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Measurement of Radium Content and Radon Exhalation Rates in Egyptian Construction Material Samples Using the Sealed-Cup Technique and LR-115 Detectors

Ahmed Saad Hussein[12*]

[1]Nuclear Security Program, Naif Arab University for Security Studies, Riyadh, KSA.
[2]Radiation Protection Department, Nuclear Power Plants Authority, Cairo, Egypt.

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Abstract: As a potential source of indoor radiation arising from radon, a range of construction material samples were collected from the local market in Egypt (black cement, white cement, gypsum, sand, clay brick, red brick, cement brick, marble, limestone, and ceramic) and studied for radium content, radon exhalation rates, and alpha index. The sealed–cup technique based on LR-115 nuclear track detectors was used. Radium content was found to vary from 1.93 (Gypsum S5) to 360.21 (Cement brick S13) Bq kg^{-1} with a mean value ***of*** 68.76 Bq kg^{-1}. Mass exhalation rate was found to vary from $5.75x10^{-9}$(Gypsum S5) to $1073.43x10^{-9}$ (Cement brick S13) Bq $kg^{-1}h^{-1}$ with a mean value of 206.70 x10-9 Bq $kg^{-1}h^{-1}$. Area (surface) exhalation rate varied between $0.30x10^{-6}$(Gypsum S5) and $55.80x10^{-6}$ (Cement brick S13) Bq $m^{-2}h^{-1}$ with a mean value of 10.65 x 10^{-6} Bq $m^{-2}h^{-1}$. Alpha index varied from 0.01 (Gypsum S5 & S6) to 1.8 (Cement brick S13) with a mean value of 0.34. These results were shown to be less than their corresponding world limits except for cement brick. The radium content and both mass and area exhalation rates in the studied samples do not pose a risk to human health except for cement brick.

Keywords: Radium content, area, and mass exhalation rates, construction material, Egypt, LR-115 detectors.

1 Introduction

Radium (^{226}Ra) and radon (^{222}Rn) are associated with naturally occurring uranium (^{238}U), present in all types of rocks, soil, construction materials, and groundwater. The radium content of a sample contributes to the level of environmental radon because radon is produced directly from the natural alpha decay of parent ^{226}Ra. Higher values of ^{226}Ra contribute significantly to enhanced levels of indoor radon and radon decay progeny [1]. Radon and its progeny in the indoor air environment contribute more than 50% of the annual natural background radiation dose to people in modern society [2-4].

High concentrations of radon and its progeny have been shown to contribute to lung cancer, bone cancer, sores, lymphoma, kidney damage, born defects, cataract formation, leukemia, and anemia [5-7]. Nowadays, one may assume that most inhabitants in urban cities typically spend about 80% of their time indoors, which means that indoor exposure to radon and its progeny from construction materials could be relevant to health [3]. Under such conditions, it is important to ensure that levels of radium and radon in dwellings are safe by modern standards, especially when planning future construction [3,8]. The soil under the dwellings is the main radon source in the indoor air. Secondary sources include radon exhalation from naturally occurring radium in wall materials that may infiltrate into living spaces. Indoor levels are highly influenced by ventilation. The term exhalation designates the rate of escape of radon from a material to the atmosphere. Radon exhalation is a complex phenomenon depending upon parameters such as ^{226}Ra content in construction materials, sample moisture, sample grain size, temperature, and atmospheric pressure [9].

Recent public concerns about radium and radon concentrations in construction materials have been addressed by measurements using passive measuring techniques [10-12]. Numerous studies in Egypt have been conducted to measure radon concentration in construction materials and their radon exhalation rates using similar passive measuring techniques [13-18].

The purpose of the present study was to measure radium content, radon exhalation rates (surface and mass), and

*Corresponding author e-mail: aomar@nauss.edu.sa

alpha index of twenty samples in ten types of the most used construction materials collected from the local market in Egypt. The measurements were performed using a standard sealed-cup technique with LR-115 nuclear track detectors.

The sealed-cup technique with the LR-115 nuclear track detector is an efficient, cheap, and useful tool for evaluating long-term radon exposures. Results from this study could be valuable for assessing the radiological characteristics of construction materials and the implications for public health and safety.

