JMME. Vol. 13, April, 2021
pp. 39 – 46
ISSN: 2006-1919

EFFECTS OF THERMAL TREATMENT OF AMA NGUZU CLAY FOR REFRACTORY BRICKS PRODUCTION

O.M. Chima,
Department of Mechanical Engineering, Michael Okpara University of Agriculture, Umudike, Nigeria.
Email: chimamelford@gmail.com.

ABSTRACT

The effects of thermal treatment on the properties of clay bricks produced from Ama Nguzu clay were studied through transformation processes in the clay matrix upon firing. The clay sample was collected, processed and characterized for mineralogical and chemical oxide composition using x-ray diffractometer (XRD) and x-ray Florence (XRF) respectively. The processed clay was used to develop test samples of the refractory brick. They were dried at 110^{0}C and thereafter fired at different temperatures of 900^{0}C, 1000^{0}C, 1100^{0}C and 1200^{0}C at the rate of 4^{0}C/min for 1 hour. The fired samples were tested for the refractory properties. The XRD results indicated quartz as the dominant mineralogical phase while that of XRF showed the presence of major oxides such as alumina, silica and iron oxide. The results obtained for the refractory property tests showed that the linear shrinkage, bulk density, modulus of rupture and refractoriness respectively increased from 4.77 – 6.75%, 1.605 – 1638g/cm^3, 10.94 – 19.93N/mm^2 and 1450^{0}C - 1650^{0}C when the fired temperature increased from 900^{0}C - 1200^{0}C. With the same trend in the fired temperature, the apparent porosity, water absorption and thermal shock resistance decreased from 39.5 – 30.25%   24.61 – 22.14% and 31 – 28 cycles respectively. It was found that the properties are interrelated and are either directly or inversely related to the fired temperature. It was also found that the properties of the clay depended on other factors such as the mineral phase and chemical oxide constituents which are related to the fired temperature. Therefore, it was concluded that the firing temperature range of 1100 - 1200^{0}C is most appropriate to yield optimum performance for the refractory brick understudied. However, low porosity due to over firing does not yield impressive spalling resistance.

Key words: *Clay, Properties, Temperature, Refractory, Performance, Oxide.*

1.0 INTRODUCTION

Refractories are materials which possess high melting point alongside with other high temperature characteristics that make them suitable to function as heat resisting barriers between high and low temperature zone [1]. The principal raw materials used in the production of refractories are normally the oxides of silicon, magnesium, calcium and zirconium. Other non-oxide refractories are carbide, nitrides, borides, silicates and graphite [2]. Clay is the commonest raw material used in the development of refractory brick in our local industries [3]. Such application depends on the clay structure, composition and processing method [4]. The fired temperature is one of the factors that determine the quality of the refractory brick [5]. The clay material contains a lot of mineralogical phases and oxides, which undergo several transformation stages upon firing. When clay is heated, it undergoes processes such as drying, dehydration, burning out of carboneous matter, quartz inversion, oxidation, sintering and vitrification. These processes take place at various respective temperature ranges of 30 – 150^{0}C, 150 - 300^{0}C, 350 - 450^{0}C, 500 - 600^{0}C, 538 - 983^{0}C, and 900 - 1000^{0}C. [6].

The firing processes in brick production involve heating the material to an elevated temperature usually above 900^0C. This develops the inter particle bond, the strength, the pore structure and the colour of the product [6]. The bond often results in fusion of more constituents of the composition. Thermal treatment of clay can lead to changes in the chemical and physical structure depending on the clay minerals and type of exchange ion. Different temperature ranges in which significant changes occur in the structure of clay mineral may be identified to ensure proper processing method that guarantees the desired properties of the product [7]. The thermal process involved in the production affects the vitrification and sintering reactions. The quality of refractory product is dependent on both reactions [3]. Hence, optimum firing in thermal treatment of clay is required to ensure that the desired properties of the product for specific application are guaranteed.

