

Calculating CR-39 Response to Radon in Water Using Monte Carlo Simulation

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Abstract

Introduction

CR-39 detectors are widely used for Radon and progeny measurement in the air. In this paper, using the Monte Carlo simulation, the possibility of using the CR-39 for direct measurement of Radon and progeny in water is investigated.

Materials and Methods

Assuming the random position and angle of alpha particle emitted by Radon and progeny, alpha energy and angular spectrum that arrive at CR-39, the calibration factor, and the suitable depth of chemical etching of CR-39 in air and water was calculated. In this simulation, a range of data were obtained from SRIM2008 software.

Results

Calibration factor of CR-39 in water is calculated as $6.6 \text{ (kBq.d/m}^3\text{)}/\text{(track/cm}^2\text{)}$ that is corresponding with EPA standard level of Radon concentration in water ($10\text{-}11 \text{ kBq/m}^3$). With replacing the skin instead of CR-39, the volume affected by Radon and progeny was determined to be 2.51 mm^3 for one m^2 of skin area. The annual dose conversion factor for Radon and progeny was calculated to be between $8.8\text{-}58.8 \text{ nSv}/\text{(Bq.h/m}^3\text{)}$.

Conclusion

Using the CR-39 for Radon measurement in water can be beneficial. The annual dose conversion factor for Radon and progeny was calculated to be between $8.8\text{-}58.8 \text{ nSv}/\text{(Bq.h/m}^3\text{)}$.

Keywords: Alpha Particle, Angular Spectrum, Annual Dose, Calibration Factor, CR-39, Energy Spectrum, Radon, Suit Etching Depth

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1. Introduction

Solid State Nuclear Track Detectors (SSNTDs) have many applications in all branches of science and technology. They are considered as the best options to measure the outdoor Radon concentration [1]. One of the most important items to measure Radon concentration is the study of the energy and angle of Radon and progeny alpha in media that arrive at the CR-39 (Columbia Resin-39) detector and real calibration coefficient of CR-39. The CR-39 has been widely used for Radon and progeny measurement in air [1]. The CR-39 suitable depth of chemical etching is the depth that varying track density has any particles fluctuates and rapid loss. Previously, by use stopping power data and fitting an equation with 16 coefficients to the data, the alpha particles energy in a distance from the emitted point was presented by Nikezic et al [2]. The main disadvantage of the Nikezic method is running energy calculating code for every particle in the main Monte Carlo program that increases the running time of simulation. For reducing the running time of the Monte Carlo simulation program, the alpha particles' energy was introduced as a function of distance traveling in water and CR-39. Previously for simulation of alpha particles, the random number with Gaussian distribution was used [3]. The energy and angular spectrums of Radon and progeny alphas can be simulated because they arrive at the detector with different angles and energy. The OriginPro8 (OriginLab Corp., Northampton, Ma, USA) is used to draw, fit, and analyze the data. To simulate alpha track with different energies which are emitted from Radon and progeny around the CR-39 detector in different angles, the stopping power and range data in every media were needed. The SRIM2008 software was used for extracting this data. This software is the most suitable code in comparison with FLUKA code [4, 5]. No similarity was observed between simulated and measured alpha spectrum of Radon and progeny with energy less than 5 MeV in lost simulations [6]. For investigation of the

accuracy of the method, this problem was resolved. Previously, theoretical and experimental CR-39 calibration coefficients for Radon and progeny in air were calculated [7,8]. The CR-39 calibration coefficient of Radon and progeny in water for Radon measurement in water was calculated. According to the EPA standards, the limiting levels for Radon concentration in air and water are 150 Bq/m^3 and 10 kBq/m^3 , respectively [9]. The goal of CR-39 calibration coefficient measurement in water is to investigate probability of Radon measurement in water according to the EPA standards. Moreover, the annual dose conversion factor of skin in water due to Radon and progeny alpha activity in water was calculated. In this work, using the Monte Carlo method, the probability of using the CR-39 in water and introducing the suitable depth of chemical etching was investigated.

