CHARACTERIZATION OF KAOLIN FROM NARAGUTA CLAY DEPOSIT IN JOS NORTH LOCAL GOVERNMENT AREA OF PLATEAU STATE FOR THE PRODUCTION OF CERAMIC MATERIAL

Akande, H. F., Mumah, S.N., Mudi, K. Y. and Samuel, F.
Department of Chemical Engineering, Kaduna Polytechnic, Kaduna, Nigeria
Contact: +234-8033686645, hassan.akande@kadunapolytechnic.edu.ng

ABSTRACT

Clay Samples from Naraguta deposit in Jos North Local Government Area of Plateau State, Nigeria have been studied for its application as raw material for ceramic material production. The effects of firing temperature on the physical properties were also studied. Naraguta clay, samples were obtained randomly from different points within each deposit from underground local mines pit at a depth of 10, 20 and 30 cm and labelled samples 1, 2 and 3 respectively. The samples were air-dried for several days and crushed using a crusher. Each crushed sample was thoroughly mixed, coned and quartered. The samples were subsequently balled milled to form a powder. The powder was pressed to form pellets which were dried at 105 ^oC until a constant weight was achieved. The samples were then heated to temperature 750, 800, 850, 900, and 950 ^oC in a furnace, respectively at a rate of 5 ^oC/min with soaking time of 1h at each maximum temperature. The elemental analysis was carried out using X-ray fluorescence (XRF) while the structural analysis was done using X-ray diffraction (XRD).The property tests such as bulk density, apparent porosity, compressive strength and shear stress were carried out using standard techniques such as American Society for Testing and Materials. The results of the elemental analysis revealed that the clays were composed majorly of silica (SiO_2), 49 – 51%; alumina (Al_2O_3), 31.49 – 33.30% with the presences of some other oxides. The XRD spectra of the clay showed that the components of the clay samples were predominantly kaolinite constituting about 80% of the particles with some traces of mica and quartz. The physical properties (bulk density, compressive strength and shear strength) of the clay sample initially increased with increased firing temperature from 750 ^oC to 800 ^oC, and then the properties decreased with further increase in firing temperature until it reached 950 ^oC because of the transformation of the clay due to dehydroxylation of kaolin to meta-kaolin. The results of both the physical and chemical analysis fall within the acceptable standards for materials for ceramic material production.

Key Words: *Kaolin, Ceramic Material, Chemical and Physical Analysis, X-Ray fluorescence, X-Ray Diffraction*

1. INTRODUCTION

Ceramics are inorganic and nonmetallic solids with a range of useful properties, including very high hardness and strength, extremely high melting points, and excellent electrical and thermal insulation. The best-known ceramics are pottery, glass, brick, porcelain, and cement (Emmanuel 2015). Clay remains the major raw material in the ceramic industry. It has also found wide applications in the paint and paper industries. This versatility depends principally on the composition and physical properties. The properties of a particular ceramic depend not just on the materials from which it is made but also on the crystalline structure**.** The clay used for the production of ceramics should have the following specifications; plastic enough for shaping, dry without excessive cracking and warping, have low and wide vitrification range, low carbonate content, and a good spread in particle size. (Angelica *et al*., 2008). Low and wide vitrification range is desirable for favourable unit firing cost and for ease of firing. Because the fluxing action of carbonates tends to shorten the firing range and trigger localized flux spots, the low carbonate content is desirable, and CaO and MgO resulting from the firing of typical carbonate-bearing clay can hydrate relatively easily with no damage to the fired material.

In the fabrication of ceramic products, raw materials selection plays a vital role in the final product. The properties of the components present in the clay and the firing temperature affect the physicochemical properties of the resulting ceramics (Jordan et al., 1999) critically.

The sintering process has been widely used in producing ceramic components. Sintering processes can be divided into two types: solid-state sintering and liquid phase sintering. Solid-State Sintering (SSS) happens when the powder compact is fully densified in a solid-state at sintering time, whereas fluid phase sintering occurs when the powder compact is still in a liquid phase during sintering (Kang, 2005).

Sintering of ceramic mass is therefore critical in adjusting desired properties and occurs in general during liquid-phase formation. The presence of components such as low-melting clays is significant in the process. Fluxes (alkaline oxides, mainly K_2O and Na_2O), in reaction with silica and alumina, promote liquid phase formations and therefore assist the densification process (Hamisi *et al.*, 2014). Evaluating the phase transformations which take place during the firing process is, therefore, crucial in determining the quality of the clay used in the formulation of ceramic materials (Sousa and Holanda, 2005).

