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A Study on Radiation Hazard of Granite and Marble Widely Used in Jordan Using Gamma Ray Spectrometer

A. N. Akour*, S. Shakhatreh

Department of Basic Scientific Sciences, Al-Huson College, Al-Balqa Applied University, Salt, P.O. Box 50, Al-Huson 21510, Jordan

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ABSTRACT

Granite and marble are widely used in building construction, so possible radioactive nuclides inside them may contribute to the exposure dose to human health. The purpose of this study was to investigate the natural radioactivity concentration and assess the radiological risk limits and health care. The samples of marble and granite were pulverized into small, fine, smooth pieces and counted with the GAMMA-X (GMX) spectrometer to measure the radioactivity concentrations of ^{238}U, ^{232}Th, and ^{40}K. The radiological dose, internal and external hazards, and radium equivalent activity were calculated with a standard formula. The results showed that the radioactive concentrations of ^{238}U, ^{232}Th, and ^{40}K in granite were higher than those in marble. The external hazard for granite samples was below unity, while its internal hazard exceeded unity. The radium equivalent activity did not exceed the critical legal level of 370 Bq/kg as a safe level. For marble, the external and internal hazards and radium equivalent activities showed good agreement with the safe construction level. Its external and internal hazards were less than unity, whereas the radium equivalent activities were less than the critical legal level.

INTRODUCTION

Marble, granite, and stone are widely used troughout the world as building and ornamental materials. Jordan is one of the countries that imports a large amount of these materials from other countries. They are commonly used in the kitchen, shelves and as floor slabs. Since Jordan is one of the countries that do not depend on nuclear energy for electricity, the radiation dose studies for the public are mostly focused on natural radioactive materials such as soil [1], sand [2], rocks and water [3], dust [4,5] and sediments [6], where all these materials contribute to a significant exposure dose but mostly under the safety area levels. This study is the first one in Jordan on our known that holds the exposure dose for building materials, granite and marble that are used in Jordan. Since igneous rocks such as granite have higher radioactivity than other kinds of rocks, it is necessary to determine their radioactivity contribution dose for the public. People spend most of their time indoors, so it may increase the exposure dose of the public.

Natural primary radioactivity, such as ^{40}K, ^{238}U, ^{232}Th and its daughter has been linked to a variety of cancers, including lung and blood cancers [7] and induced genetic damage in the livers. This calls for us to determine the exposure dose from several materials around the public, mainly the mostly indoor-used materials such as granite and marble. Many researchers have investigated the radioactivity concentrations and their hazards in some materials, such as soil [1] where the internal and external hazards are so negligible; building materials [8] where the mean values of internal and external hazard are 0.366 and 0.266, respectively; marble and granite [9,10] where the internal and external hazard for marble is within the safe levels while the granite hazard mostly exceeds the unit. These studies [8-10] also reported different radioactivity concentrations from one region to

*Corresponding author.
E-mail address: abdulrahmanakour1@yahoo.com
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another. Our study includes local and nonlocal samples that were imported from Brazil, India, Portugal, Saudi Arabia, and Turkey.

This study aimed to shed light on the granite and marble radioactivity concentrations and their relevant external and internal hazards, as well as the absorbed dose rate determination using the GAMMA-X (GMX) spectrometer. This study also presents baseline data on environmental radioactivity that will be used in Jordania as well as its comparison with that of other countries.

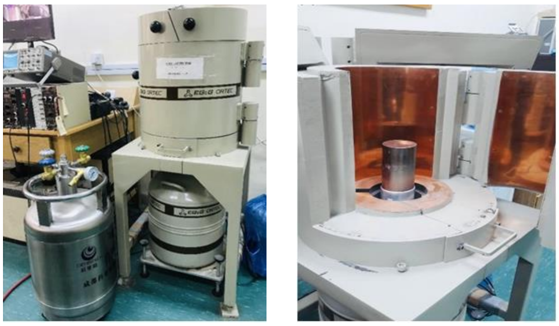
MATERIALS AND METHODS

Nine commercial marble and granite samples were taken from a factory located in Al-Rumtha and were imported from some countries (Table 1). The samples were pulverized to obtain small fine, smooth pieces such as dust and this was done in the Stone Quarry and Al-Hoson College Mining Laboratory. The pulverized samples were then put in a standard suitable vessel specified for the GMX spectrometer, which was tightly closed and left for about one month to reach secular equilibrium. The mean weight of the samples was 53.31±2.15 g.

Table 1. Comparison of radium equivalent activity (Ra_{eq}), external hazard (Hext) and internal hazard (Hin) for granite and marbles between our study and Tunisia and China.

