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A Study of Radon and thoron Gases Release from Kirkuk governorate in some Building Materials Using Passive and Active Methods RAD7

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Keywords
radioactive pollution, Building Materials, CR-39, Radon gas, RAD7.

Abstract
Radon is the most important natural radioactive factor harmfully influencing the human population, because radon is radioactive gas comes from the natural decay of uranium deposits in soil, rocks, and water, which is harmful on human and environment. this study was concerned with the use of calibrated long-tube technique equipped with calibrated CR-39 passive track detector, and the use of RAD7 active solid state detector for measuring activity concentration and exhalation rate of radon and thoron gases from 20 building materials which are used in construction in the Kirkuk governorate. The lower and higher values of the radon activity concentration , radon exhalation rate by using CR-39 plastic track detector have been recorded for samples of (Iraqi plaster and Jordan paint) was found to be (21.32±4.61 Bq.m-3, 94.1±9.7 Bq.m-3) , (4.79 , 29.178 mBq.m-2.h-1), (0.145 ,5.29 mBq.Kg-1.h-1) respectively. The lower and higher values of the thoron activity concentration , thoron exhalation rate by using CR-39 plastic track detector have been recorded for samples of (Iraqi plaster and Jordan paint) was found to be(7.10±2.66 Bq.m-3 , 78±8.86 Bq.m-3),(1.59 mBq.m-2.h-1 , 21.178 mBq.m-2.h-1),(0.055 mBq.Kg.-1.h-1, 1.29 mBq.Kg-1.h-1) respectively. Finally, it was concluded that the levels of the radon and thoron exhalation rates in Kirkuke governorate building materials are within the internationally acceptable values.

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1. Introduction

Radon is a natural radioactive inert gas [1,2], it is colorless odorless taste less gas [3]. Its density is higher than air by 7.5 times [4]. Radon is a noble gas with a slight ability to form compounds under lab conditions. The density of radon is 9.73 g/l [5] . Its half-life is 3.825 days which is long enough to allow it to move through the soil and enter the atmosphere reaching the human environment [6]. Radon is perhaps the most important natural radioactive factor harmfully influencing the

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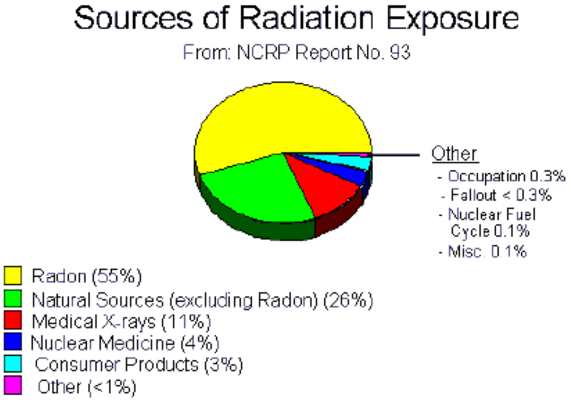
human population, because radon is radioactive gas comes from the natural decay of uranium deposits in soil, rocks, and water, which is harmful on human and environment [7]. There are three naturally occurring isotopes of radon gas. The first one is(^{222}Rn)and called Radon (Radium emanation) which is produced from the decay of ^{238}U series which represents about 99.3% of the natural uranium within the earth's crust .Radon's half-life is about (3.825 days) with decay constant of about (0.1812 day^{-1}) [8]. The second isotope (^{220}Rn) is called Thoron (Thorium emanation) which is naturally produced during the decay of ^{232}Th series. The third isotope (^{219} Rn) is called Actinon (Actinium emanation) which is produced through decay of ^{235}U series [9]. The assessment of radiological risk related to inhalation of radon and radon progeny is based mainly on the integrated measurements of radon. The international commission for Radiological protection has suggested that areas where 1% or more of the building have indoor radon concentration higher than the 10 times of national average should be considered as "radon prone" areas. Therefore, it is desirable not only to measure the radon but also to find out the sources of radon especially in the houses [10]. Radon exhalation from building materials depends not only on the radium concentration, but also on factors such as:

- The fraction of radon produced which is released from the material’s grain to its interstitial space, also known as the emanation power of fraction
- The porosity of the material
- The surface preparation and building material covering [11] .