According to Emmanuel (2015), Nigeria is immensely endowed with ceramic raw materials like alumina, coal, clay, feldspar, kaolin, quartz, silicon, and zirconium, located throughout the country's geographical zones. Local demands for ceramic products are very high, considering the population of Nigeria which is presently about 180 million, yet the supply is met mostly through importation. Despite the fact that both raw materials and human capacity are available for research, production and development of high-quality ceramic products of international standard, the country imports more than 50 containers a day (Emmanuel, 2015). The author also reported that the housing deficit in *Nigeria* is about 17 million units with an annual growth *of* 2 million units. With massive developments in *real estate sector* in the country to meet the increasing *demands* for decent housing, *the demand for ceramic tiles which are used for floor, walls, ceiling and even roof* is *high* in the country.

Next is the power sector with its high demand for electrical porcelain insulators. Another area worth considering is ceramics for home use. This includes dinner wares, pottery wares and decorative wares. Other products such as spark plug ceramics, beryllium oxide ceramics, chemical or refractory porcelain, engine and turbine combustion chambers can be categorized for industrial applications (Emmanuel, 2015).

It makes economic sense to invest in the production of ceramics in Nigeria at this time with all these demands in the different sectors, even as the raw materials are readily available in commercial quantities. This will not only help meet the growing demand for these products but will also help create jobs and help the state economy to grow. Clay bodies are widely distributed on the Precambrian basement complex of Nigeria. Clay occurs in deposits of greatly varying nature. No two deposits have the same clay, and frequently different samples of clay from the same deposit differ. Due to its small size, complex structural structure and relative slow kinetics of forming and transition, clay can be challenging to define.

Naraguta clay deposit is located in Jos North Local Government Area of Plateau State, North Central Nigeria. Clay samples were collected from three different locations in the upper part of the area. The study areas are bounded between latitudes 8˚30E and 9˚00E and longitudes 9˚30N and 10˚00N. Fresh samples in lump form were obtained randomly from different points within each deposit.

This work is therefore to evaluate the properties of kaolin from Naraguta clay deposit in Jos North Local Government Area of Plateau State, Nigeria to determine its suitability for ceramic materials production.

EXPERIMENTAL PROCEDURE

Sample Preparation

The material preparation was carried out using the procedure adopted by Ituma *et al*. (2018) and Lydia *et al*. (2018). In this experiment, 5 kg of fresh samples in lump form were obtained randomly from different points within each deposit from underground local mines pit at a depth of 10, 20 and 30 cm and labelled samples 1, 2 and 3 respectively. The samples were air-dried for several days and crushed using a set of Denver crushers by Denver Equipment Co. England. Each crushed sample was thoroughly mixed, coned and quartered. These were packaged in small polythene bags as representatives of the samples for the required test. The clay sample was subsequently ball milled to form a powder. The powder was pressed to form pellets which were dried at 105 ^{o}C until a constant weight was achieved. The samples were then heated to 750, 800, 850, 900, and 950 ^{o}C, respectively at a rate of 5 ^{o}C /min with soaking time of 1h at each maximum temperature.

Sample Characterization

The elemental compositions of the clay samples were determined using X-ray fluorescence (XRF). The clay samples were mixed inside a cleansed crucible and heated to 500 °C for 8 min and allowed to cool at room temperature until the sample fused and later used for the analysis. The chemical composition of the sample is presented in Table 1.

The X-Ray Diffraction (XRD) patterns and phase identifications of the samples were carried out using X-ray diffractometer (XRD); PW 1800 diffractometer, Philips, the Netherlands with graphite monochromatized copper Kα-radiation.

