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Natural Radioactivity and Hazard Assessment of Imported Ceramic Tiles in Nigeria

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ABSTRACT: The natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K) content of ceramic wall and floor tiles commonly used in Nigeria have been determined by a gamma ray spectroscopy system using a high purity germanium detector. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K ranged from 52 ± 2 to 105 ± 3, 56 ± 1 to 115 ± 2 and 185 ± 9 to 893 ± 17 Bq kg^{-1} with mean values of 72 ± 14, 84 ± 18 and 629 ± 198 Bq kg^{-1}, respectively for the wall tiles. For the floor tiles, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K varied from 41 ± 2 to 131 ± 4, 59 ± 1 to 127 ± 2 and 351 ± 11 to 979 ± 16 Bq kg^{-1} with mean values of 74 ± 31, 82 ± 24 and 618 ± 231 Bq kg^{-1}, respectively. The errors quoted in the means are standard deviations. The values of radium equivalent activity (Ra_{eq}) calculated varied from 194 to 328 Bq kg^{-1} for the wall tiles, and 176 to 306 Bq kg^{-1} for the floor tiles. The radium equivalent activities of all the samples examined were lower than the recommended limit of 370 Bq kg^{-1} for building materials. The mean values of the external hazard index (H_{ex}), the activity index (I_γ) and the alpha index (I_α) of the wall and floor tiles are less than unity, which is a commonly accepted limit.

Key Words: Ceramic tiles, natural radionuclides, activity concentration, hazard assessment.

INTRODUCTION

Our world is radioactive and has been since it was created. Many radionuclides occur naturally in terrestrial soil and rocks and in building materials derived from them. Upon decay, these radionuclides produce an external radiation field to which all human beings are exposed. In terms of dose, the principal primordial (half-life comparable to the age of the earth) radionuclides are ^{40}K, ^{232}Th and ^{238}U. Both ^{232}Th and ^{238}U head series of radionuclides that produce significant human exposure (UNSCEAR 2000). Radioactive decay of these isotopes produces alpha, beta, and gamma radiations. The concentrations of the radioisotopes in the earth's crust and in building materials determine the dose of natural radiation, both outside and inside of building.

The concentration of ^{226}Ra (a daughter isotope of ^{238}U) is crucial because its daughter isotope ^{222}Rn easily penetrates into places of permanent residence for people. The isotope ^{40}K (half life time 1.28 x 10^9 years) is the only radioactive isotope among the three constituents of natural potassium (0.012%). Natural uranium consists of 99.28% of the isotope ^{238}U (half-life time 4.51 x 10^9 years). The only one natural isotope of thorium, ^{232}Th, undergoes alpha decay with a half-life time of 1.41 x 10^{10} years and initiates the thorium series isotopes (Charewicz *et al* 2000). The world-wide average concentrations of ^{238}U, ^{232}Th and ^{40}K in soil are about 33, 45 and 420 Bq kg^{-1} (UNSCEAR 2000).

All building materials contain small amounts of radioactive substances. The most important naturally occurring radionuclides present in building materials including soil are ^{226}Ra, ^{232}Th and ^{40}K (Khan *et al* 1998). The radionuclides are not uniformly distributed; they can vary considerably according to the geological

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origin of the raw materials. Hence the knowledge of the natural radioactivity in building materials is important for the determination of population exposure to radiations (Xinwei 2004).

The level of activity in ceramic tiles, like other construction materials give rise to external and internal indoor exposure. The external radiation exposure is caused by gamma radiation originating from ^{226}Ra, and ^{232}Th and their decay products and ^{40}K, while internal radiation exposure, mainly affecting the respiratory tract, is due to the short-lived decay products of radon which are exhaled from building materials into room air (Stoulos *et al* 2003, Khan and Khan 2001).

Ceramic tiles are one of the commonly used decorative building materials. In recent time, the use of the ceramic tiles in indoor decoration has increased in Nigeria. In most dwellings and work place, ceramic floor tiles are used instead of concrete flooring and flexy tiles. Most of the tiles used in Nigeria are the imported type. The raw materials of ceramic tile include kaoline, plasticity clay, feldspar, quartz, gellibackite, steatite etc. These are crushed to powder, press moulded, calcined at high temperature and finally turned into ceramic tile.

The aim of the present study is to determine the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in imported ceramic wall and floor tiles collected from suppliers and assess the radiological hazards associated with these tiles. The result obtained is compared with results of ceramic tiles in literature.

