

Radioactivity concentrations in common brands of cement in Nigeria

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ABSTRACT Radioactivity concentrations of ^{238}U , ^{232}Th and ^{40}K in common brands of Portland cement in Nigeria have been measured using a shielded 7.6 cm \times 7.6 cm NaI(Tl) detector coupled to a Canberra series 10 plus multichannel analyzer. The cement samples show significant spread in their radioactivity concentrations, even among samples of the same brand. The weighted means of the activity concentrations of ^{238}U , ^{232}Th and ^{40}K in the different brands were 52.4, 4.1 and 91.8 Bq/kg, respectively. The radium equivalent ranged between 29.1 and 128.1 Bq/kg with a weighted mean value of 68.5 Bq/kg. The values are lower than corresponding values in cement samples in most other countries as contained in recent publications. They are also lower than the UNSCEAR recommended maximum of 370 Bq/kg. The safety criteria developed by ITB Poland for radioactivity in building materials, especially for radon build-up in homes were however, exceeded in some of the cement samples.

Keywords: cement / Nigeria / gamma spectroscopy / radioactivity concentration / radiation exposure

RÉSUMÉ Radioactivité des ciments de grande diffusion au Nigéria.

Les concentrations radioactives en ^{238}U , ^{232}Th et ^{40}K dans des ciments de Portland de grande diffusion au Nigéria ont été mesurées en utilisant un détecteur NaI(Tl) de 7,6 cm \times 7,6 cm couplé à un analyseur multicanal Canberra série 10+. Les échantillons de ciment montrent une dispersion significative de la radioactivité au sein même d'une même marque. Les moyennes pondérées des concentrations en activité de ^{238}U , ^{232}Th et de ^{40}K dans les différentes marques étaient 52,4, 4,1 et 91,8 Bq/kg, respectivement. L'équivalent en radium varie entre 29,1 et 128,1 Bq/kg avec une valeur moyenne pondérée de 68,5 Bq/kg. Ces valeurs sont inférieures à celles mesurées dans des échantillons de ciment de la plupart des autres pays qui sont rapportées dans les publications récentes. Elles sont également inférieures à la valeur maximum de 370 Bq/kg recommandée par l'UNSCEAR. Les critères de sûreté développés par ITB Pologne pour la radioactivité des matériaux de construction, particulièrement pour le radon dans les maisons étaient cependant dépassés dans certains échantillons de ciment.

1. Introduction

Radioactivity in cement, a vital component of building materials, is of significant importance in the assessment of population exposures. This is more so because most

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individuals spend up to 80% of their time indoor. Elevated indoor external dose rates may result from high activities of radionuclides in building materials (Zikovsky and Kennedy, 1992). A radium equivalent of 370 Bq/kg in building materials will produce an exposure of about 1.5 mSv/y to the inhabitants (UNSCEAR, 1982). Internal exposure may also arise from inhalation of radon, a naturally occurring radioactive gas found in soils, rocks and water. Two isotopes of this gas, ^{222}Rn ($T_{1/2} = 3.8$ d) and ^{220}Rn , ($T_{1/2} = 55.6$ s) are gaseous members of ^{238}U and ^{232}Th decay series, respectively. Being a gas, radon escapes readily from the walls and floors into the atmosphere and accumulate in indoor area. Since ^{238}U , ^{232}Th and ^{40}K are ubiquitous radionuclides in the earth, the source of most raw materials for cement production; these radionuclides may find their ways into cement.

As a developing nation, the use of cement plays an important role in the national economy of Nigeria. This is evident in the yearly consumption of cement which is about 10 million tones, corresponding to about 75 kg/capita. The consumption has been on the increase at annual rates of between 10 and 15% (Esubiyi, 1995). Accurate knowledge of radioactivity concentration in cement is important for checking its radiological effects on man and on the environment. Data are however, still quite sparse on the radioactivity of the different brands of cement in Nigeria (Farai and Ademola, 2005). The objective of this work is to measure the radioactivity concentration of ^{238}U , ^{232}Th and ^{40}K in the various brands of Portland cement found in the Nigerian market and assess the health implications of the use of such cement products for the construction of buildings.

2. Materials and method

2.1. Sample collection and preparation

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). There is presently no accurate statistics on the number and production capacities of the local cement manufacturing companies. This is so because since the downturn of the economy in the last one decade, many of business ventures merely import finished cement products in bulk from their countries of origin and bag them with different brand names.

For the present survey, samples of ten common brands of cement were collected from the open market. The sampling exercise covered mainly South Western (SW) geopolitical zone and the Federal Capital Territory (FCT) where most of the brand names were found. However, samples of some brands found to be predominant in the North Central (NC) and South Eastern (SE) geopolitical zones were also

Table I
Sampling data and ranges of radioactivity concentrations.
Informations sur les prélèvements et variations des concentrations en radioactivité.

