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Determination of ^{222}Rn and ^{238}U Concentrations in Some Selected Samples of Rock for Sulaimani Area (Kurdistan Region-Iraq)

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Abstract

Different rock samples collected from the Mountains located to the west and north of Sulaimani district. Solid state nuclear track detector technique, CR-39 passive detectors included, is used for analyzing concentrations Radon. The uranium concentrations determined from the secular equilibrium between uranium and radon activity. The concentrations of Radon and Uranium have been calculated. The maximum and the minimum concentrations determined (4774.524±17.665 and 224401.714) Bq/m^3 for Radon and (0.313±0.143 ppm, 14.728±6.731 ppm) for uranium, respectively. The higher concentration values of both Byara (Mountain of Balkha) and Penjween is due to sample rocks formation compared to the values appeared from Ranya and Chaqchaq regions. The highest annual effective dose was 0.8μSv/hr, which is higher than the value 1mSv ~ 0.114 μSv/hr recommended by the International Commission on Radiological Protection ICRP..

Introduction

There are three groups (igneous, sedimentary, and metamorphic) of rocks. Igneous rocks (from fire) create when molten rock, called magma, hardens [1]. They include many different minerals, such as silica, aluminum, and iron. Molten rock forms when rocks are to dissolve by the heat inside the earth. The molten rock is less intensive than solid rock and tends to move up towards the earth's surface. When magma breaks out to the surface of the earth in a volcano, it is called lava. Sedimentary rocks, such as sandstone and shale, are formed by the pressing together of sediment materials.[1] Uranium represents 0.00016% of the earth's crust and the average concentrations of uranium in the earth crust was about (2-4) parts per million (ppm) [2,3]. It is found mostly as uranite. Uranium is a heavy element, but its combination with oxygen makes it less dense than the magma, and it tends to rise to the surface. Uranium deposits are found on the outer rock surfaces and along cracks in the rock material. Much of the uranium present during igneous rock formation does not enter at all into the crystal structure of any of the minerals.They deposited on the outer rock surfaces at the end of the crystallization process. The motion of radon gas out of rocks as well as the soil-influenced by the amount and location of the uranium underground, and the pathway for Radon movement to the surface.[1,4]

Out of these the relatively stable isotope with half- life of 3.8 day, the most stable isotope is ^{222}Rn, which is a decay product of ^{226}Ra from the natural series decay of ^{238}U [5]. There are two other isotopes of radon: first

the ^{219}Rn derived from the most stable isotope of actinium ^{227}Ac. It is an alpha emitter with a half-life of 3.96 seconds. Secondly, the ^{220}Rn which is a natural decay product from the most stable thorium isotope (^{232}Th). It has a half-life of 55.6 seconds and also emits alpha radiation [4,5].

As radon itself decays from the natural radioactive series thorium and uranium, it produces new radioactive elements called radon daughters or decay products. Unlike the gaseous radon itself, radon daughters are solids and stick to surfaces, such as dust particles in the air. If such contaminated dust inhaled, these particles can stick to the airways of the lung and increase the risk of developing lung cancer [6].

Radon is the second most frequent causes the lung cancer. As declared by United States Environmental Protection Agency EPA, Smoking cigarette causes 21,000 lung cancer deaths per a year in the United States, according to EPA estimates [7].

If a closed volume supplies with Radon constantly, the concentration of short-lived isotopes will increase until equilibrium. The rate of decay of each decay product will equal that of the Radon itself. The equilibrium factor is 1 when both activities are similar, meaning that the decay products have stayed close to the radon parent long enough for the equilibrium to be reached, within a couple of hours. Because of their electrostatic charge, radon progenies join to surfaces or dust particles, whereas gaseous radon does not. Attachment removes them from the air, usually causing the equilibrium factor in the atmosphere to be less than one. In high concentrations, airborne radon isotopes contribute significantly to human health risk [8].

So due to this effects it will be necessary to study Rn concentrations in most of the materials and surrounding air of our lives such as rocks, soil, water ….etc, especially that uses in construction.

