal etching conditions for each batch of detector samples. For this reason, the dependence of the etching characterization of the studied plastic on normality (c) and temperature (T) of the etchant solution was extensively studied using fission track diameters. Cellulose Nitrate (LR-115) NTDs were also calibrated for radon-concentration measurements using a well-known active detector for standardization. Radon level determinations in some houses were measured, and the annual equivalent doses were calculated using LR-115 detectors.

2 Experimental

2.1 Materials

In the present work, Cellulose Nitrate sheets $[C_6H_7O_2(OH)_3(NO_2)]$ of LR-115 type II plastic detectors with a thickness of 12 μm and manufactured by Kodak Pathe Company, France, were used.

2.2 F, F and α -irradiation facilities

The plastic two sets of LR-115 detectors (Kodak Pathe Company, France), each set of detectors of (3×3) cm² in area and a thickness of 12 μ m, were irradiated with fission fragments of ²⁵²Cf and alpha particles from a ²⁴¹Am thin source of activity 0.1 μ Ci, respectively. All these sources were in disk form, and their activities were known. The detectors were exposed using 2π geometries by placing detectors directly at the normal incidence of the source in air. Detectors of set 1 were exposed to fission fragments, and detectors of set 2 were irradiated by alpha particles at varying distances in front of the air collimator opening of about 0.5 mm allowed alpha particles' energy to enter through the plastic foils at normal incidence and to have a minimum value of energy dispersion as well. Exposure time was adjusted and calculated using the activity of the sources to avoid overlap of etched tracks in LR-115 detectors and to have a track density of order nearly 10^4 tracks/cm².

2.3 Rn irradiation chamber

Irradiation of detectors by Rn-gas was also carried out in a specialized irradiation chamber attached to pc and designed for such purpose [20]. The schematic diagram of the irradiation chamber is illustrated in Figure 1, where the chamber is connected to a radium source by adjusting the AB-5 device. A calibration curve that relates Rn concentration (CRn) to the track density was obtained using LR-115 track detectors.

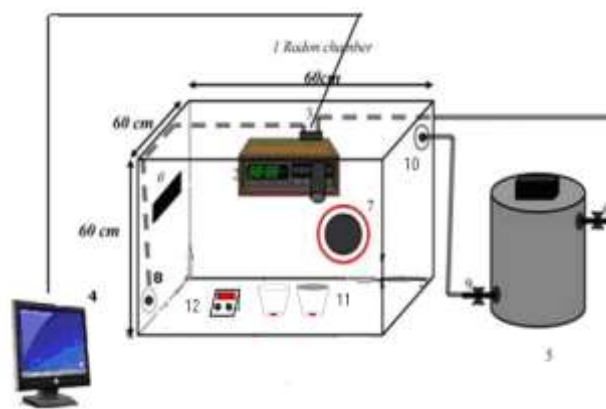


Fig.1: Schematic diagram of the Rn-irradiation circuit.

- | | |
|------------------------|---------------------|
| 1- Radon chamber. | 2- AB-5 Monitor. |
| 3- AB-5 internal pump. | 4- Computer. |
| 5- Radon source. | 6- Rectangular port |
| 7- Circular port. | 8- Outlet tubes. |
| 9- Two valves. | 10- Inlet tube. |
| 11- LR-115 | 12- Hygrometer. |

3 Etching Equipment Set-ups and Track Counting

3.1 Bulk Etch Rate (V_B) Track Diameter Measurements

For the etching process, a Julabo water bath with shaker and available temperature accuracy of about $\pm 1.0^\circ\text{C}$ was used. LR-115 foils were immersed in a chemical etching solution of 2.5N NaOH. After etching, detectors were rinsed in distilled water for more than one hour in order to remove etch products from the surface and then thoroughly washed with distilled water using an ultrasonic cleaner for a sufficient period of time. Finally, detectors were carefully dried using a cold air dryer.

