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ORIGINAL PAPER

Assessment of radioactivity levels in some cement produced locally in Iraq

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Abstract

Purpose Due to the rapid expansion of the construction industries, cement production is ever-increasing worldwide and maintaining the qualities of the local cement became vital for safe and durable structures. In this regard, the radiometric assessment of cement plays a significant role.

Method The specific activities of the naturally occurring radionuclides present in ten Portland cement samples that can cause possible health hazards were determined using a gamma-ray spectrometer equipped with (3 × 3) inch NaI (Tl) detector with 1024 channels.

Result and conclusion The measured activity concentrations of *Ra-226*, *Th-232,* and *K-40* were found in the range of 12.38–55.75 (average of 31.29 Bq/kg), 6.29–57.54 (average of 15.97 Bq/kg), and 112.63–266.37 (average of 168.24 Bq/kg), respectively. The recorded average activity concentration of these radionuclides in the cement was below the stipulated world standard by UNSCEAR (Sources and Effects of Ionizing Radiation, United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR 2000 Report to the General Assembly, with Scientific Annexes, vol. I, 2000). The obtained mean activity concentration of Ra_{eq} in the cement (67.101 Bq/kg) was much lower than the recommended guideline of 370 Bq/kg. In addition, the mean value of the calculated absorbed dose rate (59.947 $nGy{\cdot}h^{-1}$), annual effective dose (0.220 $mSv{\cdot}y^{-1}$), activity concentration index (0.240), alpha index (0.156), excess lifetime cancer risk (0.771×10^{-3}) external (0.084) and internal (0.300) hazard index of the studied samples were much below the recommended safety limits. It is affirmed that the cement sold in the local market can safely be used for building constructions.

Keywords Cement · Concentration · Absorbed dose · Alpha index · Health hazards

Introduction

Geologically, the radioactive materials are widely distributed throughout the earth's crust that presents at different levels in the rocks and soils. It is known that the earth is radioactive because of its configuration and origin. However, the concentrations of the natural radioisotopes in the cement to a large extent are based on the soil structures, compositions, and qualities. In fact, the radioactivity of the rocks that form the soil decides its radioactivity [1]. To determine the effects of radiation exposure due to extraterrestrial and terrestrial sources, it is important to know the distribution of radionuclides and radiation concentrations in the environment. The natural radioactivity in the solid produces gamma radiation and changes the background radiation level in the environment [2]. Commonly, the primitive radionuclides are present in various types of isotopes around the world wherein the living systems are continuously exposed to this natural ionizing radiation [3]. The radionuclides that exist on the earth's crust are the primary sources of naturally occurring radioactive compounds worldwide. Most of these radionuclides are chain members or radioactive decline chain members [4].

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Some studies revealed that the human exposure to the radioactivity of the structural building materials can pose a severe health risk, where both external and internal radiation exposure due to the inhalation of the radium gaseous progeny are responsible for such health hazards unless inhibited [5, 6]. More than 60 radionuclides that are found in nature can be divided into three groups including the primordial, cosmogenic, and human-generated [7]. The estimation of the radiation in the construction materials became significant to identify the environmental risks to public health. Thus, it appears necessary to establish the standard levels of radiation and national guidelines consistent with the international recommendations [8]. The characteristic level of the radioactivity in the manufactured construction materials can contribute to both internal and external exposure even at low activity levels [9]. The consequence of the external exposure to the natural environmental gamma radiation depends on the geological and geographical conditions at various ground levels [10]. Certainly, the evaluation of the population exposure due to the internal radiation is highly significant. In addition, a detailed understanding of the concentrations of the natural radionuclides in the building materials is essential [11]. The local cement in Iraq contains *Ra-226*, *Th-232*, and *K-40* as the most common natural radionuclides. In-depth knowledge related to the radiological parameters including the radioactive contents of the building materials is vital for determining the potential radiation exposure to the Iraqi nationals because the majority of them spend most of their lives in their homes and offices [12].

