



Natural Radioactivity in Different Commercial Ceramic Samples Used in Palestinian Buildings as Construction Materials

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Abstract:

This work presents the radioactivity concentrations of ^{226}Ra , ^{232}Th and ^{40}K for 36 commercial ceramic samples collected from 12 different companies and used in the construction of Palestinian buildings. The studied samples were analyzed and the concentrations of radioisotopes were determined by γ -ray spectrometry using hyper-pure germanium (HPGe) detector in Bq/kg dry-weight. Detected average values of concentrations of ^{226}Ra , ^{232}Th and ^{40}K were 73.7 , 58.2 and 624.0 Bq/kg respectively. The different radiation indices which indicate hazardous radiation were also determined. The results showed that the average radium equivalent (Ra_{eq}), the absorbed dose rate (D_r), the annual effective dose equivalent (AEDE), the external hazard index (H_{ex}), and the radioactivity level index (I_γ) were: 205.2 Bq/kg, 97 nGy/hr, 0.97 mSv/yr, 0.55 and 1.49 respectively. The mean values of Ra_{eq} , AEDE and H_{ex} obtained in this study were in good agreement with those of the international values while the mean values of D_r and I_γ were higher than the international values. The measured activity concentrations for these radionuclide were compared with the reported data obtained from similar materials used in other countries and with typical world values.

Key words: Natural radioactivity, building materials, ceramic, γ -ray spectrometry

دراسة النشاط الإشعاعي الطبيعي لعينات من السيراميك التجاري والمستخدم كمواد بناء في البنايات الفلسطينية

الملخص:

لقد تم في هذا البحث دراسة تركيز النويدات ذات النشاط الإشعاعي الطبيعي لعناصر الراديوم-226، الثوريوم-232، ونظير البوتاسيوم-40، لست وثلاثين عينة تجارية من الخزف (السيراميك) ، جمعت من إنتاج 12 شركة عالمية تستخدم كمواد بناء في المنازل الفلسطينية. استخدم جهاز كاشف الجرمانيوم عالي النقاوة بهدف التعرف على تركيز الانوية المشعة لتلك العينات بوحدات بيكريل / كغم. لقد وجد أن متوسط تركيز هذه النويدات: 73.7، 58.2 و 624 بيكريل / كغم، للراديوم - 226، للثوريوم - 232 وللپوتاسيوم - 40 على التوالي. لقد تم تقدير الأضرار الإشعاعية الناجمة عن النشاط الإشعاعي الكلي، وذلك بحساب بعض المعاملات الإشعاعية المختلفة. أظهرت النتائج أن معدلات نشاط الراديوم المكافئ (Ra_{eq})، معدل الجرعة الممتصة (D_p)، مكافئ الجرعة الفعالة السنوي (AEDE) معامل الاخطار الخارجي (H_{ex}) ومعامل مستوى الإشعاع ($I\gamma$) هو: 205.2 بيكريل / كغم، 97 نانوجراي / ساعة، 0.97 ملي سيفرت / سنة، 0.55 و 1.49 على التوالي. وبإجراء مقارنه علميه وجدنا أن متوسط القياسات لنشاط الراديوم المكافئ، مكافئ الجرعة السنوي ومعامل الأخطار الخارجي هو أقل من المستوى المتوسط بينما معدل الجرعة الممتصة ومعامل مستوى الإشعاع فإنها مرتفعة عن الحدود العالمية. وأخيرا فقد تم مقارنة تركيز النويدات الطبيعية الناتجة مع دراسات أخرى في العديد من دول العالم.

Introduction:

Radioactive elements are generally classified into two types: Naturally occurring and artificially produced radioactive elements. Naturally occurring radioactive materials (NORM) are found in building materials such as cement, granite, ceramic, stones ...etc. They are the main components of external and internal radiation exposure in dwellings. The external radiation exposure by gamma radiation originated from the members of uranium and thorium decay chains and from potassium-40. The internal exposure affecting the respiratory tract, is due to radon and its daughters which are exhaled from building

materials (Zalewski *et al.* , 2001; Kathren, 1998). The study of the radioactive components in building materials (e.g. ceramic) is a fundamental link to understand the behavior of radioactivity in the ecosystem because these materials emit radiation by the disintegration of natural radionuclides and contribute to the total absorbed dose via ingestion, inhalation and external irradiation (Kathren, 1998; Chowdhury *et al.*, 2005). The level of contribution to the background radiation depends on the amount of the radioactive elements in the building materials, but this amount or concentration varies

with the type and the components of the material. Investigations had shown that ^{226}Ra , ^{232}Th and ^{40}K were present in building materials in varied concentrations. Generally, dose contributions from building materials to the doses in dwellings are small compared to those from underlying bed rock and soil (Al-Jundi *et al.*, 2005).

