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Preliminary Investigation of Naturally Occurring Radionuclide in Some Six Representative Cement Types Commonly used in Cameroon as Building Material

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ABSTRACT

The present study aimed at the determination of the specific activity of naturally occurring radioactive materials and the evaluation of the radiological health hazards radioactivity in twenty four cement samples presenting six cement types commonly used in Cameroon for building construction have been analyzed. A high purity germanium detector (HPGe) spectrometer was used for quantification of gamma emitting radionuclide in the cements to demonstrate the radiological health hazards. Terrestrial absorbed dose rate (D), annual effective dose equivalent (AEDE), radium equivalent activity (Ra_{eq}), external (H_{ex})/internal (H_{in}) hazard index, activity gamma (I_γ) and alpha index (I_α) caused by gamma emitting natural radionuclide are determined from the obtained values of ^{226}Ra, ^{232}Th and ^{40}K. The calculated values of the absorbed dose rate and the indoor annual effective dose are slightly higher comparably to the recommended worldwide values. The details of the samples preparation procedure and the gamma-ray spectrometry technique are presented, together with the preliminary investigated results of specific activity of naturally occurring radionuclide chains for six representative cement type analyzed in this current work.

Keywords: *Radioactivity, Cement, Broad Energy Germanium Detector, Radiological Health Hazard Parameters.*

1. INTRODUCTION

Gamma radiation emitted from naturally occurring radioisotopes, such as ^{40}K and the radionuclides from the ^{232}Th and ^{238}U series and their decay products called terrestrial background radiation exist at trace levels in all ground formations. The main external source of irradiation to the human body are from ^{238}U, ^{235}U, and ^{332}Th, the parents of the three natural decay series, called the uranium (U) series, the actinium series and the thorium (Th) series, respectively. These series each consists of many daughter products which are generated through successive decay of parent radionuclides. In the three long-lived series, decay cascades produce radioactive daughter nuclides which eventually result in stable isotopes of ^{208}Pb, ^{207}Pb and ^{206}Pb.

Natural uranium is a composite of the isotopes ^{238}U (99.28%), ^{234}U (0,0057%), and ^{235}U (0.72%) While on a mass basis there is far more ^{238}U than ^{235}U in a natural sample and the activity ratio is approximately 21:1 [1]. The behaviour and distribution of these decay series radionuclides in the environment are based on their biogeochemistry and half-life ($t_{1/2}$), and the nature of their surroundings. The naturally occurring radioisotopes are present in different concentrations in sediments reflecting the origin of the sediments that are used in formation of cements. The activity concentrations of ^{226}Ra, ^{214}Bi, ^{214}Pb (from ^{238}U decay series), ^{228}Ac, ^{212}Bi, ^{212}Pb (from ^{232}Th decay series) and ^{40}K were measured using an HPGe spectrometer.

Studies conducted on natural radioactivity have shown that presence of potassium (^{40}K) and other daughter radionuclides from Thorium (^{232}Th) and Uranium (^{238}U) decay series in various components in the environment constitute potential exposure to the global population [2]. The primordial radionuclides are predominant in almost all raw and produced materials widely used in building industry including; cement, bricks, sands, tile, limestone, gypsum and those derived from rocks and soils [2, 3]. Natural radiation in building materials related to external and internal exposure. The external exposure comes from direct terrestrial gamma-rays radiation and the internal exposure from inhalation of radioactive inert gas Radon (^{222}Rn, a daughter product of ^{226}Ra). The presence of these natural radionuclides in building raw materials depend on geological/geographical conditions and the geochemical characteristics of the materials themselves.
Cement is commonly used in construction of building both in urban and rural areas in Cameroon. Due to its high production rate and widely used by the population, the radiation originating from it deserves special attention as this work seeks to achieve. Some of the raw materials used in the production of cement includes; limestone (CaCo3), shale ash and ion oxide which contain elements like gypsum, silicates and aluminates that have ionization tendency [4].

