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## Natural Radioactivity Levels and Radiation Hazards for Gypsum Materials Used in Egypt

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### ABSTRACT

The gypsum of building materials which are one of the potential sources of indoor radioactivity because of the naturally occurring radionuclides in them. External as well as internal exposures are the two pathways of radiation dose imparted to the human beings from the building materials. Natural radioactivity levels of 18 samples of natural gypsum and manufactured in Egypt have been investigated by using gamma spectrometer with HPGe detector. The samples were collected from local market in Egypt. From the measured  $\gamma$ -ray spectra, activity concentrations were determined. The activity ranged from  $10.7 \pm 3.4$ - $170.4 \pm 5.1$  Bq kg<sup>-1</sup> for <sup>226</sup>Ra, with mean  $91.97$  Bq kg<sup>-1</sup> and from  $3.1 \pm 0.7$ - $70.8 \pm 3.6$  Bq kg<sup>-1</sup> for <sup>232</sup>Th with mean  $42.27$  Bq kg<sup>-1</sup> and from  $301.3 \pm 6.3$ - $689.1 \pm 8.7$  Bq kg<sup>-1</sup> <sup>40</sup>K with mean  $499.28$  Bq kg<sup>-1</sup>. The radium equivalent activity  $R_{eq}$ , the external hazard index  $H_{ex}$  and the absorbed dose rate in air  $D$  in each sample was evaluated to assess the radiation hazard for people living in dwelling made of materials studied. Gypsum sample have radium equivalent dose  $R_{eq}$  ranged from  $38.41$  to  $324.7$  Bq kg<sup>-1</sup>. These values are lower than the limit of  $370$  Bq kg<sup>-1</sup> adopted by OECD the Organization for Economic Cooperation and Development. The absorbed dose rate in indoor air are lower than the international recommended values of  $55$  n Gy h<sup>-1</sup> in sample G1, G2, G11, G17 and G18. The sample G3, G4, G5, G6, G7, G8, G9, G10, G12, G13, G14, G15 and G16 are higher than of  $55$  n Gy h<sup>-1</sup> this samples are not acceptable for use as building materials as defined by the OECD criterion.

**Key words:** Gypsum, gamma spectrometer, building materials

### INTRODUCTION

Gypsum is a naturally occurring mineral that is made up of calcium sulfate and water (CaSO<sub>4</sub>·2H<sub>2</sub>O) that is sometimes called hydrous calcium sulfate. It is the mineral calcium sulfate with two water molecules attached. By weight it is 79% calcium sulfate and 21% water. Gypsum has 23% calcium and 18% sulfur and its solubility is 150 times that of limestone, hence it is a natural source of plant nutrients. Gypsum naturally occurs in sedimentary deposits from ancient sea beds. Gypsum is mined and made into many products like drywall used in construction, agriculture and industry. It is also a by-product of many industrial processes. Gypsum is also used as a generic name for many types of sheet products made of a non-combustible core with a study surfacing that adds strength. These include drywall, ceiling tiles, partitions, etc., whose strength is directly related to its thickness and a few trace materials. Gypsum has been known for centuries as a building material. The earliest known use of gypsum as a building material was in Anatolia around 6000 B.C.

Gypsum has been found on the interiors of the great pyramids in Egypt which were erected in about 3700 B.C. Gypsum is found on every continent in the world and is one of the most widely used minerals. Gypsum mines are located all across North America and some Western States contain huge deposits of powdery gypsum rock.

Synthetic gypsum is generated as a byproduct in Flue-Gas Desulfurization (FGD) systems used to reduce sulfur dioxide emissions from coal-fired electric powerplants. These FGD systems not only keep the air clean, but, they also can provide a sustainable, ecologically sound source of very pure gypsum. Synthetic gypsum also is generated by various other acid-neutralizing processes. In the United States, most gypsum is used to manufacture wallboard and plaster for homes, offices and commercial buildings. An average new American home contains more than 7.31 metric tons (t) of gypsum or, in other terms, more than 571 m<sup>2</sup> (6,144 square feet) of gypsum wallboard (Mineral Information Institute, 2001). Worldwide, gypsum is used in Portland cement which is used in concrete for bridges, buildings, highways and many other structures that are part of our everyday life. Gypsum also is extensively used as a soil conditioner on large tracts of land in suburban areas and in agricultural regions.

