

## Performance, Measurements and Potential Radiological Risks of Natural Radioactivity in Cements Used in Jordan

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### ABSTRACT

This study examined the cement properties and potential radiological risks of natural radioactivity in cements used in Jordan. The results showed that all cement types were compliant with the Jordanian and European standard specifications (JSS. 30-1/2007) and (EN. 19-1/2000), respectively. The maximum value of compressive strength was found in white Portland cement. Ambient temperature for all samples occurred between 330 min and 500 min and was released with approximately the same rate values. The activity concentration of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K were detected in all cement samples. The maximum value of activity concentrations (in Bq/kg ± SD) of <sup>226</sup>Ra was 79.52 ± 4.67, while it was for <sup>232</sup>Th and <sup>40</sup>K 30.99 ± 2.85 and 354.70 ± 19.64, respectively. The highest value of the dose rate was 117 ± 8 nGy/h. The annual effective dose due to absorption and radium equivalent crest values were 574 ± 8 μSv/h and 343 ± 16 Bq/kg, respectively. The external hazard, activity concentration index and alpha index were less than unity for all samples. All measurements and calculations were within the recommended international standard values (ISVs), except for S3 & S5 samples. The results were compared with those of other studies from different countries all over the world.

**KEYWORDS:** Cement, Compressive strength, Gamma ray spectroscopy, Ambient temperature, Natural radioactivity, Radiation hazard parameters.

### INTRODUCTION

Cement is considered as one of the most important building materials around the world. It is used in different types of construction: houses, offices, factories, shops, ... etc. in every city or town. There are different types of cement related to the composition percentages.

Usually, cement is composed of natural sources (e.g. soil and rock), industrial products (e.g. phosphate,

oil industry) and waste products (e.g. minerals, slags, oil shale, coal fly ash, ... etc.).

Five different types of cement from Lafarge Cement Company were used in this study. Ordinary Portland cement is the most widely used type of cement in concrete construction, with no exposure to sulfates in soil and groundwater (Neville and Brooks, 2010). White Portland cement is used for agricultural purposes. Due to its low content of soluble alkalis, white concrete sometimes requires a pastel white finish to prevent staining, particularly in tropical regions. White cement contains small amounts of iron oxide and manganese

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oxide, together with chalk or limestone, free of specified impurities. Special precautions are required during the grinding of clinker to avoid contamination. Therefore, the cost of white cement is high (twice that of ordinary Portland cement) (Neville and Brooks, 2010).

Portland fly ash cement is the most common type of pozzolan. There are two sub-classes recognized by BS EN 197-1: 2000: Class IIA has a fly ash content of 6 to 30 percent and Class IIB, which is used in this study, having a fly ash content of 21 to 35 percent (Neville and Brooks, 2010). Portland-pozzolanic cements are made by blending pozzolans with Portland cement. Pozzolans are described as siliceous or siliceous and aluminous materials which possess little or no cementitious value, but will, in finely divided form and in the presence of moisture, chemically react with lime (liberated by hydrating Portland cement) at ordinary temperatures to form compounds possessing cementitious properties. The pozzolan content is limited to between 15 and 40 percent of the total mass of the cementitious material (Neville and Brooks, 2010).

Blast-furnace cement is made by blending Portland cement clinker with granulated blast-furnace slag, which is a waste product in the manufacture of pig iron.

Slag contains lime, silica and alumina, but not in the same proportions as Portland cement. Its composition can vary a great deal. Portland blast-furnace cement can be referred to as slag cement. The amount of slag should be between 25 and 70 percent of the mixture's mass or between 50 and 85 percent for the manufacturing of low-heat Portland blast furnace cement. The maximum lime/silica ratio is 1.4 and the ratio of the mass of CaO plus MgO to the mass of SiO<sub>2</sub> must exceed 1.0. Typical uses are in mass concrete and in sea-water construction because of a better sulfate resistance (due to a lower C<sub>3</sub>A content) than ordinary Portland cement. Blast-furnace cement is commonly used in countries where slag is widely available (Neville and Brooks, 2010).

Cement contains natural radionuclides, since it consists of soil and rock. These radionuclides are mainly uranium <sup>238</sup>U, thorium <sup>232</sup>Th and <sup>40</sup>K; a radioactive isotope of potassium (Khan, 2003). The most important radiological part in <sup>238</sup>U chain starts from radium <sup>226</sup>Ra, which makes <sup>226</sup>Ra the interesting radionuclide, instead of <sup>238</sup>U (Turhan, 2008). Measuring the activity concentrations of these radionuclides from different sources becomes more important for researchers to

investigate their risks on human health, which can lead to various types of cancer, such as osteosarcoma (bone cancer), lung carcinoma (lung cancer), ... etc., as well as other disorders in the body's systems.

The level of natural radioactivity in cement varies according to its constituents and their percentages, beside its high dependence on the characteristics of raw materials and their geological sources (Turhan, 2008). Therefore, the periodic study of cement radioactivity is essential in determining radiological parameters and estimating their hazards to human health.

Several studies have measured activity concentrations of natural radionuclides and examined their effects on human health in different samples, such as: food, soil, water and detergent powders (Hamadneh et al., 2017; Okoor et al., 2019), using gamma ray spectroscopy.

The goal of this study is to study the performance of five different types of cement used in Jordan, as well as to detect gamma radiation from <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K using gamma ray spectroscopy in these samples and evaluate the radiant hazard factors and the annual effective dose. All measurements are compared with the recommended international standard values all over the world reported by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000), Organization for Economic Cooperation and Development (OECD 1979) and the report of the European Commission (EC, 1999). Results for different samples manufactured in different factories (Arabia Company, Ashamaliya Company and Al-Rajhi Company) were mentioned to compare with the present study's results.

