



ISSN: 0975-833X

## RESEARCH ARTICLE

### ASSESSMENT OF RADIOLOGICAL HAZARDS OF SOME SELECTED PORTLAND CEMENT USED IN NIGERIA

<sup>1</sup>Oladapo O. O., <sup>2</sup>Oni E. A. and <sup>3</sup>Akerele O. O.

<sup>1</sup>Department of Science laboratory Technology, Ladoke Akintola University of Technology, P.M.B. 4000 Ogbomoso

<sup>2</sup>Department of Pure and Applied Physics, Ladoke Akintola University of Technology, P.M.B. 4000 Ogbomoso

<sup>3</sup>National Institute of Radiation Protection and Research, Ibadan, Nigeria

#### ARTICLE INFO

##### Article History:

Received 17<sup>th</sup> September, 2012  
Received in revised form  
21<sup>th</sup> October, 2012  
Accepted 11<sup>th</sup> November, 2012  
Published online 18<sup>th</sup> December, 2012

##### Key words:

Radionuclides concentration,  
Nigerian Portland cements,  
Activity,  
Radiological hazards.

#### ABSTRACT

This work assess through analysis, the natural radionuclide level and the associated radiological hazards of some selected Portland cement used in Nigeria. The natural radionuclide content of Portland Cement widely used in Nigeria were measured using a highly shielded Canberra Na(Tl) detector, a type of gamma-ray spectrometry. A total of 15 samples from 5 batches of the product were analyzed. The mean value of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K concentrations determined were  $33.85 \pm 9.79$  Bqkg<sup>-1</sup>,  $20.05 \pm 3.93$  Bqkg<sup>-1</sup> and  $78.01 \pm 25.01$  Bqkg<sup>-1</sup> respectively, being lower than the permissible global values of 52.2 Bqkg<sup>-1</sup>, 41.0 Bqkg<sup>-1</sup> and 230.0 Bqkg<sup>-1</sup> respectively by UNSCEAR. On the basis of hazard indices, internal hazard index of 0.2766, external hazard index of 0.1850 were obtained for the product and are found to be much lower than unity. The absorbed dose rate of 31.0030nGyh<sup>-1</sup> and the annual effective dose rate of 0.1521mSvy<sup>-1</sup> are much below the recommended standard by UNSCEAR. All the cement sample analyzed met the safety criteria by UNSCEAR and hence do not pose any radiological hazards to human health.

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#### INTRODUCTION

Man is continuously exposed to ionizing radiation from Naturally Occurring Radioactive Materials (NORM). The origin of these materials is the earth crust, but they find their way into building materials, air, water, food and the human body itself. In many parts of the world, building materials containing radioactive materials have been used for generations. As individuals spend more than 80% of their time indoor, the internal and external radiation exposure from building materials creates prolonged exposure situation (ICRP, 1999). The Earth is naturally radioactive, and about 90% of human radiation exposure arises from natural sources such as cosmic radiation, exposure to radon gas and terrestrial radionuclides (Lee *et al.*, 2004). Everyday, each of us is exposed to natural occurring quantities of radiation. Besides bricks, granite and sands; cement as one of the main components in building materials is a combination of rocks and soil known to contain natural radioactivity mainly radionuclides from the <sup>238</sup>U and <sup>232</sup>Th decay chains and the radioactive isotope of potassium <sup>40</sup>K. During the last three decades, there has been an increasing interest in the study of the radioactivity of building materials. Several surveys have been conducted to establish the radioactivity concentrations in raw material, industrial by-product and building materials and their radon exhalation rate (European Commission, 1999).

Cement manufacturing started in Nigeria in the early 1950s, the domestic industry has never been able to produce more than 30% of the national consumption level. As at 1993, the remaining 70% was imported (Esubiyi, 1995). Portland cement, which is one of the major building material used in Nigeria is a closely controlled chemical combination of Calcium, Silicon, Aluminum, Iron and small amounts of other compounds, to which gypsum is added in the final grinding process to regulate the setting time of the concrete. Some of the raw materials used to manufacture cement are Limestone, shells and chalk or marl, combined with shale, slate or blast furnace slag, silica sand and iron ore, Lime and silica make up approximately 85 percent of the mass (Odunike *et al.*, 2008). Various work on the various activity concentration of Portland cement and fibre cement used in Nigeria have been carried out (Farai & Ejeh, 2006; Ademola, 2008b; Akinloye & Okeya, 2009). The aim of this work is to determine through analysis, the associated radiological hazards of some selected Portland cement used in Nigeria.

