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Refractory properties of snail shell particulate modified termite clay

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ABSTRACT

This research studied the possibility of enhancing the refractory properties of termite-based clay with the addition of particulate snail shells. The clay was sourced from Federal University of Technology, Akure Campus while the snail shells were obtained from farmland in Akure, Ondo State. The prepared sample compositions consist of 100 wt. % clay (control sample) and those with varying proportions of 3-15 wt. % particulate snail shell. Samples produced were air dried for 48 hours. The dried samples were oven dried at a temperature of 110˚C for 2 hours and finally fired in a furnace at a temperature of 900˚C for 2 hours to increase strength for handling. The fired samples were subjected to physical and mechanical tests to determine their refractory properties. Representative samples were characterized via the stereo microscope in order to ascertain the amount and sizes of pores in the ceramic matrix composite after firing and also were characterized via ED-XRF in order to ascertain the relative abundance of the elemental constituents of the clay. Results obtained showed that termite clay admixed with 9 - 12 wt. % particulate snail shells exhibited the desired properties of a good refractory material suitable for low duty work.

Keywords: Termite Clay, Snail Shell, Composites, Refractory Properties, Fired Clay.

1. INTRODUCTION

Exploration of very high-temperature environments has become necessary due to the development of high-performance materials, systems, and processes. Ceramic refractory bricks are increasingly being used in high-combustion applications because of their superior pyrolytic, chemical, and thermomechanical qualities. Ceramic materials with high fusion points fall under the category of refractories. Refractory materials are those that are difficult to fuse and are mostly made from ceramics. Almost all ceramics melt at temperatures above 2000°C and when heated to high temperatures, they have the capacity to maintain their physical form and chemical composition. They are beneficial at high temperatures due to this feature (Iliya and

Oluwole, 2023).

A large number of refractory materials are made from naturally occurring high-melting-point oxides or compounds, like dolomite, chromite-magnesite, zirconia, aluminosilicate, carbon, and magnesia. Fireclay, a type of aluminosilicate refractory material, is one of the preferred refractory materials due to its availability (Samad et al., 2021). Clays are hydrated silico-aluminous minerals with a stacking of two different types of layers that contain silicon in tetrahedral coordination and aluminum in an octahedral environment, respectively. Clays can form with water, colloidal suspensions, and plastic pastes due to their large specific surface, plate-like structure, and physicochemical surface (Iliya and Oluwole, 2023; Boch and Niepce, 2007).

According to Shuaib-Babata and Mudiare, (2017) it is a blessing for a nation's economic stability to hinge on the availability of clay deposits as the primary source of refractories in the region. Clay deposits in Nigeria occupy an estimated 1 billion tons of proven reserves which are available in the nation's states. Previous studies found that, when subjected to refractory tests, the action of termite clay-based materials was in strong contrast with the firebricks obtained (Legese et al., 2021; Eze et al., 2020).

It was therefore, proposed that further improvement can be obtained by adding additives such as alumina cement, snail shell and coconut. Previous research on refractory has shown that for most applications, no single clay material has the capacity to provide all of those properties needed; hence, deficiencies in the properties of clay products limit their industrial applications. This has also resulted in the use of mixtures of various clays and certain local agro-waste materials as additives to clay content to enhance the desired properties of refractory materials (Utami et al., 2021; Chima et al., 2017).

Snail shells, which are agro waste, are usually been discarded around the country as a by-product of domestic households, restaurants, and eateries after the snail flesh is separated from the shells. According to some authorities, major environmental and ecological issues have been posed by the accumulation of poorly handled wastes and have resulted in the over-swelling of dumping sites with discarded snail shells, leading to environmental hygiene challenges and also causing many degrees of injuries to humans and animals in the environment (Nwakonobi et al., 2015).

While several works have been carried out in the field of refractory materials for more than two centuries, resulting in the variety of refractory materials available on the world market today, most developing nations still have to invest their hard-earned foreign reserves in refractory materials. In the light of this situation, there has been an ongoing upsurge of interest in the imperative of looking inward to build good refractories using local materials. Therefore, these bio-shell wastes need to be recycled for processing into green engineering products, such as adding to clay to improve refractoriness for sustainable economic benefit (Kolawole et al., 2017). It was on this basis that this work was planned to purify the termite clay that is readily available in Nigeria and incorporate pulverized snail shells to improve the refractory properties.

2. MATERIALS AND METHODS

The materials used for this research were Plaster of Paris, thread, termite hill clay from Federal University of Technology Akure and snail shells from farmland in Akure. The equipment and machines were, Denver Pulverizing machine, Denver ball mill, Sieve shaker and set of sieves, Electronic Weighing balance, Compression machine, X-ray fluorescence (XRF), Muffle furnace, Modified Lee disk, Hydraulic compression testing machine, Laboratory oven, kiln, conical flask, retort stand, hotplate, Pyrometer, Stereo-microscope.

