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Experimental Evaluation of Radiological Hazards in Ceramic Tiles Used in the Jos-South, Area of Plateau State, Nigeria

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ABSTRACT

Radiological hazards resulting from 5 different brands of ceramic tiles commonly used in homes and offices in Jos-south area of plateau state were assessed in this study by carrying out radioactivity measurement of natural radionuclides (40 K, 226 Ra and 232 Th) in them using a NaI(Tl) gamma ray spectrometer. The ceramic tiles had mean activity concentrations of 44.5, 59.6 and 417.4 Bq kg⁻¹ for 226 Ra, 232 Th and 40 K respectively. The average radium equivalent is within the recommended limit of 370 Bq kg⁻¹ for building materials. The radiological hazard indices such as the gamma absorbed dose rate, annual effective dose, external radiation index (H_{ex}), internal radiation hazard index (H_{in}), Alpha index (Ia), Gamma index (I γ), excess lifetime cancer risk, and annual gonadal dose equivalent were evaluated. The results obtained show that the radiological parameters do not exceed the recommended safety limits and hence pose no significant radiological hazard when used as building material.

Key Words: Ceramic tiles, Gamma ray spectrometry, Radiological hazards, Natural Radionuclides.

1. INTRODUCTION

The ceramic industry encompasses a varied group of producers and products. One of the main subsectors is the production of ceramic tiles. Due to the increasing number of houses being built as human population increases, demand for tiles is increasing by the day. Tiles have been found essential in housing, hospitals, industries, hotels, restaurants, cinema halls, shopping malls as well as other public places.

Ceramic is composed of two main components: the tile body and the tile glaze. The tile body is derived from clay minerals mined from the Earth's crust and is usually produced with variable purity. The second component of the tile is the shiny tile glaze, which typically consists of one or more layers with a total thickness between 75 and 500 microns, covering the upper side of the ceramic tiles. This substance is usually a glass material glaze composed of inorganic raw materials, with silica (SiO_2) as the main component [2].

Ceramic tiles are generally preferred worldwide due to their ornamental properties along with their relatively cheap cost. They are made of a mixture of earthly materials that has been pressed into shape and fired at high temperature. Body of ceramic tiles is a mixture of different raw materials including clays, quartz materials and feldspar, and may be glazed or left unglazed. Zircon is a common opacifying constituent of glazes applied to ceramic tiles and is also used as an opacifier in porcelain tiles by incorporating it directly into the mixture used for forming the body of the tile. Due to the presence of zircon in the glaze, ceramic tiles can show natural radioactivity concentration significantly higher than the average values for building materials [7,16, 20].

Indoor and outdoor gamma dose rate associated with commercial building materials in Jos, Plateau Nigeria including shops where tiles are sold was assessed by [11], using a hand held survey meter. These form of detectors do not indicate the radionuclides that are present but only the overall radioactivity levels. It is important to carry out gamma ray spectrometric analysis for the different

brands of ceramic tiles that are commercially available for use as walling and flooring materials to assess their associated radiological hazards. The activity concentration of natural radionuclides (226 Ra, 232 Th and 40 K) in different brands of tile samples used for building purposes in Nigeria was evaluated by [10]. However, so many new brands of tiles are being produced/imported to the country which requires to be measured in order to access the radioactivity contents.

This paper is designed to add and enhance the existing information on the natural radioactivity of brands of ceramic tiles presently used in Nigeria with particular interest in Jos south local government area of plateau state. The study employs the use of gamma-ray spectrometry method to measure the activity concentrations of natural radionuclides in six brands of ceramic tiles that are used as flooring/walling materials in Jos-South area of Plateau State, Nigeria in order to assess the radiological hazards associated with their use as building materials.

2. MATERIALS AND METHODS

2.1 Sample Preparation and Measurement

Jos south is the industrial center of Plateau State due to the presence of industries like Grand Cereals and Oil Mills, Zuma Steel West Africa, Nasco group etc. Most building materials used in this region are sourced from the popular building materials market located in Bukuru, which is the biggest building materials distribution outlet in plateau state. Customers also come from different towns in the state and from neighboring states to buy materials for construction purposes.