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According to the rule that exposure should be "as low as reasonably achievable", the radium equivalent, the external hazard index, the absorbed dose and the annual effective dose were assessed and compared with results of other studies and with the worldwide average value in the United Nations Scientific Committee on the Effects of Atomic Radiation report [5]. In this regard, the evaluation of natural radioactivity was determined in some cement types used in Cameroon focusing on the radiation risk assessment of the population and the radionuclides of interest. The distribution of natural radionuclide γ-ray activities and their respective annual effective dose rates, produced by ^{40}K, ^{238}U, ^{226}Ra, ^{235}U and ^{232}Th, were determined.

This subject is important in environmental radiological protection, since cements are widely used as building material. The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the different type of cements were measured by means of a Broad Energy Germanium Detector (BEGe). The most important radiological parameters including the absorbed dose rate, annual effective dose equivalent, radium equivalent activity, the external and internal hazard index, activity gamma and alpha index were also determined and evaluated for each cement sample.

2. SAMPLING AND SAMPLE PREPARATION

A total of six main types of cement manufactures (a number of four samples for each cement type) and used in building construction in Cameroon were collected. For each of the represented cement types, four samples were taken to make a composite sample. These samples were used without any process of homogenization since they are in powdered form. The samples were oven-dried at 110^{0}C for 24 hrs to ensure that moisture is completely removed. The samples were sieved, packed in plastic containers, labelled, weighted and hermetically sealed with a plastic tape. The samples containers used were selected in order to match the container geometry used for efficiency calibration. The sealed samples were then stored for 30 days to enable them attain a state of secular equilibrium with their short lived progeny.

3. EXPERIMENTAL

Each sample was subjected to a low background gamma-ray spectrometer consisting of Broad Energy Germanium Detector (BE6530) manufactured by Canberra Industries. The resolution of this detector is 0.5 keV at 5.9 keV for ^{55}Fe, 0.75 keV at 122 keV for ^{57}Co and 2.2 keV at 1332 keV for ^{60}Co. The detector is placed in a low-level Canberra Model 747 lead shield with thickness of 10 cm.

The energy distributions of the radioactive samples were generated by the computer inbuilt Multiport II Multichannel Analyzer (MCA). Each sample was counted for 86400 seconds for effective peak are statistics of above 0.1%. The background was also counted under the same conditions for 48 hrs and subtracted from the photo peak area for the measured samples during analysis process.

Following the sample analysis process, the specific activity concentration in Becquerel per kilogram (Bq.kg^{-1}) for each radionuclide was calculated automatically by Genie-2000 software based on the following equation:

$$A_{sp} = \frac{\frac{N_S}{t_s} - \frac{N_B}{t_B}}{\varepsilon(E_i) \times P_{\gamma_i} \times M_S \times C} \qquad (1)$$

where: N_s is the net counts of the radionuclide in the samples; N_B is the net counts of radionuclide in the background; $P_{\gamma i}$ is the gamma emission probability (gamma yield); $\varepsilon(E_i)$ is the peak efficiency of the detector at energy Ei; t_s is the sample counting time; t_B is the background measuring time; M_s is the mass of the sample (kg) and C is the cascade summing correction coefficient.

Assuming a state of secular equilibrium between ^{238}U and ^{232}Th and their respective decay daughter products, the following relatively intense gamma-ray transitions were used to measure the activity concentrations for the above-mentioned radionuclides.

a) ^{226}Ra concentration was calculated as a weighted mean of the activity concentrations of the gamma-rays of ^{214}Pb (295.1 keV, 351.9 keV), ^{214}Bi (609.3 keV and 1120.29 keV) and its specific gamma-ray at 186.2 keV. Interference correction due to the presence of 185.7 keV energy peak of ^{235}U has been taken into account and subtracted accordingly.
b) The gamma-ray photopeaks used for the determination of the ^{232}Th contents were 338.4 keV, 911.2 keV and 969.11 keV of ^{228}Ac and 238.6 keV of ^{212}Pb.
c) ^{40}K was directly determined by using 1460.8 (10.7%) gamma-ray.

