JKAU: Sci., **Vol. 23** No. 1, pp: 39-50 (2011 A.D./1432 A.H.)

DOI: 10.4197 / Sci. 23-1.4

Natural Radioactivity in Different Commercial Ceramic Samples Used in Yemeni Buildings

Sahar A. Amin*, Mustafa Naji

Department of Physics, Faculty of Science,
Taiz University, Taiz, Yemen.
Department of Physics, Faculty of Education,
Aden University, Aden, Yemen.

Abstract: In this work we calculated the radioactivity concentrations of the natural radioactive nuclides ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K for 10 commercial samples collected from 10 different companies which are used in the construction of Yemeni buildings.

Gamma ray spectroscopy is used to analyze the samples and the concentrations of radioisotopes were determined using hyper-pure germanium (HPGe) detector in Bq/kg dry-weight. The average concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K were found to be 131.4, 83.55, 131.88 and 400.7 Bq/kg respectively. Different hazard indices were also determined. The results showed that the average radium equivalent activity (Ra_{eq}), the absorbed dose rate (D_r), the annual effective dose equivalent (AEDE), the external hazard index (H_{ex}) and representation level index (I_γ) were: 307.52Bq/kg, 139.31nGy/hr, 1.40mSv/yr 0.83 and 2.15 respectively. The mean value of (Ra_{eq}) obtained in this study was in good agreement with that of the international value while the mean values of the other indices were found to be higher than the international reference values. The measured activity concentrations for these radionuclides were compared with the reported data obtained from similar materials used in other countries.

Keywords: Natural Radioactivity; Building Materials; Ceramic; Gamma Ray Spectrometry; Hazard Indices.

Introduction

Humans are exposed every day to natural radionuclides which are ^{232}Th, ^{226}Ra and ^{40}K. They pose external exposure risk by their gamma emission and internal exposure risk due to radon and its progeny, which originates from the three well known series Uranium (^{238}U), Thorium (^{232}Th) and Actinium (^{238}U). The contribution of ^{235}U to the environmental dose is very small because the ratio of ^{235}U /^{238}U is less than 1% [1]. Several other radionuclides are also present in the environment, one of the most important is Potassium (^{40}K). It contributes significantly to radiation exposure because it is a gamma and beta emitter [2]. The members of the radioactive decay chain of ^{232}Th (14%), ^{235}U and ^{238}U (55.8%), along with ^{40}K (13.8%) are responsible for the main contributions to the dose from natural radioactivity while a mere of 0.3% is due to the effect of ^{87}Rb [3].

Rocks and soil consist of radionuclides in varying concentrations, depends on the local geological and geophysical conditions[4].

The study of the concentrations and distributions of the natural radionuclides in rocks and soil allows to understand the radiological implication of these elements due to the γ-ray exposure of the body and irradiation of lung tissue from inhalation of radon and its daughters[5-7]. In particular, it is also important to assess the radiation hazards due to the use of rocks and soils in the construction of dwellings [8-10]. It is therefore important to measure the concentration of radionuclides in rocks that are used as building materials for assessing the radiological risks to human health.

The objective of this study is to determine the activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in ceramic samples collected from different Yemeni companies. The radium equivalent activity, the air absorbed dose rate, the annual effective dose rate, the representative index and the values of both external and internal hazard indices were evaluated and compared with previous international values.

Sample Preparation

Ten commercial foreign samples from 10 different ceramic tile manufacturing companies used in Yemeni buildings were used in this study. The country of origin and sample code are listed in Table-1. Collected samples were air-dried, dry-weighted, sieved through a fine mesh, well sealed in standard Marinelli beakers (30ml) for about four weeks in order to establish secular equilibrium between ^{232}Th and ^{226}Ra with their progeny. After that period , the samples were measured using a closed end-coaxial Canberra p- type (model 707) high-purity germanium (HPGe) detector with a relative efficiency of 30%. The germanium crystal is located inside the lead shield.

