

## Radiation dose estimation from the radioactivity analysis of cement used in Nigeria

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Studies have been carried out using gamma-spectrometric techniques to determine the natural radioactivity in cement used in Nigeria and associated radiation hazard. The mean values of  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  are 140.5, 30.1 and 62.7  $\text{Bqkg}^{-1}$  respectively. The measured activity concentrations of these radionuclides were compared with the reported data of other countries. The outdoor dose rates, indoor dose rates, annual outdoor effective dose rates, annual indoor effective dose rates, external and internal hazard indices, Radium equivalent and representative level index were calculated and compared with the internationally approved values. The  $R_{\text{aeq}}$  and  $I_{\text{gamma}}$  values of all the studies samples are in agreement with the internationally accepted values. The values of both external and internal hazard indices are less than unity. The use of cement as building materials in Nigeria is in accord with the OECD criterion.

### I. INTRODUCTION

There has been an increasing demand for cement production throughout the world with the ever-increasing growth in human civilization. Portland cement is the most common type of cement in general usage in many parts of the world, as it is a basic ingredient of concrete, mortar, stucco and most non-specialty grout. It is a fine powder produced by grinding Portland cement clinker (more than 90%), a limited amount calcium sulphate which controls the set time, and up to 5% minor constituents [1]. Since cement has been used as building materials, and several research has been done to investigate the radionuclides present in cement. This area is still a point of interest in the western world and the developing countries such as Nigeria because of the danger in radiation poses to the health of the populace. It is therefore imperative to study in detail, the radiological characteristics of the various cement use in Nigeria. Furthermore, detailed knowledge of the radiological characteristics in cement allows better determination of the radiation exposure, both occupational and of the public, due to slag, fly ash, silica fume, klin, car and truck tyres used as raw materials during cement production [2]. Recently, considerable attention has been given to low-level exposures arising from natural radioactive isotopes present in all building materials particularly  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  [3-22]. The main objectives of the present work are:

- (i) Radiometric study of cement and determine the concentrations of naturally occurring radionuclides (U, Th and K) in cement.

- (ii) To determine the dose emitted from cement in order to assess the radiological impact of cement on the populace.
- (iii) Estimation of hazard indices with implication to the preparation concrete for buildings and roads construction.

### II. EXPERIMENTAL

The analysed cement samples were collected in various areas. Table I shows the various brands collected and the areas. The samples were coded to avoid mix up. They were then taken to the laboratory from their collection points. The collected samples were stored in polyethylene bags. The samples were dried for 24 hr in an air circulation oven at 60°C [21]. About 250 g of each sample was packed in a plastic containers and sealed for 15 days before radioactive determination of Uranium and Thorium to attain radioactive equilibrium with their daughter products and to prevent radon loss. After attainment of secular equilibrium between  $^{232}\text{Th}$ ,  $^{238}\text{U}$  and their daughter products, the samples were subjected to gamma-ray spectrometric using 7.6 cm by 7.6 cm NaI (TL) crystal detector coupled to a 12.7 cm dia photomultiplier tube and the associated data acquisition and analysis system. The detector was used to estimation of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in cement from Nigeria. This was carried out at the centre for energy research and development situated at Obafemi Awolowo University Ile-Ife. The energy resolution of the detector was 8% ( $^{137}\text{Cs}$  661 KeV) and 25% efficiency. The detector is maintained in a vertical position in a lead cylindrical shield of 10 cm thickness.

**TABLE I.** Samples and locations.

| Samples       | Locations     |
|---------------|---------------|
| Burham (6)    | Lagos         |
| Eagel (6)     | Port_harcourt |
| Dangote (6)   | Ado-Ekiti     |
| Ashaka (6)    | Egor          |
| Elephant (6)  | Ado-Ekiti     |
| Ibeto (6)     | Awka          |
| Diamond (6)   | Akure         |
| Gold (6)      | Hesa          |
| Wapco (6)     | Gboko         |
| Horseband (6) | Abeokuta      |

Natural radionuclides of relevance to this work are mainly  $\gamma$ -ray emitting nuclei in the decay series of  $^{232}\text{Th}$ ,  $^{238}\text{U}$  and  $^{40}\text{K}$ , while  $^{40}\text{K}$  can be measured by its own  $\gamma$ -rays,  $^{232}\text{Th}$  and  $^{238}\text{U}$  are not  $\gamma$ -ray emitters. However it

is possible to measure  $\gamma$ -rays of their decay products. Decay product for  $^{238}\text{U}$  ( $^{214}\text{Bi}$ , 2.615 MeV) and  $^{232}\text{Th}$  ( $^{228}\text{Ac}$ , 1.765 MeV) were used by assuming the decay series to be in secular equilibrium. Although activity concentrations of  $^{40}\text{K}$  were measured by its own  $\gamma$ -rays (1.465 Mev). The absorbed gamma dose rate in air 1 m above the ground surface for uniform distribution of radionuclide was computed on the basis of Beck *et al.* relationship [23,24].