Through the present work, V_B was measured using the fission-fragments irradiation method in which plastic samples were carefully cut and then irradiated first with ^{252}Cf open source. Also, the plastic detectors were first exposed to a ^{241}Am radioactive source. The V_B value was calculated using the following relationship [28]

$$V_B = D_{ff}/2t_e \quad (1)$$

where D_{ff} is the fission track diameter and t_e is the etching time.

Track parameter measurements of fission and alpha track diameters of LR-115 detectors were performed using an optical microscope (Olympus BH-2) with 600X amplification. At least 100 tracks in each case of exposure were counted and analyzed. An eyepiece was attached to the microscope with a screw micrometer of each division of $0.22\ \mu\text{m}$.

3.2 Sparking Process

It is an electronic measurement technique for track counting. LR-115 film is used in spark equipment and placed between two thick metallic plates, and a thin layer of Al foil evaporated on a polystyrene plastic backing. The LR-115 detector was first etched through track holes before sparking started. As soon as a high voltage is applied to the electrodes, the discharge through each spark hole takes place, which can be counted through a counter. Atypical circuit of the home-made spark counter, used in this study, consists of a 5 kV DC-power supply and a NIM-module single channel analyzer connected to a scalar counter, and is shown in Figure 2.

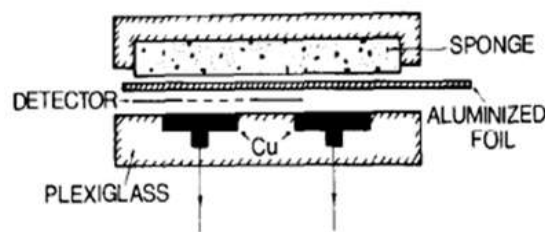


Fig. 2: Side-by-side-electrodes spark counter.

4 Results and Discussion

To enhance the shape of the track and to remove all etchant from the holes, one has to first apply the pre-sparking method at a higher voltage ($\sim 1\ \text{kV}$). This can successfully be done without any effect on track diameter. Figure 3 represents two photomicrographs of 2 MeV α -tracks in LR-115 detectors etched in 2.5N NaOH at 50°C for (a) $h=5\ \mu\text{m}$ and (b) $6\ \mu\text{m}$, without pre-sparking. From inspection of Figure 3, one can easily notice that some etchant residue is still around the track holes, although careful cleaning was carried out. Such a residue makes track boundaries less well defined and consequently increases the uncertainty of track parameter measurements.



Fig. 3: Photomicrograph of 2 MeV α -tracks in LR-115 detectors, etched in 2.5N NaOH at 50°C for (a) $h = 5\ \mu\text{m}$ and (b) $6\ \mu\text{m}$, without pre-sparking.

Figure 4 provides a photomicrograph of pre-sparked (900 V) for (a) 1.0 MeV, (b) 3.0 MeV and (c) 3.5 MeV α -particle tracks in LR-115 detectors etched in 2.5N NaOH at 50°C for $h = 4\ \mu\text{m}$, respectively. An improvement of track shape and contrast is clearly seen in Fig. 4.

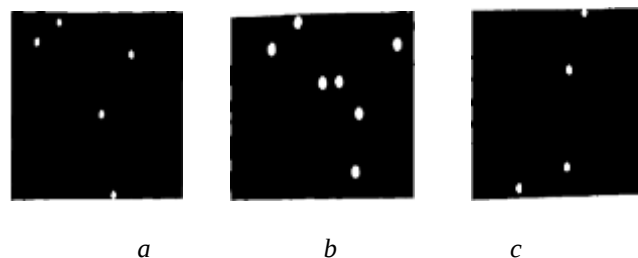


Fig. 4: Photomicrographs of tracks of α -energies (a) 1.0 MeV, (b) 3.0 MeV and (c) 3.5 MeV etched in 2.5N NaOH at 50°C for $h = 4\ \mu\text{m}$, sparked at $V = 900\ \text{volt}$.

