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# Impact of Superficial Building Materials on Indoor Radon Level

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**Abstract:** This study is undertaken to determine the activity concentration of $^{226}$Ra, $^{232}$Th, $^{40}$K and radon emanation coefficient of some superficial materials commonly used in dwelling for interior decoration. Eighty samples of four different materials were collected from local suppliers and analyzed, employing high resolution gamma-ray spectroscopy. Mean activity concentrations for $^{226}$Ra ranged from 18.5 Bq/kg in marble, 60 Bq/kg in granite, 67 Bq/kg in ceramic up to 164 Bq/kg in porcelain, while for $^{232}$Th it ranged from 12 Bq/kg in marble, 53 Bq/kg in ceramic, 54.5 Bq/kg in granite up to 64 Bq/kg observed in porcelain samples and for $^{40}$K mean activity concentrations ranged from the lowest value of 137 Bq/kg in marble samples up to the highest value of 1042 Bq/kg in granite samples. The mean activity concentrations of $^{40}$K obtained for ceramic and porcelain samples were 499 and 525 Bq/kg, respectively. Radium equivalent activity values for the materials examined were lower than the international recommended maximum value of 370 Bq/kg, except for two granite samples where the values were found to be 433 and 565 Bq/kg. The annual indoor effective doses received by an individual from the examined samples did not exceed the exemption dose criterion of 0.3 mSv. The results also show that all the examined materials, apart from granite, appear to have low radon emanation coefficients.
**Keywords:** Natural radioactivity; Radon emanation coefficient; Annual effective dose.

## Introduction

Natural radionuclides in building materials are sources of external and internal exposure in dwellings, workplaces and industrial buildings. External radiation exposure is caused by the gamma emitting radionuclides. Internal radiation exposure, mainly affecting the respiratory tract, is due to the short-lived decay products of radon, which are exhaled from building materials into the room air. Therefore, radiation exposure of the population may be increased appreciably by the use of building materials containing above-normal levels of natural radionuclides to levels comparable or even greater than the exposure from medical radiation [1-3].

In most cases, the natural radioactivity content of building materials is determined by gamma-ray spectrometry. The external radiation exposure is estimated either by direct exposure measurements or by mathematical calculations. More complicated is the evaluation of the internal radiation exposure. Radon emanation from a material depends not only on the radium content of the material, but also on many other factors, including for example grain size, permeability, material density and ventilation rate. Several methods for the measurement of radon emanation and exhalation were developed and reported in the literature [4-5].

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In recent years, there has been a significant increase in the use of granite, marble and glazed tiles as covering or decorative building materials for home interiors instead of conventional or mosaic tiles. Because of their appearance, availability, attractive colors, polished surface and durability against external conditions and high scratch resistance, granite and marble tiles are commonly used as wall and floor covering, facing materials for buildings, kitchen countertop, vanity tops and inner and outer decorative materials. Ceramic and porcelain tiles are also used in bathrooms, toilets and kitchens. Radionuclides' concentrations and radon emanation or exhalation coefficients of various covering materials used in different countries have been reported in the literature. These include granite [1, 3, 6-7], marble [8-10], ceramic and porcelain [11-15].

However, covering or decorative building materials derived from rock and soil include primordial radionuclides such as $^{226}$Ra, $^{232}$Th, and $^{40}$K varying from one country to another and from one location to another in the same country [35]. While many reports on activity concentration and radon emanation coefficients of these materials have been published, their large variability has made the existing data insufficient to conclude which types of tiles, for example, have a higher activity level than others.

The purpose of this study is to determine: radioactivity present in these materials, their radon emanation and exhalation coefficients and their possible contribution to the indoor radon concentration. The results obtained are also used to assess any possible radiological hazard to occupants of the dwellings using such materials. In addition, the results are compared with the corresponding results available in the literature.

## Materials and Methods

In the present work, samples of covering and decorative building materials were collected from local tile manufacturing companies and agencies supplying raw construction materials. The materials collected were granite, marble ceramic and porcelain tiles. The samples were crushed, ground, homogenized, sieved through 0.15 mm mesh, sealed in cylindrical plastic containers of 5.0 cm diameter and 4.0 cm height, dry-weighed and marked according to the name and producer company. The samples were then stored for almost a month in order to achieve secular equilibrium between the series parents and the subsequent daughters following radon in the decay sequence.

A total of 80 samples including granite, marble, ceramic and porcelain were involved in the measurements. Details concerning the petrographic features, mineral chemistry, geochemistry and physico-mechanical properties of the investigated samples are available in the literature for marble and granite rocks [5, 3, 17 and 18] and for ceramic and porcelain tiles [19-20, 11].

Measurements were performed using a High Purity Germanium (HPGe) detector supplied by EG&G Ortec. The detector is an n-type gamma-x ray (GMX) detector, operated at 3500 V, with a useful energy range of 3 keV to 10 MeV, a standard energy resolution of 2.02 keV and a relative efficiency of 56.9% at 1.33 MeV of $^{60}$Co. The absolute efficiency calibration of the detector was performed using the radionuclide specific efficiency method, in which the uncertainty in gamma-ray intensities, the influence of coincidence summation and self-absorption effects of the emitting gamma photons are avoided. The IAEA certified reference materials RG-set (RGU-1, RGTh-1, RGK-1), each sealed in a cylindrical plastic container identical to those of samples, were used in the calibration process. Spectra were collected for 12 hours. Areas under the energy peaks of interest were used for drawing the peak efficiency curve between log of efficiency versus log of peak energy. A polynomial was fitted to the curve and the result was stored for further use.

The gamma-ray transition lines 295.20 keV (19.2%) and 351.90 keV (37.1%) from ($^{214}$Pb) and 609.30 keV (46.1%) and 1764 keV (16%) from ($^{214}$Bi) were used as the signature of $^{226}$Ra. The gamma-ray transition lines 238.6 keV (43.6%) from ($^{212}$Pb), 583.14 keV (86.0%) from ($^{208}$Tl) and 911.07 keV (29.0%) from ($^{228}$Ac) were used as the signature of $^{232}$Th. The activity concentration of $^{40}$K was measured directly from its only 1460 keV (10.7%) transition line.

