

Radioactivity Measurement in Different Types of Fabricated Building Materials Used in Palestine

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Abstract

The natural radionuclides (^{238}U , ^{232}Th and ^{40}K) and the manmade radiation levels were measured in samples of different types of fabricated building materials in Palestine. Concentration of radionuclide in samples were determined by γ -ray spectrometry using hyper-pure germanium (HPGe) detector in Bq/Kg dry weight . In this paper samples of commonly building materials (granite, clay brick (karmeed), lime stone, marble, cement, white cement, sea sand, gravel powder, gravel, glue ceramic, gypsum powder and hydrated lime) used in Palestinian buildings were collected . the concentration values of ^{238}U , ^{232}Th and ^{40}K in these samples ranged between 13.9-97.3, 7.2-78.6 and 2.0-1139.0 Bq/Kg respectively.

The ^{137}Cs isotope was detected in some samples. Radium equivalent activity (R_{eq}), dose rate in air (D_r) , external hazard index (Hex), radioactivity Level index (I_γ) and annual gonadal equivalent dose (D) in all samples were calculated. The activity concentration data were discussed are compared with other experimental values in some countries.

Key words: Building materials, Radioactivity, γ -ray spectroscopy.

Introduction

Studies and surveys of natural environmental radiation are of great importance, because it is the main source of exposure to human kind and other living organisms. Building materials are one of the sources which cause direct radiation exposure. The exposure of the public to natural sources of radiation has been estimated by the United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR, 1996) with an effective dose equivalent to 2.4 mSv/yr of which 1.1 mSv/yr is due to the basic background radiation and 1.3 mSv/yr is due to exposure to radon⁽¹⁾

Studies of the radiation dose from this source and its effects on health improve the understanding of radiation damage and therefore are of great value as a reference when standards and regulators control actions on a radiation protection are established. Calculations of the environmental dose level produced by natural and artificial radionuclides enable us to establish an important component of the environmental dose received due to external radiation originating from terrestrial radionuclide of natural origin. The important radiological consequence of the natural radioactivity in building materials are two-fold: irradiation of the body by γ -rays and irradiation of the lung tissue by ^{222}Rn decay products. The uranium and thorium series and ^{40}K isotope are usually present in fabricated building materials. Therefore, the radionuclide contents in building materials were measured by several researchers⁽¹¹⁻²⁷⁾.

The present study has been carried out to provide information about the radionuclide concentrations in various building materials used in Palestine. The measurements helps to implement precautionary measures whenever the dose is found to be above the recommended limits and knowledge of γ -radioactivity is required by the building construction association to adopt preventive measures to mitigate the harmful effects of ionizing radiations.

This work aims to measure the gamma activity due to ^{238}U , ^{232}Th and ^{40}K from some types of fabricated building materials used in Palestine buildings and to compare the radionuclides concentrations in different samples with many studies in the world.

Experimental Procedure

1. Sampling and sample preparation:

Twenty four fabricated sample of 12 different building materials used in Palestine where collected for radioactivity measurement. The samples were crushed, sieved through <2mm meshes, homogenized and dried in an oven at 80 °C for 12 hr to eliminate moisture. The prepared samples were placed in 1000 ml Marinelli beaker, which is then sealed for 4 weeks to reach secular equilibrium between ^{226}Ra and ^{232}Th and their progeny to be ready for γ -ray measuring using HPGe detector⁽²⁾. Each sample was analyzed after secular equilibrium of ^{226}Ra and ^{232}Th with their decay products was obtained using HPGe detector and MCA with 8000 channel.