Experimental Methods:

In this study, several of samples were collected at different locations of Sulaimani governorate, like Sulaimani, Ranya and Sharazoor (Penjween, Garmek, and Byara).

Figure(1) shows the image of locations of the collecting samples (studied area) taken from the Google Earth-Satellite.

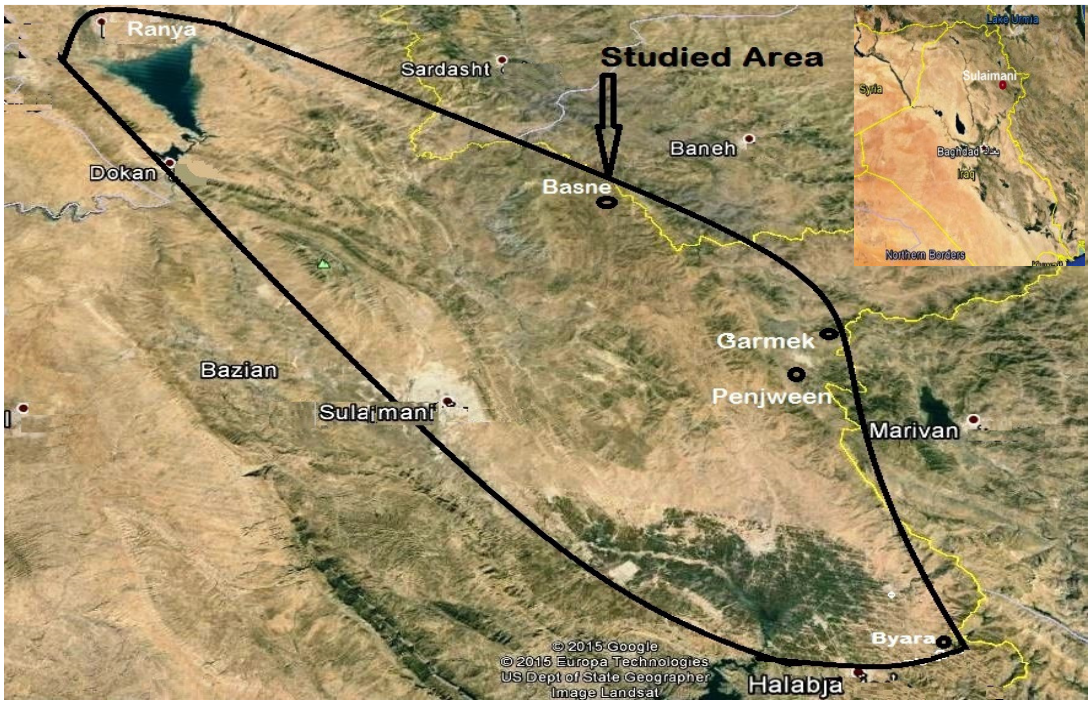

Fig. (1) Satellite image shows the surveyed area of Sulaimani (Kurdistan Region - Iraq)

For the measurement of radon concentration it is necessary to determine the diffusion constant (D) for the system from the relation [9]:

$$\rho = D\, C_o\, T \qquad \text{----------} \quad (1)$$

Where, ρ is the track density Tr\cm^2

D, Diffusion constant

C_o, Rn concentration in air space of the closed-cup tube (B\cm^3)

T, Radiate time

Diffusion constant (D) could bedetermined from the relation below by using the dimensions of the closed-cup technique [10]:

$$D = 1/4\ (r)\ [2\cos\theta t - (r \setminus R\alpha)\ ] \qquad \text{----------} \quad (2)$$

Where r - radius of closed-cup tube for the diffusion volume (3.25cm)

θt - threshold angle for the CR-39 detector (35°),[11]

$R\alpha$ - a range of α-particle in air from radon.