The values are

$$D_{\text{out}} (\text{nGyh}^{-1}) = 0.0417A_{\text{k}} + 0.462A_{\text{Ra}} + 0.604A_{\text{Th}}$$

where  $A_{\text{k}}$ ,  $A_{\text{Ra}}$  and  $A_{\text{Th}}$  are specific activities concentration of  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  in  $\text{Bqkg}^{-1}$  respectively. Radioactivity and absorbed dose rates in air from cement are given in Table II.

In order to estimate the annual effective dose rates; the conversion coefficient from the absorbed dose in air to effective dose ( $0.7 \text{ SvGy}^{-1}$ ) was used [23,24].

The outdoor and indoor occupancy factor (0.2 and 0.8) proposed by UNSCEAR were used. The outdoor and indoor effective dose rates were calculated using the formulae

$$\text{Outdoor Effective dose rate (mSvy}^{-1}) = D_{\text{out}} (\text{nGyh}^{-1}) \times 8760(\text{hy}^{-1}) \times 0.2 \times 0.7 (\text{SvGy}^{-1}) \times 1\text{E}-6 [23,24]$$

$$\text{Indoor Effective dose rate (mSvy}^{-1}) = D_{\text{in}} (\text{nGyh}^{-1}) \times 8760 (\text{hy}^{-1}) \times 0.8 \times 0.7 (\text{SvGy}^{-1}) \times 1\text{E}-6 [23,24].$$

**TABLE II.** Radioactivity in cement, dose rate, annual effective dose rate ( $\text{mSvy}^{-1}$ ), radium equivalent activity ( $R_{\text{aeq}}$ ). External land internal hazard indices ( $H_{\text{ext}}$  and  $H_{\text{in}}$ ) and representative level (I gamma r).

| $^{40}\text{K}$<br>$\text{Bqkg}^{-1}$ | $^{238}\text{U}$<br>$\text{Bqkg}^{-1}$ | $^{232}\text{Th}$<br>$\text{Bqkg}^{-1}$ | $D_{\text{out}}$<br>$\text{nGyh}^{-1}$ | $D_{\text{in}}$<br>$\text{mSvy}^{-1}$ | $D_{\text{effout}}$<br>$\text{Bqkg}^{-1}$ | $D_{\text{ineff}}$<br>$\text{Bqkg}^{-1}$ | $H_{\text{ext}}$<br>$\text{Bqkg}^{-1}$ | $H_{\text{in}}$<br>$\text{Bqkg}^{-1}$ | $R_{\text{aeq}}$<br>$\text{Bqkg}^{-1}$ | I<br>$\text{Bqkg}^{-1}$ |
|---------------------------------------|----------------------------------------|-----------------------------------------|----------------------------------------|---------------------------------------|-------------------------------------------|------------------------------------------|----------------------------------------|---------------------------------------|----------------------------------------|-------------------------|
| 185.1                                 | 34.1                                   | 0.1                                     | 23.53                                  | 28.24                                 | 0.03                                      | 0.14                                     | 0.13                                   | 0.22                                  | 48.50                                  | 0.18                    |
| 30.0                                  | 40.1                                   | 0.1                                     | 20.02                                  | 24.03                                 | 0.02                                      | 0.12                                     | 0.12                                   | 0.23                                  | 42.95                                  | 0.15                    |
| 85.4                                  | 35.1                                   | 75.0                                    | 65.08                                  | 78.09                                 | 0.08                                      | 0.38                                     | 0.40                                   | 0.50                                  | 148.93                                 | 0.52                    |
| 84.0                                  | 21.2                                   | 140.0                                   | 97.86                                  | 117.43                                | 0.12                                      | 0.58                                     | 0.62                                   | 0.67                                  | 227.87                                 | 0.80                    |
| 200.4                                 | 32.0                                   | 37.0                                    | 45.49                                  | 54.59                                 | 0.06                                      | 0.27                                     | 0.27                                   | 0.36                                  | 100.34                                 | 0.36                    |
| 89.2                                  | 31.0                                   | 111.0                                   | 85.09                                  | 102.10                                | 0.10                                      | 0.50                                     | 0.53                                   | 0.61                                  | 196.60                                 | 0.69                    |
| 66.4                                  | 27.3                                   | 71.3                                    | 58.45                                  | 70.14                                 | 0.07                                      | 0.34                                     | 0.36                                   | 0.44                                  | 134.37                                 | 0.47                    |
| 112.0                                 | 29.5                                   | 48.0                                    | 47.29                                  | 56.75                                 | 0.06                                      | 0.28                                     | 0.29                                   | 0.37                                  | 106.76                                 | 0.38                    |
| 500.3                                 | 27.2                                   | 89.2                                    | 87.31                                  | 104.77                                | 0.11                                      | 0.51                                     | 0.52                                   | 0.61                                  | 193.23                                 | 0.70                    |
| 52.0                                  | 23.3                                   | 55.2                                    | 46.27                                  | 55.58                                 | 0.06                                      | 0.27                                     | 0.29                                   | 0.35                                  | 106.24                                 | 0.37                    |