The present work aims to investigate radon gas concentrations, surface exhalation rate and mass exhalation rate in commonly building construction materials used in Iraq.

Radon and its progenies are proved to be a health hazard and they contribute around (50-55%) to the total radiation population exposure , as illustrated in Fig.(1-1), where by estimated contribution of thoron and its decay products to the annual effective from radon is about 8%. However, the behavior of thoron and its progenies and their effects on human health are not completely understood [12]

Figure 1: Sources of Radiation Exposure; from NCRP Report [13].



2. Experimental part

The samples under study were collected from various locations in Kirkuk governorate ,The radon and its daughters were detected by a passive technique of solid state nuclear track detectors using the " sealed can technique and active detector RAD7. A total of 20 different types of building materials that are widely used in Kirkuk (brick, paint ,Ceramic, cement, soil,) were sampled from different factories in Kirkuk governorate. The samples were cleaned from strange things, and powdered by using a special machine to obtain a homogenous grain size and then dried in an oven at 110 C^0 for 48 hours to evaporate all the moisture content and to maintain the actual weight [14], After drying, the powder of each sample were divided into nearly three equal volumes and then put inside a plastic containers of volume 115.5cm^3 and the sample surface area was 38.5cm^2 as shown in Fig. 2. These containers were made of PVC (poly vinyl choired) which is an impermeable and a tight container to prevent the escape of radiogenic gases, radon and thoron [15].

Having prepared the sample setup, the next step was to install radon diffuser within the containers. For this purpose a long-tube techniques (made of the PVC material of 3.5cm in radius and 27cm in height) contain pieces of size (2cm x 1cm) of CR-39 plastic track detectors were placed at a distances of 25cm and ~0.3cm from the surface of powder samples to discriminate and evaluate radon and thoron with high degree of resolution and accuracy [16], then the long-tube techniques hermetically scaled.

The CR-39 detector pieces, each (2cm x 1 cm) of size, were placed into position on a holder ~0.3cm from the open end of tube, while the upper detector was at about 25cm attached the rubber stopper surface at the closed end as shown in Fig. 2. The detectors were placed such that the sensitive surface of the lower detector faced upwards and that of upper detector downwards .This arrangement ensures that the lower detector dose not register alpha-particles directly from the material surface. The upper detector records mostly the radon component product from the material source while both radon and thoron alpha-particles registered by the lower detector. The differences in the track densities of two detectors is the thoron content of the materials source mixture.

The long-tube technique were also provided with two valves which can be used to connect the input and output of RAD7 monitor for measuring the radon activity concentration, as in Figs.(3) and (4). All the tubes were tightly closed from the top, sealed and putted in large cork boxes, and then left at room temperature.

The container was then sealed for three months; during that time, α particles emitted by radon and their daughters bombarded the CR-39 track detectors. After the irradiation, the detectors were developed in NaOH solution 6.25N at 70C° for 5 hours; after chemical etching, α particle track densities were determined by an optical microscope 400X. The radon concentration CR_n in $Bq.m^{-3}$ was determined by the following equation[17]:

$$C_{Rn}=\frac{\rho x}{Ft} \qquad (1)$$

Where ρx is the track density in (tracks.mm^{-2}), t is the exposure time in (days) and F is the calibration factor of (0.66Tr.cm^{-2} d^{-1}/Bq.m^{-3})

Figure 2: Experimental setup for measuring radon and thoron exhalation rate from building materials[18].

