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Estimation of natural radioactivity in local and imported polished granite used as building materials in Saudi Arabia

J.H. Al-Zahrani

Physics Department, Faculty of Science, King Abdulaziz University, Jeddah, Saudi Arabia

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ABSTRACT

Measurements of natural radioactivity in local and imported samples of commercial granites used in Saudi Arabia were carried out by using gamma-ray spectroscopy with hyper-pure germanium detector. The activity concentrations measured of granite samples were determined for ^{226}Ra(from 1.53 to 77.16 Bq kg^{-1}), ^{232}Th (from 0.51 to 89.82 Bq kg^{-1}) and ^{40}K (from 19.47 to 1632.37 Bq kg^{-1}). The corresponding average activity concentrations for ^{226}Ra,^{232}Th and ^{40}K were 28.82, 34.83 and 665.08 Bq kg^{-1}, respectively. The radiological hazard parameters (radium equivalent, gamma index, external index, internal index, absorbed dose and annual effective dose) were calculated to assess the radiation hazards associated with granite samples. The obtained results are lower than the recommended limits. The results were compared with the published data of other countries. The measurements will help in the development of standards and guidelines for the use and management of these materials.

1. Introduction

Granite is a form of igneous rocks, consists mainly of quartz, mica, and feldspar, used for interior and exterior decorative as building and ornamental materials, these rocks, due to their composition contains some radionuclides. The radionuclides of ^{238}U and ^{232}Th radioactive series are the sources of both external and internal exposures in building materials. Gamma-rays cause the external exposure from radionuclides of ^{238}U and ^{232}Th radioactive series and 40 K, while alpha particles mainly cause the internal exposure due to inhalation of radon and the short-lived products of the above radioactive series, which are deposited in the respiratory tract tissues (Ahmad, Jaafar, Bakhash & Rahim, 2015; Papadopoulos et al., 2013). Thus, the study of the natural radioactivity and the radon emissions from the granite materials is an essential subject to radiological environmental protection because it facilitates the possibility to assess any related health risk. Several studies refer to natural radioactivity levels along with radiation risks of polished granite which are both used as decorative building material, as: (Llope, 2011; Mittal, Guin, Sharma, & Sengupta, 2013; Hameed, Pillai, Satheeshkumar, & Mathiyarasu, 2014; Asaduzzaman, Khandaker, Amin, & Bradley, 2016). In Saudi Arabia, local and imported Polished Granites are used as building materials, decorating and expensive materials due to its hard, tough, elegant look and different colors. Therefore, it is important to measure the concentration of radionuclides in granite used in building materials, where the inhabitants spend about 80% of their time indoors. As a result, the objectives of this study are: 1) To determine the specific radioactivity concentrations of ^{238}U, ^{232}Th, and ^{40}K in local and imported granites used as building materials in Saudi Arabia. 2) To assess the possible radiological risks to human health and compare the results with the recommended limits of UNSCEAR data.

2. Material and methods

In this study, a total of 24 polished granite samples, local (11 samples) and imported (13 samples) are the most used as a decorative inner cover on walls and floors were collected from several commercial companies, construction sites and local suppliers in Saudi Arabia. The collected samples were crushed and milled to a fine powder, homogenized, oven dried at 110° C for 6 h, and then each sample packed in a cylindrical 1000-ml plastic Marinelli beaker, sealed for one month to achieve secular equilibrium between ^{226}Ra and ^{232}Th with their decay products (Ahmad, Jaafar, &

E-mail address: Jalzhrani@kau.edu.sa.

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Alsaffar, 2015). The activity concentrations of ^{226}Ra,^{232}Th and ^{40}K for all homogenized and equilibrium samples were measured by a gamma ray spectrometry by using High purity Germanium (HPGe) detector (25% relative efficiency) with coaxial-type vertical dipstick cryostat. It is surrounded by lead and copper, which provides an efficient suppression of background gamma radiation present at the laboratory. The system has a resolution (FWHM) of (3.0–3.5 keV) for 1332.5 keV gamma-ray peak of ^{60}Co and a peak to a Compton ratio of 41:1., and it was given high voltage through preamplifier which was then connected to amplifier to computer based Multi channel analyzer through ADC (analogue to digital converter). The spectra were analyzed by commercially available software GE- NIE-2000 obtained from Canberra, USA. For gamma-ray measurements, each prepared granite sample was placed directly over the detector and counted for a 36000sec period. Additionally, under the same conditions as for the samples, the background distribution in the environment around the detector was determined. The ^{226}Ra specific activities were estimated from ^{214}Bi (609.3 keV) and ^{214}Pb (295.2 and 352.0 keV), whereas, for ^{232}Th the specific activities of ^{228}Ac (911.1 keV), ^{212}Pb (583.1 keV) and ^{208}Tl (238.6 keV) were used. The specific activity of ^{40}K was determined directly from its gamma emission at 1460.83 keV (Ahmad, Jaafar, & Alsaffar, 2015). Table 1 presents the code number, origins, colors of the local and imported granite samples used in Saudi Arabia buildings (see Table 1).

The activity concentrations of ^{238}U, ^{232}Th series, and ^{40}K were calculated using the following equation:

$$A \left(Bq\ kg^{-1}\right) = N/\varepsilon\ \beta_M \tag{1}$$

where: N is the net gamma counting rate (counts per second), ε is the detector efficiency of the specific γ−ray, β is the absolute transition probability of Gamma−decay and M is the mass of the sample (kg).

