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Effect of Etching Solution on Nuclear Track Detector CR-39

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Abstract

In this research, we studied the effect of two types of etching solutions and a number of etching factors on the characteristics of the nuclear track detector CR-39. The first type of solution was prepared from dissolving NaOH in (Ethanol+water) by different normalities under different temperatures and compared with the second type of the solution prepared from dissolving NaOH in water(NaOH/water) by different normalities under different temperatures. All detectors were irradiated with alpha particle (α-particle) with 4.7MeV emitted from ^{241}Am source in during (20 Sec). The results showed that the use of the solution NaOH/Ethanol+water reduces the time of appearance particle tracks because of the increase in the bulk etch rate V_B and track etch rate V_T and significantly de-sensitizes the plastic. Activation energy of the bulk etch rate E_B and Track etch rate E_T was also calculated for both solutions , They were 0.78eV ,0.71eV respectively for NaOH/Ethanol+water and 0.74eV , 0.64eV respectively for NaOH/water.

Keywords: CR-39Nuclear track detector, Molecular structure,; Etching characteristics ; NaOH, ;Ethanol; Activation Energy.

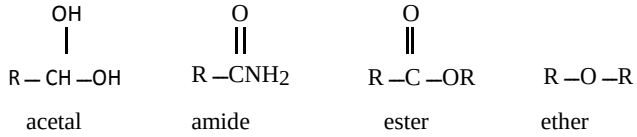
INTRODUCTION

The CR-39 plastic detector belongs to organic detectors; it is a kind of multi-carbon detectors (Polly carbonate). The commercial name of this polymer (CR-39) and the most commonly used name Solid State Nuclear Track Detector (SSNTD) [8,20]. It consists of polymeric material and is prepared from polymerization (diethylene glycol bis(allyl carbonate)) ($C_{12}H_{18}O_7$) it is a hydrocarbon structure [10] Amorphous[4] it is molecular weight 274.27 g/mol and it is density 1.32g.cm^{-3} and refractive index n=1.504, and ionization potential I=1.702.2 eV [9] and the hydrogen proportion is estimated 48.6% , 32.4% C , and 18.9% O the detector is homogenous and isotropic and is visually transparent and high purity and insoluble and infusible except if the chemical bonds are broken [3,21]. And its high sensitivity in detecting charged particles ,the main reason for the high sensitivity of the detector is the presence of carbon bonds in the polymer CR-39 where it is weak and break easily when exposed to radiation [3,12].It is also has a low detection threshold[18].

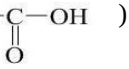
As a result of the features of this detector, it has been used in many applications: It is used to measure radon and thoron concentrations in building materials, indoor building, water, soil, food and agricultural materials [17,20,1,16, 11].

The detectors are also used to distinguish between heavy charged particles such as nuclear fission fragments and light charged particles[7] And in determining the age of geological areas, rocks and archeology[10] And in biological applications as a measure of radiation dose of the radioactive nucleus in the bronchial tissues of the lungs and blood from the inhalation of radioactive nuclei are present in the smoke of the tobacco or the minutes of uranium in the air of mines[14,15] It is also used to determine the alpha concentration in the blood of women who suffer from poor fertility (infertility)[22].

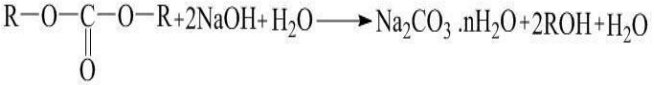
There are a lot of organic polymers that are used as nuclear detectors, and polymers with heterogeneous series are more affected by solutions than other, the most importantly those containing the bonds consisting of groups of acetal , amide, ester and ether and their effect velocity is classified as follows:



acetal amide ester ether

So the fastest polymers dissolve the container on the aggregates of acetal and then come containing the amide groups and then the polymers containing the ester and ether[23]. Polymers can be degraded by energetic agencies (ultra sound, ultraviolet, gamma ray, alpha ray, etc) or by chemical agencies such as (NaOH, KOH, LiOH, etc)[24] . In the case of CR-39 the alkaline etchant attacks the ester group () for the polymer chain.