2. Instrument and Calibration:

The gamma system used in analysis of the collected samples are Ortec coaxial HPGe detector. The detector has a photo peak efficiency of 15% and energy resolution of 1.85 KeV, full width at half maximum (FWHM) for the 1333 KeV gamma line of ^{60}Co . To reduce gamma-ray background, a cylindrical lead shield with a fixed bottom and movable cover shielded the detector used. The lead shield contained three inner concentric cylinders of lead, copper and cadmium.⁽³⁾

The calibration of the spectrometer was carried out using standard sources: ^{241}Am (60KeV), ^{109}Cd (88KeV), ^{57}Co (122KeV), ^{60}Co (1173 KeV and 1333KeV), ^{139}Ce (166KeV), ^{203}Hg (279KeV), ^{113}In (392KeV), ^{85}Sr (514KeV), ^{137}Cs (662KeV) and ^{88}Y (898 KeV and 1836 KeV) in the energy range(60-1850) KeV. Also, efficiency calibration curve was made using different energy peaks covering the rang up to 2500 KeV.⁽⁴⁾ The standard source packed in Marinelli beaker has same geometry as that used for measured samples. An empty bottle with the same geometry was taken for subtracting the background. The measuring time of both activity and background measurements ranges between 35000 and 70000 seconds. The background spectra were used to correct the net γ -rays peak areas for studies isotopes. Absolute efficiency calibration curves were calculated to study isotopes by using the last standard sources and chemically pure KCl dissolved in distilled water at different concentrations.⁽⁵⁾

The absolute efficiency of the detector for each gamma-ray energy was then calculated using the following formula.⁽⁶⁾

$$Eff. = Z/(A.I) \text{ cps/Bq} \dots \dots \dots (1)$$

Where *Eff.* is the absolute photo peak efficiency, Z is the net counts per second for the standard, A is the activity of the radionuclides used in Bq and I is the absolute intensity of the gamma transition. The efficiency calibration curve by using the standard source is shown in figure(1).

The gamma-ray transitions used to measure the concentration of the assigned nuclides in the series are as follows⁽⁷⁾:

1. ²¹⁴Pb (351.9 KeV), ²¹⁴Bi (609.3, 768.4, 934.1 , 1120.3 1238.1 and 1764.8 KeV) and ²²⁶Ra 186.2 (KeV) for the uranium series.
2. ²²⁸Ac (209.5 , 338.5, 463.5 , 911.1 and 968.9 KeV), ²⁰⁸Tl (583.1, and 860.1 KeV) and ²¹²Bi (727.2 , 785.4 and 1620.6 KeV) for the thorium series.
3. ⁴⁰K(1460.8 KeV) for potassium.
4. ¹³⁷Cs (661.3 KeV) for cesium.

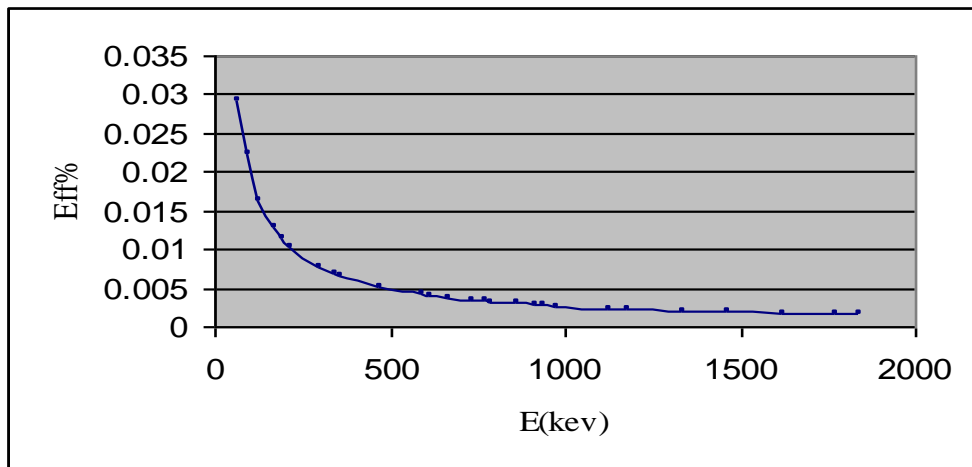


Fig. (1): The photopeak efficiency of HPGe detector

Calculation of the Basic Radiation Quantities

1. The Activity Concentration (C)⁽⁸⁾:

The radioactivity concentration was calculated by γ -ray spectrometry with the following simple regression:

$$C = \text{Net Area}(c/s) / (I\gamma \cdot \zeta \cdot M) \text{ Bq/Kg} \dots\dots\dots(2)$$

Where C : activity concentration of the gamma- spectra line in Bq/Kg.
Net area (c/s) : the net detected counts per second corresponding to the energy.
 ζ : counting system efficiency of the energy and M : mass of the sample in Kg.