## **TYPES AND SOURCES OF GYPSUM**

There are several types of naturally occurring gypsum and many industrial processes also produce gypsum as a by-product of their systems such as phosphoric acid and citric acid manufacture. Mined Gypsum Mined gypsum is found at various locations around the world. In North America there are gypsum deposits from Canada to Texas and in many Western States. Chemically raw mined gypsum is primarily calcium sulfate hydrated with water molecules in its chemical structure. Other materials and chemicals in mined gypsum may be small amounts of sand or clay particles and a few trace elements. The trace elements may be boron or iron to arsenic and lead and varies with each deposit. Many deposits in Canada have arsenic while those in Texas may have very little. Primarily mined gypsum is very safe to use and a great amendment for many soils.

## **MATERIALS AND METHODS**

Gypsum samples were collected at a total of 18 samples of natural and manufactured building materials used in Egypt. The gypsum samples were dried, pulverized and packed in 200 g by mass in cylindrical plastic container of radius 3 inch and height 3.5 inch which sits on the 3×3 inch HPGe detector with high geometry. The containers are sealed for about four weeks to ensure radioactive equilibrium between the parent radionuclides and their gaseous daughter decay products in the uranium and thorium series (Jeambrun *et al.*, 2012).

Each sample containing soil grain weighing about 200 g was stored in standardized polyethylene containers. The containers were sealed to avoid any possibility of out gassing of radon and kept for a period of 1 month to make sure the samples attained the radioactive equilibrium between <sup>226</sup>Ra with its decay products in the uranium series. It was assumed that <sup>232</sup>Th is in secular equilibrium with <sup>226</sup>Ra. For the measurement of activity concentrations of naturally occurring radionuclides of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in soil samples, a High Purity Germanium (HPGe) detector based gamma ray spectrometer with a relative efficiency of 30% was employed. HPGe detector was coupled with a Canberra multichannel analyzer (MCA). The resolution (FWHM) of the spectrometry system was 1.8 at 1332 keV gamma-ray line of <sup>60</sup>Co. Spectrum of every sample was collected for 54000 sec (15 h). Spectrum analysis was performed with computer software and activity concentrations of 3 natural radionuclides were determined. To reduce the background

effect, the detector was shielded in a 10 cm wall lead covering lined with 2 mm copper and 2 mm cadmium foils. The concentration of  $^{238}\text{U}$  was estimated from 1.76 MeV transition line of  $^{214}\text{Bi}$  while that of  $^{232}\text{Th}$  was estimated from 2.615 MeV of  $^{208}\text{Tl}$  and gamma energy value of 1.465 MeV was used to determine the concentration of  $^{40}\text{K}$  in all the samples. In order to determine the background radiation distribution in the environment around the detector, an empty sealed container was counted for 10 h (Jonsson, 2010).

## RESULTS AND DISCUSSION

**Norm in building materials:** For  $^{238}\text{U}$ , a gamma-ray line of energy 1001.03 keV, of 234 MPa, was used to find the concentrations. Gamma-ray lines of energies 295.09, 351.87, 609.31, 1120.27 and 1764.49 keV resulting from the decay of daughters  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  radionuclides were used to determine the activity concentrations of  $^{226}\text{Ra}$  which it is in secular radioactivity equilibrium with its short half-life daughters; the gamma-ray lines at 911.07, 968.97, 583.10 and 2614.48 keV from the decay of short half-life daughters  $^{228}\text{Ac}$  and  $^{208}\text{Tl}$  were used to determine the activity concentrations of  $^{232}\text{Th}$ , respectively (since there is secular radioactivity equilibrium in  $^{232}\text{Th}$  series); while the 1460.80 transition was used to determine the activity concentrations of  $^{40}\text{K}$ .  $^{235}\text{U}$  and  $^{226}\text{Ra}$  emit gamma rays of energy 185.7 keV (57.2%) and 186.2 keV (3.6%), respectively. The detector energy resolution is not sufficient to easily separate these peaks. Therefore, the concentrations of  $^{235}\text{U}$  was calculated by subtracting the fraction of  $^{226}\text{Ra}$  using Eq. 1 (Ebaid, 2010):

$$A_{\text{U-235}} = [1.75(\text{CR}_{\text{T-186}})/\epsilon_{186}] - 0.063A_{\text{Ra-226}} \quad (1)$$

