Levels of natural radioactive nuclides in some building construction materials

Shadiah S. Baz and A. Abbady

Physics Department, Girls Faculty of Science, Jeddah, King Abdulaziz University, Saudi Arabia
ssb_998@hotmail.com

Abstract: The radiological hazards associated with naturally occurring radioactive nuclides in sandstone, granite and shale samples from the south of the eastern desert in Egypt had been studied using gamma spectroscopy. The average activity concentration of ^{226}Ra, ^{232Th} and ^{40}K were (7.1±2.8, 27.22±3.6 and 495.4± 18.8 Bq/kg) in sandstone. For granite samples, the average concentrations were (26.2±3.7, 42.2±6.1 and 812.4 ±26.9 Bq/kg) respectively. The corresponding average values in shale were (22.1±4.3, 38.3± 5.3 and 4357 ±19.9 Bq/kg) for ^{226}Ra, ^{232}Th and ^{40}K. Granite samples exhibit an enhanced activity concentrations compared to other materials, while sandstone samples had the smallest value. The ^{40}K recorded the highest activity concentration in all measured samples. The average radium equivalent activity (Ra_{eq}: 84.1±10.3, 149.1±15.6 and 110.4±11.2 Bq/kg) for sandstone, granite and shale were found to be less than the acceptable limits for public exposure control recommended by the European Commission. The calculated average dose rates for the three types were: 42.4±3.9, 74.2±6.3, 53.6±5.1 respectively. These values are lower than the estimated average global terrestrial radiation of 55 nGyh-1 except granite samples where the obtained value was 74.2±6.3 nGyh-1.
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Key words: Natural radioactivity concentration, construction, Granite, Sandstone, Shale, Dose rate.

1. Introduction

Studies and surveys of low level radiation exposure arising from naturally occurring radio nuclides are of great importance. The interest of these studies is due to that the exposure from these nuclides contributes about 10% of the average annual dose to human body from all sources. The activity concentrations of natural radionuclide's in various environmental samples had been extensively studied by many explorers [Francis Otoo *et al.,* 2012, Harb *et al.,* 2014, El-Bahi,2004] In controlling the natural radiation exposure for the residents of dwellings, it is necessary to determine the levels of natural radioactivity from building construction materials since they contain various amounts of natural radioactive nuclides and cause direct radiation exposure. Some types of these materials such as; Brick, Cement, Granite stones and Gypsum may produce significant external dose rates in the range of $nGyh^{-1}$ (Louizi and Proukakis 1994).

Methods of measuring concentrations of radioactive nuclides in the environmental media can be applied using different detectors. Gamma ray spectrometers are the most convenient one in many larger analytical laboratories. NaI(Tl) detector have become a powerful tool for radiations detection and energy measurements since they give good efficiency for gamma-rays detection. In order to achieve the best possible gamma-energy resolution with a NaI scintillation spectrometer, a selected suitable electronics must be chosen.

The aim of this article is to measure natural radioactive nuclides in some construction building materials such as: sandstone, granite and shale collected from the eastern desert in Egypt. The analysis had been carried out by gamma-rays spectrometer using NaI(Tl) crystal as a detector.

Samples preparation:

The selected samples had been collected from the eastern desert south Egypt. The sampling site has rocky nature consisting mainly of igneous, metamorphic and sedimentary rocks. For detection procedure, the collected materials should be prepared and shaped in the same geometry of the calibration sample. The preparations start by crushing large grain samples to small pieces. Then they were dried at 105°c to eliminate any traces of water. Afterwards, the samples were ground to a fine grain size powder. Using an electric shaker, the powdered samples were mixed very well to obtain homogenous samples. The cone shaped procedure was done to obtain good representative samples with the required weight. The prepared powder samples were sealed tightly in cylindrical containers and stored for a period of four weeks to allow ^{226}Ra and ^{232}Th decay series to reach radioactive equilibrium with the short-lived progenies. (Adukpook *et al.,* 2011).