$R\alpha$ can be calculated from the relation [12]:

$$R\alpha = (0.005\ E\alpha + 0.285)\ E\alpha^{3/2} \qquad \text{----------} \quad (3)$$

= 4.019 cm

Then (D) = (0.058241) Tr cm^{-2} d^{-1} / Bq m^{-3}

We calculate Rn concentration in the sample using [13]:

$$Cs = \lambda_{Rn} C_o h t \setminus L \quad \text{---------- (4)}$$

Where, Cs - Radon is the concentration of the samples (Bq\m^3)

λ_{Rn}, Decay constant for Rn (0.1814 day)

h, Height of air space in the closed-cup tube (8.5cm)

L, Thickness of the sample in the closed-cup tube (2 cm)

T, Time of radiate (60 days)

Through this relation [13] the activity of Radon in the sample (A_{Rn}) can calculated:

$$A_{Rn} = Cs\ V \quad \text{---------- (5)}$$

Where A_{Rn} - Activity of Radon

V - The volume of sample ($V = \pi r^2 L$) = 81.388 cm^3

Uranium concentration determined by the number of atoms of radon using the relation below,

$$A_{Rn} = \lambda_{Rn} N_{Rn} \quad \text{---------- (6)}$$

Where At_u is the Mass number of uranium, and N_{avo} is Avogadro number (6.02×10^{23} mol^{-1})

We can calculate the concentration of uranium by ppm Cu (ppm) the relation below is used:

$$Cu(ppm) = M_u / M_s \quad \text{---------- (7)}$$

M_u is calculated from $M_u = N_u At_u / N_{avo.}$, which is the weight of uranium in the samples

N_u is the number of atoms of uranium and M_s is the mass of samples (100 gm).

In this work, the SSNTD technique used a plastic detector CR-39 depending on the extend time for detecting emitted α-particles from the ^{222}Rn gas of the samples which produced from the natural decay of ^{238}U. The CR-39 detectors of thickness 500 μm were cutting by (1x1.5) cm then exposed to (100 gm) of the rock samples inside the closed-cup tube as shown in Fig. (2a) and Fig. (2b)

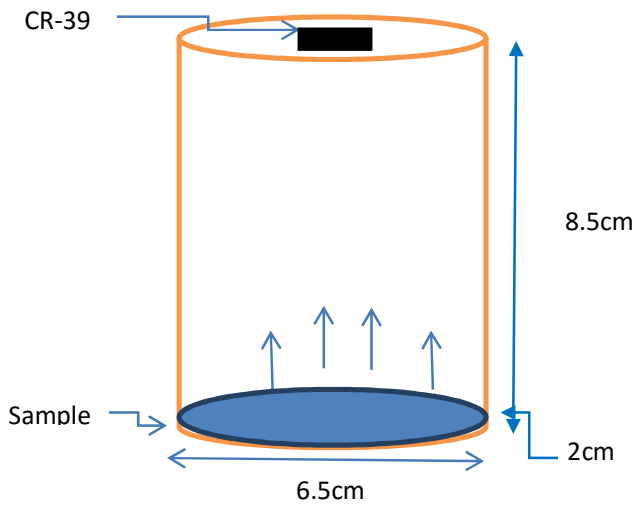

Fig.(2a) Schematic Diagram of Closed-Cup tube for SSNTD Technique.

Fig.(2b) : Closed-Cup tube for SSNTD technique

After closing the tubes cup perfectly they stored for 60 days, then the CR-39 detectors removed from the tube and chemically etched using molarities (6.25M) of NaOH. The etching process continued for 6 hrs at temperature 70 C^{o}. The detectors are washed and then dried. The optical microscope (Olympus) with magnification power 400x used to observe the trace of α – particles with high deep on a CR-39 plastic detector [11,14].