3.1 Assessment of Dose

3.1.1 Absorbed Dose Rate in Air (*D*)

In order to assess any radiological hazard, the exposure to radiation arising from radionuclides present in cement can be determined in terms of many parameters. A direct connection between radioactivity concentrations of natural radionuclides and their exposure is known as the absorbed dose rate in the air at 1 metre above the ground surface. The mean activity concentrations of ^{226}Ra (of the ^{238}U serie), ^{232}Th, and ^{40}K (Bqkg^{-1}) in the soil samples are used to calculate the absorbed dose rate given using

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the following formula provided by UNSCEAR [6] and European Commission [7]. UNSCEAR and the European Commissions, the dose conversion coefficients were calculated for the standard room centre.

$$D\ (\text{nGy.h}^{-1}) = 0.92A_{Ra} + 1.1A_{Th} + 0.08A_K \quad (4)$$

Where D is the absorbed dose rate in nGy.h^{-1}, A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K, respectively. The dose coefficients in units of nGy.h^{-1} per Bq.kg^{-1} were taken from the UNSCEAR (2000) report [6-8] and determined by the Monte Carlo simulation using the model standard room.

3.1.2 Annual Effective Dose Equivalent (AEDE)

The absorbed dose rate in air at 1 metre above the ground surface does not directly provide the radiological risk to which an individual is exposed [9]. The absorbed dose can be considered in terms of the annual effective dose equivalent from indoor terrestrial gamma radiation which is converted from the absorbed dose by taking into account two factors, namely the conversion coefficient from absorbed dose in air to effective dose and the indoor occupancy factor. The annual effective dose equivalent can be estimated using the following formula [6, 10]:

$$AEDE\ (\mu\text{Sv.y-1}) = D\ (\text{nGy.h}^{-1}) \times 8760\ \text{h} \times 0.8 \times 0.7\ \text{Sv.Gy}^{-1} \times 10^{-6} \quad (2)$$

The values of those parameters used in the UNSCEAR report (2000) are 0.7 Sv.Gy^{-1} for the conversion coefficient from absorbed dose in air to effective dose received by adults and 0.8 for the indoor occupancy factor [6].

3.1.3 Radium Equivalent Activity (Ra_{eq})

Due to a non-uniform distribution of natural radionuclides in the soil samples, the actual activity level of ^{226}Ra, ^{232}Th and ^{40}K in the samples can be evaluated by means of a common radiological index named the radium equivalent activity (*Raeq*) [9, 11]. It is the most widely used index to assess the radiation hazards and can be calculated using Equation 4 given by Beretka and Mathew (1985). This estimates 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-ray dose rate [11].

$$Ra_{eq}\ (\text{Bq.kg}^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (3)$$

Where A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in Bq.kg^{-1}, respectively.

The permissible maximum value of the radium equivalent activity is 370Bq.kg^{-1} [6, 8] which corresponds to an effective dose of 1 mSv for the general public and to the radiation dose rate of 1.5 mGy.y^{-1} [6, 12].

3.1.4 External and Internal Hazard Index (Hex and H_{in})

To limit the radiation exposure attributable to natural radionuclides in the samples to the permissible dose equivalent limit of 1 mSv.y^{-1}, the external hazard index based on a criterion have been introduced using a model proposed by Krieger [13] which is given by [2, 6].

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (4)$$

In order to keep the radiation hazard insignificant, the value of external hazard index must not exceed the limit of unity. The maximum value of *Hex* equal to unity corresponds to the upper limit of radium equivalent activity 370 Bq.kg^{-1} [13, 14] measured dimensions and calculated densities.