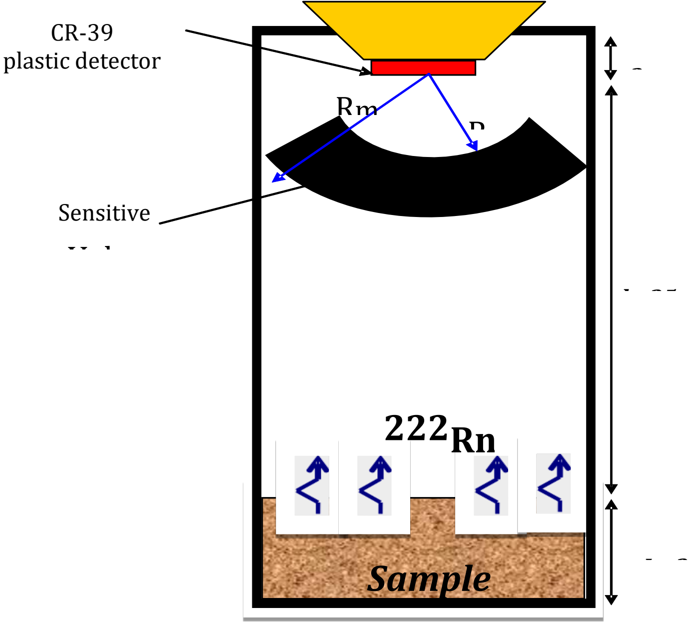
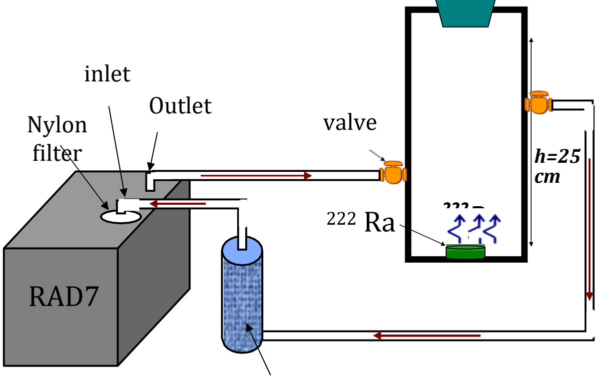


Figure 3: Radon and thoron chambers construction exhalation rate .

Figure 4: Experimental setup for measurement of radon released from building material using RAD7 radon monitor



The radon exhalation rate in terms of area EA in (Bq.m-2.h-1) was calculated as follows [19,20]:

$$E_A = \frac{CV\lambda}{A\left[T + \lambda^{-1}\ \{e^{\lambda t} - 1\}\right]} \qquad (2)$$

Where C is the integrated radon exposure measured by the CR-39 SSNTD (Bq.m-3.h), V is the effective volume of the container (m^3), λ is the decay constant of radon (h-1), T is the exposition time (h), and A is the area covered by the container (m^2) [10].

The radon exhalation rate in terms of mass E_M in (Bq $kg^{-1}h^{-1}$) was calculated as:

$$E_m = \frac{CV\lambda}{M\left[T + \lambda^{-1}\{e^{\lambda t} - 1\}\right]} \qquad (3)$$

Where M is the mass of the sample (kg).

3. Results and Discussion

The average radon and thoron activity concentration measured by CR-39 through the long-tube chamber were shown in Table 1 varies from (21.32±4.61 Bq.m^{-3} to 94.1±9.7 Bq.m^{-3}) and (7.10±2.66 Bq.m^{-3} to 78±8.86 Bq.m^{-3}) respectively, as in Fig.(5,7). The corresponding exhalation rate values in terms of area and mass varies from (4.79 to 29.178 mBq.m^{-2}.h^{-1}) and from (0.145 to5.29 mBq.Kg^{-1}.h^{-1}) for Radon and from (1.59 to 21.178 mBq.m^{-2}.h^{-1}) and from (0.055to 1.29 mBq.Kg^{-1}.h^{-1}) for thoron for materials under investigation, as in table .2 . Jordan paint demonstrate the highest mean area and mass exhalation rates for radon were (94.1±9.7 Bq.m^{-3} ,29.17 mBq.m^{-2}.h^{-1}, 5.29 mBq.Kg^{-1}.h^{-1}) while for thoron were (78±8.86 Bq.m^{-3}, 21.178 mBq.m^{-2}.h^{-1} and 1.29 mBq.Kg^{-1}.h^{-1}) respectively.

