

**Measurement of natural radioactivity in some types of cement samples
by using (HPGe) detector**

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Abstract

In the present work, we have measured specific activity concentrations in ten cement samples from different origins by using high purity germanium (HPGe) detector. The results have shown that, the specific activity, for ^{238}U was ranged from $(22.23\pm 4.7 \text{ Bq/kg})$ in Turki origin to $(9.39\pm 3 \text{ Bq/kg})$ in Iraq (Najif) origin, for ^{232}Th was ranged from $(24.56\pm 4.9 \text{ Bq/kg})$ in Turki origin to $(8.98\pm 2.9 \text{ Bq/kg})$ in Iraq (Najif) origin, for ^{40}K was ranged from $(317.81\pm 17.8 \text{ Bq/kg})$ in Turki origin to $(67.34\pm 8.2 \text{ Bq/kg})$ in Iraq (Kabisa) origin. The study shows the cement samples do not pose any significant source of radiation hazard and are safe for use in the construction of dwellings.

Key Words: HPGe, cement samples, natural radioactivity.

قياس النشاط الإشعاعي الطبيعي لبعض أنواع نماذج الاسمنت باستخدام كاشف الجرمانيوم

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الخلاصة

تم في هذا البحث قياس النشاط الإشعاعي النوعي لعشرة نماذج اسمنت من مختلف المنشآت باستخدام كاشف الجرمانيوم عالي النقاوة . بينت النتائج ان النشاط الاشعاعي النوعي لليورانيوم- ^{238}U تراوحت قيمته بين $(22.23\pm 4.7 \text{ Bq/kg})$ تركي المنشئ الى $(9.39\pm 3 \text{ Bq/kg})$ لاسمنت عراقي (نجف) المنشئ بالنسبة الى ^{238}U ، وبين $(24.56\pm 4.9 \text{ Bq/kg})$ تركي المنشئ الى $(8.98\pm 2.9 \text{ Bq/kg})$ عراقي (نجف) المنشئ بالنسبة الى ^{232}Th ، وبين $(317.81\pm 17.8 \text{ Bq/kg})$ تركي المنشئ الى $(67.34\pm 8.2 \text{ Bq/kg})$ عراقي (كبيسة) المنشئ بالنسبة الى ^{40}K . بينت الدراسة ان جميع نماذج الاسمنت قيد الدراسة لا تشكل أي مصدر للخطر وهي امينة للاستعمال في بناء المساكن.
الكلمات الافتتاحية: كاشف الجرمانيوم عالي النقاوة ، نماذج الاسمنت ، النشاط الإشعاعي الطبيعي.

Introduction

Our world is radioactive and has been since it was created. Over 60 radionuclides can be found in nature. Radionuclides are found in air, water and soil, and additionally in us, being that we are products of our environment. Every day, we ingest/inhale nuclides in the air we breathe, in the food we eat and the water we drink. Radioactivity is common in the rocks and soil that makes up our planet, in the water and oceans, and even in our building materials and homes. All building raw materials and products derived from rock and soil contain various amounts of mainly natural radionuclides of the uranium and thorium series, and the radioactive isotope of potassium (^{40}K). It has long been known that some construction materials are naturally more radioactive than others. The natural level of radioactivity in construction materials, even of low-level activity, gives rise to external and internal indoor exposure [1]. The external radiation exposure is caused by gamma radiation originating from members of the uranium and thorium decay chains and from 40 potassium, however, the internal radiation exposure, mainly affecting the respiratory tract, is due to the short-lived daughter products of radon which are exhaled from construction materials into room. Thus, the knowledge of radioactivity in building materials is important to estimate the radiological hazards on human health [2]. The most important naturally occurring radionuclides present in cements are (^{238}U), (^{232}Th) and (^{40}K) as mentioned above [3].