Method and experimental procedures:

Samples under investigation had been analyzed in the laboratory of gamma-ray spectrometry built in the faculty of science, Qena, Upper Egypt. The

applied spectrometer consist basically of NaI(Tl) crystal 3x3 inch, Ortec connected with a micro-computer multichannel analyzer. The system automatically computes and display the total counts, background counts net total counts and the counting rate in each peak. Details and characteristics of the applied spectrometer were mentioned in previous articles (Ahmed N.K 2005, Abbady *et al.,* 2006). The applied spectrometer was calibrated using standard sources with well specific energies such as ^{60}Co, ^{137}Cs and ^{133}Ba. The absolute efficiency was determined using multinuclides standard source QCY48 (Harb *et al.,* 2008). To reduce the background counting, the sample and the detector were housed in a lead chamber and another lead cylinder around the crystal. The real measuring time was fifteen hours for each sample to obtain clear and well shaped gamma spectra suitable for analysis. The spectra were analyzed either with the computer program Maestro 2 (EG & ORTEC) or manually using a spreadsheet to calculate the net area under each peak. Background measurements were also made for the same period and subtracted from the sample spectra. The activity concentration A_{Ei} (Bq/Kg) of each radionuclide in any given sample was calculated from the measured spectrum. Where the parent nuclides were in secular equilibrium with their daughter nuclides, the activity concentrations of the parent nuclides were estimated from their respective daughter concentrations. The transition gamma lines (609.3 1764.6 KeV) of ^{214}Bi and (583.3, 2614.5 KeV) of ^{208}Tl were used to determine the activity concentration of ^{226}Ra and ^{232}Th respectively. ^{40}K was determined directly with its only 1460.7 KeV peak transition line.

The specific activity A_{Ei} (Bq/Kg) of a nuclide i and for a peak at energy E is given by:

$$A_{Ei} = \frac{N}{t_c I_\gamma(E_\gamma) \in (E_\gamma) M} \quad (1)$$

Where N is the number of counts in a given peak energy E, t_c is the real counting time in seconds, $I_\gamma(E_\gamma)$ is the number of gamma photons per disintegration 0f the nuclide i for a transition at energy E and M is the mass of the sample in kilogram.

Estimation of radiation hazards:

Radium equivalent activity Raeq

The main objective of calculating Ra_{eq} is to make an estimate of radiation dose likely to be externally due to gamma radiation. This Ra_{eq} provides useful guidelines in regulating the safety standards on radiation protection for the general public. The Ra_{eq} was calculated using the following equation:

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

Where A_{Th}, A_{Ra}, A_K are the mean activities of ^{232}Th, ^{226}Ra and ^{40}K (Bq/Kg) in the sample respectively. The Ra_{eq} is the summation of above mentioned radio nuclides and based on the assumption that, 1 Bq/Kg of ^{226}Ra, 0.7 Bq/Kg of ^{232}Th and 14 Bq/Kg of ^{40}K produce the same gamma radiation dose rate (Molanca *et al.,* 1993). The maximum value of Raeq must be 370 Bq/Kg to keep the external dose to 1.5 mSv/y. The construction building materials whose Ra_{eq} exceeds 370 Bq/Kg is discarded to reduce radiation hazards associated with these materials.

Several models have been developed according to values of activity concentrations of individual radionuclides to limit radiation dose from buildings as low as possible. The absorbed dose rates (DR) due to gamma radiation in air at 1m above the ground surface, assuming uniform distribution of the naturally occurring radionuclides have been calculated according to UNSCEAR 2000 as follows:

$$DR(nGh^{-1}) = 0.427A_{Ra} + 0.662A_{Th} + 0.0432A_K \quad (3)$$

Where A_{Ra}, A_{Th}, and A_K are the mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively in Bq/Kg.

3. Results and Discussion:

The activity concentrations in (Bq/Kg) for ten sandstone, five granite and six shale samples calculated for each of ^{226}Ra, ^{232}Th and ^{40}K respectively, are listed in table (1) and shown in Fig(1).

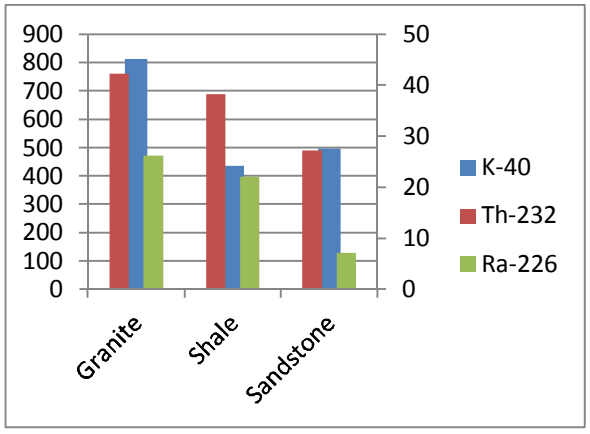

Fig (1): The calculated mean activity concentrations of K^{40}, Th^{232} and Ra^{226} nuclides in granite, shale and sandstone samples.

