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Evaluation of Natural Radioactivity in Granite and Marble used as Flooring Materials in Baghdad Province / Iraq

Zaid Zeyad*, Yaaroub Faleh Khalf AL-Fatlawy
Department of Biology, College of Science, University of Baghdad, Baghdad, Iraq.

Abstract

Twenty four samples of various granites and marble used common as flooring materials were collected from many factories and market in Baghdad Province, Iraq. Natural radioactivity of ^{226}Ra, ^{232}Th and ^{40}K in the samples was measured by using high purity germanium detectors with an efficiency of 40%. The Specific activity concentration in granite samples ranged from 9.20 to 79.64 Bq/kg, 10.50 to 229.40 Bq/kg and 185.30 to 1391.20 Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The estimated of radiological parameters: radium equivalent activity (R_{eq}), activity concentration Index (I) and hazard indices (internal (H_{in}) and External (H_{ex})) that associated with the radiation hazard in the samples. The results of activity specific concentrations and radiological parameters of ^{226}Ra, ^{232}Th and ^{40}K were compared with the reported data of the worldwide limit of building materials.

Keywords: Natural radioactivity, granite, high purity germanium detectors, hazard indices.

تقييم النشاط الاشعاعي الطبيعي في الجرانيت والرخام المستخدم في محافظة بغداد/ العراق

زيد زياد*، يعرب فالح خلف الفتلاوي
قسم علوم الحياة، كلية العلوم، جامعة بغداد، بغداد، العراق

الخلاصة

تم جمع اربع وعشرون عينة من الجرانيت والرخام التي تستخدم بصورة شائعة كمواد ارضيات من مختلف المعامل والأسواق في مدينة بغداد. وتم قياس النشاط الاشعاعي للراديوم والثوريوم والبوتاسيوم بأستخدام كاشف الجرمانيوم عالي النقاوة ذو الكفاءة ٤٠ %. وتراوحت الفعالية النوعية في عينات الجرانيت من 9.20 الى 79.64بيكرل/كغم و 10.50 الى 229.40 بيكرل/كغم و 185.30 الى 1391.20 بيكرل/كغم للراديوم والثوريوم والبوتاسيوم على التوالي. بينما تراوحت الفعالية النوعية في عينات الرخام من ٠.٢٤ الى ١٦.٢ بيكرل/كغم و ٠.١٧ الى 38.23 بيكرل/كغم و ٢.٨٢ الى ٨٨٦.٥٦ بيكرل/كغم للراديوم والثوريوم والبوتاسيوم على التوالي. أيضا تم حساب معاملات الخطورة الاشعاعية في العينات : مكافئ الراديوم، معامل التركيز الفعال، معاملات الخطورة (الداخلية والخارجية). و تمت مقارنة نتائج الفعالية النوعية ومعاملات الخطورة الاشعاعية مع المحددات العالمية لمواد البناء.

Introduction

The human population is always exposed to ionizing radiation due to background radiation sources. Besides man-made radiation, the main source of background radiation is natural radioactivity [1]. Natural radioactivity has existed since the beginning of the universe due to the long half-life of the natural radio element found in the earth's crust [2]. The radionuclides of ^{238}U, ^{232}Th and ^{40}K can be found almost in all types of flooring materials are produced from the earth's crust [3]. These

*Email : zadnan61@gmail.com

radioactive elements pose exposure hazards externally due to their emissions of gamma ray and internally due to radon and its daughter which alpha particles emitting [4,5].

The human body contains a slight quantity of naturally occurring radioactive elements in muscles, bones and tissue, in particular radioactive gases like Radon is concentrated in the different portions of the body and contribute to the internal dose by ingestion and inhalation [6]. Because people spend most of their time (about 80%) indoors, Its important knowledge of natural radioactivity to determining the amount of public exposure in flooring materials [7].