4.1 Dependence of bulk-etch rate V_B on both T and C of NaOH Solution using $F. F.$ source

Bulk etch rate, V_B , of LR-115 type II detectors was determined at temperatures of 40, 45, 50, 55, and 60 °C using different etchant concentrations of an aqueous solution of NaOH, viz., 2, 4, 5, 6, 7, and 9 normality. Using Eq.1, V_B values were determined under different conditions of temperatures and concentrations. Figure 5 represents the variation of V_B with C at different values of T (from 40 to 60 °C). It is clear that (see Fig. 5) the dependence of V_B on etchant concentration and temperature follows the Arrhenius equation as described through the following figure.

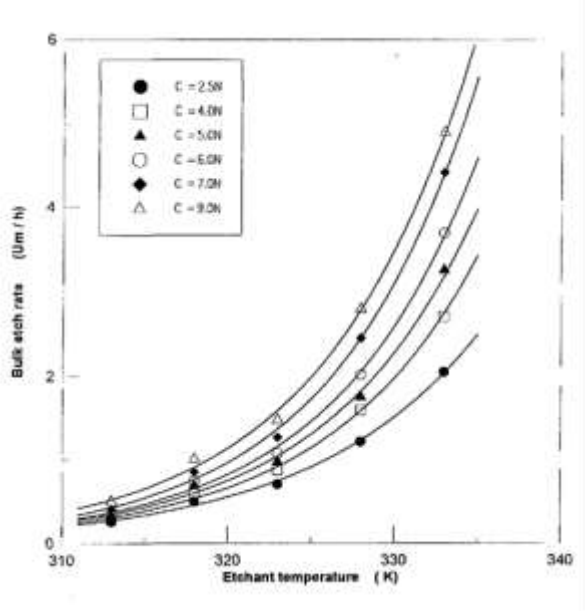


Fig. 5: Relation between bulk etch-rate and etching temperature for LR-115 detectors etched in 2.5, 4, 6, 6.7, and 9N and at different T .

It is described by the Arrhenius equation as follows

$$V_B(C, T) = AC^n \exp(-E_b/kT) \quad (2)$$

where the A , n , and E_b are constant.

The average values of A , n , and E_b are 1.417×10^{14} , 0.5, and 0.93 eV, respectively.

In the case of $C = 2.5N$, the V_B - T dependence is also extracted and written in the form:

$$V_B(2.5N, T) = 1.205 \times 10^{14} \exp(-0.92/kT) \quad (3)$$

Comparing the measured V_B values with those calculated from the last equations, one finds that a good agreement was obtained, and the maximum discrepancy was about 5 percent.

4.2 Registration Characteristics by LR-115 NTDs

Figure 6 shows the mean track diameter as a function of alpha energies using LR-115 plastics etched in 2.5 N NaOH at 45 °C, and for $h=2.04$ and $2.55 \mu m$. Also, at the same depth, h , track diameter has an inverse dependence on energy beyond the Bragg peak.

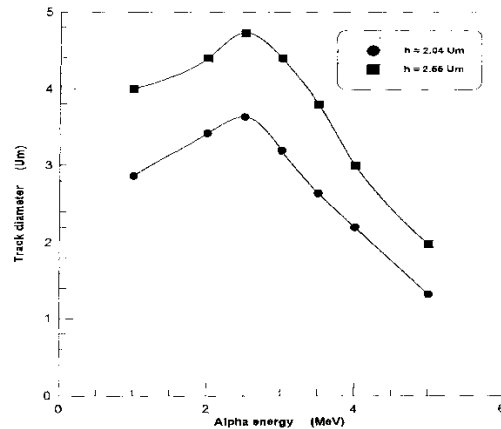


Fig. 6 : Relation between track diameter and alpha energies using LR-115 detectors etched in 2.5N NaOH at 45 °C and at values of $h=2.04$ and $2.55 \mu m$, respectively.

Figure 7 represented a behavior of response function V , with residual range of α -energy for two removal thicknesses h of the detector at $T=45$ °C. The response V with range, which is the same behavior for all of the removal h 's at temperatures of etching, viz, 50, 55, and 60 °C.

