NATURAL RADIOACTIVITY MEASUREMENTS IN LOCAL AND IMPORTED COMMERCIAL GRANITES USED AS ORNAMENTAL STONES

ABDEL MOEZ A. SADEK; ABDEL HADI A. ABBAS and ANAS M. EL-SHERIF

ABSTRACT

$^{238}\text{U}$, $^{232}\text{Th}$, $^{226}\text{Ra}$, and $^{40}\text{K}$ radioactivities were determined in commercial granite samples collected from local market using a NaI (Tl) gamma-ray spectroscopy system. $^{238}\text{U}$, $^{232}\text{Th}$, $^{226}\text{Ra}$, and $^{40}\text{K}$ activity concentrations of the studied commercial granites range between 31 and 351.33 BqKg$^{-1}$, 22.2 to 102.35 BqKg$^{-1}$, 22.2 to 168.35 BqKg$^{-1}$ and 881.49 to 1460.67 BqKg$^{-1}$, respectively. The highest values of activity concentrations of $^{238}\text{U}$, $^{226}\text{Ra}$ and $^{40}\text{K}$ were recorded in Lahm Mafroum commercial granites. High radioactivity of Lahm Mafroum is attributed to the presence of meta-autonite, zircon, allanite, monazite, sphene and apatite.

Absorbed Dose Rate (D), annual effective dose equivalent (AEDE), radium equivalent activity (Raeq), external (Hex) and internal (Hin) hazard index, in addition to activity gamma index ($I_{\gamma}$) caused by gamma emitting natural radionuclide are determined from the obtained values of $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$. Fairly, many of the studied commercial granites do not satisfy the universal standards.

INTRODUCTION

Usually granites have a commercial name, petrographic and technological characterizations, and identification of the producer country (Abirochas, 2003; Pivko, 2003; Anjos et al., 2005). 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 with marble. Distinct types of commercial granites have different geographical origins and mineralogical compositions and may be either magmatic or metamorphic rocks. Thorium, uranium and potassium concentrations of granitic rocks are intimately related to their mineral compositions and general petrologic features (Whitfield et al., 1959; Rogers and Ragland, 1961; Doventon and Prensky, 1992). Uranium and thorium in igneous and metamorphic rocks are usually found in a few accessory minerals such as apatite, sphene and zircon. Other highly radioactive minerals, like monazite, allanite, uraninite, thorite and pyrochlore, are widespread in nature, but they are very minor constituents of rocks. Uranium tends to be highly mobile near the surface whereas thorium is relatively stable. Uranium is easily oxidized to a water-soluble form and can be readily leached from pegmatites and granites and re-deposited in sediments at large distances from the source rock. Thorium is much less soluble than uranium and potassium and does not move except by mechanical means such as wind and erosion processes. Both thorium and uranium contents tend to be high in felsic rocks and to increase with alkalinity or acidity, with their highest concentrations found in pegmatites. The potassium content of rocks also increases...
with acidity. It is usually found in potash feldspars, such as microcline and orthoclase, or in micas, like muscovite and biotite. Rocks that are free of these minerals have very low K-activity. The petrologic features of granitic rocks associated with effects of weathering and metamorphism produce expressive alterations in the relationship between the natural radionuclides (Th, U, K, Th/U and Th/K).

The study of the concentrations and distributions of the natural radionuclides in rock and soil allows the understanding of the radiological implication of these elements due to the gamma ray exposure of the body and irradiation of lung tissue from inhalation of radon and its daughters. In particular, it is also important to assess the radiation hazards arising due to the use of rock and soil in the construction of dwellings. Therefore, it is important to measure the concentration of radionuclides in rocks used as building materials for assessing the radiological risks to human health and for the use and management of these rocks. Several authors have studied the natural radioactivity level of rocks from different places (Xinwei, 2004; Xinwei et al., 2006).