The radium equivalent activity (R_{eq}) is given as (Beretka & Mathew, 1985; UNSCEAR, 2000):

$$Req = ARa + 1.43ATh + 0.077AK \tag{2}$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bqkg^{-1}, respectively. The estimated values of 1 Bqkg^{-1} of ^{226}Ra, 0.7Bqkg^{-1} of ^{232}Th and 13Bqkg-1of ^{40}K produce the same gamma-ray dose]. For safe use, the maximum value of Ra_{eq} in building materials must be less than 370 Bqkg^{-1} to keep the external dose below 1.5 mSv.y^{-1} (UNSCEAR, 2000).

Representative level index (I_r) is used to estimate the standard of gamma radiation hazard associated with the natural radionuclides in specific building materials. It is calculated using the following formula (Asaduzzaman et al., 2016; UNSCEAR, 2000):

$$I_r = C_{Ra}/300 + C_{Th}/200 + C_K/3000 \le 1 \tag{3}$$

where C_{Ra}, *Seth* and C_K (in Bq/kg) are the concentration of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

These two hazard indices were used to measure the external and internal hazards due to the emitted gamma radiation. For safe use of material in the construction of dwellings, H_{ex} and H_{in} should be less than unity and can be defined as (Beretka & Mathew, 1985; UNSCEAR, 2000):

$$Hex = ARa/370 + ATh/259 + AK/4810 \le 1 \tag{4}$$

$$Hin = ARa/185 + ATh/259 + Ak/4810 \le 1 \tag{5}$$

The absorbed dose rate in the air is due to gamma-ray emission from the three nuclides ^{226}Ra, ^{232}Th and ^{40}K in the building materials, can be defined in units of nGyh^{-1} using the formula proposed by (Asaduzzaman et al., 2016; UNSCEAR, 2000):

$$D \left(nGy\ h^{-1}\right) = 0.0417\ Ak + 0.462\ ARa + 0.604\ ATh \tag{6}$$

where ARa, ATh, and AK are the defining activities of ^{226}Ra, ^{232}Th, and ^{40}K in Bqkg^{-1} and their corresponding dose conversion factors. By using the standard room model with dimensions of 4 m × 5 m × 2.8 m, the average world value of the absorbed dose rate is 55 nGyh^{-1} (Ahmad, Jaafar, & Alsaffar, 2015). The annual effective dose is calculated by applying the dose conversion factor of 0.7 SvGy^{-1} with 0.8 as an indoor occupancy factor (Jibiri, Isinkaye, Bello, & Olaniyi, 2016; UNSCEAR, 2000):

$$D_{eff}(mSv/y) = D\left(nGyh^{-1}\right) \times (0.7Sv/Gy \times 8760h/year \times 0.8) \times 10^{-6} \tag{7}$$

where, Deff = 0.39 mSvy^{-1} as the annual effective dose indoor for individuals, the recommended upper limit is one mSvy^{-1} (ICRP, 1990), 8766 h is the number of hours in 1 year.

3. Results and discussions

3.1. Activity concentration

Table 1 summarized the obtained activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for the measured granite samples together with their corresponding total uncertainties. ^{226} Ra range varies from 1.53 Bqkg^{-1} to 77.16 Bqkg^{-1}; ^{232}Th varies from 0.51 Bqkg^{-1} to 89.82Bqkg^{-1}. In most samples, ^{40}K represents the highest radioactivity concentration; it varies from 19.47 Bqkg^{-1} to 1632.37 Bqkg^{-1}. From all the samples measured in this study, the Iranian white sample(G1) presents the lowest activity concentrations for ^{226}Ra and ^{40}K, whereas, the Indian green sample (G11) presents the lowest activity value for ^{232}Th. The range activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were less than the typical ranges of activity concentration in building materials reported by (UNSCEAR, 2008) as 100–500 and 40–350 and 1200–1800 Bq kg$^{-1,}$ respectively. As shown in Table 1, the radioactivity in granite samples varied from one sample to another; this depends on the nature of the region from which samples were collected (Harb, El-Kamel, El-Mageed, Abbady, & Rashed, 2014). The average values for all the three nuclides ^{226}Ra, ^{232}Th, and ^{40}K were 28.82, 34.83 and 665.08Bqkg-^{1}, respectively. ^{226}Ra and ^{232}Th values are lower than the world average value 50 Bq kg^{-1} for building materials of ^{226}Ra, ^{232}Th, whereas, ^{40}K is higher than the world average value 500 Bqkg^{-1} (UNSCEAR, 2008). Fig. 1 shows the activity concentration of ^{226}Ra,^{232}Th, and ^{40}K in the present granite samples. Any granite rock could contain naturally occurring radioactive elements such as radium, thorium, and potassium. Some granite contains more of these items than others, depending on the chemical composition and the formation of the molten rock. Geologists provide an explanation of this behavior in the course of partial melting and fractional crystallization of magma, which enables U and Th to be concentrated in the liquid phase and become incorporated into the more silica-rich products. For that reason, igneous rocks of the granite composition are strongly enriched in U and Th with different concentrations (US-EPA, 1993). Table 1, showed that samples with dark colors (G: 4,6,7,14,19,24) for Ra and (G: 4,5,6,7,14,17,19,20,22,23,24) had high values of radiation. The ^{226}Ra, ^{232}Th and ^{40}K average concentrations of selected granite samples in

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