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RESEARCH PAPER

Evaluation of Radon (^{222}Rn) Exhalation Rates from Imported Granite Tiles Used as a Building Materials in Erbil Governorate, Kurdistan Region -Iraq

Hindren R. Awla[1], Habeeb H. Mansour [1]

1Department of Physics, College of Education, Salahaddin University-Erbil, Kurdistan Region, Iraq

A B S T R A C T:

The present study aims to evaluate the activity concentration of effective radium content (Ra_{eff}), and radon exhalation rates (^{222}Rn) of 21 imported granite samples that are used as building materials in Erbil Governorate. The measurements were carried out using a Rad7 and LR-119 type II plastic track detectors as an active and passive measuring, respectively. The radon activity concentrations for both detection methods were varied from 25.9 ± 6.85 to 427 ± 38.3 Bq.m^{-3}, and from 31.23 ± 5.8 to 433.63 ± 35.2 Bq.m^{-3}with an average value of 99.35 ± 13.84 Bq.m^{-3} and 103.16 ± 9.46 Bq.m^{-3}, respectively. The effective radium content, radon exhalation rate in terms of surface and mass were calculated using LR-115 technique, and it was found to vary from (0.2 ± 0.04 to 2.94 ± 1.5) Bq.kg^{-1}, (59.07±7.67 to 819.95±28.5) mBq.m^{-2} h^{-1}, and (1.5±1.1 to 22.23±4.5) mBq.kg^{-1}.h^{-1} respectively. The results show that some of the granite samples exceeded the radioactively acceptable limit, which means that they may be harmful to the human health.

KEY WORDS: radon gas,LR-115 and Rad 7 radon detectors, radon exhalation rates, effective radium content, granite tiles.
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1.INTRODUCTION:

Measurement of radon gas ^{222}Rn exhalation rates from granite samples used in the construction of dwellings is useful for assessing the sources contributing to the risk of indoor radon to populations of humans (Menon et al., 2015). Radon(^{222}Rn), the radioactive gas of half-life 3.8 day, originates from ^{226}Ra, which is a decay product of ^{238}U series, and found in every kind of soils and rocks (Shashikumar et al., 2019)

Radon 222Rn and its progenies may cause a serious health risk, in particular when concentrated in some enclosed areas like caves, underground mines, and cellars; when using building materials with high concentrations of uranium, such as granite tiles; or in badly designed and poorly ventilated houses (Najam et al., 2013). On the other hand, the survey of radon gas exhalation rate from construction materials are useful for understanding the individual concentration of each material to the total exposure of the indoor radon. Many investigations have studied radon exhalation rate from building materials especially from granite rocks, and reported the typical range of radon exhalation rates in different countries (Abbasi and Mirekhtiary, 2013, Del Claro et al., 2019, Kuzmanović et al., 2019).

Nowadays, Exposure to ^{222}Rn in indoor air is addressed in health regulations in many countries. As a radiotoxic and carcinogenic gas, radon could have an effect on the quality of indoor air (Nhan et al., 2012). The epidemiological research established a strong relationship between long-term exposure to high ^{222}Rn concentrations and lung cancer(WHO, 2009).

There are several techniques for the measurement of radon and its progenies activity concentration using passive and active methods (Azeez et al., 2021, Omar, 2020, Nastro et al., 2018). For passive measurement, solid state nuclear track detectors (SSNTDs) have been

* **Corresponding Author:**
Hindren R. Awla
E-mail: handrenramazan@gmail.com
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widely used, like CN-85, CR-39, LR-115, etc, which can register alpha particles of different energies emitted by radon gas and its progenies over long time scales (Najam et al., 2013, Yousef et al., 2015a, Mahmood et al., 2014). On the other hand, active methods are usually used for short term measurement which include scintillation methods like a Lucas cell, Rad7 (solid state detectors), surface barrier detector and Ionization Chamber, etc (Stajic and Nikezic, 2015, Hassan et al., 2011, Amasi et al., 2015).

