

## MEASUREMENTS OF NATURAL RADIOACTIVITY IN SOME KINDS OF MARBLE AND GRANITE USED IN RIYADH REGION

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Thirty seven samples of marble and twenty samples of granite from both local and foreign locations were collected from different factories in Riyadh Region. The samples were crushed, dried in controlled furnace for around twenty four hours, and then stored for four weeks in ½ l plastic Marinelli beakers. The gamma radiation of the samples was measured, employing high resolution  $\gamma$ - ray spectroscopy with an accumulating time for about 80000 sec. each. From the measured  $\gamma$ -ray spectra, activity concentrations were determined for marble samples  $^{238}\text{U}$  (0.71- 44.1Bq/kg),  $^{232}\text{Th}$  (0.10-32.0 Bq/kg),  $^{226}\text{Ra}$  (0.36-32.4 Bq/kg) and  $^{40}\text{K}$  (0.68-897.1 Bq/kg) while for granite  $^{238}\text{U}$  (0.03-314.4 Bq/kg),  $^{232}\text{Th}$  (0.02-186.4 Bq/kg),  $^{226}\text{Ra}$  (0.03-147.0 Bq/kg) and  $^{40}\text{K}$  (0.28 – 1531.7 Bq/kg). The radium equivalent activity, radiation hazard indices and absorbed dose rate for all samples were estimated.

**Keywords:** *Radioactivity Concentration, Radium Equivalent, Marble, Granite, Hazard, Annual Dose Rate, Natural Radioactivity.*

### INTRODUCTION

The knowledge of the natural radioactivity of building materials is important for the determination of population exposure to radiations, as most of the residents spend about 80% of their time indoors [1,2]. The presence of the radioisotopes in materials causes external exposure to the people who live in the building.  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  can also increase the concentration of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  and of its daughters in the building.  $^{40}\text{K}$  and part of the above-mentioned radionuclides cause external exposure while the inhalation of  $^{222}\text{Rn}$ ,  $^{220}\text{Rn}$  and their short lived progeny leads to internal exposure of the respiratory tract to alpha particles [3,4]. Due to the health risks associated with the exposure to indoor radiation, many governmental and international bodies such as the

International Commission on Radiological Protection (ICRP)[5], the World Health Organization (WHO) etc. have adopted strong measures aimed at minimizing such exposures. In this context, limits have been set on the concentrations of radionuclides in various building materials and the use of materials with abnormally high levels of radioactivity has been banned. Because of its polished surface and its availability in a variety of attractive colors, marble is widely used as a building/construction material. It is mostly used as a lining on walls and floors in dwellings. The presence of other minerals in marble gives it a variety of attractive colors. However, granites, usually suitable as building and ornamental materials for interior and exterior use, are hard natural stones that require harder tools to be cut, shaped and polished, compared to marble. Usually marble and granites have a commercial name, petrographic and technological characterizations, and identification of the producer country. The objective of the present study is to determine the specific radioactivity concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$  and  $^{40}\text{K}$  in some kinds of marble and granite used in Riyadh region aiming to contribute and establish a baseline map of environmental radioactivity levels. The average radium equivalent activity ( $Ra_{eq}$ ), the total absorbed dose rate ( $D$ ), the external ( $H_{ex}$ ), internal ( $H_{in}$ ) and the representative ( $I_{yr}$ ) hazard indices have been estimated and compared with the recommended limits from UNSCEAR data. Additionally, the correlations between thorium, uranium and potassium will be shown in this paper with an aim to correlate the petrographic characteristics of commercial marble and granites with their corresponding dose rates for natural radioactivity. These results are of great interest in the environmental radiological protection study, since marble and granites are widely used as building and ornamental materials, including indoor covering. Local authorities can limit the use of building materials that cause a significant increase in radiation exposure due to higher levels of indoor radon, and external gamma exposure. According to international recommendations quoted in the Basic Safety Series No. 115 from the IAEA, the use of building materials containing enhanced concentrations of NORM should be controlled and restricted under the application of the radiation safety standards. Several publications deal with measuring low levels of naturally occurring radioactive isotopes in marble and granite rocks [6-16].

## MATERIAL AND METHOD

Saudi Arabia is becoming a relatively large market for local and foreign marble and granite usage. New practices have been introduced in house construction that includes usage of granite tiling of houses. Even when old houses are renovated conventional tiles are replaced with marble and granites.