The flooring materials (granite, marble, etc.) are a part of building materials that contain several amounts of natural radioactive elements. Materials obtain from rock and soil have mainly natural radionuclides of the uranium (^{238}U) and thorium (^{232}Th) chains, and the radioactive isotope of Potassium (^{40}K) [8].

In the uranium series, the segment of the decay chain starting from radium (^{226}Ra) which is the most important isotope radiologically and, therefore, reference is frequently made of radium instead of uranium [9].

Materials and Methods

Collection and preparation

In total, 24 flooring materials' samples were collected from different markets and factories. The samples were chosen in terms of the widely and most common type found in Iraqi markets. The types of materials are: 10 samples of granite (GR), 14 samples of marbles (MR). The samples were classified in the following Table-1.

Each sample was pulverized into small pieces, then into fine powder using jaw crusher. The samples were dried at 100 °C for 2 hours to ensure that any moisture was removed from the samples and then to obtain uniform particle sizes, a 650μm mesh was used to sieve the samples after that samples were weighted (1kg) and transferred to a Marinelli beaker (1 L)[10].

The samples were stored and kept sealed for about one month before measurements in order to achieve the secular equilibrium for ^{238}U and ^{232}Th with their respective progenies [11].

Table 1- The Type, Symbol, Company or Commercial name, Color and Country of Origin granite materials.

No.	Type	Symbol	Company or Commercial name	Color	Country of Origin
1	Granite	GR1	Gendola C116	Red buffer	China
2	Granite	GR2	Tone brown	Black + Cherry	India
3	Granite	GR3	Robby	Cherry	India
4	Granite	GR4	Norway	Black+orange	Brazil
5	Granite	GR5	Imperial	Red	China
6	Granite	GR6	Tiger (skin red)	Pink	China
7	Granite	GR7	Coffee	Brown + black	China
8	Granite	GR8	Galaxy	Black shine	India
9	Granite	GR9	Sea wave	Black+ gray	China
10	Granite	GR10	Imperial	Gray	China
11	Marble	MR11	Non	Gray	Vietnam
12	Marble	MR12	Non	Beige	Italy
13	Marble	MR13	Afyon sugar	White	Turkey
14	Marble	MR14	Leo Beige	Beige	Omani
15	Marble	MR15	Non	Black	Brazil
16	Marble	MR16	Non	Dark red	India
17	Marble	MR17	Non	White	Vietnam
18	Marble	MR18	Mugla	Silver	Turkish
19	Marble	MR19	Kermanshah	Beige	Iran
20	Marble	MR20	Dream Pink	Pink	Iran
21	Marble	MR21	Lilac	White + Gray	Turkish
22	Marble	MR22	Graffiti	Black	Iran
23	Marble	MR23	Manksha	White	Turkish
24	Marble	MR24	Baroda Green	Green	India

Spectroscopy Analysis

The specific activity concentration was obtained from an average of gamma-ray photo peaks at several energies. The gamma-ray lines at 295.2 and 351.9 keV from lead (^{214}Pb) and at 609.3 keV from bismuth (^{214}Bi) were used to determine the specific activity of ^{226}Ra. The gamma-ray lines of 238.63 KeV of lead (^{212}Pb), the 583.2 KeV from Thallium (^{208}Tl) and at 911.2 keV from Actinium (^{228}Ac) were used to determine the specific activity of ^{232}Th. The specific activity of ^{40}K was estimated directly by its own gamma-ray line at 1460.8 keV. These specific activity estimated by using the equation [10]:

$$\boldsymbol{A} = \frac{\boldsymbol{N-BG}}{\boldsymbol{T}.\boldsymbol{I_\gamma}(\boldsymbol{E_\gamma}).\mathcal{E}(\boldsymbol{E_\gamma}).\boldsymbol{M}}$$

Where A: is the specific activity of radioactive elements measured in (Bq/kg) units, B.G: is the net peak area of the background, Ɛ (Eγ): is the detector efficiency at energy (Eγ), Iγ (Eγ): is the abundance at energy (Eγ), M: is the mass of the soil sample (kg), T: the measured time, which is equal (3600 s).

