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ORIGINAL ARTICLE

Gamma radiation measurements of naturally occurring radioactive samples from commercial Egyptian granites

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Abstract Due to the widespread use of granites as building and ornamental materials, measurements of natural radioactivity for a total 27 selected samples of commercial granites used in Egypt were carried out by using a high pure germanium detector. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of commercial granites ranged from 25 to 356, 5 to 161, and 100 to 1,796 (Bq kg^{-1}), respectively. The concentrations of these radionuclides are compared with the international recommended values. To evaluate the radiological hazard of the natural radioactivity, the radium equivalent activity, the absorbed dose rate, the effective dose rate, and the hazard index have been calculated. The radium equivalent activity Ra_{eq} varied from 41 to 669 (Bq kg^{-1}) which exceeds the permitted value (370 Bq kg^{-1}) and the internal hazard index H_{in} varied from 0.2 to 2.8 which is higher than 1. The absorbed dose rate due to the natural radioactivity in the samples under investigation ranged from 19 to 310 (nGy h^{-1}). The total effective dose rates per person indoors were determined to be 0.09 to 1.5 (mSv year^{-1}).

Keywords Granites · Radium, thorium and potassium concentrations · Annual effective dose · Hazard index

Introduction

Exposure to ionizing radiation is generally undesirable at all levels by the public, although no harmful effects are presently proven for very low exposure (UNSCEAR 1993). Radiation and exposure to it, originates from two major sources namely, naturally occurring and man-made sources. The earth's crust can be divided into two distinct categories such as virgin and modified natural sources. Virgin sources of radiation are of cosmic and primordial (terrestrial) origin and have existed on the earth since primordial times. Modified natural sources are mainly from activities like mining, usage of fossil fuel, production of fertilizer or usage of natural material for building constructions. Natural radiation is the largest contributor to the collective radiation dose to the world population.

Many natural rocks contain radioactive elements such as ^{40}K, ^{232}Th, ^{238}U and ^{226}Ra. These radioactive elements contribute to the background radiation levels. Around the world though, there are some areas that have high background radiation levels due to high concentrations of radioactive minerals in soil. Building materials are possible sources of radiation exposure in particular if they contain large amounts of either naturally occurring radionuclides or man-made radionuclides (EC 1999; OECD 1979). Granites, usually suitable as building and ornamental materials for interior and exterior use, are hard natural stones, shaped and polished, compared to marble. Distinct types of commercial granites have different geological origins and mineralogical compositions and may be either magmatic or metamorphic rocks. In terms of natural radioactivity, granites exhibit an enhanced elemental concentration of uranium (U) and thorium (Th) compared to the very low abundance of these elements observed in the mantle and the crust of the Earth. 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 granitic composition are strongly enriched in U

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and Th (on an average 5 ppm of U and 15 ppm of Th), compared to the Earth's crust (average 1.8 ppm for U and 7.2 ppm for Th) (Mason and Moore 1982), the upper continental crust (average 2.7 ppm for U and 10.5 ppm for Th) (Rudnick and Gao 2003) and rocks of basaltic or ultramafic composition (0.1 ppm of U and 0.2 ppm of Th) (Mason and Moore 1982; Faure 1986; Tzortzis et al. 2004). The knowledge of the natural radioactivity of building materials is important for the determination of population exposure to radiations, as most of the residents spend about 80% of their time indoors. Therefore, it is important to measure the concentration of radionuclides in granites used in building materials collected from different places and to assess the possible radiological risks to human health. The measurements will also help in the development of standards and guidelines for the use and management of these materials.

Materials and methods

Sample collection and preparation

A total of 27 different kinds of commercial "granites" used in Egypt have been collected. The measured samples were crushed, homogenized and sieved to about 100 meshes by a crushing machine. The samples were then placed for drying at 105°C for 24 h to ensure that moisture is completely removed. The samples were stored in cylindrical plastic containers for four weeks to reach secular equilibrium between ^{232}Th, ^{226}Ra and its short lived daughter products (El-Arabi 2005; Amin et al. 2008). Spectra for different samples were measured with a high-purity germanium (HPGe) detector of high-resolution γ-ray spectrometer.

