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Assessment of Natural Radioactivity and Radiological Hazards in Ceramic Samples Importer for the Local Market in Benghazi-Libya

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Abstract

The natural radioactivity of 15 ceramic samples imported from Italy, China, and Egypt to the local market of Benghazi city-Libya, was measured by the means of a gamma-ray spectroscopy system with an HPG detector. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were ranged from 75.75 to 85.63 Bq.kg^{-1}, 44.10to 65.20 Bq.kg^{-1} and 819.52 to 959.27 Bq.kg^{-1}, respectively. The radium equivalent, external hazard index, gamma radiation index, absorbed dose rate, annual effective dose, and cancer risk factor were calculated using the above measurements in order to assess the radiological hazard associated with the studied ceramic samples. The average values of these radiological indices for the ceramic samples were found to be; 232.29 Bq.kg^{-1}, 0.63, 1.71, 107.11 nGy.h^{-1}, 0.13 mSv.y^{-1} and 0.43, respectively. In general, the average value of radium equivalent for ceramic samples is not exceeded the recommended value of 370 Bq.kg-1, the external hazard index is lower than unity, and for all samples the annual effective dose below the recommended world limits. On the other hand, some radiological hazard indices were reported to be higher than the recommended world limits. The ceramic samples concerned in this study might be recommended to be utilized as building materials for dwelling construction.

Keywords: Natural radioactivity; Radiological hazard; Absorbed dose rate; HPGe detector; Ceramic

Introduction

Ceramic is commonly used in most modern buildings for the interior decoration of walls and floors. The largest contribution to the global population exposure to radiation arises from especially radionuclides from natural sources, present in the terrestrial crust. The materials use of containing naturally radionuclides in building construction may enhance the natural radiation background [1]. External exposure results are from gamma-emitting radionuclides existing in the walls, floor, and ceiling of buildings [2]. The presence of natural radionuclides in building materials may lead to an increasing in radiation exposure of the inhabitants of dwellings built with such materials [3,4]. It has been found that exposure to the radiation from building materials is about 80% of the radiation dose that a person receives within a year [5,6]. Building materials are normally extracted from rocks, sand, and soil that contain varying levels of radionuclides depending on the raw materials from which they are derived [7]. The activities of ^{226}Ra, ^{232}Th and ^{40}K in the building raw materials and their products mainly depend on geological, geochemical characteristics, and geographical conditions of those materials [8]. The quantitative determination of the level of concentrations of radioactive isotopes emitted from ceramic as a building material is essential to estimate the related radiologic hazards to human health and to build up reference information of radiologic hazard in ceramic imported for the local market in Benghazi city-Libya.

Materials and Methods

Samplings

Ceramic is one of the most important building materials and has been collected from building materials stores in Benghazi city-Libya. Three main types that were selected to be radiologically investigated are those that are common in the local market (imported from Italy, China, and Egypt). Totally 15 samples were collected, crushed, grinded, and oven-dried at *105* C^o (sample preparation process has been done at Arabian Gulf Oil Company laboratories). The stability and equilibrium of the radionuclides was the second stage of the sample preparation; the samples were sieved, weighed, and packed in Marinelli beakers (the volume of beakers is one liter). Samples were carefully sealed and stored for a month to reach secular equilibrium between ^{226}Ra and its progenies before starting the measurement process.