The lowest mean area and mass exhalation rates for radon was registrars for Iraqi plaster where (21.32±4.61 Bq.m^{-3} , 4.79 mBq.m^{-2}.h^{-1} and 0.145 mBq.Kg^{-1}.h^{-1}) respectively, while the lowest mean area and mass exhalation rates for thoron was registrars for Iraqi plaster where (7.10±2.66 Bq.m^{-3} , 1.59 mBq.m^{-2}.h^{-1} and 0.055 mBq.Kg^{-1}.h^{-1}) respectively as shown in Figs.(9).

Table (1) shows the results obtained for radon and thoron activity concentration measured by two methods passive method using CR-39 and active method using RAD7 solid state detector. The agreement between the results of calculation of the two methods was good for radon activity concentration measurement in all samples under the study while for thoron activity concentration measurement this agreement is not quite clear. The fluctuation of thoron data-measured by RAD7 solid state detector may be coming to non uniform of internal concentration of thoron gas inside the long-tube volume due to the small size and low power of the fan. However, the agreement of radon concentration in this two methods.

Finally, one can notice that for all samples investigated, radon exhalation rates value was found to be quite lower than the world average value (1250 mBq.m^{-2}.h^{-1}) which reported in the UNSCEAR report [14, 15]. Hence it is concluded that materials under the study may be used for construction purposes as they do not pose any health hazards due to low exhalation rate of radon and thoron.

For the calculation of the overall uncertainty, the following sources of errors were considered:

Counting statistics, counting statistics of the background and uncertainty of the detector calibration.

Table 1. Radon and thoron
Concentration for the some Building Materials

Sample Code	**Sample**	**Country of Origin**	**Radon activity concentration (Bq.m^{-3})**		**Thoron activity concentration (Bq.m$^{-3)}$**
			measured by CR-39 detector	measured by RAD7	measured by CR-39detector
1	Brick	Iran	46.77±6.83	6.38±40.82	5.16±26.72
2	Brick	Iraq	29.47±5.42	5.08±25.875	18.94±4.35
3	Secondary roof	Morocco	9.38±88.43	83.225±9.12	47.06±6.86
4	Dye	Iran	7.46±55.68	45.6±6.75	39.77±6.30
5	Ceramic	India	38.18±6.17	29.45±5.426	5.35±28.63
6	Ceramic	Turkish	28.43±5.33	20.75±4.55	14.21±3.77
7	Burke	Turkish	5.33±28.43	25.3±5.02	15.66±3.77
8	Dye	China	46.61±6.82	22.45±4.73	16.27±4.03
9	Plaster	Iraqi	4.61±21.32	14.51±3.80	7.10±2.66
10	Ceramic	Spanish	7.75±60.1	54.6±7.38	34.7±5.89
11	Dye	Jordanian	9.7±94.1	93.52±9.67	78±8.86
12	Burke	Iraqi	4.91±24.15	23.05±4.80	16.1±4.01
13	Ceramic	Iranian	5.41±29.37	21.11±4.95	18.35±4.28
14	Halan stone	Iraqi	5.81±33.8	32.05±5.66	25.3±5.02
15	Cement	Turkish	7.54±57	56.02±7.48	32.59±5.70
16	Dust	Iraqi	4.99±24.97	15.74±3.96	12.49±3.53
17	Granit	Chinese	7.06±49.86	48±6.92	19.95±4.46
18	Cement	Iraqi	37.46±6.12	29.35±5.41	24.79±4.99
19	Soil	Iraqi	68.78±8.29	53.1±7.28	39.30±6.62
20	Ceramic	Iranian	4.74±22.52	17.94±4.18	15.01±3.87

Figure 5. Bar diagram showing variation in radon concentration for building materials samples measured by CR-39.