#### MATERIALS AND METHODS

##### Sample Preparation

For this study, a total of fifteen (15) cement samples of five (5) batches were used. These include; Dangote, Pure Chem, Burham, Magen Roi, Elephant Portland cements. They were collected from Ogbomoso and Ibadan, both locations lie in the

**Table 1: Radionuclide concentrations in Nigeria Portland Cement (BqKg<sup>-1</sup>)**

Cement Sample	<sup>40</sup> K	<sup>226</sup> Ra	<sup>232</sup> Th
Dangote	44.78 ± 20.28	17.88 ± 3.80	58.38 ± 25.08
Magen Roi	28.21 ± 4.04	25.80 ± 3.49	74.80 ± 41.47
Pure Chem	25.46 ± 6.54	20.38 ± 3.52	78.27 ± 25.94
Burham	32.90 ± 8.39	17.39 ± 5.25	89.72 ± 15.02
Elephant	37.90 ± 9.71	18.81 ± 3.59	88.87 ± 17.97
Mean Value	33.85 ± 9.79	20.05 ± 3.93	78.01 ± 25.10

**Table 2: The mean specific radioactivity of natural radionuclides of some Nigeria and Foreign countries' cement in Bqkg<sup>-1</sup>**

No	COUNTRY SITE	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K	REFERENCE
1	Australia	52	48	11.5	Beretka & Mathew, 1985
2	Australia	26.7	14.2	210	Sorantin & Steger, 1984
3	Bangladesh	120.2	132.4	505.7	Mollah et al., 1986
4	Finland	40.2	19.9	251	Mustonen, 1984
5	Hungary	30.5	21.8	220	Gallyas & Torok, 1984
6	India	59.2	14.8	107.4	Londhe et al., 1984
7	Iraq	60	31.6	349	Bahjat & Fouad, 2000
8	Italy	46	42	316	Scichetti et al., 1984
9	Italy	67	138	584	Esposito & Pelliccion, 1985
10	Nigeria	9.40	6.01	27.86	Akinloye & Okeya, 2009
11	Nigeria	52.4	4.1	91.8	Farai & Ejeh, 2006
12	Nigeria	43.8	21.5	71.5	Ademola, 2008b
13	Nigeria	33.85	20.05	78.01	This work
14	Poland	48.1	48.1	185	Marcinkowski & Pensko, 1979
15	Spain	74	31	241	Krisiuk et al., 1971
16	USA	-	14.8	126	Londhe et al., 1984
17	USSR	25.9	14.8	148.1	Londhe et al., 1984
	Permissible Global Level	52.20	41.0	230.0	UNSCEAR 1982, 1988

**Table 3: Ra<sub>eq</sub>, D, H<sub>in</sub>, and E values for some selected Nigeria Portland Cement**

SAMPLE	Ra <sub>eq</sub> (Bqkg <sup>-1</sup> )	D (nGyh <sup>-1</sup> )	H <sub>in</sub>	H <sub>ex</sub>	E (mSvy <sup>-1</sup> )
Dangote	74.4350	33.9223	0.3232	0.2020	0.1664
Magen Roi	70.3400	31.7354	0.2677	0.1913	0.1557
Pure Chem	60.0823	27.3359	0.2326	0.1637	0.1341
Burham	64.0481	29.4470	0.2636	0.1746	0.1444
Elephant	71.0192	32.5769	0.2960	0.1934	0.1598
Mean Value	67.9845	31.0030	0.2766	0.1850	0.1521

Southwestern Nigeria. The cement samples were collected, air-dried, crushed and made to pass through a 0.5 mm mesh sieve. The cement samples were stored in cylindrical air-tight containers and the containers were labeled and sealed. These samples were then left for about four (4) weeks before counting in order to attain a state of secular radioactive equilibrium after their progeny (Veiga *et al.*, 2006). Then the samples were analyzed to determine the radionuclide concentration in the samples.

