NATURAL RADIOACTIVITY AND ASSOCIATED RADIATION HAZARDS IN LOCAL PORTLAND AND POZZOLANIC CEMENTS USED IN JORDAN

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Abstract

Activity concentration of the natural gamma-emitting radionuclide (^{40}K, ^{226}Ra, and ^{232}Th) in at least forty samples of local Portland and Pozzolanic cement types is measured using gamma spectrometric techniques.

The range of the mean specific activity (minimum and maximum values) due to all the three radionuclide is found. Radiological hazards of the different samples is estimated using five approaches; the representative level index, the external hazard index, the internal hazard index ,the radium equivalent index, and the absorbed dose rate. Some of the measured radiological hazard parameters is compared to a similar parameters in different countries.

Keywords: *radiological hazards, portland cement, pozzolanic cement*

1. INTRODUCTION AND OBJECTIVES

The global demand for cement as a building material is so great. Cement is an important construction material for houses and buildings built in urban areas of Jordan. Portland cement is the most common type of cement used in construction applications, but it is an expensive binder due to the high cost of production associated with the high energy requirements of the manufacturing process itself (Pakou et al., 1994). Other cheap inorganic materials with cementitious properties such as natural pozzolans e.g. volcanic tuff (UNSCEAR, 2000; Ackers et al., 1985) and clay (Beretka and Mathew, 1985), and waste products from industrial plants e.g. slag (Khan et al., 1998), fly ash (Tufail et al., 1992; Tzortzis et al., 2003) and silica fume (Ziqiang and Mingqiang 1988) can be used as a partial replacement for Portland cement i.e. blended cements (Projin et al., 1984). In addition, to reduce the cost of binder, there are potential technological benefits from the use of pozzolanic materials as those blended with Portland cement in concrete applications. These include increased workability, decreased permeability (Sciocchetti et al., 1984), and increased resistance to sulphate attack (Malanca et al., 1993), improved resistance to thermal cracking and increased ultimate strength and durability of concrete (UNSCEAR, 1993; Mollah et al., 1986; Mustonen, 1985). The first objective of the present work is to assess the natural radioactivity of cement since workers are exposed to for a long time especially in mines and at manufacturing sites as well as people, that spend about 80% of their time inside offices and homes (Mollah et al., 1986; Paredes et al., 1987) result in exposure to cement or its raw materials being necessary reality.

The second objective of the present work is to calculate the radiological parameters (the representative level index, the external hazard index, the internal hazard index, the radium equivalent index, and the absorbed dose rate) which are related to the external gamma-dose rate and their effects on humans. The results of concentration levels and radiation equivalent activities are compared with similar studies carried out in other countries.

2. MATERIALS AND METHODS

2.1. Preparation of samples

Twenty five samples of all cement types produced by Cement Jordanian factory (Portland cement, Portland Pozzolanic cement (5%), Portland Pozzolanic cement (25%), White cement, and Sulphate

Resistant Cement (S.R.C)) were collected for investigation (Sideris et al., 2006; Massazza 1999). The percentage values in parentheses above indicate the percent of Pozzolana in cement. For comparison with products from other factories, 8 samples were taken from the ordinary Portland cement from (Arabia Company, Ashamaliya Company, Al-rajhi Company) and 4 samples were taken of Portland Pozzolanic cement (25%) from (Arabia Company, Al-rajhi Company) and 4 samples were taken of Sulphate Resistant Cement from (Arabia Company, Ashamaliya Company, Al-rajhi Company) .

Each sample, about1-kg in weight dried in an oven at about 110 ºC to ensure that moisture is completely removed. The samples crushed, homogenized, and sieved through a 200 mesh, which is the optimum size to be enriched in heavy minerals. Weighted samples placed in a polyethylene beaker, of 350-cm^3 volume. The beakers completely sealed for 4 weeks to reach secular equilibrium where the rate of decay of the radon daughters becomes equal to that of the parent.

2.2. Instrumentation and calibration

Measurements were performed using a High Purity Germanium (HPGe) detector supplied by EG&G Ortec. The detector is an n-type gamma-X ray (GMX) detector, operated at 3500 V, with useful energy range from 3 keV to 10 MeV, and a standard energy resolution of 2.02 keV and a relative efficiency of 56.9% at 1.33 MeV of 60Co. The absolute efficiency calibration of the detector was performed using the IAEA standard "soil-6" source within a Petri-dish of 90 mm in diameter and 10 mm thick. Its spectrum was collected for 12 h. Areas under the energy peaks of interest were used for drawing the peak efficiency curve between log of efficiency versus log of peak energy. A polynomial was fitted to the curve and the result was stored for further use. The gamma ray transition lines 186.21 keV (^{226}Ra), 295.20 keV, 351.90 keV (^{214}Pb) and 609.30 keV (^{214}Bi) were used to determine the activity concentration of ^{238}U. However, the interference of the ^{235}U peak at 185.9 keV, with the ^{226}Ra peak at 186.21 keV was resolved according to the method reported by Ribeiro et al. (2001). The gamma-ray transition lines 583.14 keV (^{208}Tl) and 911.07 keV (^{228}Ac) were used to determine the activity concentration of ^{232}Th. The activity concentration of 40K was measured directly from its only 1460 keV transition line.