2. Materials and Methods

2.1. Calculation of Alpha Energy versus Distance

If Radon or progeny in the distance "d" from CR-39 emits an alpha particle with energy E_1 , then it will hit CR-39 with E_2 energy. The distance d must be smaller than the alpha particles range in the media. The d distance is equal to the difference between alpha range R_1 with energy E_1 and alpha range R_2 with energy E_2 as presented in Equation (1).

$$d=R_1-R_2=R_{\max}-R \quad (1)$$

If E_1 is the initial energy then R_1 will be the maximum range. To determine d, first, alpha particles range data from 0-10 MeV is extracted by using SRIM2008. Then, using Originpro8 software, a nonlinear equation is fitted to the range data. To detect Radon with chemical etching technique, alpha particles of very low energy are not important. Thus, the equation of the alpha range from 0.5 to 10 MeV (without straggling) is as Equation (2).

$$R=aE+bE^2+cE^3 \quad (2)$$

By substituting R in Equation (1) in Equation 2, distance traveling versus alpha energy is obtained as Equation 3.

$$d=R_{\max}-(aE+bE^2+cE^3) \quad (3)$$

Equation 3 expresses the distance travelled by alpha particles as a function of energy. To calculate alpha energy in "d" distance from the emission point, the inverse of Equation (3) should be obtained. This can be done in OriginPro8 software by exchanging the travelled distance and energy columns data and fitting a new equation as Equation (4).

$$E=E_0-a'x-b'x^2-c'x^3 \quad (4)$$

In Equation 4, E_0 is alpha initial energy and a' , b' , c' coefficients for Radon and progeny alpha's in CR-39, water, and air are obtained and are shown in Table 1.

Table 1. Alpha energy of Radon and progeny at distance d from emitted point in CR-39, water, and air.

Radon and Progeny	E (MeV), d (μm) CR-39
^{222}Rn	$E=5.49-0.10148d - 0.000589d^2 - 0.00004206d^3$
^{218}Po	$E=6-0.10054d + 0.000004332d^2 - 0.000040172d^3$
^{214}Po	$E=7.69-0.09899d + 0.000798d^2 - 0.00002608d^3$
Air E(MeV) , d(mm)	
^{222}Rn	$E=5.49-0.081d - 0.0009333d^2 - 0.000007997d^3$
^{218}Po	$E=6-0.07312d- 0.00075 d^2 - 0.00000821d^3$
^{214}Po	$E=7.69-0.05612d - 0.000024d^2 - 0.0000079d^3$
Water E(MeV) , d(μm)	
^{222}Rn	$E=5.49-0.0792d - 0.000915d^2 - 0.00000764d^3$
^{218}Po	$E=6-0.07221d - 0.000734d^2 - 0.000007885d^3$
^{214}Po	$E=7.69- 0.065d - 0.0000054d^2 - 0.0000052d^3$

2.2. Input Data and Practical Notes

To study the depth of alpha particles due to Radon progeny in CR-39, the equilibrium factor must be specified. Equilibrium factor shows that the percentage of cumulative activity of Radon and progeny due to Radon progeny is defined as ratio of Radon and progeny activity to Radon progeny activity. Equilibrium factors for ^{218}Po and ^{214}Po are considered to be 0.723 and 0.217, respectively [10]. Using SRIM 2008 software, alpha range and stopping power data was easily extracted and the data can be used reliably. The energy of alpha particles for Radon and progeny are reported as follows [11]: ^{222}Rn (5.49 MeV), ^{214}Po (7.69 MeV), and ^{218}Po (6.0 MeV). Radon is a noble gas and is distributed homogeneously in the air, while ^{218}Po and

^{214}Po are solid and are seated gradually on CR-39 surface. Because of the slow movement of Radon progeny for sitting on a surface, their distribution can be assumed homogenous in environments. In total, almost 100% ^{214}Po and 20% of ^{218}Po sit on surfaces. Due to the short life time of Radon progeny, a lot of them will emit their own alpha particles before sitting on CR-39 surface.

2.3. Monte Carlo Simulations

Monte Carlo simulation of alpha particles around the CR-39 and inside it is described as a flowchart in Figure 1.

