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DETERMINATION OF THE RADIOLOGICAL HEALTH INDEXES OF USING NATURAL KAOLIN

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ABSTRACT

Kaolin (China clay) is used for various purposes such as in the manufacture of fine porcelain, pottery making, paper industry etc and is thus a valuable geological material. A qualitative and quantitative determination of the natural radionuclides contained in samples obtained from Delta State Nigeria was carried out using a well - calibrated highly efficient NaI(TL) detector. The consequent radiological health indexes were then calculated. The radionuclides detected belong to the ^{238}U and ^{232}Th decay and the non series ^{40}K. The mean specific activities of ^{226}Ra, ^{228}Ra and ^{40}K varied from 95.3 - 96.3, 99.7 – 99.9 and 537.2 – 543.6 Bqkg^{-1} respectively. These levels are slightly higher than corresponding values obtained by other investigators elsewhere not in Nigeria. Radium-equivalent activity was found to be 280.3 Bqkg^{-1} The external hazard index is 0.76 while the mean annual effective dose equivalent is 0.16 mSvy^{-1} . Since the hazard index is less than unity and the determined dose equivalent is below the internationally recommended limit of 1mSvy^{-1} for members of the public, it is concluded that the use of natural kaolin of Nigerian origin does not pose any radiological effect of consequence

Key words: Nigerian kaolin, wide applications, radiologically friendly.

INTRODUCTION

Kaolin (Chinese kaoling, high ridge") or china clay is a pure, soft, white clay of variable but usually low plasticity that retains its white color when fired. The term kaolin may be extended to include other porcelain clays not discolored by firing. The material was first obtained from a hill called Kaoling and was sent to Europe in the early 18th century. Its chief constituent is the mineral kaolinite, a hydrous aluminum silicate ($Al_2Si_2O_5(OH)_4$) formed by the decomposition of aluminum silicates, particularly feldspar (Truman and Thomas , 2008). The proportions of the compound components have been found to be 46% SiO_2, 40% Al_2O_3 and 14% H_2O. Its structure may be flake-like, lathlike, fibre-like or hollow tube-shaped.

In its pure form, it is used in the manufacture of fine porcelain and china; impure varieties are used in making pottery, stoneware, and bricks (Truman and Thomas, 2008). The mineral is also used in the paper industry as fillers in the bulk. Its whiteness is employed in improving paper brightness and opacity. It is applied as white wares providing strength and plasticity in ceramics products (Turhan,2009). It is used to improve the optical, mechanical and rheological properties of paints and it is also used in pharmaceuticals for the production of human and veterinary medicinals for the treatment of digestion problems. Recent investigation (Mokobia, 2010a using this geological material for luminescence) has revealed the possibility of dating.

The radioactive contamination of geological materials has attracted great attention especially as these naturally occurring radioactive materials (NORM) are known not only to have reached hazardous levels (El – Dine et al., 2001) but have been found to be capable of resulting in

much larger radiological exposure to the public relative to that caused by the nuclear industry for instance (Mokobia et al, 2006). This contamination has health implications.

An aspect of this pertains to ^{222}Rn (radon) a member of the ^{238}U/^{226}Ra (Uranium) decay series (Mokobia, 2004a; 2010). This short-lived radioactive gas (0.83days) decays into much shorter-lived ^{218}Po (Polonium) and ^{214}Pb (Lead) emitting α particle which causes cancer of the lungs if breathed in (Mokobia, 2004b). The other aspect has to do with the accumulation of ^{239}Pu (Plutonium) when ^{238}U interacts with atmospheric neutrons through (n, γ) nuclear reactions. These reactions result in the production of fissile materials (Mokobia et al, 2006) ^{233}U and ^{239}Pu as illustrated in the following equations:

[1]

$$^{238}U(n,\gamma)\ ^{239}U \xrightarrow{\beta^-} {}^{239}Np \xrightarrow{\beta^-} {}^{239}Pu \quad [2]$$

This gradual production of ^{239}Pu presents a crucial but unrecognized problem in radiation protection especially as it is reportedly toxic even in minute quantities. Several other radionuclides in this decay chain are known to be radiotoxic (Ahmed, 2004).

Talking specifically about kaolin, Zoltan and Richard (2005) have indicated that this mineral has low toxicity to aquatic species and that long-term exposure to it in humans may lead to a relatively benign pneumoconiosis usually referred to as kaolinosis.