2 Materials and methods

In the present investigation, the "sealed-cup technique" was used to study the radium content and radon exhalation rates [1,9,13,19,20]. Different construction material samples were collected from the local market in Egypt. The samples were dried at room temperature to 1mm grain size, each sample was divided into five equal volumes, weighed (200 gm), and then placed in cylindrical containers made of aluminum. Each sample container was capped tightly within an inverted cylindrical cup of 3.5 cm radius and 11 cm high. A piece of LR-115 type II (Kodak Pathé, France).

detector with an area of 2.25 cm2 was affixed to the top center of the inverted cup. The experimental arrangement is shown in Figure 1. The cups were left undisturbed at room temperature for three months of exposure time. During this time alpha particles bombarded the LR-115 detector inside the inverted cup through the alpha-decay of the samples.

After the irradiation period, the irradiated detectors were collected and chemically etched in 2.5 M NaOH solution at 60 ^oC for 2 hours. The etching was carried out to reduce the thickness of the LR-115 detectors to about 5 µm [19]. After etching, the LR-115 detectors were washed in distilled water using an ultrasonic cleaner and dried in air.

Subsequently, alpha particle tracks were counted using an optical microscope. The background of the LR-115 detector was counted and subtracted from the count of all detectors.

The corrected track density was converted into Bq m-3 using a calibration constant (the sensitivity factor). The calibration constant η for radon measurement using a cylindrical cup equipped with LR-115 nuclear track detectors was reported by several authors [1,13,21,22].

It can be estimated experimentally or theoretically. The radon measuring devices were calibrated at the National Institute for Measurements and Standards (NIS), Cairo, Egypt. The calibration coefficient adopted in the present work for LR-115 was $\eta = 0.034 \pm 0.003$ α- tracks $cm^{-2} d^{-1}$/ Bq m^{-3} of radon [13,23,24].

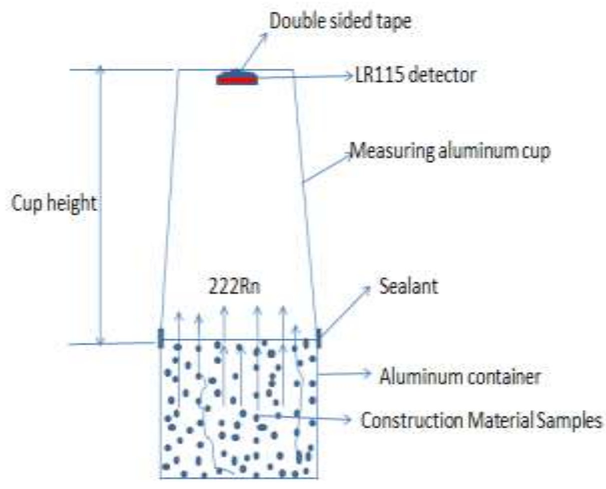

Figure 1 : Schematic diagram of the passive radon device.

Fig.1. Schematic diagram of the passive radon device.

The radon activity concentration (C_{Rn}) is calculated by using the equation [21,22,25]:

$$C_{Rn} = \rho/(\eta\ t) \quad (1)$$

where ρ the track density (tracks cm^{-2}) , η the calibration coefficient(the sensitivity factor) of LR-115 detector(tracks cm^{-2} d^{-1} / Bq m^{-3}) and t the exposure time (d).The value of η depends on the height and radius of the measuring cylinder cup[13]. An effective equilibrium (about 98%) for members of the radon decay series is reached in about 30 days [26]. Once radioactive equilibrium is established, one may use the radon alpha analyses for the determination of the steady-state activity of radium. The radium content (C_{Ra}) is calculated by using the equation:

$$C_{Ra} = C_{Rn}/((1- e\text{-}(-\lambda_{Rn}\ t))) \quad (2)$$

The mass exhalation rate (E_M) of the sample release of the radon can be calculated by using the expression [9]:

$$E_M\ (Bq\ kg^{-1}\ d^{-1}) = C_{Ra}\ (\lambda_{Ra}/(\lambda_{Rn}))\ 1/T_e \quad (3)$$