Prior to the sample measurements, an empty container, identical to those of samples and reference materials, was put into the shielded detector and measured for 12 hours to determine the laboratory background which was later subtracted from the measured gamma-ray spectrum of each sample. Measured background

and counting time were then used to determine the minimum detectable activity (MDA) for each peak of interest according to the Currie method [21]. The values obtained are given in Table 1.

TABLE 1. Background counting rates and minimum detectable activities (MDA) obtained with the standard sources.

| Nuclide | Energy (keV) | Background count/h | MDA (Bq/kg) |
|---|---|---|---|
| $^{214}$Pb | 295.2 | 48.8 | 0.19 |
| $^{214}$Pb | 351.9 | 85.6 | 0.25 |
| $^{214}$Bi | 609.3 | 68.8 | 0.23 |
| $^{214}$Bi | 1764.1 | 66.5 | 0.22 |
| $^{228}$Ac | 911.1 | 10.2 | 0.09 |
| $^{208}$Tl | 583.2 | 10.4 | 0.09 |
| $^{212}$Pb | 238.6 | 23.6 | 0.13 |
| $^{40}$K | 1460.8 | 798211.8 | 24.5 |

## Calculation and Analysis

Investigation of the radiation contents of the samples was carried out by placing sample containers sequentially on the top of the detector cap and counting for at least 12 h for each sample. The specific activity $A_i$ (Bq/kg) of a radionuclide i of peak energy $E_i$ was calculated using the formula [5,12]:

$$A_i = \frac{C_i}{\varepsilon(E_i).I.t.M}, \quad (1)$$

where $C_i$ is the net peak area of gamma-ray at energy $E_i$, $\varepsilon(E_i)$ is the absolute efficiency for gamma rays at a particular energy, $I$ is the absolute transition probability of gamma decay, $t$ is the counting live-time and $M$ is the dried sample mass.

Radium equivalent activity $Ra_{eq}$ is an index that was introduced to account for the radiation hazard from a radionuclide mixture within a material [22-23]. Assuming that 370 Bq/kg of $^{226}$Ra, 259 Bq/kg of $^{232}$Th and 4810 Bq/kg of $^{40}$K produce the same gamma-ray dose rate, $Ra_{eq}$ was calculated using the following relation[16]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (2)$$

where $A_{Ra}$, $A_{Th}$, and $A_K$ are the specific activities in Bq/kg of $^{226}$Ra, $^{232}$Th and $^{40}$K, respectively.

A radium equivalent activity of 370 Bq/kg in building materials will produce an external gamma radiation dose rate of 1.5 mSv/year; i.e., to keep the external gamma-ray dose below that value, a building material is considered to be permissible if the condition $Ra_{eq} < 370$ is satisfied.

However, the indoor-absorbed dose rate $D$ (nGy/h) due to the external gamma radiation from marble, ceramic, porcelain and granite tiles utilized as building materials, may be calculated from the activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K present in those materials, using the following relation [24]:

$$D = 0.12 \times A_{Ra} + 0.14 \times A_{Th} + 0.0096 \times A_k \quad (3)$$

The indoor annual effective dose $E_{eff}$ (mSv/y) was estimated from the value of $D$ by adopting a conversion factor of 0.7 Sv/Gy and an annual exposure time of 7000 h/y, considering that people on average spend 80% of their time indoors, using the following formula [25-26,23]:

$$E_{eff} = 0.7\ (SvGy^{-1}) \times 7000(h\ y^{-1}) \times D\ (Gy/h) \quad (4)$$

For superficial and other building materials with restricted fractional mass usage, similar to those examined in this study, the exemption dose criterion is 0.3 mSv/y. Effective doses exceeding the dose criterion of 1 mSv/y should be taken into account in terms of radiation protection.

The radon emanation coefficient of samples, defined as radon escape to production ratio, was calculated based on two gamma spectroscopic measurements [4, 27-28]. The first measurement was carried out directly after sealing the samples, while the second measurement was carried out after attaining to secular equilibrium between radon and its progeny. Assuming that radon activities are always equal to those of its progenies, gamma rays emitted from $^{214}$Pb and $^{214}$Bi were counted in both measurements. Based on the theoretical $^{222}$Rn ingrowth curve, Fig. 1, and assuming that all radon atoms at the time of sample container sealing (t = 0) exist in the solid phase, the net count rate of gamma ray $C$(t) at time t (day) can be given as:

$$C(t) = C_{eq}(1 - e^{-\lambda t}) + C_o e^{-\lambda t} \quad (5)$$

where $\lambda$ is the decay constant of radon, $C_{eq}$ and $C_o$ are the net count rates at t >30 days and t = 0, respectively[4]. As counting rate is proportional to radon activity through a calibration constant, which cancels out when activity ratio is calculated, the radon emanation coefficient F (%) was calculated using direct counting rate ratios instead of measured activities as [23]:

$$F = \frac{C_{eq} - C_o}{C_{eq}} \times 100 \quad (6)$$

The detection limit of $F$ is related to the obtained total number of counts within the measurement time or to $^{214}$Pb and $^{214}$Bi activities of the samples.

Assuming that $A_{Ra}$ (Bq kg$^{-1}$) is the concentration of $^{226}$Ra, then the radon mass exhalation rate $E_m$ (Bq kg$^{-1}$ h$^{-1}$) is calculated as [22, 27, 5 and 12]:

$$E_m = F A_{Ra} \lambda \quad . \quad (7)$$

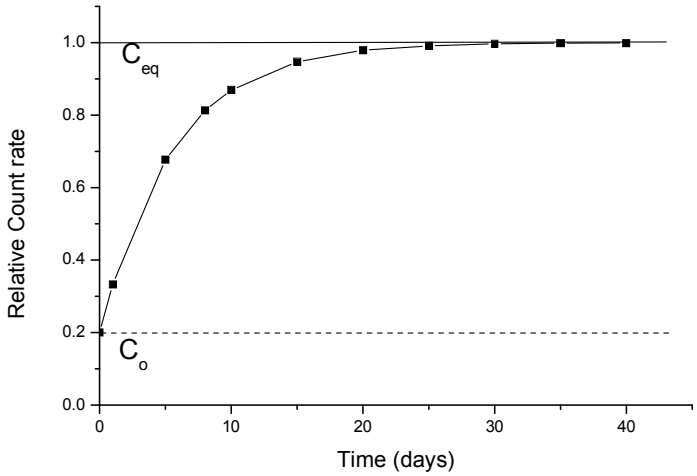


FIG. 1. Radon ingrowth curve.

