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Assessment of Natural Radioactivity Levels in Cement Samples Commonly Used for Construction in Lagos and Ogun State, Nigeria

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ABSTRACT

The activity concentrations of natural radionuclides ^{238}U, ^{232}Th and ^{40}K in the cement samples used for construction in Lagos and Ogun State Nigeria were measured by gamma spectrometry using a well-type NaI(Tl) detector. Radiological hazard assessments due to these natural radionuclides were carried out. The average activity concentrations of ^{238}U, ^{232}Th and ^{40}K determined in the samples were 2.16, 7.82 and 114.3 Bq/kg respectively. The mean absorbed dose rate, the mean annual effective dose, excess lifetime cancer risk and annual gonadal equivalent dose in the samples analyzed were 9.59 nGy/h, 17.66 µSv/y, 67.99 $(MPY)^{-1}$ and 69.07 µSv/y respectively. Values of radium equivalent activity (Ra_{eq}), external (H_{ex}) and internal (H_{in}) hazard indices and gamma activity concentration index (I_{γ}) were also estimated. The results obtained in all cases are well below the world average. Therefore, cements produced in this part of the country are safe and considered to have negligible radiological impact on the health of the individuals exposed to them.

1. Introduction

Cement is one of the most important materials used for construction of buildings in Nigeria. The advancement in the building industry results in most buildings in Nigeria being constructed from cement blocks and bricks. Cement also finds applications in concrete making, plastering and smoothening of building surfaces. Portland cement is obtained from grinding a clinker, produced by burning a mixture of limestone, clay, and gypsum at high temperatures (1450 – 1600 ºC) for the materials, and approximately 2000 ºC for the combustion fumes (Kpeglo et al., 2010). The cement dust is a gray powder having an aerodynamic diameter ranging from 0.05 to 5.0 µm (Kpeglo et al., 2010). This falls within the range of respirable particles (HPA, 2009). Varying degrees of respiratory symptoms have earlier been associated with exposure to Portland cement in people who are occupationally exposed (Yang et al., 1996; Noor et al., 2000).

The knowledge of the radioactivity level in building materials is important in order to estimate the radiological hazards on human health (Estokova and Palascakova, 2013). All building raw materials derived from rock and soil contain various amounts of natural radionuclides of the ^{238}U and ^{232}Th series, and the radioactive isotope of ^{40}K (Turhan, 2008). The natural radioactivity in construction materials, results into external and internal indoor exposure (Paulikovà and Kopilčáková, 2009). The external radiation exposure arises from gamma radiation generated by members of the ^{238}U and ^{232}Th decay chains and ^{40}K, while the internal radiation exposure, is due to radon and its short-lived products emanating from the construction materials (Khan and Khan, 2001).

Currently, the standards and guidelines with regard to acceptable levels of natural radioactivity in building materials are not defined by the regulatory authority in Nigeria. A number of studies have been carried out on building materials in Nigeria (Oleru, 1984; Farai and Ademola, 2005; Ademola, 2008; Akinloye and Abodunrin, 2008, Akinloye and Okeya, 2009). The data available on the subject are inadequate as the basis for policy makers, if accurate reference level that will incorporate general public health in every State in the country, is to be drawn. The main objective of this study is to investigate the radiological risk from the major brands of cements used for construction in these highly populated States in Nigeria. Hence, the activity concentrations of the cement samples due to ^{232}U, ^{232}Th and ^{40}K were determined.

2. Materials and Methods

A total of sixteen (16) cement samples representing the eight (8) brands, which is more than 95% of the different brands of cement manufactured and used in Lagos and Ogun State, Nigeria, for building construction purposes. These states are important sea and land transnational transport routes. The samples were collected from different suppliers, who are the main dealers in cement industry in the country. The cement samples collected were in powdery form as prepared by the individual manufacturer for the use of the building professionals. The samples were packed with identification labels, and taken to the laboratory where they were dried, pulverized and sieved through a 0.2 mm mesh for homogeneity. A-200 g weight of each sample was packed into the standard cylindrical container with a dimension of 7.0 cm by 6.5 cm. The samples were hermetically sealed with adhesive tape for thirty days which was sufficient time required to attain secular radioactive equilibrium (Ademola et al., 2014). These containers were of the same size and geometry as the one used for calibration and background measurement.