2. Radium Equivalent Activity (Ra_{eq})⁽⁹⁾

Distribution of ²²⁶Ra, ²³²Th and ⁴⁰K in building materials is not uniform. Uniformity in respect of exposure to radiation has been defined in terms of radium equivalent activity Ra_{eq} in Bq/Kg to compare the specific activity of materials containing different amount of ²²⁶Ra, ²³²Th and ⁴⁰K . It is calculated through the following equation:

$$Ra_{eq} = 1.43 C_{Th} + C_{Ra} + 0.077C_k \dots\dots\dots(3)$$

Where C_{Th} : ²³²Th activity concentration in Bq /Kg.
 C_{Ra} : ²²⁶Ra activity concentration in Bq /Kg.

C_k : ^{40}K activity concentration in Bq /Kg

It is assumed that 370 Bq/Kg of ^{226}Ra , 259 Bq/Kg of ^{232}Th and 4810 Bq/Kg of ^{40}K produces the same gamma ray dose rate.

3. The Dose Rate in Air (D_R)^(10,11)

The dose rate in air due to the radioactivity where calculated using the following equation:

$$D_R = 0.104 C_{Ra} + 0.130 C_{Th} + 0.09 C_k \dots \dots \dots (4)$$

Where D_R (10^{-8} Gy/hr) is the absorbed dose rate in air surrounded by an infinite thickness of building material.

4. External Hazard Index (H_{ex})⁽⁹⁾

A modified quantity of radium equivalent activity is external hazard index (H_{ex}), which is defined as follows:

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_k/4810 < 1 \dots \dots \dots (5)$$

The values of H_{ex} must be lower than unity in order to keep the radiation hazard insignificant. The maximum value of unity for H_{ex} corresponds to the limit of 370 Bq/kg for Ra_{eq} . The upper limit of radioactivity levels to general population can be exposed is 0.5 nGy/yr.

5. Radioactivity Level Index (I_γ)⁽¹²⁾

This index can be used to estimate the level of γ -radiation hazard associated with the natural radionuclides. The represented level of radiation hazard index may be defined as:

$$I_\gamma = C_{Ra}/150 + C_{Th}/100 + C_k/1500 \dots \dots \dots (6)$$

May be used to estimate the level of γ -radiation hazard associated with the natural radionuclides in the used samples.

6. The Annual Gonadal Equivalent Dose (D)⁽¹³⁾

The annual gonadal equivalent dose (AGED) for the resident of a house built using a material with given activity concentration of ^{226}Ra , ^{232}Th and ^{40}K was calculated using the following equation:

$$D (\mu\text{SV/yr}) = 3.09C_{Ra} + 4.8C_{Th} + 0.314C_K \dots \dots \dots (7)$$

Where C_{Ra} , C_{Th} and C_K are defined in the last text. This model considers a house to be a cavity with infinitely thick walls, which it possible to compare AGED of a house containing concentration of ^{226}R , ^{232}Th and ^{40}K equal with the world average values :25,25 and 370 Bq/Kg respectively with those obtained using a given material

Results and Discussion

Figure 2, show the partial γ -ray spectrum of gravel samples represented of high concentration of ^{238}U .

work indicates higher values were obtained for some type of cement, gravel, glue, ceramic, gypsum powder and hydrated lime for ^{238}U series, marble, cement and hydrated lime for ^{232}Th series and granite and clay brick for ^{40}K isotope compared to the international average values⁽¹⁴⁾. The ^{137}Cs was measured in some building materials range between 0.14 Bq/Kg and 1.5 Bq/Kg detected in gravel and granite respectively. On the basis of the half-lives of uranium and thorium, the radon and thoron production and hence their gamma emitting daughters in building materials during the life time of a building may be considered to be constant⁽¹¹⁾.