MATERIALS AND METHODS

Ceramic wall and floor tiles were collected from suppliers in Nigeria. The samples were pulverized and dried. A mass of 100 g of each sample was placed in a cylindrical container of height 28 mm and diameter 70 mm. The containers were sealed and left for more than 4 weeks in order to allow for Ra and its short-lived progeny to reach radioactive equilibrium.

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the samples were measured using coaxial HPGe (Hyperpure Ge) detector of model GC3018 – 7500SL (serial number 693085). The relative efficiency of the detector is 30% and has a resolution of 1.8 keV at 1.33 MeV (0.875 keV at 122 keV). The detector is maintained in a vertical position in a lead cylindrical shield of 10 cm thickness. The detector is coupled to a computer through a preamplifier base (model 2002CSL).

The energy calibration of the detector was performed using point sources of known energy from Amersham Buchier GmbH & Co. KG. Calibration was done for gamma energy range of 20.0 keV to 2.87 MeV. The efficiency calibration was done using reference sources R6/120/85 (QCY-48 solution) from Amersham, 97-273 (Cr-51 solution) and 12024 (Cs-137 solution). These are aqueous solution which contains well known concentrations (Bq kg^{-1}) of several isotopes emitting gamma- rays in a wide range of energies between 60 keV and 2 MeV. Dry quartz sand was used as sample matrix in order to simulate soil sample. This is free of natural radioactivity. The quartz sand was carefully mixed with the nuclide solutions and dried at low temperature until its initial mass was attained.

The activity concentrations of ^{226}Ra and ^{232}Th were determined from the photopeak of ^{214}Pb (295.21 keV) and ^{212}Pb (238.63 keV), respectively while the activity concentration of ^{40}K was determined from the 1460.8 keV photopeak following the decay of ^{40}K. Each sample was counted for 43200 s (12 h).

RESULTS AND DISCUSSION

Activity concentration

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the ceramic wall and floor tiles are presented in Table 1. The activity concentration levels of the wall tiles varied between 52 ± 2 to 105 ± 3, 56 ± 1 to 115 ± 2 and 185 ± 9 to 893 ± 17 Bq kg^{-1} with mean values of 72 ± 14, 84 ± 18 and 629 ± 198 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The floor tiles had activity concentrations from 41 ± 2 to 131 ± 4, 59 ± 1 to 127 ± 2 and 351 ± 11 to 979 ± 16 Bq kg^{-1} with corresponding mean values of 74 ± 31, 82 ± 24 and 618 ± 231 Bq kg^{-1}, respectively for ^{226}Ra, ^{232}Th and ^{40}K. The errors quoted in the mean values are the standard deviations which depict the extent of the spatial spread in the concentration of the radionuclides in the samples. The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in all the samples examined exceeded the corresponding typical world averages of 50, 50, 500 Bq kg^{-1} for building materials (UNSCEAR 1993). The activity concentrations of ^{40}K were higher than that of ^{226}Ra, and ^{232}Th in all the samples. This could be due to it abundance in the earth crust from which majority of the raw materials is extracted.

Assessment of the radiological hazards

The knowledge of the activity concentrations of materials used in building is important in the assessment of the possible radiological hazards to human health. The radiological hazard due to the use of the ceramic tiles has been calculated using some indices

Radium equivalent activity

A common index called the radium equivalent activity (Ra_{eq}) was used to compare the radiological effects of the ceramic tiles (Beretka and Mathew 1985). The index compares the activities of materials containing different amounts of radium, thorium and potassium. The index is based on the estimation 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 rates, and therefore Ra_{eq} can be written as (Beretka and Mathew 1985):

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

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. The maximum value of Ra_{eq} must be less than 370 Bq kg^{-1} in order to keep the external dose to be less than 1.5 mGy y^{-1} (Beretka and Mathew 1985, NEA-OECD 1979).