The chemical reaction can be represented as follows:



The product ROH collects on the form of drops on the surface of the etchant solution, sodium carbonate undergoes hydrolysis in aqueous solution

$$Na_2CO_3 \longrightarrow 2Na^{+} + CO_3^{--}$$

$$2HOH \rightleftharpoons H_3O^{+} + OH^{-}2$$

$$H_3O^{+} + CO_3^{--} \rightleftharpoons H_2O + HCO_3^{--}$$

$$HCO_3^{--} + H_3O^{+} \rightleftharpoons H_2CO_3 + H_2O$$

In the case of etching at temperatures carbon dioxide may be expelled from the undissociated carbonic acid :

$$H_2CO_3 = H_2O + CO_2^{\bullet}$$

Alternatively hydrated salts of sodium carbonate may be deposited. These are seen as a crusty white deposit around the etchant/air interface around the edges of the etch tank. This deposit slowly dissolves when disturbed [3]. The addition of alcohol to the etchant solutions significantly reduces the sensitivity of the plastic[18].

METHODOLOGY

CR-39 detector of 200µm thickness was used from TASKRAK (Track Analysis System Ltd.(TASL), Bristol, UK) and have been prepared in pieces of 1×1 cm^2. The detectors were irradiated in air with normally incident alpha particles emitted from ^{241}Am with energy 4.7MeV.

After irradiation, these detectors were etched in NaOH/ethano+water and NaOH/water at different concentrations and etching temperatures. To avoid the effect of decomposition products on the etching rate of the detector during etching, replace the etching solution NaOH/water every four hours, for NaOH/ethanol+water change every two hours to avoid the change in the concentration of the etching solution. After each etching step, detectors were washed with distilled water and then dried softly to avoid scratching the detector, to begin the process of microscopic observation to measure the track openings diameters with a magnification of 40× attached to digital camera [MDCE-5A] connected to a computer.

The Bulk etching rate (V_B) is an important parameter of the detector, and it can be measured by many variables, There are different methods have been established for the determination of the bulk etch rate, in our study we are used a direct measurement of thickness change of the CR-39 before and after etching.

The following equations have been used to determine V_B, V_T, V, S, η. The bulk etch rate V_B is measured by the change of the thickness of the detector using the relation[9]:

$$V_B = \frac{1}{2}\frac{\Delta h}{\Delta t} \quad (1)$$

Where: h is the removal layer thickness (µm) , t is etching time(hr)

The track etch rate V_T measured by using the relation:

$$V_T = V_B\left[\frac{4V_D^2 + V_D^2}{4V_B^2 - V_D^2}\right] \quad (2)$$

Where: V_D is the rate of diameter growing of the track(µm)

The etch rate ratio V can be determine by using the relation:

$$V = \frac{V_T}{V_B} \quad (3)$$

The etching efficiency η and etching sensitivity S can be determine from the following equations:

$$\eta = 1 - V\frac{1}{\ldots} \quad (4)$$

$$S = V - 1 \quad (5)$$

RESULTS AND DISCUSSION

Effect of the type of etchant solution on the track diameter :

CR-39 detectors irradiated with alpha particles from ^{241}Am source were etched with NaOH/ethanol+water for various concentration and temperatures were used to achieve the optimum processing conditions, CR-39 were also etched in aqueous solution of NaOH for various concentration and temperatures.