Gamma spectrometric analysis

In the present work, the measurements of natural radioactivity levels were performed by γ-ray spectrometry, using a high-purity germanium (HpGe) detector connected to a multichannel analyzer. The γ-ray activities were measured using a Canberra coaxial hyper pure germanium detector (HpGe) model GX3015. A 4096 Canberra multichannel analyzer was used to store the spectra measured from standard and investigated samples. The resolution was 1.9 keV for the 1332.5 keV γ-ray transition of ^{60}Co with photo peak relative efficiency of 37%. A nine-cylinder lead shield with a fixed bottom and movable lead cover was used around the detector to reduce the γ-ray background. The calibration and efficiency determination was obtained using 138G Marinelli beakers, source no. 566-69-7 manufactured by Isotope products laboratories, containing mixed sources of ^{109}Cd, ^{57}Co, ^{123m}Te, ^{113}Sn, ^{137}Cs, ^{88}Y, ^{60}Co, thus allowing some peaks to occur in the measurement range.

The ^{226}Ra activities (or ^{238}U activities for samples assumed to be in radioactive equilibrium) were estimated from ^{214}Pb (295.2, 351.9 keV) and ^{214}Bi (609.3, 1120.3, 1764 keV). The γ-ray energies of ^{228}Ac (338.4, 911 keV) and ^{208}Tl (583.2 keV) were used to estimate the concentration of ^{232}Th. The natural abundance of ^{235}U is only 0.72% of the total uranium content and hence was not considered in the present study. The activity concentration of ^{40}K was measured directly by its own γ rays (1460.8 keV). Errors arise due to a number of factors, like efficiency calibrations, peak area determination, and random uncertainties associated with background and sample counts. Each sample was measured during an accumulating time between 10 and 16 h. Prior to the samples measurement, the environmental gamma background at the laboratory site has been determined with an empty cylindrical beaker under identical measurement conditions. It has been later subtracted from the measured γ-ray spectra of each sample.

Results and discussion

Activity mass concentration

The activity concentrations were calculated from the intensity of each line taking into account the mass of the sample, the branching ratios of the γ-decay, the time of counting and the efficiencies of the detector. Activity concentrations, calculated from the intensity of several γ-rays emitted by a nuclide, are grouped together to produce a weighted average activity per nuclide.

The radioactivity concentration in the environmental samples was obtained as follows:

$$A = \frac{(\mathrm{cps})_{\mathrm{net}}}{m \times I \times E_{\mathrm{ff}}} \quad (1)$$

where A is the activity concentration in Bq kg^{-1}, $(\mathrm{cps})_{\mathrm{net}}$ is the net count per second and equal to $(\mathrm{cps})_{\mathrm{Sample}} - (\mathrm{cps})_{\mathrm{B.G.}}$, I is the intensity of the γ-line in a radionuclide, E_{ff} is the measured efficiency for each γ-line observed for the same number of channels either for the sample or the background, and m is the mass of the sample in kilograms. The values of the mean concentration of Ra, Th and K obtained for 27 different commercial granite types are shown in Table 1. The concentration obtained for ^{226}Ra and ^{232}Th ranged from 25 ± 4 to 356 ± 22 and from 5 ± 2 to 161 ± 13 Bq kg^{-1}, respectively, while the concentration of ^{40}K ranged from 100 ± 9 to 1796 ± 56 Bq kg^{-1}. From all the samples measured in this study, "Pearl Aswan" appears

Table 1 Activity concentration in Bq kg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K, absorbed dose rate in nGy h^{-1} and the effective dose in mSv year^{-1} of Egyptian commercial granites