Result and discussion:

The various rocks of samples which have been collected from different locations of Sulaimani area with geological information [15] as shown in Table (1):

Table (1): Locations and geological information for each collecting sample

Samples	Locations	Type
1	Sulaimani-Ranya-Gorago	Limestone
2	Sulaimani -Ranya- Bosken	Limestone
3	Sulaimani -Ranya-Kewarash	Limestone
4	Sulaimani -Ranya-Tenoka	Limestone
5	Sulaimani-Chaqchaq	Limestone
6	Sulaimani-Azmir1	Marly limestone
7	Sulaimani-Azmir2	Marly limestone
8	Sulaimani-Penjween1	Shale and marble
9	Sulaimani-Penjween2	Shale and marble
10	Sulaimani-Basne	Red clay stone
11	Sulaimani-Dukan	Weathered limestone
12	Sulaimani-Byara-Balkha	Siliceous shale and marble
13	Sulaimani-Pejween-Hangazhala1	Shilair Phylite
14	Sulaimani-Penjween-Hangazhala2	Siliceous marble
15	Sulaimani-Garmek-Tatan	Shilair Phylite
16	Sulaimani-Penjween-Mlakawa1	Limestone and slightly Phylite
17	Sulaimani-Penjween-Mlakawa2	Shilair Phylite and shale

Figure (3) showed the observed α-particles from one field of CR-39 detectors. Through the optical microscope, type (Olympus) with magnification power of 400x, Equations 4 and 7 used to calculate the concentrations of radon and uranium respectively.

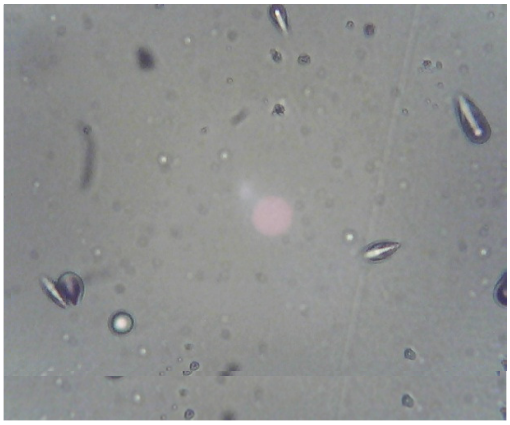

Fig.(3): Microscopic photo of α-Tracks.

Table (2) shows the tracking density of α– particles, Rn-concentrations and U-concentrations of the samples.

Table (2) Rn-concentrations and U-concentrations of the rock samples

Samples	ρ (Tr/cm^2)	(C_o) Bq/m^3	(Cs) Bq/m^3	C_U (ppm)
1	628.9	179.971±3.959	3264.666±16.649	0.321±0.167
2	503.12	143.976±3.167	2611.733±13.319	0.257±0.133
3	754.68	215.965±4.751	3917.600±19.979	0.385±0.2
4	471.67	134.977±2.969	2448.474±12.487	0.241±0.125
5	911.76	260.916±5.74	4733.014±24.138	0.466±0.242
6	5086.8	1455.676±32.024	26405.955±134.67	2.598±1.353
7	6342.8	1815.102±39.932	32925.943±167.922	3.239±1.687
8	17584	5031.965±110.703	91279.843±465.527	8.981±4.678
9	15448.8	4420.941±97.26	80195.862±408.998	7.890±4.11
10	6845.2	1958.872±43.095	35533.939±181.223	3.496±1.821
11	817.57	233.962±5.147	4244.066±21.644	0.418±0.217
12	22168.4	6343.870±139.565	115077.802±586.896	11.322±5.898
13	14632.4	4187.314±92.12	75957.869±387.385	7.473±3.893
14	1949.59	557.909±12.273	10120.466±51.614	0.996±0.518

15	4727.23	1352.778±29.761	24539.400±125.15	2.414±1.257
16	6782.4	1940.901±42.699	35207.939±179.56	3.464±1.804
17	16516.4	4726.453±103.982	85737.852±437.263	8.435±4.394