Table 1:
Activity concentrations (Bq kg^{-1}) of samples of ceramic wall and floor tiles

S/N	**Country of Origin**	**^{266}Ra**	**^{232}Th**	**^{40}K**
Wall tiles				
1	China	70 ± 3	92 ± 1	584 ± 13
2	China	70 ± 3	73 ± 1	643 ± 14
3	China	59 ± 3	115 ± 2	493 ± 12
4	China	70 ± 3	96 ± 1	502 ± 12
5	Spain	72 ± 3	65 ± 1	763 ± 14
6	Spain	76 ± 3	69 ± 1	850 ± 15
7	China	61 ± 2	78 ± 1	754 ± 12
8	Spain	60 ± 3	56 ± 1	700 ± 16
9	China	52 ± 2	76 ± 1	521 ± 12
10	Spain	74 ± 3	74 ± 1	893 ± 17
11	China	92 ± 3	84 ± 1	784 ± 15
12	GDR	76 ± 3	103 ± 1	375 ± 11
13	China	105 ± 3	115 ± 2	761 ± 14
14	China	73 ± 3	82 ± 1	185 ± 9
Mean ± Std. Div.		**72 ± 14**	**84 ± 18**	**629 ± 198**
Floor tiles				
15	Spain	73 ± 3	71 ± 1	797 ± 14
16	China	131 ± 4	66 ± 2	764 ± 15
17	China	98 ± 3	127 ± 2	351 ± 11
18	China	68 ± 3	100 ± 2	406 ± 13
19	Brazil	58 ± 3	84 ± 1	491 ± 12
20	China	49 ± 2	59 ± 1	540 ± 12
21	China	41 ± 2	67 ± 1	979 ± 16
Mean ± Std. Div.		**74 ± 31**	**82 ± 24**	**618 ± 231**

The calculated values of Ra_{eq} are presented in Table 2. All the samples had Ra_{eq} values below this limit. The values ranged between 194 and 328 Bq kg^{-1} for the wall tiles and 176 and 306 Bq kg^{-1} for the floor tiles. The mean values are 241 and 239 Bq kg^{-1}, respectively. Based on their Ra_{eq} values, all the ceramic tiles examined produce radiation levels considered safe for dwelling and workplace. Table 3 compares the reported values of radium equivalent activities for ceramic tiles in other countries of the world with those determined in this study. As shown in the table, Ra_{eq} of ceramic tiles vary from country to country and that obtained in this study compares wall with results from other countries.

Table 2:
Radium equivalent activity, Ra_{eq} (Bq kg^{-1}), external hazard index, H_{ex}, activity index, I_γ and alpha index, I_α of samples of ceramic wall and floor tiles

S/N	**Country of origin**	**Ra_{eq}**	**H_{ex}**	**I_γ**	**I_α**
Wall tiles					
1	China	245	0.66	0.89	0.35
2	China	224	0.60	0.81	0.35
3	China	262	0.71	0.94	0.29
4	China	246	0.66	0.88	0.35
5	Spain	223	0.60	0.82	0.36
6	Spain	241	0.65	0.88	0.38
7	China	231	0.62	0.84	0.31
8	Spain	194	0.52	0.71	0.30
9	China	202	0.54	0.73	0.26
10	Spain	249	0.67	0.91	0.37
11	China	273	0.74	0.99	0.46
12	GDR	252	0.68	0.89	0.38
13	China	328	0.89	1.18	0.52
14	China	205	0.55	0.72	0.36
Mean ± Std. Div.		**241 ± 34**	**0.65 ± 0.09**	**0.87 ± 0.12**	**0.36 ±0.07**
Floor tiles					
15	Spain	235	0.64	0.86	0.36
16	China	284	0.77	1.02	0.66
17	China	306	0.83	1.08	0.49
18	China	243	0.66	0.86	0.34
19	Brazil	215	0.58	0.78	0.29
20	China	176	0.47	0.64	0.25
21	China	212	0.57	0.80	0.21
Mean ± Std. Div.		**239 ±44**	**0.65 ± 0.12**	**0.86 ± 0.15**	**0.37 ±0.16**

External Hazard Index

The gamma-rays emanating from building materials can pose a health hazard. This is usually given in terms of the external hazard index, H_{ex} (UNSCEAR 1982, Beretka and Mathew 1985, Xinwei 2004);

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \qquad (2)$$

For safe use of a material in the construction of dwellings, H_{ex} should be less than unity. The values obtained for the external hazard index were found to lie in the range of 0.52 and 0.89 with a mean value of 0.65 for the wall tiles. For the floor tiles, the values ranged between 0.47 and 0.83 with a mean value of 0.65.