Materials and Method

In the present work, we have adopted the gamma spectroscopic method using high purity germanium (HPGe) detector for the detection and measurement of the specific activity of the radioactive elements in some types of cement samples, the cement samples were crushed to small pieces then to fine powder by using an electrical mill. (1 kg) of about (300 mm) grain by using special sieves (mesh). The samples were dried at (50 °C) for one hour and they were packaged in a marinelli beaker, the sealed marinelli beaker were kept for one month before measurements in order to achieve the secular equilibrium for ^{238}U and ^{232}Th with their respective progenies

Each cement sample under investigation was about of (1 kg) in weight ,which was washed with distilled water to remove the formalin liquid (conservator substance), and was cut and put in the marenilli beaker uniformly and then examined with high purity germanium detector for a period of (3) hours.

The energy calibration of germanium detector system was found by using some standard radioactive sources with known energies. These sources should be counted for a long enough period in order to produce well-defined photopeaks and then calibrated according to their energies

[4]. In the present work, standard radioactive source (^{22}Na) with a period of (1080 s.) was used for the system calibration, the Gamma-ray spectra for this source is shown in Figure (1).

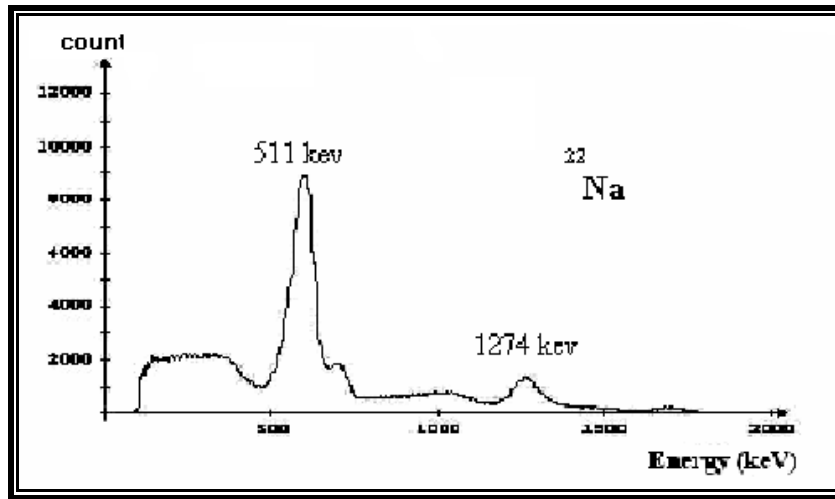


Figure (1) Gamma-ray spectrum for (^{22}Na) source

The efficiency of scintillation detector is defined as the ratio of the number of pulses (counts per unit time) recorded by the detector to the number of radiation quanta (photons) emitted by the source (disintegration per unit time) the following equation [5]:

$$\text{Efficiency (\%)} = \frac{\text{cps}}{\text{dps}} \times 100\% \quad \dots (1)$$

where

cps : count per second

dps : disintegration per second (the source activity)

For this purpose, spectra of standard radioactive sources with known energies and activities are accumulated for a long enough time by the detector to produce well-defined photopeaks. In the present work, the high purity germanium detector (50 %) efficiency.

The specific activity for each detected radionuclide (radioactive element) had been calculated using the following equation [6]:

$$\text{Specific Activity} = \frac{\text{Net area under the peak}}{W \times I_{\gamma} \times \text{Eff.} \times T} \quad \dots (2)$$

Where:

T : Measuring time (s)

Eff.: Percentage efficiency

I_γ : Percentage intensity of gamma-ray **W**: Mass of the sample (kg)

Net area under the peak: (Total counts – Background)

Measurement of Parameters

1. Radium Equivalent Activity (Ra_{eq})

To represent the activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K by a single quantity, which takes into account the radiation hazards associated with them, a common radiological index has been introduced. The index is called radium equivalent activity (Ra_{eq}) which is used to ensure the uniformity in the distribution of natural radionuclides ²³⁸U, ²³²Th and ⁴⁰K and is given by the expression [7]:

$$Ra_{eq} \text{ (Bq/kg)} = A_U + 1.43A_{Th} + 0.077A_K \dots\dots\dots (3)$$

Where ,A_U, A_{Th} and A_K are the specific activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in (Bq/kg) respectively.