The main objectives of this study were to measure the activity concentration of radon, radium effective content and the radon exhalations rate of granite tile samples, which are widely used as construction materials and decorations, including the inner cover of Erbil governorate houses. The measurements were verified using active (Rad7) and passive (LR-115) methods. The results are helpful for the estimation of radiological parameters which are of great interest in the study of environmental radiation protection in Iraqi Kurdistan Region.

2.MATERIALS AND METHODS

2.1.Samples Collection and Experiment Set up:

Twenty-one samples of imported granite tiles from different countries that widely used in construction were collected from the local market of Erbil governorate. Since radon concentrations are largely affected by moisture, (Lee et al., 2021), the samples were powdered by using a special machine to obtain a granite powder, then dried in an oven for 48 hours at 110 ^oC to evaporate any moisture content and getting a real weight (Ari, 2022). After drying, the powder of each sample was divided into nearly three equal volumes and then put inside a plastic container type polyvinyl chloride (PVC) of volume 115.5 cm^3 and sample surface area was 38.5 cm^2. The containers are closed tightly by a calibrated diffusion tube as a radon chamber made of the same material (PVC) of 27 cm in high and 7 cm in diameter (Habib, 2000). Then held in the big Flynn box to be storage at room temperature's and to get secular equilibrium between radium (^{226}Ra), radon (^{222}Rn) and radon its progenies.

2.2.Measurement Methods

To emphasize the accuracy of the results, the calibrated diffusion tube as shown in Fig.1, provided with two types of detectors used for measurement of radon gas activity concentration released from the samples under the study. One is Rad 7 solid state detector: which is an active device for radon monitoring, manufactured by DURRIDGE Company, USA, connected throughout a closed loop contains desiccant tube and diffusion tube. While the other is LR-115 type II nuclear track detector (Kodak brand 12 μm thick) used as a passive method. A piece of this type of 1cm x 2cm area were fixed on the bottom of the rubber stopper of each tube chamber with tape such that sensitive side of the detector faced the specimen to start the measurement and after the process of time of sculler equilibrium between radium and radon has attend , both valves are opened and the Rad 7 pump draws air from the diffusion tube to the Rad 7 chamber after passing through desiccant tube filled with calcium sulfate ($CaSO_4$) and an inlet filter to the internal hemisphere detection chamber, then, the air returns to the tube chamber from the outlet button on Rad 7 device. The semiconductor detector in Rad 7 converts the α radiation of radon progenies directly to an electrical signal using an alpha technique which is able to discriminate the electrical pulse generated by α particles from ^{218}Po with 6.0 MeV and ^{214}Po with energy 7.0MeV. In this way, it is possible to use only the polonium ^{218}Po peaks for radon (^{222}Rn) which is achieved in about five times the half-life ($t_{1/2}$) of polonium ^{214}Po. By employing this method, it can be possible to measure radon activity concentration released from samples under the study.

The activity concentration is obtained from a calibration factor determined from radon chamber run by the US EPA and the DURRIGE Co., were equal to 123 (cpm/ Bq. L^{-1}) with uncertainty (2%) for normal mode. Continuous to our experiment, and to attend the passive method for radon gas detection throughout the diffusion tube, pieces of calibrated LR-115 were exposed to radon gas(^{222}Rn) for a period of about 67 days, were retrieved and etched for 2 hours in 2.5N NaOH solution maintained at a temperature of (60 ± 1)^oC in a constant temperature water bath to reveal the tracks(Agba et al., 2017). Then, the detectors were steeped in distilled water for 2 minutes, washed, and dried with fresh air. Etched alpha particle tracks on sensitive surface of the LR-115 detectors were scanned under the Motic digital microscope at a 400X of magnification, after subtracting the back ground which is equal to 23±4.8 $tracks.cm^{-2}$.

To measure a background in diffusion tube before sampling, we connected the RAD7 outlet to the open end of the drying tube and the diffusion tube valves, making a closed loop. The background value in this experiment is 4.0 ± 0.7 Bq.m^{-3} and then subtracted from the actual measurements.