Table 1 shows the brands of ceramic tile samples measured. The wall/floor tiles used for this work were purchased from the Building materials market Bukuru. All samples were pulverized and dried The dried samples, each a mass of 200 g, were packed into cylindrical plastic containers of base diameter (7 cm) that could sit on the 7.6 cm by 7.6 cm NaI (Tl) detector. The plastic containers including an empty one were sealed tightly with caps and wrapped with thick Sellotape around their screw necks. A Canberra lead-shielded 76 mm×76 mm NaI (Tl) detector crystal (Model No. 802 series, Canberra Inc.) coupled to a Canberra Series 10 plus multichannel analyser (MCA) (Model No. 1104) through a photomultiplier tube/preamplifier/amplifier was used for the radioactivity measurements. The detector has a resolution of 8 % full-width at half-maximum at ¹³⁷Cs energy of 0.662 MeV. This was good enough to distinguish the gamma ray energies of interest in the present study. The equipment was calibrated against reference source from Rocketdyne Laboratories, CA, USA. The following gamma transitions were used: ⁴⁰K, 1.461 MeV; ²²⁶Ra, 1.760 MeV (²¹⁴Bi) and ²³²Th, 2.614 MeV (²⁰⁸Tl). Each of the sample containers was placed on top of the detector and counted for10 h. To account for background radiation, the empty plastic container was counted for 10 h in the same counting geometry with those of the samples. The areas under the peaks (background) were subtracted from those of the samples (gross count) to obtain the net count for each sample.

Names of tiles used in Jos South	Country of origin
Lion King super vitrified floor tiles (BN) ceramics	Nigeria
Polygold glazed wall tiles (China)	China
VIAENY wall tiles	China
PEEYDON ceramics floor tiles	Spain
Time ceramics	Nigeria
FORMIGRES Ceramic floor tiles	Brazil

Table

1: The different tiles and their country of origins

3. RESULT AND DISCUSSIONS

3.1 Determination of radioactivity concentration

The calculations of the activity concentration (Ac) values for radionuclides from the 226 Ra and 232 Th series and 40 K present in ceramic tile samples can be determined as:

$$A_{c} = \frac{C_{net}}{\gamma \times \varepsilon \times m \times t} - \dots - (1)$$

Where C_{net} represent peak net counts, γ represent the emission probability of specific energy peak, ε is the absolute efficiency of the full energy peak of the detector, *m* is the mass sample in (Kg) and *t* is the time of count. Table 2 presents the radioactivity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K for the tiles samples measured and their average value respectively. The observed activities concentration of the radionuclides content in the tiles ranged from 313.1 to 567.8 Bq/kg for ⁴⁰K, 34.9 to 98.4 Bq/kg for ²³²Th and 34.2 to 57.9 Bq/kg for ²²⁶Ra respectively. The Lion king super vitrified tiles was noted to have the highest value for ⁴⁰K with a value of 567.8 Bq/kg, while PEEYDON ceramics had 98.4 Bq/kg which was the highest for ²³²Th. The highest value for ²²⁶Ra was recorded for VIAENY with 57.9 Bq/kg.

The mean values observed for all the tile samples for 40 K, 232 Th and 226 Ra were 417.4 Bq/kg, 59.6 Bq/kg and 44.5 respectively. Comparing these with the world average values of; 40 K: 400 Bq/kg, 232 Th: 35 Bq/kg and 226 Ra: 30 Bq/kg it is observed that the mean values of the activity concentrations measured in this study are slightly higher than the world average [22]. It is also observed that the distribution of the activity concentration in the samples is not uniform; this is likely due to their region of origin and processing composition. The higher activity concentrations found in the studied ceramic tiles may be due to the presence of zircon in the glaze [7].