Most of the naturally occurring radionuclides exist in rocks and soils at concentrations that are not of concern to human health or the environment (Elles and Lee 2002). But there are areas with relatively high natural concentrations of U, Ra and Th due to their geological evolution. Natural and anthropogenic accelerated radionuclides mobilization also occurs in areas with high background concentrations (Barnett et al., 2000). U and Th are long-lived radionuclides with a suite of radioactive daughter products which can pose a human-health and ecological risk. Radiation of natural origin is responsible for most of the total radiation exposure to the general population. Quantification of background levels of radionuclides is necessary to evaluate the potential environmental risk, to determine the boundary of areas of high natural background and to establish its cleanup level (Elles and Lee 2002; Taboada et al., 2006; El-Aassy et al., 2012).

**SAMPLING AND EXPERIMENTAL TECHNIQUES**

Thirty six commercial granite samples used as building materials were collected from local markets representing local and imported types. Different locations of the local samples were recorded on Fig.(1). They subjected to radiometric analysis using NaI (Tl) gamma-ray spectrometers for determination of their radioelements concentration and activity.

![Fig. 1 : Different localities of commercial granite (El-Ramly, 1972)](image)

These samples were prepared for gamma-ray spectrometric analysis in order to determine their uranium, thorium, radium and potassium contents by using multichanel analyzer of gamma-ray detector (Gamma-Spectrometer technique). The instrument used in determination of the four radioactive elements consists of a Bicron scintillation detector NaI (Tl) 76x76 mm, hermetically sealed with the photomultiplier tube in aluminum housing. The tube is protected by a copper cylinder protection of thickness 0.6 cm against induced X-ray and a chamber of lead bricks against environmental radiation. Uranium, thorium, radium and potassium are measured by using four energy regions.
representing Th-234, Pb-212, Pb-214 and K-40 at 93 kV, 239 kV, 352 kV, and 1460 kV for uranium, thorium, radium and potassium, respectively. The measurements were carried out in sample plastic containers, cylindrical in shape, 212.6 cm$^3$ volumes with 9.5 cm average diameter and 3 cm height. The rock sample is crushed to about 1 mm grain size, and then the container is filled with about 300-400 gm of the crushed sample sealed well and left for at least 21 days to accumulate free radon to attain radioactive equilibrium. The relation between the percentage of Rn-222 accumulation and time increase till reaching the steady stage after about 38 days (Matolin, 1991).

ASSESSMENT OF DOSE

Absorbed Dose Rate in Air ($D$)

The absorbed gamma dose rates in air at 1m above the ground surface for the uniform distribution of radionuclides ($^{226}$Ra, $^{232}$Th and $^{40}$K) were calculated by using Eq.(1) on the basis of guide lines provided by UNSCEAR (2000) (rg ün et al., 2007).

$$D \text{ (nGy/h)} = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K} \quad (1)$$

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

Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent (AEDE) was calculated from the absorbed dose by applying the dose conversion factor of 0.7 Sv/Gy and the outdoor occupancy factor of 0.2 (UNSCEAR, 2000, rg ün et al., 2007).

Radium Equivalent Activity ($Ra_{eq}$)

The radium equivalent activity for the samples was calculated. The exposure to radiation (Tufail et al., 1992) can be defined in terms of the radium equivalent activity ($Ra_{eq}$), which can be expressed by the following equation:

$$Ra_{eq} = A_{Ra} + 10/7A_{Th} + 10/130A_{K} \quad (2)$$

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

External and Internal Hazard Index ($H_{ex}$ and $H_{in}$)

To limit the annual external gamma-ray dose (Saito and Jacob, 1995; Saito et al., 1998; UNSCEAR, 2000) to 1.5 Gy for the samples under investigation, the external hazard index ($H_{ex}$) is given by the following equation:

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_{K}/4810 \quad (3)$$

The internal exposure to $^{222}$Rn and its radioactive progeny is controlled by the internal hazard index ($H_{in}$), which is given by Nada (2003):

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_{K}/4810 \quad (4)$$

These indices must be less than unity in order to keep the radiation hazard insignificant (Lakehal et al., 2010; Baykara et al., 2010).

Activity Concentration Index ($I_{\gamma}$)

Another radiation hazard index called the representative level index, $I_{\gamma}$, is defined as follows (NEA-OECD, 1979):

$$I_{\gamma} = A_{Ra}/150 + A_{Th}/100 + A_{K}/1500 \quad (5)$$

where $A_{Ra}$, $A_{Th}$ and $A_{K}$ are the activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K, respectively in Bq/kg (Abbady et al., 2005). The safety value for this index is $\leq 1$ (El Galy et al., 2008; El-Aassy et al., 2012).