Energy resolution of 1.85KeV, full width to half maximum (FWHM) for the 1332KeV gamma line of Co-60[11]. A mixed radionuclide gamma-ray reference standard consists of a solution in 4M HCl of the ten radionuclides which are ^{241}Am (60KeV), ^{109}Cd (88 KeV), ^{57}Co(122 KeV), ^{139}Ce (166 KeV), ^{203}Hg (279 KeV), ^{113}Sn (392 KeV), ^{85}Sr (514 KeV), ^{137}Cs (662KeV), ^{60}Co (1173 and 1333KeV), ^{88}Y (898 and 1836 KeV) of activities 1131, 644, 577, 738, 1886, 2054, 3838, 2496, 3377, 3382, 6288 and 6651 γ-rays/s.g, respectively as in the reference certificates number QCY48 issued by GE Healthcare limited is used to calibrate HPGe detector.

An empty beaker with the same geometry was measured to subtract the background. The measuring time for both background and activity measurements was 8 hrs to accumulate many counts under the peaks[12]. The total uncertainty in the calculated activity concentration is in most cases below 5%. Fig.1 shows the energy spectrum for samples C-9 and C-4.

Table 1: Tile manufacturing companies of ceramic and the country of origin used in Yemeni buildings.

Companies	*TAULELL*	*CERYPSA*	الفجيرة	رأس الخيمة	*ROMAN*	الجودة	*ATLAS*	*ENGLI*	ردفان	*MERAN*
Country of origin	*Italy*	*Spain*	*Emarate2*	*Emarate1*	*Indonesia*	*Saudi*	*India1*	*India2*	*Yemen*	*China*
Sample code	C-1	C-2	C-3	C-4	C-5	C-6	C-7	C-8	C-9	C-10

Results and Discussion

Radioactivity Determination

Figure 2 illustrates a comparison between different ceramic samples manufactured by different companies for the concentration values of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K respectively. The sample concentrations and its average activities are shown in Table 2. The results show that the average concentrations of ^{238}U, ^{232}Th, ^{226}Ra, ^{224}Ra and ^{40}K radionuclides in the different ceramic samples are 207.2 Bq/kg, 75.2 Bq/kg, 169.13 Bq/kg, 86.7Bq/kg and 400.7 Bq/kg respectively.