## Results and Discussion

### Activity Concentration

Tables 2-5 list the activity concentration (in Bq/kg) of $^{226}$Ra, $^{232}$Th, $^{40}$K, the radon emanation coefficient F (%), the radon mass exhalation rate $E_m$ (Bq kg$^{-1}$ h$^{-1}$), the radium equivalent activity $Ra_{eq}$ (Bq/kg), the total absorbed dose rate D (nGy/h) and the annual effective dose $E_{eff}$ (mSv) for all the examined samples. For comparison purposes, summaries of the above data are presented in Table 6. As it can be seen in the tables, substantial variability was observed both between and within the examined materials. The $^{226}$Ra, $^{232}$Th and $^{40}$K activity concentrations were different in different types of granite, marble, ceramic and porcelain samples available in the local market. This is due to the different composition of these materials and uneven distribution of the radionuclides within the samples. The variation observed in similar materials is also a function of the local geology as building materials are extracted from different regions of earth crust and are used directly in building construction [29].

Mean concentration values of $^{226}$Ra, $^{232}$Th and $^{40}$K measured in marble samples are significantly lower than those found in other material samples, as shown in Fig. 2. Mean activity concentrations of $^{226}$Ra ranged from 18.5 Bq/kg in marble, 60 Bq/kg in granite, 67 Bq/kg in ceramic up to 164 Bq/kg in porcelain. The minimum measured value of $^{226}$Ra activity concentration was 1.0±0.1 Bq/kg in the marble sample (M8), whereas the maximum value was 228±5 Bq/kg in the porcelain sample (P8). Granite and ceramic were also highly populated with $^{226}$Ra, but still their activities are less than that observed in porcelain tiles, characterized by the presence of zircon mineral that has a relatively high uranium activity concentration [13].

The mean activity concentration of $^{232}$Th ranged from 12 Bq/kg in marble, 53 Bq/kg in ceramic, 54.5 Bq/kg in granite up to 64 Bq/kg observed in porcelain samples. The minimum observed $^{232}$Th activity concentration was 1.6±0.2 Bq/kg in the marble sample (M5) and the maximum was 228±3 Bq/kg in the granite sample (G19). It is also observed from the results that $^{232}$Th has relatively the least concentration value in comparison to those of $^{226}$Ra and $^{40}$K in the samples examined.

TABLE 2. Activity concentration (Bq/kg) of $^{226}Ra$, $^{232}Th$, $^{40}K$, radon emanation coefficient F (%), radon mass exhalation rate $E_m$ (Bq $kg^{-1}$ $h^{-1}$), radium equivalent activity $Ra_{eq}$ (Bq/kg), total absorbed dose rate D (nGy/h) and annual effective dose $E_{eff}$ (mSv) for granite samples.

| Sample Code | Country of origin | Activity concentration (Bq/kg) | | | F (%) | $E_m$ (Bq/kg.h) | $Ra_{eq}$ (Bq/kg) | D (nGy/h) | $E_{eff}$ (mSv) |
|---|---|---|---|---|---|---|---|---|---|
| | | $^{226}Ra$ | $^{232}Th$ | $^{40}K$ | | | | | |
| G1 | Jordan | 38±2 | 46±1 | 989±39 | 5.2 | 0.015 | 179 | 20.5 | 0.10 |
| G2 | S. Africa | 22±1 | 33±1 | 324±12 | 23 | 0.038 | 94 | 10.4 | 0.05 |
| G3 | S. Africa | 18±1 | 14±1 | 63±4 | 19 | 0.026 | 43 | 4.7 | 0.02 |
| G4 | China | 89±3 | 112±6 | 1253±52 | 5.9 | 0.040 | 346 | 38.4 | 0.19 |
| G5 | S. Arabia | 38±2 | 46±1 | 1076±48 | 16.3 | 0.047 | 187 | 21.3 | 0.10 |
| G6 | S. Arabia | 32±1 | 76±3 | 1662±59 | 18.1 | 0.044 | 269 | 30.4 | 0.15 |
| G7 | S. Arabia | 43±2 | 35±2 | 284±28 | 32 | 0.104 | 115 | 12.8 | 0.06 |
| G8 | S. Arabia | 28±1 | 18±1 | 1282±54 | 38 | 0.081 | 153 | 18.2 | 0.09 |
| G9 | Italy | 42±1 | 65±2 | 1133±46 | 22 | 0.070 | 222 | 25.0 | 0.12 |
| G10 | Italy | 57±2 | 84±3 | 1152±45 | 11.4 | 0.049 | 266 | 29.7 | 0.15 |
| G11 | Italy | 67±2 | 35±2 | 1071±56 | 1.8 | 0.009 | 200 | 23.2 | 0.11 |
| G12 | Italy | 53±2 | 37±2 | 1250±47 | 18.8 | 0.075 | 202 | 23.5 | 0.12 |
| G13 | Italy | 106±3 | 157±5 | 1331±55 | 2.1 | 0.017 | 433 | 47.5 | 0.23 |
| G14 | India | 14±1 | 87±2 | 926±24 | 7.4 | 0.034 | 210 | 22.7 | 0.11 |
| G15 | India | 61±2 | 48±1 | 1322±35 | 5.9 | 0.027 | 231 | 26.7 | 0.13 |
| G16 | Spain | 71±1 | 43±1 | 1358±36 | 16 | 0.086 | 237 | 27.6 | 0.14 |
| G17 | Spain | 116±2 | 76±2 | 1334±34 | 11 | 0.097 | 327 | 37.4 | 0.18 |
| G18 | Spain | 59±1 | 121±2 | 1438±38 | 8 | 0.036 | 343 | 37.8 | 0.19 |
| G19 | Finland | 152±1 | 228±3 | 1123±31 | 16 | 0.184 | 565 | 60.9 | 0.30 |
| G20 | Finland | 62±1 | 54±1 | 1368±33 | 4 | 0.019 | 245 | 28.1 | 0.14 |
| G21 | Finland | 45±1 | 39±1 | 942±26 | 18 | 0.061 | 173 | 19.9 | 0.10 |
| G22 | Egypt | 81±3 | 55±2 | 663±21 | 14 | 0.086 | 211 | 23.8 | 0.12 |
| G23 | Egypt | 32±5 | 63±2 | 1375±42 | 4.8 | 0.012 | 228 | 25.9 | 0.13 |
| G24 | Egypt | 118±4 | 59±2 | 288±12 | 22 | 0.196 | 225 | 25.2 | 0.12 |