The tests in this study were carried out to determine the mechanical properties and to study the hazard of contamination of structures of cement types. Therefore, the engineer can take the decision to choose a recommended type of cement with good mechanical properties, low cost and less radiation.

## MATERIALS AND METHODS

### Cement Types

Fig. 1 shows the occurrences of cement raw materials in different locations in Jordan. These locations are distributed throughout Jordan in the northeast (Zarqa) and in the south (Tafila and Ma'an).

Five different types of cement from Lafarge Cement Company in compliance with Jordanian standard

specifications (JSS. 30-1/2007) and European standards (EN. 19-1/2000) were used in this study as a binder. Oxide rates, percentages of compound composition (calculated by Bogue equations) and compressive strengths of these types are shown in Table 1.

Locally available natural sand coming from a quarry

located at the Jerash province with particles smaller than 2mm, a fineness modulus of 2.25 and a specific gravity of 2.60 g/cm<sup>3</sup>, complying with Jordanian standard specifications (JSS. 96/1987), was used for mortars.

The water utilized came from clean and fresh potable supplies.

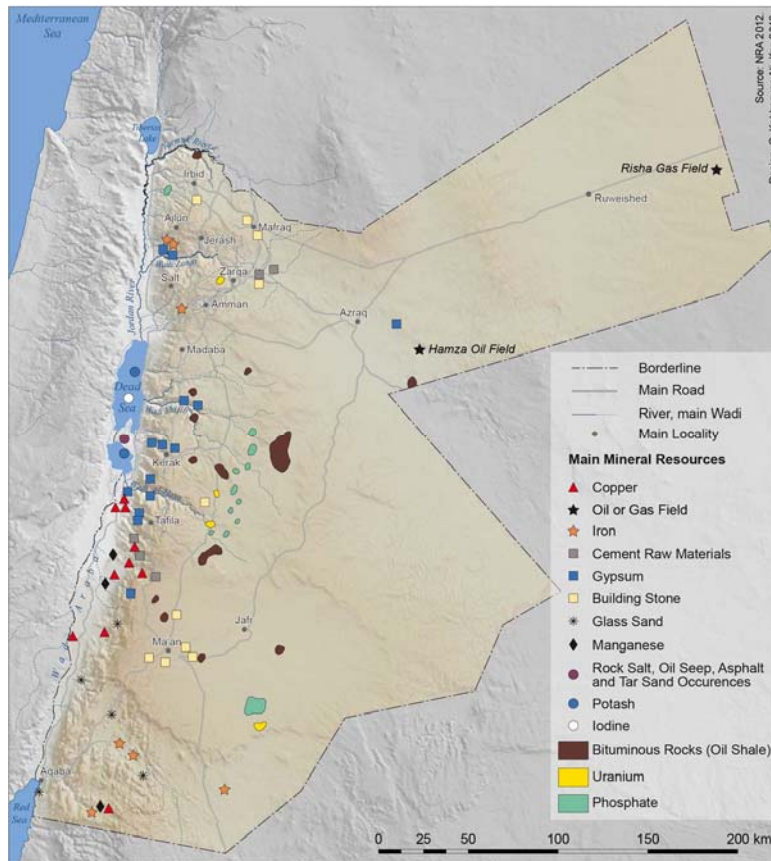


Figure (1): Locations of main mineral resources in Jordan (Atlas of Jordan, 2014)

Table 1. Cement sample types, description, compositions and number of collected samples

| Cement Type       | Description                | No. of Collected Samples | CaO | SiO <sub>2</sub> | Al <sub>2</sub> O <sub>3</sub> | Fe <sub>2</sub> O <sub>3</sub> | MgO | SO <sub>3</sub> | K <sub>2</sub> O | others | C <sub>3</sub> S | C <sub>2</sub> S | C <sub>3</sub> A | C <sub>4</sub> AF | Compressive Strength (MPa) |
|-------------------|----------------------------|--------------------------|-----|------------------|--------------------------------|--------------------------------|-----|-----------------|------------------|--------|------------------|------------------|------------------|-------------------|----------------------------|
| CEM I 52.5 N      | Ordinary Portland cement   | 5                        | 63  | 20               | 6.0                            | 3.0                            | 1.5 | 2.0             | 1.0              | 3.5    | 54               | 17               | 11               | 9                 | 55 to 61                   |
| CEM I 52.5 N      | White Portland cement      | 3                        | 67  | 22               | 2.5                            | 2.1                            | 2.0 | 3.0             | 1.2              | 0.2    | 77               | 5                | 3                | 6                 | 52 to 65                   |
| CEM II B-V 42.5 N | Portland fly ash cement    | 3                        | 70  | 12               | 5.0                            | 4.0                            | 4.5 | 3.0             | 1.0              | 0.5    | 65               | 15               | 10               | 10                | 48.5 to 60                 |
| CEM II B-P 42.5 N | Portland Pozzolanic cement | 5                        | 54  | 24               | 6.3                            | 5.0                            | 4.8 | 3.1             | 0.9              | 1.9    | 59               | 16               | 12               | 8                 | 45.5 to 51.7               |
| CEM III B 42.5 N  | Blast-furnace cement       | 3                        | 51  | 27               | 10.4                           | 2.3                            | 0.8 | 1.8             | 0.2              | 6.5    | 56               | 15               | 12               | 8                 | 36.3 to 47                 |

### **Paste and Mortar Preparation**

All pastes were prepared with a (W/C) ratio according to standard consistency of plain cement specimens by Vicat Apparatus according to ASTM C187-04.