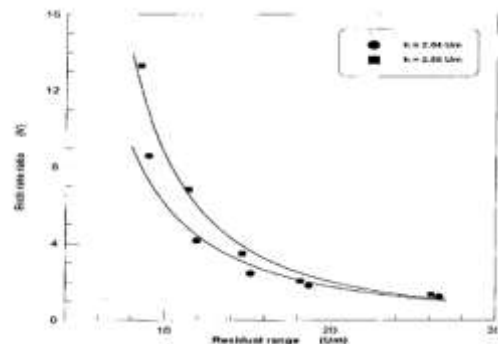


Fig. 7: Variation of etch-rate ratio, V as a function of residual range, R of LR-115 detector etched in 2.5N NaOH for fixed removal h at 45 °C.

Figure 8 represented a behavior of excess rate ratio ($V-1$) which is related to the response function V , with restricted energy loss that is related to the rate of energy loss (dE/dX) range of α -energy for two removal thicknesses h of the detector at $T=45$ °C. It is well known that the excess

etch-rate ratio (V-I) or track response is a strong function of the restricted energy loss (REL) for NTDs. Values of REL were calculated using the computer code of Henke and Benton 1993. This calculation was evaluated at $W_0 = 350$ eV, and the function (V-I) was then studied with (REL)₃₅₀ for LR-115 detectors etched in 2.5N NaOH at a temperature, $T=45^\circ\text{C}$.

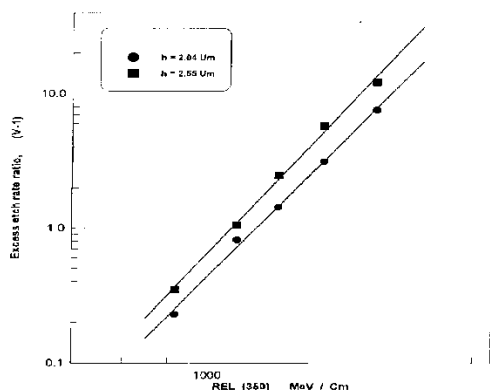


Fig.8: Variation of excess etch rate ratio (V-I) with restricted energy loss REL₃₅₀ of LR-115 detector etched in 2.5 N NaOH at 45°C .

From inspection of figures 15 through 18, the relation between (V-I) and (REL)₃₅₀ obeys a power law, and such a behavior agrees well with some previous work done using CR-39 detectors.

4.3 Radon Level Determinations in Some Egyptian Sites

Radon is a naturally occurring radioactive gas that is formed from the radioactive decay of uranium and found in small amounts in most rocks and soil. It is a member of the noble gas family found in soil and is released during the decay of radium.

4.3.1 Determination of radon calibration factor k

Hazard from radon exposure represents about half of the total hazard from background radiation. During the current study, it is useful to examine the use of the LR-115 detectors in radon-concentration determination and radon-dose evaluation. Track density-radon concentration calibration of LR-115 NTDs is the first and necessary step to be carried out for dose calculation. LR-115 detectors were irradiated in a radon chamber at different radon concentrations for two days. The detectors were put in cups of dimension 3.5cm in radius, and 8cm height, and entered the radon calibration chamber as mentioned above.

The AB-5 monitor was programmed to give the radon

concentration regularly (every hour). At the end of the exposure time, the detectors were taken out, etched in 2.5N NaOH solution, and counted using an optical microscope to determine the corresponding track density. The experiment was repeated using different radon concentration values.

The track density (ρ_o) of LR-115 detectors is proportional to radon concentration (C_o) as given in the equation:

$$\rho_o = KC_o \quad (4)$$

Where K is the calibration factor of the detector.

Figure 9 shows the relation between the track density of LR-115 and radon concentration up to 50000 Bq/m^3 as measured by the AB-5 monitor. From this figure, it is clear that the track density is directly proportional to radon concentration. The proportionality constant leads to $K=0.0599 \text{ track cm}^{-2}\text{d}^{-1}/\text{Bqm}^{-3}$.

