GAMMA SPECTROMETRIC ANALYSIS OF DIFFERENT BRANDS OF CEMENT USED IN NIGERIA.

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Abstract

The natural radionuclide content of common brands of cement consumed in Nigeria is presented in this work. Samples of 9 brands of grey ordinary Portland cement (OPC) and 5 brands of white cement were collected and analyzed for their radiological content by gamma spectrometry using a 7.6 x 7.6 NaI(Tl) detector. The total average content of ^{226}Ra, ^{232}Th, and ^{40}K for all the cement brand samples were 38.1, 27.9, and 301.1 Bq.kg^{-1} respectively and are lower when compared to the world average (50, 50 and 500 Bq.kg^{-1}) in building materials. The evaluated radium equivalent activities, external and internal hazard indices were lower than the recommended safe limit and are comparable with results from similar studies concluded in other countries. The evaluated mean gonadal dose equivalent of two cement brand samples were found to be higher than the world average in soil while others are less than the world average in soil.

Keywords: natural radioactivity, gamma spectroscopy, exposure, gonads, cement

1. Introduction

Man and the biota are continuously irradiated by ionizing radiation from many sources broadly classified as either natural or artificial. Most of the total radiation exposure of man and his environment come from natural sources (UNSCEAR, 2000). This natural radiation comes from two main sources: cosmogenic radionuclides (^{3}H, ^{14}C etc) and long lived primordial radionuclides and their daughters (^{40}K, ^{238}U, ^{235}U, and ^{232}Th). The amount of the cosmogenic radionuclides is basically constant because of equilibrium between their rate of creation by cosmic radiation and their radioactive decay (Mikhail, 2008). Although the amount of primordial radionuclide keeps decreasing slowly with time due to radioactive decay, quite a significant amount still remain in the earth crust today due to their long half-lives. These natural (primordial) radionuclides are known to be distributed in rocks and soil across the earth in varying concentrations depending on the geography and geological formations. The inhomogeneous distribution of these radionuclides in geological formations like soils and rocks suggests that man made products derived from these substances will contain traces of these radionuclides whose concentrations will be dictated by the origin of the soil and rocks they are derived from. One of such materials derived from rocks is Portland cement. Portland cement is a major building material used worldwide; it is derived from mixing natural clay, limestone and gypsum at high temperature (Kpeglo et al., 2011). Cement when mixed with fine and coarse aggregate in the right proportion is used for making concrete block- a basic building material worldwide.

The use of cement as a basic building material cuts across all social and economic strata in Nigeria. It is used (when mixed with other materials such as fine sand) majorly in making concretes, concrete and sand blocks and for floor, wall and even roof finishing. Over 90% of structures used as homes, offices, and commercial centers in Nigerian urban areas are constructed majorly using cement as binding material for concrete blocks. In rural areas where clay/mud blocks and other locally sourced materials are used, it is common to see walls and floors plastered using cement paste. Consequently, the yearly consumption of cement of 10 million tons (Farai and Ejeh, 2006) has been on the increase due to rapid structural development by government at all levels and the rapid increase in population and the consequent need for more homes and public structures. About 20 years ago, a large proportion of cement used in Nigeria are imported (Esubiyi, 1995) while lesser proportion comes from the local industries. Today, the number of local cement industries has swollen such that more than 50% of locally consumed cement is manufactured locally. Generally, the local producing companies are located in areas where the raw materials are obtained while the imported product comes majorly from Asian and European countries. Although the brand names of some of the cements may suggest that they are manufactured in Nigeria, research (Avwiri,2005) has shown that some local companies only package imported cement dust using different brand names. The diverse sources of these cements imply that their natural radionuclide content will vary. Many research conducted and completed all over the world (El-Bahi, 2004; Khalid and Hasan, 2001; Farai and Ejeh, 2006 etc) has shown that natural radionuclides is present in Portland cement in varying concentration from country to country. Due to the diverse application of cement in building constructions, it could account for the bulk of indoor background radiation exposure to the populace. Furthermore, the grain size of cement is such that it is aerodynamic (Kalacic, 1973), which could easily pass through respiratory track, or get blown by air into food and water. Consequently, the presence of radionuclide in cement does not only pose potential external radiation hazard but could also cause internal radiological contamination as well.