650 g of cement had been mixed with a measured quantity of water which conformed to the numerical limits of specification. The cement paste was prepared as described and quickly formed into an approximate sphere with gloved hands. Then, it was tossed six times through a free path of about 150 mm (6 in.) from one hand to another to produce a nearly spherical mass that may be easily inserted into the Vicat ring with a minimum amount of additional manipulation. The ball, resting in the palm of one hand, was pressed into the larger end of the conical ring, held in the other hand, completely filling the ring with paste. The excess at the larger end was removed by a single movement of the palm of the hand. This process must not exceed a period of 30 seconds after the completion of mixing. Trial pastes with varying percentages of water were made until this normal consistency is obtained (ASTM C187-04).

Fresh mortars were prepared with a C/sand ratio of 0.3 and a water/cement (W/C) ratio of 0.50 by weight according to ASTM C109/C109M-02.

For all pastes and mortars, water was added to cement and mixed for about 2 min. After mixing, fresh pastes were cast at Vicat's mould to determine the setting times.

For mortar specimens, cement and sand were first mixed in dry conditions for 2 min before water was added, after which they were mixed for an additional time of 3 min. Flow table measurements were carried out immediately after mixing (ASTM C230/230M-08).

After mixing, the flow mold was placed at the center of the flow table. A layer of mortar of about 25 mm (1 in.) thickness was placed in the mold and tamped 20 times with the tamper. The tamping pressure should be just sufficient to ensure uniform filling of the mold. Then, the mold with mortar and tamp was filled as specified for the first layer. The mortar was cut off to a plane surface flush with the top of the mold by

drawing the straight edge or the edge of the trowel with a sawing motion across the top of the mold. The table top is cleaned, being especially careful to remove any water from around the edge of the flow mold. The mold is lifted away from the mortar one minute after completing the mixing operation in Fig. 2 (ASTM C1437-07).

Immediately, the table was dropped 25 times in 15 s, unless otherwise specified. If using the caliper specified, the diameter of the mortar is measured along the four scribe lines and recorded as the number of caliper divisions (estimated to one tenth of a division). If a non-specified caliper is used, the diameter of the mortar is measured along the four lines scribed and recorded to the nearest millimeter. The flow is the resulting increase in average base diameter of the mortar mass, expressed as a percentage of the original base diameter (ASTM C1437-07).

After flow testing, mixed mortars consisting of 1 part cement and 2.75 parts sand, proportioned by mass and water content to obtain a flow of  $110 \pm 5$  in 25 drops of the flow table, were placed into compressive test cubes of two-inch or (50-mm) size and compacted by tamping in two layers. The cubes were left to sit in the molds for one day, stripped and immersed in water until being tested.

Vibration table was used for compaction. The specimens were demolded after 24 h, weighed and measured dimensionally before curing in fresh water at  $23 \pm 2^\circ\text{C}$ . Fig. 3 shows the Ambient temperature measurements.

Five mortar specimens were cast in two-inch (50-mm) test cubes to determine the ambient temperature every 15 minutes for the first 24 h after casting. "DM 6801A+" digital thermometers with Type-K thermocouples were used to measure the mortar ambient temperature. Thermometers are connected to the thermocouples inserted in the middle of the mortar tube (i.e., 2.5-cm depth). A digital stopwatch was used to measure the time that elapsed after the first contact between the cement and water. Software implemented on a chatting camera recorded the measurements during the first 24 h after casting (as shown in Fig. 3).



Figure (2): Flow table for mortar workability



Figure (3): Ambient temperature measurements

### Mortar Absorption

Moisture plays a critical role in cement mortars; it is required to hydrate the cement particles to develop appropriate mechanical and permeability properties to withstand the applied loads of environmental action. However, in almost every degradation mechanism of mortar, water is present. There are standard methods to measure the moisture condition in mortar, such as relative humidity probes, etc... (Hewlett, 2004).

Absorption is one of the methods which could be used as an indicator of water content. To determine the mortar absorption, five two-inch (50-mm) test cubes were cast. A Heraeus high temperature-drying cupboard oven with downward door opening and analog display (as shown in Fig. 4) was used in this test.

After 28 days of curing, each cube weight ( $W_1$ ) was measured before being placed in the oven under  $110^\circ\text{C}$  to a period for 24 h to completely remove remaining free water.

After 24 h, the cube was extracted from the oven by dragging the hollow plate with a lab tong and picking up

using a thermal lab glove.

The cube weight ( $W_2$ ) was immediately measured after extraction by a digital scale with 0.02 g accuracy.

A hollow steel plate, four sheets of thermal blocker and two ceramic plates were installed above the scale to prevent the temperature of the cube from affecting the measurement.

Absorption was calculated as  $\Delta W = W_1 - W_2$ .



Figure (4): High-temperature cupboard oven

### Radioactive Measurement

Nineteen samples of five different types of cement were collected. Each sample was air-dried for two days and oven-dried at about 105<sup>0</sup> C to remove remaining moisture. Next, the sample was cooled at room temperature in a moisture-free place (Baykara, 2011). Finally, the samples were filled in standard cylindrical Marinelli beakers and sealed for 5 weeks to ensure secular equilibrium between the radionuclides <sup>226</sup>Ra, <sup>222</sup>Rn and their short-lived radionuclide daughters.

Gamma analysis was performed using gamma-ray spectroscopy. Samples were measured in the Jordanian Atomic Energy Commission (JAEC) laboratories. A high-purity germanium detector (HPGe, 50% relative efficiency) with a beryllium window, a built-in ADC and a multichannel analyzer coupled with Genie 2000 software is used. The method of this examination has been ISO-17025 accredited since 2012 at JAEC.