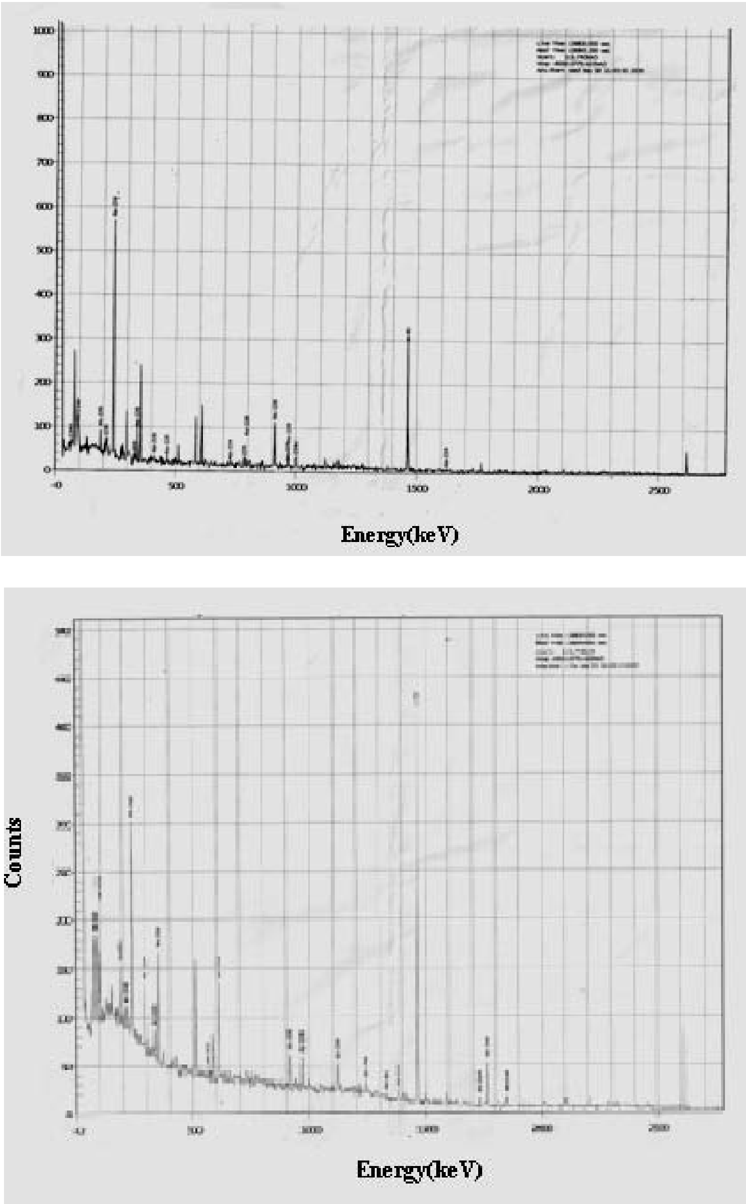

Fig. 1. Energy spectrum of the samples C-9 and C-4 measured using HPGe detector. The measuring time was 8hrs.

Table 2. Activity concentrations in Bq/kg of commercial ceramic samples used in Yemeni buildings.

Sample Lab No	Activity concentrations in (Bq/kg)				
	U-238	**Th-232**	**Ra-226**	**Ra-224**	**K-40**
C-1	253 ± 19.5	48 ± 2.8	13 5 ± 12	14 ± 0.88	547 ± 22
C-2	233 ± 14	42 ± 1.7	92.3 ± 6.5	31± 1.33	816 ± 23
C-3	97 ± 8	13 ± 1	60 ± 7	94 ± 4	463 ± 28
C-4	153 ± 10	61 ± 3	103 ± 10	57 ± 2.2	869 ± 41
C-5	153 ± 13	47 ± 2	114 ± 11	46 ± 2.9	223 ± 21
C-6	0	26 ± 1.1	0	76 ± 3	258 ± 18
C-7	516 ± 28	227 ± 9	452 ± 25	234 ± 10	237 ± 17
C-8	560 ± 40	267 ± 11	549 ± 32	288 ± 15	194 ± 19
C-9	89 ± 9	21 ± 2	125 ± 14	25 ± 1.8	376 ± 26
C10	18 ± 5	0	61 ± 3	2 ± 0.2	24 ± 4
Average constration	207.2	75.2	169.13	86.7	400.7
Range	0 - 560	0 - 267	0 - 549	2 - 288	24 - 869

Results of the average activity concentration of ^{238}U, ^{232}Th and ^{226}Ra in the samples were higher than the world reference values as has been reported in UNSCEAR (1993)[13], but it is lower for ^{40}K. Some values obtained in the present study are lower than the international values; while others are higher. A comparison with previous studies for different countries has been reported as shown in Table-3.

Table 3. A comparison between different ceramic samples activity concentration values for ^{226}Ra, ^{232}Th, and ^{40}K respectively in different countries around the world.

Countries	Activity Concentration (Bq/kg)						Reference
	^{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	------	[19]
Pakistan	63.1-123.9	83.4	-----	------	144.1-834	403.5	[20]
Algeria	-----	55	------	41	-----	410	[21]
Egypt (Cleopatra Factory)	71.2-86	76.1	63.3-68.7	66.2	900-1018	962	[17]
Cameroon	11.3-13.13	12 ± 1	18.63-22.64	20 ±2	-----	319 ± 19	[25]
Italy	-----	56∓5	-----	43∓4	-----	440∓40	[26]
Egypt Lecico & El-Gawhra Factories	41.7-60.7	52.2	30.7-47.1	39.1	195-680	480	[22]
Qena (Egypt)	40-230	126	10-130	72	80-600	300	[23]
Palestine	45.4-102.0	73.7	38.8-78.3	58.2	363-871.2	624	[24]
Yemen	0-549	131.88	13-267	83.55	24-869	400.7	Present work
UNSCEAR	-----	50	-----	50	-----	500	[13]