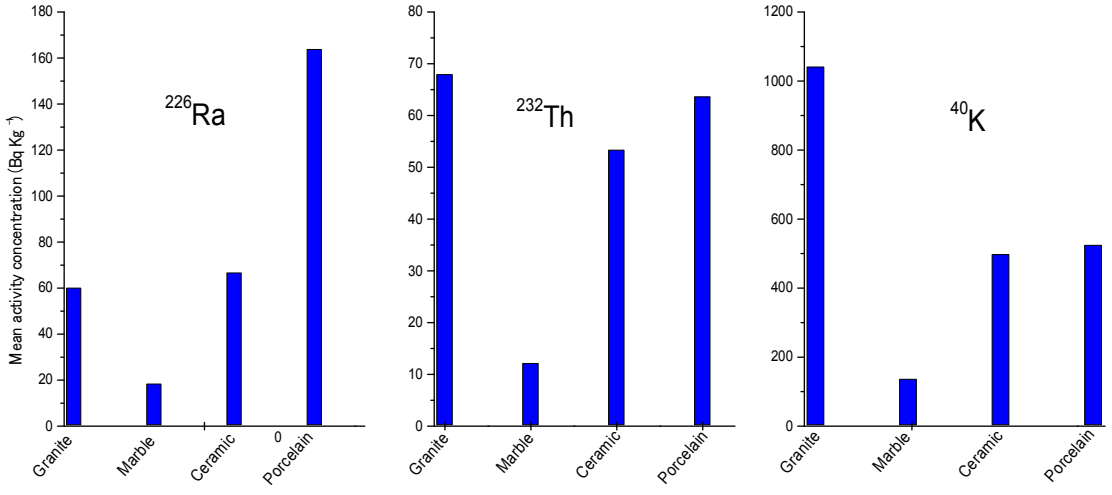


FIG. 2. Mean activity concentration of $^{226}Ra$, $^{232}Th$ and $^{40}K$ in granite, marble, ceramic and porcelain samples.

TABLE 3. Activity concentration (Bq/kg) of $^{226}Ra$, $^{232}Th$, $^{40}K$, radon emanation coefficient F (%), radon mass exhalation rate $E_m$ (Bq $kg^{-1}$ $h^{-1}$), radium equivalent activity $Ra_{eq}$ (Bq/kg), total absorbed dose rate *D* (nGy/h) and annual effective dose $E_{eff}$ (mSv) for marble samples.

| Sample Code | Country of origin | Activity concentration (Bq/kg) | | | F (%) | $E_m$ (Bq/kg.h) | $Ra_{eq}$ (Bq/kg) | D (nGy/h) | $E_{eff}$ (mSv) |
|---|---|---|---|---|---|---|---|---|---|
| | | $^{226}Ra$ | $^{232}Th$ | $^{40}K$ | | | | | |
| M1 | Jordan | 19±1 | 14±1 | 93±2 | 9.7 | 0.014 | 46 | 5.1 | 0.03 |
| M2 | Jordan | 16±2 | 7.5±1.2 | 28±7 | 1.7 | 0.002 | 29 | 3.2 | 0.02 |
| M3 | Jordan | 20.4±0.8 | 13±1 | 85±2 | 11.7 | 0.018 | 46 | 5.1 | 0.02 |
| M4 | Jordan | 28±2 | 8.2±0.4 | 42±2 | 8.5 | 0.018 | 43 | 4.9 | 0.02 |
| M5 | Jordan | 19±2 | 1.6±0.2 | 29±6 | 13.9 | 0.020 | 24 | 2.8 | 0.01 |
| M6 | Italy | 5.1±0.2 | 2.2±0.1 | 28±2 | 5.2 | 0.002 | 9 | 1.0 | 0.01 |
| M7 | Italy | 14±1 | 18±1 | 116±18 | 2.8 | 0.003 | 49 | 5.3 | 0.03 |
| M8 | Italy | 1.0±0.1 | 3.9±0.7 | 28±3 | ND | ND | 8 | 1.1 | 0.01 |
| M9 | Italy | 37±3 | 19±1 | 31±2 | 7.4 | 0.021 | 67 | 7.4 | 0.04 |
| M10 | Italy | 16±2 | 2.4±0.4 | 33±3 | 14.9 | 0.018 | 22 | 2.6 | 0.01 |
| M11 | Spain | 13.6±0.1 | 12.2±0.1 | 36±3 | 33 | 0.032 | 34 | 3.7 | 0.02 |
| M12 | Spain | 12±1 | 4.6±1.2 | 94±4 | 19.8 | 0.018 | 26 | 3.0 | 0.01 |
| M13 | Malaysia | 17±2 | 16±2 | 247±6 | 28 | 0.036 | 59 | 6.7 | 0.03 |
| M14 | S. Arabia | 31±2 | 29±2 | 31±4 | 17.1 | 0.040 | 74 | 8.0 | 0.04 |
| M15 | S. Arabia | 28±3 | 13±1 | 47±3 | 0.9 | 0.002 | 50 | 5.6 | 0.03 |
| M16 | S. Arabia | 19±1 | 35±4 | 56±4 | 2.7 | 0.016 | 73 | 7.7 | 0.04 |
| M17 | Greece | 11±1 | 2.3±0.6 | 108±7 | 31 | 0.026 | 21 | 2.7 | 0.01 |
| M18 | Greece | 53±4 | 32±2 | 774±21 | 5.8 | 0.023 | 158 | 18.3 | 0.09 |
| M19 | Greece | 25±2 | 42±2 | 988±31 | 1.1 | 0.002 | 161 | 18.4 | 0.09 |
| M20 | China | 13±1 | 2.2±0.1 | 162±11 | 25.4 | 0.025 | 29 | 3.4 | 0.02 |
| M21 | Turkey | 29.4±1.6 | 2.4±0.2 | 34.4±3.1 | 1.8 | 0.004 | 35 | 4.2 | 0.02 |
| M22 | Turkey | 2.3±0.2 | 4.4±0.3 | 26±2 | ND | ND | 9 | 1.1 | 0.01 |
| M23 | Turkey | 15.2±0.6 | 12.6±0.7 | 377±26 | 16.7 | 0.019 | 62 | 7.2 | 0.04 |
| M24 | Egypt | 11.8±1.2 | 4.8±0.6 | 28±2 | 8.7 | 0.008 | 21 | 2.4 | 0.01 |
| M25 | Egypt | 5.5±0.4 | 3.2±0.1 | 45±4 | 9.2 | 0.004 | 14 | 1.5 | 0.01 |