Brand	No of samples	Sampled geopolitical zones	Activity concentrations (Bq/kg)		
			^{40}K	^{238}U	^{232}Th
Dangote	4	SW, FCT	150.3–180.2	65.3–85.3	5.0–6.2
Elephant	6	SW, FCT	BDL–23.4	80.7–95.9	5.4–6.4
Ashaka	2	FCT	88.3–113.9	62.0–68.0	4.6–4.9
Rock	2	NC	72.0–98.8	6.5–27.6	5.1–6.2
Burcham	4	FCT, SW	88.7–132.9	BDL–15.6	BDL–10.5
Continental	4	FCT, SW	95.5–116.7	39.4–150.4	BDL–6.4
Gateway	4	SW	106.1–127.0	76.2–82.0	5.2–6.1
Elepvont	2	NC	118.5–159.3	6.4–15.9	4.8–5.4
Super camel	4	NC, SE	140.2–181.5	21.4–35.0	5.0–8.0
Aalborg white	2	NC	62.1–95.6	29.2–55.3	4.8–6.5

BDL – Below Detection Limit.

collected. In all, 34 samples, comprising of two from each sampling point, were investigated. The number of samples of each brand collected, shown in Table I, is fairly representative of the relative abundance of each brand in the market. The Elephant brand was found to be quite ubiquitous. Apart from Aalborg White which was labeled as being imported from Denmark, it could not be clarified if all samples of the same brand were from the same bulk source. The samples were pulverized and packed 200 g by mass in clean radon impermeable cylindrical plastic containers which are 8 cm in height and 7 cm in diameter such that they fit the NaI detector crystal with good geometry. The containers were then hermetically sealed for upwards of 2 months to ensure secular equilibrium between ^{226}Ra (^{238}U series) and ^{232}Th and their respective gaseous gamma emitting progenies.

2.2. Experimental methods

All measurements were made in a low-level gamma counting spectroscopy comprising of a 7.6 cm \times 7.6 cm NaI (Tl) detector (Model 8020 by Bicorn), which is coupled to a Canberra Series 10 plus Multichannel Analyzer (MCA) (Model 1104) through a preamplifier base. The detector assembly has a resolution of about 8% at 0.662 MeV of ^{137}Cs which is capable of distinguishing the gamma ray energies of interest in the mixed spectrum of a typical cement sample. A reference pulverized soil sample, prepared from Rocketdyne Laboratories, California, USA which is traceable to a mixed standard gamma source (No. 48722-356) by Analytics

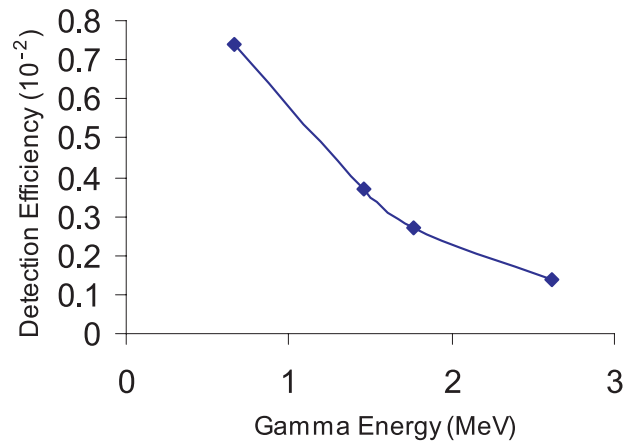


Figure 1 – *Detection efficiency as a function of gamma energy.*
Efficacité de détection en fonction de l'énergie gamma (en MeV).

Inc., Atlanta, Georgia was counted for 10 hrs to determine the detection efficiency E_p of the system, which at constant matrix and geometry, is defined as:

$$E_p = \frac{C}{tA_r Y m} \quad (1)$$

where C is the net count above the background after counting a reference sample of known activity A_r (Bq/kg) and mass m (kg) for a time t (s) and Y is the gamma yield. The efficiency was determined for the energy range 0.66–2.62 MeV. The reference sample has a fairly similar matrix as the pulverized cement samples. The cement samples too were counted for 10 hrs (36,000 s) at same geometry.

The efficiency of the detection system as a function of gamma energy is shown in Figure 1. The detection limits of the detection system (IAEA, 1989; Oni, 2004) are given in Table II together with the gamma energies used for the measurement of the three primordial radionuclides. The radioactivity concentrations of the radionuclides in each cement sample were obtained by relating the detection efficiency obtained from calibration, to the net count under each photopeak at the end of each counting exercise.