Calculation of radiological parameters (Hazard index)

The natural radioactivity distribution in the samples under current study was not uniform. Thus, a common hazard index has been introduced to estimate the real activity level of ^{226}Ra, ^{232}Th and ^{40}K in the samples and other radiation hazards associated with these radionuclides [12].

1. Radium Equivalent Activity (Ra_{eq})

To represent the activity concentrations of ^{238}U (^{226}Ra), ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them, The index is named radium equivalent activity (Ra_{eq}) and which was used to confirm the uniformity in the dissemination of natural radionuclides ^{238}U (^{226}Ra), ^{232}Th and ^{40}K and it is given by the following equation [13,14]:

$$Ra_{eq}\,(Bq/kg) = A_{Ra} + 1.43A_{Th} + 0.077A_K$$

Where: A_{Th}, A_{Ra} and A_K are the specific activities concentration of ^{226}Ra, ^{232}Th and ^{40}K in (Bq/kg) units.

2. Activity Concentration Index (I_γ)

The activity index (I_γ) for ending building materials intended for use in building construction [14,15]:

$$I_\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000}$$

Where A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the ending product. If the value of the activity concentration index is < 1 unit or less, the corresponding material can be used as building material, so far as the radioactivity is concerned without restriction. If the value exceeds 1unit, the corresponding material can not be used in the building.

3. External (H_{ex}) and Internal (H_{in}) Hazard Index

The internal hazard index (H_{in}) is given by the following equation [16]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1$$

The external hazard index (H_{ex}) is assumed by the following equation [17]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1$$

These index values must be lower than unity in order to must the radiation hazard to be not harmed.

Result and Discussion

Specific Activity

The results of specific of natural radioactivity concentration (^{226}Ra, ^{232}Th and ^{40}K) in the present study revealed the highest specific activity in granite samples of ^{226}Ra was 145.3 Bq/Kg for Chinese sample (GR10) while the lowest specific activity was 9.2 Bq/Kg for Indian sample (GR8), the results presented in Table-2.

The specific activity of ^{226}Ra in marble samples ranged 0.24 Bq/Kg for Omani sample (MR14) to 16.2 Bq/Kg for Iranian sample (MR22).

The specific activity results of ^{232}Th in show the highest specific activity granite samples was 229.4 Bq/Kg for Indian sample (GR3) while the lowest specific activity was 0.٣٦ Bq/Kg for Indian sample (GR8).

The specific activity of ^{232}Th in marble samples shows the maximum value was 38.3 Bq/Kg for Brazilian sample (MR15) while the minimum value was 0.17 Bq/Kg for Italian sample (MR12).
The highest specific activity of ^{40}K in in granite was 1391.2 Bq/Kg for Brazilian sample (GR4) while the lowest specific activity was 185.3 Bq/Kg for Indian sample (GR8). In marble, the highest specific activity was 886.56 for Brazilian sample (MR15) while the lowest specific activity was 2.82 for Iranian sample (MR22).

The variation of the specific activity concentration (^{226}Ra, ^{232}Th and ^{40}K) in the present study of flooring materials due to the geochemical characteristics (mineral content, materials) and geology of the region [18].

Table 2- The specific activity in granite and marble sample.

No.	٢٢٦Ra (Bq/Kg)	^{232}Th (Bq/Kg)	٤٠K (Bq/Kg)
GR1	71.9	116.111	1241.18
GR2	11.09	88.09	1245.02
GR3	68.17	229.4	1335.7
GR4	60.988	140.58	1391.2
GR5	71.39	102.9	1095.17
GR6	28.4	66.7	1151.1
GR7	34.1	71.7	1028
GR8	9.2	10.5	185.3
GR9	37.5	67.5	879.1
GR10	79.64	173.26	1210.5
MR11	1.3	0.84	12.4
MR12	3.48	0.17	86.7
MR13	1.55	0.41	12
MR14	0.24	0.24	7.73
MR15	12.23	38.23	886.56
MR16	11.35	23.42	486.12
MR17	2.4	0.72	86.4
MR18	3.4	1.45	97.1
MR19	2.712	0.63	3.5
MR20	1.2	1.5	83.2
MR21	1.95	1.82	20.43
MR22	16.21	2.52	2.82
MR23	1.4	0.64	48.2
MR24	9.12	1.73	32.313