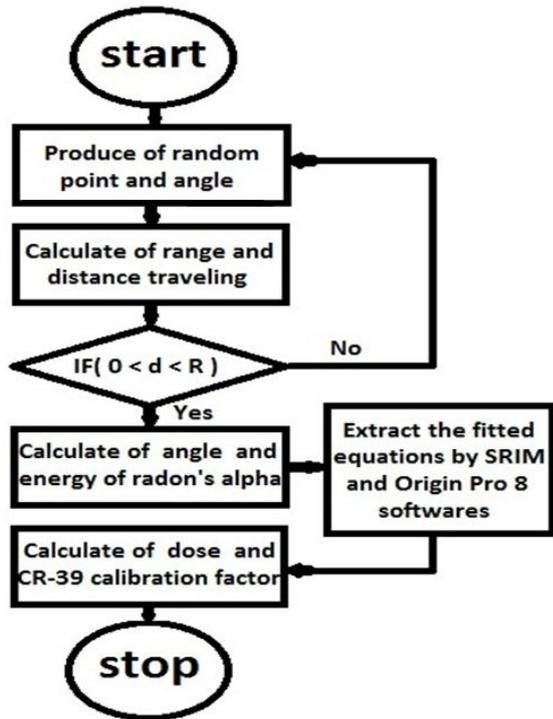


Figure 1. The flowchart of Monte Carlo code for simulation of alpha particles around the CR-39 detector and inside it.

The CR-39 with surface area 1×1 cm is put on the x-y plane. A surface area with dimension $(1+2R) \times (1+2R)$, vertical on y-axis, at distance R (range) from CR-39 is considered. The useful volume around the CR-39 will be as Equation 5.

$$V = R \times (1+2R) \times (1+2R) \quad (5)$$

Outside this space, the travelling distances will be greater than alpha range and alpha particles will not arrive to the CR-39. A random point P1 (Y_1, X_1, Z_1) is created in the active space. From this point, alpha particle is emitted in a direction with the random angles θ and ϕ . If the traveled distance by the alpha particle (or d) is smaller than its range in environment, then it will arrive at detector plane in point p2 (Z_2, X_2) with angle θ and energy E less than its initial energy E_0 . Otherwise it will not arrive to the detector plane. The numbers of alpha particles per day in active space are as Equation 6.

$$N = 0.5 \times 86400 \times C \times F \times R \times (1+2R) \times (1+2R) \quad (6)$$

In Equation 6, the 0.5 shows that half of the alpha particles are emitted forward to CR-39 and the 0.5 of those are emitted backward. The 86400 is for one day Radon activity. C is Radon concentration, F is equilibrium factor, and active space around CR-39 is calculated using $R \times (1+2R) \times (1+2R)$ and R is the alpha particle range of ^{222}Rn or ^{218}Po or ^{214}Po . With plotting the range data using SRIM 2008 software for CR-39 with the chemical formula C10H18O7 and fitting an equation to them, alpha particle range in CR-39 is calculated as Equation 7.

$$R = 3.34773E+0.34937E2+0.02142E3 \quad (7)$$

Monte Carlo simulation of alpha particles in CR-39 using Equation 7 is available. The reflected alpha particles from CR-39 are ignorable [12]. The alpha particles have low curvature path [12].

3. Results

3.1. Radon and Progeny Alpha Spectrum in Air

According to the Monte Carlo code, the alpha energy spectrum that reaches the CR-39 detector in dimensions of 1×1 cm due to 1 kBq/m³ daily activity Radon and progeny is as Figure 2. This spectrum is according to the obtained results [6].

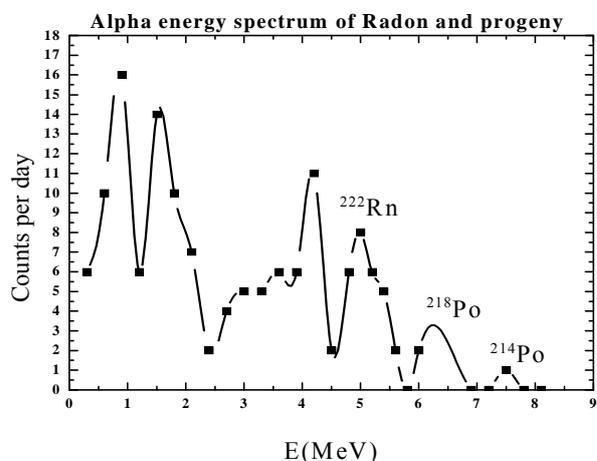


Figure 2. The energy spectrum of Radon and progeny alpha particle in the air.