Many works have been done in Nigeria on building materials including cements without indication of brand name and their acceptability. Thus, one is not sure about the nature and the distribution of the cement referred to in those works. In this work, the natural distribution of the cement brands considered is emphasized using the Nigeria map for the first time. Furthermore, this work considered brands that make up of more than 95% of those presently used in Nigeria including those used as tiles adhesive. This work could serve as indicator of the indoor radiation exposure of Nigerian people due to cement.

In the light of the diverse application of cement in Nigeria building construction industries and the fact that an average Nigerian spends about 80% of their time indoors, the knowledge of radiological content and associated hazard from cement is thus a necessity. The aim of this study is to quantify the natural radionuclide content of cements available in local market in Nigeria and estimate the potential radiological hazard to the dwellers of buildings constructed from such cements. The data from this study may be used by Nigerian authority for the development and implementation of radiation protection guidelines for the use and management of cements in the country. The data in this work could also assist builders in Nigeria in considering radiological factor when making choices for cements rather than the traditional factors of cost and availability.

2. Materials and methods

2.1 Sample Collection

In order to collect cement brand samples that represent a fair proportion of major cement brands used in Nigeria, a survey was carried out. The survey includes visiting major cement suppliers and sites where buildings were under construction throughout the six geopolitical zones in Nigeria. The survey revealed that the acceptability of a particular brand of cement is dictated by its availability and cost. The availability of a particular brand in a region is closely related to the proximity to the manufacturer or distributor and the cost of all local packaged cement brands are almost the same. The survey revealed that 9 grey cements and 5 white cements used as tile adhesive are generally used in different parts of Nigeria. The color of each of the brands is given in table 1. The cement brands and the places in Nigeria where they are mostly used are shown in fig. 1. 6 samples of each brand of the 14 brands were collected from major suppliers and building construction sites. The samples were collected into clean plastic containers, sealed, marked and transported to the laboratory.

2.2 Gamma spectrometry

Six samples of each of the 14 brands of cements collected from construction site and major distributors in locations across the country where they are mostly found as shown in figure 1 were collected for radioactivity measurement. Each sample was air dried and pulverized into powdered form. 200 g of each powdered sample was put in a cylindrical polystyrene container and sealed with tapes to prevent radon permeability and left for more than four weeks in order to allow for radon and its short lived progeny to reach radioactive equilibrium. After this period, the radioactivity measurement was carried out for 7 h using a 7.6cm × 7.6cm NaI(TI) detector with a resolution of 8% at 662 keV and housed in 10cm thick lead shield to reduce background gamma radiation. The power supply and the data acquisition of the energy spectra were achieved by using an integrated specrtroscopy system from Bicron. The system utilized SAMPO S 100 software package from Canberra. The energy calibration of the detector was performed using IAEA standard point sources (^{109}Cd, ^{57}Co, ^{137}Cs, ^{54}Mn, and ^{22}Na) of gamma energy range between 83 keV and 1275 keV being the energy range of the radionuclide to be identified. To simulate the cement samples, 200 g of IAEA-375 reference soil was used. The radioactivity concentrations of ^{226}Ra were determined from the photopeaks of 609.32 keV (^{214}Bi), 1120.20 keV (^{214}Bi) and 352.6 keV (^{214}Pb) and that of ^{232}Th from 969.3 keV (^{228}Ac) and 583.78 keV (^{208}Tl) while the radioactivity of ^{40}K was determined from 1460.3 keV photopeak following the decay of ^{40}K.