Table (2): The activity mean concentration in (Bq/kg) of fabricated building materials used in Palestine

Sample Code	C_U	C_{Th}	C_K	C_{Cs}
1	35.1	20.5	639.5	1.50
2	32.8	7.2	1139	0.17
3	13.9	13.9	45.9	-
4	36.0	54.7	2.0	0.21
5	85.1	67.6	18.5	-
6	29.8	28.9	31.5	-
7	20.6	18.8	26.3	-
8	24.7	37.7	252.4	-
9	97.3	26.4	3.2	0.14
10	65.9	36.2	2.4	-
11	78.2	9.8	2.7	0.17
12	58.2	78.6	2.9	-
Average	48.1	33.4	180.5	

The activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in Bq/Kg of building material samples in the present study as well as in other studies for many different countries were compared in table (3). Some values obtained in the present study were noticeably low, and others were higher than international values. This is due to the fact that the fabricated building material used in the study regions were generally around the international levels.

Table (3): Comparison levels of radionuclide in building materials in (Bq/kg) dry weight

Type of Building Material	Country	No. of samples	U-238	Th-232	K-40	Ref. No.
Granite	Egypt (central region)	8	39.9	43.5	1062.0	16
	Hong Kong	-	202.0	140.0	1030.0	17
	U.K	-	89.0	81.0	1106.0	18
	Brazil	2	48.6	288.0	1335.0	19
	Germany	32	100.0	80.0	1299.0	18
	Austria	22	55.4	25.0	911.0	20
	Turkey	40	15.9	33.8	359.0	21
	Global Av.	-	33.0	34.1	-	22
	Palestine	2	35.1	20.5	639.5	Present work
Clay brick (Karmeed)	Egypt (central region)	19	24.1	24.0	258.0	16
	Egypt (Alexandria)	-	134.3	68.0	276.6	23
	Hong Kong	-	143.0	158.0	850.0	17

Continue Table (3): Comparison levels of radionuclide in building materials in (Bq/kg) dry

	U.K	25	52.0	44.0	703.0	18
	Brazil	8	51.7	65.3	747.0	19
	Germany	109	59.0	67.0	673.0	18
	Austria	32	38.3	44.7	635.0	20
	Global Av.	-	67.0	67.0	700.0	22
	Palestine	2	32.8	7.2	1139.0	Present work
Limestone	Egypt (central region)	6	14.5	3.4	8.7	16
	Brazil	1	24.3	7.0	205.0	19
	Austria	4	9.0	2.8	34.0	20
	Australia	2	-	11.1	-	12
	Palestine	2	13.9	13.9	45.9	Present work
Marble	Egypt (central region)	14	8.7	0.7	9.7	16
	Egypt (Qena)	-	205.0	115.0	865.0	11
	Brazil	3	16.4	24.5	413.0	19
	Austria	8	7.3	1.7	27.0	20
	Jordan	8	28.7	6.2	42.3	13
	Palestine	2	36.0	54.7	2.0	Present work
Cement	Egypt (central region)	13	25.3	9.6	40.3	16
	Egypt (Alexandria)	-	82.2	15.0	55.1	23
	Hong Kong	-	19.2	18.9	127.0	17
	U.K	-	22.0	7.0	141.0	18
	Brazil	1	61.7	58.5	564.0	19
	Germany	14	<26.0	<18.0	241.0	18
	Austria	18	26.7	14.2	210.0	20
	Australia	7	51.8	48.1	115.0	12
	Pakistan	-	26.1	28.7	279.9	24
	Cuba	-	22.8	10.6	467.0	25
Palestine	2	85.1	67.6	18.5	Present work	
White cement	Egypt (Qena)	-	72.0	46.0	250.0	11
	Cuba	-	44.5	21.8	99.0	25
	Palestine	2	29.8	28.9	31.5	Present work
Sand	Egypt (central region)	25	7.1	4.0	75.4	16
	Egypt (Alexandria)	-	33.1	11.1	82.1	23
	Hong Kong	-	24.3	27.1	841.0	17
	Jordan	10	31.9	32.9	38.8	13
	Australia	3	3.7	40.7	44.4	12
	Pakistan	-	21.5	31.9	520.0	24
	Cuba	-	16.7	15.6	188.0	25
	USA (San Francisco)	2	104.0	88.4	115.0	26
	Nether Land	4	8.1	10.6	200.0	27
	Global Av.	-	15.0	15.0	260.0	22
Palestine (Sea Sand)	2	20.6	18.8	26.3	Present work	
Gravel	Egypt (central region)	24	10.0	1.8	50.4	16
	Brazil	1	10.3	12.6	933.0	19
	Jordan	8	68.1	12.8	34.3	13