Where:

- $\text{CR}_{\text{T}}$  = Total count rate (counts  $\text{sec}^{-1}$ ) in the 186 keV energy peak
- $\epsilon$  = Detection efficiency
- $A_{\text{Ra-226}}$  = Activity concentration of  $^{226}\text{Ra}$

Determination of activity concentrations was calculated using the Eq. 2 (Amrani and Tahtat, 2001):

$$A = C/M\beta\epsilon \quad (2)$$

Where:

- $C$  = Net peak area of specific gamma ray energy (count  $\text{sec}^{-1}$ )
- $M$  = Mass of the samples (kg)
- $B$  = Transition probability of gamma-decay
- $\epsilon$  = Detector efficiency at the specific gamma-ray energy

During the last three decades, there has been an increasing interest in the study of the radioactivity of different building materials. Several national surveys were conducted to establish the radioactivity concentrations in raw material, industrial by-products and building materials and their radon exhalation rate (EC, 1999). The production process and the origin of the raw materials are the most important factors that determine the radionuclide activity concentrations in the construction materials. Different types of building materials were found to contain radionuclide concentration of over two to three orders of magnitude.

In order to compare the specific activities of materials containing different amounts of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , an index, called the radium equivalent concentration  $R_{\text{eq}}$  was defined based on the fact that  $370 \text{ Bq kg}^{-1}$  of  $^{226}\text{Ra}$ ,  $259 \text{ Bq kg}^{-1}$  of  $^{232}\text{Th}$  and  $4810 \text{ Bq kg}^{-1}$  of  $^{40}\text{K}$  produce the same gamma dose rate. Therefore, the  $R_{\text{eq}}$  of building material can be written as:

$$R_{\text{eq}} = C_{\text{Ra}} + 1.43C_{\text{Th}} + 0.077C_{\text{K}} \quad (3)$$

where,  $C_{\text{Ra}}$ ,  $C_{\text{Th}}$  and  $C_{\text{K}}$  are the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , respectively, in  $\text{Bq kg}^{-1}$ . The raw materials commonly used in the construction industry may contain NORM at different activity concentrations according to their place of origin.

**$^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  concentration in building materials:** The activity concentrations of NORM in building materials vary according to the type and origin of the building material. The typical activity concentration [ $\text{Bq kg}^{-1}$ ] in the most common building materials in Europe, e.g., concrete and sand-lime bricks is 40, 30 and 400 and 10, 10, 330 for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , respectively (EC, 1999). (Table 1-2) Previous typical values of NORM in building materials used, both as structural materials or covering layers. Table 3 the activity concentration range ( $\text{Bq kg}^{-1}$ ) of gypsum in some countries (EC, 1999). Table 4 Compare the activity concentration range ( $\text{Bq kg}^{-1}$ ) of gypsum in Egypt Previous and present works (Table 5).

**Radon exhalation from in bilding materials:** The radon emanation power or emanation coefficient, denoted by  $\zeta$ , is defined as the fraction of  $^{222}\text{Rn}$  produced by the disintegration of  $^{226}\text{Ra}$  in the grains of the material that can escape from it. The emanation power is dimensionless and ranges from 0 (no radon escapes from the material) to 1 (all radon escapes). The rate of radon exhalation is proportional to the gradient of the radon concentration in the internal pores:

$$E = -D \cdot dC/dx|_{x=1} \quad (4)$$

Table 1: Concentrations in common building materials and industrial by-products used for building materials in Europe (EC, 1999)

| Material                          | $^{226}\text{Ra}$ | $^{232}\text{Th}$ | $^{40}\text{K}$ |
|-----------------------------------|-------------------|-------------------|-----------------|
| <b>Typical activity</b>           |                   |                   |                 |
| Concrete                          | 40                | 30                | 400             |
| Aerated and light-weight concrete | 60                | 40                | 430             |
| Clay (red) bricks                 | 50                | 50                | 670             |
| Sand-lime bricks                  | 10                | 10                | 330             |
| Natural building stones           | 60                | 60                | 640             |
| Natural gypsum                    | 10                | 10                | 80              |
| <b>Maximum activity</b>           |                   |                   |                 |
| Concrete                          | 240               | 190               | 1600            |
| Aerated and light-weight concrete | 2600              | 190               | 1600            |
| Clay (red) bricks                 | 200               | 200               | 2000            |
| Sand-lime bricks                  | 25                | 30                | 700             |
| Natural building stones           | 500               | 310               | 4000            |
| Natural gypsum                    | 70                | 100               | 200             |