Concentrations of Radon in the air of closed-cup tube ranged between (134.977±2.969 and 6343.870±139.565) Bq/m^3 but concentrations of Radon in the rock samples ranged between (4774.524±17.665 and 224401.714±830.286)Bq/m^3. The minimum value appeared in the location of Ranya–Tenoka, Gorago, Bosken, Kewarash, Sulaimani-Chaqchaq and Sulaimani-Dukan because the compositions of the rock sample. These regions composed of limestone without clay that have a lower concentrations of uranium [16,17]. The maximum value appeared in the regions of Byara (mountains of Balkha), Penjween1, Penjween2, Hangazhala2 and Mlakawa2. The rock samples in these regions composed of the Siliceous shale and marble enrichment with a high concentration of uranium according to the geological information [15]. The high value appeared from the rock samples of Sulaimani (mountains of Azmir) when the structure of these regions was Marly limestone [15]. In general, high levels of radon are associated with granite igneous rocks, shale and dirty quartz sedimentary rocks, phosphate deposits and some beach sands, which may contain high levels of radon progenitors[18]. Figure (3) shows the radon concentrations of the rock samples in this work.

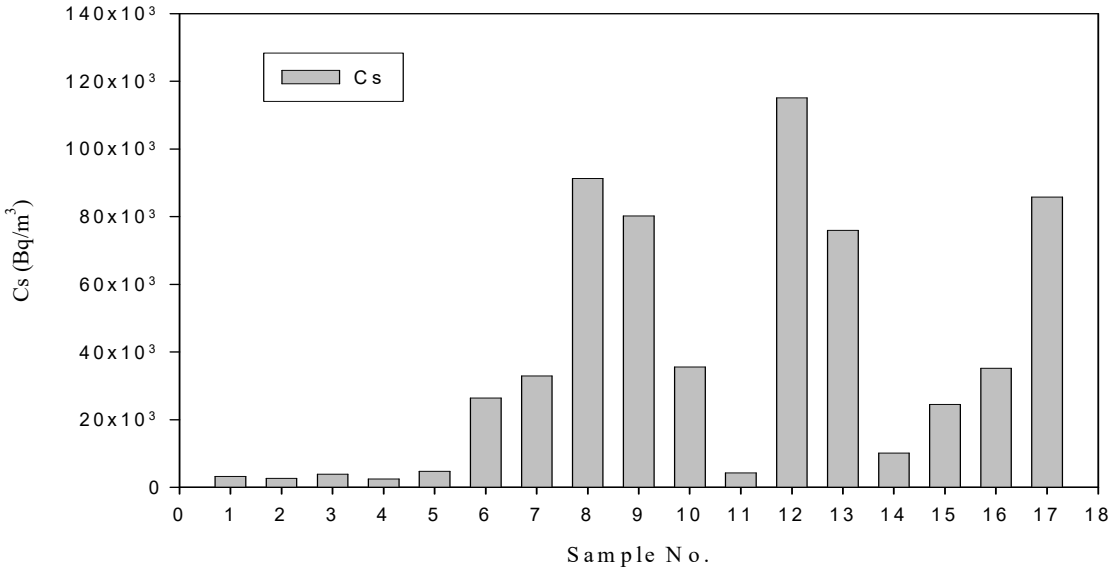

Fig.(3) Radon concentrations of rock the samples

The results compared to other works done in the Shale in the locations Marine, North Carolina, and South Carolina. The high Radon concentration appeared on the rocks of type Granite and Metamorphic. However, the minimum values appeared in Florida, North Carolina, South Carolina and Sweden due to the structure of rocks which was composed of limestone as appeared in our results. Table (3) shows the data from these regions [18].