The activity concentration (A) was calculated using Eq. (1).

$$A = \frac{R}{\varepsilon(E).m.P_{\gamma}(E)} \quad (1)$$

where, R is the count rate at energy E,  $\varepsilon(E)$  is the efficiency of the detector at energy E, m is the mass of the sample in kilogram and  $P_{\gamma}(E)$  is the gamma yield of the nuclide at energy E.

The activity concentration of <sup>226</sup>Ra was calculated from the gamma lines of its daughters <sup>214</sup>Bi (604.9 keV) and <sup>214</sup>Pb (351.9 keV). The activity concentration of <sup>232</sup>Th was determined by the gamma line of its daughter <sup>228</sup>Ac (911.6 keV). The activity concentration of <sup>40</sup>K was detected directly through its gamma line of 1461.8 keV.

Natural radionuclides in building materials cause an indoor air dose rate (D) due to gamma ray emission, defined in Eq. (2) according to the report of EC 1999 as (EC, 1999):

$$D (nGy/h) = 0.92A_{Ra} + 1.1 A_{Th} + 0.08 A_K \quad (2)$$

where  $A_{Ra}$ ,  $A_{Th}$  and  $A_K$  are the activity concentrations of <sup>226</sup>Ra, <sup>234</sup>Th and <sup>40</sup>K in Bq/kg, respectively.

The annual effective dose (AED) was calculated using Eq. (3) (UNSCEAR, 2000):

$$AED(\mu Sv/y) = D(nGy/h) \times 8760(h/y) \times 0.8 \times 0.7(Sv/Gy) \times 10^{-3} \quad (3)$$

where 0.7 (Sv/Gy) represents the conversion coefficient from the dose rate (D) in air to effective dose and 0.8 is the indoor occupancy factor.

A common operator is used to calculate the radiological hazards related to the radionuclides <sup>238</sup>U, <sup>234</sup>Th and <sup>40</sup>K called radium equivalent ( $Ra_{eq}$ ). It is defined in Eq. (4) (Bereka and Mathew, 1985).

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (4)$$

This value should be less than 370 Bq/kg to be accepted as a safe value (OECD, 1979).

The external ( $H_{ex}$ ) hazard index was calculated using Eq. (5).

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (5)$$

The radiation hazard index is negligible if the value of this index is less than unity (UNSCEAR, 2000).

The activity concentration index (I) was studied to resemble the meeting with the dose criterion by calculating the average of the activity concentrations, as shown in Eq. (6) (UNSCEAR, 2000).

$$I = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \quad (6)$$

The alpha index ( $I_{\alpha}$ ) was calculated using Eq. (7) to determine the excess alpha radiation related to the radon inhalation from building materials (Righi and Bruzzi, 2006).

$$I_{\alpha} = \frac{A_{Ra}}{200 \text{ Bq / kg}} \quad (7)$$

$I_{\alpha}$  value should be less than unity; otherwise, there will be a radon exhalation causing an indoor radon concentration exceeding the recommended limit; 200 Bq.m<sup>-3</sup> (Al-Hwaiti, 2015; EC, 1999; Righi and Bruzzi, 2006; Sharma, 2016).

## RESULTS AND DISCUSSION

### PASTE RESULTS

#### Standard Consistency

Vicat's apparatus was used to determine the water/cement ratio (W/C) for the standard consistency of plain cement paste. Results showed that W/C ratio used in the preparation of all cement material pastes was 0.29.

#### Setting Times

Table 2 summarizes the initial and final setting times for the five different types of cement pastes. The results showed that all cement types are compliant with Jordanian standard specifications (JSS. 30-1/2007) and European standards (EN. 19-1/2000). For ordinary

Portland (Type I) cement, the un-sieved heavy metals have no significant effect on initial and final setting times, and the sieved heavy metals cause a decrease in initial and final setting times (Alqam, 2012).

### MORTAR RESULTS

#### Flow Table

Table 3 shows the variation in the average value of the four measured diameters of the flow table test for the five different types of cement mortars. The average flow diameters complied with Jordanian standard specifications (JSS. 30-1/2007) and European standards (EN. 19-1/2000). The results of the flow table tests indicated that the workability of fresh mortar is improved with increasing heavy metal use (Alqam, 2012).

**Table 2. Setting time variations for different types of cement pastes**

| Sample | Description                | Initial setting time (min) | Final setting time (min) |
|--------|----------------------------|----------------------------|--------------------------|
| S1     | Ordinary Portland cement   | 165                        | 215                      |
| S2     | White Portland cement      | 435                        | 470                      |
| S3     | Portland fly ash cement    | 410                        | 450                      |
| S4     | Portland-Pozzolanic cement | 430                        | 475                      |
| S5     | Blast-furnace cement       | 170                        | 220                      |

**Table 3. Mortar average flow diameters for different types of cement**

| Sample | Description                | Average diameter (mm) |
|--------|----------------------------|-----------------------|
| S1     | Ordinary Portland cement   | 182                   |
| S2     | White Portland cement      | 115                   |
| S3     | Portland fly ash cement    | 160                   |
| S4     | Portland-Pozzolanic cement | 173                   |
| S5     | Blast-furnace cement       | 180                   |

#### Ambient Temperature

Fig. 5 shows the results of ambient temperature in different types of cement. The obtained results indicated that peak values of ambient temperature occurred almost between 330 min and 500 min from the moment of adding water for all cement specimens. Temperature release rate values were almost the same for all cement specimens before and after the peak values.