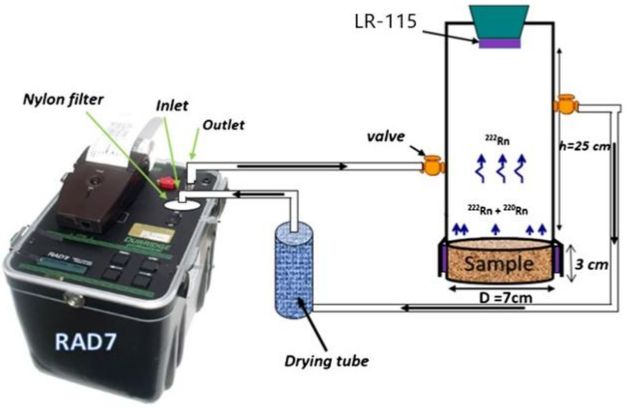

Figure (1): The schematic diagram of equipment used in this work, to measure the activity concentration of ^{222}Rn in granite samples, using the RAD7 and LR-115 detectors.

3.LABORATORY MEASUREMENTS

3.1.Radon concentration (C_{Rn})

The activity concentration of radon C_{Rn} in different granite samples are calculated by Eq. (1) for passive method (Thabayneh, 2015):

$$C_{Rn}(Bqm^{-3}) = K_{Rn}\left(\frac{\rho}{T_{eff}}\right) \dots\dots\dots (1)$$

where C^{Rn} activity concentration of radon for granite samples in Bq.m^{-3}; K_{Rn} is the calibration factor equal to 0.0301 in ((tracks. cm^{-2}d^{-1}) per Bq. m^{-3}) (Azeez, 2019), ρ is the density of alpha track in (tracks/cm^2) and T_{eff} is the effective exposure time (61.48days), which is the correction exposure time needed only for the closed system and can be calculated by Eq. (2) (Durrani and Ilic, 1997, Duggal et al., 2012):

$$T_{eff} = T - \frac{1}{\lambda}\left[1 - e^{-\lambda T}\right] \dots\dots\dots (2)$$

Where T is the exposure time (67 days) and λ is, decay constant of radon gas =0.181days^{-1}. The obtained results are shown in Table 1.

3.2.Effective Radium Content

The effective radium content Raeff (Bq.kg^{-1}) is the specific concentration that produced radon by disintegration radium, able to escape from the granite samples into the surrounding air. The following equation (3) can be used to calculate the (Ra_{eff}) of granite samples(Shakir Khan et al., 2011, Somogyi, 1990):

$$Ra_{eff}\ (Bq.kg^{-1}) = \left(\frac{\rho}{K_{Rn} \times T_{eff}}\right)\left(\frac{hA}{M}\right) \dots (3)$$

Where (ρ) is the observed track density on the LR-115 detector (Tracks/cm^2), A is the cross-sectional area of the chamber (38.5 cm^2), h is the distance between the detector and top surface of the granite samples, M is the mass of the granite samples in kg, (Teff and K_{Rn}) are defined in equations (1 and 2).

3.3.The Radon Exhalation Rate

The radon exhalation rate is the flux of radon released from the surface of material samples. The surface exhalation rate (E_A) in the granite samples was measured (Kassi et al., 2018):

$$E_A\ (\mathrm{Bq.m^{-2}}.h^{-1}) = \frac{\rho.V\lambda}{A \times T_{eff} \times K_{Rn}} \dots\dots\dots (4)$$

Where V is the radon chamber's volume (9.61× 10^{-4} m^3) and (λ) is the decay constant of radon= 0.18145 day^{-1}. Also, the mass exhalation rate (Em) can be calculated (Kassi et al., 2018):

$$E_m\ (Bq.kg^{-1}.h^{-1}) = \frac{\rho.V\lambda}{M \times T_e \times K_{Rn}} \dots\dots\dots (5)$$

All factors are defined in equations (3,4). Where, M is the mass of the granite samples in kg. Table 2 presents the obtained results for both E_A and E_M.

Table (1): Details on samples used in this investigation and CRn the radon activity concentration levels measured using LR-115 and Rad 7 detector for 21 granite samples.