Apparent porosity

Apparent porosity of the clay material was carried out by preparing a clay brick measuring 5x5x4 cm. The brick was then dried in an oven at 100 °C, fired in a furnace at a temperature of 900 °C. The fired specimen was cooled and then transferred into a desiccator, and the dry weight noted and recorded as x. The specimen was then immersed in a water-filled beaker, allowed to soak in boiled water for 30 min while being agitated to free trapped air bubble. The specimen was then allowed to cool in a desiccator and then soaked weight recorded as y. The specimen was weighed suspended in water using beaker placed on a balance, and the suspended weight noted and recorded as z. The apparent porosity was calculated using Equation (1) (Abubakar *et al*., 2014)

$$\text{Apparent porosity} = \frac{\mathrm{y-x}}{\mathrm{y-z}} \times 100\% \qquad (1)$$

Bulk density

The samples were dried for 24 h, the test pieces were put in an oven and dried at a temperature of 110 °C for 6 h. They were allowed to cool and weighed by means of weighing balance and their dried weights (D) recorded in turn. They were transferred to a beaker and heated for 40 min and then cooled, the soaked weight (W) was recorded. Water was put in another beaker and each of the test pieces suspended in the water so that their suspended weight (S) were recorded. The bulk density was calculated using 2 (Ovat and Bisong 2017).

$$\text{Bulk density} = \frac{\mathrm{D\rho W}}{\mathrm{w-s}} \qquad (2)$$

Where D = Dried weight
W = Soaked weight
S = Suspended weight
ρW =Density of water

Loss on ignition

The loss on ignition was calculated by measuring 50 g of the sample, dried in an oven at 110 °C and cooled in a desiccator. A porcelain crucible was cleaned, dried and weighed (A). The dried sample was introduced into the crucible and weighed (B). The crucible containing the sample was placed in a muffle furnace and heated to a temperature of 900 °C for 3 h, cooled in a desiccator and then weighed (C). The loss on ignition was calculated from Equation (3) (Abubakar *et al.*, 2014)

$$\text{LOI} = \frac{\mathrm{B-A}}{\mathrm{B-C}} \qquad (3)$$

Compressive and Shear Strength of the Kaolin Sample

The compressive strength was determined according to the ASTM Standard Test Method (D2166-13). Clay samples were ball milled to form a powder. The powders were pressed to form pellets which were dried at 105°C until a constant weight was achieved. The samples were then transferred into a furnace and heated to a temperature of 750, 800, 850, 900, and 950°C, respectively, at a rate of 5°C/min. The samples were cooled to room temperature. The samples were placed on a compressive tester and load was applied by turning the handwheel of the tester at a uniform rate of 10 N/m^2 until failure occurred. The manometer readings were recorded, and the compressive strength (CS) and Shear strength were calculated using Equation (4) (Abubakar *et al*., 2014).

$$CS = \frac{maximum\ laod\ (N)}{cross\ sectional\ area\ (m^2)} \qquad (4)$$

RESULTS AND DISCUSSION

The chemical composition of kaolin is essential because of its influence on the behaviour of ceramic masses during thermal treatment. Table 1 shows the oxide compositions of the clay samples.

Table 1: Chemical Composition of the Kaolin Sample

Oxide	Kaolin Sample 1 (%ppm)	Kaolin Sample 2 (%ppm)	Kaolin Sample 3 (%ppm)
CuO	0	0	0
NiO	0	0	0
Fe_2O_3	0.483	0.387	0.694
MnO	0	0	0

Oxide	Kaolin Sample 1 (%ppm)	Kaolin Sample 2 (%ppm)	Kaolin Sample 3 (%ppm)
Cr_2O_3	0.011	0.009	0.017
TiO_2	1.112	0.801	1.483
CaO	0.083	0.106	0.093
Al_2O_3	31.494	32.878	33.301
MgO	0.328	0.317	0
ZnO	0.004	0.004	0.006
SiO_2	49.038	51.102	50.673
Total	82.553	85.604	86.267
LOI	17.447	14.396	13.733

From Table 1, it could be deduced that the silica contents of the samples are 49.04, 51.10 and 50.67% of clay sample 1, 2 and 3, respectively. This showed that all the clay samples met the requirements for the manufacture of ceramics, as reported by Abubakar *et al*. (2014). In their work, they reported that a clay material having silica contents of between 40 – 65 % satisfies the requirements for the manufacture of refractory bricks and ceramics. This may be due to the presence of higher bond impacted into the matrix of the clay by the silica content.