Where λ_{Ra}, λ_{Rn}, and T_e are constant radium decay, radon decay, and the effective exposure time, respectively. The effective exposure time Te given by:

$$T_e = [\ t - 1/(\lambda_{Rn})\ (\ 1 - e\text{-}(-\lambda_{Rn}\ t\ ))] \quad (4)$$

The surface exhalation rate (E_A) of the sample for the release of radon can be calculated by using the expression [9] :

$$E_A(\ Bq\ m^{-2}\ d^{-1}) = E_M\ (\ M/A\ ) \quad (5)$$

Where M is the mass of the sample in kg and A the area of the cross-section of the cylindrical cup in m^2.

The excess alpha radiation resulting from the radon inhalation and originating from the construction materials is assessed through the alpha index (Iα) which is defined as follows [27,28]:

$$I_\alpha = (C_{Ra})/200 \qquad (6)$$

The recommended exemption and upper levels of ^{226}Ra concentrations in construction materials are 100 Bq kg^{-1} and 200 Bq kg^{-1} respectively. The recommended limit concentration of ^{226}Ra is 200 Bq kg^{-1}, for which Iα=1 [27].

3 Results and Discussion

Table 1 shows measured values for radium content and the mass and surface exhalation rates for construction materials collected from the local market in Egypt. The lowest value of radium content was 1.93±0.10 Bq kg^{-1} in gypsum sample S5, while the highest value was 360±43.2 Bq kg^{-1} in cement brick S13. These variations may be ascribed to the different radioactive contents of the materials. Other factors that may also have been crucial, include the emanation factor, the diffusion coefficient of radon in those materials, and the porosity, as well as the density of the materials. Radium content values were much lower than the maximum permissible value of 370 Bq kg^{-1} for building materials recommended by the Organization for Economic Cooperation and Development [3,29]. Figure 2 shows the comparisons between the values of radium consecration for different construction materials.

The radon mass and area exhalation rates were found to vary from 5.75×10^{-9}(Gypsum S5) to 1073×10^{-9} (Cement brick S13) Bq $kg^{-1}h^{-1}$ and 0.30×10^{-6} (Gypsum S5) to 55.8×10^{-6} (cement brick S13) Bq $m^{-2}h^{-1}$, respectively. A significant correlation can be seen between radon mass and area exhalation rates versus the radium content of the studied samples (Table 1). The overall values of radon area exhalation rate were less than and below the world average value of 57.6 Bq $m^{-2}h^{-1}$[3]. The results show that this construction material is safe as far as the health hazards of radium are concerned, with levels for cement brick samples approaching world average values.

Figures 3 and 4 present the radium concentration vs mass and area exhalation rates. The relations in both figures are linear between the concentration values of radium and the exhalation rates (mass and area).

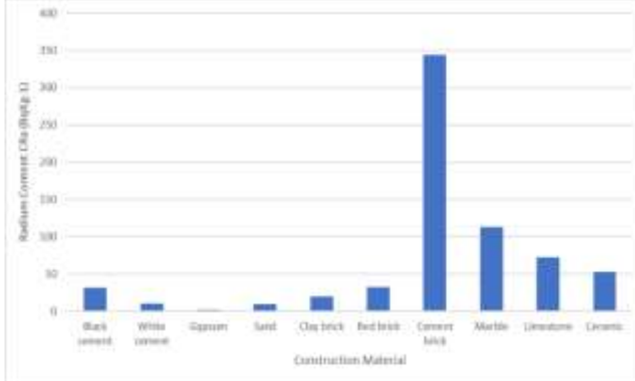

Fig.2. The relation between the sample name and radium concentration (C_{Ra}) of the construction materials.

Table 1 also represents the alpha index I_α where the values range from 0.01(Gypsum S5) to 1.80(Cement brick S13). These values, which were less than the unity values, indicated that the construction materials are safe from the environmental radiation hazards point of view; except possibly for cement brick samples where I_α =1.64 and 1.80. Therefore, reducing the use of cement bricks whenever possible is recommended.