Sample	Origen	Color	Density ($kg.m^{-3}$)	C_{Rn} ($Bq.m^{-3}$) LR-115	C_{Rn} ($Bq.m^{-3}$) RAD 7
01Ch	China	Beige-black	1390.87	75.33±7.3	84.0±16.4
02Ch	China	Pink	1217.55	58.79 ±6.2	52.7±10.1
03Ch	China	Beige	1255.77	64.31±7.1	41.7±11.4
04Ch	China	White-black	1243.29	67.98±5.3	59.2±11.9
05Ch	China	Black	1403.61	51.45±5.9	47.8±10.1
06Ch	China	Maple red	1207.68	235.2±13.8	246±18.3
07Ch	China	Dark red	1337.06	97.38±9.1	83.1±9.46
08In	India	Raw black	1175.27	69.82±7.7	57.3±12
09In	India	Black galaxy	1586.9	56.96±4.1	42.7±9.11
10In	India	Red – black	1324.06	226±19.6	257±23.5
11In	India	Raw beige	1286.02	71.66±9.7	64± 13.4
12In	India	Red brown	1264.53	45.93±3.9	47.5±12.1
13In	India	Red	1230.12	433.63±35.2	427±38.3
14In	India	Black-beige	1302.22	38.58±2.6	35±10.6
15Ve	Vietnam	Beige-black	1275.36	64.31±5.5	42.8±10.2
16Ba	Brasilia	Yellow-brown	1253.78	220.49±18.6	205±27.4
17Ba	Brasilia	Baltic brown	1311.93	121.27±15.3	124±12.1
18No	Norway	Emerald blue	1308.89	31.23±5.8	29.5±6.85
19Uk	Ukraine	Black pearl	1318.77	49.61±4.3	38.2±10.5
20Eg	Egypt	White – black	1301.62	42.26±3.2	61.9±8.38
21Ir	Iran	White	1222.32	44.1±8.4	43.6±8.6
Averag			1296.07	103.35±9.46	99.35± 13.84

4.RESULTS AND DISCUSSION

The values of radon activity concentration C_{Rn} and radon exhalation rates from materials, in general, depend on many physical and chemical properties like the porosity, bulk density, the chemical composition, and the radium ^{226}Ra content, etc. In this study, we determined the bulk density, and the emanated radon activity concentration from 21 granite samples using both passive and active methods. The values are given in Table (1), It is clear that the radon activity concentrations measured by LR-115 detector vary from (31.23 ±5.8 to 433.63±35.2) $Bq.m^{-3}$with an average value of 103.35±9.46 $Bq.m^{-3}$. Whereas, their values by using RAD7 solid state detector ranged between (29.5±6.85 to 427±38.3) $Bq.m^{-3}$, with an average value of (99.35 ± 13.84) $Bq.m^{-3}$. The lowest and highest values were found in samples 18No and 13In, which were imported from Norway and India, respectively. The emanated radon gas ^{222}Rn from the granite samples under the study in both methods was shown in Fig. (2). It is clear that only four samples (13In,10In,06Ch and 16Ba) of all the studied granite samples have radon concentration exceeding the action levels of 148 $Bq.m^{-3}$ as proposed by EPA(EPA, 2003). Meanwhile, five granite samples (13In,06Cn,10In,16Ba and 17Ba) of all the studied granite samples have C_{Rn} exceeding the action level of 100 $Bq.m^{-3}$ suggested by WHO(WHO, 2009). The linear correlation of determination of (R^2=0.984) for both methods were recorded as seen in Fig. (3).

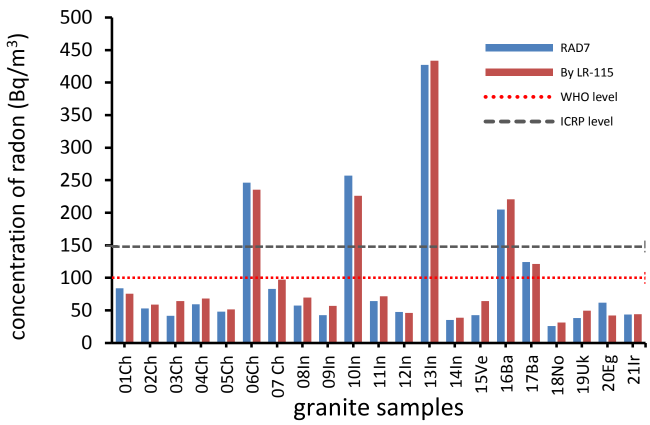

Figure (2): Activity concentrations of radon gas by using both passive (LR-115) and active (RAD7 detector) methods for granite samples.