In brick production, firing demands enormous energy which takes high percentage of the production cost. This demands that the production condition should be optimized by good temperature management to ensure minimum production cost with excellent quality of product. This is because over-firing of the brick could consume energy which has direct implication on the production cost and the quality of the brick. On the other hand, under-burning results in poor quality of the brick which makes the product brittle. Therefore, it is obvious that poor temperature management is a major challenge that is facing industrial production of refractory brick. This has prompted many research studies to focus on this area [7, 8, 9, 10, 11, 12].

Although much work has been done on this area, it has been found that there is need to focus this study on the interrelationship between the thermal treatment, mineralogical and chemical composition as it relates to the physiomechanical and thermal behaviour of the brick product. The knowledge from this study could be utilized in improving manufacturing standards of quality products and also to optimize the production condition in our local brick industries.

2.0 MATERIALS AND METHODS

The clay sample was mined from AmaNguzu clay deposits in Nguzu, Edda, Ebonyi State, Nigeria. It was prepared for X-ray Diffraction (XRD) and X-ray Fluorescence (XRF), analyses, in accordance to the standard procedure. The analyses were carried out to determine the mineralogical phase composition, and proportions of the constituent chemical oxides in the clay sample before firing. The test samples of the clay were crushed and ground to fine particles. The mineralogical phase was identified with the aid of computerized Empyrean X-ray diffraction equipment that uses Cu Kα radiation at a scan speed of 4^0/min 2ϴ.in National Steel Raw Material Exploration Agency, Kaduna. The results were interpreted using International Centre for Diffraction Data (ICDD) software which revealed the main mineralogical phases. The chemical oxide analysis was carried out at Defense Industries Cooperation of Nigeria (DICON), Kaduna. Quantitative analysis of the major minerals within the samples was done by X-ray Fluorescence Spectroscopy using a Magi X Pro XRF Spectrometer.

The raw clay sample was air dried and crushed and ground in a mortar to an average particle size of 0.425mm. The sample was soaked in a container of water and thoroughly mixed together to ensure proper dissolution. The dissolved clay was then filtered through a sieve to get rid of unwanted particles and to obtain finer particle size. The water content was removed to obtain processed clay which was further dried at 100^0C. Finer particles of the processed clay was achieved through further grinding and sieving using smaller sieve size of 18µm. Good plasticity of the clay material required for moulding was achieved by mixing the clay with 35% weight of water and blended to produce homogeneous clay body. The appropriate quantity of the plastic clay mass was put into the respective moulds designed with various dimensions for different tests specified in the study. The moulding clay mass was compacted using an improvised wooden piece which imparted the required compacting pressure on the mass. The moulded test pieces were dried at 110^0C and fired at various firing temperatures of 900, 1000, 1100 and 1200^0C in line with the set objectives of the study.

The fired samples were analyzed for refractory properties such as linear shrinkage, apparent porosity, bulk density, water absorption, modulus of rupture, thermal shock resistance and refractoriness. The linear shrinkage was determined by obtaining the percentage change in length after firing. The expression was mathematically presented as shown in equation 1.

$$\frac{Ld - Lf}{Ld} \text{ x } 100 \qquad (1)$$

The Archimedes' method was used to determine the water absorption, apparent porosity and bulk density as stated in previous literature. [15, 23]. Hence, the samples were weighed dry to obtain dry weight (W_1) and thereafter introduced into boiling water for 4 hours. The samples were then reweighed when suspended in water to get the suspended weight (W_2) and weighed in air to obtain the saturated wet weight (W_3). The values of the respective properties were computed as shown in equations 2 – 4.

$$\text{Water Absorption} = \frac{W3 - W1}{W1} \text{ x } 100 \qquad (2)$$

$$\text{Apparent porosity} = \frac{W3 - W1}{W3 - W2} \text{ x } 100 \qquad (3)$$

$$\text{Bulk density} = \frac{W1}{W3 - W2} \qquad (4)$$

The modulus of rupture was determined using electrical transversal strength testing machine. A breaking load was applied to ensure that the brick material is fractured. The values of the breaking load, P (kg) of the specimen at fracture, the distance between supports of the transversal machine L (cm), height H (cm) and the width B (cm) of the broken pieces were determined and recorded. The modulus of rupture was then calculated as:

$$\text{MOR (MPa)} = \frac{3PL}{2BH^2} \qquad (5)$$

The thermal shock resistance and refractoriness were determined using prism spalling test method according to ASTM C-484 standard and pyrometric cone equivalence (PCE) method respectively. [1] and [22] described these standard test procedures in detail.