TABLE 4. Activity concentration (Bq/kg) of $^{226}Ra$, $^{232}Th$, $^{40}K$, radon emanation coefficient F (%), radon mass exhalation rate $E_m$ (Bq $kg^{-1}$ $h^{-1}$), radium equivalent activity $Ra_{eq}$ (Bq/kg), total absorbed dose rate *D* (nGy/h) and annual effective dose $E_{eff}$ (mSv) for ceramic samples.

| Sample Code | Country of origin | Activity concentration (Bq/kg) | | | F (%) | $E_m$ (Bq/kg.h) | $Ra_{eq}$ (Bq/kg) | D (nGy/h) | $E_{eff}$ (mSv) |
|---|---|---|---|---|---|---|---|---|---|
| | | $^{226}Ra$ | $^{232}Th$ | $^{40}K$ | | | | | |
| C1 | Jordan | 33±3 | 36±3 | 420±28 | 4.8 | 0.012 | 117 | 13.0 | 0.06 |
| C2 | Jordan | 70±5 | 52±5 | 537±40 | 2.8 | 0.015 | 154 | 20.8 | 0.10 |
| C3 | Turkey | 78±7 | 61±4 | 486±54 | 3 | 0.018 | 203 | 22.6 | 0.11 |
| C4 | Turkey | 53±11 | 44±4 | 414±24 | 5.5 | 0.022 | 148 | 16.5 | 0.08 |
| C5 | Spain | 92±6 | 42±4 | 726±23 | 5 | 0.035 | 208 | 23.9 | 0.12 |
| C6 | Spain | 72±5 | 56±5 | 464±22 | 2 | 0.011 | 188 | 16.5. | 0.08 |
| C7 | Egypt | 106±8 | 63±4 | 348±21 | 12 | 0.096 | 223 | 24.9 | 0.12 |
| C8 | Egypt | 81±5 | 55±4 | 364±14 | 14 | 0.086 | 188 | 20.9 | 0.10 |
| C9 | Italy | 79±3 | 66±3 | 692±38 | 2.8 | 0.017 | 227 | 25.4 | 0.12 |
| C10 | Italy | 48±4 | 42±2 | 462±40 | 0.5 | 0.002 | 144 | 16.1 | 0.08 |
| C11 | Italy | 56±5 | 44±4 | 441±40 | 0.4 | 0.002 | 153 | 17.1 | 0.08 |
| C12 | Syria | 53±8 | 56±5 | 522±42 | 0.5 | 0.002 | 173 | 19.2 | 0.09 |

| Sample Code | Country of origin | Activity concentration (Bq/kg) | | | F (%) | $E_m$ (Bq/kg.h) | $Ra_{eq}$ (Bq/kg) | D (nGy/h) | $E_{eff}$ (mSv) |
|---|---|---|---|---|---|---|---|---|---|
| | | $^{226}Ra$ | $^{232}Th$ | $^{40}K$ | | | | | |
| C13 | Syria | 75±6 | 64±6 | 590±46 | ND | ND | 212 | 23.6 | 0.12 |
| C14 | Syria | 65±6 | 54±5 | 664±54 | 0.8 | 0.004 | 193 | 21.7 | 0.11 |
| C15 | China | 61±4 | 53±5 | 386±44 | 3.2 | 0.015 | 167 | 18.4 | 0.09 |
| C16 | China | 72±6 | 58±6 | 568±38 | 3.1 | 0.017 | 199 | 22.2 | 0.11 |
| C17 | UAE | 103±8 | 62±3 | 669±42 | 6.3 | 0.050 | 243 | 27.5 | 0.13 |
| C18 | India | 45±4 | 42±3 | 347±18 | 8.2 | 0.028 | 132 | 18.0 | 0.09 |
| C19 | India | 28±2 | 64±5 | 374±20 | 17 | 0.036 | 148 | 15.9 | 0.08 |

TABLE 5. Activity concentration (Bq/kg) of $^{226}Ra$, $^{232}Th$, $^{40}K$, radon emanation coefficient F (%), radon mass exhalation rate $E_m$ (Bq $kg^{-1}$ $h^{-1}$), radium equivalent activity $Ra_{eq}$ (Bq/kg), total absorbed dose rate *D* (nGy/h) and annual effective dose $E_{eff}$ (mSv) for porcelain samples.

