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The Evaluation of Radioactivity Levels and Radiological Hazards from Granite used for Construction Materials

M. Almeshari[*]

Department of Diagnostic Radiology, College of Applied Medical Sciences, University of Ha'il, Ha'il, Saudi Arabia.

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Abstract: The activities associated with ^{226}Ra, ^{232}Th, and ^{40}K were quantified on granite rock samples obtained from the protected region southeast of Aswan City. Utilizing γ-ray spectroscopy, the amount of gamma radiation in these samples was measured. with NaI(Tl) detectors. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were determined based on the measured γ-ray spectra. For ^{226}Ra, the activity concentrations ranged from 10±2 to 90±11 Bq kg-1, with an average value of 26 ± 4.8 Bq kg-1. Similarly, for ^{232}Th, the activity concentrations ranged from 98±6 to 160 ±30 Bq kg-1, with an average value of 122 ± 11 Bq kg-1. Lastly, for ^{40}K, the activity concentrations ranged from 72 ± 3 to 102 ± 9.4 Bq kg-1, with an average of 89 ± Bq kg-1. Several parameters were established to assess the potential radiological risk associated with the radioactivity present in the samples, including the radium equivalent activity, absorbed dose rate, and external hazard index. The radium-equivalent activities (Ra_{eq}) observed in all granite samples were below the established threshold of 370 Bq kg-1. The airborne absorbed dose rates, determined by evaluating the three radionuclide concentrations, exhibited a range of 73.6 to 117.7 nanograys per hour. The researchers calculated the yearly effective dose rates, both external and internal. The results showed that the range for external dose rates was between 0.09 and 0.145 mSv.y-1, while the range for internal dose rates was between 0.361 and 0.578 mSv.y-1. The average values for external and internal dose rates were determined to be 0.11 and 0.44 mSv.y-1, respectively. The significance of this topic lies in its importance to the environment, as granites are extensively used as construction and decorative materials. The findings are deliberated upon and juxtaposed with the relevant data published in existing literature.

Keywords: Natural radioactivity, Granite, Radioactivity, Building materials, Absorbed dose.

1 Introduction

Several global surveys were done to quantify the terrestrial gamma-ray exposure rates at locations above the Earth's surface. To attain an accurate interpretation of the survey results, it is essential to understand the levels of radioactivity in the underlying rock, as the soil originates from it. The main source of outdoor gamma radiation exposure is the presence of trace amounts of terrestrial radionuclides found in various types of geological formations. The primary origins of extrinsic gamma radiation encompass ^{238}U, ^{232}Th, their respective decay byproducts, and ^{40}K. Consequently, the levels of natural environmental radiation are predominantly influenced by geological and geographical variables [1]. Elevated radiation levels commonly characterize igneous rocks, for example, granite, whereas sedimentary rocks generally exhibit lower radiation levels. Nevertheless, it is important to note that specific instances exist where shale and phosphate rocks exhibit elevated levels of radionuclides [2]. The variability of absorbed dose in the air due to external gamma radiation emitted by radionuclides is influenced by local geological and geographical factors.

The evaluation of dose rates in air associated with the use of stones as tiling materials can be achieved by assessing the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in each sample. This approach was documented in studies such as [2] [3] [4] [5]. Numerous surveys were conducted globally to measure terrestrial gamma-ray dose rates above ground in different geographical areas. However, to accurately interpret the findings of these surveys, it is critical to understand the rocks' levels of radioactivity, as the soil originates from them. The main source of outdoor gamma radiation exposure is the presence of trace amounts of terrestrial radionuclides inside various types of geological formations. The primary origins of extrinsic gamma radiation encompass ^{238}U, ^{232}Th, and their subsequent decay byproducts, in addition to ^{40}K. Hence, the levels of natural environmental radiation are predominantly influenced by geological and geographical factors [1]. Igneous rocks, such as granite, are commonly correlated with elevated

[*]Corresponding author e-mail: m.almeshari@uoh.edu.sa

radiation levels, whereas sedimentary rocks tend to exhibit comparatively lower amounts of radiation. Nevertheless, it is worth noting that there exist specific instances where shale and phosphate rocks exhibit elevated levels of radionuclides [2] [6] [7]. The extent to which radionuclide-emitting external gamma radiation contributes to the absorbed dosage in the air is subject to variation based on local geological and geographical factors.