	Austria	6	13.9	14.8	171.0	20
	Pakistan	-	24.8	9.9	51.3	24
	Cuba	-	20.2	12.7	98.0	25
	USA (San Francisco)	4	44.3	141.0	25.9	26
	Nether Land	4	9.7	12.6	140.0	27
	Palestine	2	97.3	26.4	3.2	Present work
Gypsum	Egypt (central region)	10	3.1	0.7	7.5	16
	U.K	-	22.0	7.0	141.0	18
	Brazil	1	6.3	-	154.0	19
	Germany	23	<18.0	<10.0	96.0	18
	Austria	16	47.8	5.4	151.0	20
	USA (San Francisco)	12	37.0	18.5	7.4	26
	Nether Land	4	6.5	6.0	28.0	27
	Palestine	2	78.2	9.8	2.7	Present work

Figures (3), (4), and (5) show a comparison between different building materials for concentration values for ^{238}U , ^{232}Th and ^{40}K respectively.

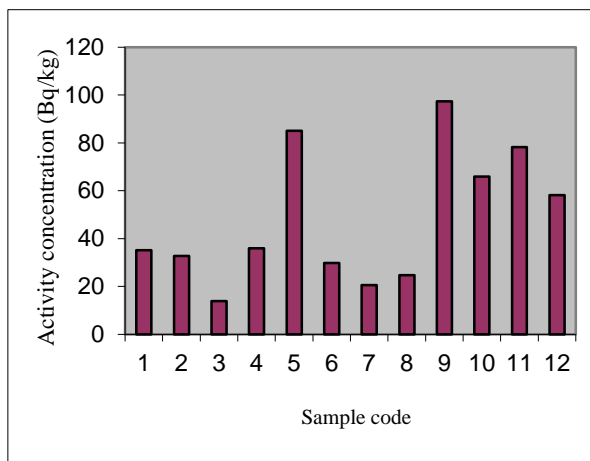


Fig. (3): A comparison between different building materials concentration values for U-238 series

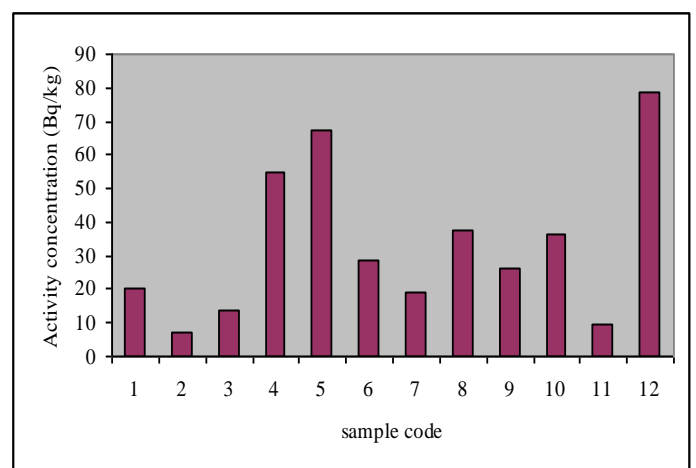


Fig. (4): A comparison between different building materials concentration values for Th-232 series