Based on the abovementioned factors and the need for human health protection, this study made a radiometric evaluation involving the activities of the natural radionuclides (*Ra-226*, *Th-232*, and *K-40*) present in the cement manufactured in Iraq. Various radiological parameters were determined to assess the radiation risks associated with the cement and its compounds commonly produced locally and used for building construction in Iraq. The results were analyzed, discussed, interpreted, and compared with other findings. It is asserted that the achieved information can be used to establish some standards and guidelines for managing these materials production and environmental mitigation.

Preparation of samples

All the cement samples were collected from the domestic market in Iraq and dried for 1 day in the air at ambient temperature to maintain the constant weight. Each cement sample of weight 1 kg was milled to get the fine powder and strained through a sieve hole of diameter 2 mm. All 10 samples were dried for 24 h in the oven at 110 °C and packed in the Marinelli beakers (1000 mL) with a plastic cover for their radiological tests. To achieve secular equilibrium between ^{226}Ra and its daughters, samples were kept in hermetically sealed containers for 30 days (short-lived). In cement samples, the natural radionuclides ^{226}Ra, ^{232}Th, and ^{40} K were determined. Whereas ^{226}Ra and its daughters are expected to reach a secular equilibrium prior to the gamma spectrum [13, 14].

Table 1 Radionuclide energy and source efficiency

Radionuclide	Energy (keV)	Efficiency (%)
^{60}Co	1173.0	0.00603
^{60}Co	1332.50	0.00528
^{137}Cs	661.66	0.01383
^{241}Am	59.54	0.040

Measurements and system control

The radioactivity measurements were performed using the γ-ray spectroscopy equipped with NaI (Tl) detector having 1024 channels. The spectra were recorded at every 20,000 s. The obtained spectral data were analyzed using computer software. The positive operating voltage of 670 V(DC), 60% of efficiency, and resolution of 7.5% at the peak of 662 keV were used. The detector was inserted with a lead shield to reduce from the background radiation. The standard radioactive sources were employed to calibrate the system and to calculate the efficiency of the detector. This research focused on conventional sources such as ^{60}Co, ^{137}Cs, and ^{241}Am to calibrate energy lines because energy, channel number, and efficiency are roughly linear, as shown in Table 1. A Marinelli beaker of one liter filled with ^{152}Eu standard source was used to determine absolute efficiency of the detector. The concentration of the radionuclides including *K-40*, *Ra-226,* and *Th-232* in the prepared cement samples was determined from the recorded spectra, where the analysis of *K-40* was based on its single peak at 1460 keV. The peak of *Th-232* was observed at 911.16 keV of *Ac-228* and 2615 keV of *TI-208*. The concentration of *Ra-226* was determined using the gamma-lines of *Pb-214* at 351.92 keV and *Bi-214* at 609.32 keV [15, 16].

Radiation indices evaluation

I. *Absorbed dose rate in cement*

There is concern that some of the buildings will expose the entire body to excessive radiation doses from gamma rays emitted by ^{214}Pb and ^{214}Bi progeny of ^{226}Ra and ^{232}Th decay chains, and ^{40} K also contributes to the total body radiation dose. The radiation exposure induced by the radionuclides present in cement can be defined in terms of many parameters to determine any radiological hazard. The absorbed dose rate in the air at a height of one meter above the ground is a direct relationship between the radioactivity concentrations of natural radionuclides and their rate of exposure. The values of D were estimated via the relation [13]:

$$D = 0.462A_{\mathrm{Ra}} + 0.604A_{\mathrm{Th}} + 0.042A_K \tag{1}$$

where D is absorbed dose rate in cement, A_{Ra} , A_{Th}, andA_K are the corresponding activity concentrations of *U-238, Th-232*, and *K-40.* where the dose conversion coefficients (nGyh^{-1} per Bqkg^{-1}) for ^{226}Ra, ^{232}Th, and ^{40} K are 0.462, 0.604, and 0.0417, respectively.