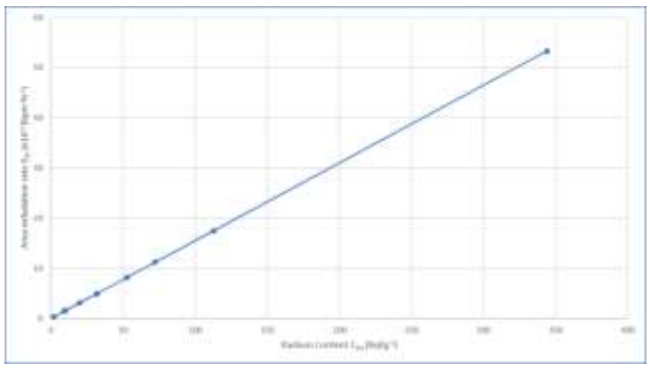

Fig. 3. The correlation between radium concentration (C_{Ra}) and mass exhalation rate (E_M) of construction materials.

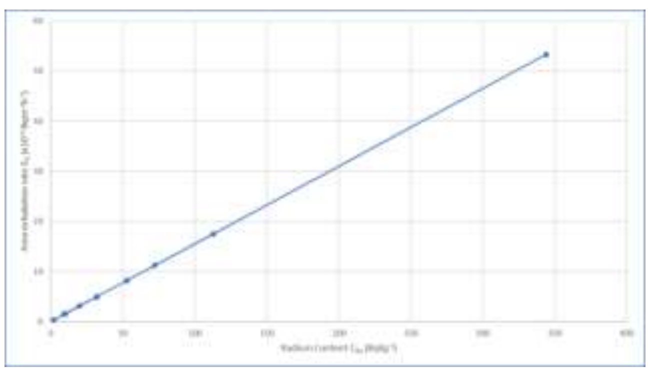

Fig. 4. The correlation between radium concentration (C_{Ra}) and area exhalation rate (E_A) of the construction materials.

Table 1: Radium content (C_{Ra}), mass exhalation rate (E_M), area exhalation rate (E_A) of, and Alpha index(I_α)in construction material samples in Egypt.

Sample Type		C_{Ra} (Bq kg^{-1})	Radon Exhalation		I_α
			E_M(Bq kg^{-1}h^{-1}) x10^{-9}	E_A(Bq m^{-2}h^{-1}) x10^{-6}	
Black cement	S1	29.80±3.33	88.80	4.61	0.15
	S2	33.70±4.38	100.00	5.21	0.17
White cement	S3	11.20±1.15	33.40	1.74	0.06
	S4	9.18±1.06	27.40	1.42	0.05
Gypsum	S5	1.93±0.10	5.75	0.30	0.01
	S6	2.05±0.12	6.11	0.32	0.01
Sand	S7	7.84±0.97	23.40	1.21	0.04
	S8	10.70±1.11	31.80	1.65	0.05
Clay brick	S9	18.50±2.23	55.10	2.86	0.09
	S10	21.50±2.98	64.20	3.33	0.11
Red brick	S11	26.00±3.88	77.60	4.03	0.13
	S12	38.30±4.94	114.00	5.93	0.19
Cement brick	S13	360.00±43.20	1073.00	55.80	1.80
	S14	328.00±41.30	978.00	50.80	1.64
Marble	S15	117.00±15.00	384.00	18.10	0.58
	S16	109.00±14.00	326.00	16.90	0.55
Limestone	S17	78.60±7.86	234.00	12.20	0.39
	S18	66.30±7.16	197.00	10.30	0.33
Ceramic	S19	49.10±5.40	146.00	7.60	0.25
	S20	56.40±6.15	168.00	8.73	0.28
Average		**68.76±9740**	**206.70±291.37**	**10.65±15.10**	**0.34**

Table 2: Comparison of radium content (C_{Ra}) in construction material samples from different countries using different techniques.