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The activity concentrations of natural radionuclides were determined by high efficiency gamma-ray spectrometry using well-type NaI detector. The detector is coupled to a computer based UNISPEC Multi-Channel Analyser (MCA), with serial number 22060316, mounted inside a cylindrical lead shield (100 mm thick). The radionuclides were identified using gamma ray spectrum analysis software, Gene 2K. The signal pulse from the MCA is always amplified with a pre-amplifier model 2002CSL (serial number 13000742). In order to reduce the uncertainty in gamma-ray intensities as well as the influence of coincidence summation and self-absorption effects of the emitted gamma rays, the absolute efficiency calibration of the gamma spectrometry system was undertaken using the radionuclide specific efficiency method (Kpeglo et al., 2010), since the accuracy of all quantitative results will depend on the attainable accuracy of the system calibration. The energy calibration was carried out with point sources (^{60}Co and ^{137}Cs) from IAEA. A resolution graph was drawn for the calibration as shown in Figure 1.

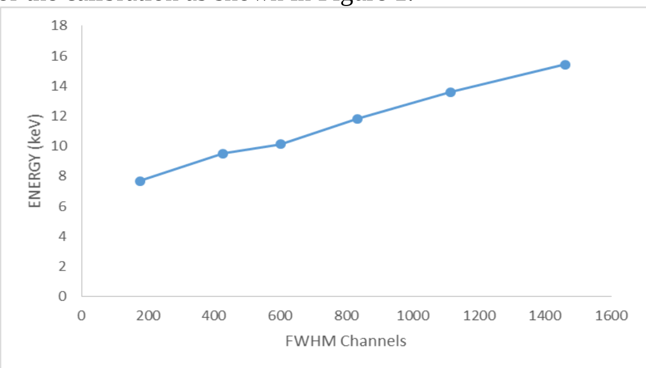

Figure 1. Curve showing energy against the channel for the gamma peak used in calibrating the spectrometer.

The same geometry was used to determine the peak area of the samples and references. Each sample was measured with a counting time of 28,800 s. Background measurements were taken and subtracted in order to get net counts for the sample. The specific radioactivity of ^{40}K was measured directly by its gamma energy at 1460.8 keV, while activities of ^{238}U and ^{232}Th were calculated based on the weighted mean value of their respective decay products in equilibrium: ^{226}Ra (186 keV), ^{214}Pb (295 keV; 351 keV) and ^{214}Bi (609 keV) for ^{238}U, and ^{212}Pb (238.6 keV) and ^{228}Ac (911 keV) for ^{232}Th.

3. Results and Discussion

3.1. Activity Concentrations

The activity concentrations of the radionuclides assessed in this work are below detection level (BDL) in some samples. The results of the activity concentrations of ^{238}U, ^{232}Th and ^{40}K from the different brands of cement analyzed and the mean values are presented in Table 1. The mean ranged from 0.82±0.17 – 14.76±2.14, 0.04±0.01 – 5.81±0.58 and 23.25±1.35 – 159.74±9.23 Bq/kg for ^{238}U, ^{232}Th and ^{40}K. The overall mean values of 7.82, 2.16 and 114.3 Bq/kg were respectively obtained for ^{238}U, ^{232}Th and ^{40}K in all the samples analyzed. The dominant gamma activity measured in all the samples is from ^{40}K. All the values obtained are below the world average of 30 Bq/kg, 35 Bq/kg and 400 Bq/kg respectively for ^{238}U, ^{232}Th and ^{40}K (UNSCEAR, 2008).

3.2. Radium Equivalent Activity (Ra_{eq})

This is a common index term which take into account the radiation hazards associated with the activities of the radionuclides, ^{238}U, ^{232}Th and ^{40}K found in environmental samples. In this work, Ra_{eq}, was computed using equation 1 as found in (Ademola, 2008):

$$Ra_{eq}\,(\mathrm{Bq/kg}) = C_{Ra} + 1.43C_{Th} + 0.077C_K \qquad 1$$

where C_{Ra}, C_{Th} and C_K are the concentrations in Bq/kg for ^{238}U, ^{232}Th and ^{40}K respectively.