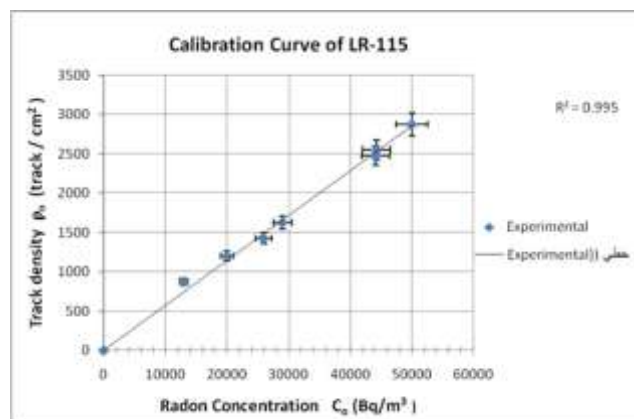


Fig. 9: Radon concentration versus track density for LR-115 NTD etched in 2.5N NaOH for 2 hours at 60°C .

4.3.2 Determination of equilibrium factor, F

The equilibrium factor (F) is defined as the ratio of Potential Alpha Energy Concentration (PAEC) of actual air-radon (also radon progeny) mixture to the PAEC in secular equilibrium with radon.

Thus, F is given as:

$$F = \frac{(\text{PAEC})_{\text{air-radon mixture}}}{(\text{PAEC})_{\text{equilibrium}}} \quad (5)$$

To determine the F factor, the detectors were exposed to radon and/or radon daughters in different configurations [15, 19] as in Figure 9.

The basic arrangements are the closed-mode and open-mode configurations. In closed-mode configuration, the detectors were kept in the lower part of a cup covered with a semi-permeable membrane. The membrane has a role to

let radon only enter the cup and prevent thoron, radon daughters, and dust. In an open-mode configuration, the detector was exposed freely to radon as well as to radon daughters.

The equilibrium factor (F) was determined by NTD based on using can and bare method. In this method, two similar detectors were exposed to radon, one in a can-mode configuration (in can detector) and the other in a bare-mode configuration (bare detector). The equilibrium factor (F) can be found as a function of the track density ratio (D/D_0) between bare (D) and in-can (D_0) detectors, respectively [20-28].

Many authors suggest an equation to determine the equilibrium factor for some other types of NTDs. So in our work, we used an equation determined by Abo-El Maged *et al.* [20-21], where they used a mathematical solution based on solving the equation of F as a function of ventilation rate with (D/D_0). The problem of finding the F factor between radon and its daughters as a function of the track density ratio (D/D_0) is solved graphically for LR-115. This equation depends on the track density and cup dimension. The F equation can be written as:

$$F = \exp \left[a + b \left(\frac{D}{D_0} \right) + c * \ln \left(\frac{D}{D_0} \right) \right]$$

(6)

Where $a=1.862$, $b=-4$, and $c=9.095$ for the dimensions of the cup (3.5cm radius and 8cm height).

Figure 10 (a and b) shows, respectively, the closed and open configurations and cup dimensions.

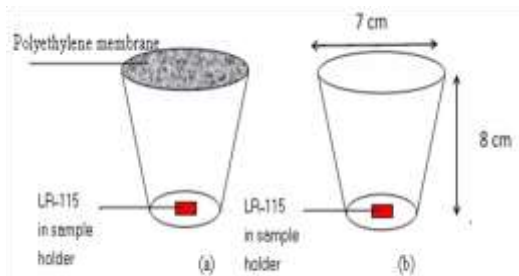


Fig. 10: Schematic diagrams a and b for closed and open cups, respectively, used to determine the equilibrium factor.

A set of ten cups containing LR-115 detectors was used in this study. Five of them were in an open mode configuration; the other five were covered by a polyethylene membrane and used in a closed mode configuration. Each cup contained LR-115 detectors and was exposed to a constant radon concentration using the radon chamber. Exposure time lasted for six days under the condition that every 24 hours, two cups (one open cup and

one closed) were taken away from the chamber.

Each detector was then etched in 2.5 M NaOH solutions at 60°C for a duration of 2 hours, and then the track density ratio (D/D_0) was calculated.