3. Results and discussion

3.1. Radionuclide concentrations

The measured activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in various brands of cement considered in this work is given in table1. From the table the mean activity concentrations varied generally from 28.8 Bq.kg^{-1} to 49.5 Bq.kg^{-1}, from 19.4 Bq.kg^{-1} to 32.8 Bq.kg^{-1}, and from 205.9 Bq.kg^{-1} to 452.9 Bq.kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K respectively. The mean concentrations of the grey ordinary Portland cement (OPC) brands varied from 24.9 Bq.kg^{-1} to 46.3 Bq.kg^{-1}, from 19.4 Bq.kg^{-1} to 32.5 Bq.kg^{-1}, from 205.9 Bq.kg^{-1} to 421.9 Bq.kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K respectively. While the mean concentrations of the white cement used as tile adhesive varies from 35.5 Bq.kg^{-1} to 49.5 Bq.kg^{-1}, from 27.4 Bq.kg^{-1} to 32.8 Bq.kg^{-1}, and from 283.8 Bq.kg^{-1} to 452.9 Bq.kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K respectively. The mean concentrations of the radionuclides in OPC brands were found to be low when compared with those of white cements. The variation of the mean activity concentrations of the cement brands could be attributed to the variations in the geological origin of the raw material used in their production. All the white cement brands considered in this work are imported while only Burham and Bua are imported OPC. From the result it can be concluded that imported white cements in Nigeria tend to have higher activity concentrations when compared with the local OPC brands. Generally, the mean activity concentration of ^{40}K was the highest in all the samples when compared with the other two radionuclides. This is typical and expected from any geologically derived material due to the relative abundance of ^{40}K in the natural environment (IAEA, 2003). The range of radionuclide concentrations in the cement brands were found to be below world average of 50 Bq.kg^{-1}, 50 Bq.kg^{-1}, and 500 Bq.kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K respectively in building materials (UNSCEAR, 1993), while the upper limits of the range was found to be higher than the world average of 35 Bq.kg^{-1} 30 Bq.kg^{-1} and 400 Bq.kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K respectively in soil (UNSCEAR, 2000).

3.2 Radium equivalent activity

Radium equivalent activity is a single quantity that combines the radiological effects of ^{226}Ra, ^{232}Th, and ^{40}K in material used for buildings (Krisiuk, et al., 1971; OECD, 1979; Beretka and Mathew 1985; Roy, et al., 2000; Sam and Abbas, 2001). It is a weighted sum of activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K based on the assumption that 370 $Bq.kg^{-1}$ of ^{226}Ra, 259 $Bq.kg^{-1}$ of ^{232}Th, and 4810 $Bq.kg^{-1}$ of ^{40}K produce the same gamma radiation dose rates (Krisiuk, et al., 1971 and OECD, 1979). The radium equivalent is calculated using equation (Beretka and Mathew 1985);

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_{K} \quad (1)$$

where C_{Ra}, C_{Th}, and C_{K} are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K respectively. Any material whose radium equivalent activity concentration exceeds 370 $Bq.kg^{-1}$ is not recommended for safe use as building material (Sam and Abbas, 2001). The evaluated Ra_{eq} for the cement is given in table 2. For the grey cement, the mean Ra_{eq} varies from 84.6 $Bq.kg^{-1}$ to 114.6 $Bq.kg^{-1}$, while for the white cement it varies from 101.5 $Bq.kg^{-1}$ to 125.5 $Bq.kg^{-1}$. Generally, the Ra_{eq} for all the cement considered is less than the limit, 370 $Bq.kg^{-1}$. A comparison of the mean radium equivalent activities in this work and those obtained from recent published work from other countries is given in table3. The variation in the Ra_{eq} from other countries can be attributed to the difference in the Geology and consequent geochemical constituent of the rock from which the cements were derived.

3.3 Internal and external gamma indices

Other indices used for testing the suitability of any material for safe use as building material are the external hazard (H_{ex}) and internal hazard (H_{in}) indices which are defined according to Beretta and Mathew (1985) as:

$$H_{ex} = {C_{Ra}}/{370} + {C_{Th}}/{259} + {C_{K}}/{4810} \quad (2)$$

$$H_{in} = {C_{Ra}}/{185} + {C_{Th}}/{259} + {C_{K}}/{4810} \quad (3)$$

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

The activity limits in terms of these limits is 1 for safe use of the material in building construction.
The external gamma indices for all the cements considered in this work are less than unity. The obtained values of the external hazard index for all the samples are given in table 2. It varies from 0.21 for Sokoto cement locally produced, to 0.34 for magen roi, an imported white cement.