It may be noted that ^{238}U is replaced by the decay product ^{226}Ra, although there may be disequilibrium between ^{238}U and ^{226}Ra. The ratio is given as 1.03 (UNSCEAR, 2000). The results obtained are presented in column 2, Table 2.

Radium Equivalent Activity index (Ra_{eq}) provides useful guidelines needed in regulating the safety standards on radiation protection for general public residing in common dwelling. Radium Equivalent Activity index represents a weighted sum of activities of the natural radionuclides. The mean value from all samples is 19.7 Bq/kg, a value distinctively lower than the world average of 370 Bq/kg (UNSCEAR, 2000). Materials with Ra_{eq} values higher than 370 Bq/kg have been regarded unfit for construction so as to avoid radiation hazard (Sam and Abbas, 2001).

Table 1. Mean Activity Concentrations (Bq/kg) from Eight Brands of Cements.

SAMPLE	^{238}U	^{232}Th	^{40}K
S	9.21±1.55	0.77±0.08	123.18±7.12
P	10.72±1.06	2.29±0.51	108.07±6.26
MR	5.42±1.19	0.04±0.01	153.90±8.83
D1	14.76±2.14	5.81±0.58	139.10±7.87
DO	0.82±0.17	BDL	94.39±5.49
B	6.30±0.99	4.95±0.57	23.25±1.35
E	4.78±0.70	0.56±0.07	112.18±6.50
G	10.56±1.50	2.86±0.32	159.74±9.23
MEAN	7.82	2.16	114.3

3.3. Hazard Index

Indices which represent external and internal radiation hazards have been defined by Beretka and Mattew (1985). External hazard (H_{ex}) was determined using the relation in equation 2:

$$H_{ex} = \frac{C_U}{370\ \mathrm{Bq/kg}} + \frac{C_{Th}}{259\ \mathrm{Bq/kg}} + \frac{C_K}{4810\ \mathrm{Bq/kg}} \qquad 2$$

In order to quantify the health impact from radon and its progeny which may give rise to internal radiation exposure, internal hazard index (H_{in}) was determined using equation 3:

Table 2. Radium equivalent activity, external and internal hazard index, absorbed dose, annual effective dose, excess lifetime cancer risk, annual gonadal dose and gamma activity concentration index in eight brands of cement.

SAMPLE	Ra_{eq} (Bq/kg)	H_{ex}	H_{in}	D (nGy/h)	H_E (nSv/y)	ELCR (10^{-6})	AGED (μSv/y)	I_γ
S	19.79	0.053	0.078	9.63	17.74	68.28	70.33	0.074
P	22.30	0.060	0.089	10.7	19.62	75.53	76.59	0.079
MR	17.33	0.047	0.061	8.82	16.23	62.48	65.24	0.070
D1	33.84	0.091	0.131	16.1	29.59	11.39	113.82	0.115
DO	8.08	0.022	0.024	4.31	7.94	30.57	32.16	0.034
B	15.16	0.041	0.058	6.97	12.83	49.40	47.42	0.045
E	14.21	0.038	0.051	7.13	13.13	50.53	52.30	0.055
G	26.95	0.073	0.101	13.1	24.20	93.15	94.74	0.098
MEAN	19.71	0.053	0.074	9.59	17.66	67.99	69.07	0.071

$$H_{in} = \frac{C_{Ra}}{185\ Bq/kg} + \frac{C_{Th}}{259\ Bq/kg} + \frac{C_K}{4810\ Bq/kg} \quad 3$$

The concept of hazard index is used to evaluate the potential hazards associated with both radiological and non-radiological hazards (Akinloye and Okeya, 2009). The hazard indices (H_{ex} and H_{in}) for materials used in bulk amounts should be less than unity in order to limit the gamma radiation dose to 1.5 mSv/y. The mean values of H_{ex} and H_{in} obtained are presented in columns 3 and 4 in Table 2. The mean values of 0.053 and 0.074 which are less than unity as required were obtained for H_{ex} and H_{in} from all samples.