The effective radium content, surface exhalation rate and mass exhalation rate for the granite samples under the study were measured using LR-115 SSNTD, as illustrate in Table (2). Effective radium content (Ra_{eff}) in the granite samples varied from (0.2±0.04 to 2.94±1.5) $Bq.kg^{-1}$, with an average value of 0.68±0.075 $Bq.kg^{-1}$. Furthermore, the surface radon exhalation rate was seen to change from 59.07±7.67 to 819.95±28.5 $mBq.m^{-2}h^{-1}$, with an average value of 195.07±12.9 $mBq.m^{-2}h^{-1}$.

Table (2): Effective radium content (Ra_{eff}), mass exhalation rate EM (^{222}Rn), Surface exhalation rate E_A (^{222}Rn), Effective annual dose and estimate life cancer rick of radon gas released by granite samples, calculated using passive method (LR-115).

Sample code	Ra_{eff} ($Bq.Kg^{-1}$)	E_A ($mBq.m^{-2}h^{-1}$)	E_M ($mBq.kg^{-1}.h^{-1}$)
01Ch	0.452±0.067	142.45±11.9	3.41±1.8
02Ch	0.403±0.06	111.18±10	3.04±1.74
03Ch	0.427±0.065	121.6±11	3.23±1.6
04Ch	0.456±0.067	128.55±11.3	3.44±1.8
05Ch	0.306±0.055	97.28±9.85	2.31±1.5
06Ch	1.625±0.12	444.72±21	12.28±3.5
07Ch	0.608±0.077	184.14±13.5	4.59±2.1
08In	0.496±0.07	132.03±11.4	3.74±1.2
09In	0.299±0.054	107.71±10.3	2.26±1.4
10In	1.424±0.19	427.35±20.3	10.76±3.2
11In	0.465±0.068	135.51±11.6	3.51±1.5
12In	0.303±0.055	86.86±9.3	2.29±1.4
13In	2.941±1.5	819.95±28.5	22.23±4.5
14In	0.247±0.049	72.96±8.53	1.86±1.2
15Ve	0.421±0.065	121.61±11	3.18±1.6
16Ba	1.467±1.02	416.92±20	11.09±3.6
17Ba	0.771±0.087	229.31±15.12	5.82±2.1
18No	0.199±0.044	59.07±7.67	1.5±1.1
19Uk	0.313±0.056	93.81±9.6	2.37±1.5
20Eg	0.271±0.052	79.91±8.9	2.04±1.4
21Ir	0.30±0.054	83.39±9.1	2.27±1.5
Average	0.68±0.075	195.07±12.9	5.11±2.03