3.2 Radiological Assessment

3.2.1 Radium Equivalent Concentration

The Gamma-ray radiation hazards resulting from ²²⁶Ra, ²³²Th and ⁴⁰K can be estimated using different indices such as Radium equivalent (Ra_{eq}). It is the most widely used radiation hazard index (Beretka & Mathew, 1985; Krieger, 1981) [1, 12]. Ra_{eq} is a weighted sum of activities of the above three radionuclides based on the estimation that 370 Bq/kg of ²²⁶Ra, 259 Bq/kg of ²³²Th and 4810 Bq/kg of ⁴⁰K produce the same g-ray dose rates [17].

$$Ra_{ea} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad \dots \quad (2)$$

Table 2, column 3, summarizes the Ra_{eq} results for all the samples studied. These values ranged from 131.9 Bq kg⁻¹ (VIAENY) to 269.9 Bq kg⁻¹ (Polygold) which are are less than the maximum admissible value of 370 Bq kg⁻¹ [21], which is equivalent to an external dose of 1.5 mSv yr⁻¹ [14]. Thus, all the materials selected for this study would not pose a significant radiological hazard when used in building constructions.

Tile name	Activity concentration in (Bq/kg)			Ra _{eq}
	⁴⁰ K	²³² Th	²³⁶ Ra	
PEEYDON	564.0 ± 12	98.4 ± 3	46.7 ± 2	230.8
ceramics				
Time ceramics	421.6 ± 11	$40.9~\pm~1$	34.2 ± 1	125.2
Lion King super	567.8 ± 12	60.6 ± 2	54.6 ± 2	185.0
vitrified				
VIAENY	313.1 ± 10	34.9 ± 1	57.9 ± 2	131.9
Polygold glazed	209.5 ± 9	56.7 ± 2	30.4 ±1	127.6
tiles				
FORMIGRES	428.5 ± 11	65.8 ± 2	43.2 ± 2	170.3
ceramics				
Mean Value	417.4	59.6	44.5	161.8

Table 2. Activity concentration and Radium equivalent activity for the ceramic tile samples

3.2.2 Estimation of absorbed and annual effective dose

The absorbed dose rate in air in a room has been calculated according to [5]. The specific dose rates (in units $nGyh^{-1}$ per Bq kg⁻¹) for ²²⁶Ra, ²³²Th and ⁴⁰K are given for different materials. Dose rate indoors are calculated according to [5, 6, 23], for materials under investigation, when used as tile on all walls.

For conversion of the activity concentration to absorbed dose rate in air at 1 m above the ground surface for uniform distribution of naturally occurring radionuclides, dose coefficients of 0.12 nGy h^{-1} per Bq kg⁻¹ for ²²⁶Ra, 0.14 nGy h^{-1} per Bq kg⁻¹ for ²³²Th, and 0.0096 nGy h^{-1} per Bq kg⁻¹ for ⁴⁰K were used.

$$D(nGy h^{-1}) = 0.0096C_k + 0.12C_u + 0.14C_{Th} \qquad -----(3)$$

Where A_{Ra} is the specific activity concentration of ²²⁶Ra, C_{Th} is the specific activity concentration of ²³²Th, C_K specific activity concentration of ⁴⁰K in Bq kg⁻¹, and D is the dose rate in nGy h⁻¹.

The following relation was used to calculate the annual effective dose, $H_E (Sv y^{-1})$:

$$H_E = D (nGy h^{-1}) \times CF \times 8760 h y^{-1} \times OF$$
 ------(4)

where CF and OF are the conversion factor and outdoor occupancy factor, respectively.

Annual estimated average effective dose equivalent received for indoor exposure to whole body is calculated using a conversion factor of 0.7 Sv/Gy, which is used to convert the absorbed dose rate to annual effective dose with an indoor occupancy factor of 0.8 [22].

Table 3, column 2, shows values of the absorbed dose rates for the different brands of tiles. Values ranged from 14.8 nGyh⁻¹ for VIAENY Tiles to 28.7 for nGyh⁻¹ lion king tiles with a mean value of 22.2 nGyh⁻¹, which is lower than the world average value of 86 nGyh⁻¹. Consequently, mean value of the annual effective dose estimated for the tiles is 0.11 mSv y⁻¹ is less than the world indoor average value of 0.49 mSv y⁻¹ [22] and 1.0 mSv y⁻¹ limit set by the International Committee on Radiation Protection (ICRP).



Figure 1. The contributions of ⁴⁰K, ²³²Th and ²²⁶Ra to the absorbed dose rates.

3.2.3 Internal and External Hazard Indices

Radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its progeny products are quantified by the internal hazard index (H_{in}), which is determined by equation 5. Also to limit the external gamma radiation dose from ceramic materials to 1mSv y⁻¹, the external hazard index (Hex) is determined by equation 6, [1,16].