Measurements

The activity concentrations of natural radioactivity (^{226}Ra, ^{232}Th, and ^{40}K) in ceramic samples were measured by using high purity germanium HPGe detector (Canberra model EGPC-30185z2022 and digital multichannel analyzer multiport MCRAD-5008). To ensure a low background environment, the HPGe detector was enclosed within a *10 cm* lead shield coated internally with a *2 mm* copper layer. The background radioactivity was determined by utilizing an empty container with the same geometry as that used for the samples; this was sealed and saved for a month before determining the background measurement. After performing a correction for the background spectra, the activity concentrations of natural radioactivity in the samples (measured in $Bq.kg^{-1}$) were calculated based on the count spectra of each sample using the gamma-ray photon peaks. The ^{226}Ra activity concentration was calculated indirectly using the gamma-ray peak values of its Radon daughters: ^{214}Pb (351.99 keV) and ^{214}Bi (609.72 keV). The gamma-ray peaks of ^{228}Ac (911.07 keV) were used to determine the activity concentration of ^{232}Th. The ^{40}K activity concentration was measured from its energy photopeak of high probability (1460.83 keV). The activity concentration A measured in $Bq.kg^{-1}$ for the natural radionuclides in the measured samples was calculated using the formula [9]:

$$A = \frac{N}{\varepsilon\, I_\gamma t\, m}$$

In the above expression, N is the corrected net photo-peak area at energy peak, ϵ is the absolute efficiency at photopeak energy, and t is the time of the sample spectrum collection in seconds. In this context I_γ is the gamma-ray emission probability corresponding to the peak energy and *m* is the mass (kg) of the measured sample.

Calculation of radiological hazards

Radium equivalent activity ($Ra_{eq.}$)

Radium equivalent activity $Ra_{eq.}$ is the weighted sum of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K based on the assumption that 370 Bq.kg^{-1} of ^{226}Ra, 259 Bq.kg^{-1} of ^{232}Th and 4810 Bq.kg^{-1} of ^{40}K produce the same gamma radiation dose rates [10]. Radium equivalent activity $Ra_{eq.}$ is calculated using the equation [11]:

$$Ra_{eq.} = A_{Ra} + 1.43\, A_{Th} + 0.077\, A_K$$

where A_{Ra}, A_{Th} and A_K are the specific activity concentrations (Bq.kg^{-1}) of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

External hazard index (H_{ex})

The external hazard index H_{ex} is an important factor used to estimate the radiation dose from building materials in dwellings. The external hazard index H_{ex} is calculated via the following equation [12]:

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

With A_{Ra}, A_{Th} and A_K being the activity concentrations for ^{226}Ra,^{232}Th, and ^{40}K in Bq.kg^{-1}.

Gamma Index (I_γ)

The representative level index (I_γ) is calculated to assess the level of gamma radiation hazards related to the natural radionuclides with the accord of the following:

$$I_\gamma = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500}$$

Here, once again, A_{Ra}, A_{Th} and A_K are the activity concentrations for ^{226}Ra,^{232}Th, and ^{40}K in Bq.kg^{-1} [13,14].

Absorbed dose rate (D)

The absorbed dose rate D in the air at one meter above the ground due to ^{226}Ra, ^{232}Th, and ^{40}K can be estimated by the formula [15]:

$$D\ (n\ Gy.h^{-1}) = 0.427\, A_{Ra} + 0.662\, A_{Th} + 0.043\, A_K$$

with the same above-mentioned definition of A_{Ra}, A_{Th} and A_K.

The annual effective dose (E_{out})

The annual effective dose equivalent received by humans is estimated from the dose rate D, occupancy factor which is defined as the level of human occupancy factor and has taken to be 20% (0.2) of 8760 hours in a year, and the conversion factor of 0.7 $Sv.Gy^{-1}$ is used to convert the absorbed dose in the air to the effective dose. The annual effective dose equivalent is estimated by [16]:

$$E_{out}(mSv.y^{-1}) = D(\text{nGy}.h^{-1}) \times 8760 \times 0.2 \times 0.7 \times 10^{-6}$$

Excess Lifetime cancer risk ($ELCR$)

Excess Lifetime cancer risk ($ELCR$) was calculated by using the following relation [17]:

$$ELCR = E_{out} \times D_L \times R_F$$

where E_{out} is the above-mentioned annual effective dose Equivalent, D_L is defined as the average duration of life (approximately 66 years) and R_F is the risk factor (Sv^{-1}) [17]. For stochastic effects, ICRP 60 uses values of 0.05 for the public [18].