Assessment of Exposure Risk

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 10Bq/kg of ^{226}Ra, 7Bq/kg of ^{232}Th and 130Bq/kg of ^{40}K produce the same γ-ray dose rates. The equivalent radioactivity is generally defined as[14]

$$Ra_{eq}(Bq/kg) = C_{Ra} + 1.43C_{Th} + 0.077C_{K} \ldots\ldots\ldots(1)$$

Where C_{Ra}, C_{Th} and C_K are the activities of ^{226}Ra, ^{232}Th and ^{40}K respectively in Bq/kg. Eq. (1) is based on the fact that 370Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same gamma ray dose equivalent[25]. The maximum value of Ra_{eq} must be < 370 Bq/kg in order to keep the external dose <1.5 mGy/hr[15]. The Ra_{eq} values are shown in Table (4). The measurements range from (57.05 – 945.75) Bq/kg with an average value 307.52 Bq/kg, which is slightly less than the safe limit.

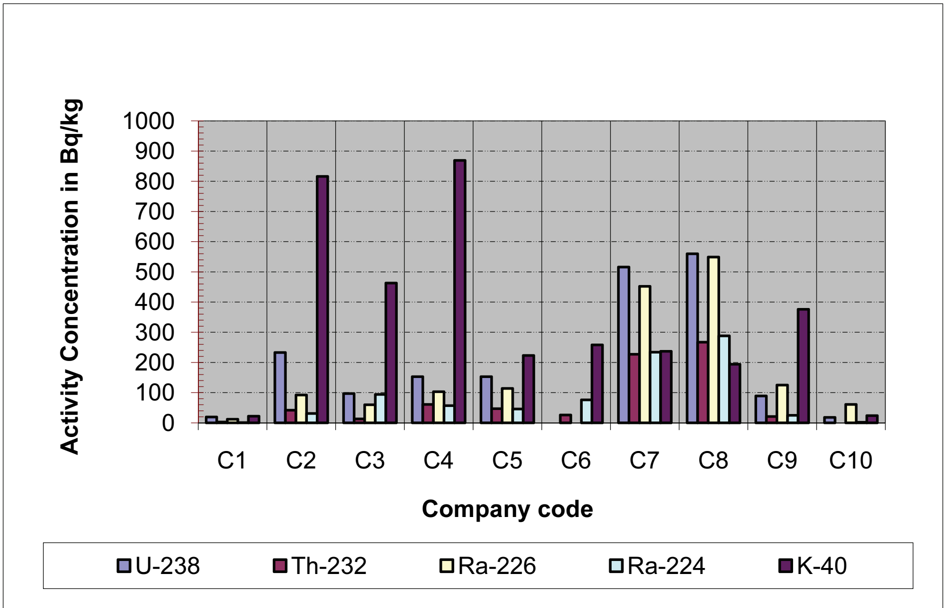

Fig. 2. **A comparison between different ceramic samples manufactured by different companies is shown in for the concentration values of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K respectively .**

Table 4. Radiation indices of the commercial ceramic samples from different companies used in Yemeni buildings.

Company cod	Ra_{eq}(Bq/Kg)	D_r(nGy/hr)	AEDE mSv/yr	H_{ex}	Iγ
C-1	245.76	113.05	1.13	0.66	1.75
C-2	215.19	102.47	1.02	0.58	1.58
C-3	114.24	54.23	0.54	0.31	0.84
C-4	257.14	121.90	1.21	0.69	1.88
C-5	198.38	89.43	0.90	0.54	1.38
C-6	57.05	28.36	0.282	0.15	0.43
C-7	794.86	353.52	3.55	2.15	5.44
C-8	945.75	419.56	4.22	2.55	6.46
C-9	183.98	83.52	0.83	0.50	1.29
C10	62.85	27.08	0.27	0.17	0.42
Range **Average**	57.05- 945.75 307.52	27.08-419.56 139.31	0.27-4.22 1.40	0.15-2.55 0.83	0.42 -6.46 2.15

The Absorbed Dose Rate

The absorbed 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}, C_K) in Bq/kg at a height of 1m above the ground was calculated by the following equation (UNSCEAR, 1998)[12].