The level of natural background radiation is generally higher in buildings (population-weighted world average 84 nGy/hr) than outdoors (population-weighted world average 59 nGy/hr) due to the NORM content of buildings materials (UNSCEAR, 2000; Risica and Nuccetelli, 2001). The worldwide average annual effective dose from natural sources is estimated to be 2.4 mSv of which ~1.1 mSv is due to the basic background radiation and 1.3 mSv is due to exposure to radon (UNSCEAR, 1988).

The content of natural radionuclides in building materials is caused by many factors: geological origin and composition of soil, its density and porosity, content of water in soil, diffusion rate and permeability rate, rate of emanation and exhalation, etc. Therefore, there is significance in the effective dose equivalent in closed space due to the type of building material and construction of the building object (Popovic and Todorovic, 2006).

The aim of this study is to determine the activity of the naturally occurring radionuclides and the γ -ray absorbed dose in the fabricated foreign imported ceramic tile samples used in the Palestinian buildings in order to establish the radiation background database.

Ceramic materials play very important role in our lives, because they are used in most of our houses as building materials. Ceramic consists of earthy materials like limestone, feldspar, glaze, clay and sand glass, which are normally used in the ceramic industry and final commercial products (El-Shershaby *et al.*, 2004). Ceramic materials are non metallic, inorganic compounds, primary oxides, also carbides, nitrides and silicides (Anderson, 1995).

In this study, the final commercial products of ceramic used in decoration of large areas in Palestinian houses and buildings from different companies and factories were analyzed to investigate the radioactivity content. For this purpose, plates of ceramic samples were grinded mechanically and homogenized into powder material for investigation.

Materials and Techniques

Thirty six commercial foreign samples from 12 different ceramic tile manufacturing companies used in Palestinian buildings were used for radioactivity measurement. The companies are: Cleopatra (SC_1)-Egyptian; Prima (SC_2)-Egyptian; Venus (SC_3)-Spanish; Halcon (SC_4)-Spanish; Caya(SC_5)-Spanish; Atlas Concord (SC_6)-Italian; Piemme (SC_7)- Italian; Aspendos (SC_8)- Turkish; Toprak (SC_9)- Turkish; Lung(SC_{10})-Chinese; MGreez (SC_{11})-Chinese; Negev (SC_{12})-Israeli. Collected samples were air-dried, dry-weighted, sieved through a fine mesh (< 2 mm), hermetically sealed in standard 1000

ml plastic Marinelli beakers for about four weeks in order to establish secular equilibrium between ^{226}Ra , ^{232}Th with their progeny. After the holding period, the samples were measured with an n-type coaxial lead shielded intrinsic Ortec high-purity germanium (HPGe) detector with an efficiency of 15% and an energy resolution of 1.85 KeV, full width to the half maximum (FWHM) for the 1333 KeV gamma line of ^{60}Co (Chiozzi *et al.*, 2002; Tzortiz *et al.*, 2003). The relative efficiency calibration of the spectrometer was carried out using the following standard sources: ^{241}Am (60 KeV), ^{109}Cd (88 KeV), ^{57}Co (122 KeV), ^{60}Co (1173 and 1333 KeV), ^{139}Ce (166 KeV), ^{203}Hg (279 KeV), ^{113}Sn (392 KeV), ^{85}Sr (514 KeV) ^{137}Cs (662 KeV) and ^{88}Y (898 and 1836 KeV). The calibration efficiency curve beyond 1850 KeV was constructed using different energy peaks of Ra-226 in order to cover the range from 60 to 2500 KeV (Helmer, 1982). The standard source packed in Marinelli beaker has the same geometry as that used for measured samples. Absolute efficiency calibrations were calculated to study isotopes by using the last standard source and chemically pure KCL dissolved in distilled water at different concentrations (El-Tahawy *et al.*, 1992). An empty bottle with the same geometry was taken for subtracting the background. The measuring time of both activity and background measurements ranged between 12 to 20 hours. The background spectra were used to correct the net γ -rays peaks areas for studies isotopes. The γ -ray transitions used to measure the concentration of

the assigned nuclides in the series are: ^{214}Pb (352 KeV), ^{226}Ra (186 KeV) and ^{214}Bi (609, 1120 and 1765 KeV) for uranium series; ^{228}Ac (338.5 and 911 KeV), ^{208}Tl (583 and 860 KeV) and ^{212}Bi (727 and 1621 KeV) for thorium series and ^{40}K (1461 KeV) for potassium isotope (Chowdhury *et al.*, 2005; EML, 1990).