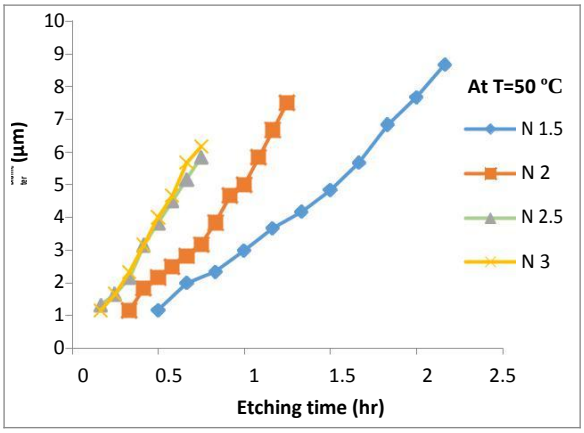


Figure 1. Average alpha-particle track diameter as a function of etching time in CR-39 detectors etched in NaOH/ethanol+water at 50°C with different concentration

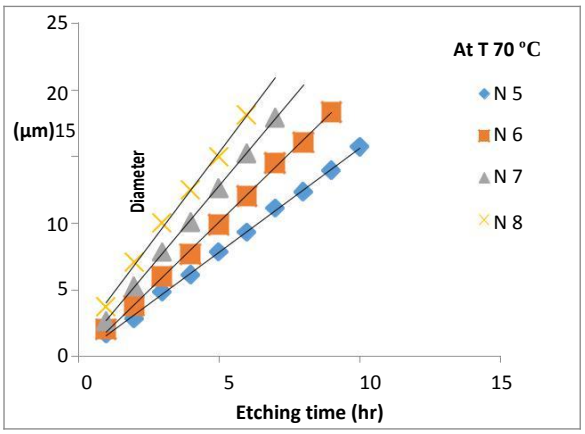


Figure 2. Average alpha-particle track diameter as a function of etching time in CR-39 detectors etched in NaOH/water at 70°C with different concentration

Figs. 1 and 2 show average track diameters of alpha particles as a function of etching time for (1.5,2,2.5,3)N NaOH/ethanol+water solution at temperature 50°C ,and (5,6,7,8)N NaOH/water solution at temperature 70°C. As was expected, track diameter is seen to increase with an increase in the concentration of the etching solution, also is seen track diameter in NaOH/ethanol+water bigger with less etching time compare with NaOH/water.

Table 1

V_D, V_B, V_T, V, η and S values of CR-39 detectors for the listed concentration of NaOH/water solution at 70ºC.

Conc.	V_D	V_B	V_T	V	η	S
5	1.63	1.063	4.12	3.87	0.741	2.87
6	2.16	1.29	7.13	5.48	0.817	4.48
7	2.62	1.48	11.65	7.82	0.872	6.82
8	3.79	2.14	17.58	8.19	0.877	7.19

Table 2

V_D, V_B, V_T, V, η and S values of CR-39 detectors for the listed concentration of NaOH/ethanol+water solution at 50ºC.

Conc	V_D	V_B	V_T	V	η	S
1.5	4.21	3.009	8.82	2.93	0.659	1.93
2	5.63	3.99	11.9	2.98	0.664	1.98
2.5	8.10	5.32	19.96	3.75	0.733	2.75
3	11.6	7.38	32.31	4.37	0.771	3.37

Effect of the type of etchant solution on the bulk etch rate:

The Bulk etch rate (V_B) is considered a fundamental parameter in the formation of the track, where it is an etching and detecting parameter [2]. As previously mentioned, there are different methods to measure bulk etch rate (V_B), we are used a direct measurement of thickness, the thickness of the removed layer was measured for successive etching times and for each normality of solution. From Fig . 3. The removal thickness layer during a three hour time was found to (3.7, 12) µm for solutions NaOH/water and NaOH/ethanol+water respectively. The amount of removal thickness layer for

the solution NaOH/ethanol+water bigger than NaOH/water.