| Sample Code | Country of origin | Activity concentration (Bq/kg) | | | F (%) | $E_m$ (Bq/kg.h) | $Ra_{eq}$ (Bq/kg) | D (nGy/h) | $E_{eff}$ (mSv) |
|---|---|---|---|---|---|---|---|---|---|
| | | $^{226}Ra$ | $^{232}Th$ | $^{40}K$ | | | | | |
| P1 | Spain | 117±7 | 62±3 | 584±33 | 0.2 | 0.002 | 251 | 28.3 | 0.14 |
| P2 | Spain | 96±8 | 53±4 | 442±25 | ND | ND | 206 | 23.2 | 0.11 |
| P3 | Spain | 192±8 | 68±4 | 395±29 | 0.4 | 0.006 | 320 | 36.4 | 0.18 |
| P4 | Spain | 182±7 | 58±2 | 612±44 | ND | ND | 312 | 35.5 | 0.17 |
| P5 | Spain | 78±4 | 72±5 | 504±52 | ND | ND | 220 | 24.3 | 0.12 |
| P6 | Italy | 89±4 | 44±3 | 283±28 | 0.5 | 0.003 | 174 | 19.6 | 0.10 |
| P7 | Italy | 146±6 | 98±3 | 557±35 | 0.4 | 0.004 | 329 | 36.6 | 0.18 |
| P8 | Italy | 228±5 | 58±4 | 696±24 | ND | ND | 365 | 42.2 | 0.21 |
| P9 | Italy | 214±8 | 76±6 | 484±24 | ND | ND | 360 | 41.0 | 0.20 |
| P10 | China | 186±6 | 64±4 | 528±36 | ND | ND | 318 | 36.3 | 0.18 |
| P11 | China | 212±6 | 65±4 | 655±51 | 0.4 | 0.006 | 355 | 40.8 | 0.20 |
| P12 | China | 227±8 | 46±3 | 564±47 | 0.2 | 0.003 | 336 | 39.1 | 0.19 |

TABLE 6. Mean, median, minimum, maximum and standard deviation values of the measured data for granite, marble, ceramic and porcelain samples.

| Material | Statistics | Activity concentration (Bq/kg) | | | F (%) | $E_m$ (Bq/kg.h) | $Ra_{eq}$ (Bq/kg) | D (nGy/h) | $E_{eff}$ (mSv) |
|---|---|---|---|---|---|---|---|---|---|
| | | $^{226}Ra$ | $^{232}Th$ | $^{40}K$ | | | | | |
| Granite | | | | | | | | | |
| | Mean | 60 | 68 | 1042 | 14 | 0.061 | 238 | 27 | 0.13 |
| | Median | 55 | 54.5 | 1142.5 | 15 | 0.044 | 224 | 25 | 0.12 |
| | Min. | 14 | 14 | 63 | 1.8 | 0.012 | 43 | 4.7 | 0.02 |
| | Max. | 152 | 228 | 1662 | 38 | 0.196 | 565 | 60.9 | 0.30 |
| | S.D. | 35 | 47.6 | 420.5 | 9 | 0.048 | 109 | 11.7 | 0.06 |
| Marble | | | | | | | | | |
| | Mean | 18.5 | 12 | 137 | 12 | 0.016 | 47 | 5.3 | 0.03 |
| | Median | 16 | 8 | 42 | 9 | 0.018 | 35 | 4.2 | 0.02 |
| | Min. | 1 | 1.6 | 26 | ND | 0.0 | 8 | 1 | 0.01 |
| | Max. | 53 | 35 | 988 | 33 | 0.04 | 161 | 18.4 | 0.09 |
| | S.D. | 11.5 | 11.4 | 241 | 9.8 | 0.011 | 39 | 4.5 | 0.02 |

| Material | Statistics | Activity concentration (Bq/kg) | | | F (%) | $E_m$ (Bq/kg.h) | $Ra_{eq}$ (Bq/kg) | D (nGy/h) | $E_{eff}$ (mSv) |
|---|---|---|---|---|---|---|---|---|---|
| | | $^{226}Ra$ | $^{232}Th$ | $^{40}K$ | | | | | |
| Ceramic | | | | | | | | | |
| | Mean | 67 | 53 | 499 | 5 | 0.025 | 180 | 20 | 0.10 |
| | Median | 70 | 55 | 464 | 3 | 0.012 | 188 | 20.8 | 0.10 |
| | Min. | 28 | 36 | 347 | ND | 0.0 | 117 | 13 | 0.08 |
| | Max. | 106 | 66 | 726 | 17 | 0.096 | 243 | 27.5 | 0.13 |
| | S.D. | 21 | 9 | 123 | 4.8 | 0.027 | 35 | 3.9 | 0.02 |
| Porcelain | | | | | | | | | |
| | Mean | 164 | 64 | 525 | 0.18 | 0.0020 | 296 | 33.6 | 0.17 |
| | Median | 184 | 63 | 543 | 0.1 | 0.0035 | 319 | 36.4 | 0.18 |
| | Min. | 78 | 44 | 283 | ND | 0.0 | 174 | 19.6 | 0.10 |
| | Max. | 228 | 98 | 696 | 0.5 | 0.0060 | 365 | 42.2 | 0.21 |
| | S.D. | 56 | 14.5 | 115 | 0.2 | 0.0024 | 65.5 | 7.7 | 0.04 |

The mean activity concentration of $^{40}K$, in the studied samples, ranged from the lowest value of 137 Bq/kg for marble samples up to the highest value of 1042 Bq/kg for granite samples. The mean activity concentrations obtained for ceramic and porcelain samples were 499 and 525 Bq/kg, respectively. The lowest observed $^{40}K$ activity concentration was 26±2 Bq/kg in the marble sample (M22) and the highest was 1662±59 Bq/kg in the granite sample (G6).

It can be seen from the results that the mean activity concentrations measured in the marble samples were significantly lower than the typical world average values of 50, 50 and 500 Bq/kg for $^{226}Ra$, $^{232}Th$ and $^{40}K$, respectively [30]. In addition, the mean activity concentrations of $^{226}Ra$ and $^{232}Th$ measured in granite, ceramic and porcelain samples were slightly higher than the quoted values, while the mean value of $^{40}K$ was found to be higher than the world average for granite samples only. However, the natural radioactivity levels measured in all samples are comparable to those measured in other countries and the average worldwide ranges [31].