3.2. Monte Carlo Simulation Result of Angular Distribution and Penetration Region of Radon and Progeny Alpha's in CR-39 Detector

Information about the incident angle of Radon and progeny is shown in Table 2. This table shows that the most of the ^{218}Po alpha particles incident with angles less than 50° and about 35° . The ^{218}Po alpha incident angle remains constant between 40° and 50° . However, between 60° and 80° is much less than the previous period. The lowest frequency is observed in 50° . The most intensity of Radon alpha is in the incident angle of 100° . The frequency decreases almost uniformly with increasing of incident angle (Table 2). The alpha particle between 40° and 80° was related to ^{222}Rn alpha's with 4.5 MeV energy. More information from the incident angle and the penetration of alpha particles in CR-39 is given in Table 2. Table 2 shows that the ^{214}Po alpha penetration in CR-39 is between 7 to 51 μm . The most intensity of ^{214}Po is related to

the 10° - 40° . According to Table 2, the observed track in the depth of 7-12 μm and with 10° - 40° , is relevant to ^{214}Po alpha's. The ^{218}Po alpha particles penetrate in 0° - 5° and 15° - 55° with 25-52 μm penetration. Moreover, ^{222}Rn alpha's is seen in 0° - 20° and 0° - 60° angles. The highest frequency of ^{218}Po alpha's is related to 0° - 5° . Therefore, with regard to Table 2, the observed tracks in the depth of 32-52 μm with 0° - 5° are related to ^{218}Po alpha's. The most frequency of ^{222}Rn alpha's is in 30° - 50° . The observed tracks in the depth of 42-49 μm with 30° - 50° are related to ^{222}Rn alpha's. The tracks with 35° - 56° and the depth of 49-56 μm are certainly related to ^{214}Po . In 6.4-7.4 MeV, energy interval related to this depth is not due to the ^{218}Po alpha activity (or 6 MeV) and ^{222}Rn alpha activity (or 5.49 MeV). The simulation results show that the maximum depth penetration of Radon and progeny in CR-39 is about 52 μm (Figure 3).

Table 2. Incident angle and depth penetration of Radon and progeny alpha's in CR-39.

222Rn alpha's				
Angle (degree)	2-18	30-50	20-30	75-80
Depth(μm)	25-30	42-49	30-38	28-42
214Po Alpha's				
Angle (degree)	0-60	25-55	30-50	70-75
Depth(μm)	7-12	18-25	49-51	24-44
alpha's 218 Po				
Angle (degree)	30-60	3-30	30-50	50-70
Depth(μm)	25-35	32-52	51-55	14-45
Radon and progeny alpha's				
Angle (degree)	0-80	3-80	5 - 80	35-65
Depth (μm)	0-28	28-42	42-49	49-56
Energy(MeV)	0-3.7	3.7-5.5	5.5-6.4	6.4-7.47

3.3. Introducing the Suit Depth for Chemical Etching

The variations of alpha tracks versus depth in CR-39 are shown in Figure 3. The alpha tracks until 25 μm have little variation. For depth

larger than 25 μm , the change in alpha tracks is very high. Therefore, the suitable depth for chemical etching is 25 μm . In this depth, according to Table 2, all Radon and progeny alpha's arrive in all incident angle.

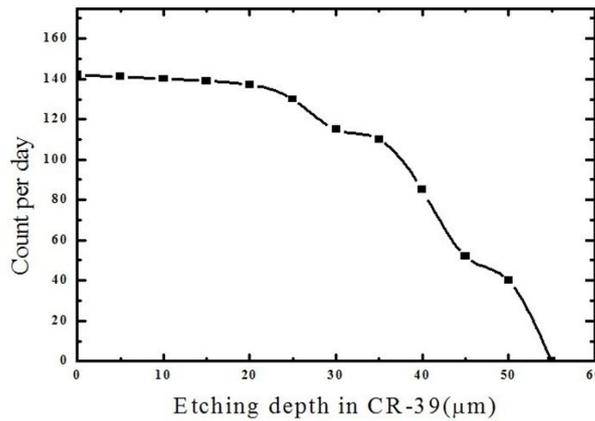


Figure 3. The variation of Radon and progeny alpha tracks versus depth in CR-39.

Table 3. The CR-39 sensitivity and calibration coefficient of Radon and progeny alpha's in air and water.

Media	Sensitivity Coefficient (track/cm ²)/ (Bq.d/m ³)	Calibration Coefficient (kBq.d/m ³)/ (track/cm ²)
CR-39 in Air	0.142	0.0077
CR-39 in Water	0.00015	6.67

3.4. Alpha Calibration Coefficient of CR-39 Detector in Air and Water

Sensitivity coefficient (S) is equal to the latent tracks in unit area per second in the CR-39 detector due to unit concentration of Radon and progeny as Equation (8) [13].

$$S = (N/A) / (t \times C) \tag{8}$$

In Equation (8), N is the tracks in CR-39 detector with surface A (m²) at time t (s) due to Radon and progeny activity with concentration C (Bq/m³). According to Equation (8), the sensitivity coefficient of alpha is non-dependent on Radon concentration. By measuring S, t, A and N, the Radon concentration can be obtained using Equation (8). In this paper, by using Equation (8) and Monte Carlo simulation result, the CR-39 sensitivity to Radon and progeny alpha particles in air and water are calculated (Table 3). Radon concentration in water is considered to be 10 kBq/m³.