2. Absorbed Gamma Dose Rate (D_γ)

Outdoor air gamma absorbed dose rate (D_γ) in (nGy/h) due to terrestrial gamma rays at hight (1 m) above the ground surface which can be computed from specific activities A_U, A_{Th} and A_K of ²³⁸U, ²³²Th and ⁴⁰K in (Bq/kg) respectively using the following relation [8]:

$$D_{\gamma} \text{ (nGy/h)} = 0.462A_U + 0.604A_{Th} + 0.0417A_K \dots\dots\dots (4)$$

3. Annual Effective Dose Rate (AED)

The estimated annual effective dose equivalent received by a member was calculated by using a conversion factor of (0.7 Sv/Gy), which was used to convert the absorbed rate to human effective dose equivalent with an outdoor occupancy of 20 % and 80 % for indoors [9]:

$$(AED)_{indoor} \text{ (mSv/y)} = D_{\gamma} \text{ (nGy/h)} \times 10^{-6} \times 8760 \text{ h/y} \times 0.80 \times 0.7 \text{ Sv/Gy} \dots\dots (5)$$

$$(AED)_{outdoor} \text{ (mSv/y)} = D_{\gamma} \text{ (nGy/h)} \times 10^{-6} \times 8760 \text{ h/y} \times 0.20 \times 0.7 \text{ Sv/Gy} \dots\dots (6)$$

4. External Annual Dose (EAD)

The external annual effective dose was calculated by using the following equation [10]:

$$EAD = (0.92A_U + 1.1A_{Th} + 0.08A_K) \times (10^{-9} \text{ Gy/h}) \times (0.7 \text{ Sv/Gy}) \times (24 \times 365) \text{ h/y} \times 0.8 \dots\dots (7)$$

5. Activity Concentration Index (I_γ)

The activity index (I_γ) for soil samples was calculated by using the following equation [10]:

$$I_{\gamma} = \frac{A_U}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \dots\dots\dots (8)$$

6. External (H_{ex}) and Internal (H_{in}) Hazard Indices

Beretka and Mathew [11] defined two other indices that represent internal and external radiation hazards. The external hazard index is obtained from (Ra_{eq}) expression through the supposition that its allowed maximum value (equal to unity) corresponds to the upper limit of Ra_{eq} (370 Bq/kg). The external hazard index (H_{ex}) can then be defined as:

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad \dots\dots\dots (9)$$

Internal exposure to ²²²Rn and its radioactive progeny is controlled by the internal hazard index (H_{in}) as given below [12]:

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad \dots\dots\dots (10)$$

This index value must be less than unity in order to keep the radiation hazard to be insignificant.

Results and Discussion

Our present investigation is based on the study of (10) samples of different kinds of cement samples, which was available in the local market, some of them were Iraqi made and the others from different countries like, (Egypt, Iran, Lebanon, Jordan and Turkiyi).

Table (1) presents specific activity concentration for cement samples in different countries, from Table (1) it can be noticed that:

The highest value of specific activity concentration of (²³⁸U) in cement samples was found in Turkiyi origin, which was equal to (22.23±4.7 Bq/kg), while the lowest value of specific activity concentration of (²³⁸U) in cement samples was found in Iraq (Najif) origin, which was equal to (9.39±3 Bq/kg), see Figure (2), with an average value of (15.084±3.8 Bq/kg). The present results show that values of specific activity concentrations of (²³⁸U) in the studied were lower than the value of the specific activity concentration of (²³⁸U) global limit which is equal to (35 Bq/kg) [13].

The highest value of specific activity concentration of (²³²Th) was found in Turkiyi origin, which was equal to (24.56±4.9 Bq/kg), while the lowest value of specific activity concentration of (²³²Th) in cement samples was found in Iraq (Najif) origin, which was equal to (8.98±2.9 Bq/kg), see Figure (3), with an average value of (15.426±3.8 Bq/kg). The present results show that values of specific activity concentrations of (²³²Th) in the studied regions in cement samples were lower than the value of the specific activity concentration of (²³²Th) global limit which is equal to (30 Bq/kg) [13].