The current investigation employed gamma spectrometric techniques to assess radioactivity levels in granite samples collected from the protection region located southeast of Aswan City. This study's goal was to investigate the distribution patterns and assess the radiation danger indices associated with the subject of investigation. The findings of this investigation were compared with granite samples obtained from different geographical regions.

2 Experimental procedure and methods

2.1 Sampling preparation

A total of twenty granite samples were obtained from the protective region located in the southeastern vicinity of Aswan City. These samples were subjected to measurements to ascertain their levels of natural radioactivity. Before the collecting process, a substantial rock was subjected to fragmentation by the utilization of a hammer, subsequently resulting in the formation of smaller rock fragments. These fragments were subsequently placed in black plastic bags for storage and transportation purposes. The sample sizes varied between approximately 0.5 and 1 kilograms, with all samples being granite rocks. Subsequently, the rock samples were carefully conveyed from the designated collection sites to the laboratory, where they were meticulously encased in black, thin-walled plastic bags to ensure their secure transportation. The air-drying process was applied to every sample, subsequently followed by the procedures of crushing, grinding, and sieving using a mesh with a size of 1.0 mm. The objective of this procedure was to achieve the segregation of the larger particles from the intended finely textured powder. The larger grains were discarded, while the finely ground rock was held in cylindrical polyethylene beakers with a volume of 350 cm^3 apiece. The duration of the storage period spanned four weeks, providing an ample timeframe for the achieving of secular equilibrium between radon and thoron and their corresponding decay products. This temporal allocation aligns with the guidelines set forth by the International Atomic Energy Agency (IAEA) and ensures the suitability of the samples for further examination [8].

2.2 Sample counting

The technique of gamma-ray spectrometry was employed to conduct measurements of radioactivity. The spectrometer had a scintillation detector measuring 3 x 3 in size. The detector system was well sealed to prevent any external interference and consisted of several components, including a high-resolution NaI(Tl) crystal, a photomultiplier tube, an interior shield made of either magnetic or light-blocking material, an aluminum enclosure, and a 14-pin connector that was connected to a PC-MCA device known as the Canberra Accuspes. The detector possesses the following specifications: (1) It exhibits a resolution of 7.5% at the 662 keV peak of ^{137}Cs, (2) It incorporates a 0.5 mm thick aluminum window with a density of 147 mg/cm2, (3) It includes a 1.6 mm thick oxide reflector with a density of 88 mg/cm2, (4) It is equipped with a magnetic or light shield composed of lined steel, and (5) It operates at a positive voltage of 902V DC.

In order to mitigate the interference resulting from gamma rays, a cylindrical lead shield with a stationary base and an adjustable lid was used to safeguard the detector. The lead shield was composed of an inside cylindrical structure constructed from copper, with a thickness of 0.3 mm. The inclusion of a copper layer effectively mitigated the X-ray emission at 73.9 keV originating from the lead material upon exposure to external radiation. Additionally, the lead shield demonstrated a notable reduction in the intensity of the soft component of cosmic rays, effectively lowering it to a minimal level. A 100 mm thickness of lead shielding was used [9].

The energy calibration of the detecting system was conducted by utilizing the kilo electron volt (keV) values associated with the radioactive isotopes ^{60}Co (1173.2 and 1332.5 keV), ^{133}Ba (356.1 keV), ^{137}Cs (661.9 keV), and ^{226}Ra (1764.49 keV). Additionally, a calibration curve for efficiency was generated, encompassing energy peaks reaching a maximum of roughly 2000 keV. To ensure measurement quality, daily calibrations were executed for both efficiency and energy for each sample. The period of measurement for activity or background was 43,200 seconds. Every gamma-ray spectra examined obtained offline was conducted using a specialized software application known as Genie 2000.