In addition to the external hazard, radon and its short-lived products are also hazardous to the repository organs. To account for this threat the maximum permissible concentration for ^{226}Ra must be reduced to half of the normal limit (185 Bqkg^{-1}). The internal exposure to carcinogenic radon and its short-lived progeny is quantified by the internal hazard index (H_{in}) given by the expression [15].

3.1.5 Activity Concentration Index (Iγ)

Because of more than one radionuclide contribute to the dose; it is practical to present investigation levels in the form of an activity index. The European Commission has proposed in their guidance document the induction of an activity concentration index used to assess safety requirement for buildings materials [7]:

$$I_\gamma = \frac{A_{Ra}}{200(Bq/kg)} + \frac{A_{Th}}{300(Bq/kg)} + \frac{A_K}{3000(Bq/kg)} \quad (6)$$

where A_{Ra}, A_{Th} and A_K are the thorium, radium, and potassium activity concentrations (Bq/kg).

Values of index $I_\gamma \leq 0.5$ corresponds to a dose rate criterion of 0.3 mSvy^{-1}, whereas $2 \leq I_\gamma \leq 6$ corresponds to a criterion of 1 mSv/y [7, 15, 16]. Thus the material with $I_\gamma > 6$ should be avoided to use as building material, since these values correspond to the dose rates higher than 1 mSv/y which is highest then recommended values [7, 17].

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Due to radon inhalation originated from buildings materials [16]. The alpha index was determined using the following formula:

$$I_\alpha = \frac{A_{Ra}}{200(Bq/kg)} \quad (5)$$

Where A_{Ra} is the specify activity concentration of ^{226}Ra assumed in equilibrium with ^{238}U.

The recommended exemption and upper of ^{226}Ra activity concentration in building materials are 100 and 200 Bq/kg, respectively as suggested by many countries in the world [18]. These considerations reflected in the alpha index. The recommended upper limit activity concentration of ^{226}Ra is 200 Bq/kg, for which I_α =1.

4. RESULTS AND DISCUSSIONS

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K and radium equivalent activity in different cement types used in Cameroon have been calculated and presented below in Table 1. The comparison of the mean values of ^{226}Ra, ^{232}Th, ^{40}K and radium equivalent (Ra_{eq}) activities in the cement samples collected in Cameroon with data from other countries are reported in Table 2. The radiation hazard indices in the investigated cement have been estimated and summarized in Table 3.

Table 1: Specific activity concentration of ^{226}Ra, ^{232}Th and ^{40}K and radium equivalent activity in the investigated cement samples used in Cameroon

Sample N^0	Sample name	Sample number	Mean, Specific activity concentration (BqKg^{-1})			Ra_{eq} (Bqkg^{-1})
			^{226}Ra	^{232}Th	^{40}K	
01	CW1	04	16.62±0.75	15.86±0.74	186.10±8.79	53.63±7.46
02	CW2	04	21.20±0.93	12.50±0.84	236.76±11.23	57.31±9.08
03	CW3	04	42.72±1.37	32.46±0.93	218.16±9.24	105.94±10.92
04	CW4	04	36.55±1.30	31.48±1.07	285.68±11.95	103.56±11.54
05	CW5	04	47.61±1.39	23.18±0.72	182.33±7.80	94.80±9.77
06	CW6	04	37.31±1.33	32.17±0.93	ND	83.31±5.38
Average worldwide values			32	45	420	370

ND=Not Detected

The uncertainty of the average values is quoted at 1 sigma standard deviation.