Results and Discussion

In this study, the activity concentrations of radionuclide were estimated by means of gamma-ray spectrometry technique (HPGe detector) and estimation of the gamma dose rate from these radionuclides. The obtained average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for each of the measured samples are summarized in Table 1. On the other hand, Figure 1, shows a comparison between the activity concentrations in $Bq.kg^{-1}$ of the interested radionuclides for the all-ceramic samples under investigation.

Table 1: The activity concentrations ($Bq.kg^{-1}$) of the radionuclides (^{226}Ra, ^{232}Th and ^{40}K) of the investigated sample.

Country	Ra_{eq}	^{226}Ra	^{232}Th	^{40}K
Italy	I1	87.23	64.19	954.63
	I2	89.98	65.20	951.32
	I3	88.17	64.17	956.99
	I4	86.01	63.55	959.27
	I5	84.85	61.43	956.45
China	C1	96.43	43.42	866.21
	C2	96.73	47.83	869.32
	C3	95.43	45.74	866.12
	C4	95.98	46.22	867.25
	C5	94.63	44.10	866.77
Egypt	E1	74.17	52.97	819.52
	E2	78.2	53.1	820.63
	E3	76.14	52.66	821.47
	E4	75.75	54.71	823.09
	E5	79.73	56.62	824.24
	Avg.	86.63	54.39	881.55
	Min.	75.75	44.10	819.52
	Max.	96.73	65.20	959.27
	P.L	50	50	500

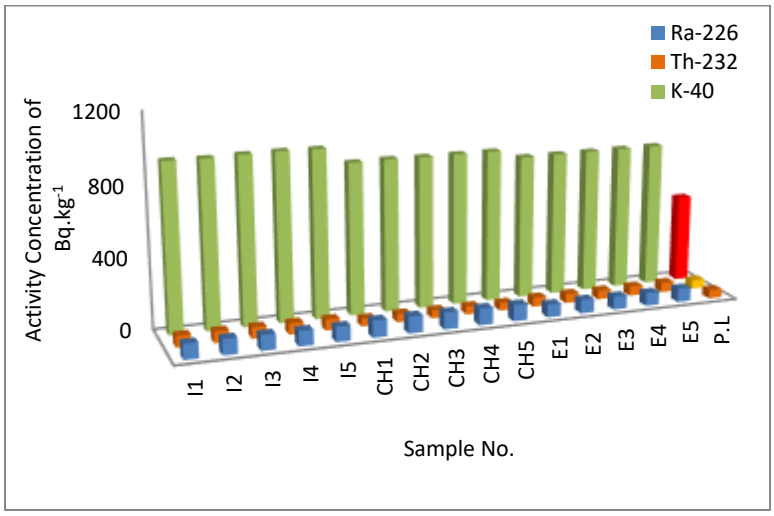

Figure 1: The activity concentrations of ceramic samples.

From the obtained results, it is obvious to recognize that, the values of activity concentrations in the studied ceramic samples varied from 75.75 to 96.73, 44.10 to 65.20 and from 819.52 to 959.27 Bqkg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K respectively. The results reveal that the values of activity concentrations of radionuclide for all-ceramic samples are higher than the recommended limit for building materials 50, 50, 500 Bqkg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K respectively [16].

The present work focused on a comprehensive quantitative evaluation of radiological hazards of ceramic samples, which used as building materials and collected from building materials stores in Benghazi city-Libya. The radium equivalent, external hazard, Gamma radiation index, absorbed dose rate, annual effective dose and cancer risk factor were calculated and listed in Tables 3 and 4. From Table 3, it may report that the obtained values of Ra_{eq} ranged from 213.02 Bq.kg^{-1} to 256.47 Bq.kg^{-1}. The average value of Ra_{eq} was 232.29 Bq.kg^{-1}. The obtained values were found to be less than the recommended value of 370 Bq.kg^{-1} [16], for all-ceramic samples. As shown in Table 3, the maximum 0.57 and minimum 0.69 values of H_{ex} have an average value amount of 0.63. Moreover, all values of H_{ex} for the ceramic samples are lower than unity. The obtained gamma index ranged from 1.57 to 1.89. The average value of the gamma radiation index was 1.71, the obtained values for all the measured samples exceed the recommended value of $I_\gamma \geq 1$ *[16]*.