II. *Radium equivalent activity (Ra_{eq})*

The real activity level of *Ra-226*, *Th-232*, and *K-40* in the cement samples was due to the inhomogeneous distributions of the natural radionuclides in the soils which were obtained using the common radiological index $\mathrm{Ra_{eq}}$ given by the expression [17]:

$$\mathrm{Ra_{eq}} = A_{\mathrm{Ra}} + 1.43A_{\mathrm{Th}} + 0.077A_k \tag{2}$$

where the symbols have their usual meaning.

III. *Activity concentration index (ACI)*

It is convenient to provide the ACI because more radionuclides can contribute to the measured doses. The radiative forcing of the building materials was assessed by calculating the ACI following the expression [18]:

$$\mathrm{ACI} = A_{\mathrm{Ra}}/300 + A_{\mathrm{Th}}/200 + A_k/3000 \tag{3}$$

where the symbols have their usual meaning.

IV. *Annual effective dose equivalent (AEDE)*

The effective annual outdoor dose was estimated using the equation:

$$\mathrm{AEDE}\left(\frac{\mu Sv}{y}\right) = D\left(\frac{nGy}{h}\right) \times 8760\left(\frac{h}{y}\right) \times 0.2 \times 0.7\left(\frac{\mathrm{Sv}}{Gy}\right) \times 10^{-3} \tag{4}$$

where D refers to dose rates are absorbed in the air, the conversion factor of the absorbed dose rate in the air is equal to 0.7, and the factor of outdoors effective dose occupancy is 0.2 [5].

V. *External and internal hazard index (H_{ex}& H_{in})*

The indices H_{ex} and H_{in} are also called the representation level index, the value of this indicator H_{ex} must be less than one unit to conform to the maximum radium equivalent dose (370 Bq/kg) to keep the radiological hazard negligible. Furthermore, internal exposure to alpha particles from radon inhaled through construction materials can harm the lungs and other respiratory organs. This indicator's value must be less than one unit for its significance to be neglected. It demonstrates that the radiation dose is less than the 1 $\mathrm{mSv{\cdot}y^{-1}}$ limit set by the International Atomic Energy Agency [19], and it is calculated from the following two equations [11]:

$$H_{\mathrm{ex}} = \frac{A_{\mathrm{Ra}}}{370} + \frac{A_{\mathrm{Th}}}{259} + \frac{A_K}{4810} \leq 1 \tag{5}$$

$$H_{\mathrm{in}} = \frac{A_{\mathrm{Ra}}}{185} + \frac{A_{\mathrm{Th}}}{259} + \frac{A_K}{4810} < 1 \tag{6}$$

VI. *Alpha index($I\alpha$)*

This index is necessary to calculate exposure to alpha radiation from the inhalation of building materials. *Ra-226* activities should not exceed the limit of 200 Bq/Kg. To avoid exposure to the internal radon concentration of more than 200 $\mathrm{Bq{\cdot}m^{-3}}$. According to several countries throughout the world, the recommended exemption, and the highest concentration of *Ra-226* in building materials are 100 and 200 Bq/kg, respectively. *Ra-226* is recommended to have a maximum activity concentration of 200 Bq/kg, where $I\alpha < 1$. The alpha index was determined via the following formula [20, 21].

$$I\alpha = \frac{A(\mathrm{Ra})}{200} \tag{7}$$

where A (Ra) is the *Ra-226* activity concentration activity (Bq/Kg) in the cement specimens.

VII. *Excess lifetime cancer risk (ELCR)*

The values of ELCR for the studied cement specimens were calculated by considering a linear relationship to the doses without a threshold according to the international radiation protection commission [22, 23] practice. For the low dose limit, the value of the fatal cancer risk factor of the ICRP is 0.05 Sv^{-1}. The cancer risk was estimated using the relations [24, 25].

$$\mathrm{Risk} = \mathrm{Dose(Sv)} \times \mathrm{Riskfactor}\left(\mathrm{Sv}^{-1}\right) \tag{8}$$

$$\mathrm{Risk} = \mathrm{Dose(Sv)} \times 70(\mathrm{yr}) \times 0.05\left(\mathrm{Sv}^{-1}\right) \tag{9}$$

where the estimated cumulative annual dose exposure for the lifetime is 70 yr.