**Radiological Hazard Indices**

The radium equivalent activity ($Ra_{eq}$) of the examined samples was calculated according to Eq. (2) to evaluate the relative radiological hazard due to external irradiation. The lowest mean value found was for marble (47 Bq/kg), while the highest was for porcelain (296 Bq/kg). The other mean values obtained were 180 and 238 Bq/kg for ceramic and granite, respectively. It is clear that all the estimated mean values of $Ra_{eq}$ (Fig. 3) were lower than the recommended maximum value of 370 Bq/kg. However, the exceptions are two granite samples, where $Ra_{eq}$ was found to be 433 and 565 Bq/kg for the samples (G13) and (G19), respectively.

The indoor absorbed gamma dose rate (D) in air was calculated, using the measured activity concentration data, according to Eq. (3). The values obtained are listed in Tables 2-5. The obtained mean values were 27, 5.3, 20 and 33.6 nGy/h for granite, marble, ceramic and porcelain samples, respectively. The lowest observed D value was 1 nGy/h for the marble sample (M6) and the highest value was 60.9 nGy/h for the granite sample (G19). All the estimated D values for the samples examined were lower than the world average indoor gamma absorbed dose rate of 84 nGy/h [25]. However, the correlation of D values to those of $Ra_{eq}$ is shown in Fig. 4, for granite samples. The significant correlation between the two indices (R = 0.99) is expected due to the common specific activity concentrations of $^{226}Ra$, $^{232}Th$ and $^{40}K$, utilized in their evaluation (Eqs. 2 and 3) although weighting or conversion factors as well as units were different.

Finally, the annual effective dose $E_{eff}$ was calculated from the sample D values according to Eq. (4). The estimated $E_{eff}$ mean values were 0.13, 0.03, 0.10 and 0.17 mSv for granite, marble, ceramic and porcelain samples, respectively. The minimum observed $E_{eff}$ value was 0.01 mSv for many marble samples and the maximum value was 0.3 mSv for the granite sample (G19). It can be seen from Tables 2-5 that all the estimated $E_{eff}$ values did not exceed the exemption dose criterion of 0.3 mSv [25].

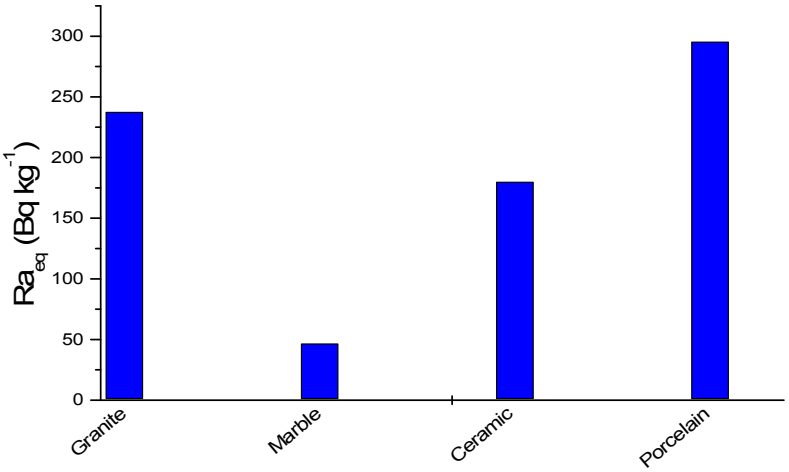


FIG. 3. Mean values of radium equivalent activity ($Ra_{eq}$) for granite, marble, ceramic and porcelain samples. (All the measured values are less than the recommended maximum value of 370 Bq kg$^{-1}$).

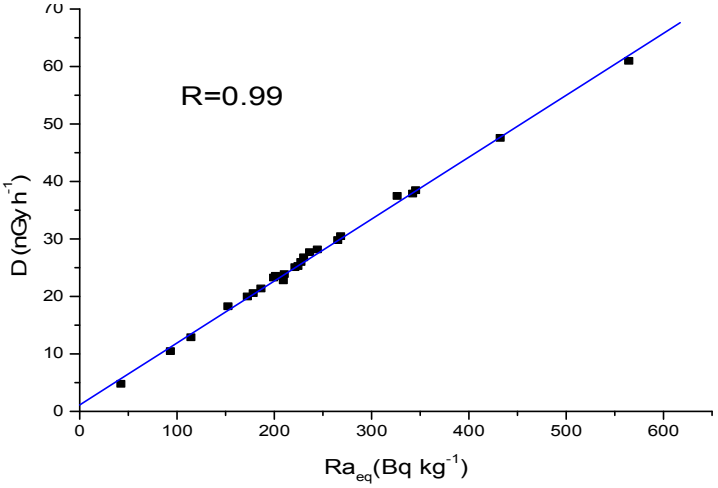


FIG. 4. The correlation of the absorbed dose rate (D) values to those of radium equivalent activity ($Ra_{eq}$), for granite samples.

**Radon Emanation Coefficient and Exhalation Rate**

The radon emanation coefficient F (%) and radon mass exhalation rate $E_m$ (Bq kg$^{-1}$ h$^{-1}$) of the examined samples are listed in Tables 2-5. The emanation coefficient varied from a non-detectable (ND) value in some marble, ceramic and porcelain samples to a maximum value of 38 % in the granite sample (G8). Although the values of F in granite samples have shown a wide dispersion (range: 1.8-38% and standard deviation of 35), it is agreed that granite has higher radon emanation coefficients than other examined marble, ceramic and porcelain samples. The mean radon emanation coefficient estimated for the examined materials (Fig. 5) were 0.18, 5, 12 and 14% in porcelain, ceramic, marble and granite, respectively. The values observed here are in good agreement with the corresponding values reported in the literature [12, 32] as shown in the figure.

However, the radon exhalation rates ranged from a non-detectable (ND) value to a maximum value of 0.196 Bq/kg.h observed in the granite sample (G24). Many marble and ceramic samples and half of the porcelain samples did not show any detectable radon exhalation, while higher values occurred among granite samples. The mean radon exhalation rate for granite was 0.061 Bq/kg.h, while much lower mean values were observed as 0.016, 0.025 and 0.002 Bq/kg.h for marble, ceramic and porcelain, respectively. This evidence confirms the theory about the difficulties facing radon to escape from condensed solid matrices such as ceramic and porcelain mainly due to reduction of the grain micro-porosity during the manufacturing process that blocks radon emanation. Although high

radium activity concentrations are potentially associated with both ceramic and porcelain, it is highly likely that radon produced through the decay of such radium remains locked up in glass-like matrices [33]. Generally, the natural stones of magmatic origin such as granites are considered as significant sources of radon emanation [3]. The results also confirm that no correlation is present between radon exhalation rate and material radium content and that the emanation rate is not constant within a given type of material or from one type to another with approximately the same radium content.