To accomplish the dispersion of background radiation originating from naturally existing radioactive elements in the area surrounding the detector, an unoccupied polystyrene container was subjected to the same counting procedure. Measuring and subsequently removing the background determined the activity concentration.

2.3 Radiation hazard indices for building materials

When making decisions on construction materials, it is advisable to select those with minimal levels of particular radioactivity. While the effective dose equivalent caused by radioactivity present in construction materials, measuring at 0.41 mSv, may not be as substantial in comparison to other sources, such as two mSv, it remains crucial to conduct comparisons among various materials to mitigate avoidable exposure. To analyze the radiological impacts of construction materials that contain radium (Ra), thorium

(Th), and potassium (K), the potential radiation risks associated with these particular radionuclides were assessed using two distinct indices. The predominant index employed is the radium equivalent activity (Ra_{eq}), which serves as a comprehensive indicator of the activities associated with radium (Ra), thorium (Th), and potassium (K). The values of 370 Bq/kg for radium (Ra), 259 Bq/kg for thorium (Th), and 4810 Bq/kg for potassium (K) are employed to make estimations regarding the corresponding gamma-ray dosage rates. The calculation of the Ra_{eq} is determined by the following formula:

$$Ra_{eq} = A_{Ra} + (A_{Th} \times 1.43) + (A_K \times 0.077) \quad (1)$$

The calculation of the absorbed dose rate in the air (measured in nomograms per hour) caused by the average specific activity concentrations of uranium-238, thorium-232, and potassium-40 (measured in Becquerel per kilogram) is performed utilizing the formula provided by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [10].

$$D = 0.427\ S_U + 0.662\ S_{Th} + 0.0432\ S_K \quad (2)$$

In this context, SU denotes the average activity concentration of 238U, STh denotes the average activity concentration of ^{232}Th, and SK denotes the average activity concentration of 40K in rock samples. Equation (2) is employed to compute the absorbed dose rate in the atmosphere, specifically at an elevation of 1.0 m above the Earth's surface, by utilizing the measured amounts of radionuclides found in environmental substances. The representative level index (Iγr), an additional radiation hazards indicator, is determined by adding the activity concentrations of radium (ARa) divided by 150, thorium (ATh) divided by 100, and potassium (AK) divided by 1500 [11]:

$$I\gamma = A_{Ra}/150 + A_{Th}/100 + A_K/1500 \quad (3)$$

Where the activity concentrations in Bq/kg for ^{226}Ra, ^{232}Th, and ^{40}K, are represented by A_{Ra}, A_{Th}, and A_K

3 Results and Discussion

3.1 ^{238}U, ^{232}Th, and ^{40}K activity concentrations

The calculation of radioactive concentrations and the count rates for each detected photopeak, which pertain to the specific activity or activity per unit mass of radionuclides found, are dependent on the achievement of secular equilibrium within the samples. The attainment of secular equilibrium was observed in the relationship between ^{232}Th and ^{238}U, as well as their respective decay products. The authors of the study employed the average concentrations of decay products ^{214}Pb and ^{214}Bi to determine the concentration of ^{238}U [12]. Similarly, the content of ^{232}Th was measured using the average concentrations of ^{212}Pb and ^{228}Ac in the samples. As a result, precise measurements of radionuclide concentrations for 238U and 232Th were achieved, whereas the measurement of ^{40}K concentration was deemed dependable. The findings of this study reveal that the range of specific gamma-ray activity associated with ^{226}Ra in granite spans from 10 to 90 Bq.kg-1. The lowest and highest activity for the isotopes ^{232}Th and ^{40}K are measured to be 98 Bq.kg-1 and 160 Bq.kg-1 and 72 Bq.kg-1 and 102 Bq.kg-1, respectively. Table 1 displays the activities of the ^{226}Ra (^{238}U) series, ^{232}Th series, and ^{40}K, which are measured in units of Bq.kg-1. It is observed that in granitoid rocks, uranium (U) and thorium (Th) tend to collect in accessory minerals such as zircon, monazite, apatite, sphene, and allanite [13]. Additionally, these elements may be present as impurities inside minerals such as fluorite, apatite, hematite, and mica. Thorium can undergo adsorption onto several substrates, including clays, iron oxides, and hydroxides. Uranium can undergo direct crystallization from silicate magmas, resulting in the formation of uraninite, particularly in environments characterized by high temperatures. Other auxiliary minerals, including pyrochlore, brannerite, euxenite, thorite, thorianite, apatite, sphene, zircon, xenotime, and monazite, are frequently observed in conjunction with biotite and have the potential to contain uranium.