The highest value of specific activity concentration of (⁴⁰K) was found in Turkiyi origin, which was equal to (317.81±17.8 Bq/kg), while the lowest specific activity concentration of (⁴⁰K) in cement samples was found in Iraq (Kabisa) origin, which was equal to (67.34±8.2 Bq/kg), see Figure (4), with an average value of (152.364±12 Bq/kg). The present results show that values of specific activity concentrations of (⁴⁰K) in cement samples were lower than the value of the specific activity concentration of (⁴⁰K) global limit which is equal to (400 Bq/kg) [13].

The highest value of radium equivalent activity (Ra_{eq}) was found in Turki origin, which was equal to (81.822 Bq/kg), while the lowest value of radium equivalent activity in cement samples was found in Iraq (Najif) origin, which was equal to (31.219 Bq/kg), with an average value of (48.875±10.3 Bq/kg). The present results show that the values of radium equivalent activity in cement samples were lower than the value of the radium equivalent activity global limit which is equal to (370 Bq/kg) [13].

The highest value of the absorbed gamma dose rate (D_γ) was found in Turki origin, which was equal to (38.357 nGy/h), while the lowest value of the absorbed gamma dose rate in cement samples was found in Iraq (Najif) origin, which was equal to (14.629 nGy/h), with an average value of (22.64±4.8 nGy/h). The present results show that the values of absorbed gamma dose rate in cement samples were lower than the value of the absorbed gamma dose rate global limit which is equal to (55 nGy/h) [13].

The highest value of indoor annual effective dose rate (AED)_{in} was found in Turki origin, which was equal to (0.188 mSv/y), while the lowest value of indoor annual effective dose rate in cement samples was found in Iraq (Najif) origin, which was equal to (0.072 mSv/y), with an average value of (0.111±0.02 mSv/y). The present results show that values of indoor annual effective dose rate in the studied in cement samples were lower than the values of the indoor annual effective dose global limit which is equal to (1 mSv/y) [13].

The highest value of outdoor annual effective dose rate (AED)_{out} was found in Turki origin, which was equal to (0.047 mSv/y), while the lowest value of indoor annual effective dose rate in cement samples was found in Iraq (Najif) origin, which was equal to (0.018 mSv/y), with an average value of (0.028±0.006 mSv/y). The present results show that values of outdoor annual effective dose rate in cement samples were lower than the value of the outdoor annual effective dose rate global limit which is equal to (1 mSv/y) [13].

The highest value of external annual effective dose (EAD) was found in Turki origin, which was equal to (0.358 mSv/y), while the lowest value of external annual effective dose in cement samples was found in Iraq (Najif) origin, which was equal to (0.137 mSv/y), with an average value of (0.211±0.04 mSv/y). The present results show that the values of the external annual effective dose in cement samples were lower than the value of the outdoor annual effective dose global limit which is equal to (1.5 mSv/y) [13].

The highest value of the activity concentration index (I_γ) was found in Turki origin, which was equal to (0.303), while the lowest value of the activity concentration index in cement samples was found in Iraq (Najif) origin, which was equal to (0.115), with an average value of (0.192±0.048). The present results show that values of the activity concentration index in

cement samples were lower than the value of the activity concentration index global limit which is equal to (1) [13].

The highest value of internal hazard index (H_{in}) was found in Turkey origin, which was equal to (0.281), while the lowest value of internal hazard index in cement samples was found in Iraq (Najif) origin, which was equal to (0.110), with an average value of (0.173±0.03). The present results show that values of internal hazard index in cement samples were lower than the value of the internal hazard index global limit which is equal to unity [13].

The highest value of external hazard index (H_{ex}) was found in Turkey origin, which was equal to (0.221), while the lowest value of external hazard index in cement samples was found in Iraq (Najif) origin, which was equal to (0.084), with an average value of (0.132±0.02). The present results show that values of external hazard index in cement samples were lower than the value of the external hazard index global limit which is less than unity [13].

Conclusions

This study showed that the analyzing cement samples by using high purity germanium (HPGe) detector from different kinds of cement samples. The specific activity concentrations in (^{238}U , ^{232}Th and ^{40}K) are below the specific activity concentration of global limit, The study shows all the cement samples do not pose any significant source of radiation hazard and are safe for use in the construction of dwellings.