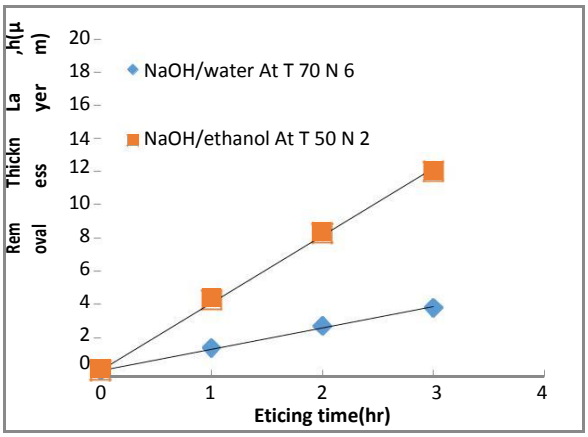
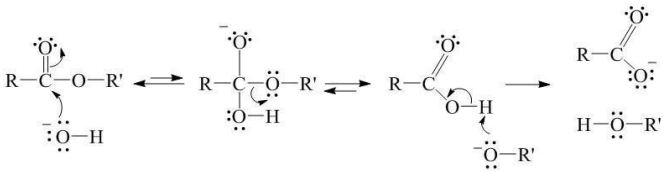
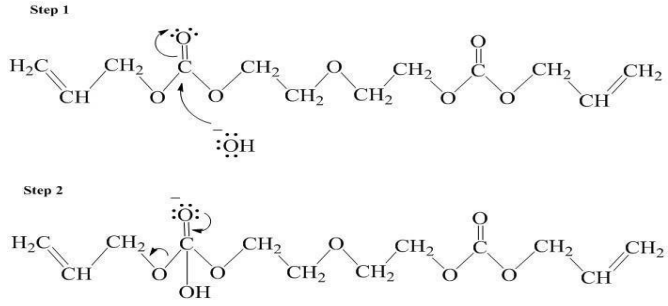


Figure 3. Change the removal thickness layer with etching time in CR-39 detectors etched in NaOH/ethanol+water at 50°C with different concentration

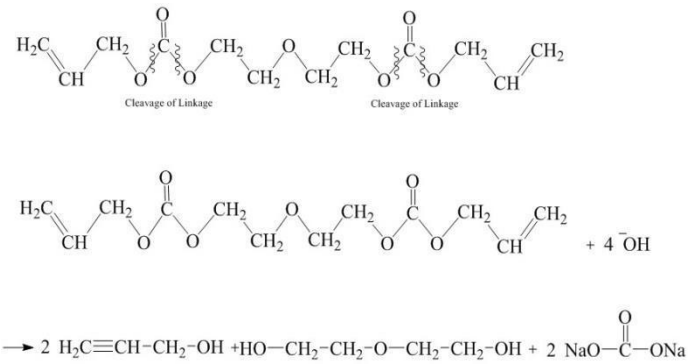
Although both etching solution NaOH/water and NaOH/ethanol+water has the same etching mechanism (mechanical). The treatment of the nuclear track detector CR-39 with basic solutions leads to scission of the carbonate ester bond by hydroxide ion through hydrolysis of ester. The hydroxides ion causes the scission of the ester linkage [13,24]:



Therefore, the mechanism of attacking the carbon atom of the alkoxycabonyl group by hydroxide ion is as follows:



The following reaction shows the position of cleavage by hydroxide ion (OH^-):



The large increase in the bulk etch rate using NaOH/ethanol solution compared to the bulk etch rate using NaOH/water, this is due to the miscibility of ethanol with the organic products of the CR-39 during the etching solution. Sodium carbonate is one of the products during the process of etching CR-39 and the rate formation of sodium carbonate during etching process is greater for the NaOH/ethanol solution than NaOH/water solution and a layer of sodium carbonate precipitate was accumulated on the surface of the CR-39 due to the insolubility of sodium carbonate and saturation in ethanol, which increases the amount of material removed from the surface of the detector significantly. Finally, the resulting is an increase in the bulk etch rate(V_B)[6, 24, 5].