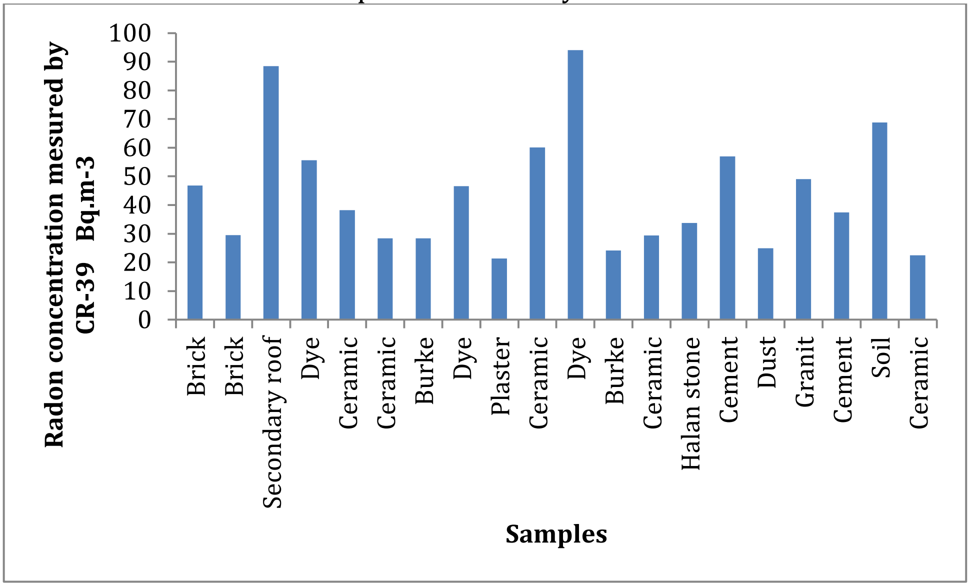


Figure 6. Bar diagram showing variation in radon concentration for building materials samples measured by RAD7.

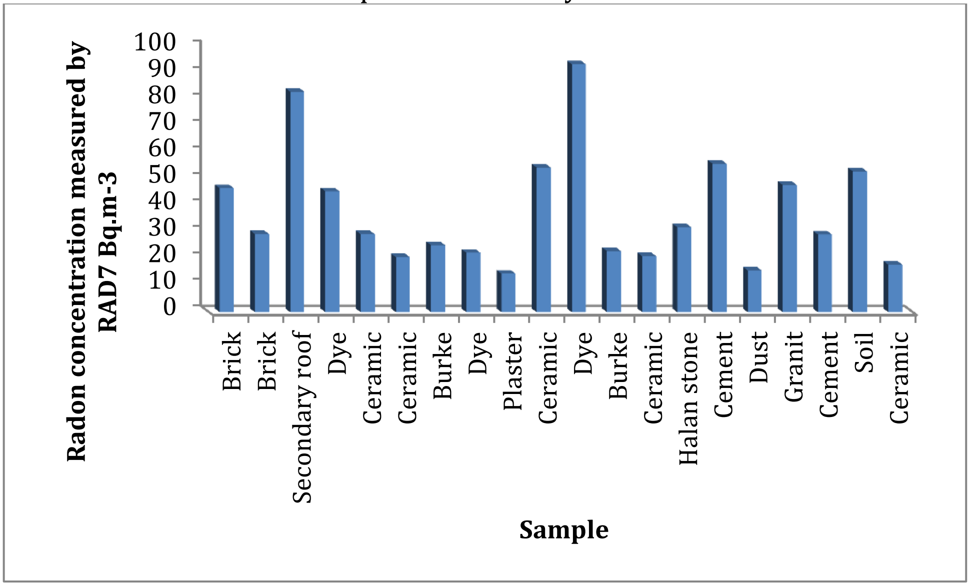


Figure 7. Bar diagram showing variation in thoron concentration for building materials samples measured by CR-39