A total of 37 different kinds of marble and 20 different kinds of granites were collected from local suppliers. These samples either local or imported are widely used as building and ornamental materials in Riyadh region. A description of the sample preparations method can be found in [17]. Radioactivity in marble and granite samples was measured using a low-level counting system consisting of a high-purity Ge detector (HPGe). It is explained elsewhere [18].

The concentration of  $^{238}\text{U}$  was calculated from the activity of  $^{234}\text{Th}$  (63.29 keV) with an emission percentage of 4.471%. While for  $^{226}\text{Ra}$  was determined by measuring its gamma emitters  $^{214}\text{Pb}$  (295.213 and 351.921 keV) with an emission percentage of 19.3% and 37.6% respectively and  $^{214}\text{Bi}$  (609.312 keV) with an emission percentage of 46%. To calculate the concentration of  $^{232}\text{Th}$  the activity of the  $^{228}\text{Ac}$  ( $T_{1/2} = 6.15$  h) lines at 338.4 and 911.21 keV with an emission percentage 11.1% and 26.6%, respectively were used. The concentration of the  $^{40}\text{K}$  was calculated using the gamma line at energy 1460.75 keV of emission percentage 10.7%. The total uncertainty ( $\sigma_{\text{tot}}$ ) of the calculated activities is composed of the counting statistical ( $\sigma_{\text{st}}$ ) and weighted systematic error ( $\sigma_{\text{sys},i}$ ) calculated by the following formula [19].

$$\sigma_{\text{tot}} = \left( \sigma_{\text{st}}^2 + \frac{1}{3} \sum_i \sigma_{\text{sys},i}^2 \right)^{\frac{1}{2}} \quad (1)$$

The minimum detection limits of the setup for  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$  and  $^{40}\text{K}$  were measured to be 0.023, 0.019, 0.014 and 0.167 Bq/kg, respectively.

## RESULTS AND DISCUSSION

More than 60 samples were investigated. About 18 resolved energy photo peaks were observed. Eight photo peaks were from the uranium series, eight were from the thorium series and one was from  $^{40}\text{K}$ . There were more than one sample for some marble types. The activity concentrations of such samples were averaged. The average radionuclide activity concentration in Saudi and foreign marble samples are reported in Table (1). The maximum activity value of  $^{238}\text{U}$  was in sample (m-34) (Light Emprado)  $44.1 \pm 0.1$  Bq/kg and the minimum value was in sample (m-10) (Paly sandro grey)  $0.71 \pm 0.04$  Bq/kg. While for  $^{232}\text{Th}$  the maximum level was observed in sample (m-3) (Guatemala green)  $32.0 \pm 0.1$  Bq/kg and the minimum level in sample (m-8) (Cecelia perlato)  $0.10 \pm 0.03$  Bq/kg. The result given in maximum concentrations of  $^{226}\text{Ra}$  was in sample (m-34) (Light Emprado)  $32.40 \pm 0.6$  Bq/kg and the minimum value was in sample (m-4) (Guatemala grey)  $0.36 \pm 0.06$  Bq/kg. The value of  $^{40}\text{K}$  ranged from  $897.1 \pm 0.1$  Bq/kg in sample (m-3) (Guatemala green) to  $0.68 \pm 0.05$  Bq/kg in sample (m-10) (Paly sandro grey).

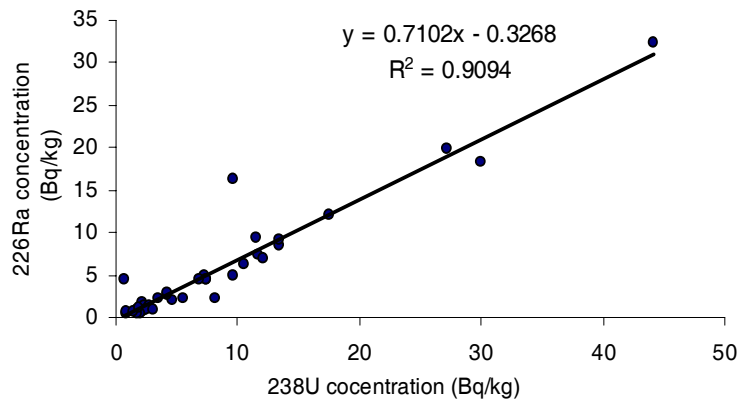
In order to observe a correlation between uranium and radium in marble a graph was plotted Figure 1, and a good correlation coefficient was determined (0.9094) which indicates the secular equilibrium between the parent and the daughter except for the samples (m-3, m-5, m-10 and m-33). Figure 2 represents good linear correlation (correlation coefficient=0.948) between thorium and potassium concentrations except for the samples (m-3, m-20, and m-35).