### Measurement

In this analysis, gamma ray spectroscopy method was adopted. The spectrometer used for gamma counting consists of a highly-shielded Canberra NaI(Tl) detector enclosed in a 100 mm thick lead blocks coupled to a Canberra Multichannel Analyzer (MCA) with a PC via an interface. The collector is located in the centre of the lead shield in order to minimize the effect of scattered radiation from the shield (UNSCEAR, 1982). The Energy and Efficiency calibration of the gamma spectrometer were carried out using the International Atomic Energy Agency (IAEA) reference source material. Accurate energy and efficiency of the gamma spectroscopy system were made quantity radionuclides present in the sample since the accuracy of all quantitative results depend on the attainable accuracy of the systems calibration. The transition line of

1460 keV for <sup>40</sup>K, 1764 keV for <sup>214</sup>Bi and 2614 keV of <sup>208</sup>Ti were used to determine the concentration <sup>40</sup>K, <sup>238</sup>U and <sup>232</sup>Th respectively. Finally, counting was carried out for a period of 36000s, first with an empty Marinelli beaker of identified geometry as the sample to determine the background spectrum. Thereafter, the sealed sample of cement were counted for the same period of 36000s.

### RESULT AND DISCUSSION

The results of this study highlights the presence and level of the radionuclide concentrations determined for each batch of Portland cement measured. Table 1 shows the mean values and the standard deviations of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in each batch. The specific activities determined for <sup>226</sup>Ra, ranged between 25.46 and 44.78 Bqkg<sup>-1</sup> with a mean value of 33.85 ± 9.79 Bqkg<sup>-1</sup>. Also, the specific activities for <sup>232</sup>Th ranged from 17.39 to 25.80 Bqkg<sup>-1</sup> with an average of 20.05 ± 3.93 Bqkg<sup>-1</sup> for <sup>40</sup>K, it ranges between 58.38 and 89.72 Bqkg<sup>-1</sup> with a mean value of 78.01 ± 25.10 Bqkg<sup>-1</sup>. Table 2 presents the comparison of the data obtained in the work with the permissible global levels of natural radionuclide concentrations in cement for <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K including the data reported by other investigators for various cement. For instance, comparison of the data from this work with the fibre cement showed that the values obtained in this work is higher than the result from fibre cement. It is evident that the data

obtained from some selected samples of Nigeria Portland cement in this work are smaller than the permissible global level of 52.20, 41.00 and 230.00 BqKg<sup>-1</sup> for <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K respectively. From the data, it is indicated that <sup>40</sup>K is the highest contributor to the radionuclide content while <sup>232</sup>Th is the least contributor.

### Assessment of the radiological Hazards of the Materials

In assessing the radiological hazard of cement, it is usually undertaken by determining the limit of the external gamma radiation dose, external hazard index and internal hazard index (Ademola, 2008a; Hayunbu, 1995; UNSCEAR, 1982). This concept of the hazard index has been used for the evaluation of potential hazard associated with both radiological and non-radiological hazards. External hazard is associated with gamma radiation emitted by radionuclides of concern and it is determined using equation 1;

$$H_{ex} = 0.0027C_{Ra} + 0.00386C_{Th} + 0.000208 C_K \dots\dots 1$$

Where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the activity concentrations in BqKg<sup>-1</sup> of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K respectively. For materials used in bulk amounts, the external hazard index should be less than unity in order to limit external gamma radiation dose of 1.5 mSv<sup>-1</sup> (Ademola, 2008, Beretka and Mathew, 1985). For superficial and other materials with restricted use, a different dose criterion index [I] is applied (European Commission, 1999). Also, the external index is gotten by using specific dose rates and 1.0 mSv<sup>-1</sup> as the limit. Materials that are used in bulk amounts like concrete or brick should be less than 1 but for superficial and other materials with restricted use, the limit is 6 (European Commission, 1999). Portland Cement falls into the latter category. In this work, the radium equivalent activity ( $Ra_{eq}$ ) was also calculated since it gives a single index to describe the gamma output from different mixtures of radium, thorium and potassium in the material. The  $Ra_{eq}$  is calculated by the equation described (Ademola, 2008a; Yang *et al.*, 2005). The equation is stated in equation 2