The activity concentration in Bqkg^{-1} (A) in the environmental samples was obtained as follows:

$$A=\frac{Np}{e \times E \times m} \quad (1)$$

where N_p is the difference between counts per second of the sample and counts per second of the background, e is the abundance of the γ-peak in a radionuclide, E is the measured efficiency for each gamma-ray peak, and m is sample mass in kilograms.

2.3. Absorbed gamma dose rate.

The absorbed dose rates due to gamma radiations in air at 1 m above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were calculated based on guidelines provided by UNSCEAR (2000). The conversion factors used to compute absorbed gamma dose rate (D) in air per unit activity concentration in Bq/kg (dry weight) corresponds to 0.462 nGy/h for ^{226}Ra, 0.604 nGy/h for ^{232}Th and 0.042 nGy/h for ^{40}K. Therefore D can be calculated as follows (UNSCEAR, 2000):

$$D= 0.462\ C_{Ra}+0.604\ C_{Th}+0.0417\ C_k \quad (2)$$

where C_{Ra}, C_{Th} and C_k are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively.

2.4. Radium equivalent index, Ra_{eq}

In comparing the radioactivity of materials that contain Ra, Th and K a common index termed radium equivalent activity is required to obtain the total activity and is also used to assess the gamma radiation hazards. Since 98% of the radiological effects of the uranium series are produced by radium and its daughter products. The Ra_{eq} of a sample can be expressed as (Beretka and Matthew, 1985)

$$Ra_{eq}= CRa +1.43\ C_{Th} +0.077\ C_K \quad (3)$$

where C_{Ra}, C_{Th} and C_K is the specific activity in Bqkg^{-1}.

2.5. Representative level index (Iγr)

Another radiation hazards index primarily used to estimate the level of γ radiation associated with different concentrations of some specified radionuclides. It is defined from the following formula (NEA-OECD, 1979; Alam et al., 1999):

$$RLI\ (I\gamma r) = (1/150)\ A_{Ra} + (1/100)\ A_{Th} + (1/1500)\ A_K \quad (4)$$

where A_{rea}, A_t and A_K is the respective activity concentration in Be kg−1.

2.6. External Hazard index, H_{ex}

In the literature a number of criterion formulae have been derived over the years to assess the radiation dose rate due to exposure to gamma radiation from the natural radionuclides contained in building materials. The merits of these have been reviewed by the OECD's Nuclear Energy Agency (1979). Krieger (1981) proposed the following conservative model based on infinitely thick walls without windows and doors to serve as a criterion for the calculation of external hazard index, H_{ex}, defined as:

$$H_{ex} = (C_{Ra}/370) + (C_{Th}/260) + (C_K/4810) \leq 1 \quad (5)$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively.

2.7. Internal Hazard index. H_{in}

The internal hazard index H_{in} is used to control the internal exposure to ^{222}Rn and its radioactive progeny. The internal exposure to radon and its daughter products is quantified by the internal hazard index H_{in}, which is given by the following equation (Krieger, 1981):

$$H_{in} = (C_{Ra}/185) + (C_{Th}/260) + (C_K/4810) \leq 1 \quad (6)$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively.

3. RESULTS AND DISCUSSION

The distribution of natural radionuclides in different brands of cements, is presented in Table1. It can be seen from the Table1, that the activity of ^{226}Ra varies from 40.3 to 75.6 $Bqkg^{-1}$ and the arithmetic mean is 66.5$Bqkg^{-1}$. The activity concentration of ^{232}Th varies from 9.7 to 23.9$Bqkg^{-1}$ and the arithmetic mean is 18.2$Bqkg^{-1}$. The activity concentration of ^{40}K varies from 150.5 to 233.8 $Bqkg^{-1}$ and the arithmetic mean is 227.2 $Bqkg^{-1}$. The mean values are slightly higher than the corresponding worldwide average values which are 35and 30 $Bqkg^{-1}$ for ^{226}Ra whereas ^{40}K and ^{232}Th values are lower (400$Bqkg^{-1}$). Since the distribution of the natural radionuclides in each cement type is not uniform, a common index termed radium equivalent activity (Ra_{eq}) is required to obtain the total activity and is also used to assess the gamma radiation hazards. The radium equivalent of the total activity of each cement type is shown in Table 2. The highest value of Raeq was seen with white cement (125.49$Bqkg^{-1}$) and the lowest with Portland Pozzolanic Cement (5%) with an average value (78.08$Bqkg^{-1}$).