Table 2: Most common industrial by-products used in building materials in Europe (EC, 1999)

| Country          | $^{40}\text{K}$ | $^{232}\text{Th}$ | $^{226}\text{Ra}$ | References                                    |
|------------------|-----------------|-------------------|-------------------|-----------------------------------------------|
| China            | 35.00           | 35.00             | 370               | Erdtmann and Soyka (1979) and Langmuir (1978) |
| Nordic countries | 40.00           | 49.00             | 80                | Erdtmann and Soyka (1979)                     |
| West Germany     | 14.00           | 18.50             | 259               | Ahmed (2004)                                  |
| Finland          | 37.00           | 43.00             | 1034              | Ahmed (2004)                                  |
| Bangladesh       | 88.10           | 68.20             | 256.4             | Mollah <i>et al.</i> (1996)                   |
| Spain            | 14.06           | 17.39             | 266.77            | IAEA (1994)                                   |
| Italy            | 67.00           | 138.00            | 580               | Pelleceion (1985)                             |
| USSR             | 14.80           | 140.80            | 259               | Ahmed (2004)                                  |
| India            | 22.00           | 9.30              | 233               | Ahmed (2004)                                  |

Table 3: Compare the activity concentration range ( $\text{Bq kg}^{-1}$ ) of gypsum in some countries

| Material           | Typical activity concentrations |                   |                 | Maximum activity concentrations |                   |                 |
|--------------------|---------------------------------|-------------------|-----------------|---------------------------------|-------------------|-----------------|
|                    | $^{226}\text{Ra}$               | $^{232}\text{Th}$ | $^{40}\text{K}$ | $^{226}\text{Ra}$               | $^{232}\text{Th}$ | $^{40}\text{K}$ |
| Phosphogypsum      | 390                             | 20                | 60              | 1100                            | 160               | 300             |
| Blast furnace slag | 270                             | 70                | 240             | 2100                            | 340               | 1000            |
| Coal fly ash       | 180                             | 100               | 650             | 1100                            | 300               | 1500            |

Table 4: Compare the activity concentration range ( $\text{Bq kg}^{-1}$ ) of gypsum in Egypt Previous and present works

| Countries | $^{226}\text{Ra}$ | $^{232}\text{Th}$ | $^{40}\text{K}$ | References      |
|-----------|-------------------|-------------------|-----------------|-----------------|
| Egypt 1   | 91.97             | 42.27             | 499.28          | Present work    |
| Egypt 3   | 105.00            | 45.00             | 500.00          | Ahmed (2005)    |
| Egypt 4   | 31.70             | 55.00             | 116.00          | El-Taher (2010) |

Table 5: Activity concentration range ( $\text{Bq kg}^{-1}$ ) of gypsum in Egypt current works