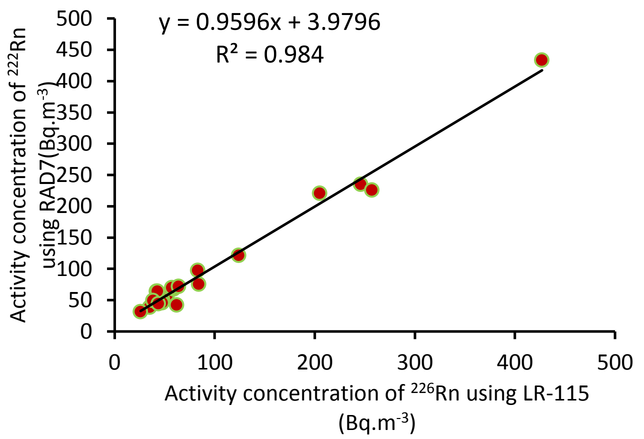

Figure (3): The linear relationship between the active and passive techniques

In addition, the values of mass exhalation rate Em for granite samples under the study were measured and ranged from 1.5±1.1 to 22.23±4.5 $mBq.kg^{-1}.h^{-1}$, with an average of 5.11±2.03 $mBq.kg^{-1}.h^{-1}$. Fig. (5) shows a linear relation between the effective radium content Ra_{eff} in granites and the radon activity concentration C_{Rn}, mass exhalation rate E_M, and surface exhalation rate E_A emanated from granite samples, with the correlation coefficients (R^2) of 0.9975, 0.9975 and 1.0, respectively. This indicates that the C_{Rn} and exhalation rates of radon from granite samples, depends mainly on radium concentration, and on the difference in the bulk densities of the samples as shown in Fig. (6). Furthermore, Fig. (4) shows an important straight line between mass and surface exhalation rate of radon from granite samples under the study with R^2=0.9975.

The range values of radon activity concentration and surface radon exhalation rates in the present study were compared with the results of the literature as shown in Table (3) for granite samples in different countries.

5.CONCLUSION

In this research work a positive correlation has been observed between the results of passive and active methods for radon activity concentration measurement in imported granite tile samples using Rad 7 solid state detector and LR-15 plastic track detector, respectively. Radon concentration levels, radon exhalation rates, and the effective radium contents, were determined from the samples. The results showed that granite samples contain different radon concentration levels. In general, about 20-25 % of the samples from India, China, and Brasilia registered higher levels than the action levels recommended by ICRP-2010 (ICRP, 2010), and WHO(WHO, 2009). Finally, we recommend that citizens who use granite rocks in construction or indoor decoration, to check the indoor radiation doses for the health safety.

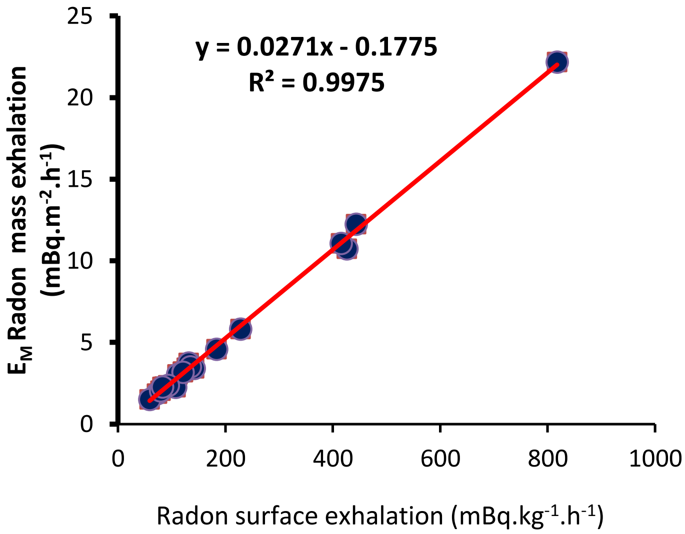

Figure (4): linear relationship between mass and surface radon exhalation.