An Al_2O_3 content higher than 30 % is necessary for increasing the refractory and mechanical resistance (Benea and Gorea, 2004). From Table 1, it could be deduced that the alumina contents of the clay samples were found to be between 31 – 33%, an indication that the samples fell within the range of standards for the manufacturing of ceramics (above 30%) and refractory bricks (25 – 44%). From Table 1 also, the Fe_2O_3 contents of the clay samples are 0.483, 0.387 and 0.694 % for sample 1, 2 and 3, respectively. These values of the Fe_2O_3contents also fell within the range of standards (0.4 – 2.4 %) for the manufacture of refractory bricks. Such level of Fe_2O_3 usually alters the colour of the clay to reddish-brown, thereby making it attractive as a ceramic material as reported by Marcel and Bernea (2004). Marcel and Bernea (2004) reported that the Fe_2O_3 content of clay influenced the high-temperature characteristics of the clay such as the fired strength.

The Loss on Ignition (LOI) values of the clay samples was within 13.73 – 17.45%. The values, which fell within the range of 8 – 18 % for ceramics and refractory bricks products as reported by Omowumi (2000). Omowumi (2000) reported that the LOI characteristics of the clay materials should be low in other not to impact adverse effects on the porosity of the materials especially those used for refractory bricks.

The bulk density, compressive strength and shear stress of the fired clay are presented in Table 2, 3 and 4, respectively.

Table 2: Compressive Strength of the Kaolin Sample

Temperature (^oC)	Compressive Strength (N/m^2)
750	7.86
800	7.88
850	7.32
900	7.43
950	8.21

Table 3: Shear Strength of the Kaolin Sample

Temperature (^oC)	Shear Strength (N/m^2)
750	1.76
800	2.27
850	1.24
900	1.23
950	2.86

Table 4: Bulk Density of the Kaolin Sample

Temperature (^oC)	Bulk Density (g/cm^3)
750	1.55
800	1.63
850	1.59
900	1.60
950	1.56

From Tables 2 - 4, it could be observed that the properties (bulk density, compressive strength and shear strength) of the fired bricks produced from the kaolin sample initially increased with increased firing temperature from 750^{o}C to 800 ^{o}C, and then the properties decreased with further increase in firing temperature until it reached the minimum at 950 ^{o}C for the considered firing temperatures. The observed trend could be as a result of the transformation of the clay sample used for producing ceramic from one phase to another. As the dehydration of kaolinite completes by ~150 °C, followed by dehydroxylation at ~500-600 °C and its structural breakdown occurs in the temperature range ~800-900 °C, depending upon the particle size and amount and type of the impurities present (Francisca, 2014). Also, the bulk density of 1.56 g/cm^3 obtained at a fired temperature of 950 °C showed that the sample is suitable for producing ceramic materials. The apparent porosity of the clay samples is presented in Table 5.

Table 5: Apparent porosity of the clay sample

Clay samples	**1**	**2**	**3**
Apparent porosity	4.1	5.8	8.2

The apparent porosity of the clay samples as revealed in Table 5 were found to be 4.1, 5.8 and 8.2 for samples 1, 2 and 3, respectively. These low porosities exhibited by these clay materials fell within the standard for the production of fired clay and refractories as reported by Tiffo *et al.* (2015). Tiffo *et al.* (2015) reported that low porosity found in a clay sample leads to an increase in their mechanical strength. Therefore, these samples are found suitable for the manufacture of refractories, ceramic materials and siliceous fired clays.

The phases present in the clay samples and their crystallinity were identified using XRD, as shown in Figure 2.