The activity concentrations for granite are reported in Table (2). Maximum concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$  and  $^{40}\text{K}$  were  $314.4 \pm 0.4$ ,  $186.4 \pm 0.2$  Bq/kg,  $147.0 \pm 0.3$  Bq/kg for (G-18) (Azoles bahea) and  $1531.7 \pm 0.1$  Bq/kg for (G-13) (Carmen red) respectively.

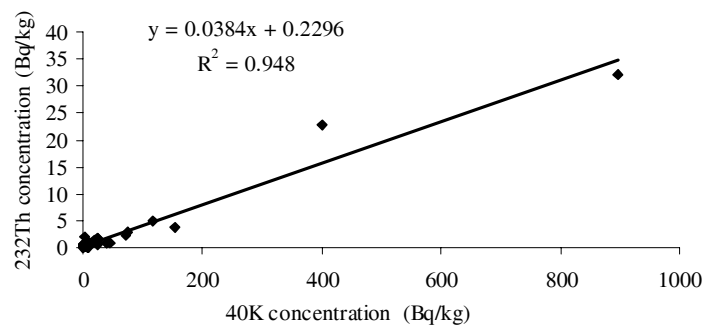
**Table 1.** Activity concentration for different marble samples (Bq/kg).

Sample No.	Commercial name	Origin	Activity (Bq/kg)			
			<sup>238</sup> U	<sup>232</sup> Th	<sup>226</sup> Ra	<sup>40</sup> K
m-1	Marble	Saudi	17.50±0.1	1.70±0.1	12.00±0.2	23.10±0.1
m-2	Cream marvel	Iran	13.50±0.1	0.18±0.05	8.50±0.2	3.30±0.1
m-3	Guatemala (green)	India	29.90±0.01	32.0±0.1	18.20±0.1	897.10±0.1
m-4	Guatemala (grey)	India	0.88±0.04	0.22±0.03	0.36±0.06	3.70±0.1
m-5	Golden camel	Pakistan	9.60±0.1	0.87±0.11	16.30±0.3	7.90±0.1
m-6	Karara	Italy	3.50±0.1	0.19±0.05	2.30±0.1	6.00±0.1
m-7	Karara (pure)	Italy	2.20±0.01	0.32±0.05	1.70±0.1	6.00±0.1
m-8	Cecelia perlato	Italy	1.70±0.01	0.10±0.03	0.87±0.06	7.50±0.1
m-9	Paly sandro clasico	Italy	10.50±0.1	0.79±0.07	6.20±0.1	44.00±0.1
m-10	Paly sandro (grey)	Italy	0.71±0.04	0.56±0.05	4.50±0.2	0.68±0.05
m-11	Fir de entco	Italy	4.70±0.1	2.30±0.2	2.00±0.08	70.50±0.4
m-12	Rosa lorca	Italy	0.93±0.04	0.74±0.09	0.62±0.07	10.80±0.1
m-13	Gallo royal	Italy	13.50±0.1	2.90±0.3	9.10±0.2	74.20±0.5
m-14	Yellow broctal	Italy	7.40±0.1	2.00±0.01	4.40±0.1	2.50±0.1
m-15	Krefenento notch	Italy	2.40±0.01	1.70±0.2	1.30±0.1	23.10±0.2
m-16	Red Krefenento	Italy	2.80±0.1	0.19±0.03	1.30±0.1	2.10±0.1
m-17	Brachia damascata	Italy	7.30±0.1	0.44±0.07	5.00±0.1	23.40±0.2
m-18	Brachia	Italy	2.50±0.01	0.23±0.04	1.00±0.1	6.10±0.1
m-19	Black Marconi a	Italy	27.2±0.1	0.28±0.06	19.80±0.4	8.40±0.1
m-20	Sheboleno	Italy	4.30±0.1	3.70±0.4	2.60±0.1	154.50±0.1
m-21	Esttoaryo	Italy	4.20±0.1	0.16±0.04	2.90±0.1	6.20±0.1
m-22	Rosa	Portugal	2.20±0.01	0.75±0.08	0.65±0.06	39.50±0.3
m-23	White mola	Turkey	2.50±0.01	0.29±0.05	0.97±0.07	3.50±0.1
m-24	Afeon shegar	Turkey	2.10±0.01	0.25±0.04	0.43±0.06	2.90±0.1
m-25	Russo lefanto	Turkey	9.60±0.1	0.22±0.06	4.80±0.1	1.20±0.1
m-26	Bianco Rosa	Turkey	3.10±0.1	0.14±0.04	0.96±0.07	1.20±0.1
m-27	Crystal	Greek	1.70±0.01	0.46±0.07	0.44±0.06	1.60±0.1
m-28	Bocshyaro	Spain	11.70±0.01	0.31±0.05	7.40±0.1	4.30±0.1
m-29	Russo basin	Spain	7.50±0.01	1.40±0.1	4.50±0.1	17.80±0.1
m-30	Red fetcano	Spain	5.50±0.1	5.00±0.5	2.20±0.1	116.2±0.8
m-31	Emparo oro	Spain	11.6±0.1	1.30±0.1	9.30±0.2	18.70±0.1
m-32	Russo elicanty	Spain	1.40±0.01	0.95±0.11	0.62±0.06	25.60±0.2
m-33	Cream marvel	Spain	6.80±0.1	0.41±0.06	4.50±0.1	5.20±0.1
m-34	Light Emprado	Spain	44.10±0.1	0.99±0.12	32.40±0.6	16.40±0.2
m-35	Azoles makoba	Brazil	8.20±0.1	22.80±0.2	2.30±0.1	401.8±0.3
m-36	Yellow sena	France	1.90±0.01	0.86±0.10	1.10±0.1	39.10±0.1
m-37	Silvia (yellow) sand	Egypt	12.10±0.1	0.48±0.02	6.90±0.1	1.30±0.1