Figure 11 represents the variation of F with the track density ratio (D/D_0), where F shows an increase with increasing the density ratio. An average value of about $F = 0.45$ was obtained.

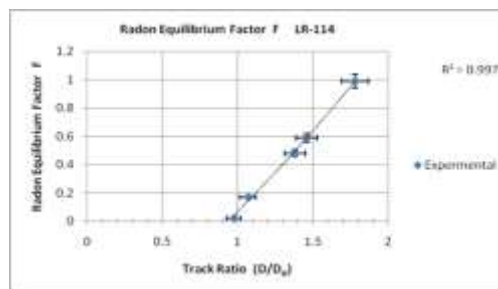


Fig. 11 Track density ratio (D/D_0) versus equilibrium factor (F) for LR-115 NTD etched in 2.5N NaOH for 2 hours at 60 °C.

4.3.3 Radon Concentration Measurements

The use of LR-115 detectors in radon-concentration determination and dose evaluation in some dwellings at Menoufia district, Egypt, was carried out. Eleven houses were chosen in this study, and 11 pairs of cups were used and exposure for eight months.

Figure 11 provides a map from Google that shows the area of study where the Menoufia governorate can be easily seen. House codes are represented by the symbol H (from H1 up to H11).

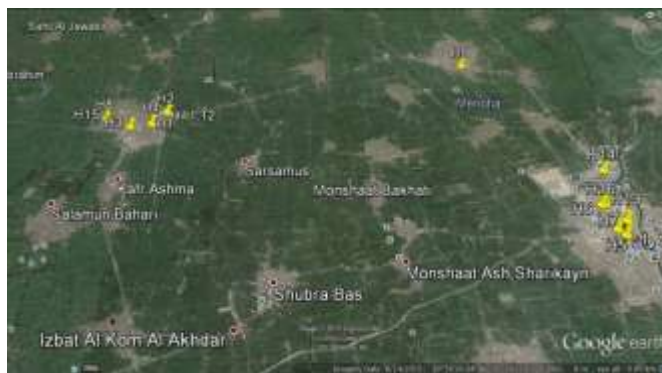


Fig. 12 Google map shows the locations of house zones in Menoufia Governorate, Egypt.

Figure 13 shows a histogram for the measured radon concentration in houses included in this study. Where data

are extracted from both open and closed cans.

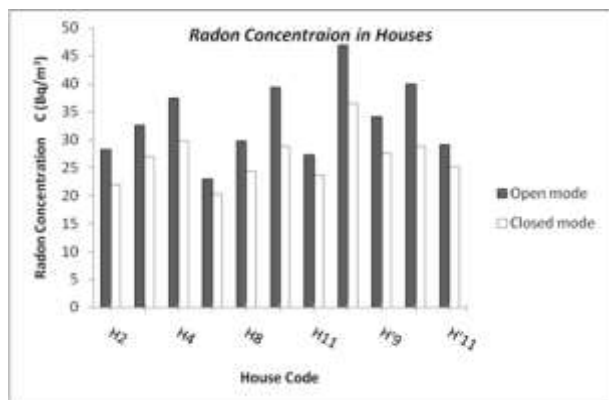


Fig. 13: Histogram for radon concentrations in 11 houses at Menoufia governorate, Egypt.

In case of closed cans, radon concentration ranges from 20.32 to 36.53 Bq/m³, while values from 23.02 to 47.07 Bq/m³ were obtained from open cans.

Table 1 Contains radon concentration values in 11 houses located within the studied area.

Table 1: Radon concentrations in eleven studied houses in both bare and closed can modes.

House code	Radon concentration Bq/m ³	
	Open can mode	Closed can mode
H ₁	28.3	22.01
H ₂	32.62	26.99
H ₃	37.53	29.75
H ₄	23.02	20.32
H ₅	29.85	24.39
H ₆	39.42	28.77
H ₇	27.36	23.61
H ₈	47.07	36.53
H ₉	34.08	25.26
H ₁₀	39.99	28.82
H ₁₁	29.13	25.26

Figure 14 shows that average Radon Concentrations in two kinds of floor zones (ground and first floor). It is clear that the radon concentration on the ground floor is smaller than first floor may be due to the ground floor having more ventilation than first in two modes (open and closed).