Figs. 4 and 5 show values of V_B increase with the increasing concentration of the solution and note this increase is exponential. It was found that the change in the bulk etch rate with used concentrations for etching solution NaOH/ethanol+water in the Fig. 4 follow the relationship:

(6)

Where:
a=3.53 , b=-1.95 , c=1.076

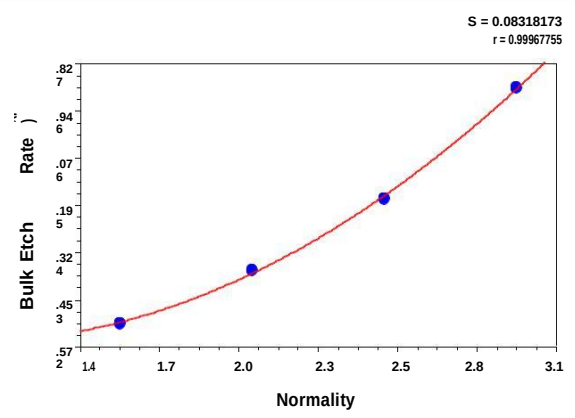


Figure 4. Change the bulk etch rate VB with the concentration of the etching solution of the NaOH/ethanol+water at 70°C

For etching solution NaOH/water in Fig. 5 follow the relationship:

(7)

Where: a=6.96 , b=2.21 , c=-3.62

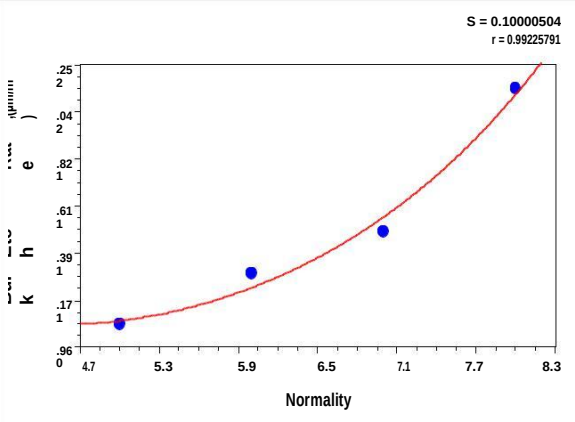


Figure 5. Change the bulk etch rate VB with the concentration of the etching solution of the NaOH/ water at 70°C

Activation Energy:

Activation energy is the energy required in activating the reaction between the detector material and the etching solution. Figs. 6 and 7 shows $\ln(V_B)$ as a function of $1/T(10^3\ K^{-1})$ and $\ln(V_T)$ as a function of $1/T(10^3\ K^{-1})$ respectively for NaOH/water and NaOH/ethanol+water .

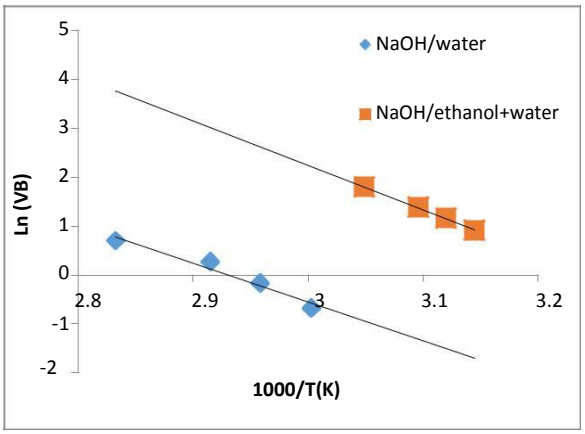


Figure 6. Represents the relationship between $\ln(V_B)$ and 1000/T for NaOH/water and NaOH/ethanol+water

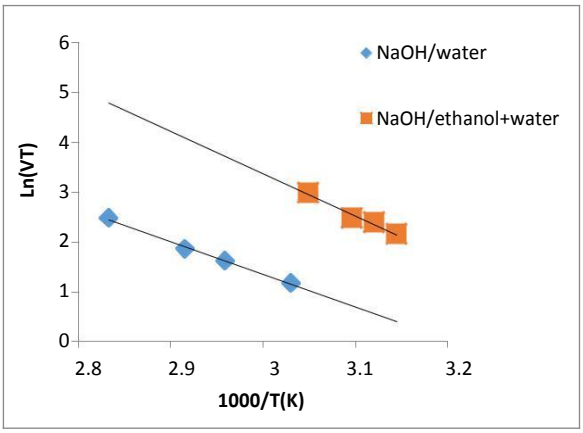


Fig. 7. Represents the relationship between $\ln(V_T)$ and 1000/T for NaOH/water and NaOH/ethanol+water

Table 3.