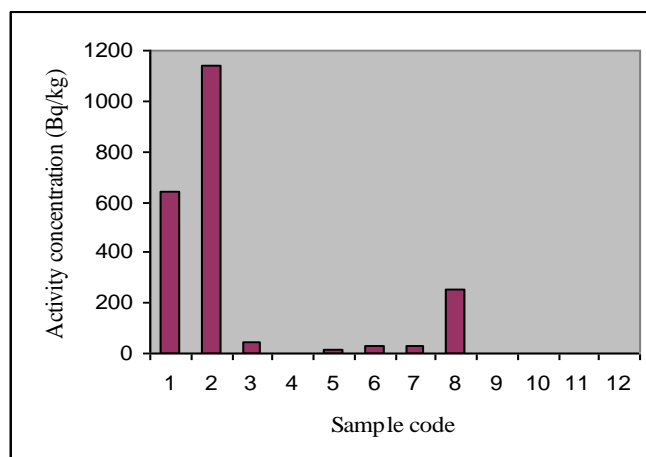


Fig. (5): A comparison between different building materials concentration values for K-40 isotope

Table (4), summarizes the radium equivalent (Ra_{eq}) for samples under investigation. Figure (6) is the Ra_{eq} results in all samples. From these results it can be noticed that the lowest value of Ra_{eq} is 37.3 Bq/Kg calculated in limestone, while the highest value is 183.3 and 170.8 Bq/kg calculated in cement and gravel respectively. The high values can be rendered to the high concentration of two radionuclide ^{238}U and ^{232}Th in these materials.

Table (4): The dose rate in air, the average radium equivalent, the external hazard index, the level of radiation hazard index and the annual gonadal equivalent dose for different building material samples

Sample code	Dose rate (10^{-8} Gy/hr)	Ra_{eq} (Bq/kg)	H_{ex}	I_{γ}	D ($\mu\text{Sv/yr}$)
1	63.9	113.7	0.31	0.87	394.7
2	61.9	130.8	0.35	1.05	489.1
3	7.4	37.3	0.10	0.26	115.5
4	11.0	114.4	0.31	0.79	340.5
5	19.3	183.3	0.50	1.26	551.4
6	9.7	73.6	0.20	0.51	222.8
7	7.0	49.5	0.13	0.34	224.8
8	28.2	98.0	0.26	0.71	313.2
9	13.8	135.3	0.37	0.92	412.1
10	11.8	117.9	0.32	0.81	355.7
11	9.6	92.4	0.25	0.62	283.4
12	16.5	170.8	0.46	1.18	508.7
average	21.7	109.8	0.30	0.69	351.0

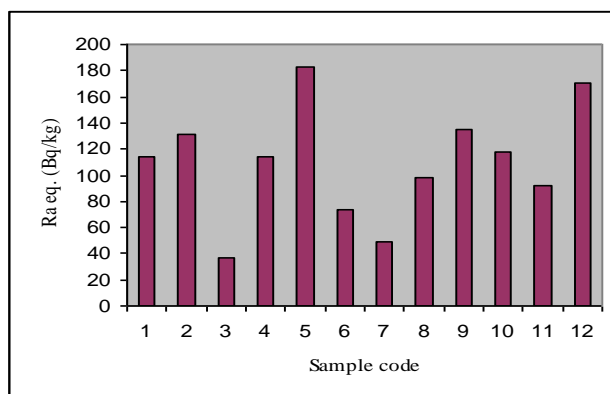


Fig. (6): The Ra_{eq} values of different building materials samples

The dose rate in air, the external hazard index, the radioactivity level index and the annual gonadal equivalent dose for the samples under investigation are represented in table (4), from the results, it can be noticed that granite and clay brick produce higher dose rate in air, results which in reasonable accord with other published data^(1,11). Some of the building materials caused excessive radiation doses to the total body due to the γ -ray emitted by the progeny of ^{226}Ra , ^{232}Th chains and ^{40}K also contribute to the total body radiation dose⁽¹¹⁾

According to ICRP ⁽¹⁵⁾, the upper limit of the external hazard index arising from building materials to the world general population can be exposed is 1.5 mGy/yr. The values for all studied samples range between 0.10 and 0.50 which are far under the criterion limit and less than unity ($H_{ex}<1$).