Table 2: Comparison of the mean values of ^{226}Ra, ^{232}Th, ^{40}K and radium equivalent (Ra_{eq}) activities in the studied cement samples collected in Cameroon with data from other countries

Country	No	Specific activity (Bqkg^{-1})			Ra_{eq} (Bqkg^{-1})	References
		^{226}Ra	^{232}Th	^{40}K		
Algeria	12	41	27	422	112	[19]
Cameroon	24	16.62÷47.61	12.50÷32.46	ND÷285.68	53.63÷105.94	present work
Egypt	85	78.00	33.30	37.00	151.00	[20]
Brazil	01	61.70	58.50	564.00	188.80	[21]
Ghana	50	35.94	25.44	251.00	90.12	[2]
Italy	07	38.00	22.00	218.00	92.00	[22]
Nigeria	22	43.80	21.50	71.7	80.10	[23]
Netherlands	06	27.00	19.00	230.00	71.90	[24]

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Turkey	06	50	40	324	62-312	[10]

Table 3: Values of Radiation Hazard Parameters in the Investigated Cement Type

Cement	D(nGy/h)	AEDE (mSv/y)	H_{ex}	H_{in}	Iα	Iγ
CW1	47.624±2.207	0.234±0.011	0.145	0.190	0.083	0.20 ± 0.03
CW2	52.195±2.678	0.256±0.013	0.155	0.212	0.106	0.23 ± 0.04
CW3	92.461±3.023	0.454±0.014	0.286	0.401	0.214	0.39 ± 0.04
CW4	91.108±3.329	0.447±0.016	0.28	0.379	0.183	0.38 ± 0.04
CW5	83.886±2.695	0.412±0.013	0.256	0.385	0.238	0.38 ± 0.04
CW6	69.712±2.247	0.342±0.011	0.225	0.326	0.187	0.29 ± 0.02
worldwide	60	0.410	<1	<1	≤0.5	=1

The specific activities of the natural radionuclides of ^{226}Ra, ^{232}Th and ^{40}K were determined by using gamma-ray spectroscopy in different cement types used in Cameroon. The mean activity concentrations together with their associated uncertainties reported at 1 sigma standard deviation are shown in Table 1. The specific activity concentrations in the investigated cement types used in Cameroon were found to varies from16.62 ± 0.75 to 47.61 ± 1.39 Bqkg^{-1} for ^{226}Ra; 12.50 ± 0.84 to 32.46 ± 0.93 Bqkg^{-1} for ^{232}Th and "ND" to 285.68 ± 11.95 Bqkg^{-1} for ^{40}K, respectively. While the lowest activity concentration of ^{226}Ra was observed in CW1 cement sample the highest value was in CW5 cement sample. For ^{232}Th the lowest activity value was observed in CW2 cement sample and the highest value was obtained in Portland cement. In the case of ^{40}K, CW4 cement sample showed the highest activity value and was not detected in CW6 cement type. As can be seen from Table 1, the specific activity values of ^{226}Ra, ^{232}Th and ^{40}K determined in cement varied from one sample to another. These variations in activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the investigated cements types used in Cameroon may depend on the uranium, thorium and potassium content under the earth crust from where the raw materials for particular brand of cement were obtained.

Recommended by UNSCEAR the world's mean values of ^{226}Ra, ^{232}Th and ^{40}K specific activity are 32, 45 and 420 Bqkg^{-1}, respectively [6]. The activity concentrations of ^{232}Th and ^{40}K observed in this study were significantly lower compared to the world average values, whereas the mean concentration of ^{226}Ra values observed in CW3, CW4, CW5 and CW6 cement type were relatively higher than the recommended values.

The distribution of ^{226}Ra, ^{232}Th and ^{40}K in the studies cement types was not uniform. Due to this non-uniformity of natural radionuclides in this study, the radium equivalent activity (Ra_{eq}) was calculated to compare the specific activity of the studied cement samples and the results are summarized in Table 1. The calculated radium equivalent ranged between 53.63 ± 7.46 to 105.94 ± 10.92 Bqkg^{-1}. It was clearly shown that obtained values of Ra_{eq} in the studied cement types are lower than the recommended maximum value of 370 Bqkg^{-1}, which corresponds to an annual effective dose of 1mSv. This shows that investigated cement types are within the recommended safety limit when used as building construction materials.

For comparison purposes the activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K as well as the calculated radium equivalent have been compared with the data reported by other countries. As can be seen from Table 2 some of the observed values of ^{226}Ra, ^{232}Th, ^{40}K and the evaluated radium equivalent were lower than the reported data of other countries while some were higher but varied in within the reported mean values of other countries.