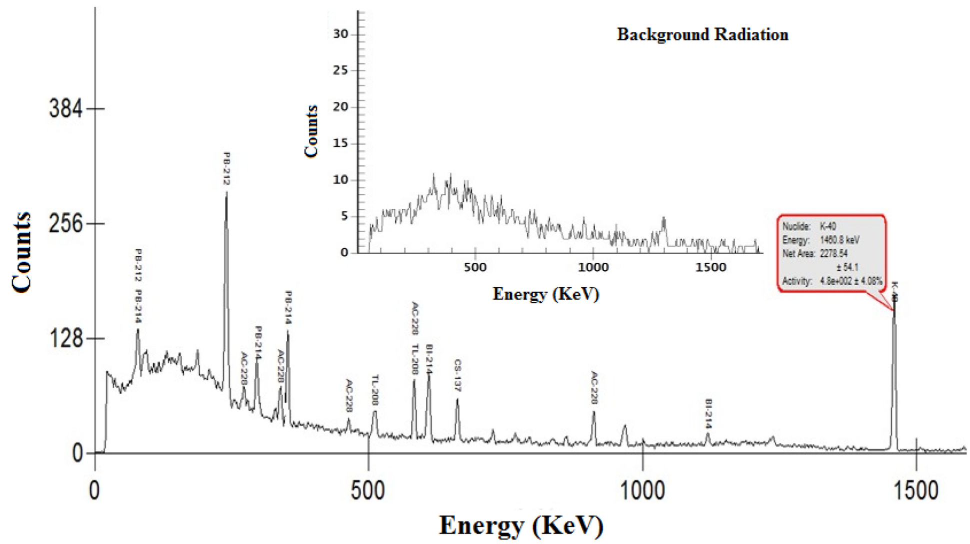

Fig. 1 Radioactivity spectrum of CSu8 sample (Inset: spectrum background)

Table 2 The radioactivity concentration of *U-238*, *Th-232*, and *K-40* in the cement specimens of Iraq

Activity Concentration (Bq/kg)				
Samples number	Sample code	U-238	Th-232	K-40
1	1CK	55.75 ± 9.12	10.78 ± 1.30	171.39 ± 6.47
2	CKr2	27.60 ± 2.23	7.20 ± 0.86	182.11 ± 9.45
3	CKrb3	26.41 ± 4.43	10.70 ± 1.57	141.30 ± 8.62
4	CKsh4	12.38 ± 2.62	7.66 ± 0.97	175.37 ± 7.12
5	5Cal	33.90 ± 5.73	6.70 ± 1.14	172.19 ± 6.81
6	CGe6	23.86 ± 2.95	6.29 ± 0.61	155.56 ± 5.74
7	7Can	29.56 ± 3.07	28.3 ± 7.21	185.26 ± 10.25
8	CSu8	41.37 ± 6.57	57.54 ± 8.64	266.37 ± 11.32
9	CB9	23.72 ± 2.88	17.82 ± 3.52	112.63 ± 8.75
10	CF10	38.42 ± 4.96	6.72 ± 2.13	120.27 ± 5.41
	Average	31.29 ± 4.45	15.97 ± 2.79	168.24 ± 7.99