RESULTS AND DISCUSSION

Most of the studied commercial granite facies (coarse-, and fine-grained granites) have the same mineralogy (quartz, plagioclase, K-feldspar, biotite and muscovite) and only differ in their modal proportions. In the fine-grained facies the amounts of biotite are less than in the coarse-grained facies and muscovite more than biotite. The main difference between facies is textural: the coarse-grained granite can be either porphyritic or equigranular; fine-grained facies always have an equigranular texture and finer grain size. Petrographically, most of the the studied granites are alkali feldspar granites. They are medium-, to coarse-grained and consist essentially of perthites, quartz, oligoclase, microcline and biotite. They are normally of hypidiomorphic
granular texture. Perithes form euhedral microcline and orthoclase perthite crystals. The microcline perthites may dominate strongly over orthoclase perthites. The perthites include string, flame and feather types. Quartz occurs as anhedral grains, interstitial to the perthites. A graphic texture has been observed in few samples. Oligoclases are less abundant than perthites. It occurs as small interstitial crystals between quartz and perthites. Biotite occurs as brown to reddish green subhedral flaky crystals in minor amounts. The flakes are pleochroic and mottled with iron oxides.

**Uranium Bearing Minerals**

Accessory minerals are represented mainly by zircon, allanite, monazite, sphene, apatite and fluorite. Zircon occurs as small subhedral to euhedral crystals. It is also observed as inclusions in quartz, biotite and chlorite. Some zircon crystals are occasionally metamict and surrounded by strong pleochroic haloes due to radiogenic effects (Fig.2). It is sometimes zoned, often associated and rimmed by iron oxides (Fig.3). Allanite occurs as few subhedral to euhedral reddish brown medium-grain size crystals intimately associated with biotite (Fig.4). Monazite occurs as small euhedral to subhedral, prismatic crystals enclosed within biotite and quartz (Fig.5). Their colours vary from pale yellow to yellowish brown. Sphene is found as euhedral sphenoidal shaped characterized by perfect cleavage, usually associated with mafic minerals (Fig.6). Fluorite occurs as colorless or violet anhedral grains. Opaques are disseminated as fine-grained within the mica or feldspar plates.

**Secondary Uranium Minerals**

In the oxidation zone, U$^{4+}$ is oxidized to uranyl ion U$^{6+}$ which is soluble and mobile in solutions and easy to combine with other cations such as Ca$^{2+}$, Cu$^{2+}$ and Pb$^{2+}$, and anions such as those present in the form of sulphates and phosphates tend to form secondary uranium minerals such as uranophane, beta-uranophane, kasolite, torbernite, autunite and meta-autunite. Autunite & Meta-autunite [Ca(UO$_2$)$_2$(PO$_4$)$_2$·2H$_2$O] represents the secondary uranium minerals in highly radioactive type of the studied commercial granites (Lahm Mafroom). The aggregates of autonite and meta-autonite are soft and consist of lemon yellow to

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greenish yellow small crystallites with a micaceous habit. Occasionally meta-autonite is found as tabular square crystals often in parallel growths and terminated by bipyramidal forms. This mineral contains 11.87% P$_2$O$_5$, 46.80% UO$_2$, and the associated minerals contain 2.81% K$_2$O, 31.14% CaO, 0.42% MnO, 1.90% Fe$_2$O$_3$, 2.98% SiO$_2$, 1.59% SO$_3$, and 0.48% Al$_2$O$_3$, as detected by ESEM (Fig.7). The presence of meta-autonite crystals were confirmed by XRD technique (Fig.8).