Felsic rocks commonly exhibit elevated concentrations of thorium and uranium, with their amounts being positively correlated to the alkalinity or acidity of the rocks. Pegmatites are known to exhibit the most elevated quantities of these elements. Moreover, there exists a correlation between the acidity of the rocks and an elevation in the concentration of potassium. Potassium is frequently found in potash feldspars, including microcline and orthoclase, as well as in micas, such as muscovite and biotite. According to previous study, rocks that do not contain these minerals typically have a significantly low level of potassium activity [4].

The conventional representation of uranium-238 (^{238}U), thorium-232 (^{232}Th), and potassium-40 (^{40}K) concentrations is in the form of equivalent ground concentrations, denoted as K (%), U (ppm), and Th (ppm), respectively. The measured radioactivity of a sample containing 1 part per million (ppm) of ^{232}Th and one ppm of natural U is found to be 4.08 and 13.0 Becquerel per kilogram (Bq.kg-1), respectively. The specific activity of ^{40}K in natural potassium is 317 Bq.kg-1 when the quantity of potassium in the sample is 1% by weight.

Upon comparing the aforementioned data with the reported ranges and averages provided by Turekian and Wedepohl (1961) and Rogers and Adams (1969) [13] [14], it becomes apparent that biotite and pegmatite muscovite granite exhibit U concentrations that are representative of the norm. Conversely, other granite plutons investigated display various levels of U enrichment. During the final stages of magmatic activity, it is possible for hydrothermal solutions to retain residual elements, leading to the formation of veins that are enriched in uranium.

Table 1: Activity concentration of 226Ra, 232Th, and 40K (Bq.kg-1) for all samples under study.

Sample	^{226}Ra (Bq.kg^{-1})	^{232}Th (Bq.kg^{-1})	^{40}K (Bq.kg^{-1})	U ppm	Th ppm	K %
1	80	120	96	6.5	29.5	0.32
2	70	110	101	5.7	27.1	0.33
3	90	98	91	7.3	24.1	0.30
4	10	160	81	0.8	39.4	0.27
5	12	107	89	1.0	26.3	0.29
6	13	140	86	1.1	34.4	0.28
7	17	120	96	1.4	29.5	0.32
8	16	130	86	1.3	32.0	0.28
9	15	160	72	1.2	39.4	0.24
10	12	120	90	1.0	29.5	0.30
11	15	109	102	1.2	26.8	0.34
12	16	150	83	1.3	36.9	0.27
13	19	107	99	1.5	26.3	0.33
14	18	108	92	1.5	26.6	0.30
15	17	140	89	1.4	34.4	0.29
16	17	150	81	1.4	36.9	0.27
17	19	105	95	1.5	25.8	0.31
18	21	109	88	1.7	26.8	0.29
19	23	103	73	1.9	25.3	0.24
20	21	105	98	1.7	25.8	0.32

Furthermore, substantial quantities of uranium (U) might be deposited in the intergranular voids or in minerals formed during the latter stages of alteration, which are linked to the processes of silicification and hematitization in the surrounding rocks [15].

Upon comparing the findings of the present study with prior research conducted by El-Shershaby (2002), Arafa (2004), and Orgun et al. (2005) [16] [17] [18], it becomes apparent that the results obtained in this study exhibit lower values in comparison to those reported in other sources, specifically in various regions of Egypt plutons that are recognized for their elevated levels of radionuclide concentrations. The average concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the granitic rock samples are being compared to the findings of earlier studies, as shown in Table 2. This comparative analysis illustrates that the geological attributes of the examined locations have an impact on the levels of ^{226}Ra, ^{232}Th, and ^{40}K.