Sample no.	Commercial name	Specific activity (Bq kg^{-1})			Absorbed dose rate (nGy h^{-1})	Effective dose (mSv year^{-1})
		^{226}Ra	^{232}Th	^{40}K		
2	Pink (PK)	133 ± 12	160 ± 10	1,253 ± 41	210	1.03
2	Grigio Torgoma (GT)	166 ± 10	28 ± 4	744 ± 34	125	0.61
2	Karank Gray (KG)	81 ± 8	31 ± 4	546 ± 28	79	0.39
3	Aswam (AW)	236 ± 18	150 ± 11	1,268 ± 42	252	1.24
1	Fine Gray (FG)	96 ± 9	32 ± 3	703 ± 23	93	0.45
2	Sardo Sinai (SS)	191 ± 18	141 ± 12	1,364 ± 45	231	1.13
2	Halayeb Granit (HG)	41 ± 6	8 ± 2	280 ± 19	35	0.17
1	Rosa Sinai (RS)	223 ± 17	161 ± 13	1,552 ± 51	265	1.3
1	Red Gharda (RG)	92 ± 7	91 ± 8	1,770 ± 58	164	0.8
2	Rednefertary (RF)	106 ± 9	114 ± 7	1,796 ± 56	193	0.94
2	Rosa abou simple (RAS)	51 ± 6	17 ± 3	332 ± 18	47	0.23
1	Rosa Vittoria (RV)	110 ± 11	142 ± 14	1,589 ± 52	203	0.99
2	Charme (CH)	162 ± 10	16 ± 4	1,477 ± 49	146	0.72
2	Jungle Green (JG)	25 ± 4	5 ± 2	100 ± 9	19	0.09
2	Pearl Aswan (PA)	356 ± 22	140 ± 13	1,464 ± 48	310	1.5

to present the highest concentrations of ^{226}Ra reaching levels of 356 Bq kg^{-1}, "Rosa Sinai" exhibits the highest concentration of ^{232}Th reaching 161 Bq kg^{-1} while "Rednefertary" appears to present the highest concentration of ^{40}K reaching 1796 Bq kg^{-1}. "Jungle Green" demonstrates the lowest concentrations for all the investigated samples, showing 25 and 5 Bq kg^{-1} net activity levels for ^{226}Ra and^{232}Th, respectively, and level of 100 Bq kg^{-1} for ^{40}K. The specific activities of ^{226}Ra, ^{232}Th and ^{40}K of granites of different countries are given in Table 2 for comparison. As shown in this table, the radioactivity in granite samples varied from one country to another. When the present data for Egyptian granites is compared worldwide, it is seen that the overall values of ^{226}Ra are included in the higher range and the values of ^{40}K matches with those of the other countries, while values of ^{232}Th are little bit on the higher side as compared to other countries.

Radium equivalent activity and hazard indices

The distribution of ^{226}Ra, ^{232}Th and ^{40}K in granite is not uniform. In order to compare the activity concentration and the radiological effects of granite samples, which contain ^{226}Ra, ^{232}Th, and ^{40}K, the radium equivalent activity (Ra_{eq}) as a common index has been introduced (Amin et al. 2008)

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

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. While defining Ra_{eq} activity according to Eq. (2), it has been assumed that 370 Bq kg^{-1} ^{226}Ra or 259 Bq kg^{-1} ^{232}Th or 4,810 Bq kg^{-1} ^{40}K produce the same gamma dose rate. The Ra_{eq} calculated using the activity concentrations are plotted in Fig. 1. The values ranged between 41 Bq kg^{-1} for Jungle Green and 669 Bq kg^{-1} for Pearl Aswan with an average value of

Table 2 Comparison of the activity concentration levels of granites in different countries

Country	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	References
Brazil (average)	31	73	1,648	Anjos et al. (2005)
Turkey	43–651	51–351	418–1,618	Orgun et al. (2005)
Cyprus	1–588	1–906	50–1,606	Tzortzis et al. (2003)
Greece	64	81	1,104	Pavlidou et al. (2006)
Taiwan	42	73	1,055	Chen and Lin (1996)
Nigeria	129	131	882	Joshua et al. (2009)
India	82	112	1,908	Sonkawade et al. (2008)
Italy (average)	81	129	1,065	Righi and Bruzzi (2006)
Egypt (average)	137	82	1,082	Present work