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \qquad -----(6)$$

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where A_{Ra} , A_{Th} and A_{K} are the activity concentrations (Bq kg⁻¹) of ²²⁶Ra, ²³²Th, and ⁴⁰K, respectively.

The maximum values of H_{ex} and H_{in} should be less than unity meaning the calculated dose is less than 1 mSv per year [8]. As shown in table 3, columns 4 and 5, mean values of the external and internal hazard indices are less than 1.

3.2.4 Gamma Index

Gamma index is used to evaluate the γ -radiation hazard related to the natural radionuclide in the particular samples under investigation. The index is based on radioactivity controls of building materials on a defined dose criterion for controls and an exemption level. The European technical guidance, RP112, proposes that a dose criterion be chosen on a national basis in the range of $0.3 - 1 \text{ mSv y}^{-1}$ of the excess gamma dose to that received outdoors, and to exempt from all restrictions building materials that increase the annual effective dose to a member of the public by 0.3 mSv at the most [5]. The index is given as shown in table 4, [5]. If the activity index $I\gamma$ is 1 or less than 1, the material can be used as building material, as far as radioactivity is concerned, without restriction.

Gamma activity index ≤ 6 corresponds to annual effective dose of 1 mSvy⁻¹ and gamma activity

index ≤ 2 corresponds to an annual effective dose $\leq 0.3 \text{ mSvy}^{-1}$ if the bulk materials are used in a superficial way. The values of I γ for the ceramic tiles measured in this work are shown in table 5, Peeydon had the highest value of 0.79 while Time ceramics had the lowest value with 0.39. the mean values observed for all the measured ceramic tiles is 0.45.

Considering the criterion of unity that corresponds to annual effective of 1 mSv, all values for the ceramic samples fall below the criterion which corresponds to the protection level thus the tiles can be safely used in building construction.

$$I_{\gamma} = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_{K}}{3000} - \dots$$
(7)

3.2.5 Alpha index

For indoor radiation exposure, inhalation of the short-lived decay products of 222 Rn is of great concern. This is due to their deposition along the walls of the various airways of the bronchial tree providing the main pathway for radiation exposure in the lungs [22]. The radium content of building materials constitutes the source of emanation of radon indoors. To assess the internal hazard originating from alpha activity of building materials, the alpha index (I_a) is used and is given as [4, 18]:

where A_{Ra} is the activity concentration of 226 Ra (Bq kg⁻¹).

For safe use of material in building construction, it is required that I_{α} be less than unity. When the ²²⁶Ra activity concentration of 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 exceeding 200 Bq m⁻³. On the contrary, when the ²²⁶Ra activity concentration is 100 Bq kg⁻¹, it is unlikely that the radon exhalation from the building materials could cause indoor radon concentrations exceeding 200 Bq m⁻³ [13].

The activity concentrations of ²²⁶Ra in all the samples examined are less than the recommended exemption level of 100 Bq kg⁻¹. It is noted that the upper limit of radon concentration (I α) is equal to 1, [19]. The results of the present study show that the radon concentration varies from 0.15 for Polygold to 0.29 for VIAENY with an average value of 0.22 which is less than 1. As can be observed in Table 3 column 6, all the values of I α obtained are below the recommended limit. This shows that radon inhalation from the ceramic tiles studied would not cause any excess radiation exposure when used as a building material.

Table 3: Absorbed dose rate, Annual effective dose rate, external hazard index, internal hazard index and alpha index for the ceramic tile samples

Tile name	D(nGy h ⁻¹)	AEDR (mSv y ⁻¹)	H _{ex}	\mathbf{H}_{in}	I _α
Polygold	28.7 ± 3	0.068 ± 0.001	0.35 ± 0.04	0.43 ± 0.04	0.23
glazed tile					
Time ceramics	13.9 ± 2	0.068 ± 0.001	0.34 ± 0.04	0.43 ± 0.04	0.17
Lion King	20.5 ± 3	0.100 ± 0.002	0.10 ± 0.01	0.65 ± 0.06	0.27
super vitrified					
VIAENY	14.8 ± 2	0.073 ± 0.001	0.36 ± 0.04	0.51 ± 0.05	0.29