In this simulation, the N=14200 tracks in unit area (A=1 m²) of CR-39 per day (t=1 d) due to (C=100 kBq/m³) Radon concentration in air were created. Therefore, the sensitivity coefficient is as the follows: $S = (N/A) / (t.C)$
 $= (14200 \text{ track/cm}^2) / (1 \text{ d} \times 100 \times 1000 \text{ Bq/m}^3)$
 $= 0.142 \text{ (track/cm}^2) / (\text{Bq.d/m}^3)$

The calibration coefficient is reciprocal of sensitivity coefficient. The CR-39 sensitivity coefficient ((track/cm²)/ (Bq.d/m³)) in air is obtained as 0.142 which is less than the maximum sensitivity or 0.332 [12] and is greater than the experimental result or 0.168 [14]. One of the applications of sensitivity coefficient (Table 3) is the Radon concentration measurement. For Radon concentration measurement, the CR-39 detector is placed in the environment and tracks are counted after a time interval. According to Equation (8) and sensitivity coefficients (Table 2), Radon concentration is obtained. Then all steps for determination of calibration coefficient in air were repeated for other media. Putting the CR-39 in water will increase the CR-39 sensitivity to alpha particle [15]. The CR-39 is suitable for calculating dose distribution in water [16].

3.5. The Calibration Coefficient of CR-39 Detector

If chemical etching continues to depth of 25 μm, the track density will reduce from 140 to 130 track/cm². Dividing Radon concentration (1000 Bq/m³) to tracks density in this depth (130 track/cm²) shows that every track in CR-

39 is related to 7.7 Bq/m^3 Radon concentration in the air. The calibration coefficient, or Radon concentration related to one track in a day in a unit area of CR-39, is the inverse of the sensitivity coefficient. The calibration coefficients of CR-39 for Radon and progeny alpha's in water and air is shown in Table 3. The calibration coefficient of CR-39 changed a little from 0 to $25 \mu\text{m}$ depth.

3.6. Annual Skin Dose due to Radon and Progeny in Water

Skin dose due to Radon and progeny in water was dependent on Radon concentration and Radon progeny suspension in water. The skin dose origin is alpha due to Radon progeny that arrives on human skin in water [17]. There is uncertainty in skin dose calculation because of differences in human anatomy and skin thickness [17]. Maybe skin is covered by clothes or hair. TCRP assumed that the sensitive region of skin is also sensitive to UV radiation. However, cancer investigation of survival in nuclear bomb explosions does not show this. An effective dose equivalent of 25 mSv/year , may be taken as the reference level, is equivalent to an annual average Radon concentration of 200 Bq/m^3 . Radon is heavier than air and is soluble in water. Therefore, a sharp increase of the dose rate is clearly observed in the beginning of raining. Maybe there are cancer risks from Radon in drinking water because water containing Radon remains in the stomach for about one hour and then enters to the blood cycle. Radon dose from inhalation of Radon is about 1.1 mSv/year and Radon dose due to drinking water is about 0.002 mSv/year . An adult person drinks about 600 liters of water per year. The maximum absorbed dose in the stomach, in 1000 Bq/m^3 Radon concentration, is about 50 mSv/year for