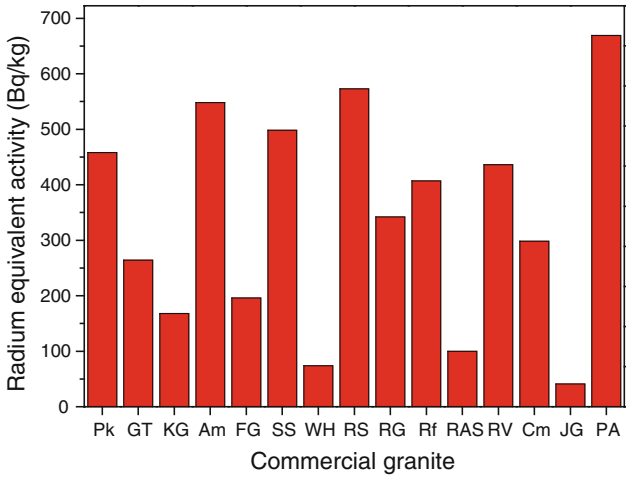

Fig. 1 Measured distribution of radium equivalent values in Egyptian commercial granites

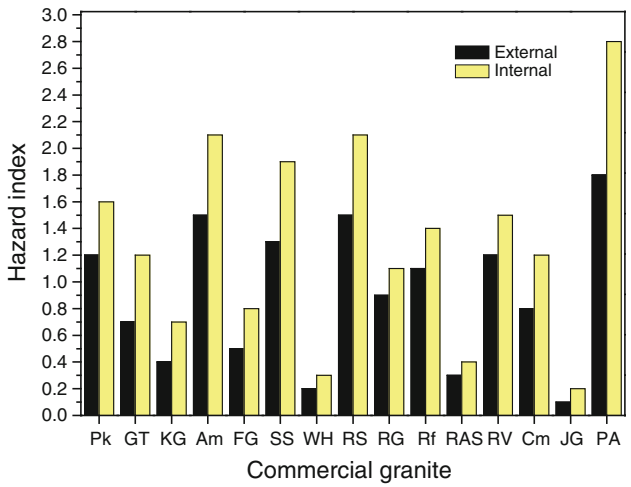

Fig. 2 Measured values of both the external and internal hazard indices for Egyptian commercial granites

338 Bq kg^{-1}. The maximum Ra_{eq} activity was found 669 Bq kg^{-1} for Pearl Aswan, 573 Bq kg^{-1} for Rosa Sinai, 548 Bq kg^{-1} for Aswam, 498 Bq kg^{-1} for Sardo Sinai, 458 Bq kg^{-1} in Pink, 436 Bq kg^{-1} for Rosa Vittoria and 407 Bq kg^{-1} for Rednefertary. These obtained values are higher than the recommended maximum value of 370 Bq kg^{-1}. This could be attributed to the high activity concentration level of ^{226}Ra in these samples.

The external hazard index (H_{ex}) is a radiation hazard index defined by Beretka and Mathew (1985). This index value must be less than unity to keep the radiation hazard insignificant, i.e., the radiation exposure due to the radioactivity from construction materials to be limited to 1.5 mSv $year^{-1}$ based on the formula (Beretka and Mathew 1985)

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

The maximum value of H_{ex} equal to unity corresponds to the upper limit of Ra_{eq} (370 Bq kg^{-1}). The values of H_{ex} for all samples ranged from 0.1 to 1.8 as shown in Fig. 2. The values of H_{ex} of Egyptian commercial granite studied in this work are less than unity except for Pink, Aswam, Sardo Sinai, Rosa Sinai, Rednefertary, Rosa Vittoria and Pearl Aswan samples.

The presence of ^{226}Ra and ^{232}Th in building materials are sources of internal exposure to ^{222}Rn and ^{220}Rn and their progenies, respectively. The contribution is of ^{220}Rn is negligible due its shorter half life than that of ^{222}Rn and the main contribution is of ^{222}Rn. In order to address the radiation hazard to respiratory organs due to ^{222}Rn and its progeny, the internal hazard index H_{in} which is given by (Beretka and Mathew 1985)

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

Also, in view of the fact that normally external hazard index calculations are based on 2π geometry whereas for internal hazard index full 4π geometry needs to be considered, which makes it necessary to reduce concentration of ^{226}Ra to half of the value used in external hazard index term. The maximum value of H_{in} must be less than one (Krieger 1981). The calculated values of H_{in} range from 0.2 to 2.8 as shown in Fig. 2. The values of H_{in} for all samples are higher than unity except for Karank Gray, Fine Gray, Halayeb Granite, Rosa abou Simple and Jungle Green samples.