3. Results and discussion

The ranges of radioactivity of each radionuclide in the ten brands are given in Table I. The arithmetic means are presented in Table III with the standard deviations given as the errors. The standard deviations give a measure of the spread in the

Table II
Radionuclide detection limits of the system.
Limites de detection du système de mesure.

Radionuclide	Energy (MeV)	Detection efficiency (λ_γ) $\times 10^{-2}$	Detection limit (Bq/kg)
^{137}Cs	0.662	1.120	1.05
^{40}K	1.460	0.441	11.57
^{214}Bi (^{226}Ra)	1.760	0.271	8.72
^{208}Tl (^{228}Ra)	2.615	0.242	4.43

Table III
Radioactivity concentrations and radiological health indices.
Concentration radioactive et critères de protection.

S/No	Brand	^{40}K (Bq/kg)	^{238}U (Bq/kg)	^{232}Th (Bq/kg)	Ra_{eq} (Bq/kg)	H_{ex}	F_1
1	Dangote	163.0 \pm 12.5	77.3 \pm 8.5	5.6 \pm 0.5	97.9 \pm 10.1	0.26	0.27
2	Elephant	13.5 \pm 8.5	75.9 \pm 8.1	5.8 \pm 0.5	85.2 \pm 9.4	0.23	0.23
3	Ashaka	101.1 \pm 18.1	65 \pm 4.2	4.7 \pm 0.2	79.5 \pm 5.8	0.21	0.22
4	Rock	85.4 \pm 14.9	17.0 \pm 14.9	5.7 \pm 0.8	31.7 \pm 17.1	0.08	0.08
5	Burcham	113.2 \pm 18.3	10.7 \pm 4.0	7.5 \pm 2.8	30.1 \pm 9.3	0.08	0.09
6	Continental	103.8 \pm 10.7	112.7 \pm 63.7	5.2 \pm 1.7	128.1 \pm 66.9	0.35	0.36
7	Gateway	117 \pm 9.7	79.3 \pm 3.1	5.7 \pm 0.4	96.5 \pm 4.4	0.25	0.26
8	Elepvont	139.0 \pm 29.0	11.15 \pm 6.7	5.1 \pm 0.4	29.1 \pm 9.3	0.07	0.08
9	Super Camel	160.6 \pm 20.7	28.8 \pm 6.7	6.7 \pm 1.4	50.7 \pm 10.2	0.13	0.15
10	Aalborg White	78.9 \pm 23.7	42.3 \pm 18.5	5.6 \pm 1.2	56.4 \pm 21.9	0.15	0.16
	Wt. Mean	91.83	52.37	4.14	68.53		

values of radioactivity concentrations. The ranges of values are wide, resulting in large values of standard deviations. The weighted means, which give less importance to values with large errors and more importance to values with small errors, are also presented in Table III. These weighted means are compared with recently published data (El-Bahi, 2004) on other countries in Table IV. The weighted mean radioactivity concentrations in cement samples from Nigeria are less than the values for most of the countries. The values obtained for Th and K are much less than the respective global average concentrations of 41.0 and 230 Bq/kg (UNSCEAR, 1982). The weighted mean for U in the cement samples is interestingly, in very close proximity to the global average of 52.2 Bq/kg.

The weighted mean concentrations of the radionuclides in Table III are in the decreasing order $C_{\text{Th}} < C_{\text{U}} < C_{\text{K}}$. This order of activity concentrations is however

Table IV
Comparison levels* of radionuclides for cement dry weight (in Bq/kg).
Comparaisons des niveaux de concentrations en radionucléides de différents ciments (en Bq/kg).

Country region	²³⁸ U	²³² Th	⁴⁰ K
California	-	8.2	73
USSR	44.4	44.4	222
Shenzhen	55.2	26.4	175
Hong Kong	19.2	18.9	127
Egypt	34.6	19.2	93
Global Average	52.2	41.0	230
Nigeria (present data)	52.4	4.1	91.8

* Source: El-Bahi (2004).

not necessarily the order of their radiological health importance. In order to examine the total exposure caused by the three radionuclides, their radioactivity concentrations were combined in terms of radium equivalent, which is defined as:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (2)$$

where C_{Ra} , C_{Th} and C_K are the respective radioactivity concentrations of U (Ra), Th and K. The mean values of the radium equivalent and the associated errors due to the spread of the values are given in Table III. The overall weighted mean of the radium equivalent has been estimated with equation (2) from the values of the weighted mean of the radionuclides. This was estimated as 68.53 Bq/kg, which is much less than the maximum permissible limit of 370 Bq/kg (UNSCEAR, 1982). The world average from Table IV is about 130 Bq/kg.