The estimated absorbed dose rate *D* values for the investigated ceramic samples range from 98.24 to 117.96 nGy.h^{-1}, but the average value of the absorbed dose rate for the ceramic samples was 107.11 nGy.h^{-1}. The absorbed dose rate values of the studied samples are higher than the world absorbed dose rate of 84 nGy.h^{-1} [16] The obtained values for the annual effective dose *E(out)* are values vary from 0.12 to 0.14 mSv.y^{-1} with an average value of 0.13 mSv.y^{-1}. This average value is lower than the 1 mSv.y^{-1} limit set by the European Commission [14]. The recorded values of excess lifetime cancer risk ELCR range from 0.40 to 0.48 with an average of 0.43, the results showed that all values of ELCR for ceramic samples are higher than the recommended value of 0.29 x 10^{-3} [16].

Table 2: The value of radium equivalent, external hazard index, external γ–radiation for ceramic samples.

Samples	Hazard index		
	Ra_{eq}	H_{ex}	I_{γ}
I1	252.53	0.68	1.86
I2	256.47	0.69	1.89
I3	253.62	0.68	1.87
I4	250.75	0.68	1.85
I5	246.34	0.66	1.82
C1	225.22	0.61	1.65
C2	232.06	0.63	1.70
C3	227.53	0.61	1.67
C4	228.85	0.62	1.68
C5	224.43	0.61	1.65
E1	213.02	0.57	1.57
E2	217.32	0.59	1.60
E3	214.70	0.58	1.58
E4	217.36	0.59	1.60
E5	224.16	0.60	1.65
Avg.	232.29	0.63	1.71
Min	213.02	0.57	1.57
Max	256.47	0.69	1.89
P.L	370	1	1

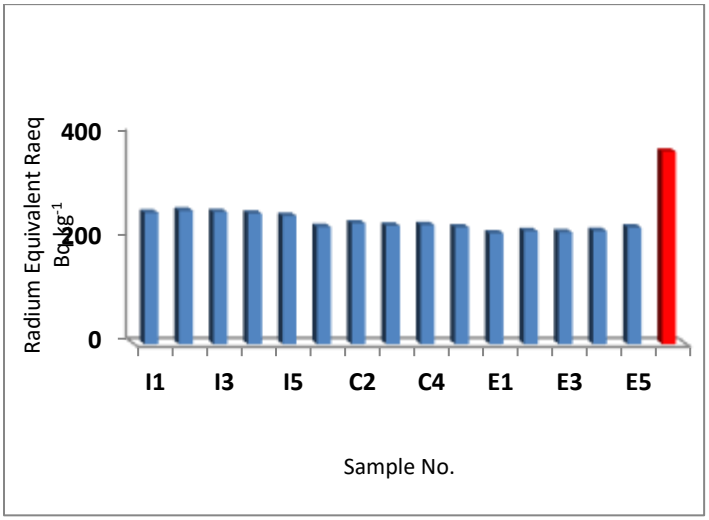

Figure 2: Radium Equivalent of ceramic samples.