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

The values 0.427, 0.662 and 0.0432 nGy h^{-1}per Bq kg^{-1} are the conversion factors for ^{226}Ra, ^{232}Th and 40K[27]. The obtained values of D_r range between (27.08-419.56) nGy/hr, with an average value of 139.31nGy/hr (Table 4). The calculated gamma dose rates in all ceramic samples are higher than the international recommended value 55 nGy/hr[12] by 153% with exception of two samples C-6 and C-10.

The Annual Effective Dose Equivalent

The annual effective dose equivalent (AEDE) for the worker or public in mSv/yr was calculated using the equation given by [16].

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

Where the values 0.49, 0.76 and 0.048 are the conversion factors for ^{226}Ra, ^{232}Th and ^{40}K. The results are presented in (Table 4). The highest value of AEDE was found to be 4.22 mSv/yr. while the lowest value was found to be 0.27. The results of some companies (sample No. C-4, C-7 and C-8) are about 2 to 320% higher than the international values (1mSv/yr) for the public; the rest is lower than the permissible value[17].

The External Hazard Index

The external hazard index H_{ex} is used to asses radiation risks attributed to radioactive materials. H_{ex} is calculated using the following equation[15].

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

The value of H_{ex} must be lower than unity to keep the radiation hazard insignificant. The maximum value of unity for H_{ex} corresponds to the limit 370 Bq/kg for Ra_{eq}[18]. The calculated values of the H_{ex} for ceramic samples are given in (Table 4). The values are found to range from (0.15-2.55) with an average value of 0.83. The results of H_{ex} for samples (C-7 and C-8) are about twice higher than the maximum value. The values of the other samples are lower than unity which are in agreement with the international values.

The Radioactivity Level Index

Radioactivity level index is used to estimate the level of γ-radiation hazards associated with the natural radionuclides. The level index I_γ is calculated by the equation given by[15]

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

The values 150, 100 and 1500 are the factors of a dose criterion of 1mSv/yr for ^{226}Ra, ^{232}Th and ^{40}K respectively. The values of I_γ are listed in Table 3. All the values exept samples C-3, C-6 and C-10 are higher than unity ($I_\gamma > 1$).

Conclusions

The natural radioactivity and related radiation hazards from the 10 investigated ceramic samples used in Yemeni buildings were determined using γ-ray spectrometry. The following conclusions can be retained in:

1. The average value of the concentrations for ^{238}U, ^{232}Th, ^{226}Ra, ^{224}Ra and ^{40}K have been found to lie within the range 0-560∓40, 0-267∓11, 0-549∓32, 24∓4-869∓41 Bq/kg, respectively.

2. The radium equivalent activity for most ceramic samples is lower than the recommended limit 370 Bq/kg set in UNSCEAR[13] report with the exception of two samples.

3. The absorbed dose rate in air was found to vary from 27.08-419.56 nGy/hr .The average value of (139.31 nGy/hr) is higher than the international values of 55 nGy/hr[12].

4. The average corresponding effective dose 1.40mSv/yr is higher than the admissible value of 1mSv/yr.

5. The results showed that the average H_{ex} is 0.83 in ceramic samples. Two samples show higher values than the international value but the other ceramic samples are in agreement with the recommended value.

6. The average value of I_γ is 2.15 is higher than the world value. The calculated value of I_γ for most of the samples is greater than unity. Thus the samples are percipience in the radioactivity as the first enhanced level.

In regard to the above results, we conclude that the ceramics used as building materials are a source of background radiation that contributes the total annual dose rate of gamma radiation in Yemeni dwellings by a value of 40% on average.

References

[1] **Walley El-Dine, N., El-Shershaby, A., Ahmed, F.** and **Abdel-Haleem, A.S.,** Measurement of radioactivity and radon exhalation rate in different kinds of marbles and granites, *Appl. Radiat. Isot.*, **55**: 853-860 (2001).

[2] **Xinwei, L., Lingqing, W., Xiaodan, J., Leipeng, Y.** and **Gelian, D.,** Specific activity and hazards of Archeozoic-Cambrian rock samples collected from the Weibei area of Shaanxi, China. *Radiat. Prot. Dosim.*, **118**: 352-359 (2006).