#### Water Absorption

Table 4 shows the variations of the water absorption percentage for the five different types of cement mortars, which could be attributed to the formation enhancement of hydrated product in case of white Portland cement (WPC type I 52.5N) or the evaporation of free water which filled the voids in case of ordinary Portland cement (OPC type I 52.5N).

#### Compressive Strength

Table 1 shows the cube compressive strength of the

five different cement mortar types at the age of 28 days. The maximum increase in compressive strength of

cement paste reached 350.2 kg/cm<sup>2</sup> for white Portland cement (WPC type I 52.5N).

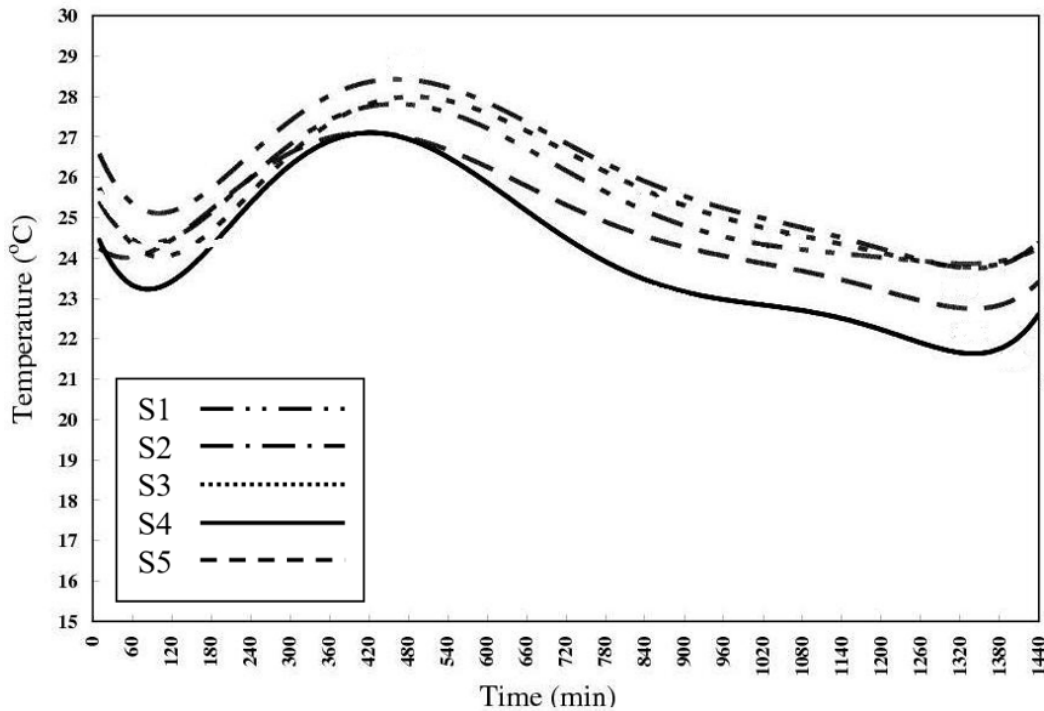


Figure (5): The ambient temperature for different types of cement mortars

Table 4. Cube water absorption variations for different types of cement mortars

| Sample | Description                | Cube Weight (gm) |       |       |                  |       |       | Absorption (%) |
|--------|----------------------------|------------------|-------|-------|------------------|-------|-------|----------------|
|        |                            | Room Temperature |       |       | after 110°C/24 h |       |       | after 110°C    |
| S1     | Ordinary Portland cement   | 270.2            | 270.4 | 273.2 | 263              | 264.2 | 267   | 2.5            |
| S2     | White Portland cement      | 262.8            | 265.6 | 266.3 | 262.3            | 265   | 265.6 | 0.2            |
| S3     | Portland fly ash cement    | 259.6            | 262   | 263.6 | 255.2            | 258.7 | 260   | 1.5            |
| S4     | Portland-Pozzolanic cement | 258.6            | 265.8 | 273.8 | 252.3            | 259.4 | 267.5 | 2.4            |
| S5     | Blast-furnace cement       | 270              | 271.1 | 272.5 | 262              | 265   | 266.8 | 2.5            |

**Activity Concentrations of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K**

Activity concentrations were analyzed for <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K. The results for each radionuclide are shown in Table 5. The values obtained were used to assess the variability and distribution of the related radionuclides in the samples and radiological hazard indices. The three radionuclides were detected in all cement samples and the results varied with the activity concentrations of R, Th and K radionuclides. The activity concentrations (in Bq/kg ± SD) of <sup>226</sup>Ra were between 27.12 ± 1.85 and 79.52 ± 4.67, where the

activities of <sup>232</sup>Th varied from 14.15 ± 1.76 to 30.99 ± 2.85 and <sup>40</sup>K radionuclide concentrations were between 123.02 ± 6.22 and 354.70 ± 19.64.

The European Commission (1999) reported the recommended world-wide average concentration values of the natural radionuclides <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K as 40, 30 and 400 Bq/kg, respectively (EC, 1999). Our data in general is lower than the international reported limits.