However, all the values obtained in this study for radium equivalent activity fall far below the criterion limit, as the use of materials whose radium equivalent activity concentration exceeds (370 $Bqkg^{-1}$) is prohibited. It is apparent that the Ra_{eq} of cement samples originating from different types show considerable variations, which are likely related to the type of raw materials used in cement manufacture. This is important in selecting suitable cement type for use in building and construction, especially those which have large variations in their activities.

There are a lot of radiation hazards indices primarily used to estimate the level of γ radiation associated with different concentrations of some specified radionuclides. The representative level index values, the external hazard index values, the internal hazard index values, as estimated using equations (4, 5, and 6) for all types of cements are listed in Table.2.

Table 1. The minimum, maximum, and mean activity concentrations of 226Ra, ^{232}Th and ^{40}K in (Bqkg^{-1}) for Cement Jordanian factory products.

Types of Cement	Specific γ- ray activity concentrations (Bqkg^{-1})								
	^{226}Ra			^{232}Th			^{40}K		
	Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.	Mean
Portland Cement	40.3	75.6	66.5	9.7	23.9	18.2	150.5	233.8	227.2
Portland Pozzolanic Cement (5%)	31.2	60.1	43.9	9.1	17.1	11.4	200.7	287.0	232.3
Portland Pozzolanic Cement (25%)	39.9	56.3	49.1	7.9	17.0	12.0	193.3	291.9	221.5
Sulphate Resistant Cement (S.R.C)	29.1	42.0	42.9	5.8	11.3	10.9	231.9	298.0	265.6
White Cement	56.2	79.2	77.2	17.9	26.4	22.9	178.6	219.0	201.9

Table 2. Radium equivalent activity, Representative level index, Gamma dose rate, External hazard index, and Internal hazard index for different brands of cement.

Types of Cement	Dose Rate (nGy/h)	Ra_{eq} (Bq/Kg)	H_{ex} (mSvy^{-1})	H_{in} (mSvy^{-1})	RLI (Bq/Kg)
Portland Cement	51.19	110.02	0.296	0.476	0.776
Portland Pozzolanic Cement (5%)	36.69	78.08	0.211	0.329	0.559
Portland Pozzolanic Cement (25%)	39.01	83.32	0.225	0.357	0.595
Sulphate Resistance Cement (S.R.C)	37.29	78.93	0.213	0.329	0.572
White Cement	57.91	125.49	0.339	0.547	0.878

The obtained results for the products show that the averages of radiation hazard parameters for all products under investigation are lower than the acceptable level 370 Bqkg^{-1}for radium equivalent Ra_{eq}, 1 for level index $I_{\gamma r}$, the external hazard index $H_{ex} \leq 1$ and 59(nGyh^{-1}) for absorbed dose as can be seen from Table 2.

Table 3 lists the comparison of activity concentrations in Portland cements in different areas of the world. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for all measured samples of Portland cement are comparable with the corresponding values and sometimes less than that of other countries. The radioactivity in Portland cement varies from one country to another because of different materials use in cement manufacture.

Table 3. Comparison between the activity concentrations of Portland cement samples from Cement Jordanian factory with that of other countries.

Country	Activity (Bq/kg)			Reference
	^{226}Ra	^{232}Th	^{40}K	
Finland	44	26	241	NEA-OECD (1979)
Brazil	61.7	58.5	564	Malanca et al. (1993)
Pakistan	31.3	26.8	212	Tufail et al. (2007)
Cuba	23	11	467	Brigido Flores et al. (2008)
Sweden	96	127	962	NEA-OECD (1979)
Australia	51.8	48.1	115	Beretka and Matthew (1985)
Austria	26.1	14.2	210	Sorantin and Steger (1984)
China	69.3	62	169	Ziqiang et al. (1988)
Germany	26	18	241	NEA-OECD (1979)
United Kingdom	22	7.0	141	NEA-OECD (1979)
Egypt	31.3	11.1	40.6	Sharaf et al. (1999)
Present work	**66.5**	**18.2**	**227.2**	---
World	35	30	400	UNSCEAR (2000)

4. CONCLUSIONS

Based on the assessment of potential radiological hazards as inferred from the calculations of radium equivalent activity, representative level index and the dose rate, the investigated cement samples fall within the category of accepted building materials and are safe to use for the construction of inhabited buildings. The results may be important from the point of view of selecting suitable materials for use in cement manufacture. The manufacturing operation reduces the radiation hazard parameters. Cement products do not pose a significant radiological hazard when used for building construction.

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