Results and discussion

The detection limit and accuracy of low activity measurements are influenced by the system's background. The background should be free of natural radionuclide spectral lines in the system components and surroundings. For each sample type, the back count should be repeated, and the background spectrum should be deleted from each spectrum. Background counting must be done on a regular basis for each sample type, and the resulting background spectrum must be subtracted from each sample spectrum (Fig. 1, Inset). In this work, the radioactivity concentration levels of the radionuclides *U-238 (Ra-226), Th-232 (Ra-228),* and *K-40* were assessed as shown in Table 2. Through the results presented in the table below, it was found that the highest value of *Ra-226* concentration is 55.75 Bq/kg in Sample No.1 and that the lowest value is 12.38 Bq/kg and was found in Sample No.4 and that its average value is 31.29 ± 4.45 Bq/kg (Fig. 2). About 60% of the recorded values from *Ra-226* were lower than the global average value of 32 Bq/kg according to UNSCEAR (2000) [26]. For the *Th-232* series, it was found that the highest value of *Ra-228* is 57.54 Bq/kg in Sample No.8 and the lowest value was 6.29 Bq/kg in Sample No.6 with an average value of 15.97 ± 2.79 Bq/kg (Fig. 2). These values are within the internationally recommended limits of 45 Bq/kg (except sample No.8) of the UNSCEAR (2000). For Sample No. 9 the level of *K-40* was the lowest (112.63 Bq/kg) and for Sample No. 8 it was highest (266.37 Bq/kg) with an average of 168.24 ± 7.99 Bq/kg. Also, despite the wide variance in the *K-40* activity level 112.63–266.37 Bq/kg, 100% of the samples (Fig. 2) showed a value less than the global average of 420 Bq/kg in accordance to the UNSCEAR (2000), radioactivity spectrum of CSu8 sample as depicted in the Fig. (1). This variation was primarily attributed to the wide geological variation in the extraction and processing of the raw materials and chemicals that are involved in cement production [27].

(i) *Absorbed dose rate*
If the activity concentrations of the radionuclides in the soil are detected by assuming their uniform distributions, then the dose of air exposure caused by these isotopes can be estimated. The calculated values were found to be lower compared to those reported in the state-of-the-art literature (Table 3). In addition, the average absorbed dose for the proposed samples was 59.947 nGy·h^{-1} which is lower than (84 nGy·h^{-1}) the one referred by UNSCEAR (2000) as illustrated in Fig. 3 [28, 29].

(ii) *Radium equivalent activity*
The activity of radium equivalent describes the risks associated with the use of buildings and other construction materials that contain *U-238, Th-232*, and *K-40.* The overall activity levels for *U-238, Th-232*, and *K-40* must be measured when the corresponding γ-rays are approximately equivalent to 10, 7, and 130 Bq/kg. The equivalent activity of Ra_{eq} in the cement samples was calculated (Table 3 and Fig. 4) which was found in the range of 36.845 to 144.162 Bq/kg with an average of 67.101 Bq/kg. The achieved values for the studied cement specimens were less than the one published by UNSCEAR (2000) which is 370 Bq/kg. It was concluded that the annual effective dose limits of the cement samples did not exceed the recommended limit of 1 mSv [30].

(iii) *Activity Concentration Index*
Table 3 and Fig. 5 show the results of this indicator wherein the obtained values were ranged from 0.138 to 0.514 with an average of 0.240. For the specimens in large quantities the value of ACI < 1 corresponded to the absorbed γ ray dose rate of *1* mSv·y^{-1}. In this work, for all samples, the obtained value of the activity concentration index was below the *γ* index limit [9, 25].

(iv) *Annual effective dose equivalent (AEDE)*
Table 3 displays the values of the *AEDE* against the safety standard for the studied cement samples. The average annual effective dose in the studied cement samples was 0.220 mSv·y^{-1} which is below the recommended limit. The effective dose rates for all samples did not exceed the recommended value of 1 mSv·y^{-1}. Thus, all the cement samples analyzed in this work agreed well with the public safety standard [4, 31].

(v) *External and Internal Hazard Index*
The external risks provide the hazard assessment of the natural gamma radiation to ensure that the effective dose of such rays does not exceed the accepted limits of 2.5–3.0 mSv·y^{-1}. The values of this indicator (H_{ex}) are presented in Table 4 and Fig. 6 which were ranged from 0.033 to 0.150 with an average of 0.084. The obtained values are lower than the index level of this indicator set by the international standard. This indicator must have a value below one unit to achieve the permissible radium dose equivalent of 370 Bq/kg to keep the radiological hazard insignificant [32, 33]. The inhalation of the alpha particles released by the short-lived isotopes like radon and thoron is very hazardous. The internal radon gas exposure is one of the most hazardous causes of lung diseases that ranged from 0.169 to 0.556 with an average of 0.3. The value of this indicator must be less than one unit to remain negligible where the radiation dose is the lowest value of the recommended limit (IAEA 2007) as shown in Table 4 and Fig. 6 [18, 34].