The cement sample S5 (Blast-furnace cement) showed the highest value of activity concentration and caused large deviations in the distributions of activity

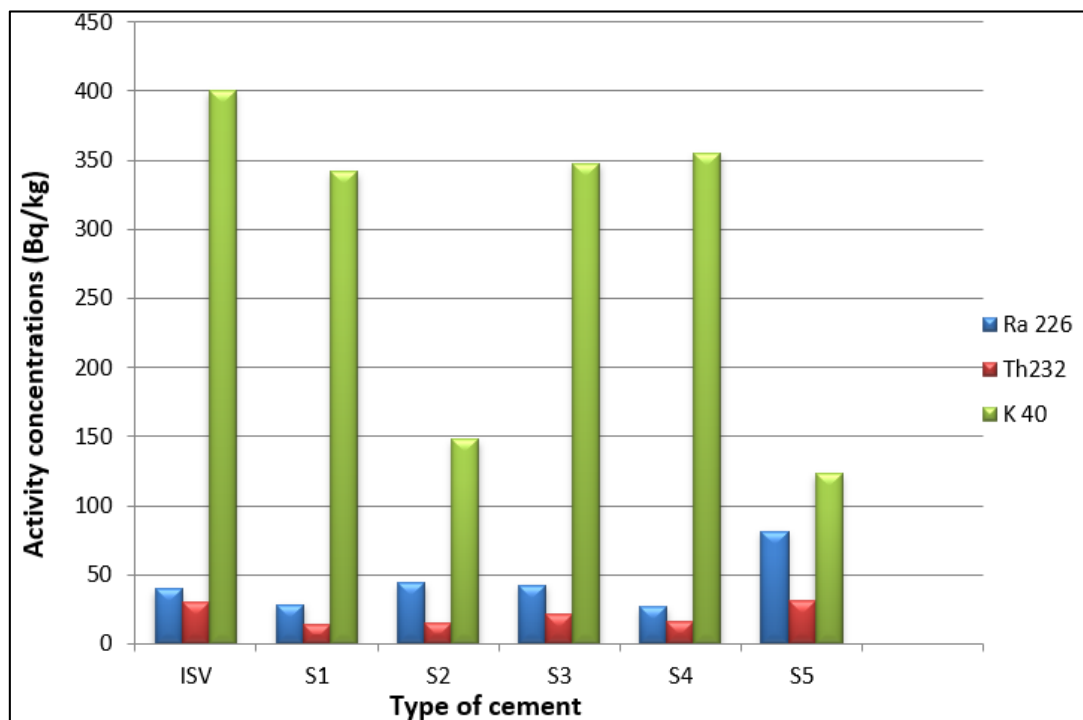


concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ . The mean values of activity concentrations for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in this sample were  $79.52 \pm 4.67$  Bq/kg,  $30.99 \pm 2.85$  Bq/kg and  $123.02 \pm 6.22$  Bq/kg, respectively. From the literature (Turhan, 2008; Paperfthymiou and Gouseti, 2008), it is known that fly ash additives to building materials can

increase  $^{226}\text{Ra}$  activity level, as we have seen the highest  $^{226}\text{Ra}$  activity concentration in this sample. Fig. 6 shows the mean values of natural radioactivity concentrations in all cement types compared to international standard values.

**Table 5. Activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  for the studied cement samples**

| Sample | Activity Concentration (Bq/kg) |                  |                   |                   |                 |                    |
|--------|--------------------------------|------------------|-------------------|-------------------|-----------------|--------------------|
|        | $^{226}\text{Ra}$              |                  | $^{232}\text{Th}$ |                   | $^{40}\text{K}$ |                    |
|        | Range                          | Mean $\pm$ SD    | Range             | Mean $\pm$ SD     | Range           | Mean $\pm$ SD      |
| S1     | 29.72-35.44                    | 28.44 $\pm$ 2.20 | 13.85-15.15       | 14.15 $\pm$ 1.76  | 321.71-373.31   | 341.71 $\pm$ 21.40 |
| S2     | 29.61-47.20                    | 38.08 $\pm$ 2.73 | 6.10-32.98        | 15.37 $\pm$ 2.59  | 133.2-163.30    | 148.25 $\pm$ 8.03  |
| S3     | 5.92-55.66                     | 42.53 $\pm$ 1.45 | 3.28- 30.69       | 21.17 $\pm$ 10.65 | 331.73-363.98   | 347.85 $\pm$ 12.12 |
| S4     | 24.43-30.94                    | 27.12 $\pm$ 1.85 | 15.26-18.24       | 16.33 $\pm$ 1.64  | 337.39-395.29   | 354.70 $\pm$ 19.64 |
| S5     | 79.68-82.98                    | 79.52 $\pm$ 4.67 | 29.81-32.30       | 30.99 $\pm$ 2.85  | 119.48-126.56   | 123.02 $\pm$ 6.22  |



**Figure (6): The means of natural activity concentrations according to cement type and comparison with international standard values (ISV)**

The average activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in cement samples produced in different factories (Arabia Company, Ashamaliya Company and Al-Rajhi Company) in Jordan were (in Bq/kg) 67, 18

and 27. The activity concentrations of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  were larger in the previous studies compared with this study, while the activity concentration is smaller for  $^{40}\text{K}$  results (Hamideen, 2018). However, all these results

were below the allowed international standard values.

**The Radiation Hazard Indices of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in the Cement Samples**

The radiological hazard parameters *D*, AED, *Ra<sub>eq</sub>*, *I<sub>α</sub>*, *I* and *H<sub>ex</sub>* for all cement samples were calculated using Eqs. (2-7) and are presented in Table 6. The dose rate values were between 64 ± 9 nGy/h and 117 ± 8 nGy/h with a mean value of 82 ± 7 nGy/h. According to the UNSCEAR 2000 report, the world population-weighted average dose rate value is given as 84 nGy/h. It can be seen from the results that dose rate values of cement samples did not exceed the worldwide average value, except for sample S3 & S5. The highest value of the dose rate was found in sample S5.