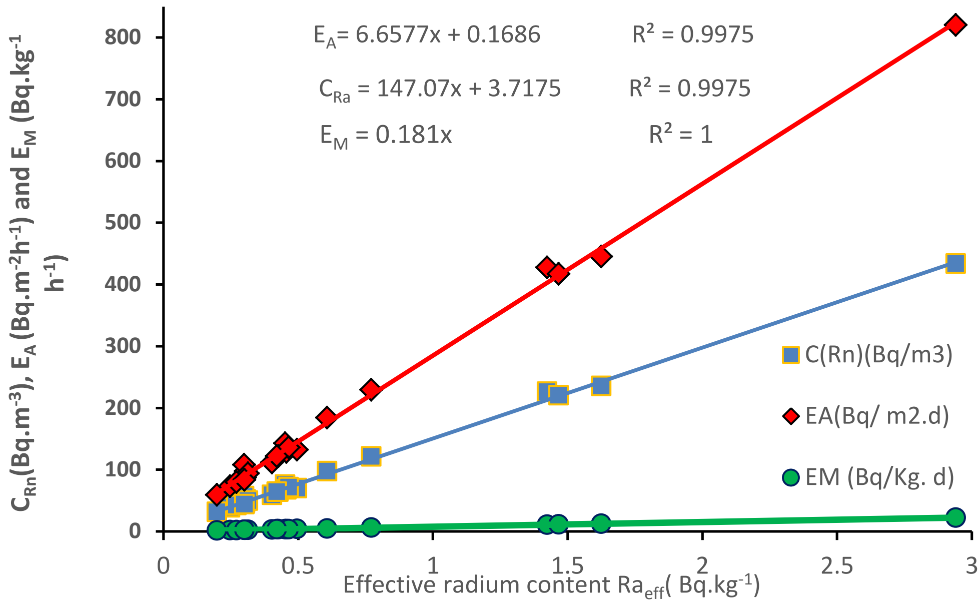

Figure (5): Relationship between Effective radium content with radon exhalation rates (mass and surface) and activity concentration of ^{222}Rn released from granite samples.

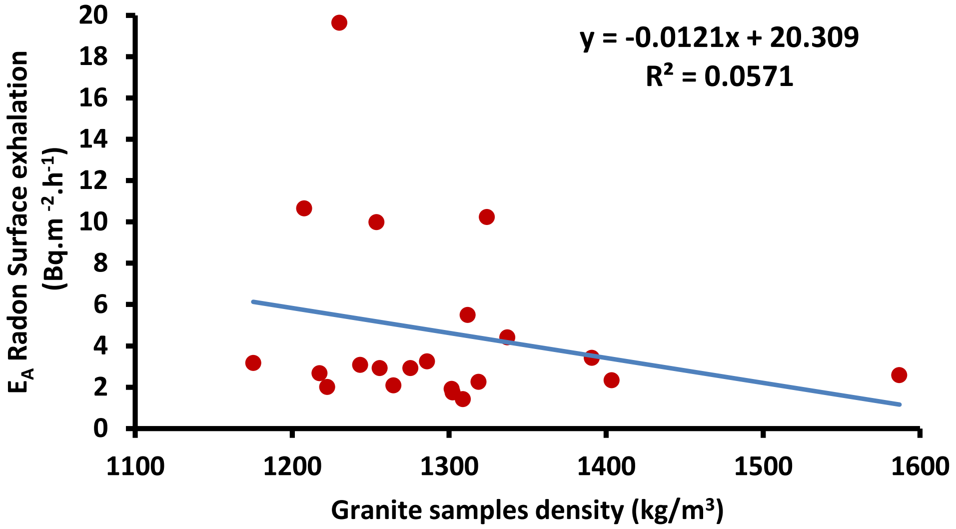

Figure (6): Relationship between surface exhalation rat in (Bq.m^{-2}.h^{-1}) with granite samples bulk density in (kg.m^{-3}).

Table (3): Some global studies on the range of radon activity concentration CRn and exhalation rates from granite samples.

Country	C_{Rn} ($Bq.m^{-3}$)	E_M ($mBq.kg^{-1}.h^{-1}$)	E_A ($mBq.\ m^{-2}.\ h^{-1}$)	Measurement Method	Reference
Iraq	34±4-19434±2332	0. 26±0. 3-168±22	-------	CR-39	(Kobeissi et al., 2012)
Brazil	15±10-1127±24	--	60-2100	Rad7	(Del Claro et al., 2019)
India	95.6±8-1140±121	13.1± 1.3–156.2±11.4	94±8.2–1127±18.6	LR-115	(Pillai et al., 2014)
Egypt	590.61±36.92 - 1319.06±52.81	7.72±0.47 – 19.38±0.76	761.24±46 - 1699.46±66.99	LR-115	(Yousef et al., 2015b)
Iran	218-1306	----	213±13-874±28	Alpha guard	(Abbasi and Mirekhtiary, 2013)
Iraq-Kurdistan	38.58±2.6-433.63±35.2	1.5±1.1 - 22.23±4.5	59.07±7.67 - 819.95±28.5	LR-115	Present study

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