(vi) *Alpha Index*
The alpha index in the samples was determined using a concentration of *Ra- 226* activity. Table 4 and

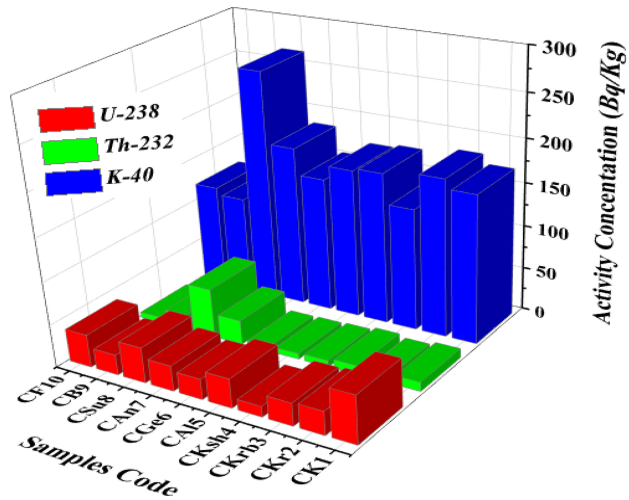

Fig. 2 Radioactivity concentration of *U-238*, *Th-232*, and *K-40* the studied cement specimens

Table 3 The values of the absorbed dose rates, radium equivalent activity, Activity concentration index, and annual effective dose of the studied cement samples

Samples code	*D* nGy·h^{-1}	Ra_{eq} Bq/k^1	ACI	AEDE mSv·y^{-1}
CK1	76.979	84.362	0.296	0.283
CKr2	48.008	51.918	0.189	0.176
CKrb3	47.470	52.591	0.188	0.174
CKsh4	33.975	36.845	0.138	0.125
CAl5	52.453	56.739	0.203	0.192
CGe6	41.423	44.832	0.162	0.152
CAn7	73.352	84.394	0.302	0.269
CSu8	122.850	144.162	0.514	0.451
CB9	50.513	57.875	0.205	0.185
CF10	52.444	57.290	0.201	0.192
Average	59.947	67.101	0.240	0.220

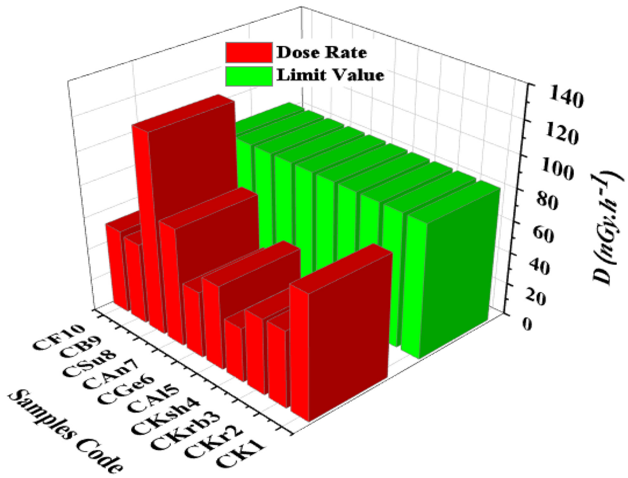

Fig. 3 Absorbed dose rate and limits of the cement samples

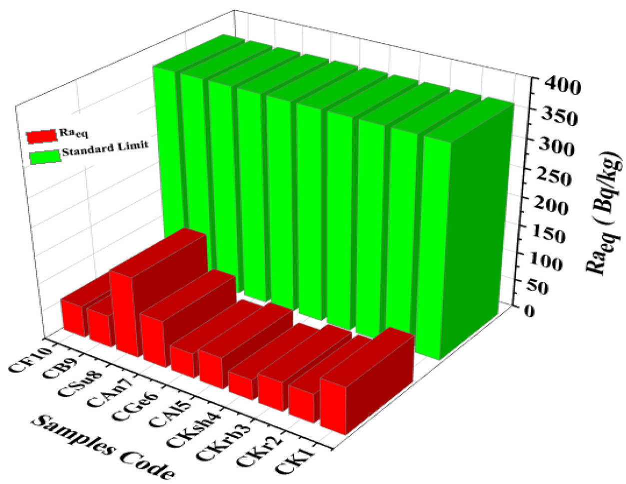

Fig. 4 Radium equivalent activity (Ra_{eq}) of samples and standard limit of cement samples