$$Ra_{eq} = C_{Ra} + 1.43 C_{Th} + 0.07 C_K \dots\dots\dots 2$$

Where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the activity concentrations in Bqkg<sup>-1</sup> of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K respectively. The  $Ra_{eq}$  values calculated for the selected Nigeria Portland Cement vary between 60.0823 and 74.435 Bqkg<sup>-1</sup> with a mean value of 67.9845. These values are smaller than suggested maximal admissible value of 370 Bqkg<sup>-1</sup> which is equivalent to an annual dose of 1.5 mSv but the European Commission report sets the limit as 0.3 – 1.0 mSv<sup>-1</sup> for safe use (European Commission, 1999; Flores *et al.*, 2005; OECD, 1979). Part of the constitute source of internal radiation is radon and its short-lived products. It is quantified by the internal hazard index described in equation 3 (Beretka & Mathew, 1985; Krieger, 1981; Xinwei, 2005).

$$H_{in} = C_{Ra} / 185 \text{ Bqkg}^{-1} + C_{Th} / 259 \text{ Bqkg}^{-1} + C_K / 481 \text{ Bqkg}^{-1} \dots\dots\dots 3$$

Where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  have the same meaning as in equation 2. It has been stated that, if the maximum concentration of radium is half of that of the normal acceptable limit, than  $H_{in}$  will be less than unity (Ademola, 2008a; Xinwei, 2005). Therefore, for the safe use of a cement in construction of dwelling,  $H_{in}$  should be less than unity. The value of  $H_{in}$  calculated for the Portland Cement studied in this

work varies from 0.2326 to 0.3232 with a mean value of 0.2766. The values are much less than unity. Moreover, to provide a characteristic of the external gamma ray, the absorbed dose rate D in the cement was calculated using equation 4 (UNSCEAR, 2000; Veiga *et al.*, 2006)

$$D(\text{nGyh}^{-1}) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K \dots\dots\dots 4$$

Where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  have the same meaning as in equation 2. The absorbed dose rate for this product ranged from 27.3359 to 33.9223 nGyh<sup>-1</sup> with a mean value of 31.0030 nGyh<sup>-1</sup> as indicated in table 3. The values are less than the global average of 55 nGyh<sup>-1</sup> (UNSCEAR, 2000). In order to assess the health effect due to the absorbed dose rate, the annual effective dose rate (E) was determined. The calculation was made by using the conversion coefficient of 0.7 SvGy<sup>-1</sup> and the indoor occupancy factor of 0.8 as described in equation 5

$$E(\text{mSvy}^{-1}) = D(\text{nGyh}^{-1}) \times 8760(\text{hy}^{-1}) \times 0.8 \times 0.7(\text{SvGy}^{-1}) \times 10^{-6} \dots\dots\dots 5$$

The E values obtained for this product ranged from 0.1341 to 0.1664 with a mean value of 0.460 mSvy<sup>-1</sup> for terrestrial radionuclides for areas of normal background radiation. From this, it is evident that the data obtained for this product give a much lower value. A summary of all the results is presented in Table 3 while comparison with the results of other investigators for cement are presented in Table 2

### Conclusion

The measurements of the natural radionuclide content of some selected samples of Nigeria Portland Cement were undertaken by means of gamma-ray spectrometry using a well shielded and well calibrated Canberra Na(Tl) detector coupled to a Canberra Multichannel Analyzer (MCA). The results of the natural radionuclide concentrations was obtained for <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in the 15 samples from 5 batches of the cement product analyzed were much smaller than the permissible global value by UNSCEAR. The radium equivalent activity, the internal hazard index ( $H_{in}$ ), the dose rate, the annual effective dose rate and the external hazard index were calculated to determine the radiological implication of employing the use of Nigeria Portland Cements. All the samples of the Portland cements analysed met the safety requirements.

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