Table 2: Comparison of the average concentrations of uranium (U), thorium (Th), and potassium (K) in granite rocks with previously published studies.

Type	**Mean concentration**		
	U Bq/kg	Th Bq/kg	K %
Granite Wadi Allaqi, Aswan, Egypt	25.93	121.8	90.77
Saudi Arabia [19]	28.82	34.83	665.08
Palestine [20]	71	82	780
Jordan [21]	41.52	58.42	897
India [22]	25.88	42.82	560.6
Iran [23]	77.4	44.5	1017.2
Spain [24]	84	42	1138
Greek [25]	74	85	881
Nigeria [26]	63.29	226.67	832.59

The radium equivalent activity (Bq.kg-1) results, representative level index, total absorbed dose rate of gamma radiation in the air, and Table 3 presents the outdoor effective dose assessment for the materials that were examined. The average values of (Ra_{eq}) and representative level index (Iγr) in granite were found to be 208 ± 14.4 and 1.5 ± 0.8 Bq.kg-1, respectively. The determined Ra-equivalent activities for the examined samples are found to be lower than the suggested maximum threshold of 370 Bq.kg-1 for construction materials, as presented in Table 3. The absorbed dose rates resulting from the presence of ^{226}Ra, ^{232}Th, and ^{40}K in granite exhibit a range of 73.6 to 117.7 nGy.h-1, with an average value of 89.6 ± 9.4 nGy.h-1.

3.2 The annual effective dose is equivalent to the measured terrestrial radiation.

To determine the annual effective doses, two key factors: the conversion coefficient that relates absorbed dosage in air to effective dose and the indoor occupancy factor. The values of these factors exhibit variability contingent upon the age composition of the population and the climatic conditions of the examined geographical area. The UNSCEAR [2] Report employed a conversion coefficient of 0.7 Sv Gy-1 for adults, along with an indoor occupancy factor of 0.8, indicating that 80% of the time is allocated to inside activities and the remaining 20% is spent outside. The parameters mentioned earlier are maintained in the current study.

The annual effective dose equivalent of gamma radiation from terrestrial, both indoors and outdoors, is calculated:

84 (nGy h^{-1}) × 8.760 (h) × 0.8 × 0.7 (Sv Gy-1) = 0.41 mSv (Indoor) (4)

59 (nGy h^{-1}) × 8.760 (h) × 0.2 × 0.7 (Sv Gy-1) = 0.07 mSv (Outdoor) (5)

The findings suggest that the annual average doses resulting from natural radioactivity in granite are higher in comparison to the global average. More precisely, the mean yearly dosages are reported as 110 ± 9.4 µSv y-1 and 441 ± 21 µSv y-1, respectively, while the global average stands at 70 µSv y-1 and 410 µSv y-1. The study's findings suggest that the amounts of radiation originating from natural sources do not pose a contamination or environmental hazard. The global mean yearly effective dose is determined to be 0.48 mSv, with the majority of individual nations falling within the range of 0.3-0.6 mSv.

3.3 External and internal hazard index.

A conservative model was put forth in Germany to establish a maximum radiation dose of 1.5 mGy y-1 from building materials [27]. The aforementioned model, as documented by (Krisuk et al., 1971), and ^{40}K activity concentrations found in various construction materials [28]. The model postulates the presence of walls that possess infinite thickness, devoid of any windows or doors. It proceeds to compute the external hazard index, denoted as Hex, by employing the mathematical expression:

$$H_{ex} = A_{Ra} / 370 + A_{Th} / 259 + A_K / 4810 \quad (6)$$

This index represents the external exposure risk from γ-rays and corresponds to a maximum activity concentration of 370 Bq kg^{-1} for the material.