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PEEYDON	24.8	0.122 ± 0.002	0.36 ± 0.04	0.75 ± 0.06	0.23
ceramics					
FORMIGRES	18.5 ± 2	0.091 ± 0.001	0.46 ± 0.05	0.58 ± 0.05	0.22
ceramics					
Mean Value	20.7	0.087	0.51	0.66	0.22

3.2.6 Annual Gonadal Dose

The organs of interest considered by [21], are the gonads, the active bone marrow and the bone surface cells. The gonads are particularly radiosensitive.

A single dose of only 0.3Gy (30 rads) to the testes may result in temporary sterility among men; for women, a 3Gy (300- rad) dose to the ovaries may lead to temporary sterility. Higher doses increase the period of temporary sterility [3]. The Annual Gonadal dose equivalent was calculated using the equation below (9). Therefore, the annual gonadal dose equivalent (AGDE, mSv y⁻¹) due to the activity concentrations of 226 Ra, 232 Th and 40 K was estimated using the formula given by [24]:

$$AGDE = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_{K}$$
 -----(9)

Table 5 shows the values of the annual effective dose equivalent calculated according to equation (9). Values observed ranged from 0.068 to 0.12 mSv y^{-1} with a mean value of 0.09 mSv y^{-1} . All the ceramic tiles measured have values lower than 1 mSv y^{-1} as recommended by the International Commission on Radiological Protection (ICRP) for members of the public.

3.2.7 Excess lifetime cancer risk (ELCR)

Radioactivity in building materials are known to produce carcinogenic effects due to accumulation in indoor air of radon and its progenies that emanate from the wall and floor of a room. The probability or extra risk of developing lung cancer due to exposure to gaseous radionuclides indoors incurred over the lifetime of an individual is estimated using the excess lifetime cancer risk (ELCR). Based upon calculated values of annual effective dose (AEDE), ELCR was estimated. Values of ELCR was estimated using the following formula [15]:

$$ELCR = AEDE \times DL \times RF$$
 ------(10)

where AEDE is the annual effective dose equivalent, DL is the average lifetime duration (70 years) and RF is the fatal risk factor per Sievert assumed to be 0.005 Sv⁻¹ in this study as per [9]. For stochastic effects, a value of 0.05 was used for the public. The world average value of excess lifetime cancer risk is 0.29×10^{-3} [22]. The result obtained are presented in Table 5. From the table, the average value of excess lifetime cancer risk is slightly higher than the world average value.

Table 4: Gamma-index values suggested by the European Commission (1999) taking in to account typical way and amounts in which the material is used in a building

Dose criterion	0.3mSvy ⁻¹	1mSvy ⁻¹
Materials used in bulk amount, eg. Bricks	$I\gamma \le 0.5$	$I_{\gamma} \leq 1$
Superficial and other materials with restricted	$I_{\gamma} \leq 2$	$I_{\gamma} \leq 6$
use: tiles, bricks and boards, etc.,		

Tile name	Ιγ	AGDE(mSv y ⁻¹)	ELCR x 10 ⁻³ (mSv y ⁻¹)
Polygold glazed tiles	0.57 ± 0.04	0.73	0.23
Time ceramics	0.39 ± 0.02	0.41	0.24
Lion king super vitrified	0.59 ± 0.04	0.60	0.35
VIAENY	0.51 ± 0.03	0.42	0.25

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PEEYDON Ceramics	0.79	0.73	0.43
FORMIGRE Ceramics	0.61 ± 0.04	0.54	0.32
		0.17	
MEAN VALUE	0.72	0.65	0.38

4. CONCLUSION

The natural radionuclide content, Radium equivalent activity (Ra_{eq}) and various hazard indices of some ceramic tiles commonly used in Jos-South and environs were determined using the NaI (Tl) gamma-ray spectrometer. The average Ra_{eq} values of the studied samples are below the world accepted values (370 Bqkg⁻¹). Values of external and internal hazard indices for all samples investigated are below unity. All the samples investigated are within the recommended safety limit specified for materials used for building construction and hence do not pose any radiation hazards to the dwellers.

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