In the light of these, an investigation of the radiological implications associated with the use of this valuable mineral is necessary. In this study, specifically, the qualitative and quantitative determinations of the natural radionuclides contained in kaolin of Nigerian origin were carried out using gamma spectrometric technique. The quantitative data obtained were then used to estimate the radiological hazard indexes associated with its wide usage.

MATERIALS AND METHODS

Samples of the mineral under study were collected from five locations around Ozanogogo (latitude $6^\circ 8' - 6^\circ 10'$ N and longitude $6^\circ 6' - 6^\circ 17'$ E) (Olobaniyi et al, 2007) Ika South Delta State Nigeria. On collection, these samples were securely tied in dark cellophane bags and properly labeled. The collected samples were washed, cleaned with acetone and air dried to constant weight at room temperature in the sample preparation laboratory at the Centre for Energy Research and Development (CERD), Obafemi Awolowo

University, Ile-Ife. They were separately crushed using a thoroughly cleaned crusher. Known masses of each were then weighed into Marinelli beakers and firmly sealed. The sealed containers were left for 28 days so that secular equilibrium between the parent and daughter nuclides present in the samples hitherto disturbed during the sample collection was restored in line with conventional practice (Mokobia et al., 2003 & 2006).

Gamma counting was subsequently carried out using a highly efficient NaI (TL) Scintillation detector. Energy and efficiency calibrations were carried out using mixed gamma-ray standard sources obtained from the National Bureau of Standards of the United States. Each sample as well as the mixed standard source and the background were counted for 10 hrs. It was ensured that the sample containers and that of the background and standard are of the same configuration so as to minimize error. Spectra evaluation was carried out using a PC based SAMPO 90 computer program. This aided the matching of the γ-energies at various levels to a library of possible isotopes and hence the qualitative determination of the radionuclides contained in the samples.

The activity concentrations (quantitative determination) of ^{238}U and ^{232}Th were

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$$A_{Ra_{eq}} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad [3]$$

The external health hazard index arising from the use of this mineral was determined by employing the equation used in Kpego, (2011):

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad [4]$$

In the equations [4] and [5], A_{Ra}, A_{Th} and A_K represent the activity concentrations of $^{238}U/^{226}Ra$, ^{232}Th and ^{40}K respectively in $Bqkg^{-1}$. The annual effective dose rate ($mSvy^{-1}$) was determined using the relationship:

$$E = TQD_t * 10^{-6} * F_t \quad [5]$$

D_t , the outdoor dose rate in air ($nGyh^{-1}$) was obtained from the relation (Farai and Jibiri, 2000):

$$D_t = 0.446A_{Ra} + 0.662A_{Th} + 0.048A_K \quad [6]$$

10^{-6} represents a conversion factor from nano to milli, T is 8760 hours per year, Q = 0.7 $SvGy^{-1}$ is the conversion coefficient from absorbed dose in air to effective dose and F_t = 0.2 is the outdoor occupancy factor (UNSCEAR,2000; 2008).

RESULTS AND DISCUSSIONS

The radionuclides detected in the gamma spectroscopic analysis belong to the ^{238} U and ^{232}Th series decay and the non series ^{40}K. Data in Table 2 shows that the specific activities of these primordial radionuclides detected in this mineral ranged from (95.3 ± 7.0 – 96.3 ± 3.4) $Bqkg^{-1}$, (99.7 ± 3. 8 – 99.9 ±5.5) $Bqkg^{-1}$ and (537.2 ± 2.5 – 543.6 ± 3.6) $Bqkg^{-1}$ respectively. These levels are comparable to those appearing in the literature for other though slightly higher (Zoltan et al, 2005; Turhan, 2009). This slight higher concentration might be attributed to higher radiological contamination of the Nigerian sample.

The contributions of these nuclides in the overall radioactivity contained in this geological sample as shown in Table 3 are 13%, 14% and 73% for ^{238}U, ^{232}Th and ^{40}K respectively. Thus ^{40}K presents the bulk of the radioactivity in natural kaolin respectively. This contribution is shown clearly in Figure 1.