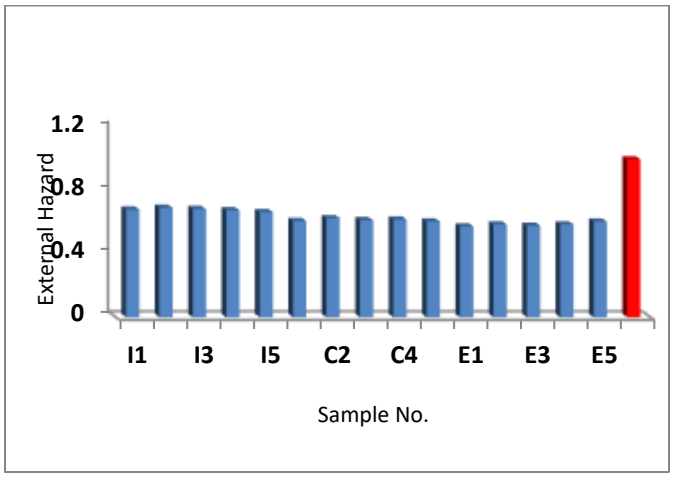

Figure 3: External Hazard of ceramic samples.

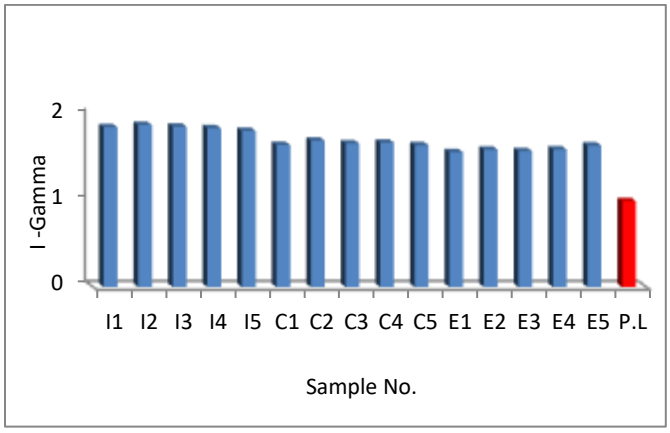

Figure 4: I-Gamma values of the ceramic samples.

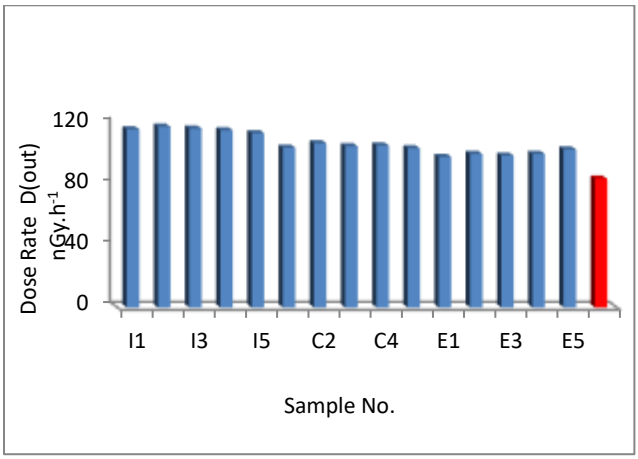

Figure 5: The absorbed dose of ceramic samples.

Table 3: The value of absorbed dose rate, annual outdoor effective dose and cancer risk factor for ceramic samples.			
Sample No.	Dose rate D_{out} (nGy.h^{-1})	Annual effective dose E_{out} (mSv.y^{-1})	Cancer risk factor $ELCR_{OUT} \times 10^{-3}$
I1	116.29	0.14	0.47
I2	117.96	0.14	0.48
I3	116.79	0.14	0.47
I4	115.56	0.14	0.47
I5	113.67	0.14	0.46
C1	104.17	0.13	0.42
C2	107.07	0.13	0.43
C3	105.12	0.13	0.42
C4	105.70	0.13	0.43
C5	103.82	0.13	0.42
E1	98.24	0.12	0.40
E2	100.12	0.12	0.40
E3	98.99	0.12	0.40
E4	100.12	0.12	0.4
E5	103.05	0.13	0.42
Avg.	107.11	0.13	0.43
Min	98.24	0.12	0.40
Max	117.96	0.14	0.48
P.L	84	1	0.29