3.0 RESULTS AND DISCUSSION

Figure 1 shows the x-ray diffraction pattern for the clay material. The results presented in Table 1 show the chemical composition of the clay material while those in Table 2 show the refractory properties of the clay brick at various fired temperatures.

From the results shown for the clay brick's properties, it was observed that the linear shrinkage values range from 4.77 – 6.75%. This range of value was found in agreement with the acceptable range for refractory bricks as reported by [13]. The material stability could be traced to the presence of quartz which acts as filler as well as a modifier of fired body properties that lowered the shrinkage. The presence of quartz decreases shrinkage and reduces the tendency to warp during firing.[2].

Table 1: Composition of the AmaNguzu Clay

Oxide	**Al_2O_3**	**SiO_2**	**K_2O**	**CaO**	**TiO_2**	**V_2O_5**	**Fe_2O_3**	**Cr_2O_3**	**MnO**
AmaNguzu Clay	42.9	51.1	0.22	0.035	1.85	0.065	2.923	0.015	0.0066
	CuO	**ZnO**	**Ga_2O_3**	**MoO_3**	**Ag_2O**	**Re_2O_7**	**WO_3**	**SiO_3**	
	0.009	0.002	0.025	0.01	0.658	0.066	0.048	0.10	

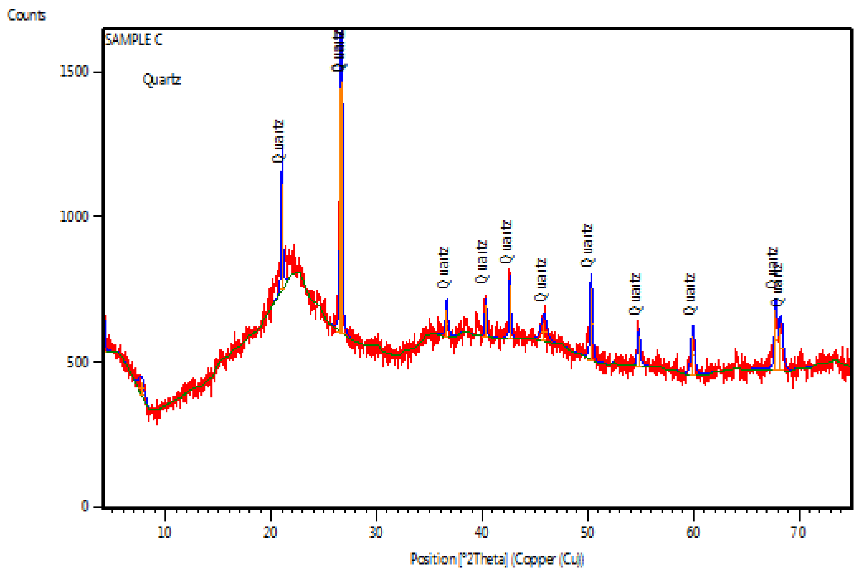

Figure 1. X-ray Diffraction Pattern for AmaNguzu Clay Material

Table 2: Analysis of Refractory Properties of the AmaNguzu clay based bricks.

Temperature (^{0}C)	Linear Shrinkage (%)	Water Absorption (%)	Apparent Porosity (%)	Bulk Density (g/cm^3)	Modulus of Rupture (N/mm^2)	Thermal Shock Resistance (cycle)	Refractoriness (^{0}C)
900	4.77	24.61	39.5	1.605	10.94	31	1450
1000	6.09	24.33	39.1	1.609	13.98	30	1500
1100	6.4	23.94	38.6	1.614	15.85	28	1600
1200	6.75	22.14	36.25	1.638	19.93	27	1650

Besides, the moderate percentage composition of silicon assisted to maintain a good dimensional stability, retained its shape and prevented shrinkage [5].