The internal hazard index (H_{in}) quantifies the internal exposure to carcinogenic radon and its short lived progeny. The values of the calculated H_{in} for the various brand of cement are also given in table 2, and are all less than 1.

3.4 annual gnadal dose equivalent (AGDE)

Sinnce the gonads are considered as the organs of interest, together with the active bone marrow and bone surface cells (UNSCEAR, 1988), the annual gonadal dose equivalent (AGDE, µS y^{-1}) due to the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K was calculated by (Al-Jundi et al., 2006):

$$AGDE = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_{K} \quad (4)$$

According to this model, a house is considered as a cavity with infinitely thick walls which makes it possible to make comparison of AGDEs of a house whose materials contains concentrations of ^{226}Ra, ^{232}Th, and ^{40}K with that of the world average of 35, 30, and 400 $Bqkg^{-1}$ respectively in soil (UNSCEAR, 2000). The implication of this is that, if a building has materials whose radioactivity of the three primordial radionuclides is lower than that of the world average, such building could act as a radiation shield for the inhabitant. On the other hand, if the radioactivity is higher than the world average, the building itself could be a source of radiation to the inhabitant whose AGDE would then be greater than the world average of 359.2 μSvy^{-1} in soil. The mean AGDE of the OPC varies from 239 μSvy^{-1} to 377.6 μSvy^{-1} with a mean of 311 μSvy^{-1}. Of all the 9 OPC only Burham and Bua has AGDE greater than the world average value. For the white cements the AGDE varies from 329.8 μSvy^{-1} to 414.8 μSvy^{-1} with JK and Magen roi having AGDE values greater than the world average.

4. Conclusion

The natural radionuclide content and their consequent radiation hazard indices were evaluated in grey and white cement used in Nigeria. Although the mean specific activities of imported cement brands are higher than the locally produced ones, their total mean activities were less than the world average in building materials. The radium equivalent activities obtained for all the cement brands considered in this work were all below the criterion limit of radiation dose (1.5 mSv/y). Calculations of both internal and external gamma indices showed that no sample exceeded the recommended exemption levels of unity.

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Table 1 Specific activities and color of various Nigerian portland cement brands.

Cement brand	Color	^{226}Rn	^{232}Th	^{40}K
Dangote	Grey	33.9 ± 16.2	32.5 ± 12.4	289.7 ± 79.3
ABS	white	40.2 ± 13.9	27.4 ± 9.82	324.9 ± 63.9
Burham	Grey	39.5 ± 18.7	29.5 ± 11.6	421.9 ± 89.1
Rak white	white	35.5 ± 17.3	29.8 ± 9.7	304.1 ± 69.9
Moulder3	White	37.5 ± 23.9	32.8 ± 21.8	283.8 ± 65.6
JK	white	49.5 ± 19.7	28.4 ± 11.2	346.3 ± 53.8
Maggen roi	white	46.7 ± 14.6	30.7 ± 8.8	452.9 ± 55.5
Unicem	Grey	43.3 ± 15.4	30.1 ± 9.9	297.5 ± 52.8
Sokoto	Grey	28.8 ± 9.9	22.3 ± 8.8	205.9 ± 47.6
Eagle	Grey	31.1 ± 11.4	26.0 ± 10.0	212.7 ± 57.9
Ashaka	Grey	24.9 ± 10.3	21.8 ± 8.9	227.1 ± 39.7
Ibeto	Grey	37.2 ± 12.7	27.4 ± 9.8	298.8 ± 48.5
Bua	Grey	46.3 ± 12.5	32.5 ± 13.0	295.8 ± 27.7