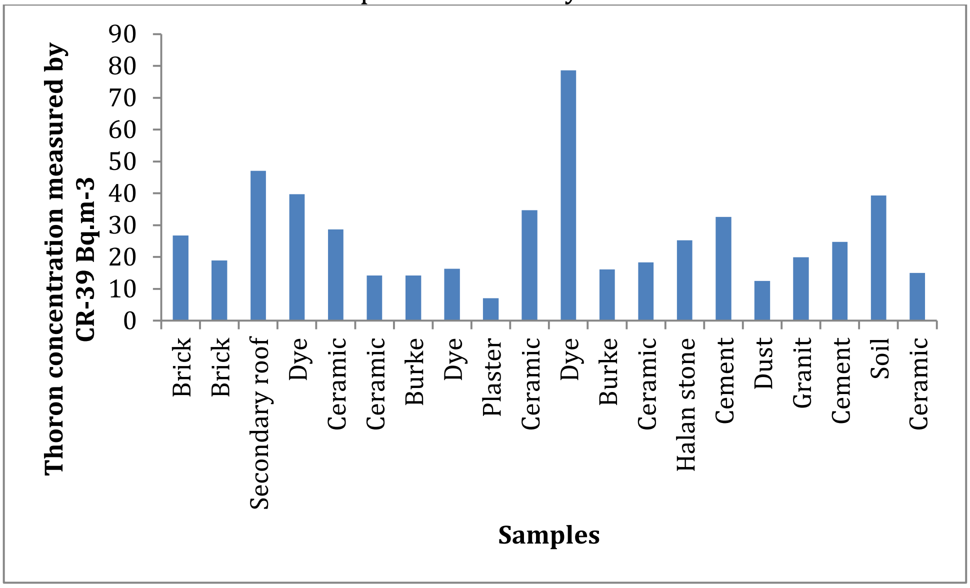


Table 2: Radon and thoron exhalation rates of the studied building material samples

Sample Code	**Radon exhalation rate by CR-39**		**Thoron exhalation rateby CR-39**	
	EA (mBq.m^{-2}.h^{-1})	EM (mBq.kg^{-1}.h^{-1})	EA (mBq.m-2.h^{-1})	EM (mBq.kg^{-1}.h^{-1})
1	10.53675	0.326622	6.012	0.186362
2	6.63075	0.3359	4.2615	0.215897
3	19.89675	0.23059	17.685	1.154021
4	12.528	0.419416	8.94825	0.299572
5	8.5905	0.254411	6.44175	0.190775
6	6.39675	0.217942	3.19725	0.108933
7	6.39675	0.273639	3.19725	0.136771
8	10.48725	0.630874	3.66075	0.220217
9	4.797	0.145	1.5975	0.055913
10	13.5225	0.379458	7.8075	0.219088
11	29.178	5.298347	21.17	1.29
12	5.43375	0.252047	3.6225	0.168032
13	6.60825	0.201919	4.12875	0.126156
14	7.605	0.207654	5.6925	0.155434
15	12.825	0.436958	7.33275	0.249833
16	5.61825	0.73256	2.81025	0. 72614
17	20.04525	0.617394	10.0125	0.308385
18	11.2185	0.269945	4.48875	0.108011
19	8.4285	0.228519	5.57775	0.151228
20	15.4755	0.44036	8.8425	0.251616

Figure 8. Radon exhalation rates (in term of area and mass) of the studied samples.