Minimum concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$  and  $^{40}\text{K}$  were  $0.03\pm 0.01\text{Bq/kg}$  for (G-6) (Saudi green),  $0.02\pm 0.01\text{Bq/kg}$  for both (G-7) (Royal gold) and (G-17) (Rosa entema),  $0.03\pm 0.01\text{Bq/kg}$  for (G-8) (spring green) and  $0.28\pm 0.03\text{Bq/kg}$  for (G-7) (Royal gold) respectively.



**Figure 1.** The correlation between  $^{238}\text{U}$  and  $^{226}\text{Ra}$  concentration in marble samples.



**Figure 2.** The correlation between  $^{40}\text{K}$  and  $^{232}\text{Th}$  concentration in marble samples.

Figure 3. shows the strong correlation between uranium and radium in granite samples under investigation (correlation coefficient=0.9898) which indicate the secular equilibrium between  $^{238}\text{U}$  and  $^{226}\text{Ra}$ . While Figure 4 shows the good correlation between the concentrations of the two radioactive isotope ( $^{232}\text{Th}$ ,  $^{226}\text{Ra}$ ) (correlation coefficient = 0.9395).

To assess the radiological risk of the building materials used, it is useful to calculate the radium equivalent activity [20, 21]

$$\text{Ra}_{\text{eq}} = (A_{\text{Th}} \times 1.43) + A_{\text{Ra}} + (A_{\text{K}} \times 0.077) \quad (2)$$

where  $A_{\text{Ra}}$ ,  $A_{\text{Th}}$  and  $A_{\text{K}}$  are the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , respectively. The calculated values of the radium equivalent  $\text{Ra}_{\text{eq}}$  for the studied

marble samples are given in Table (3). These values range from (m-4) (Guatemala grey) 0.968 Bq/kg to (m-3) (Guatemala green) 132.914 Bq/kg. The calculated values of the radium equivalent  $Ra_{eq}$  for the studied granite samples are given in Table (4).