[3] **Bruzzi, L., Baroni, M., Mele, R.** and **Nanni, E.,** Proposal for a method of certification of natural radioactivity in building materials, *J. Radiol. Prot.*, **17**: 85-94 (1997).

[4] **Florou, H.** and **Kritidis, P.,** Gamma radiation measurements and dose rate in the coastal areas of a Volcanic Island, Aegean Sea, Greece, *Radiat. Prot. Dosim.*, **45**: 277-279 (1992).

[5] **Singh, S., Rani, A.** and **Mahajan, R. K.,** ^{226}Ra, ^{232}Th and ^{40}K analysis in soil samples from some areas of Punjab and Himachal Prodesh, Indian using gamma ray spectroscopy, *Radiat. Meas.*, **39**: 431-439 (2005).

[6] **Abbady, A., Ahmed, N.K., El-Arabi, A.M., Michel, R., El-Kamel, A.H.** and **Abbady, A.G.E.,** Estimation of radiation hazard indices from natural radioactivity of some rocks, *Nucl. Sci. Techn.*, **17**: 118-122 (2006).

[7] **Tzortzis, M., Tsertos, H., Christofides S.** and **Christodoulides G.,** Gamma-ray measurements of naturally occurring radioactive samples from Cyprus characteristic geological rocks, *Radiat. Meas.*, **37**: 221-229 (2003).

[8] **Khatibeh, A.J.A.H., Ahmed, N.** and **Matiullah, Kenawy, M.A.,** Natural radioactivity in marbles stones, *Jordan Radiat. Meas*., **28**: 345-348 (1997).
[9] **Kumer, R., Sengupta, D.** and **Prasad, R.,** Natural radioactivity and radon exhalation studies of rock samples from Surda Copper deposites in Singhbluan sheat zone, *Radiat. Meas*., **36**: 551-553 (2003).
[10] **Matiullah Abad A., Rehmar, S., Rhman S.** and **Feheem M.,** Measurement of radioactivity in the soil of Bahawalpur division, *Pakistan. Radiat. Prot. Dosim*.**, 1112**: 443-447 (2004).
[11] **Chiozzi P., Pasquale V.** and **Verdoya, M.,** Naturally occurring radioactivity at the Alps-Apennines transition, *Radiat. Meas.*, **35**: 147-154 (2002).
[12] **UNSCEAR,** Sources, Effects, and Risks of Ionizing Radiation, Report to the General Assembly, with scientific annexes, New York: UN (2000).
[13] **UNSCEAR,** Exposure From Natural Sources of Radiation, Report to the General Assembly, New York: UN (1993).
[14] **Brigido O., Montalvan, A.** and **Tomas, J.,** Natural radioactivity in some building materials in Cuba and their contribution to the indoor gamma dose rate, *Rad. Prot. Dos.,* **113**(2): 218-222 (2005).
[15] **Beretka, J.** and **Mathew, P.,** Natural radioactivity of Australian building materials, industrial wastes and by products, *Health Phys.,* **48**: 87-95 (1985).
[16] **Malanca A., Pessina, V.** and **Dallara, G.,** Radionuclide Content of Building Materials and Gamma Ray Dose Rates in Dwellings of Rio Grande Do Norte, Brazil, *Rad. Prot. Dos.,* **48**(2): 199-203 (1993).
[17] **Hilal, M.,** Nuclear Spectroscopic measurements for different environmental radiations., Ph. D. thesis, Faculty of Science, University of Man soura, Egypt*,* 138 (2002).
[18] **[18] Hyu N., and Luyen T.,** Study on external exposure doses from terrestrial radioactivity in southern Vietnam, *Rad. Prot. Dos.,* **118**(3): 331-336 (2005).
[19] **Xinwei, L.,** Radioactivity level in Chinese building ceramic tile, *Rad. Prot. Dos.,* **112**(2): 323-327 (2004).
[20] **Tufail, M., Ahmed, N., Almakky, S., Zafar, M.** and **Khan, H.,** Natural radioactivity in the ceramic used in dwellings as construction material, *Sci. Total Environ*., **127**(3): 243-253 (1992).
[21] **Amarani, A.** and **Tahtat, M.,** Natural radioactivity in Algerian building materials**,** *Appl. Radiat. Isot.,* **54**(4): 687-689 (2001).
[22] **Higgy, R.,** Measurements and analyses of low level natural radioactivity in building materials and evaluation of its environmental impact. Ph. D. thesis, Faculty of Science, Ain Shams University, Ciro, Egypt, 130-136 (1995).
[23] **Ahmed, N.,** Measurements of natural radioactivity in building materials in Qena City-Upper Egypt, *4th Radiation Physics Conference,* Alexandria-Egypt, 15-19 (1999).
[24] **Dabayneh, K.M.,** Natural Radioactivity in Different Commercial Ceramic Samples Used in Palestine Buildings as Construction Materials, *Herbron University Research Journal*, **3**(2): 49-58 (2008).
[25] **Ngachin, M., Garavglia, M., Giovani, C., Kwato, Njock, M.G.** and **Nourreddine, A.,** Assessment of natural radioactivity and associated radiation hazards in some Cameroonian building materials, *Radiat. Meas*., **42**: 61-67(2007).
[26] **Righi, S.** and **Bruzzi, L.,** Natural Radioactivity and radon exhalation in building materials used in Italian dwellings, *J. of Environmental Radioactivity*, **88**: 158-170 (2006).
[27] **UNSCEAR,** United Nations Scientific Committee on the Effect of Atomic Radiation, Sources, effect and risk of ionizing radiation, New York: UN (2000).