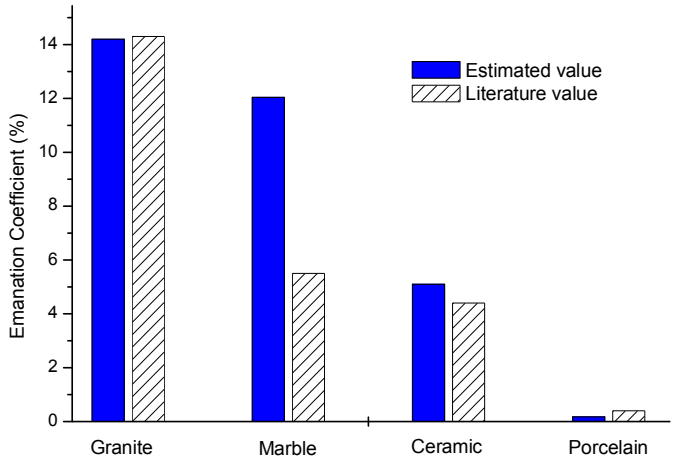


FIG. 5. Mean radon emanation coefficient estimated for the examined materials.

To assess the contribution of superficial materials to indoor radon concentration, a 5×4×3 $m^3$ sealed room with its entire floor tiled with one of the studied materials, is assumed. The radon concentration $C_{Rn}$ ($Bq/m^3$) due to radon exhalation from the floor can be obtained by [36]:

$$C_{Rn} = \frac{E_x . A}{(\lambda_{Rn} + \lambda_v)V} \quad (8)$$

where $A$ is the floor area ($m^2$), $V$ is the room volume ($m^3$), $\lambda_{Rn}$ is the radon decay constant ($h^{-1}$), $\lambda_v$ is the air ventilation rate ($h^{-1}$) and $E_x$ is the radon exhalation rate ($Bq.m^{-2}.h^{-1}$) which is given by [18]:

$$E_x = \frac{1}{2} A_{Ra} . \lambda_{Rn} . \rho . F . d \quad (9)$$

where $\rho$ is the material density ($kg.m^{-3}$) and $d$ is the material or tile thickness (m).

Based on the measured values of $A_{Ra}$, $\rho$ and F, the values of $E_x$ were calculated for two samples of each material examined: one with the highest radium activity concentration and the second with the highest radon emanation coefficient. Assuming that outdoor air has a negligible radon concentration, the calculated $E_x$ values were used to estimate the indoor radon concentration released by 20 $m^2$ floor tiles into a 60 $m^3$ room for various air change rates (ACH) as shown in Table 7. It can be seen from the table that the data obtained ranged from a minimum value of 1.3 $Bq/m^3$ for porcelain tiles up to a maximum value of 326 $Bq/m^3$ for granite tiles.

TABLE 7. Radon concentration ($Bq.\ m^{-3}$) due to radon exhalation from selected floor tile samples, for different air changes per hour (ACH).

| Sample Code | Density ($kg.m^{-3}$) | Thickness (m) | $E_x$ ($Bq.m^{-2}.h^{-1}$) | Radon concentration ($Bq.m^{-3}$) | | |
|---|---|---|---|---|---|---|
| | | | | ACH = 0 | ACH = 0.5 | ACH = 1 |
| G19 | 2684 | 0.03 | 7.41 | 326.42 | 4.87 | 2.45 |
| G8 | 2560 | 0.03 | 3.10 | 136.56 | 2.04 | 1.00 |
| M18 | 2545 | 0.03 | 0.89 | 39.21 | 0.60 | 0.30 |
| M11 | 2502 | 0.03 | 1.27 | 55.94 | 0.83 | 0.42 |
| C7 | 2311 | 0.01 | 1.11 | 48.90 | 0.73 | 0.37 |
| C19 | 2284 | 0.01 | 0.41 | 18.10 | 0.27 | 0.14 |
| P12 | 2412 | 0.008 | 0.03 | 1.3 | 0.02 | 0.01 |
| P6 | 2368 | 0.008 | 0.03 | 1.3 | 0.02 | 0.01 |

## Conclusions

This study presented data concerning the activity concentration of the natural radionuclides and radon emanation coefficients for different types of covering and decorative building materials. The mean activity concentration values of $^{226}Ra$, $^{232}Th$ and $^{40}K$ measured in marble samples were significantly lower than the typical world average values, while those measured in granite, ceramic and porcelain samples were higher than the quoted values, but comparable with values measured in other countries [31]. The isotopic gamma analysis and radiological assessment of the examined materials indicated that some granite samples have the potential to significantly influence indoor external gamma dose level. Radium equivalent activity values for all of the examined materials were lower than the international recommended maximum value of 370 Bq/kg, except for two granite samples, where $Ra_{eq}$ was found to be 433 and 565 Bq/kg. The annual indoor effective doses received by an individual from the examined samples did not exceed the exemption dose criterion of 0.3 mSv.

The results also show that all the examined materials, apart from granite, appear to have low radon emanation coefficients. Although most of the ceramic and porcelain samples have shown high activity concentrations of radium, their emanation coefficients are generally law. Their contribution to indoor radon concentrations is expected to be negligible, similar to that of marble, but still have the potential to significantly influence indoor external gamma dose rate. In general, for houses with adequate ventilation, all covering and decorative materials make no significant contribution to indoor radon and gamma radiation constitutes their major contribution to the indoor dose. Although the values obtained for radon concentration were calculated under the worst case scenario with key parameters ($^{226}Ra$ content and F) having their highest values, it is safe to anticipate that the contribution to indoor radon concentrations is not likely to be a health concern in most cases, especially when compared with the international action limit of 200 $Bq/m^3$ [35].

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