Material type	C_{Ra} Bq kg^{-1}	Used technique	Country	References
Black Cement	31.70	LR-115 detector.	Egypt	Present work.
	27.70	LR-115 detector.	Egypt	13
	77.30	LR-115 detector.	Egypt	15
	0.45	CR-39 detector.	Iraq	30
	0.44	CR-39 detector.	Saudi Arabia	31
	93.20	CR-39 detector.	Saudi Arabia	32
White cement	10.20	LR-115 detector.	Egypt	Present work.
	10.40	LR-115 detector.	Egypt	13
	29.00	LR-115 detector.	Egypt	15
	17.50	HPGe detector.	Egypt	16
	0.18	CR-39 detector.	Iraq	30
	0.26	CR-39 detector.	Saudi Arabia	31
	1.63	.CR-39 detector.	Saudi Arabia	32
	33.00	HPGe detector.	Malaysia	33
Gypsum	1.99	LR-115 detector.	Egypt	Present work.
	1.83	LR-115 detector.	Egypt	13
	8.15	HPGe detector.	Egypt	16
	52.00	LR-115 detector.	Egypt	15
	0.07	CR-39 detector.	Iraq	30

Material type	C_{Ra} Bq kg^{-1}	Used technique	Country	References
	16.70	CR-39 detector.	Saudi Arabia	31
	2.02	CR-39 detector.	Saudi Arabia	32
	33.30	NaI(Tl) detector.	Saudi Arabia	34
	73.90	CR-39 detector.	Saudi Arabia	12
Sand	9.23	LR-115 detector.	Egypt	Present work.
	8.52	LR-115 detector.	Egypt	13
	139.00	LR-115 detector.	Egypt	15
	9.25	Alpha GUARD.	Egypt	17
	16.60	HPGe detector.	Egypt	16
	59.90	CR-39 detector.	Saudi Arabia	31
	12.30	NaI(Tl) detector.	Saudi Arabia	34
	3.93	NaI(Tl) detector.	India	35
	38.00	HPGe detector.	Malaysia	33
Clay brick	20.00	LR-115 detector.	Egypt	Present work.
	17.50	LR-115 detector.	Egypt	13
Red brick	20.00	LR-115 detector.	Egypt	Present work.
	23.10	HPGe detector.	Egypt	16
	174.00	NaI(Tl) detector.	India	35
	33.00	HPGe detector.	Malaysia	33
Cement brick	344.00	LR-115 detector.	Egypt	Present work.
	351.00	LR-115 detector.	Egypt	13
	288.00	HPGe detector.	Egypt	16
	181.00	NaI(Tl) detector.	India	35
	46.00	HPGe detector.	Malaysia	33
Marble	113.00	LR-115 detector.	Egypt	Present work.
	0.25	Alpha GUARD.	Egypt	17
	0.49	CR-39 detector.	Iraq	30
	3.50-19.00	CR-39 detector.	Syria	36
	3.40	CR-39 detector.	Saudi Arabia	32
	12.70	NaI(Tl) detector.	Saudi Arabia	34
	70.10	CR-39 detector.	Saudi Arabia	12
Limestone	72.40	LR-115 detector.	Egypt	Present work.
	0.65	Alpha GUARD.	Egypt	17
	1.63	CR-39 detector.	Saudi Arabia	32
	28.60	NaI(Tl) detector.	Saudi Arabia	34
	10.00	HPGe detector.	Malaysia	33
Ceramic	52.80	LR-115 detector.	Egypt	Present work
	51.10	HPGe detector.	Egypt	16
	53.00-75.00	CR-39 detector.	Syria	36
	2.79	CR-39 detector.	Saudi Arabia	32
	47.80	CR-39 detector	Saudi Arabia	33

Much information has been published regarding radium content for construction materials. A large discrepancy has been seen in the reported values of radium content (Table 2). This discrepancy is attributed to variations in the radioactive content of these materials. Most of the radium content values and the radon exhalation rates in the samples studied were found to be consistent with data obtained by other investigators [13,16].

4 Conclusions

It is concluded that these materials may be used for construction purposes, and they do not pose any significant health hazards. It is important to study radon exhalation rates and to understand the contribution of the construction material towards total radon concentrations in dwellings. It can be concluded that the studied construction materials are safe from a radiological health perspective. It is necessary to reduce the use of cement bricks as much as possible. It is possible to establish a national database for construction materials available in a local market in Egypt using this technique with low cost for large-scale screening measurements.

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