Environmental Impacts

The average activity concentrations for granite are shown in Table (1), in which $^{238}$U, $^{226}$Ra, $^{40}$K and $^{232}$Th were 351.33 Bq kg$^{-1}$, 168.35 Bq kg$^{-1}$, 1460.67 Bq kg$^{-1}$ for (Lahm Ma-

from) and 102.35 Bq kg$^{-1}$ for (Abradour), respectively. Minimum average activity concentrations of $^{238}$U, $^{232}$Th, $^{226}$Ra and $^{40}$K were 31.00 Bq kg$^{-1}$ for (Rosa Nasr), 22.22 Bq kg$^{-1}$ for (Fard Ghazal), 22.20 Bq kg$^{-1}$ for (Rosa Nasr) and 881.49 Bq kg$^{-1}$ for (Ramady), respectively. Average activity concentrations of the studied commercial granites were lower than activity concentration of commercial Chinese acidic granite (Xinwei et al., 2006) (Table 1). The lowest activity concentrations were recorded in Rosa Nasr and Ramady where the highest values were in Lahm Mafroum, Ahmer Aswan and Al Shaieb. High radioactivity of Lahm Mafroum is attributed to the presence of meta-autonite, zircon, allanite, monazite, sphene and apatite (Figs.2-6).

The average absorbed $\gamma$ dose rate (D) values for commercial granite samples are shown
in Table (1). The values obtained in all the studied samples ranged between 81.50 and 262.27 nGy h\(^{-1}\) with an average 154.00 nGy h\(^{-1}\). These estimated values of absorbed γ dose rate in the studied samples are comparably higher than the world average value 57 nGy h\(^{-1}\) (Tzortzis et al., 2003, Abbady et al., 2005).

Furthermore, the average values of annual effective dose for all commercial granite samples were also listed. The values obtained varied between 0.10 and 0.32 mSv y\(^{-1}\). The mean value (0.19) found to be less than that 0.48 mSv y\(^{-1}\) [recommended by UNSCEAR, 2000] as the worldwide average of the annual effective dose.

Table 1: Results of radionuclide concentrations, the dose rate (D), the annual effective dose equivalent (AEDE), radium equivalent activity (Ra\(^{eq}\)), external (H\(_{ex}\)) and internal (H\(_{in}\)) hazard indices and gamma index (I\(_{\gamma}\)) for samples.

<table>
<thead>
<tr>
<th>S.No.</th>
<th>Location</th>
<th>Commercial Name</th>
<th>eU Bq kg(^{-1})</th>
<th>eTh Bq kg(^{-1})</th>
<th>Ra Bq kg(^{-1})</th>
<th>K Bq kg(^{-1})</th>
<th>D (nGy h(^{-1}))</th>
<th>AEDE (mSv h(^{-1}))</th>
<th>Ra(^{eq}) Bq kg(^{-1})</th>
<th>H(_{ex})</th>
<th>H(_{in})</th>
<th>I(_{\gamma})</th>
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<tbody>
<tr>
<td>1</td>
<td>Ahmer Aswan.1</td>
<td>74.4</td>
<td>56.56</td>
<td>33.3</td>
<td>139.01</td>
<td>128.88</td>
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<td>212.72</td>
<td>0.71</td>
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<td>84.84</td>
<td>344.1</td>
<td>1205.85</td>
<td>296.27</td>
<td>0.56</td>
<td>558.00</td>
<td>1.72</td>
<td>2.86</td>
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<tr>
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<td>56.56</td>
<td>44.4</td>
<td>1424.15</td>
<td>122.19</td>
<td>0.15</td>
<td>234.75</td>
<td>0.68</td>
<td>0.85</td>
<td>1.93</td>
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</tr>
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<td>48.48</td>
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<td>1361.55</td>
<td>114.70</td>
<td>0.14</td>
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<td>0.64</td>
<td>0.81</td>
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<td>66.63</td>
<td>66.6</td>
<td>1203.65</td>
<td>169.47</td>
<td>0.21</td>
<td>263.43</td>
<td>0.97</td>
<td>1.40</td>
<td>2.62</td>
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<td>6</td>
<td>Ahmer Aswan.6</td>
<td>124</td>
<td>66.63</td>
<td>44.4</td>
<td>1314.16</td>
<td>153.59</td>
<td>0.19</td>
<td>245.64</td>
<td>0.87</td>
<td>1.21</td>
<td>2.39</td>
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<td>289.99</td>
<td>0.93</td>
<td>1.34</td>
<td>2.53</td>
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</tbody>
</table>

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Values of eU, eTh and eRa in ppm, as well as K, in %, were converted to activity concentration, Bq kg\(^{-1}\), using the conversion factors given by Polish Central Laboratory for Radiological Protection (Małeckowski et al., 2004; Said et al., 2010).
The radium equivalent activity Ra\textsubscript{eq} for Egyptian and imported commercial granites ranges between 155.66 and 373.05 Bq Kg\textsuperscript{-1} with 264.15 Bq Kg\textsuperscript{-1} as a mean value. These values are lower than the recommended maximum value of 370 Bq kg\textsuperscript{-1}, with the exception of Lahm Mafroum Ra\textsubscript{eq} is significantly higher than the maximum permitted value.