The result also indicates that linear shrinkage of the clay brick increased from 4.77% to 6.75% as the firing temperature increased from 900^0C to 1200^0C. This indicates that linear shrinkage has a direct linear relationship with the fired temperature. Shrinkage in clay body sets in as chemically bonded water is lost. Comparison of the level of shrinkage with fired temperature shows that linear shrinkage was higher in the temperature range of 1100 -1200^0C than at 900 - 1000^0C. This could be traced to the commencement of sintering process within the temperature range of 1100 -1200^0C. This must have caused solid state reaction that led to condensed mass which decreased the original linear dimension [2].

The result for apparent porosity test confirmed that values within the range of 39.50 – 38.60% were obtained between 900^0C and 1100^0C. However, upon increase of the fired temperature to 1200^0C, the apparent porosity value reduced drastically to 30.26%. This indicates that when clay body is fired above 1100^0C, there is both loss of water, chemical reactions and structural and phase transformations. These changes led to collapse of the interlayer spaces which gave rise to changes in the pore spaces and consequently decreases in the apparent porosity of clay brick. [7, 2].

It has been reported that sintering occurs between 1125^0C - 1200^0C. [2, 14]**.** This process assists in consolidation of the particles. The solid-state reaction forms solid bonds between these consolidated particles which reduces the free surface, eliminates grain boundaries with pore volumes, leading to condensed mass. This consequently reduces the apparent porosity and water absorption capacity while bulk density is increased.

Other possible cause of decrease in apparent porosity value is attributed to the oxide composition. The presence of some low temperature fluxing agent such as K_2O as observed in Table 1 is believed to have affected the porosity values. This oxide easily forms liquid phase at elevated temperature which contributes to the densification of clay body by filling the intra and inter granular area at high temperature [2,15]. The low porosity is useful in conferring high resistance to abrasion, to slagging and to the attack of gases which could occur in the blast furnace [16].

The results indicate that water absorption capacity of the clay brick decreased from 24.61% - 22.14% as the fired temperature increased from 900^0C - 1200^0C. It also confirmed that bulk density values increased from 1.605 – 1.638g/cm^3 in the same trend with fired temperature. Comparison of the effect of thermal treatment on the three properties shows a strong correlation. This confirms that the properties of clay bricks are interrelated. Water absorption is a measure of available pore space which is expressed as a percentage of the dry brick weight [11]. Upon increase in the fired temperature, pore volume reduces to increase the bulk density and decrease the water absorption capacity.

The modulus of rupture is a measure of the transverse strength. From the results obtained, it was observed that the value of modulus of rupture progressively increased from 10.94 – 19.93 as the fired temperature increased from 900^0C - 1200^0C. This shows that the effect of the fired temperature on one property has direct implication on other properties. The results show that the strength of clay body generally increases with decrease in porosity as fired temperature increases as confirmed by other authors [15,17].

Hence, the mechanical strength is found to have a close relationship with apparent porosity of clay material.

These pores were observed as stress raisers in line with the Griffith theory of fracture mechanics. This states that the magnitude of stress amplification (stress concentration factor) depends on the crack orientation and geometry. [18]

From the results, it is evident that as the fired temperature increased, the pore sizes reduced. Consequently, the length of the pore is reduced. This process reduces the stress concentration factor which makes the material to have more resistance to mechanical stress due to load impact.ie the degree to which the stress is amplified is reduced. The effect of porosity on this mechanical property was also based on the fact that decrease in porosity with increase in fired temperature is assumed to have increased the effective cross sectional load bearing area such that the property depended on the maximum contact solid area.