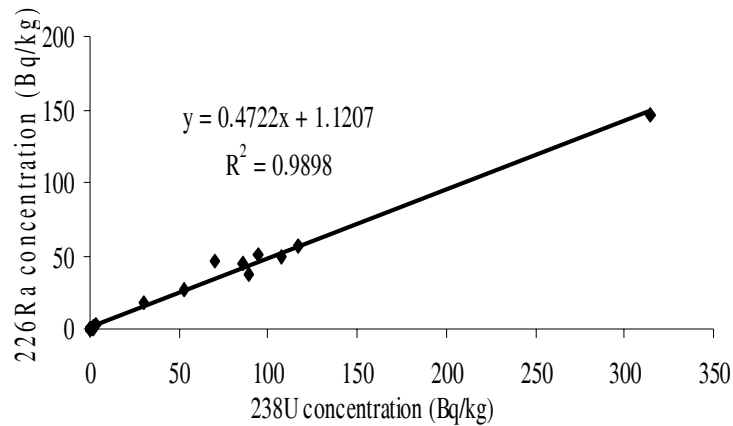
**Table 2.** Activity concentration for granite samples (Bq/kg)

Sample No.	Commercial name	Origin	Activity (Bq/kg)			
			$^{238}\text{U}$	$^{232}\text{Th}$	$^{226}\text{Ra}$	$^{40}\text{K}$
G-1	Salmon	Saudi	52.6±0.2	41.9±0.4	27.3±0.5	1484.9±0.1
G-2	Bianco sardo	Saudi	94.7±0.2	49.1±0.5	51.1±0.9	1379.0±0.9
G-3	Sweet gold	Saudi	1.9±0.1	0.56±0.02	0.82±0.05	12.3± 0.9
G-4	Najran brown	Saudi	0.14±0.04	0.10±0.03	0.10±0.03	0.91± 0.08
G-5	Saudi brown	Saudi	0.11±0.04	0.31±0.09	0.06±0.01	0.48± 0.04
G-6	Saudi green	Saudi	0.03±0.01	0.07±0.01	0.05±0.01	0.44± 0.04
G-7	Royal gold	Saudi	0.05±0.01	0.02±0.01	0.06±0.03	0.28± 0.03
G-8	Spring green	Saudi	0.05±0.03	0.06±0.01	0.03±0.01	0.51± 0.05
G-9	Yezoa osoloto	India	3.8±0.1	3.1± 0.3	2.6± 0.1	102.2± 0.7
G-10	Robe	India	85.6±0.2	66.7±0.7	44.1±0.8	1382.3±0.9
G-11	Rosa beta	China	89.2±0.2	73.1±0.7	37.1±0.7	1226.3±0.8
G-12	Rosa beta	Italy	29.9±0.1	32.0±0.3	18.2±0.3	897.1± 0.6
G-13	Carmen red	Italy	116.9±0.2	44.6±0.4	56.1±0.1	1531.7±0.1
G-14	Lebrado square	Italy	70.2±0.2	48.1±0.5	46.1±0.9	1267.8±0.8
G-15	Rosa yoryeno	Italy- Spain	107.1±0.2	85.1±0.9	48.8±0.9	1413.1±0.9
G-16	Karara	Turkey	0.06±0.01	0.13±0.07	0.06±0.03	1.1±0.1
G-17	Rosa entema	Portugal	0.56±0.07	0.02±0.01	0.07±0.04	1.3±0.1
G-18	Azoles bahea	Brazil	314.4±0.4	186.4±0.2	147.0±0.3	1194.1±0.7
G-19	Bradezio (grey)	Brazil	1.5±0.1	3.0± 0.5	0.25±0.03	13.0± 0.5

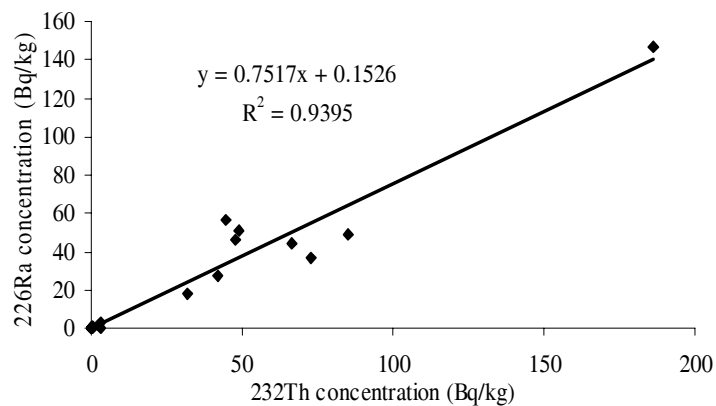
These values range from 0.089 Bq/kg (Royal gold) (G-7) to 504.61 Bq/kg (G-18) (Azoles bahea).