Values of bulk activation energy and track activation energy for different etchants

Etchant	E_B(eV)	E_T(eV)
NaOH/water	0.74	0.64
NaOH/ethanol+water	0.78	0.71

The average values of E_B and E_T was determined from the slopes of the straight lines. We found that the value of $E_B > E_T$ for two solutions are due to the fact that when the charged particles fall on the detector material, damage is created in the detector material, which leads to the scission of the polymer chain, the broken of the bonds, the formation of the free radicals and low molecular weight of the detector CR-39[9]. Because of the broken molecular chains and the formation of free radicals in the damaged regions, there is a smaller energy barrier to activate the interaction between hydroxide ions and molecular chains, so that the damaged regions along the particle path are attacked at a faster rate than the detector surfaces by the solution. This means that the E_T is less than the E_B. also it can be seen that E_B and E_T are higher for NaOH/ethanol+water than for NaOH/water.

CONCLUSIONS :

We have shown that the etching solution NaOH/ethanol+water reduces the time of appearance of the track due to the increase in the bulk etch rate V_B and track etch rate V_T compared to the NaOH/water solution, The addition of alcohol to the etchant solutions significantly reduces the sensitivity of the plastic. E_T has found to be lower than the bulk activation energy E_B, for CR-39 detector in two solutions. It also can be seen that E_B and E_T are higher for NaOH/ethanol+water than for NaOH/water.

REFERENCES:

[1] Abumurad, K. M., Kullab, M. K., Al-Bataina, B. N., Ismaial, A. M., & Lehlooh, A. D. (1994). Estimation of radon concentrations inside houses in some Jordanian regions. Mu'tah Lil-Buhooth Wa Al-Dirasat, 9(5), 9-21.

[2] Al-O'bey, A.A.I., MSc. Thesis, College of Education , University of Mosul, Iraq (2000).

[3] Amin, S. A., (1981) . " Etching Properties of the CR-39 Polymeric Nuclear Track Detector " , Ph.D. Thesis, University of Bristol, UK.

[4] Cassou, R. M., & Benton, E. V. (1978). Properties and applications of CR-39 polymeric nuclear track detector. Nuclear Track Detection, 2(3), 173-179.

[5] Chan, K. F,; Ng, F. M. F.; Nikezic, D. and Yu, K. N.)7002(, " Bulk and Track Etch Properties of CR-39 SSNTD Etched in NaOH/Ethanol " , Nucl. Instr. and Meth. in Physics Research, B 263, pp. 284-289.

[6] Chan, K. F., Siu, S. Y. M., McClella, K. E., Tse, A. K. W., Lau, B. M. F., Nikezic, D., ... & Yu, K. N. (2006). Alpha-particle radiobiological experiments using thin CR-39 detectors. Radiation protection dosimetry, 122(1-4), 160-162.

[7] Chatry C.; Serbat A.; Laugier J.; Gasiot J. and Vanderschneren J. (1990), "Energy and Angle Response of CR-39 to Protons", In Proc. 15th Int. Conf. on Particle Tracks in Solids, Hamburg, Germany.

[8] Dodiuk, H., & Goodman, S. H. (Eds.). (2013). Handbook of thermoset plastics. William Andrew.

[9] Durrani , S.A. , Oxford ; New York Pergamon Press, 1987. xiii, 304 p. : ill. ; 24 cm. International series in natural philosophy ; v. 111.