Atlas	Grey	39.5 ± 16.6	19.4 ± 8.3	254.8 ± 68.3

Table2. Mean radiation hazard indices of Nigerian Portland cement

Sample	Ra_{eq} (Bq kg^{-1})	H_{ex}	H_{in}	AGDE(μSvY^{-1})
Dangote	102.7	0.28	0.37	331.8
ABS	104.4	0.28	0.39	340.8
Burham	114.1	0.31	0.41	377.6
Rak white	101.5	0.27	0.37	329.8
Moulder3	106.2	0.29	0.39	342.0
JK	116.7	0.32	0.45	380.2
Magen roi	125.5	0.34	0.47	414.8
Unicem	109.3	0.30	0.41	353.1
Sokoto	76.5	0.21	0.28	246.8
Eagle	84.6	0.23	0.31	271.5
Ashaka	73.5	0.20	0.27	239.3
Ibeto	99.4	0.27	0.37	323.4
Bua	115.6	0.31	0.44	371.9
Atlas	86.9	0.23	0.34	283.4

Table 3. Comparison of mean radium equivalent Ra_{eq} (Bq kg^{-1}) in Portland cements

Country	Ra_{eq} (Bq kg^{-1})	References
Malaysia	188	Ibrahim (1999)
Zambia	79	Hayumbu et al., (1995)
South Korea	80.8	Lee et al., (2001)
China	127.7	Xinwei(2004)
Greece	221.6	Papaefthymiou and Gouseti (2008)
India	580.1	Sonkwade et al. (2008)
Egypt	291.9	Ahmed (2005)
Lebanon	93.8	Kobeiss et al. (2008)
Turkey	246.1	Baykara et al. (2011)
Ghana	90.1	Kpeglo et al., (2011)
Nigeria	101.2	This work

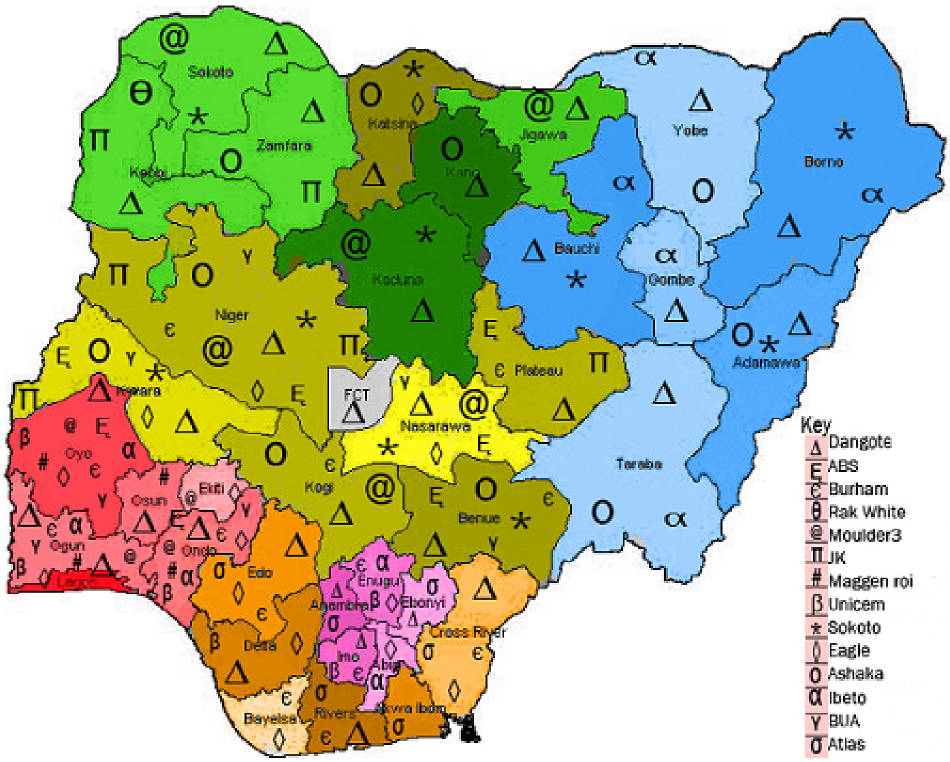

Fig.1. Distribution of major cement brands in Nigeria.

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