It is assumed that 370 Bq/kg of  $^{226}\text{Ra}$ , 259 Bq/kg of  $^{232}\text{Th}$  and 4810 Bq/kg of  $^{40}\text{K}$  produce the same gamma-ray dose rate [22]. To limit the external  $\gamma$ -radiation dose from building materials to 1.5 mSv/yr, the external hazard index ( $H_{ex}$ ) is defined by some workers [20,23] as:

$$A_{Ra}/370 + A_{Th}/259 + A_K/4810 \leq 1 \quad (3)$$



**Figure 3.** The correlation between  $^{238}\text{U}$  and  $^{226}\text{Ra}$  concentration in granite samples.



**Figure 4.** The correlation between  $^{40}\text{K}$  and  $^{232}\text{Th}$  concentration in granite samples.

The value of this index must be less than unity for the radiation risk to be negligible [23]. For the maximum value of  $H_{\text{ex}}$  to be less than unity, the maximum value of  $Ra_{\text{eq}}$  must be less than 370 Bq/kg. According to the calculated equation of  $H_{\text{ex}}$  [20,23] the values of  $H_{\text{ex}}$  for the studied marble samples range from 0.002 (m-24, m-4) to 0.359 (m-3), values which indeed are less than unity. While the values of  $H_{\text{ex}}$  for the studied granite are less than unity except for (G-18) as its value reaches 1.363 (see Table 3 and Table 4).

**Table 3.** Radium equivalent (Bq/kg), the external, internal, representative hazard indices and dose rate (nGy/h), for different marble samples

Sample No.	Commercial name	Origin	Ra <sub>eq</sub>	H <sub>in</sub> ≤ 1	H <sub>ex</sub> ≤ 1	I <sub>yr</sub> ≤ 1	D nGyh <sup>-1</sup>
m-1	Marble	Saudi	16.211	0.076	0.043	0.112	10.061
m-2	Cream marvel	Iran	9.044	0.047	0.024	0.061	6.316
m-3	Guatemala (green)	India	132.914	0.408	0.359	1.039	77.561
m-4	Guatemala (grey)	India	0.968	0.003	0.002	0.007	0.719
m-5	Golden camel	Pakistan	18.151	0.093	0.049	0.122	5.247
m-6	Karara	Italy	3.051	0.014	0.008	0.021	1.991
m-7	Karara (pure)	Italy	2.581	0.011	0.006	0.018	1.492
m-8	Cecelia perlato	Italy	1.584	0.006	0.004	0.011	1.197
m-9	Paly sandro clasico	Italy	10.711	0.045	0.028	0.078	7.321
m-10	Paly sandro (grey)	Italy	5.375	0.026	0.014	0.036	0.721
m-11	Fir de entco	Italy	10.671	0.034	0.028	0.083	6.962
m-12	Rosa lorca	Italy	2.204	0.008	0.006	0.018	1.423
m-13	Gallo royal	Italy	18.941	0.075	0.051	0.139	11.521
m-14	Yellow broctal	Italy	7.551	0.032	0.021	0.051	4.782
m-15	Krefenento notch	Italy	5.569	0.018	0.015	0.041	3.317
m-16	Red Krefenento	Italy	1.771	0.008	0.004	0.012	1.493
m-17	Brachia damascata	Italy	7.463	0.033	0.021	0.053	4.656
m-18	Brachia	Italy	1.781	0.007	0.004	0.012	1.577
m-19	Black Marconi a	Italy	20.881	0.111	0.056	0.141	12.711
m-20	Sheboleno	Italy	19.727	0.061	0.053	0.157	11.757
m-21	Esttoaryo	Italy	3.631	0.017	0.009	0.025	2.261
m-22	Rosa	Portugal	4.353	0.014	0.012	0.038	3.353
m-23	White mola	Turkey	1.661	0.007	0.007	0.004	0.011
m-24	Afeon shegar	Turkey	1.009	0.003	0.002	0.007	1.219
m-25	Russo lefanto	Turkey	5.251	0.027	0.014	0.035	4.493
m-26	Bianco Rosa	Turkey	1.254	0.005	0.003	0.008	1.547
m-27	Crystal	Greek	1.222	0.004	0.003	0.008	1.148
m-28	Bocshyaro	Spain	8.182	0.042	0.022	0.055	5.648
m-29	Russo basin	Spain	7.789	0.033	0.021	0.055	5.088
m-30	Red fetcano	Spain	18.281	0.055	0.049	0.142	11.361
m-31	Emparo oro	Spain	12.545	0.059	0.033	0.087	6.901
m-32	Russo elicanty	Spain	3.941	0.012	0.011	0.031	2.488
m-33	Cream marvel	Spain	1.245	0.005	0.003	0.008	1.547
m-34	Light Emprado	Spain	35.051	0.182	0.094	0.236	21.131
m-35	Azoles makoba	Brazil	65.741	0.183	0.177	0.511	38.033
m-36	Yellow sena	France	5.382	0.017	0.011	0.031	2.488
m-37	Silvia (yellow sand)	Egypt	7.654	0.039	0.021	0.051	5.796