Fig. 7 indicated that the radioactivity concentration of *Ra-226* for all the cement specimens was below the stipulated standard of 100 Bq/kg. The values of the alpha index for the studied cement samples were ranged from 0.061 to 0.278 with the mean value of 0.156. Assessment of the internal hazard, originating from the alpha activity of building materials, requires calculations of the alpha index or internal hazard index. The alpha indices have been proposed to assess the exposure level due to radon inhalation originating from building materials [12]. *Ra-226* is advised to have a maximum activity concentration of 200 Bq/kg, where $I\alpha < 1$. Also, it was affirmed that the exhalation of radon from the cement samples can cause an internal concentration below these limits [21, 35].

(vii) *Excess lifetime cancer risk*

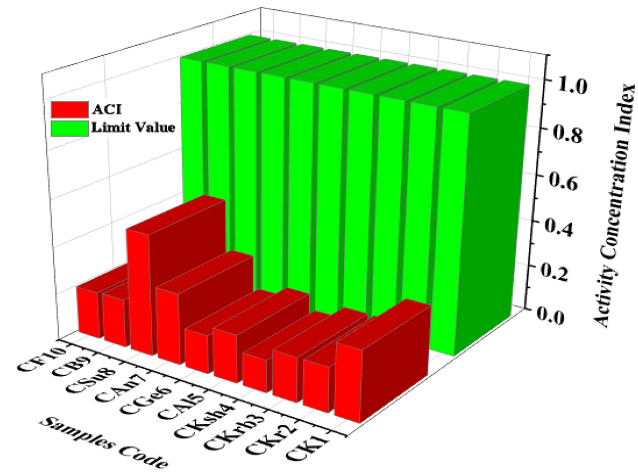

Fig. 5 Activity concentration index of samples and limit the value of cement samples

Table 4 The values of hazard indices, alpha index, and excess lifetime cancer risk of all the cement specimens

Sample code	Hex	Hin	$I\alpha$	ELCR $\times 10^{-3}$
CK1	0.150	0.414	0.278	0.991
CKr2	0.074	0.252	0.138	0.618
CKrb3	0.071	0.242	0.132	0.611
CKsh4	0.033	0.169	0.061	0.438
CAl5	0.091	0.280	0.169	0.675
CGe6	0.064	0.217	0.119	0.533
CAn7	0.079	0.346	0.147	0.944
CSu8	0.111	0.556	0.206	1.581
CB9	0.064	0.243	0.118	0.650
CF10	0.103	0.283	0.192	0.675
Average	0.084	0.300	0.156	0.771

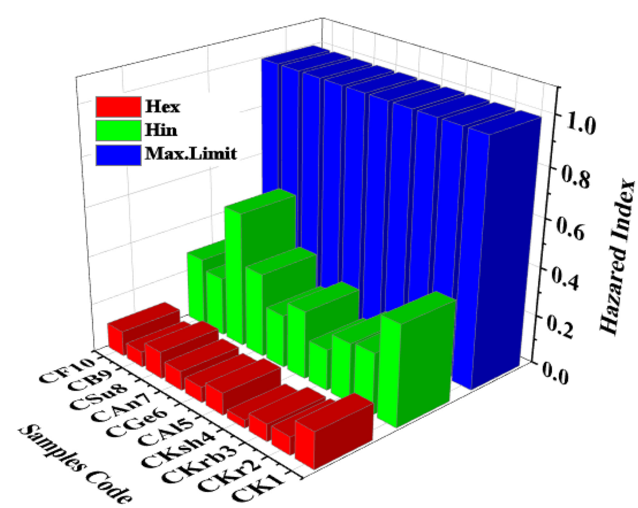

Fig. 6 The external and internal hazard index of the cement samples together with their maximum allowed limit