Table (3) Radon concentrations of some types rock in the other works

Type of rocks and locations		**Concentration of ^{222}Rn (Bq/m^3)**
Granites	Marine	817,700
	North Carolina	390,800
	South Carolina	298,800
	Sweden	92,000
Metamorphic Rocks (Shale)	Marine	
	- Sillimanite zone	503,300
	- Chlorite zone	41,000
	North Carolina	
	- Gneiss/schist	83,000
	- Metavolcanic	49,900
	South Carolina	
	- High-grade-monazite belt	53,400
	- Medium Grade	118,100
	- Low-grade	274,000
	Sweden	
	- Gneiss	26,000
	Florida	550

Limestone	North Carolina	3,440
	South Carolina	1,300
	Sweden	24,000
	North Carolina coastal plain	15,760

The uranium concentrations ranged between (0.313±0.143 and 14.728±6.730 ppm. The minimum value appeared in the location of Ranya–Tenoka, Gorago, Bosken, Kewarash, and Sulaimani-Chaqchaq, Sulaimani-Dukan. The compositions of rock samples in that regions were composed of limestone without clay. The maximum values appeared in the regions of Byara (mountains of Balkha), Penjween1, Penjween2, Hangazhala2 and Mlakawa2. The rock samples in these regions composed of the Siliceous shale and marble enrichment with a high concentration of uranium according to the geological information. They are the same locations for radon because radon produced from the natural series decay of uranium. Fig.(4) shows uranium concentrations in the rock samples.

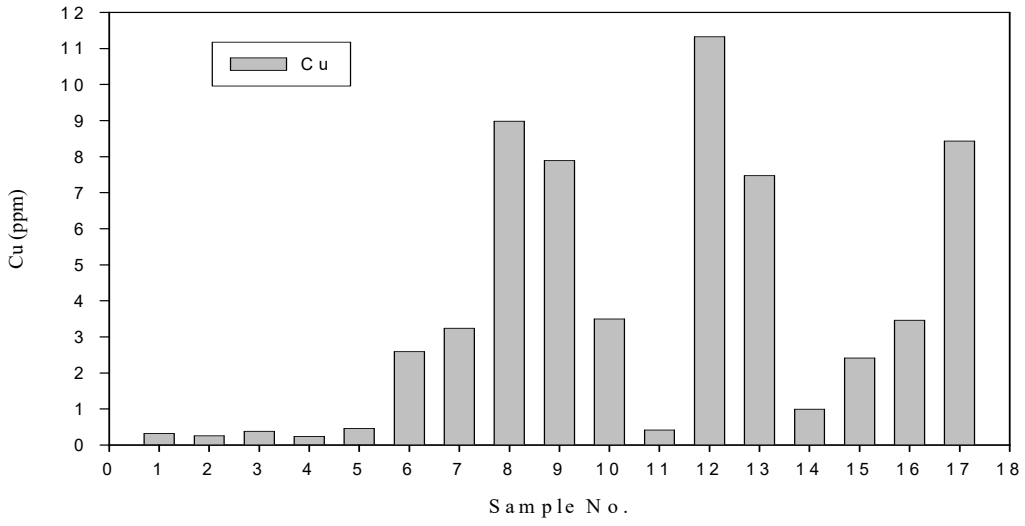

Fig.(4) Uranium concentrations of the rock samples

The background dose rate measured by Dosimeter,(UltraRadiac-CANBERRA) Model-MRAD111, the minimum value of effective doses were 0.108 µSv/hr at the locations Sulaimani -Ranya-Tenoka, Gorago, Bosken, Kewarash.The maximum value was 0.8 µSv/hr at the locations Sulaimani-Penjween1, Penjween2,Hangazhala2 , Mlakawa2 and Balkha. The values wre higher than the world recorded value of (1mSv ~ 0.114 µSv/hr, they are recommended by the International Commission on Radiological Protection (ICRP) [19,20]. These are due to the high background radiation in that regions, also we observed the same result through using a portable scintillation counter type (GS4- Scintillator counter) when the high count radiation was ranged between (250-350) count/sec and from the other low background was ranged (35-55) count/sec in that regions, this will improve our works accuracy.

Conclusion:

The radon concentrations resulting from radon emanating from rock samples have been estimated in the studied region by using Solid State Nuclear Track Detector (SSNTD) technique during CR-39 detector. Within the measured rock samples, radon and uranium concentrations from Sharazoor regions (Balkha and Penjween) have higher values than the others by comparing with the world values due to high background radiation and the natural composition of these regions..

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