The obtained results illustrates that granite samples have always the highest concentration of natural radioactive nuclides. The mean specific activity values of the same nuclides in shale samples were smaller than those of granite and higher than the values in sandstone except the value of 40K. Granite samples are strongly enriched in ^{226}Ra, 232Th and ^{40}K because partial melting and fractional crystallization of magma enables U and Th to be concentrated in the liquid phase and become incorporated into the more silica-rich products (Tzortzis *et al.,* 2003).

Radiation Hazard Indices:

The average dose rate (nGh^{-1}) and the average radium equivalent activity (Bq/Kg) of samples under investigation were calculated and listed in table (2). The average values of Ra_{eq} are below the internationally accepted value 370 Bq/Kg in all samples (UNSCEAR 2000) while the average dose rate is higher than the average global dose rate 58 nGh^{-1} for granite samples.

Table (1): Activity concentrations (Bq/Kg) in samples under investigation

Sample Type	Sample Number	Activity concentrations Bq/Kg		
		^{226}Ra	^{232}Th	^{40}K
Sandstone	S_1	9.4±2.9	31.3±4.6	540.2±22.3
	S_2	6.2±1.8	18.6±3.8	444.5±19.4
	S_3	7.1±2.6	24.1±3.5	439.1±21.4
	S_4	6.3±2.2	23.1±4.2	452.3±24.3
	S_5	7.5±3.1	25.4±4.4	466.4±26.1
	S_6	6.8±2.4	29.6±4.8	531.2±25.6
	S_7	7.2±3.2	28.3±3.9	518.7±26.8
	S_8	9.1±3.6	20.2±5.1	489.2±23.3
	S_9	6.4±2.5	23.9±4.6	495.9±20.8
	S_{10}	5.9±2.0	18.1±3.9	574.1±26.6
Mean		7.1±2.8	27.2±3.6	495.4±18.8
Granite	G_1	23.1±5.2	41.6±5.9	890.3±27.2
	G_2	29.8±5.5	42.9±6.1	955.6±31.1
	G_3	25.3±4.3	40.2±6.5	821.0±29.4
	G_4	23.7±4.8	48.8±6.2	869.3±28.3
	G_5	29.2±6.1	37.4±4.5	526.7±42.8
Mean		26.2±5.7	42.2±6.1	812.4±26.3
Shale	Sh_1	25.3±2.9	36.2±5.1	456.5±19.7
	Sh_2	21.8±3.1	33.6±4.9	443.2±18.9
	Sh_3	19.2±2.2	40.7±6.3	458.1±17.8
	Sh_4	23.6±2.8	39.1±5.9	429.4±18.2
	Sh_5	20.4±2.1	42.2±6.3	437.3±19.3
	Sh_6	22.5±3.2	38.4±5.6	387.7±16.5
Mean		22.1±4.3	38.3±5.2	435.7±19.9

Table (2): The average radium equivalent activity Ra_{eq} (Bq/Kg) and the average dose rate DR (nGy/h)

Sample Type	Average Ra_{eq}	Average DR(nGy/h)
Sandstone	84.1±10.3	42.4±3.9
Granite	149.1±15.6	74.2±6.3
Shale	110.4±11.2	53.6±5.1

The average dose rate for sandstone and shale samples are less than the average global one.

In comparing the results of this study with some published values for granite rocks, it was noted that the present results are much higher than the values reported by (Arafe 2004, Anjos *et al.,* 2005 and Xinwei *et al.,* 2006).

Conclusion:

The measured radio activities of Ra 226, Th232 and K40 in sandstone, granite and shale samples collected from the eastern desert, south Egypt were evaluated by gamma ray spectrometer. The data illustrated that the activity concentration in granite samples increases the values determined in some countries. Therefore the average dose rate due to granite samples is higher than global average dose rate limit. It should be considered that sandstone and shale rocks are more convenient as construction building materials without providing excessive exposure for inhabitants.

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