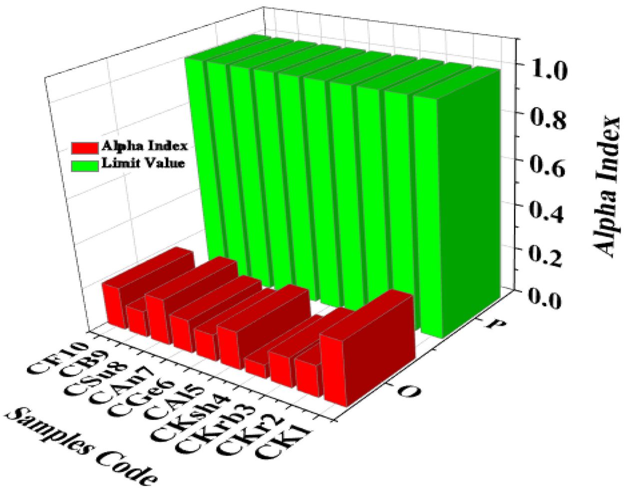

Fig. 7 Alpha and limit index of the studied cement samples

The values of the calculated cancer risk level were ranged from 0.438×10^{-3} to 1.581×10^{-3} with the mean value of 0.771×10^{-3}. Overall, these values were due to external radiation exposure where 0.7% of the fatal cancer cases may be associated with this type of irradiation. Table 4 enlists the cancer risks to the Iraqi populations caused by the studied cement specimens when used in the construction sectors [36].

Conclusion

To achieve stability, construction material can cause high indoor and outdoor γ-radiation dose exposure to humans depending on the presence of the concentration of the naturally occurring radionuclides. In fact, heavy isotopes of the radionuclides are potentially unstable and hazardous to the people because these radionuclides emit inuring radiations or particles to achieve stability. In this perception, this paper made a radiometric assessment of 10 cement samples used by various construction sectors in Iraq. The measured mean radioactivity levels of *Ra-226*, *Ra-228,* and *K-40* for the studied cement samples were 31.29, 15.97, and 168.24 Bq/Kg, respectively. The values of the *Ra-226*, *Ra-228*, and *K-40* were lower than the standard set by UNSCEAR (2000) wherein the average global values are 32, 45 and 420 Bq/Kg, respectively. The obtained results were compared with the recently reported values in the literature where the values of radium equivalent were used. The radium equivalent level shown by the CSu8 specimen was the highest (144.162) Bq/Kg and CKsh4 sample was the lowest (36.845 Bq/Kg) with the mean value of 67.101 Bq/Kg which was much below the recommended guideline of 370 Bq/Kg. Furthermore, CKsh4 and CSu8 specimens revealed the minimum and maximum absorbed dose rates of 33.975 $nGy \cdot h^{-1}$ and 122.850 $nGy \cdot h^{-1}$, respectively. The later one exceeded the global average of 84 $nGy \cdot h^{-1}$ cited by UNSCEAR 2000. About 90% of the cement samples displayed radioactivity levels below the world average. The mean value of the activity concentration index, annual effective dose, external and internal hazard index, alpha index, and excess lifetime cancer risk were discerned to be, 0.240, 0.220, 0.084, 0.30, 0.156, and 0.771×10^{-3} $mSv \cdot y^{-1}$, respectively. All the obtained values were within their world averages set by the UNSCEAR (2000). Based on the analyzes and comparison of the results, it is established that the cement manufactured in Iraq is safe for building construction without any risk of radiation exposure.

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Declarations

Conflict of interest The following authors have affiliations with organizations with direct or indirect financial interest in the subject matter discussed in the manuscript:

Ethical approval All authors have participated in (a) conception and design, or analysis and interpretation of the data; (b) drafting the article or revising it critically for important intellectual content; and (c) approval of the final version.

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