The radium equivalent, which gives the relative level of exposure rate caused by each brand of cement, is displayed in Figure 2. Continental brand gives the highest level of radiation exposure rate. Its ²³⁸U concentration (112.7 ± 63.7 Bq/kg) is much higher than the world average but its concentrations of the other radionuclides are much less with the result that its radium equivalent is much less than the maximum permissible level. Dangote brand has the next highest value of radium equivalent and also the next highest value of U concentration. Rock, Burcham and Elepvont brands have the least radium equivalents of about 30 Bq/kg each. The lowest concentrations of K (13.5 ± 8.5 Bq/kg), U (10.7 ± 4.0 Bq/kg) and Th (4.7 ± 0.2 Bq/kg) were measured in the Elephant, Burham and Ashaka brands, respectively.

RADIOACTIVITY CONCENTRATIONS IN COMMON BRANDS OF CEMENT IN NIGERIA

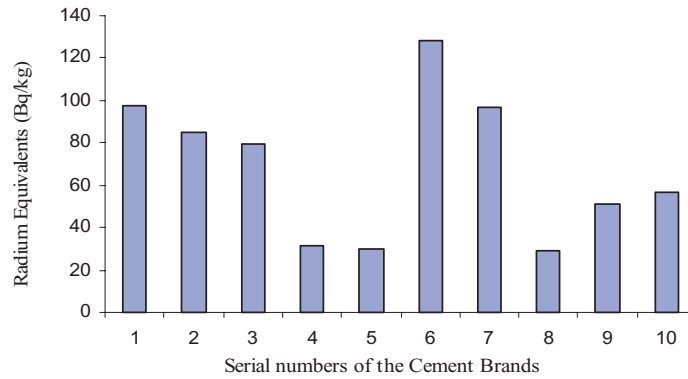


Figure 2 – Radium equivalents.
Équivalents radium.

The external hazard index H_{ext} which estimates the potential radiological hazard posed by the different samples was calculated using the following equation

$$H_{ext} = 0.0027C_{Ra} + 0.00386C_{Th} + 0.000208C_K \quad (3)$$

where C_{Ra} , C_{Th} and C_K are the activity concentrations of U, Th and K in Bq/kg, respectively. It is a dimensionless quantity and a safety criterion for materials used for building construction is that $H_{ex} \leq 1$ (UNSCEAR, 1993). The values of H_{ex} obtained for the various brands of cement surveyed in this work are presented in Table III. The values of H_{ex} ranged between 0.07 and 0.26, showing that the safety criterion was met by all the brands.

Two criteria developed in Poland for the evaluation of radiological hazard posed by building materials to the environment are the qualification coefficients F_1 (no unit) and F_2 (Bq/kg) (El-Bahi, 2004). The two quantities are defined (ITB/CLOR, 1995) as:

$$F_1 = 0.0027C_{Ra} + 0.0043C_{Th} + 0.00027C_K \quad (4)$$

$$F_2 = C_{Ra} \quad (5)$$

where C_{Ra} , C_{Th} and C_K are the values of concentration of ^{238}U , ^{232}Th and ^{40}K in Bq/kg, respectively. The quantity F_1 is dimensionless while F_2 has the dimension of Bq/kg. The safety criteria recommended by ITB, Poland are that $F_1 \leq 0.29$ and $F_2 \leq 50$ Bq/kg. The values of F_1 obtained from the present survey ranged between 0.08 and 0.36 as shown in Table III. It can be observed from the same table that F_2 (U concentration) ranged between 11.15 and 112.7 Bq/kg. It can be observed that

the values obtained are within acceptable range for F_1 and F_2 except in the Continental Brands for which the value of F_1 was 0.36. The value of F_2 for the Continental brand of cement (112.7 Bq/kg) is more than double the above Polish value of recommended maximum. The values of F_2 in the Dangote, Elephant and Ashaka brands as well as the weighted mean for all the ten brands, exceed the Polish recommended maximum value. The value of F_2 is a major determining factor in radon levels in homes constructed with these brands of cement. However, data are presently very sparse on radon levels in Nigerian homes and hence, there is for now no policy on radon levels in homes. The Polish criterion cannot be adopted or rejected without such data. Further work is urgent in this area.

4. Conclusion

Radioactivity levels in ten common brands of cement available in the open market in Nigeria have been measured in a low-level gamma counting spectroscopy. The values obtained are within UNSCEAR and ITB Poland safety criteria for radiation exposure due to radioactivity in building material, suggesting that the cement samples pose no major hazard of external radiation exposure. The criterion for safety from the point of view of radon build-up in dwellings was exceeded in more than half the number of cement sample surveyed. There is an urgent need for more studies on radon levels in homes.

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