The results for thermal shock resistance showed decrease in values from 31 – 27 cycles as the fired temperature increased from 900^0C - 1200^0C. The decrease in spalling resistance could be attributed to the decrease in the pore sizes as fired temperature increased. This is because large pore sizes impeded the movement of crack propagation as obtained at lower firing temperature level. It could also be traced to the effect of thermal conduction. The thermal conductivity of a material determines the temperature gradient set up [16]. The thermal conduction of a brick material is a function of pore sizes and their distribution in the brick structure [15]**.** When the pores are larger, it accommodates a larger quantity of air which is a poor conductor of heat and therefore offers more resistance to thermal flow. Consequently, the effect of thermal induced stress is reduced. However, as the fired temperature increased, the pore size decreased. This gave way for the crack to propagate easily across the grain boundaries. Hence, the material's ability to resist thermal stress due to thermal fluctuations was affected. The result therefore confirms previous finding that stated that low porosity due to high flux content and over-firing can give rise to poor spalling resistance. [16].

The refractoriness of the clay bricks was observed to be a function of fired temperature. This is because it increased from 1450^0C – 1650^0Cas firing temperature was increased from 900^0C – 1200^0C. The presence of oxide like alumina accounted for enhancement of refractoriness at such temperature as reported by previous authors [19]**.** This is because though alumina is said to react with silica to form mullite as shown in equation 1. The sintering of aluminosilicate raw materials such as clay leads to the synthesis of mullite ($3Al_2O_3.2SiO_2$) [20]**.**

$$3Al_2O_3 + 2SiO_2 \rightarrow \mathbf{3}Al_2O_3.2SiO_2 \quad (6)$$

Mullite phase is reported to form at the temperatures above 1100^0C [21]. This phase is considered as binding type in most refractory brick. It has high resistance to melting and minimum thermal expansion. It is also characterized by good thermal stability with other high temperature characteristics. Hence, is was noted that the formation of the phase began when the fired temperature rose to 1100^0C. This was responsible for enhancement of refractoriness at the high point of firing temperature.

4.0 CONCLUSIONS

This study confirms that Ama-Nguzu clay consists of quartz mineral with alumina and silica as the dominant chemical oxides alongside with small percentage composition of iron oxide. The increase in firing temperature showed increase in refractoriness, modulus of rupture, bulk density and linear shrinkage with decrease in water absorption and apparent porosity. The progressive increase in firing temperature yielded non-impressive values of thermal shock resistance which could make the material prone to fracture when subjected to thermal stress due to sharp temperature gradient. The presence of both the oxide and mineral constituents also influenced the properties examined. Thus, the firing temperature range of 1100 - 1200^0C can be considered most appropriate for optimum performance for the refractory brick understudied. Therefore, the interrelationship between the thermal treatment, chemical oxide and mineralogical constituents as it relates to the engineering properties of the clay brick product has been established.

REFERENCES

1. Chima, O. M., Nwoye, C. I and Nnuka, E. E. (2017) Enhancement of Refractory Properties of Blended Clay with Groundnut Shell and Rice Husk AdditiveAmerican Journal of Engineering Research. (AJER). 6(6). Pp 218 -226..

2. Kitouni, S. and Harabi, A. (2011). Sintering and Mechanical Properties of Porcelains Prepared from Algerian Raw Materials .Ceramica Vol 57 No 344. Pp 1-13.

3. Evi, D. Y and Pratiwi, I. (201718). Correlation between thermal Behavior of Clays and their Chemical and Mineralogical Composition: A Review. Global Colloquium on Geosciences and Engineering. IOP Conference Series 118 (2018) 012078 doi:10.1088/1755-1315

4. Grimshaw, R.W. (1971) The Chemistry and Physics of Clays and Allied Ceramic Materials. Ernest Benn Ltd., London, UK. 685

5. Altayework, T. B. (2013). Effect of Firing Temperature on Some Physical Properties of Burnt Clay Bricks Produced Around Addis Ababa. Master Degree Thesis. Department of Civil Engineering. Addis Ababa University, Etiopia.