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Table (1) The specific activities of radionuclides and Parameters in cement samples.

Sample Code	Origin	²³⁸ U (Bq/kg)	²³² Th (Bq/kg)	⁴⁰ K (Bq/kg)	Raeq (Bq/Kg)	D _v (nGy/h)	(A.E.D) (mSv/y)		EAD (mSv/y)	I _v	Hazard index	
							Indoor E _{in}	Outdoor E _{out}			H _{in}	H _{ex}
S ₁	Iraq (Kirkuk)	11.52±3.3	14.59±3.8	89.43±9.4	39.270	17.864	0.088	0.022	0.166	0.282	0.137	0.106
S ₂	Iraq Sulaymaniya	15.21±3.9	20.14±4.4	121.24±11	53.346	24.247	0.119	0.030	0.225	0.192	0.185	0.144
S ₃	Iraq (Kabisa)	10.14±3.1	14.47±3.8	67.34±8.2	36.017	16.233	0.080	0.020	0.150	0.129	0.125	0.097
S ₄	Iraq (L-Qaim)	14.04±3.7	18.66±4.3	121.74±11	50.098	22.834	0.112	0.028	0.212	0.181	0.173	0.135
S ₅	Iraq (Najif)	9.39±3	8.98±2.9	116.72±10.8	31.219	14.629	0.072	0.018	0.137	0.115	0.110	0.084
S ₆	Egypt	14.41±3.7	11.33±3.3	174.23±13.2	44.028	20.766	0.102	0.025	0.195	0.163	0.158	0.119
S ₇	Iran	17.46±4.1	13.46±3.6	140.54±11.8	47.529	22.057	0.108	0.027	0.207	0.172	0.176	0.128
S ₈	Jordan	17.51±4.1	11.45±3.3	122.05±11	43.281	20.095	0.099	0.025	0.189	0.156	0.164	0.117
S ₉	Lebanon	18.93±4.3	16.62±4	252.54±15.8	62.142	29.315	0.144	0.036	0.274	0.230	0.219	0.168
S ₁₀	Turkyi	22.23±4.7	24.56±4.9	317.81±17.8	81.822	38.357	0.188	0.047	0.358	0.303	0.281	0.221
Min		9.39	8.98	67.34	31.219	14.629	0.072	0.018	0.137	0.115	0.110	0.084
Max		22.23	24.56	317.81	81.822	38.357	0.188	0.047	0.358	0.303	0.281	0.221
Average		15.084 ±3.8	5.426±3.8	152.364±12	48.875±10.3	22.64±4.8	0.111±0.02	0.028±0.006	0.211±0.04	0.192±0.048	0.173±0.03	0.132±0.02
Global limit [13]		35	30	400	370	55	1	1	1.5	1	1	1

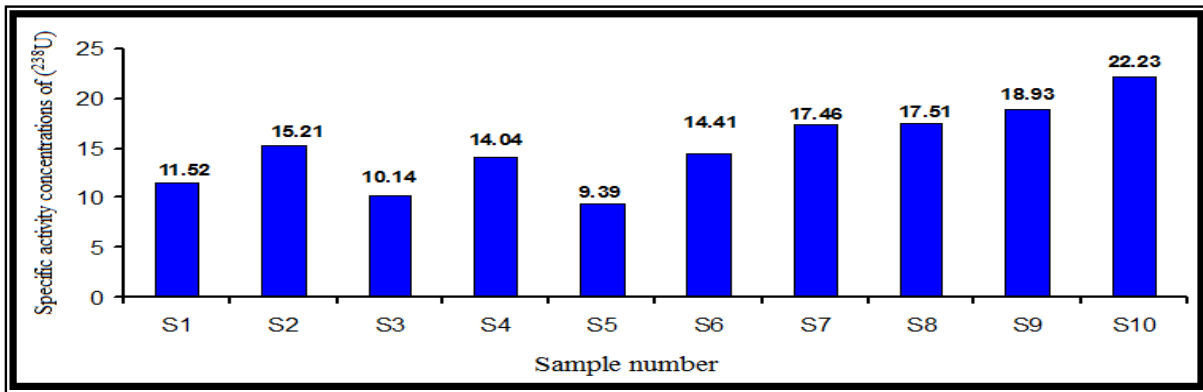


Figure (2) a histogram illustrating the change in specific activity concentrations of ^{238}U for all cement samples sites.