The level index I_γ is determined to estimate the γ -radiation hazard associated with the natural radionuclide in the used building material samples. The calculated values for most samples were less than unity ($I_\gamma<1$) and the values obtained for three samples (clay brick, cement and hydrated lime) were higher than the international levels ($I_\gamma>1$). The samples percipient in the radioactivity for first enhanced level⁽¹⁾.

The average AGED value is 351 μ Sv/yr and is slightly higher than the world average values (298 μ Sv/yr) ⁽¹³⁾. Results presented in table (4) indicate that the commonly used building materials which are examined in this work, could be used in building construction are exceeding the proposed radioactivity criteria level. In fact the ratio of AGED values obtained in this study compared to the world average values is 118%.

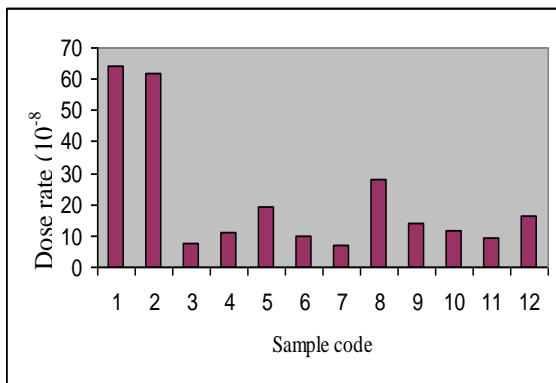


Fig. (7): The dose rate values of different building materials samples

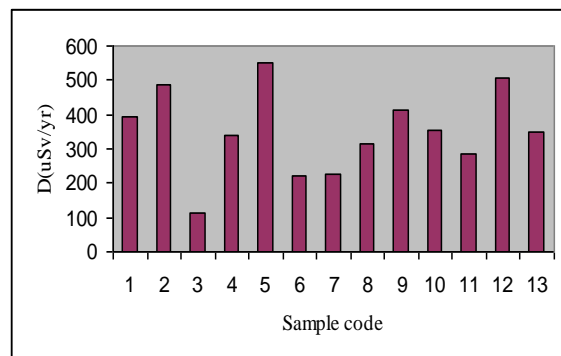


Fig. (8): The annual gonadal equivalent dose values of different building materials samples

Conclusion

Measurements of radioactivity levels of uranium, thorium and their radioactive decay series and ⁴⁰ K as well as ¹³⁷Cs in different fabricated building materials used in Palestine showed different conclusions as follow:

1. Gravel, hydrated lime and clay brick give the maximum levels of ²²⁶ Ra, ²³² Th and ⁴⁰ K respectively, among the other investigated building materials.
2. Maximum radium equivalent activity (Ra_{eq}) where found in cement and hydrated lime respectively.
3. Maximum values of the dose rate in air (10⁻⁸Gy/hr) were measured in granite and clay brick respectively.
4. Cement, hydrated lime and clay brick have radioactivity level above the proposed acceptable level ($I_\gamma>1$). These samples percipient in the radioactivity for first enhanced level.

5. ^{137}Cs radionuclide was detected in low concentration in some samples, and not found in others. The presence of fallout of cesium in some environmental regions increases the γ - radiation dose rate in materials of that region.
6. The average of calculated AGED was found to be 351.0 $\mu\text{Sv/yr}$. The average value obtained in this study was greater than the world average value (298 $\mu\text{Sv/yr}$).

Finally, we conclude that, the building materials were the major sources contributing to the total annual effective dose equivalent of γ -radiation for region populations in Palestine territories.

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