Radon and about 1.15 mSv/year for Radon progeny. Radon solubility in fat is high. Therefore, for the overweight human, the absorbed dose is greater than others. In dose calculation models, a steady distribution of radioactive source on sensitive skin cells (on the surface and inside) was assumed. The major problem is calculation of the goal cells depth. Radon dose in 100 Bq/m^3 Radon concentrations is reported to be about 1.1 mSv/year [18]. Radon dose conversion coefficient for conical and ellipsoidal bronchial cells is reported to be about 2 mSv/wlm and for spherical bronchial cells is reported to be about 10 mSv/wlm [19]. The epidemiologic and dosimetric dose conversion coefficients are reported to be about 4 and 15 mSv/wlm , respectively [20]. Radon dose conversion coefficients for conical, ellipsoidal, and spherical skin cells are reported to be about 12.92, 13.13, and 15.05 mSv/wlm , respectively. In the ICRP66 report, skin was divided to many layers. After absorbed energy calculation in these layers, dose conversion coefficient was reported to be about 15 mSv/wlm . In another report and for $42.2 \pm 3.8 \text{ Bq/m}^3$ Radon concentration, dose conversion coefficient and dose conversion factor were reported to be about $132 \pm 12 \text{ mSv/wlm}$ and $1 \mu\text{Sv}/(\text{decay/cm}^2)$, respectively. For dose calculation, the depth of skin layers must be larger than $50 \mu\text{m}$ because there aren't any sensitive cells in depth less than this. In the time of rain, Radon dose is due to ^{214}Po [21]. The effective annual Radon dose and dose coefficient factor are reported as 1.16 mSv/year and $9 \text{ nSv}/(\text{Bq.h/m}^3)$, respectively [22]. The Radon dose conversion factor of drinking water for different age groups is shown in Table 4 [23].

Table 4. Radon dose conversion factor of drinking water for different age groups.

Dose(Sv)=C×V×DCF				
Age group	Adult	Children	kids	Any age group
DCF(Sv/Bq)	10^{-8}	2×10^{-8}	7×10^{-8} [23]	0.35×10^{-8} [24]

In Table 4, C is Radon concentration (Bq/m^3), V is water volume (m^3), and DCF is dose conversion factor (Sv/Bq). The drinking water volume for different age groups per day is reported to be between 0.51 and 13.1 liter/day [24]. Therefore, Radon dose due to drinking water in 1 Bq/l is about 3.65 mSv/annual . In this paper, by replacing skin instead of CR-39, the damaged volume of 1 m^2 skin surface due to Radon and progeny annual activity and the Radon annual dose conversion factor of skin in water are 2.51 mm^3 and 28 $\text{nSv}/(\text{Bq}\cdot\text{h}/\text{m}^3)$, respectively. By assuming the maximum range (^{214}Po alpha particle), the maximum damaged volume is about 6.4 mm^3 and minimum of the Radon annual dose conversion factor of skin in water is 8.8 $\text{nSv}/(\text{Bq}\cdot\text{h}/\text{m}^3)$. By dividing the 1 m^2 skin surface to track number and adding every track dose, the Radon annual dose conversion factor of skin in water for ^{218}Po and ^{214}Po alphas are 13.3 and 8.8 $\text{nSv}/(\text{Bq}\cdot\text{h}/\text{m}^3)$, respectively.

4. Discussion

In this paper, the energy and angular spectrum of Radon and progeny alphas on CP-39 was simulated. The sensitivity and calibration coefficient of CP-39, the Radon and progeny alpha energy versus distance traveling in air and water were obtained. The suit chemical etching depth of CR-39 for Radon and progeny alpha's was simulated to be about 25 μm . Every track in 25 μm CR-39 depth is related to 7.7 Bq/m^3 Radon concentration in the air. Calibration coefficient of CR-39 in water is 6.6 ($\text{kBq}\cdot\text{d}/\text{m}^3$)/(track/cm^2) that is less than EPA standard levels for Radon

measurement in the water. Therefore, using CR-39 for Radon measurement in water is recommended. The damaged volume of 1 m^2 skin surface due to annual Radon and progeny activity and annual Radon dose conversion factor of skin in water is 2.51 mm^3 and 8.8 $\text{nSv}/(\text{Bq}\cdot\text{h}/\text{m}^3)$, respectively. The maximum annual dose for every person (2 m^2 body surfaces) that swims 1.5 hours per day in water with 8 kBq/m^3 Radon concentration is 10.9 mSv :

$$D=(8.8((\text{nSv}/(\text{Bq}\cdot\text{h}/\text{m}^3))/\text{m}^2))\times(2\text{m}^2)\times(52\times 1.5\text{ h})\times(8\times 1000\text{ Bq}/\text{m}^3)=10.9\text{ mSv} \quad (9)$$

5. Conclusion

By Monte Carlo simulation, we tried to obtain the energy and angular spectrum of Radon and progeny alphas on CR-39, calibration coefficient of CR-39 and Radon and progeny alpha energy versus distance traveling in air and water. The suit chemical etching depth of CR-39 for Radon and progeny alpha's and the annual Radon dose conversion factor of skin in water were calculated.

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