Results and Discussion

a) Radioactivity Determination.

The radioactivity concentrations of ^{226}Ra , ^{232}Th and ^{40}K for 36 commercial ceramic samples collected from 12 different foreign companies were obtained. The concentration for all samples and the average activities are presented in Table (1).

Table 1: The activity concentrations in (Bq/kg) of commercial ceramic samples used in Palestinian buildings

comp. code	Activity concentration (Bq/kg)											
	^{226}Ra				^{232}Th				^{40}K			
	S ₁	S ₂	S ₃	mean	S ₁	S ₂	S ₃	mean	S ₁	S ₂	S ₃	mean
SC ₁	82.3	90.2	85.6	86.0	71.0	66.2	74.4	70.5	720.7	830.5	767.0	772.7
SC ₂	67.2	56.8	65.3	63.1	46.0	46.1	56.7	49.6	630.1	650.0	613.2	631.1
SC ₃	44.0	48.6	43.5	45.4	44.0	40.8	31.6	38.8	481.7	511.2	522.6	505.2
SC ₄	102.6	107.9	95.6	102.0	61.2	66.7	73.0	67.0	830.0	820.0	808.0	819.3
SC ₅	84.1	79.3	76.9	80.1	81.1	70.3	59.0	70.1	622.3	757.0	714.7	698.0
SC ₆	74.7	78.5	77.2	76.8	56.0	59.6	63.4	59.7	563.0	597.0	580.0	580.0
SC ₇	50.9	56.2	61.0	56.0	44.9	39.4	39.0	41.1	443.0	511.6	470.0	474.9
SC ₈	80.2	51.3	69.5	67.0	55.3	59.7	66.0	60.3	378.8	422.3	451.7	417.6
SC ₉	62.6	58.4	56.7	59.2	40.2	43.3	39.6	41.0	380.0	370.7	338.3	363.0
SC ₁₀	82.1	90.0	77.3	83.1	66.4	81.3	59.6	69.1	798.0	861.2	825.9	828.4
SC ₁₁	69.1	66.3	73.5	69.6	61.2	30.8	68.0	53.3	601.0	541.3	572.0	571.1
SC ₁₂	88.6	101.0	99.8	96.5	76.0	78.1	80.7	78.3	840.7	903.0	870.0	871.2
Range	45.4-102				38.8-78.3				363.0-871.2			
Average	73.7				58.2				624.0			

From the results obtained, it can be seen that the average concentrations of the three radionuclides in the different ceramic samples are 73.7 Bq/kg, 58.2 Bq/kg and 624.0 Bq/kg for ^{226}Ra , ^{232}Th and ^{40}K respectively.

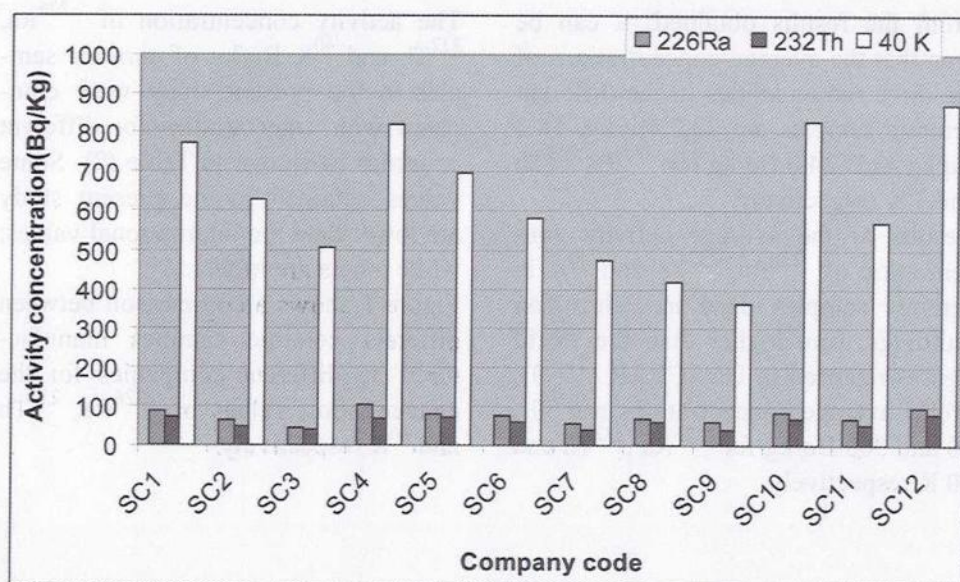
Results of the average activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in ceramic samples used in Palestinian buildings were higher than the world figures reported in UNSCEAR (1993). World average concentrations are 50, 50 and 500 Bq/kg for ^{226}Ra , ^{232}Th and ^{40}K respectively.