**Table 4.** Radium equivalent (Bq/kg), the external, internal, representative hazard indices and dose rate (nGy/h), for different granite samples.

Sample No.	Commercial name	Origin	Ra <sub>eq</sub>	H <sub>in</sub> ≤ 1	H <sub>ex</sub> ≤ 1	I <sub>yr</sub> ≤ 1	D nGy h <sup>-1</sup>
G-1	Salmon	Saudi	201.331	0.618	0.544	1.591	122.411
G-2	Bianco sardo	Saudi	227.311	0.752	0.614	1.751	140.913
G-3	Sweet gold	Saudi	2.563	0.009	0.006	0.019	1.791
G-4	Najran brown	Saudi	0.278	0.001	0.007	0.001	0.474
G-5	Saudi brown	Saudi	0.559	0.002	0.001	0.003	0.422
G-6	Saudi green	Saudi	0.126	0.005	0.003	0.008	0.234
G-7	Royal gold	Saudi	0.089	0.003	0.002	0.006	0.144
G-8	Spring green	Saudi	0.116	0.004	0.003	0.008	0.268
G-9	Yezoa osoloto	India	14.871	0.047	0.041	0.116	8.621
G-10	Robe	India	245.711	0.783	0.664	1.882	148.771
G-11	Rosa beta	China	235.913	0.737	0.637	1.196	147.111
G-12	Rosa beta	Italy	132.945	0.408	0.359	1.039	77.561
G-13	Carmen red	Italy	327.771	0.794	0.642	1.841	155.246
G-14	Lebrado square	Italy	212.333	0.698	0.573	1.633	123.991
G-15	Rosa yoryeno	Italy Spain	279.162	0.886	0.754	2.118	171.911
G-16	Karara	Turkey	0.247	0.008	0.006	0.007	0.562
G-17	Rosa entema	Portugal	0.585	0.003	0.001	0.003	0.591
G-18	Azoles bahea	Brazil	504.611	1.761	1.363	3.636	320.771
G-19	Bradezio (grey)	Brazil	5.592	0.015	0.015	0.041	3.288
G-20	Bradezio (black)	Brazil	0.622	0.001	0.002	0.005	0.384

In addition to the external hazard, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by the internal hazard index (H<sub>in</sub>) which is given by the equation [20]:

$$A_{Ra}/185 + A_{Th}/259 + A_K/4810 \leq 1 \quad (4)$$

If the maximum concentration of radium is half that of the normal acceptable limit, then H<sub>in</sub> will be less than 1.0 [20]. For the safe use of a material in the construction of dwellings, H<sub>in</sub> should be less than unity. The calculated values of H<sub>in</sub> for the studied marble samples range from 0.003 (m-24, m-4) to 0.408 (m-3). Once again, all these values are less than unity. The calculated values of H<sub>in</sub> for the studied

granite samples are less than unity except for the same sample (G-18) where it reaches 1.761 (see Table (3) and Table (4)).

Conversion factors to transform specific activities  $A_K$ ,  $A_U$  and  $A_{Th}$  of K, U and Th, respectively, in absorbed dose rate at 1 m above the ground (in nGy/h by Bq/kg) are calculated and the values are [24]:

$$D=0.446 A_U +0.662 A_{Th} +0.048 A_K \quad (5)$$

In natural environmental radioactivity situations, the effective dose is calculated from the absorbed dose by applying the factor 0.7 Sv/Gy [24]. The absorbed dose rate for all the marble and granite samples is calculated and are listed in Table (3) and Table (4). The values are varied from 0.011 (m-23) to 77.56 (m-3) nGy/h for marble, and from 0.144 (G-7) to 320.77 (G-18) nGy/h.