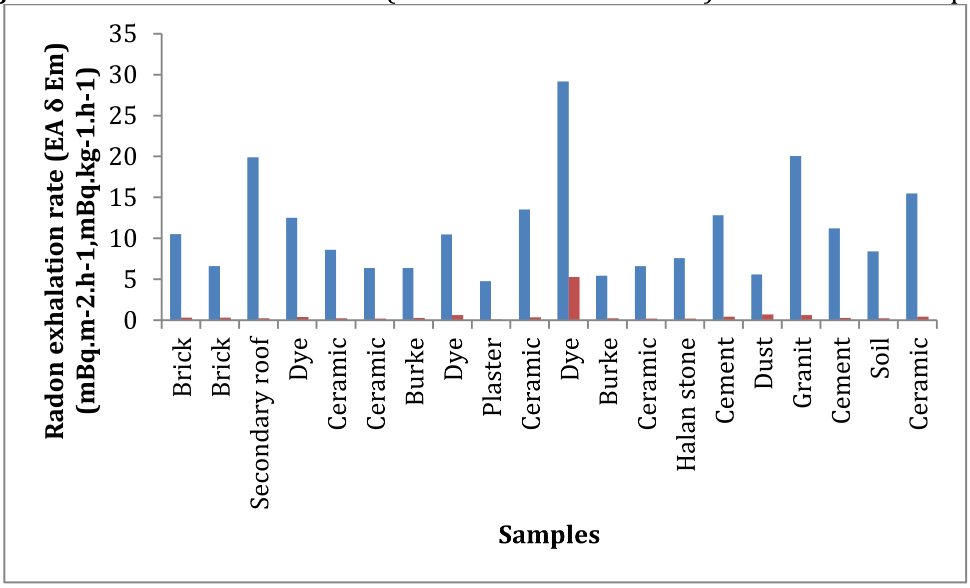
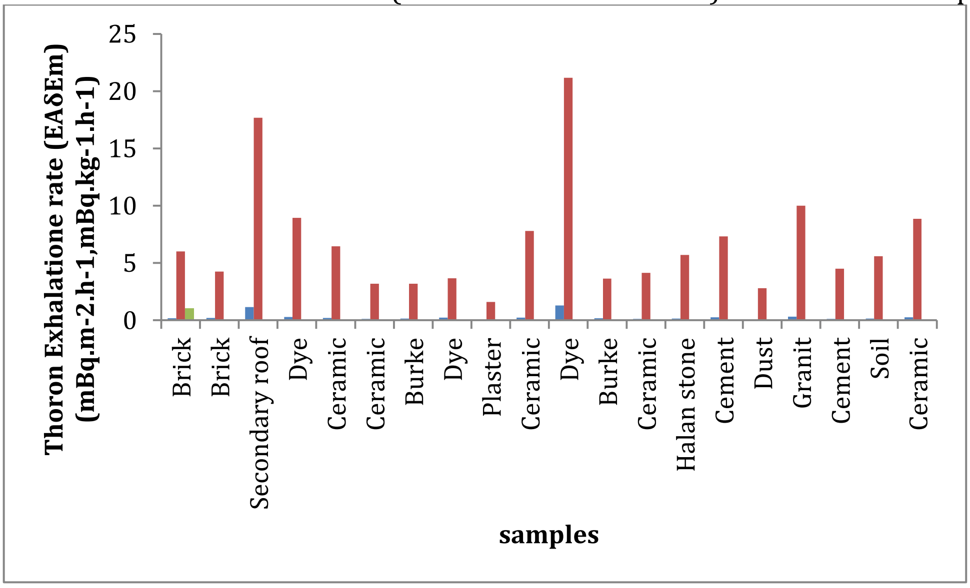


Figure 9. Thoron exhalation rates (in term of area and mass) of the studied samples.



4. Conclusion

In this study we concluded the following knowledge about measurements of radon and thoron concentration in different types of building materials which are used in Kirkuk governorate, using passive detector typeCR-39 nuclear track detectors and active detector type RAD7:

- The lowest and highest values of the radon concentration , radon exhalation rate(interm of area and mass from building material samples measured by CR-39 plastic track detector have been recorded for (Plaster Iraqi and Dye Jordanian), while the lowest and highest values of the thoron concentration and thoron exhalation rate(in term of area and mass) from building material samples measured by CR-39 plastic track detector have been recorded for samples of Plaster Iraqi and Dye Jordanian).

- Good agreement between radon activity concentrations measurement was obtained using passive CR-39 plastic track detector and active RAD7 solid state detector, while no such agreement noticed in thoron activity concentration measurement.

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