3.4. Absorbed Dose Rate in Air

The absorbed dose rate, D, nGy/h was obtained at 1 m from the ground resulting from the mean activity concentration of the radionuclides found in these cement samples using equation 4 (Ajayi, 2000):

$$D = 0.429C_U + 0.666C_{Th} + 0.042C_K \quad 4$$

The results obtained range from 4.31 – 16.08 nGy/h and are presented in column 5 in Table 2. The values are below the global average of 54 nGy/h (UNSCEAR, 2008).

3.5. Annual Effective Dose Rate

The total absorbed dose rate accumulated over a year, is reported as the annual effective dose (µSv/y), generally calculated using equation 5 (Akinloye and Abodunrin, 2008):

$$H_E = D \times T \times F \quad 5$$

Where F is the conversion factor (0.7 Sv/Gy) and T is the occupancy time given as:

$$T(h/y) = occupancy\ factor \times 24\ hour \times 365.25\ day$$

Using outdoor occupancy factor of 0.3, T was determined to be 2629.80 h/y. The results obtained for the annual effective dose and the mean values are presented in column 6, Table 2. The mean values obtained range from 7.94 – 29.59 µSv/y, with overall mean of 17.66 µSv/y from all samples. The results obtained in this work are obviously lower than the world average of 70 µSv/y according to UNSCEAR, (2008).

3.6. Excess Lifetime Cancer Risk

The Excess Lifetime Cancer Risk (ELCR) per Million Person Yearly $(MPY)^{-1}$ determined from the effective dose obtained was estimated using equation 6 according to the ICRP (1991) assumed parameters (Salih and Jaafar, 2013):

$$ELCR = H_E \times DL \times RF \quad 6$$

The assumed parameters DL (the duration of life) is 70 years and RF (the risk factor) is 0.055 Sv^{-1}. The results obtained are presented in column 7, Table 2. The ELCR is a common index term used to determine the number of cancer risk per million people yearly $(MPY)^{-1}$ from inhalation of radon and its short-lived alpha-emitting daughters released from the cement samples. Various models have been used to estimate ELCR (EPA, 1986; UNSCEAR, 1988, 1993) but the model according to ICRP (1991) was found appropriate for this study. The results of ELCR obtained ranged from 11.39 – 11.82 per million person yearly $(MPY)^{-1}$ with a mean of 67.99 $(MPY)^{-1}$.

3.7. Annual Gonadal Equivalent Dose

The annual gonadal equivalent dose (AGED, µSv/y) for the resident of a house built with any of the cement sample in this work, was estimated using (Ademola et al., 2013):

$$AGED = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_K \quad 7$$

This model considers a house as a cavity with infinitely thick walls, which makes it possible to compare AGED values of a house containing concentrations of ^{226}Ra, ^{232}Th and ^{40}K in equivalence with the world average (30, 35, and 400 Bq/kg respectively) with those obtained in a particular material. Results of the AGED estimated in this work are presented in column 8, Table 2. The organ of interest in radiation protection includes the gonad, the active bone marrow and the bone surface cells (UNSCEAR, 1988). The results obtained for each brand ranged from 32.16 – 113.82 µSv/y and the mean value of 69.07 µSv/y was obtained which is less than the world average of 298 µSv/y (Zaid et al., 1999).

3.8. External Dose Criterion

The index, gamma activity concentration index (I_γ) was derived using equation 8 according to (Stephen, 1999):

$$I_\gamma = \frac{C_{Ra}}{300\ Bq/kg} + \frac{C_{Th}}{200\ Bq/kg} + \frac{C_K}{3000\ Bq/kg} \quad 8$$

The results of the mean I_γ obtained for the samples are presented in column 9, Table 2.