The calculated internal (H_{in}) and external (H_{ex}) hazard indices for the investigated cement types as shown in Table 3 varied from 0.190 to 0.401 and from 0.145 to 0.286, respectively. These calculated internal (H_{in}) and external (H_{ex}) hazard indices in all cement samples are indeed less than the unity.

The gamma activity index (I_γ) and alpha index (I_α) used to assess safety requirement for building materials were evaluated and presented in Table 3. The obtained values for both of them ranged from 0.20±0.03 to 0.39±0.04 and from 0.083 to 0.238 respectively. The obtained values of gamma activity indexes in all cement samples were within the exemption dose criterion (0.3mSv/y) and corresponds to an activity concentration index of $I_\gamma \leq 0.5$ proposed by [7] for materials used in bulk construction.

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The results obtained for alpha index in all studied cement samples are lower than the unity. This indicates that the radon exhalation from cement would cause indoor concentration less than 200 Bq*cm^{-3}.

The estimated indoor gamma dose rate (D) values for cement samples are shown in **Table 3**. The values obtained in all the studied samples ranged from 47.624±2.207 to 92.461±3.023nGyh^{-1}. These estimated values of indoor gamma dose rate in the studied samples are comparably higher than the world average (populated-weighted) indoor absorbed dose rate of 60 nGyh^{-1} recommended by [6] with the exception of the indoor absorbed dose rate values obtained in CW1 and CW2 cement samples which are relatively lower than the world mean value.

Furthermore, Table 2 presents also the evaluated annual effective dose equivalent (AEDE) from indoor terrestrial gamma radiation for the studied cement samples. The values obtained varied from 0.234±0.011to 0.454±0.014mSvy^{-1}. The mean value found to be less that the average external annual effective dose of 0.410mSv from natural indoor radiation sources to terrestrial [6]. Comparing the estimated values in the investigated samples with the worldwide average value, it can be observed that the estimated values in Portland, CW4 and CW5 cement sample are slightly higher while those in CW1, CW2 and CW6 are below the recommended values. Comparing the estimated annual effective dose with the recommended value of 1mSv/y by ICRP [25], all the values are belong that value of ICRP.

5. CONCLUSIONS

The specific activities of ^{226}Ra, ^{232}Th and ^{40}K in the studied cement types used in Cameroon were measured and the observed mean activity values of ^{232}Th and ^{40}K are comparably lower than the typical world mean activity of 42 and 420 Bqkg^{-1}. However the mean specific activity values of ^{226}Ra were lower than the world average value of 32 Bqkg^{-1} with the exception of the observed values in CW3, CW4, CW5 and CW6 cement type which were relatively higher than the recommended average value. The observed mean activity values of ^{226}Ra, ^{232}Th and ^{40}K were also compared with the reported values by other countries and found to be within the same range.

The radium equivalent activities obtained in this study are comparably lower than the worldwide average values of 370Bq/kg. Therefore the use of the investigated cements as building materials in Cameroon is considered to be safe for the population. Absorbed dose and the indoor annual effective dose for individual living in house made with the investigated cement are compared lower than the recommended average values with the exception of some values obtained in some investigated cement which are comparably higher than the worldwide average value given by UNSCEAR [6]. The external and the internal hazard index obtained were also found to be less than unity. The activities concentrations index obtained in this study are comparably lower than the recommended values. From the obtained results, it can be concluded that the investigated cement can safely been used for building construction even though some radiological health hazards parameters are slightly higher than the worldwide values.

Since only few cement type used as building materials have been investigated in this current study, it is suggested that similar investigation should be carried out in other cement type and other building materials used in different parts of the country to have more representative values for the level of naturally occurring and technically radioactive materials, which could be useful to draw a national global picture of radioactivity on building materials in Cameroon.

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