| Sample | $A_U (\text{Bq kg}^{-1})$ | $A_{ra} (\text{Bq kg}^{-1})$ | $A_{th} (\text{Bq kg}^{-1})$ | $A_k (\text{Bq kg}^{-1})$ |
|--------|---------------------------|------------------------------|------------------------------|---------------------------|
| G1     | 7.50                      | 10.70                        | 3.10                         | 307.50                    |
| G2     | 9.70                      | 14.50                        | 11.70                        | 365.60                    |
| G3     | 43.20                     | 119.30                       | 59.80                        | 681.30                    |
| G4     | 23.50                     | 87.60                        | 41.40                        | 433.20                    |
| G5     | 48.30                     | 170.40                       | 70.80                        | 689.10                    |
| G6     | 34.60                     | 75.30                        | 39.30                        | 417.70                    |
| G7     | 74.80                     | 136.20                       | 61.20                        | 528.40                    |
| G8     | 37.90                     | 121.90                       | 49.10                        | 564.30                    |
| G9     | 22.10                     | 138.10                       | 56.70                        | 531.30                    |
| G10    | 61.90                     | 97.70                        | 67.40                        | 678.10                    |
| G11    | 19.40                     | 23.60                        | 23.80                        | 349.20                    |
| G12    | 49.70                     | 74.00                        | 52.90                        | 689.10                    |
| G13    | 61.90                     | 121.10                       | 58.10                        | 431.90                    |
| G14    | 74.60                     | 148.30                       | 63.80                        | 654.60                    |
| G15    | 10.40                     | 132.50                       | 16.50                        | 301.30                    |
| G16    | 27.20                     | 116.70                       | 31.30                        | 407.70                    |
| G17    | 25.90                     | 39.80                        | 18.20                        | 517.40                    |
| G18    | 44.60                     | 27.80                        | 35.80                        | 439.30                    |
| Min    | 7.50                      | 10.70                        | 3.10                         | 301.30                    |
| Max    | 74.80                     | 170.40                       | 70.80                        | 689.10                    |
| Mean   | 37.62                     | 91.97                        | 42.27                        | 499.28                    |

where, D is the effective diffusion coefficient and  $dC/dx$  is the change in the radon concentration. The principal factors affecting the radon exhalation rate (from a building material) per unit activity concentration of  $^{226}\text{Ra}$  are the porosity and the density of the material, the diffusion coefficient, the water content, the age and the composition of the material (Eq. 3).

For the following boundary conditions:  $C(l) = C(-l) = 0$  and  $(Dx/dC) = 0$  the radon exhalation rate E:

$$E = \zeta \cdot C_{\text{Ra}} \cdot \rho \cdot \sqrt{\lambda D / P} \cdot \tanh(\sqrt{\lambda D / P} \cdot l) \quad (5)$$

where,  $\rho$  is the density,  $l$  is the half thickness of the material,  $p$  is the porosity of the material,  $\lambda$  is the decay constant of  $^{222}\text{Rn}$  and  $\zeta C_{\text{Ra}}$  is the effective radium concentration (the fraction of the total radium which contributes to radon exhalation).

Radon exhalation from building materials has been studied since the early 70's as one of the contributors to the indoor radon concentration. In Hong Kong and the Netherlands was found to be the major contributor to the population radon dose (Stoop *et al.*, 1998; Yu *et al.*, 1992a). The radon exhalation rate from concrete varies according to the age of the concrete, the water content and the addition of fly ash. The exhalation increases almost linearly with the moisture content up to 50-60%, peaks at 70-80% and decreases steeply for higher moisture levels. The addition of fly ash to concrete generally increases the  $^{226}\text{Ra}$  activity while the radon exhalation rate slightly increases or even decreases (Roelofs and Scholten, 1994; Strandén *et al.*, 1984; Kovler *et al.*, 2005).

Several methods for exhalation measurements have been developed, as opposed to the well established gamma spectrometry procedures for the measurements of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ . Measurements of radon from the exhaling area into a closed chamber, purge and trap of the radon, radon flux measurements from the material surface and *in-situ* measurements have been reported.

**Absorbed dose rate measurement:** The absorbed dose rate (D) in air at a height of 1.0 m above the ground from the radionuclides  $^{40}\text{K}$  and also  $^{232}\text{Th}$  and the  $^{238}\text{U}$  decay series, were calculated using Eq. 4, if the naturally occurring radionuclides are uniformly distributed (Beck *et al.*, 1972; Yu *et al.*, 1992b).