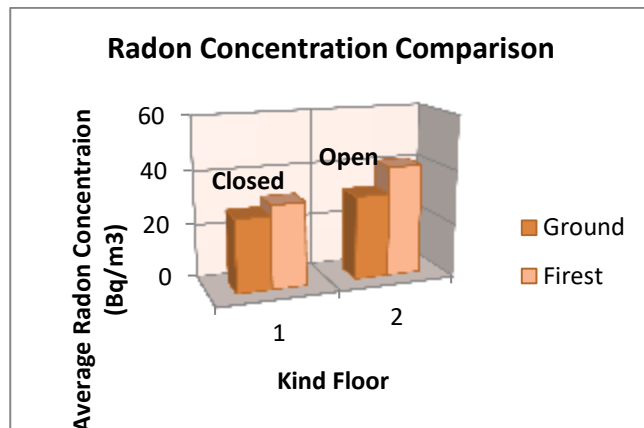


Fig. 14 Histogram Average Radon Concentrations in two kinds of floors of zone houses for Can modes at Menoufia, Egypt.

4.3.4 Radon equivalent dose determination.

The measured values for radon concentration have been used to estimate the radon equivalent dose in sites under investigation. Assuming that a person spends 7000 hours indoors per year, according to ICRP 1993[28-30].

The radon annual equivalent dose can be calculated using the equation:

$$Dose(mSv/y) = 7.85 \times 10^{-6} C_o F T \quad (7)$$

Where C_o is the radon concentration in Bq/m³, T is the occupational time

= 7000 hr per year, and F is the equilibrium factor.

So, using the average concentration calculated in this study, we found that the equivalent dose in open mode measurement is equal to 0.81 mSv/y. Where in closed mode, measurement equals 0.63mSv/y. So from this survey, we found that the average radon annual equivalent dose is 0.72 mSv/y [31-33].

5 Conclusions

The results of this work can be summarized as follows:

1-An interesting empirical formula has been found to fit the V_B data of LR-115 detectors and is written in the form:

$$V_B = AC^n \exp(-E_b/kT), \text{ Where } E_b = 0.93 \pm 0.03$$

2-Pre-track sparking methodology, introduced in this work, enhanced track Shape and contrast, which greatly improved track response evaluation.

3-The etch-pit size dependence on alpha particle energy was evaluated in the detector (LR-115), which presents different energy detection thresholds, using a ²⁴¹Am source. For the case of unknown alpha particle sources

were used such as a ^{226}Ra source and a uranium thin film as which include a higher energy, the data presented for LR-115 plastic NTDs in this work can be applied.

4-The spectrometric characteristics of LR-115 were studied. The detector is suitable to detect a certain range of energies, limited by the wide form of the energy windows for LR-115 NTDs are up to 5.0 MeV.

5-Also, the threshold for the etchable track formation between α -ions should be from the general trend of the sensitivity against the stopping power, derived from the value of 250-400 MeV/cm. Therefore, these detectors can be used in the detection of radon and its progeny monitoring, as well as Uranium and Thorium isotopes concentration, in the geological field's contamination.

6- The track registration properties in LR-115NTDs in this study, for α ions, which occurred by the chemical etching in NaOH solution, have succeeded in spreading window of the detection window as well as improving the registration response or efficiency

7-Radon calibration factor, K, and radon equilibrium factor, F, were determined and found to be $K=0.0599$ track.cm-2d-1/Bq.m-3 and $F = 0.45$, respectively.

8-The resulting radon concentration levels are within the safe criterion according to some recommended reported values.

9-The average radon annual equivalent dose was $< \text{DRAED} \geq 0.72 \text{ mSv/y}$.

Therefore, our results indicate alpha particle tracks in LR-115 NTDs, for the etching conditions employed in this work, can be used in the detection of radon and its progeny naturally, as these detectors are exposed to the environmental conditions of radioecology and radionuclides areas contaminated with different experimental conditions.

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