6. Mazen, N and Amaireh, A (2009). Production of Fire-Clay Refractory Bricks from Local Materials. European Journal of Scientific Research. 26(3). Pp 386-392.

7. Gulgun, Y., (2011). The Effect of Temperature on the Characteristics of Kaolinite and Bentonite. Scientific Research and Essays. 6(9). Pp 1928-1939.

8.Amkpa, J. A., Badarulzaman, N. A. and Aramjat, A. B. (2016). Influence of Sintering Temperatures on Physico-Mechanical Properties and Microstructure of Refractory Fireclay Bricks. International Journal of Engineering and Technology. 8(6). Pp 2588 – 2593

9. Folorunso, D. O., Aramide, F. O., Olubambi, P., Borode, J. O. (2015). The Effect of Firing Temperatures on the Performance of Insulating Firebricks Containing Different Proportions of Alumina and Sawdust. Journal of Minerals and Materials Characterization and Engineering. Vol 3. Pp 309-317.

10. Aramide, F. O. (2012). Effect of Firing Temperature on Mechanical Properties of Fired Mansonry Bricks Produced from Ipetumodu Clay. Leonardo Journal of Science. 21. Pp 70 -82.

11. Sedat, K., Sabit, E. and Hikmet, G. (2006). Firing Temperature and Firing Time Influence on Mechanical and Physical Properties of Clay Bricks. Journal of Scientific and Industrial Research. Vol 65. Pp 153-159

12. Johari, I., Said, S, Hisham, B, Bakar, A and Ahmad, Z. A (2010). Effect of the Change of Firing Temperature on Microstructure and Physical Properties of Clay Bricks from Beruas (Malaysia). Science of Sintering. 42. Pp 245 -254.

13. Omowumi, O. J. (2001). Characterization of some Nigerian clay as refractory material for furnace lining. Nigerian Journal of Engineering Measurement, pp 1-4.

14. Tasic, Z.D. (1993). Improving the Micro Structural and Physical Properties of Alumina Electric Porcelains with Cr2O3, Mno2 and ZnO Additives.

Journal of Material Science. 28(21) Pp 5693-5701.

15. Safer Ahmad, YaseenIgbal and Raz Muhammad (2017). Effects of coal and wheat husk additives on the physical, thermal and mechanical properties of clay brick. Boletin de la SocietadEspannola de Ceramica. 56(3) Pp 131-138.

16. Gilchrist, D. J, Fuel, Furnace and Refractories. (1977).International Series on Materials Science and Technology. Pergaon Press Ltd. Headington Hill Hall, Oxford . OX30BW, England. Pp 237-307.

17. Dondi, M. (2005). The role of Surface Microstructure on Resistance of Strains of Porcelain Stoneware Tiles. Journal of European Ceramic Society. 25(4). Pp 357-365.

18. Khurmi, R. S., and Sedha, R. S. (2011). Materials Science. S. Chand & Company Ltd. Ram Nagar, New Delhi, India. ISBN 81-219-0146-4. Pp 199-201.

19. Nnuka, E.E and Agbo, U.J.E. (2000). Evaluation of the Refractory Characteristics of Otukpo Clay Deposit. NSE Technical Transction. 35(1). Pp 32-40.

20. Chaouki, S., Iz-Eddine, E and Abderrahman, A. (2014). Recent Advances in Silica-alumina Refractory: A Review – Science Direct. Journal of Asian Ceramic Societies. 2(2). Pp 83 – 96.

21. Enas, M. H. A and Mahasin, F. H.A, (2015). Improving the Physical Properties of Iraqi Bauxite Refractory Bricks. Journal of Al-Nahrain University. 18 (3). Pp 67-73.

22. Chester, J. H. (1983). Refractoriness: Production and Properties. The Metal Society, London. Pp 492 – 510.

23. Demir, I. (2008). Effect of Organic Residues Addition on the Technological Properties of Clay Bricks. Waste Manage. 28. Pp 622 – 627