The Radium-equivalent activity determined is 280.3 $Bqkg^{-1}$. This value is less than the maximum internationally recommended activity of 370 $Bqkg^{-1}$ (Kpego et al., 2011). Thus the radiological hazard index arising from the medical or industrial use of natural kaolin is within the recommended safety limit. The external hazard index is 0.76 while the mean annual effective dose equivalent is 0.16 $mSvy^{-1}$. These values are each less than unity also indicating that the use of this material is radiologically safe. The determined dose equivalent is below the internationally recommended dose criterion of 1 $mSvy^{-1}$ for members of the public (IAEA, 1996; Kpego et al., 2011). These suggest that the use of this mineral is radiologically safe.

CONCLUSION

From the results above, it is concluded that the use of natural kaolin does not pose any radiological health threat to the users and other members of thus this mineral is a radiologically friendly industrial geological material.

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Table 1: Gamma rays employed in the quantitative analysis

Element	Nuclide	Gamma ray energy (keV)	Emission ratio (%)
^{238}U	^{214}Pb ^{214}Bi	351.2 609.3, 1764.7	38.9 43.3, 17
^{232}Th	^{208}Tl ^{228}Ac	583.2, 2614.7 911.1	185.77, 99.79 27.7
^{40}K		1460.8	10.7

Table 2: Activities of nuclides detected in natural kaolin and determined hazard indices

Sample	^{40}K (Bq kg^{-1})	$^{238}U/^{226}Ra$ (Bq kg^{-1})	$^{232}Th/^{228}Ra$ (Bq kg^{-1})	Ra- Eq activity (Bq kg^{-1})	External hazard Index
K1	540.2 ± 4.0	95.9 ± 4.0	99.9 ± 5.5	280.35	0.165
K2	537.2 ± 2.5	96.3 ± 3.4	99.8 ± 6.4	280.38	0.165
K3	543.6 ± 3.6	95.9 ± 3.4	99.8 ± 3.9	280.47	0.166
K4	541.0 ± 6.4	95.3 ± 7.0	99.7 ± 3.8	279.53	0.165
K5	540.6 ± 3.6	96.2 ± 7.2	99.8 ± 6.3	280.54	0.165
Mean	540.5 ± 20.2	95.9 ± 25.0	299.50 ± 25.9	280.54	0.165

Table 3: Percent (%) Contribution of ^{226}Ra, ^{228}Ra and ^{40}K to the total radioactivity in kaolin

Sample	^{226}Ra	^{228}Ra	^{40}K
K1	**13.03**	**13.57**	**73.40**
K2	13.13	13.61	73.26
K3	12.97	13.50	73.53
K4	12.95	13.55	73.51
K5	13.06	13.55	73.39
Mean	13.03	13.56	73.42

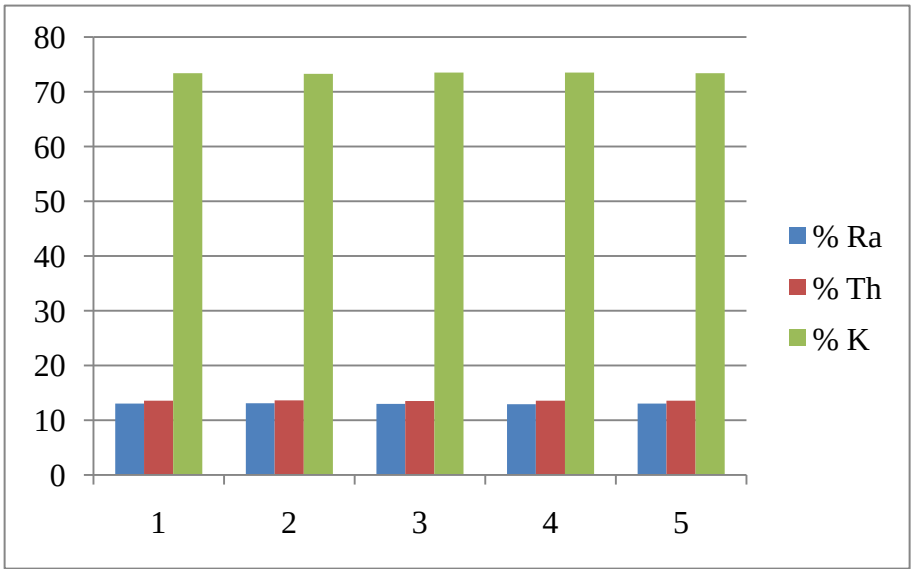

Fig. 1: Diagram showing the % contributions of Ra, Th and K in the radioactivity in natural kaolin.