$$D = 0.52813 C_{\text{Th}} + 0.38919 C_{\text{Ra}} + 0.03861 C_{\text{K}} \quad (6)$$

where,  $C_{\text{Ra}}$ ,  $C_{\text{Th}}$  and  $C_{\text{K}}$  are the activity concentration of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in ( $\text{Bq kg}^{-1}$ ), respectively. Annual effective dose rates values of the absorbed dose rate were used for the calculation of annual outdoor effective dose rate considering some correction factors. A conversion factor ( $\text{CF} = 0.7 \text{ Sv/Gy}$ ) was applied for conversion of the absorbed dose in air to the corresponding effective dose. The outdoor occupancy factor ( $\text{OF} = 0.8$ ) was taken into account, since the people spend most of their time (80%) in buildings. Finally, the ratio of indoor to outdoor gamma dose rates ( $R = 1.4$ ) was also applied as used by other investigators (Colgan *et al.*, 2008) to calculate the annual indoor effective gamma dose rate (DE) in ( $\text{Sv/y}$ ) as given follow:

$$\text{DE} = D \times \text{CF} \times \text{OF} \times R \times t \quad (7)$$

where, D is the dose rate in ( $\text{Gy h}^{-1}$ ) and  $t$  is the duration of the exposure ( $t = 365 \times 24 \text{ h}$ ). Radium equivalent activity the non-uniformity of natural nuclides concentration in building material

samples would cause in definition of an index the so-called radium equivalent activity ( $R_{eq}$ ), (Baykara *et al.*, 2011; Rizzo *et al.*, 2001) for taking into account the radiation hazards of radioactive nuclides. This quantity is calculated as given below using the activity concentration of the radionuclides (Hayumbu *et al.*, 1995):

$$R_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (8)$$

where,  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the activity concentration of  $^{226}Ra$ ,  $^{232}Th$  and  $^{40}K$  in ( $Bq\ kg^{-1}$ ), respectively. This index ( $R_{eq}$ ) is related to both internal dose due to radon and external gamma dose (Kovler *et al.*, 2005) and should have the highest value of  $370\ Bq\ kg^{-1}$  for safe use of the building materials. Hazard indexes for external gamma radiation two indexes are used in this study for assessment of excess gamma radiation from the building materials in order to be sure of the safety of building material usage. Beretka and Mathew introduced a hazard index for the external gamma radiation dose from building materials as given bellow (Table 6).

**External hazard index:** The external hazard index is obtained from  $R_{eq}$  expression through the supposition that its maximum value allowed (equal to unity) corresponds to the upper limit of  $R_{eq}$  ( $370\ Bq\ kg^{-1}$ ). This index value must be less than unity in order to keep the radiation hazard insignificant; i.e., the radiation exposure due to the radioactivity from construction materials is limited to  $1.0\ mSv\ y^{-1}$ . Then, the external hazard index can be defined as:

$$H_{ex} = (C_U/370 + C_{Th}/259 + C_K/4810) < 1 \quad (9)$$

Table 6: Absorbed dose rate ( $Bq\ kg^{-1}$ ) and Radium equivalent activity of gypsum in Egypt

| Sample | $R_{eq}\ (Bq\ kg^{-1})$ | D ( $nGy\ h^{-1}$ ) | $I_{RF112}\ (mSv\ y^{-1})$ |
|--------|-------------------------|---------------------|----------------------------|
| G1     | 38.81                   | 19.69               | 0.15                       |
| G2     | 59.38                   | 29.21               | 0.21                       |
| G3     | 257.27                  | 120.66              | 0.64                       |
| G4     | 180.16                  | 84.25               | 0.40                       |
| G5     | 324.70                  | 151.43              | 0.71                       |
| G6     | 163.66                  | 76.61               | 0.44                       |
| G7     | 264.40                  | 122.96              | 0.75                       |
| G8     | 235.56                  | 110.34              | 0.54                       |
| G9     | 260.09                  | 121.17              | 0.48                       |
| G10    | 246.30                  | 115.27              | 0.76                       |
| G11    | 84.52                   | 40.24               | 0.29                       |
| G12    | 202.71                  | 95.77               | 0.65                       |
| G13    | 237.44                  | 110.04              | 0.65                       |
| G14    | 289.94                  | 135.43              | 0.80                       |
| G15    | 179.30                  | 84.03               | 0.21                       |
| G16    | 192.85                  | 90.35               | 0.38                       |
| G17    | 105.67                  | 51.27               | 0.36                       |
| G18    | 112.82                  | 53.39               | 0.49                       |
| Min    | 38.81                   | 19.69               | 0.15                       |
| Max    | 324.70                  | 151.43              | 0.80                       |
| Mean   | 190.87                  | 89.56               | 0.50                       |