According to the European Commission, controls should be considered in terms of radiation protection, based on annual effective dose range of 0.3 – 1 mSv/y, which is gamma dose contributed by the building material to the dose received outdoors (Stephen, 1999). The dose derived for the annual effective dose in this study do not fall within the different levels stated in the recommendation, it was however determined in order to fully ascertain whether the cement samples meets the required dose criteria. The results obtained range from 0.34 – 0.115 with an average of 0.071 from all samples. The mean result obtained is below the average dose criterion, thereby affirming that the annual dose resulting from these cement samples are well below the limit that may warrant regulation. Hence, the cement samples assessed in this work are considered safe in terms of the radiological impact to the public exposed to them.

Table 3 presents the results obtained in this study and similar studies from different environments. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K obtained in this study is lower than the one obtained in Cameroon (Ngachin et al., 2007), Egypt (El-Taher, 2010), Zambia (Hayumbu et al., 1995), India (Kumer et al., 1999), Sweden (NEA-OECD, 1979), Italy (Rizzo et al., 2001), Brazil (Malanca et al., 1993),

Table 3. Comparison of the results obtained in this study with literatures.

Country	^{238}U	^{232}Th	^{40}K	Ra_{eq}	References
	(Bq/kg)				
Cameroon	27±4	15±1	277±16	70	Ngachin et al., 2007
Egypt	36.6±4.4	43.2±2.2	82±4.1	103	El-Taher, 2010
Zambia	23±2	32±3	134±13	79±11	Hayumbu et al., 1995
India	37	24.1	432.2	104.7	Kumer et al., 1999
Sweden	55	47	241	-	NEA-OECD, 1979
Italy	38±14	22±14	218±2	92±60	Rizzo et al., 2001
Brazil	61.7	58.5	564	188.8	Malanca et al., 1993
United Kingdom	22	18	155	-	NEA-OECD, 1979
Hong Kong	19.2	18.9	127	-	Yu et al., 1992
Malaysia	51±1.0	23±1.0	823±69	188±27	Nooeddin, 1999
Nigeria	2.16	7.82	114.3	19.7	Present Study

UK (NEA-OECD, 1979), Hong-Kong (Yu et al., 1992) and Malaysia (Nooeddin, 1999). The Ra_{eq} of 19.70 Bq/kg obtained in this study is far lower than the values obtained in other studies as presented in column 5, Table 3.

4. Conclusions

Cement is considered an important material for the building industry in Nigeria, resulting from advancement in the industry. This development has led to the rise in the demands for cement, leading to the rise in the number of cement factories in the country. A number of these factories are located around Lagos and Ogun State. These two States are not only well populated but serves important local and international boundary functions. Therefore, eight (8) different brands of cement produced within the vicinity of these states were sampled for gamma radionuclide content by employing a well-type NaI (Tl) detector.

The radionuclides found in all samples are natural radionuclides – ^{238}U, ^{232}Th and ^{40}K. The mean activity concentrations of these radionuclides range from 0.82±0.17 – 14.76±2.14, 0.04±0.01 – 5.81±0.58 and 23.25±1.35 – 159.74±9.23 Bq/kg with a mean of 7.82, 2.16 and 114.3 Bq/kg respectively from all samples analyzed in this study. The dominant activity is from ^{40}K, which is the usual trend in environmental samples. Absorbed dose, D, in the samples assessed range from 4.31 – 16.10 nGy/h with a mean value of 9.59 nGy/h while a range of 7.94 – 29.59 nSv/y with a mean value of 17.66 was estimated for annual effective dose in these samples. Other radiological parameters which include radium equivalent activity (Ra_{eq}), external (H_{ex}) and internal (H_{in}) hazard indices, excess lifetime cancer risk (ELCR), annual gonadal dose (AGED) and gamma activity concentration index ($I\gamma$) were also estimated in this work. The results obtained in all cases are well below the world average, and compared well with values obtained in literatures and the generally accepted limiting values. Cements produced in this part of the country can therefore be considered of negligible radiological impact to the health of the individuals exposed to them at the levels assessed in this study.

Author Contributions

Augustine Kolapo Ademola conceived and designed the research; John O. Omoboyede performed the experiments and analyzed the data; Mariam Afolake Fatai contributed materials and analysis tools; Oluwasayo Peter Abodunrin wrote the paper.

Conflicts of Interest

The authors declare no conflict of interest.

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