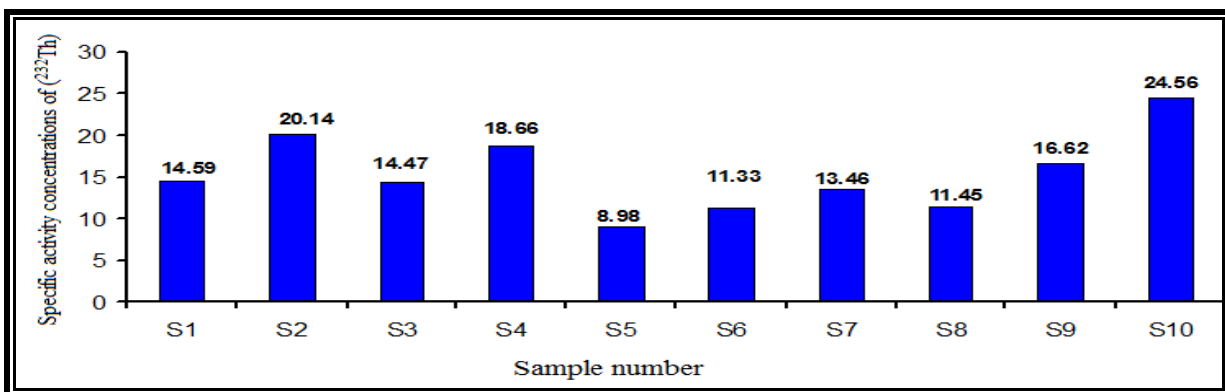


Figure (3) a histogram illustrating the change in specific activity concentrations of ^{232}Th for all cement samples sites.

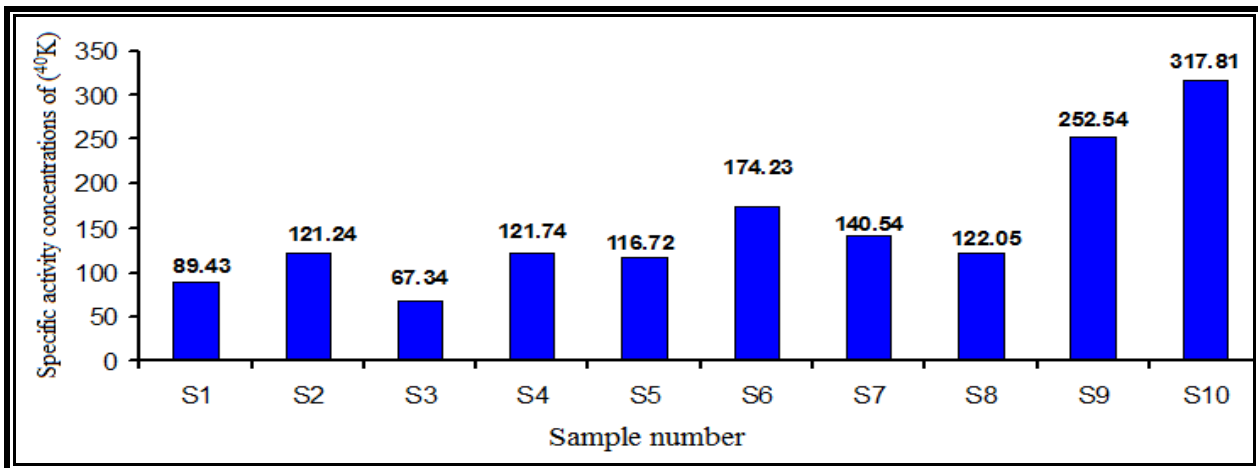


Figure (4) a histogram illustrating the change in specific activity concentrations of ^{40}K for all cement samples sites.