$$H_{in} = (C_U/185 + C_{Th}/259 + C_K/4810) < 1 \quad (10)$$

where,  $A_{Ra}$ ,  $A_{Th}$  and  $A_K$  are the specific activities of  $^{226}Ra$ ,  $^{232}Th$  and  $^{40}K$  in  $Bq\ kg^{-1}$ , respectively. The calculated values of external hazard index obtained in this study ranged from 0.12-0.79. Since, these values are lower than unity, we can say that the radiation hazard is insignificant for the population living in the investigated area. From the results it is evident that there are considerable variations in the  $R_{eq}$  of the different materials and also within the same type of materials originating from different areas. This fact is important from the point of view of selecting suitable materials for use in building and construction especially concerning those which have large variations in their activities. Large variation in radium equivalent activities may suggest that it is advisable to monitor the radioactivity levels of materials from a new source before adopting it for use as a building material. The recommended maximum levels of radium equivalents for building materials to be used for homes is  $<370\ Bq\ kg^{-1}$  and for industries is  $370-740\ Bq\ kg^{-1}$ . All the materials examined are acceptable for use as building materials as defined by the OECD criterion (OECD, 1979).

**Representative gamma index ( $I_\gamma$ ) and ( $I_a$ ):** Number of indices dealing with the assessment of the excess gamma radiation arising from building materials such as external and internal hazard indices and gamma concentration indices has been proposed by several investigators (EC, 1999; ICRP, 1994; Khan *et al.*, 2002; Komura, 1997). In this study, the gamma index ( $I_\gamma$ ) was calculated as proposed by the EC (1999):

$$I_\gamma = C_{Ra}/150 + C_{Th}/100 + C_K/1500\ (Bq\ kg^{-1}) \quad (11)$$

where,  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the activity concentrations of  $^{226}Ra$ ,  $^{232}Th$  and  $^{40}K$  in  $Bq\ kg^{-1}$ , respectively. The mean values of  $I_\gamma$  calculated from the measured activity concentration of  $^{226}Ra$ ,  $^{232}Th$  and  $^{40}K$  are presented in Table 7 for different gypsum samples. The mean calculated values of  $I_\gamma$  for the studied samples from gypsum samples varied in the range between 0.61-2.43  $Bq\ kg^{-1}$  which were less than the critical value of unity. So far, several alpha indices have been proposed to assess the exposure level due to radon inhalation originating from building materials (EC, 1999). The alpha index was determined by the following equation:

$$I_\alpha = C_{Ra}/200\ (Bq\ kg^{-1}) \quad (12)$$

where,  $C_{Ra}$  ( $Bq\ kg^{-1}$ ) is the activity concentration of  $^{226}Ra$  assumed in equilibrium with  $^{238}U$ . The recommended exemption and upper level of  $^{226}Ra$  activity concentrations in building materials are 100 and 200  $Bq\ kg^{-1}$ , respectively, as suggested by ICRP (1994). These considerations are reflected in the alpha index. The recommended upper limit concentration of  $^{226}Ra$  is 200  $Bq\ kg^{-1}$ , for which  $I_\alpha = 1$ . The mean computed  $I_\alpha$  values for the studied gypsum samples are given in Table 7 for the different gypsum samples. The values of  $I_a$  of gypsum sample are values varied in the range between 0.07-0.71 ( $Bq\ kg^{-1}$ ) with mean 0.26 ( $Bq\ kg^{-1}$ ). For the safe use of a material in the construction of dwellings,  $I_a$  should be less than unity. The mean calculated values were less than unity.