The activity concentration of ^{226}Ra , ^{232}Th and ^{40}K Bq/kg of ceramic samples in the present study were compared with other studies for different countries as shown in Table (2). Some values obtained in the present study are lower than the international values; while others are higher.

Figure 1 shows a comparison between different ceramic samples manufactured by different companies for the concentration values of ^{226}Ra , ^{232}Th and ^{40}K respectively.

Table 2: Comparison levels of radionuclides concentrations in ceramic samples (in Bq/kg) dry weight

Countries	Activity concentration (Bq/kg)						Ref
	^{226}Ra		^{232}Th		^{40}K		
	Range	Average	Range	Average	Range	Average	
China	63.5-131.4	--	55.4-106.5	--	386.7-866.8	--	Xinwei (2004)
Pakistan	63.1-123.9	83.4	--	--	144.1-834	403.5	Tufail <i>et al</i> (1992)
Algeria	--	55	--	41	--	410	Amarani & Tahtai2001
Egypt (Cleopatra Factory)	71.2-86	76.1	63.3-68.7	66.2	900-1018	962	Hilal (2002)
Egypt Lecico & El-Gawhra Factories	41.7-60.7	52.2	30.7-47.1	39.1	195-680	480	Higgy (1995)
Qena (Egypt)	40-230	126	10-130	72	80-600	300	Ahmed (1999)
Palestine	45.4-102.0	73.7	38.8-78.3	58.2	363-871.2	624	Present work
UNSCEAR	--	50	--	50	--	500	UNSCEAR (1993)

Figure 1: A comparison between different ceramic samples activity concentration values for ^{226}Ra , ^{232}Th and ^{40}K respectively

b) Assessment of Exposure Risk

1- Equivalent Radioactivity (Ra_{eq}):

The radium equivalent activity (Ra_{eq}) is a weighted sum of activities of ^{226}Ra , ^{232}Th and ^{40}K based on the assumption that 10 Bq/kg of ^{226}Ra , 7 Bq/kg of ^{232}Th and 130 Bq/kg of ^{40}K produce the same γ -ray dose rates. The equivalent radioactivity is generally defined as (Berigido *et al.*, 2005).

$$Ra_{eq} (\text{Bq/kg}) = C_{Ra} + 1.43 C_{Th} + 0.077 + C_K \dots (1)$$

where C_{Ra} , C_{Th} and C_K are activities of ^{226}Ra , ^{232}Th and ^{40}K respectively in Bq/kg. The maximum value of Ra_{eq} must be < 370 Bq/kg in order to keep the external dose < 1.5 mGy/hr (Beretka and Mathew, 1985). The Ra_{eq} values are presented in Table (3). The measurements in these ceramic samples range from 139.8 Bq/kg to 275.6 Bq/kg with an average value of 205.2 Bq/kg, which is less than the safe limit (370 Bq/kg).

2- The Absorbed Dose Rate

The absorbed dose rate in air (D_r) in nGy/hr, resulting from the natural specific activity concentration of ^{226}Ra , ^{232}Th and ^{40}K (C_{Ra} , C_{Th} and C_K) in Bq/kg at a height of 1m above the ground was calculated by the following equation (UNSCEAR, 1998).

$$D_r (\text{nG/hr}) = 0.427 C_{Ra} + 0.662 C_{Th} + 0.0432 C_K \dots (2)$$

The values obtained range between 66.9 nGy/hr and 130.6 nGy/hr, with an average value of 97.0 nGy/hr (Table (3)). The calculated gamma dose rates in all ceramic samples are higher than the international recommended value 55 nGy/hr (UNSCEAR, 1998) by 176 %.

3-The Annual Effective Dose Equivalent.