Activity index

The gamma-rays emanating from building materials can pose a health hazard. The restriction on building materials for gamma radiation is based on a dose range of 0.3 to 1 mSv y^{-1} (EC 1999, STUK 2003). In order to examine whether a building material meets these limit of dose criteria, the activity index or gamma index I_γ of the samples is found from the following Equation (EC 1999, STUK 2003, Kovler 2000):

$$I_\gamma = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000}, \qquad (3)$$

where C_{Ra}, C_{Th} and C_{K} are the activity concentrations of ^{226}Ra, ^{323}Th and ^{40}K, respectively in Bq kg^{-1}. The numerical quantities in the denominator of Equation 3 are also in units of Bq kg^{-1}, so that I_γ is a dimensionless quantity. If the activity index I_γ is 1 or less than 1, the material can be used as building material, as far as radioactivity is concerned, without restriction. In case of superficial or other materials with restricted use in house building (for example tiles), the activity index I_γ may be 6 or less than 6 (STUK 2003). The results of the activity index are presented in Table 2. The values obtained lie between the range of 0.71 and 1.18 with a mean value of 0.87 for the wall tiles and 0.64 and 1.08 with a mean value of 0.86 for the floor tiles. One of the samples of wall tiles and two of the floor tiles had values of I_γ greater than unity. Based on their I_γ values, all of the ceramic tiles examined produced radiation levels considered safe for dwelling and workplace applications (STUK 2003). Table 3 compares the values of activity index for ceramic tiles in other countries of the world with those determined in this study.

Alpha index

This is another important criterion used to assess alpha radiation due to radon inhalation originating from building materials (Xinwei 2004, Tufail *et al* 2007, Righi and Bruzzi 2006):

$$I_\alpha = \frac{C_{Ra}}{200} \qquad (4)$$

The recommended exemption level and recommended upper limit of ^{226}Ra activity concentrations are 100 and 200 Bq kg^{-1}, respectively, in building materials as suggested in many countries of the world (RPA 2000). The criterion only considers internal exposure and corresponds to a maximal ^{226}Ra activity concentration of 200 Bq kg^{-1} for building materials. When the ^{226}Ra activity concentration of a building material exceeds the value of 200 Bq kg^{-1} it is possible that the radon exhalation from this material could cause indoor radon concentration > 200 Bq m^{-3}. On the contrary, when the ^{226}Ra activity concentration is < 100 Bq kg^{-1} it is unlikely that the radon exhalation from the building materials would cause indoor radon concentration > 200 Bq m^{-3} (Xinwei 2006). The recommended upper limit concentration of ^{226}Ra is 200 Bq kg^{-1}, for which I_α is equal to one (Tufail *et al* 2007).

The alpha index has been calculated using the activity concentration of ^{226}Ra in the samples. It can be observed from Table 1 that the activity concentration values of ^{226}Ra in all the samples are less than the recommended exemption level of 100 Bq kg^{-1}, except one of the samples of the wall and floor tiles, respectively. However the two values are less than the recommended upper limit of 200 Bq kg^{-1}. The results obtained for the alpha index, which ranged from 0.26 to 0.52 with a mean value of 0.36 for wall tiles and from 0.21 to 0.66 with a mean value of 0.37 for the floor tiles are shown in the last column of Table 2. These results indicate that the radon exhalation from the ceramic tile samples would cause indoor concentration less than 200 Bq m^{-3}.

Conclusion

The activity concentrations of natural radionuclides in imported ceramic wall and floor tiles used in Nigeria had been determined. The activity index and the radium equivalent activity of the samples were calculated using the result of the activity concentrations. The average value of Ra_{eq} and I_γ of the samples were found to be less than the recommended limits.

Table 3:
Comparison of radium equivalent activity, Ra_{eq} (Bq kg^{-1}), external hazard index, H_{ex}, activity index, I_γ and alpha index, I_α.

Sample	Country	Ra_{eq}	H_{ex}	I_γ	I_α	Reference
Tiles	Austria	169	0.46	0.62	0.24	Soranti and Steger 1984
Tiles	South Korea	186	0.5	0.68	0.29	Lee *et al* 2001
Tiles	Brazil	223	0.60	0.82	0.30	Malanca *et al* 1993
Ceramic tile (wall)	Nigeria	241	0.65	0.87	0.36	Present study
Ceramic tile (floor)	Nigeria	239	0.65	0.86	0.37	Present study

The I_α calculated was less than unity for all the samples. The results obtained indicate that the use of these materials does not pose any significant radiological hazard and they are safe for use in dwellings and workplaces.

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