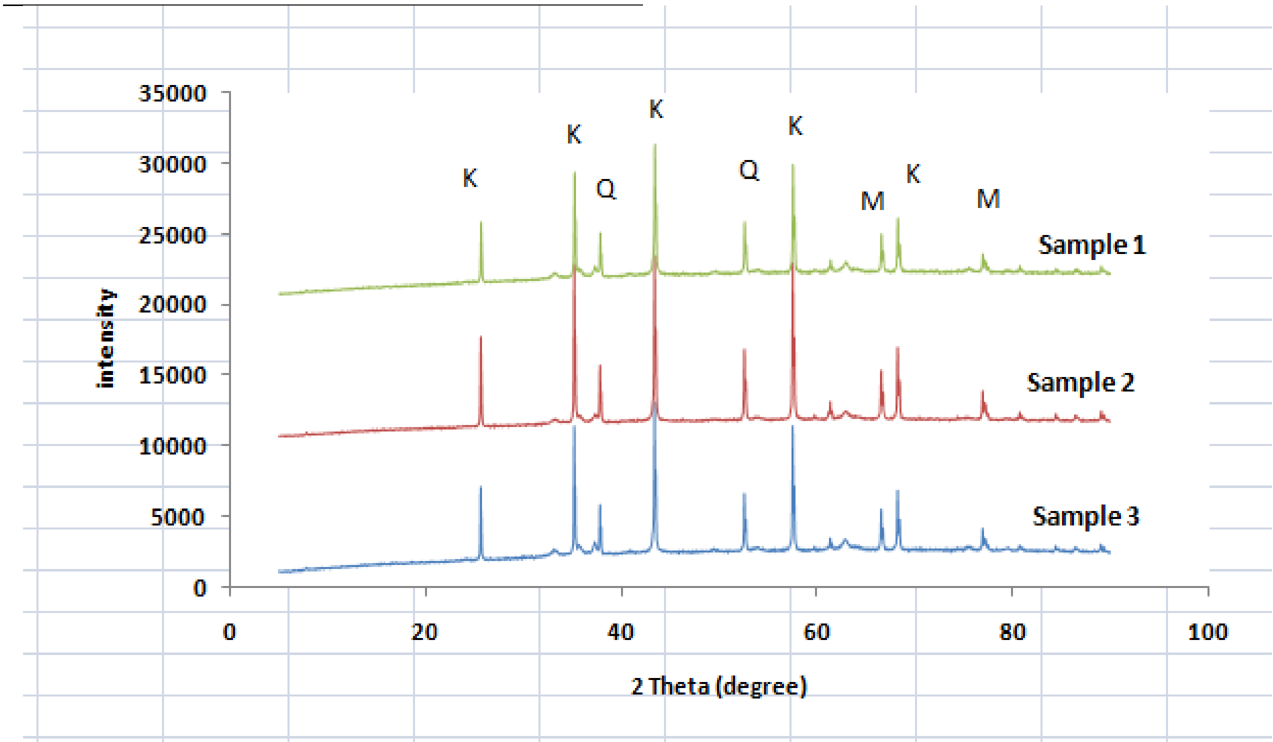

Figure 2: XRD spectra of Jos clay samples

The results, as presented in Figure, showed nine major peaks at diffraction 2θ angles, of 24.9, 33.1, 38.3, 43.8, 52.2, 57.9, 67.1, 68.9 and 78°. The presence of mica and quartz was as a result of silicate minerals present in the kaolin sample. The XRD study confirmed that the major component present in the clay samples were predominantly kaolinite constituting about 80 % of the particles. Other components include mica and quartz. The average lattice parameters from indexing of the peaks in the XRD were calculated to be 0.999 nm. The particle diameter of the kaolin sample was calculated using the Debye-Sherrer, Equation (5)

$$D = \frac{0.9\lambda}{\beta\cos\theta} \qquad (5)$$

Where λ is the wavelength of the x-ray (0.1541 nm) β is the full width at half maximum (FWHM), θ is the diffraction angle in radian, and D is the particle diameter size. The interspacing between the atoms (d) was calculated using Bragg's law,Equation (6).

$$d = \frac{\lambda}{2\sin\theta} \quad (6)$$

The average interspacing between the atoms, average crystal size and average lattice parameters of the study clay samples calculated are 0.3764 nm, 17.480 nm and 1.003 nm, respectively. These indicated that the clay samples used showed had fewer spaces between the particles. This wider angle between the unit cells of the kaolin crystal of 1.003nm is more than general well-crystallized kaolinites having a graphitic line of 001 d-spacing of 7.1 to 7.2 as reported by Aroke *et al.* (2013). The difference could be connected with a small amount of interlayer water within the kaolin crystals. However, there were some incompleteness and imperfections in some patterns in Figure 2, indicating the overshadowing amounts of other minerals of clay size with the kaolin. Some peaks related to crystallized phase were also detected, which is attributed to quartz, and mica initially present in the kaolin. This observation was similar to the findings of Francisca (2014).

CONCLUSION

Clay Samples from Naraguta deposit in Jos North Local Government Area of Plateau State were prepared and characterized using physical and chemical analysis. The results of the chemical analysis showed that kaolinite was the major components of the clay with the composition of silica (SiO_2), 49 – 51%; alumina (Al_2O_3), 31.494 – 33.301%; and other oxides. The bulk density, compressive strength and shear strength of the clay was stable at a temperature of 950 ^oC due to dehydroxylation of kaolin. It was found that the physical properties and chemical compositions of the clays are within the acceptable standard ranges for the manufacture of ceramics.

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