دراسة النشاط الإشعاعي الطبيعي لعينات من السيراميك التجاري المستخدم في المباني اليمنية

سحر أحمد أمين[1] و مصطفى ناجي[2]

قسم الفيزياء، كلية العلوم، جامعة تعز، تعز- اليمن

قسم الفيزياء، كلية التربية، جامعة عدن، عدن- اليمن

المستخلص: لقد تم في هذا البحث دراسة تركيز النويدات ذات النشاط الإشعاعي الطبيعي لعناصر الراديوم-٢٢٦، الثوريوم-٢٣٢، ونظير البوتاسيوم-٤٠، لعشرة عينات من السيراميك، جمعت من عشرة شركات مختلفة التي تستخدم كمواد بناء في المنازل اليمنية.

استخدمت تقنية مطيافية جاما لتحليل العينات باستخدام كاشف الجرمانيوم عالي النقاوة، وحساب تركيز النظائر المشعة بوحدات بيكريل/كغم. لقد وجد إن متوسط تركيز هذه النويدات ١٣١,٤، ٨٣,٥٥، ١٣١,٨٨ و ٤٤,٧ بيكريل/كغم للنويدات اليورانيوم-٢٣٨، الثوريوم-٢٣٢، الراديوم-٢٢٦، والبوتاسيوم-٤٠ على التوالي. لقد تم تقدير الأضرار الإشعاعية الناجمة عن النشاط الإشعاعي الكلي، وذلك بحساب بعض المعاملات الإشعاعية المختلفة. أظهرت النتائج إن معدلات نشاط الراديوم المكافئ (Ra_{eq})، معدل الجرعة الممتصة (D_r)، مكافئ الجرعة الفعالة السنوي (AEDE)، معامل الأخطار الخارجي (H_{ex})، ومعامل مستوى الإشعاع(I_γ) هي: ٣٠٣,٥٢ بيكريل/كغم، ١٣٩,٣١ نانوجراي/ساعة، ١,٤٠ ملي سفرت/سنة، ٠,٣٨ و ٢١٥ على التوالي. وبإجراء مقارنة علمية وجدنا أن متوسط قيمة نشاط اليورانيوم المكافئ متوافق

مع القيم العالمية، بينما متوسط القياسات لجميع المعاملات الأخرى أعلى من الحدود العالمية. و أخيرًا فقد تم مقارنة تركيز النويدات الطبيعية الناتجة مع دراسات أخرى في العديد من دول العالم.