The contributions to the dose rate in this sample from <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K were 43, 13 and 53 %, respectively. It can be interpreted from these contributions that fly ash additives, especially that of <sup>226</sup>Ra, can increase the dose rate.

The annual effective doses (in μSv/h) received from <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K due to absorption in the cement samples ranged between 313±9 and 574±8, respectively, with an overall average summation equal to 404 μSv/h. The calculated values of the annual effective dose indicate the higher exposure from sample S5, although this is still less than the international standard value. The

calculated values of *Ra<sub>eq</sub>* for all samples varied between 175±9 Bq/kg and 343±16 Bq/kg, with an average of 267±20 Bq/kg. These results were within the international standard values.

The values of external hazard index *H<sub>ex</sub>* were between 0.19 and 0.36, with an average of 0.25 ± 0.02. The obtained results of the radiation hazard index *I* were between 0.25 and 0.46, with an average of 0.33 ± 0.01. The results of alpha index *I<sub>α</sub>* varied from 0.14 to 0.40 with an average of 0.22 ± 0.02. The external hazard index, radiation hazard index and alpha index values were less than unity in all cement samples, which agrees with the recommended values. Fig. 7 and Fig. 8 show the varying distributions in all measurements for all cement samples in this study and their comparison with international standard values.

The results were compared with those reported from other countries, as shown in Table 7. The activity concentration of <sup>226</sup>Ra in this study was higher than in India, Egypt, Saudi Arabia, Pakistan and Turkey and lower than the other countries' data as was found in a previous study conducted in Jordan. The activity concentration of <sup>232</sup>Th in the cement samples was observed to be lower than those of other countries, except for Saudi Arabia, while being similar to the value reported for Turkey (2017).

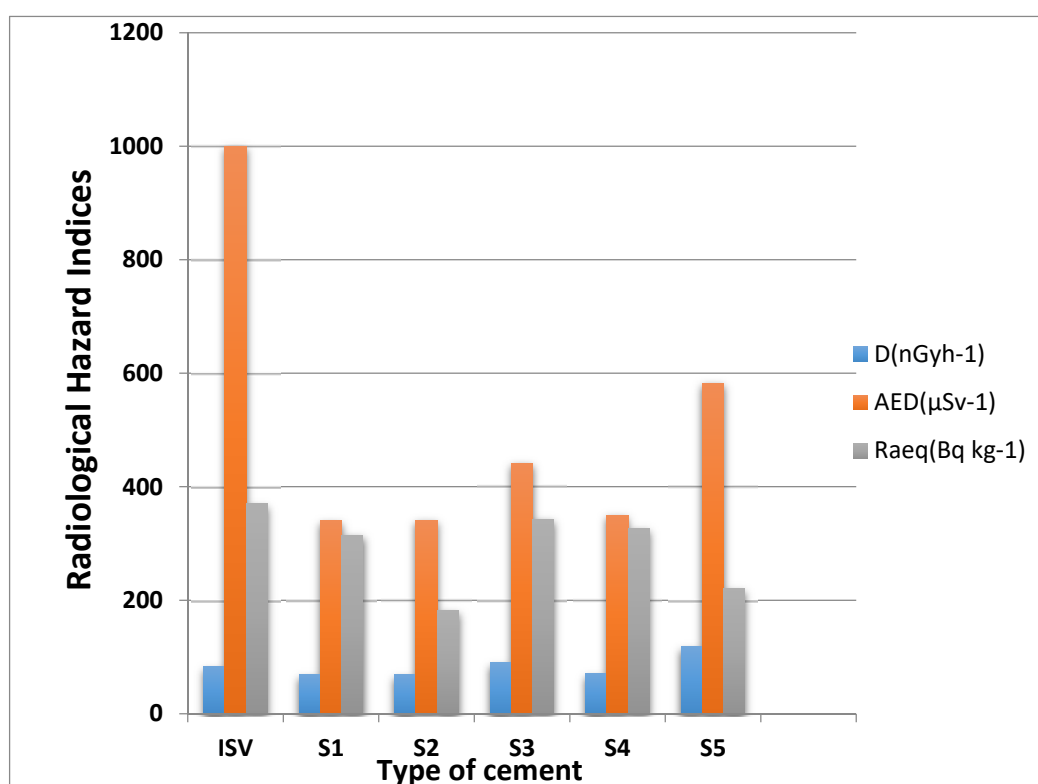
**Table 6. Dose rate, annual effective dose, radium equivalent, external hazard, radiation hazard index and alpha index in the studied cement samples**

| Sample type | D (nGy/h) | AED (μSv/h) | Ra <sub>eq</sub> (Bq/kg) | H <sub>ex</sub> | I    | I <sub>α</sub> |
|-------------|-----------|-------------|--------------------------|-----------------|------|----------------|
| ISV*        | 84        | 1000        | 370                      | 1.00            | 1.00 | 1.00           |
| S1          | 69 ±22    | 339±22      | 314±22                   | 0.20            | 0.28 | 0.14           |
| S2          | 64± 9     | 313±9       | 175±9                    | 0.19            | 0.25 | 0.19           |
| S3          | 90± 16    | 443±16      | 343±16                   | 0.27            | 0.36 | 0.21           |
| S4          | 71± 20    | 350±20      | 326±20                   | 0.21            | 0.29 | 0.14           |
| S5          | 117± 8    | 574±8       | 219±8                    | 0.36            | 0.46 | 0.40           |

\* ISV: international standard value.