Radiological parameters (Hazard index)

The radium equivalent activity results for granite samples in present study ranging from 38.48 Bq/Kg for Indian sample (GR8) to 499.06 Bq/Kg for Indian sample (GR3). In marble, the highest radium equivalent activity was 135.15 Bq/Kg for a Brazilian sample (MR15) while the lowest value was 1.18 Bq/Kg for Omani sample (MR14) the results obtained are as shown in Table-3.

All sample results of marble were within worldwide limit of 370 Bq/Kg, as recommended by (UNSCEAR, 1993) while 20% of granite and porcelain samples were higher than the permissible limit.

The values of the activity concentration index in granite samples ranged from 0.145 to 1.81 corresponding to Indian sample (GR8) and Indian sample (GR3). In marble, the highest value recorded in a Brazilian sample (MR15) with 0.527 while the lowest sample value recorded in the Omani sample (MR14) with 0.005.

The results show 50% of granite samples were higher than the worldwide limit ($I_\gamma > 1$) (UNSCEAR, 1993), while all other samples were within the worldwide limit.
The values of internal and external hazard indices for granite samples vary from 0.129 and 0.104 to 1.532 and 1.348 for Indian sample (GR8) and Indian sample (GR3), respectively. The result values

of marble samples varying from a minimum value of 0.004 and 0.003 in the Omani sample (MR14) to a maximum value of 0.398 and 0.365 recorded in a Brazilian sample (MR15).

The results of internal hazard index show 50% of the granite samples were higher than the worldwide limit ($H_{in} < 1$) while marble samples were within the worldwide limit (UNSCEAR, 1993). The results of external hazard index show 20% of the granite samples were higher than the worldwide limit ($H_{ex} < 1$), while marble samples were within the worldwide limit (UNSCEAR, 1993).

Table 3- Radium equivalent activity concentration index and hazard indices for granite and marble samples.

No.	Ra_{eq} (Bq/Kg)	I_γ	H_{in}	H_{ex}
GR1	333.51	1.234	1.095	0.901
GR2	232.93	0.892	0.659	0.629
GR3	499.06	1.819	1.532	1.348
GR4	369.14	1.370	1.162	0.997
GR5	302.87	1.118	1.011	0.818
GR6	212.42	0.812	0.650	0.574
GR7	215.79	0.815	0.675	0.583
GR8	38.48	0.145	0.129	0.104
GR9	201.72	0.756	0.646	0.545
GR10	420.61	1.535	1.351	1.136
MR11	3.46	0.013	0.013	0.009
MR12	10.40	0.041	0.037	0.028
MR13	3.06	0.011	0.012	0.008
MR14	1.18	0.005	0.004	0.003
MR15	135.16	0.527	0.398	0.365
MR16	82.27	0.317	0.253	0.222
MR17	10.08	0.040	0.034	0.027
MR18	12.95	0.051	0.044	0.035
MR19	3.88	0.013	0.018	0.010
MR20	9.75	0.039	0.030	0.026
MR21	6.13	0.022	0.022	0.017
MR22	20.03	0.068	0.098	0.054
MR23	6.03	0.024	0.020	0.016
MR24	14.08	0.050	0.063	0.038

Conclusions

The most samples of granite had ^{226}Ra, ^{232}Th and ^{40}K specific activity concentrations higher than the worldwide limit for building materials. The results of marble show that contain low natural radioactivity. The result values of hazard index (Ra_{eq}, I_γ, H_{ex} and H_{in}) for some samples of granite were found to be higher than the worldwide limit. So, these samples should be prevented used as flooring materials for constructions.

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