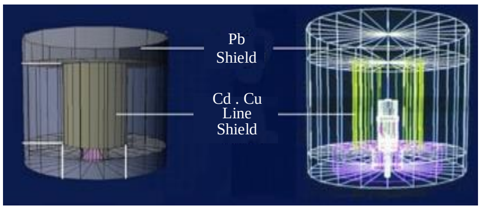
Materials	Source	Ra_{eq} range Bq/kg	H_{ext} range $*10^{-2}$	H_{in} range $*10^{-2}$
Marble	Tunisia [10]	9.04 - 110.9	3 - 50	4 - 51
Marble	Our study	14.46 - 181.04	4 - 32	5 - 37
Granite	China [12]	67 - 490	19 - 136	7 - 102
Granite	Our study	308.37 - 353.41	83 - 95	100 - 117

The measurement of radioactivity concentration was conducted with the GAMMA-X (GMX) spectrometer (Fig. 1). As the background activity was an empty vessel. The counting system consists of a Germanium (Ge) detector of n-type with a crystal length of 8.35 cm; the active diameter of the germanium crystal is equal to 6.24 cm and the 0.3 μm thickness of the bypass window. For the protection layer, about 11.6 cm of graded leads were put around the detector to minimize the background counts, along with liners of tin and copper to decrease the fluorescence of x-rays from lead to preserve the efficiency calibration (Fig. 2).

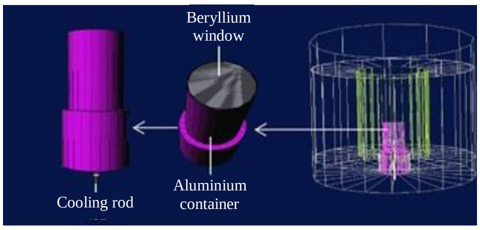


(a) (b)

Fig. 1. (a) The GMX n-type coaxial HPGe detector inside the standard lead shield and completed with LN_2 Dewar for nitrogen supply; (b) The lead shield surrounding the GMX detector to reduce the background radiation.



(a)



(b)

Fig. 2. (a) The GMX detector as viewed from outside the Pb shield; (b) The GMX detector as viewed from inside the Pb shield.

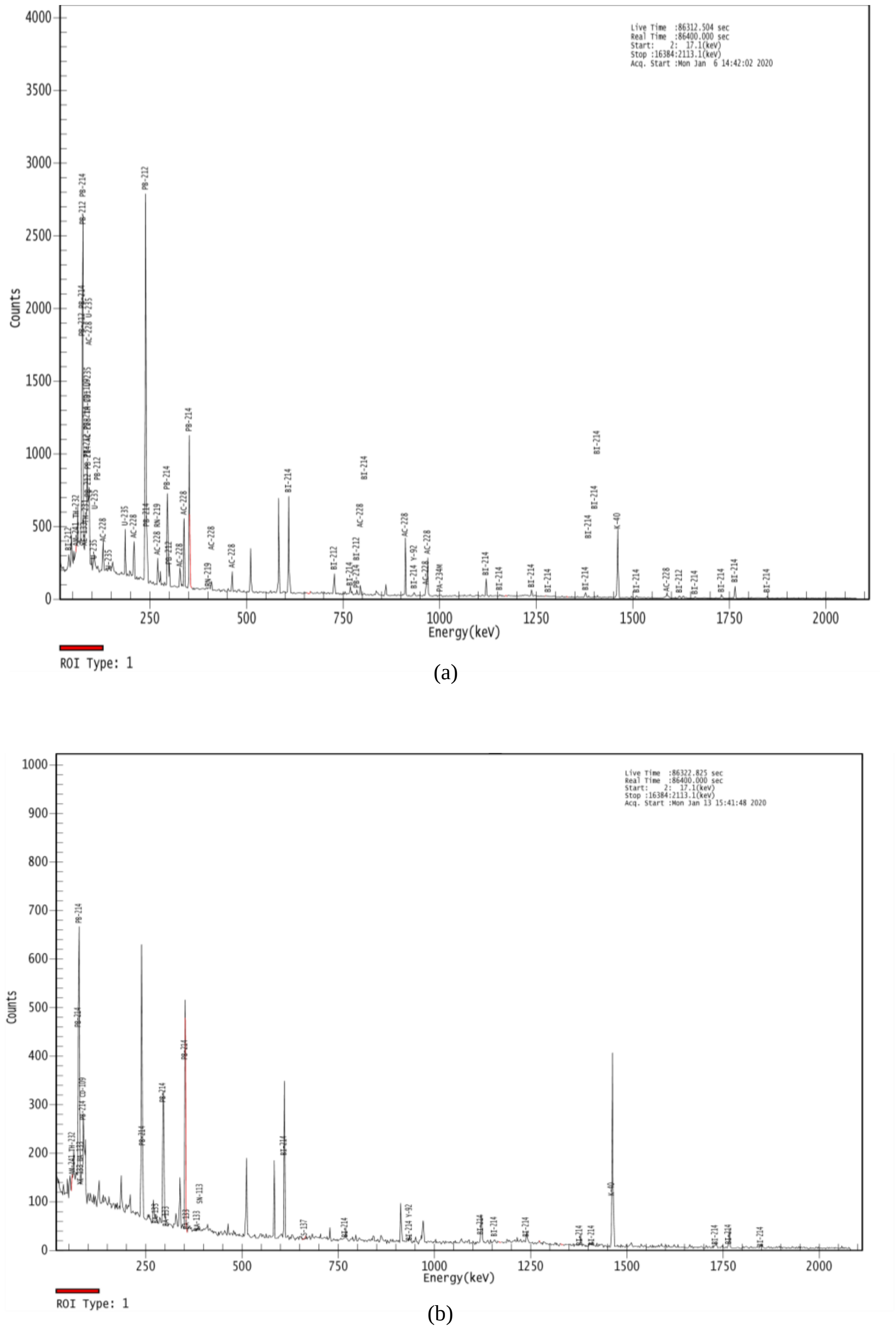
The experimental statistics was fashioned using GMX detector, employing the pattern on the HPGe crystal for a significant period, and then the analysis of spectrometry data was handled by Gennie 2K software. The calibration of energy was settled through the use of a sealed blend source containing some nuclides that emit gamma-rays covering the required energy variety.