The values of external and internal hazard indices (H\textsubscript{ex} and H\textsubscript{in}) for the studied commercial granites range between 0.45 and 1.51 and 0.53 to 2.45, respectively. External and internal hazard indices are higher than unity for most studied granites indicating that these granites can not be used as building and interior decorative material of dwelling.

The gamma activity index (I\gamma) used to assess safety requirement for building materials were evaluated and presented in Table (1). The obtained values for both of them ranged between 1.29 and 3.96 with 2.38 as an average. The obtained values of gamma activity indices in all commercial granite samples were higher dose criterion (0.3mSv\textsuperscript{-1}) and corresponds to an activity concentration index of 2 ≤ I\gamma ≤ 6 proposed by EC (1999) for materials used in bulk construction.

CONCLUSIONS

Maximum activity concentrations of \textsuperscript{238}U, \textsuperscript{226}Ra, \textsuperscript{40}K and \textsuperscript{232}Th were 351.33 Bq kg\textsuperscript{-1}, 168.35 Bq kg\textsuperscript{-1}, 1460.67 Bq kg\textsuperscript{-1} for (Lahm Mafroum) and 102.35 Bq kg\textsuperscript{-1} for (Abradour), respectively. Minimum concentrations of \textsuperscript{238}U, \textsuperscript{232}Th were 31.00 Bq kg\textsuperscript{-1} for (Rosa Nasr), 22.22 Bq kg\textsuperscript{-1} for (Fard Ghazal), 22.20 Bq kg\textsuperscript{-1} for (Rosa Nasr) and 881.49 Bq kg\textsuperscript{-1} for (Ramady), respectively. The highest values of activity concentrations of \textsuperscript{238}U, \textsuperscript{226}Ra and \textsuperscript{40}K were recorded in Lahm Mafroum commercial granites. High radioactivity of Lahm Mafroum is attributed to the presence of meta-autonite, zircon, allanite, monazite, sphene and apatite.

Absorbed Dose Rate (D), annual effective dose equivalent (AEDE), radium equivalent activity (Raeq), external (Hex) and internal (Hin) hazard index in addition to activity concentration in all the samples are given in Table (1) and Fig. (9).

Fig. 9: Contribution of \textsuperscript{238}U, \textsuperscript{232}Th and \textsuperscript{40}K radionuclides for the absorbed dose rate within the studied rock samples.
gamma index (Iγ) caused by gamma emitting natural radionuclide are determined from the obtained values of $^{226}$Ra, $^{232}$Th and $^{40}$K. Fairly, many of the studied commercial granites do not satisfy the universal standards.

REFERENCES


القياسات الإشعاعية للجريانيات التجارية المحلية والمستورد والمستخدم كصورة للزينة

عبد المعز علي صادق، عبد الهادي أحمد عباس، وأسما مالك الشرف

تم تحديد النشاط الإشعاعي الناتج من عناصر البورانيوم، الثوريوم، والروديوم في عينات جرانيتية تجارية مجمعة من السوق المحلي باستخدام نظام تحديد النشاط جاما الطيفي. وقد تراوح تركيز هذه العناصر في الجرانيت التجاري من 31 إلى 33 من 301.35 إلى 2.24، ونسبة 88.35 إلى 2.36 ونسبة 2.36 إلى 168.36. وهو يعود إلى إجراءات على مستوى المبناي واختيارات، الزركون، المونازيت، السفين والأبانيت. ودراسة العوامل المؤثرة على البيئة المحيطة وجد أن معظم هذه العوامل أعلى من المعدلات العالمية، لذا فإن ذلك يشجع على استخدام بعض هذه الأنواع من الجرانيت التجارية.