[10] Fleischer R.L., Price P.B. and Walker R.M.(1975).Nuclear Tracks in Solids: Principles and Application . University of California Press.

[11] Font, L. L., Baixeras, C., Jönsson, G., Enge, W., & Ghose, R. (1999). Application of a radon model to explain indoor radon levels in a Swedish house. Radiation measurements, 31(1), 359-362.

[12] Fujii, M., & Yokota, R. (1986). Thermosetting resins for nuclear track detection. International Journal of Radiation Applications and Instrumentation. Part D. Nuclear Tracks and Radiation Measurements, 12(1-6), 55-58.

[13] Gruhn T.A.,Li W.K., Benton E.V., Cassou R.M. and Johnson G.S. (1980) . " Etching Mechansim and Behaviour of Polycarbonates In Hydroxide Solution: Lexan and CR-39 " .In: Proc. 10th Int. Conf. Solid State Nuclear Track Detectors, Loyn, and Supp1.2, Nucl. Track . Pergaman, Oxford.291.

[14] Henshaw D.L., Aheng E., Akeitch P.A. and Rannpie P.H. (1994), "Spatial distribution of naturally accruing 210Po and 222Rn in children teeth", Inst. J.Radial.Biol.,66(6):815-826.

[15] Henshaw D.L., Fews A.P. and Webester D.J. (1980), "The microdistribution of alpha active nuclei in bronchial tissue by auto radiography using CR-39". In: Proc. 10th Int. Conf. SSNTD's", Lyon and Suppl. 2 Nucl. Tracks., (eds. H.Fracncois et al.), Pergramon, Oxford, 649–654.

[16] Kandaiya, S., Al-Najjar, S. A. R., & Piesch, E. (1988). Study of α-energy discrimination in CR-39 track etch detectors for use as a radon/thoron dosemeter. International Journal of Radiation Applications and Instrumentation. Part D. Nuclear Tracks and Radiation Measurements, 15(1-4), 595-598.

[17] Khan, A. J., Sharma, K. C., Varshney, A. K., Prasad, R., & Tyagi, R. K. (1988). Radon estimation in some Indian tobacco, tea and tooth powder using CR-39 nuclear track detector. Radiation and environmental biophysics, 27(3), 233-237.

[18] Kodaira, S., Yasuda, N., Kawashima, H., Kurano, M., Hasebe, N., Doke, T., ... & Ogura, K. (2009). Control of the detection threshold of CR-39 PNTD for measuring ultra-heavy nuclei in galactic cosmic rays. Radiation Measurements, 44(9), 861-864.

[19] Nikezic, D., & Yu, K. N. (2004). Formation and growth of tracks in nuclear track materials. Materials Science and Engineering: R: Reports, 46(3), 51-123.

[20] Nikezić, D., Baixeras, C., & Kostić, D. (1996). Sensitivity determination and optimisation of a cylindrical diffusion chamber, for radon measurements, with a CR39 detector. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 373(2), 290-298.

[21] Pereira, L. A. S., Sáenz, C. A. T., Constantino, C. J. L., Curvo, E. A. C., Dias, A. N. C., Soares, C. J., & Guedes, S. (2013). Micro-Raman spectroscopic characterization of a CR-39 detector. Applied spectroscopy, 67(4), 404-408.

[22] Salih, N. F., Jaafara, M. S., Al-Hamzawi, A. A., & Aswood, M. S(2013). The Effects of Alpha Emitters on Powder Blood for Women's Infertility in Kurdistan–Iraq. International Journal of Scientific and Research Publications, 57.

[23] Sharma, K.K. and Sharma, L.K. (1980). A textbook of physical Chemistry, Vikas Publishing House PVT Ltd.

[24] Tse, K. C. C., Ng, F. M. F., & Yu, K. N. (2006). Photo-degradation of PADC by UV radiation at various wavelengths. Polymer Degradation and Stability, 91(10), 2380-2388.