The annual effective dose equivalent (AEDE) for the public or the worker in different samples in mSv/yr was calculated using the following equation (Malanca *et al.*, 1993).

$$AEDE (\text{mSv/yr}) = (0.49 C_{Ra} + 0.76 C_{Th} + 0.048 C_K) \times 8.76 \times 10^{-3} \dots (3)$$

The results showed that the values range between 0.67 mSv/yr and 1.3 mSv/yr with an average of 0.97 mSv/yr (Table (3)). The results of AEDE for SC_1 , SC_5 , SC_{10} , and SC_{12} companies are about 10 to 30 % higher than the results international values (1 mSv/yr) for the public; the rest is lower than the permissible value for the international values (Hilal, 2002).

4- The External Hazard Index.

The external hazard index H_{ex} is used to assess radiation risks attributed to radioactive materials. Generally speaking, H_{ex} is calculated using the following equation (Beretka and Mathew, 1995)

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \dots (4)$$

The value of H_{ex} must be lower than unity in order to keep the radiation hazard insignificant (Huy and Luyen, 2005). The maximum value of unity for H_{ex} corresponds to the limit of 370 Bq/kg for Ra_{eq} (Huy and Luyen, 2005). The calculated values of the H_{ex} for ceramic samples are given in Table (3). The H_{ex} values are found to range from 0.38 to 0.74 with an average value of 0.55. These values are lower than unity

in agreement with the international assigned values.

5- The Radioactivity Level Index.

This index can be used to estimate the level of γ -radiation hazards associated with the natural radionuclide. The representative level of radiation hazard index may be defined as (Beretka and Mathew, 1985).

$$I_{\gamma} = \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500} \dots\dots\dots(5)$$

The values of I_{γ} are calculated according to equation (5) are listed in Table (3). The calculated values for all samples were found to be higher than unity ($I_{\gamma} > 1$). These samples are perceptible in the radioactivity for first enhanced level (Higgy, 1995; UNSCEAR, 1998).

Table 3: Radiation indices of the commercial ceramic samples from different companies used in Palestinian buildings:

Company code	Ra _{eq} (Bq/kg)	D _r (nGy/hr)	AEDE (mSv/yr)	H _{ex}	I _γ
SC ₁	242.8	114.8	1.14	0.66	1.77
SC ₂	182.6	87.0	0.87	0.49	1.34
SC ₃	139.8	66.9	0.67	0.38	0.03
SC ₄	260.9	123.2	1.23	0.70	1.90
SC ₅	234.1	110.7	1.10	0.63	1.71
SC ₆	206.8	97.4	0.97	0.56	1.50
SC ₇	152.9	71.6	0.71	0.41	1.10
SC ₈	185.4	86.6	0.86	0.50	1.33
SC ₉	145.8	68.1	0.68	0.39	1.05
SC ₁₀	245.7	116.9	1.17	0.66	1.80
SC ₁₁	189.8	89.7	0.89	0.51	1.38
SC ₁₂	275.6	130.6	1.30	0.74	1.91
Range	139.8-275.6	66.9-130.6	0.67-1.30	0.38-0.74	1.03-1.91
Average	205.2	97.0	0.97	0.55	1.49

Conclusion

The activity concentrations of some commercial ceramic samples used in Palestinian buildings, which were collected from different companies, were measured by using γ -ray spectrometry. This is because the ceramic is widely used as a building material in a variety of dwellings and public buildings.

The radioactivity means (in Bq/kg) of ^{226}Ra , ^{232}Th and ^{40}K for these types with values of 73.7, 58.2 and 624.0 are higher than the world figures reported in UNSCEAR (1993). The activity concentration of these samples produced an average radium equivalent activity of 205.2 Bq/kg which is lower than the safe limit (370 Bq/kg).

The different radiation indices which indicate hazardous radiation were also determined. The results showed that the average D_r , AEDE, H_{ex} and I_γ are 97.0 nGy/hr, 0.97 mSv/yr, 0.55 and 1.49 respectively in ceramic samples. The mean values of AEDE, H_{ex} are in good agreement with those of the world and the mean value of D_r is higher than the international values of 55 nGy/hr. The calculated value of I_γ for all ceramic samples was found to be higher than unity ($I_\gamma > 1$). Thus the samples are percipience in the radioactivity as the first enhanced level.

Finally, we conclude that, the ceramic used as building materials is a source of radiation that contributes the total annual dose rate of γ -radiation in Palestinian dwellings.

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