The Genie 2K Analyzer software program provides the calibration spectrum using a least-square fit for more than two pairs of energy as a function of channel variety over the whole energy range.

The GEANT4 simulation holds correction factors to investigate the experimental efficiency curve. This simulation accounts for different corrections such as self–absorption and coincidence summing. The software of Genie 2K was used to determine the efficiency at any energy level when analyzing a new undetermined spectrum, using a choice of fitting paradigms function created in the system.

RESULTS AND DISCUSSION

The ^{238}U radioactivity levels were determined from the activity concentration of ^{214}Bi at 609 keV energy, while the ^{232}Th levels were determined from ^{228}Ac at 911 keV energy. This is for the secular equilibrium of its daughters, whereas ^{40}K levels were determined at 1460 keV. The gamma spectrum for one granite sample (G1) and marble sample (M2) is shown as an example in Fig. 3. There was an explicit difference in the concentration levels between marble and granite. As shown in Fig. 4 the radioactive concentrations for ^{40}K, ^{238}U and ^{232}Th in granit were higher than those for marble. This is because granite is an igneous rock that is dispersed from the earth's core which is the source of natural radioactivity. The most significant radioactivity contribution comes from ^{40}K rather than ^{238}U and ^{232}Th in both granite and marble samples.



(a)

(b)

Fig. 3. (a) Example of gamma spectrometry spectrum for granite (sample/G1);
(b) Example of gamma spectrometry spectrum for marble (sample/M2).

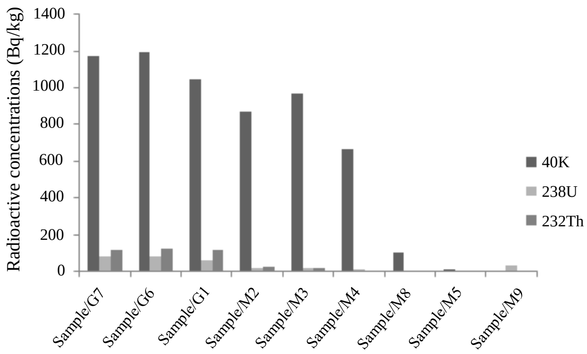


Fig. 4. The ^{40}K, ^{238}U and ^{232}Th radioactivity concentration levels for each sample.

^{40}K radioactive concentration levels in granite was varied from 1043.6 to 1191.5 Bq/kg, on the other hand the variation in marble was 7.76 to 970.6 Bq/kg. ^{238}U concentration levels in granite was varied from 65.33 to 69.53 Bq/kg while in marble was 4.00 to 31.49 Bq/kg. ^{232}Th concentration levels in granite was from 117.7 to 127.46 Bq/kg, whereas in marble was 2.07 to 22.87 Bq/kg. The concentration levels are tabulated in Table 2.

Table 2. Radiation concentration levels for each sample.