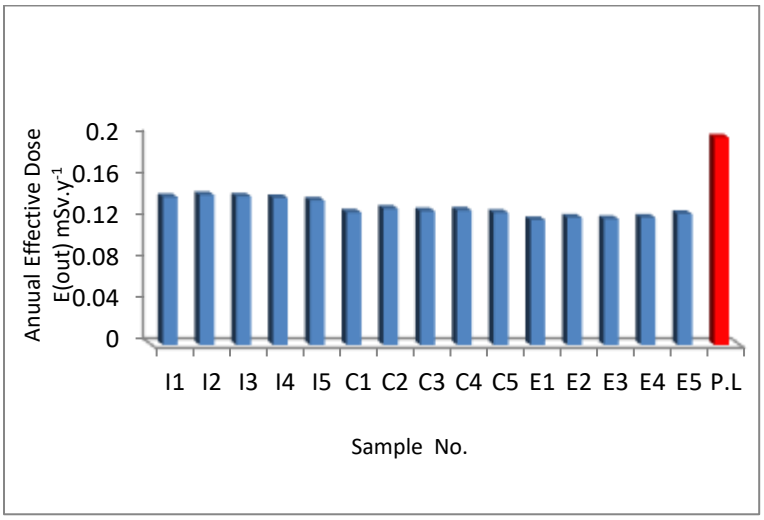

Figure 6: The annual effective dose of the ceramic samples.

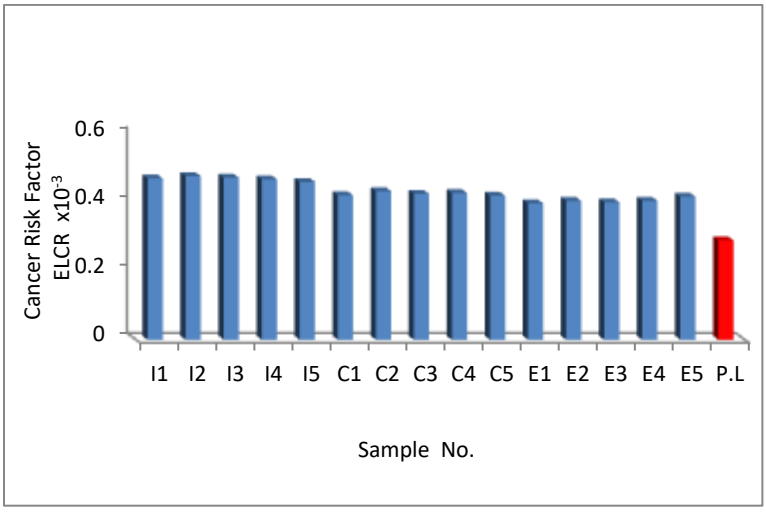

Figure 7: The cancer risk factor of the measured ceramic samples.

Conclusion

A typical high-resolution HPGe gamma-ray spectrometry system was utilized to measure the activity concentrations of radionuclide ^{226}Ra, ^{232}Th and ^{40}K. The average activity concentrations of radionuclides in this study were found to be higher than the recommended values. In addition, the measured activity concentrations were also manipulated to estimate some radiological parameters in order to qualify the associated radiological hazard from the samples at hand. The radium equivalent activities of the studied ceramic samples were below the criterion limit of 370 Bq·kg^{-1}. The values of external hazard indices for all investigated samples were found to be lower than unity. The calculated average annual effective dose equivalents from the outdoor terrestrial gamma radiation for ceramic were showed to be lower than recommended safety limit. In contrast, the absorbed dose rate, gamma radiation index and cancer risk factor are higher than recommended limit. The data reported herein can be used to enlarge the database on natural

radioactivity in ceramic used as building materials and to support technical aspects in hazard exposure reduction. In conclusion, we recommend and motivate more comprehensive researchers to include all-ceramic materials that available in the local market. This may fairly contribute to quality evaluation of the studied samples from the radiation hazards perspective.

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