As the marble and granites are widely used as building and covering materials, it is also possible to use an activity utilization index, proposed by [24], UNSCEAR and/or EC [25], that facilitates the derivation of dose rates in air from different combinations of these three radio nuclides.

According to the EC, the following gamma activity concentration index ( $I_{\gamma r}$ ) (representative level index) is derived for identifying whether a dose criterion is met:

$$I_{\gamma r} = A_{Ra}/150 + A_{Th}/100 + A_K/1500 \quad (6)$$

The index  $I_{\gamma r}$  is correlated with the annual dose due to the excess external gamma radiation caused by superficial material. Values of index  $I \leq 1$  correspond to 0.3 mSv/y, while  $I \leq 3$  correspond to 1 mSv/y. Thus, the activity concentration index should be used only as a screening tool for identifying materials which might be of concern to be used as covering material. According to this dose criterion, materials with  $I \leq 3$  should be avoided, since these values correspond to dose rates higher than 1 mSv/y [25], which is the highest value of dose rate in air recommended for population [24,26]. For the marble samples the minimum value is 0.004 (m-23) and the maximum value is 1.039 (m-3). While for the granite samples the minimum value is 0.001 (G-4) and the maximum value is 3.636 (G-18) (Azoles bahea).

## CONCLUSIONS

In view of worldwide concern about the radioactivity contents of various construction materials, experimental measurements of the activity concentrations of various marble and granite types (local and foreign) commonly used in Riyadh region have been carried out. The measured values of the activities of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in the marble samples have been found to lie in the ranges: 0.68–897.1, 0.36–32.4 and 0.10–32.0 Bq/kg, respectively. These samples were also found to have a radium equivalent activity in the range 0.968–132.914 Bq/kg. All of the samples were found to have hazard indices below 0.359 and 0.408 for the average external and internal hazard index, respectively.

The measured values of the activities of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in the granite samples have been found to lie in the ranges: 0.28–1531.7, 0.03–147.0 and 0.02–186.4

Bq/kg, respectively. These samples were also found to have a radium equivalent activity in the range 0.089 - 504.61 Bq/kg. All the samples under investigation were found to have average external and internal hazard indices less than unity except the Brazilian granite sample ( Azoles bahea) (G-18).

Therefore, the use of these marble and granite samples under investigation in the construction of dwellings is considered to be safe for inhabitants, except the Brazilian granite sample (Azoles bahea) (G-18).

Environmental monitoring should be carried out for marbles and granites where people might be exposed to radioactivity.

It emphasizes the importance of accurate information concerning commercial names and origins of these marble and granites, because simple mistakes on that can produce serious economical and social consequences in the stone market sector.

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## قياس النشاط الإشعاعي الطبيعي الصادر من بعض أنواع الرخام والجرانيت المستخدم في منطقة الرياض

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تم تجميع، من مصانع مختلفة في منطقة الرياض، سبع وثلاثون عينة من الرخام وعشرون عينة من الجرانيت المحلي والمستورد. طحنت هذه العينات وجففت لمدة أربع وعشرين ساعة ثم خزنت في أوعية مارينلي لمدة أربعة أسابيع. تم قياس إشعاعات جاما الصادرة منها باستخدام مطياف جامي ذو قدرة تحليل عالية وبزمن تخزين ٨٠٠٠٠ ثانية لكل عينة. ومن الطيف الجامي تم تحديد التراكيز للرخام ووجد أن قيمها تتراوح بين:  
 $^{238}\text{U}$ (0.71-44.1 Bq/kg),  $^{232}\text{Th}$ (0.10-32.0 Bq/kg),  $^{226}\text{Ra}$ (0.36-32.4 Bq/kg),  $^{40}\text{K}$ (0.68-897.1 Bq/kg).  
بينما للجرانيت وجد أن القيم تتراوح بين :  $^{238}\text{U}$ (0.03-314.4 Bq/kg) ,  $^{232}\text{Th}$ (0.02-186.4 Bq/kg) ,  $^{226}\text{Ra}$  (0.03-147.0 Bq/kg)  $^{40}\text{K}$  (0.28 – 1531.7 Bq/kg) .  
كما تم حساب كل من تركيز مكافئ الراديوم ومعدل الجرعة الممتصة في الهواء ومعاملات الخطورة الإشعاعية لجميع العينات.