Sample number	Origin	Radioactive concentrations (Bq/kg)		
		^{40}K	^{238}U	^{232}Th
sample/G 7	Brazil	1169.80 ± 19.1	65.33 ± 0.731	120.24 ± 1.47
sample/G 6	Brazil	1191.50 ± 19.5	65.73 ± 1.12	127.46 ± 1.54
sample/G 1	India	1043.60 ± 17.6	69.53 ± 1.12	117.70 ± 1.55
sample/M 2	India	867.45 ± 14.8	31.49 ± 7.75	22.87 ± 1.24
sample/M 3	Saudia	970.06 ± 15.9	28.20 ± 0.722	18.35 ± 1.20
sample/M 4	Saudia	667.47 ± 12.6	11.32 ± 0.564	6.82 ± 0.851
sample/M 8	Portugal	104.91 ± 5.39	7.36 ± 0.500	2.07 ± 0.540
sample/M 5	Turkey	10.74 ± 2.34	4.00 ± 0.408	Not detected
sample/M 9	Jordan	7.76 ± 3.25	12.18 ± 0.657	Not detected

The radiological dose, internal and external hazards, and radium equivalent activity were calculated as shown in Table 3. The following Eqs. (1-3) were used to determine the radium equivalent activity Ra_{eq}, the external hazard H_{ex}, and the internal hazard H_{in}.

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

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \qquad (2)$$

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \qquad (3)$$

Table 3. The radium equivalent activity, external and internal hazards for each sample.

Sample number	Ra_{eq} (Bq/kg)	H_{ext}	H_{in}
sample/G 7	343.12	0.93	1.15
sample/G 6	353.41	0.95	1.17
sample/G 1	308.37	0.83	1.00
sample/M 2	117.70	0.32	0.37
sample/M 3	118.04	0.32	0.36
sample/M 4	71.75	0.19	0.22
sample/M 8	14.64	0.04	0.05

Both the H_{ex} and H_{in} index must be less than one for the safety level of building construction [11]. For all granite samples, the internal hazard values were equal to or exceeded one while the external hazard values highly approached one; it means the granite is not advised to be used in the building materials. Otherwise, samples no. 7 and 6 that were from the same country (Brazil) showed different hazard values. Indian granite samples showed different levels but has lower hazard values than Brazillian ones; this was explicitely influenced by regional geology factors. Marble samples have an external and internal hazard of less than one so that they can be safely used in building construction. Ra_{eq} for granite samples showed a high concentration but did not exceed the legal value for safety. Other samples, especially marble samples, show low acceptable radioactive levels (Fig. 5). The results of the determination for samples no. 5 and 9 were not known since ^{232}Th was not detected (Figs. 5 and 6).

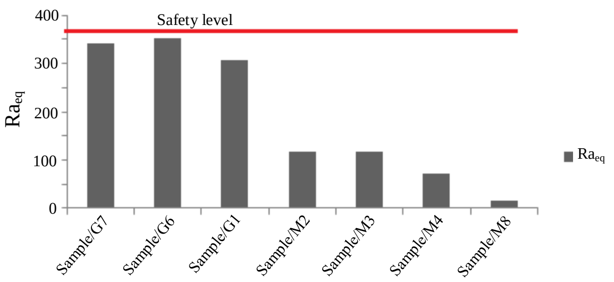


Fig. 5. The radium equivalent activity, Ra_{eq} for each sample.

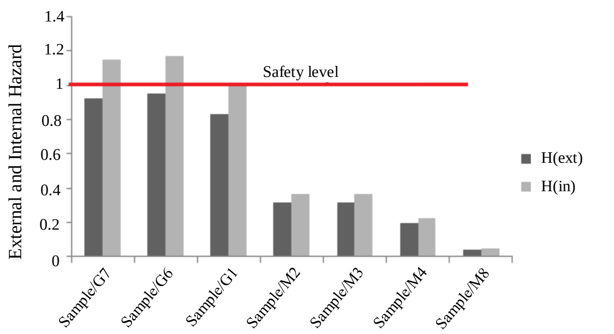


Fig. 6. External and internal hazards (H_{ex}, H_{in}) respectively for each sample.

A comparison of Ra_{eq}, H_{ext} and H_{in} for granite and marbles from Tunisia [10] and China [12] with this study was done and showed a remarkable sense Table 1. The comparison also found that the internal and external hazards for marble were suitable with the healthy building hazard level, while the granite was seemingly not safe for building materials.

CONCLUSION

Nine samples of granite and marble were investigated for their natural radioactivity concentration using gamma ray spectroscopy. The Ra_{eq} concentration and external and internal hazards were also determined. Granite resists the standard and safe condition of construction since the external and internal hazards indices were more than or approaching to one and Ra_{eq} also approached the critical legal level. On the other hand, marble showed an acceptable level of safety for construction due to the lower values of Ra_{eq} , H_{ex} and H_{in}. Our results showed a good sense compared with other studies' results.

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AUTHOR CONTRIBUTION

Dr. Abdulrahman Akour is the correspending author, he collect samples; writes the draft manuscript body (abstract, analysis, figures, tables, conclusion, involving in introduction and methodology, and involving in read and approve the final manuscript).

Dr. Saleh Shakhetreh is a coauthor in this study; he shared in preparing samples and involving in writing and revising the draft manuscript specially the introduction and methodology; also involve in read and approve the final manuscript.

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