Later, made corrections to the model by taking into account the presence of windows and doors and the finite thickness of walls [29]. They introduced a weighting factor of 0.7 in each case, which increased the maximum allowable concentrations by a factor of 2. The revised formula for Hex is

$$H_{ex} = A_{Ra} / 740 + A_{Th} / 520 + A_K / 9620 \quad (7)$$

To ensure negligible radiation hazard, Hex should be less than unity.

The internal hazard index (Hin) determines the internal exposure to radon and its daughter products, and is calculated by:

$$H_{in} = A_{Ra} / 185 + A_{Th} / 259 + A_K / 4810 \quad (8)$$

If the maximum radium concentration is decreased by 50% from the standard acceptable threshold, the value of Hin will be less than 1.0. According to (El-Taher et al., 2020), to ensure safety in the building, it is recommended that the Hin value be smaller than unity [30].

The findings of the study suggest that the granite samples used in construction materials in Egypt adhere to the prescribed threshold for radioactive criteria. The average computed values of Hex and Hin were found to be less than one for all samples examined. It is noteworthy that the International Commission on Radiological Protection (ICRP-60) advocates for the maintenance of radiation exposure levels above the natural background radiation at a minimum feasible extent while remaining below the prescribed individual dosage limits [31]. The average dosage limit for radiation workers over 5 years is 100 mSv, while for the population, it is one mSv each year. The established limits are derived from a cautious methodology that assumes the absence of a minimum dose threshold below which no effects would occur. Hence, it is recommended to refrain from utilizing granite samples that possess a Ra_{eq} value beyond 370 Bq kg-1 in the construction of residential buildings. Fortunately, the majority of the samples examined in the study exhibit Ra_{eq} values that fall within the established limit.

Table 3: The radiation hazard parameters of the granite samples under investigation

Sample	H_{ex}	H_{in}	Abs. dose rate nGy/h	Ra_{eq} Bq.kg-1	Repr. level index Iγr	Ann. Eff. Dose µSv/y
1	0.35	0.92	117.7	259.0	1.8	144.5
2	0.32	0.82	102.7	235.1	1.6	126.0
3	0.32	0.88	104.3	237.1	1.6	127.9
4	0.33	0.69	104.2	245.0	1.7	127.8
5	0.23	0.50	73.6	171.9	1.2	90.3
6	0.30	0.63	93.7	219.8	1.5	115.0
7	0.26	0.58	84.0	196.0	1.4	103.0
8	0.28	0.61	89.1	208.5	1.5	109.3
9	0.34	0.71	106.1	249.3	1.7	130.2
10	0.26	0.55	81.4	190.5	1.3	99.9
11	0.24	0.52	76.7	178.7	1.3	94.1
12	0.32	0.68	101.0	236.9	1.7	123.9
13	0.24	0.54	77.2	179.6	1.3	94.7
14	0.24	0.53	77.1	179.5	1.3	94.6
15	0.30	0.65	95.7	224.1	1.6	117.4
16	0.32	0.69	101.4	237.7	1.7	124.4
17	0.24	0.53	75.8	176.5	1.2	93.1
18	0.25	0.55	78.9	183.6	1.3	96.8
19	0.24	0.54	75.6	175.9	1.2	92.7
20	0.24	0.54	76.9	178.7	1.3	94.3

4 Conclusions

The findings demonstrate notable discrepancies in the values of Ra_{eq}, I, and gamma absorbed dose rates among various granite samples, which can be ascribed to disparities in the potassium, thorium, and radium concentrations. The granite

rocks Ra_{eq} values of the study area exhibit an average that falls below the universally recognized threshold of 370 Bq kg-1. The rates of gamma-absorbed radiation in the Egyptian rock air show a degree of comparability to the global average terrestrial radiation level of 55 nGy.h-1. This falls within the measured worldwide range of 28-120 nGy.h-1. The hexavalent chromium (Hex), hydrogen ion concentration (Hin), and the effective dose equivalent values for all samples demonstrate a notable decrease compared to the permissible threshold. Indications imply that the granite can be used in building while adhering to the prescribed threshold for radioactivity. The aforementioned findings can be used as reference values and serve as a foundational dataset for the development of a radiological map of the area.

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