Dose Assessment

The total air absorbed dose rate (nGy h^{-1}) 1 m above the ground due to the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}) was calculated Monte Carlo method using the formula (UNSCEAR 2000)

$$\text{Absorbed Dose Rate } D \text{ (nGy h}^{-1}\text{)} = 0.0417A_K + 0.462A_{Ra} + 0.604A_{Th} \quad (5)$$

Table 1 gives the estimated external gamma dose rate due to natural γ-emitters as measured in the granite. The results show that the absorbed dose rates range from 19 to 310 nGy h^{-1} with mean value of 158 nGy h^{-1}. According to the recent UNSCEAR (1993, 2000) reports, the corresponding worldwide average values range from 18 to 93 nGy h^{-1} and a typical range variability for measured absorbed dose rates in air outdoors is from 10 to 200 nGy h^{-1}. The population weighted values give an average absorbed dose rate in air outdoors from terrestrial gamma radiation of 55 nGy h^{-1}. This reveals that the mean absorbed dose rate in air outdoors from granite samples is almost two times higher than that of the worldwide average value.

Finally, to estimate the annual effective doses, account must be taken of the conversion coefficient from absorbed

dose in air to effective dose and the indoor occupancy factor. A value of 0.7 $SvGy^{-1}$ was used for the conversion coefficient from absorbed dose in air to effective dose received by adults, and 0.8 for the indoor occupancy factor, implying that 20% of time is spent outdoors, on average, around the world UNSCEAR (1993, 2000).

The effective dose rate indoors, in units of mSv $year^{-1}$, is calculated by the following formula:

$$\begin{aligned}&\text{Effective dose rate } H_E(\text{mSv/year})\\&= \text{dose rate } (\text{nGy h}^{-1}) \times 24\,\text{h} \times 365\,\text{days}\\&\quad \times 0.8(\text{occupancy factor})\\&\quad \times 0.7\,\text{Sv Gy}^{-1}(\text{conversion coefficient}) \times 10^{-6} \qquad (6)\end{aligned}$$

According to more recent regulation and especially the recommendation No. 112 issued by European Union in 1999 (EC 1999), building materials should be exempted from all restrictions concerning their radioactivity provided that the excessive gamma radiation causes the increase of the annual effective dose received by an individual by a maximum value of 0.3 mSv (EC 1999). However, in few cases the excessive annual effective dose exceeds 1 mSv $year^{-1}$, which corresponds to the dose criterion that is taken into account for radiation protection. It is therefore recommended that controls should be based on a dose range of 0.3–1 mSv $year^{-1}$, which is the building material gamma dose contribution to the dose received outdoors (Tzortzis et al. 2003).

The results obtained for annual effective doses ranged from 0.09 mSv $year^{-1}$ for "Jungle Green" to 1.5 mSv $year^{-1}$ for "Pearl Aswan" samples. The annual effective dose H_E exceeds the limit of 1 mSv for five samples as follows: Pink (1.03 mSv), Aswam (1.24 mSv), Sardo Sinai (1.13 mSv), Rosa Sinai (1.3 mSv) and Pearl Aswan (1.5 mSv).

Conclusions

Seventy-six kinds of granites used in Egypt, considered as the most popular ones, were measured for their natural radioactivity in order to assess the radiological impact when they are used as building materials. The activity concentration of ^{40}K was highest in all the samples. The average value of ^{226}Ra concentration is between 25 and 356 Bq kg^{-1}. The highest concentration of ^{226}Ra, ^{232}Th and ^{40}K were observed in Pearl Aswan, Rosa Sinai and Rednefertary samples, respectively.

The highest values of Ra_{eq} (669 Bq kg^{-1}), total absorbed dose rate (310 nGy h^{-1}) and annual effective dose (1.5 mSv $year^{-1}$) were also observed in Pearl Aswan sample.

We may therefore conclude that the all samples under investigation are within the recommended safety limit when used as building construction except some samples could be significant as a source of radiation exposure.

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