To assess the radiological hazard of cement used as building materials, the radium equivalent activity ( $R_{eq}$ ) and the external and internal indices ( $H_{ext}$  and  $H_{int}$ ) were calculated in the study.  $R_{eq}$  and  $H_{ext}$  were calculated according to Beretka and Matthew [7] as

$$R_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K$$

$$H_{ext} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \leq 1.$$

These were shown in Table II.

To address the radiation to respiratory organ due to the radioactive inert gas radon  $^{222}Rn$  a daughter product of radium and its short-lived secondary products, a formula has suggested by Krieger [25] to reduce the acceptable maximum concentration of radium to half the limit was used as follows:

$$H_{int} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 \leq 1.$$

The result was presented in Table II. The representative level index ( $I_{\gamma}$ ) was calculated using the UNSCEAR relationship as follow [12]

$$I = A_{Ra}/150 + A_{Th}/100 + A_K/1500.$$

The result was also presented in Table II.

### III. RESULTS

Radiometric analysis of cements samples are shown in Table II. The activities concentrations of  $^{40}K$  ranged from 30 (Eagle) to 500.3  $Bqkg^{-1}$  (Wapco) with an average of 140.5  $Bqkg^{-1}$ ,  $^{238}U$  ranged from 21.2 (Ashaka) to 40.5  $Bqkg^{-1}$  (Eagle) with an average of 30.1  $Bqkg^{-1}$ ,  $^{232}Th$  ranged from 0.1 (Burham), 0.1 (Eagle) to 140  $Bqkg^{-1}$  (Ashaka) with an average 62.7  $Bqkg^{-1}$ .

The calculated results show that the outdoor absorbed dose rates range from 20 (Eagle) to 97.9  $nGyh^{-1}$  (Ashaka) with mean value 57.6  $nGyh^{-1}$ , while the indoor dose rates range from 24 (Eagle) to 117  $nGyh^{-1}$  (Ashaka) with mean value 69.5  $nGyh^{-1}$ . The calculated annual outdoor effective dose rates in air varied from 0.02 (Eagle) to 0.12  $mSvy^{-1}$  (Ashaka) with mean 0.07  $mSy^{-1}$ ; while the annual indoor effective dose rates in air varied from 0.12  $mSy^{-1}$  (Eagle) to 0.58  $mSvy^{-1}$  (Ashaka) with mean 0.34  $mSvy^{-1}$ . The external hazard index ( $H_{ext}$ ) ranged from 0.12 (Eagle) to 0.62  $Bqkg^{-1}$  (Ashaka) with mean 0.35  $Bqkg^{-1}$ , while the internal hazard index ( $H_{int}$ ) ranged from 0.22 (Burham) to 0.67  $Bqkg^{-1}$  (Ashaka) with mean value 0.44  $Bqkg^{-1}$ . Radium equivalent is related to the external gamma dose and internal dose due to radon and its daughters. Radium equivalent varied from 43 (Eagle) to 227.9  $Bqkg^{-1}$  (Ashaka) with mean 130.6  $Bqkg^{-1}$ ; while representative gamma index (I) varied from 0.15 (Eagle) to 0.80  $Bqkg^{-1}$  (Ashaka) with mean 0.46  $Bqkg^{-1}$ .

### IV. CONCLUSION

Gamma-ray spectrometry study shows that cement brands in Nigeria have higher radium equivalent compared to some regions in the world. This is shown in Table III. However, the average annual outdoor effective dose rate 0.07  $mSvy^{-1}$  was lower than the average annual outdoor effective dose rate 0.46  $mSvy^{-1}$  from the terrestrial radionuclides in the world. The calculated mean outdoor dose rate was 57.6  $nGyh^{-1}$ , which was lower than the permissible level 80  $nGyh^{-1}$  [20]. The radium equivalents for all the brands were less than 370  $Bqkg^{-1}$  which is international accepted value for building residence. The values of representative gamma, external and internal indices were less than unity. Therefore the uses of various brands of cement in various construction purposes are considered to be safe for Nigerians.

**TABLE III.** Comparison of mean radium equivalent in Nigeria with various regions of the world.

| Areas     | Value     | Nigeria               |
|-----------|-----------|-----------------------|
| India     | 104.7 [5] | 130.6 (present study) |
| Australia | 114.7 [7] | 130.6                 |
| Finland   | 99.9 [6]  | 130.6                 |
| Germany   | 70.3 [25] | 130.6                 |
| Norway    | 7.4 [3]   | 130.6                 |

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