**Table 7. Comparison of the mean values of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  activities of the cement samples (Portland cement, S1-S4) with those reported in other studies**

| Country      | $^{226}\text{Ra}$ (Bq/kg) | $^{232}\text{Th}$ (Bq/kg) | $^{40}\text{K}$ (Bq/kg) | Reference                         |
|--------------|---------------------------|---------------------------|-------------------------|-----------------------------------|
| Jordan       | 38                        | 15                        | 133                     | This study                        |
| Jordan       | 67                        | 18                        | 27                      | (Hamideen, 2018)                  |
| Turkey       | 34                        | 15                        | 220                     | (Ozdis et al., 2017)              |
| India        | 24                        | 20                        | 177                     | (Sharma et al., 2016)             |
| China        | 119                       | 36                        | 444                     | (Lu et al., 2014)                 |
| Saudi Arabia | 20                        | 9                         | 158                     | (Al-Dadi et al., 2014)            |
| Slovakia     | 11.8                      | 18.4                      | 156.5                   | (Estokova and Palascakova, 2013)  |
| Pakistan     | 34                        | 29                        | 295                     | (Aslam et al., 2012)              |
| Egypt        | 36                        | 43                        | 82                      | (El-Taher et al., 2010)           |
| Greece       | 111                       | 19                        | 244                     | (Papaefthymiou and Gouseti, 2008) |
| Worldwide    | 40                        | 30                        | 400                     | (EC, 1999)                        |



**Figure (7): The means of  $D$ , AED and  $Ra_{eq}$  of the cement types and a comparison with the international standard values (ISV)**

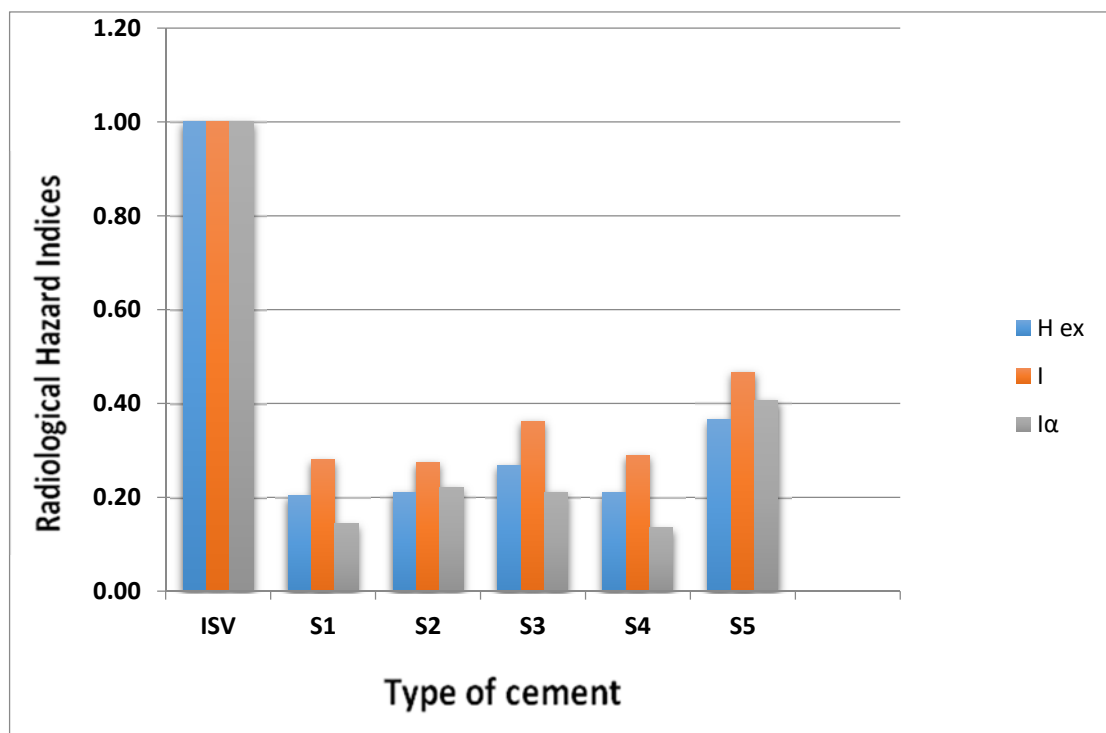


Figure (8): The means of  $H_{ex}$ ,  $I$  and  $I_{\alpha}$  of the cement types and a comparison with the international standard values (ISV)

The activity concentration of  $^{40}\text{K}$  in this study was found to be lower than those reported from other countries, except for Egypt (2010) and in a previous study conducted in Jordan (2018).

Overall, the detected and calculated values of the activity concentrations and the radiological hazard parameters were below the recommended limits. There was a clear high value in the absorbed dose rate ( $D$ ) above the recommended standard value in sample S5, which is related to blast-furnace cement. There is no recommendation for this result, as this type of cement is used under the ground in sewage systems, with no direct contact with humans. Each type of cement studied is used in a specific field as reported in this study; there are no necessary commutations to their usage.

### CONCLUSIONS

The cement types were compliant with Jordanian standard specifications (JSS. 30-1/2007) and European standards (EN. 19-1/2000). The activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  were measured and reported in

all cement samples and the reported results were within the international standard values. The calculation of dose rate, annual effective dose, radium equivalent, external hazard, activity concentration index and alpha index was carried out and the results obtained were within the recommended values, except for samples S3 & S5 (as expected for S5) which is related to the blast-furnace slag cement. There is no recommendation for this result, as this type of cement is used under the ground in sewage systems, with no direct contact after casting. The radiation results depend on the source and the area of the natural raw materials of those samples. The results overall are in agreement with other international studies and may contribute to future studies related to radiological protection for humans.

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