Table 7: External and internal hazard and Igamma, Ialpha of gypsum in Egypt

| Sample | H <sub>ex</sub> | H <sub>in</sub> | e <sub>U</sub> /e <sub>Ra</sub> | Igamma | Ialpha |
|--------|-----------------|-----------------|---------------------------------|--------|--------|
| G1     | 0.10            | 0.12            | 1.43                            | 0.31   | 0.05   |
| G2     | 0.15            | 0.17            | 1.49                            | 0.46   | 0.07   |
| G3     | 0.49            | 0.61            | 2.76                            | 1.85   | 0.60   |
| G4     | 0.31            | 0.38            | 3.73                            | 1.29   | 0.44   |
| G5     | 0.55            | 0.68            | 3.53                            | 2.30   | 0.85   |
| G6     | 0.33            | 0.43            | 2.18                            | 1.17   | 0.38   |
| G7     | 0.55            | 0.75            | 1.82                            | 1.87   | 0.68   |
| G8     | 0.41            | 0.51            | 3.22                            | 1.68   | 0.61   |
| G9     | 0.39            | 0.45            | 6.25                            | 1.84   | 0.69   |
| G10    | 0.57            | 0.74            | 1.58                            | 1.78   | 0.49   |
| G11    | 0.22            | 0.27            | 1.22                            | 0.63   | 0.12   |
| G12    | 0.48            | 0.62            | 1.49                            | 1.48   | 0.37   |
| G13    | 0.48            | 0.65            | 1.96                            | 1.68   | 0.61   |
| G14    | 0.58            | 0.79            | 1.99                            | 2.06   | 0.74   |
| G15    | 0.15            | 0.18            | 12.74                           | 1.25   | 0.66   |
| G16    | 0.28            | 0.35            | 4.29                            | 1.36   | 0.58   |
| G17    | 0.25            | 0.32            | 1.54                            | 0.79   | 0.20   |
| G18    | 0.35            | 0.47            | 0.62                            | 0.84   | 0.14   |
| Min    | 0.10            | 0.12            | 0.62                            | 0.61   | 0.07   |
| Max    | 0.58            | 0.79            | 12.74                           | 2.43   | 0.72   |
| Mean   | 0.37            | 0.47            | 2.99                            | 1.63   | 0.26   |

#### Index I<sub>RP112</sub>:

**RP112:** Activity concentration index should be used only as a screening tool for identifying materials which might be of concern, but Basis: Within the European Union, doses exceeding 1 mSv y<sup>-1</sup> should be taken into account from the radiation protection point of view:

- Dose criterion = 1 mSv y<sup>-1</sup> (excess average background)
- Occupancy factor = 7000 h y<sup>-1</sup>
- Dose conversion coefficient = 0.7 Sv Gy-1
- Average background = 50 nGy h<sup>-1</sup>~0.25 mSv y<sup>-1</sup>

“Any actual decision on restricting the use of the material should be based on a separate dose assessment”

In 2002, Denmark adopted this index for the exemption of building material (dose criterion 0.3 mSv y-1 and I = 0.5):

$$I_{RP112} = C_{ra}/200 + C_{Th}/300 + C_K/3000 \text{ (mSv y-1)} = 0.5 \quad (13)$$

Table 6 the index value I<sub>RP112</sub> are less than (0.5) in some sample (G1, G4, G6, G10, G11, G12, G17, G18) and the other sample with more than this value.

## CONCLUSION

Materials derived from rock and soil contain mainly natural radioisotopes of the  $^{238}\text{U}$  and  $^{232}\text{Th}$  series and radioactive isotope of  $^{40}\text{K}$ . Gamma ray spectrometry is powerful experimental tool in studying natural radioactivity and determining elemental concentration in various building materials. The radium equivalent activities obtained for the building materials in this study were below the criterion limit of  $\gamma$ -radiation dose ( $370 \text{ Bq kg}^{-1}$ ) adopted by the criterion (OECD, 1979). Therefore, the use of these materials in construction of dwellings is considered to be sometimes not safe for inhabitants. The obtained results show that some of the building materials used in Egypt sometimes have highly level, thus they can be not exempted from all controls concerning their radioactivity. Thus from the radiation safety, these materials are high the recommended limits for their gamma dose rates, therefore, they can not be used for all kinds of republic buildings.

There is disequilibrium between  $^{226}\text{Ra}$  and  $^{238}\text{U}$  in some samples while the equilibrium between  $^{226}\text{Ra}$  daughters is not clear, also there is equilibrium between  $^{232}\text{Th}$  daughters. The Radium Equivalent ( $\text{Bq kg}^{-1}$ ), Absorbed Dose Rate ( $\text{nGy h}^{-1}$ ), Annual Effective Dose ( $\text{mSv h}^{-1}$ ), External Hazard Index are higher than the respective world's average. The results can be considered as base values for distribution of natural radionuclides in the area and will be used as reference information for determining any future changes.

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