Sample Preparation

The termite hill clay was soaked in water for 3 days to dissolve the clay. The softened clay was stirred to allow deleterious substance to float, and these substances were decanted to form slurry in the process. The slurry was sieved to extract the clayey content using a sieve and the impurities present were discarded. The slurry was allowed to settle and the clear water gotten which was discarded leaving the filtrate. The filtrate was then dried in open air within 5 days. This treatment was necessary to remove dead organic matters. The dried clay was then oven dried, crushed with pulverizing machine and grounded into powder using the ball mill. The ground clay was sieved to pass through sieve 300 μm apertures using the sieve shaker. For the snail shell, it was washed with water, sun dried within 3 days and oven dried before crushing and grounded to powder and, sieved to 300 μm.

The performances of the developed samples were dependent on thorough mixing of the constituents. The termite clay and varying proportions of snail shell; 3, 6, 9, 12, and 15 wt. % were used to make samples in various mixing ratios. The samples were constituted by measuring the components of the samples to specifications using a laboratory weighing balance. To ensure thorough mixing, each composition was initially stirred in the dry condition for 5 minutes. Water was gradually added to the dry powder to make the combination mouldable until there was no dry powder remaining. The plastic paste was then shaped into cylindrical specimens with a height of 50 x diameter of 50 mm, as specified by the American Foundrymen Society (AFS). The samples compositions are as in (Table 1).

Table 1 Compositions of the Samples Prepared

Sample	A	B	C	D	E	F
Clay (wt. %)	100	97	94	91	88	85
Snail shell (wt. %)	0	3	6	9	12	15
Water (wt. %)	10	10	10	10	10	10

The cylindrical bricks were dried for 48 hours in the workshop environment to remove moisture and boost their green strength, making them safe to handle. The dried samples were then dried in an oven at 110°C for a further 2 hours before being allowed to cool in the oven overnight. Water and volatile components were removed from the produced samples by drying them in a furnace at 900°C using a predetermined temperature program (i.e., heating rate of ~100°C/h, dwelling time of 2h and cooling rate of ~70°C/h). The samples were allowed to cool for 24 hours in the furnace to convert them to a permanent product with acceptable strength, durability, and appearance in agreement with previous researchers (Adamu et al., 2018; Folorunso and Bello, 2021)

Test and Characterization

Analyses of the clay were carried out using XRF according to the standard procedure (Folorunso and Bello, 2021).

Porosity and bulk density

The specimens' weights, heights, and diameters were all measured. The bulk density was then estimated using the relationship, which was in agreement with ASTM 293-14a, (2014) and in accordance with Iliya and Oluwole, (2023). The apparent porosities of the samples were determined using ASTM 293-14a, (2014).

Linear shrinkage

The ASTM C67-19, (2019) standard was followed when conducting linear shrinkage tests. Before firing, the heights of the firebrick test samples were measured with Vernier calipers. In each case, three different linear measurements were obtained and the average value was calculated. The samples were then fired at 900°C at a heating rate of ~100°C/h and soaked for 2 hours, then allowed to cool to room temperature and the heights measured again.

Thermal Shock Resistance and Thermal Conductivity

Thermal shock resistance tests were performed in accordance with the ASTM C133-84, (1984) standard. For homogeneity, the prepared specimens were placed in a furnace and heated to a temperature of 900°C. This temperature was maintained for 30 minutes. The specimens were retrieved from the furnace one by one with a pair of tongs and allowed to cool in the air for 10 minutes before being examined for fractures. When no cracks (fractures) were found, the specimens were returned to the furnace and reheated for another 30 minutes before being cooled for 10 minutes again. The heating and cooling cycles were repeated until surface fractures appeared. Thermal shock resistance is defined as the number of cycles required to generate cracks.

The thermal conductivity is evaluated by cutting a test specimen from the parent samples. Each specimen was held in place by two metal discs provided by the apparatus. Each specimen's heat capacity was determined. The thermal conductivity device was turned on, and a specific temperature of 80°C was established for one portion of the metal disc, and temperature variations were monitored and recorded, as well as the time interval for temperature changes in the other section of the metal disk. The thermal conductivity was carried out using Lee's disk apparatus, and the thermal conductivity was calculated as follows and it was in accordance with Folorunso and Bello, (2021) using Equation 1.1.

$$K=\frac{m\,Cp(\theta 1-\theta 2)4x}{\pi D2(T_1-T_2)t} \quad \text{.......} \quad (1.1)$$

Where; M is mass of the disk, 0.0078kg,

C_P is specific heat capacity of the disk, 910 kJ/kgK,

θ_1, θ_2 are initial and final temperature of disk B, respectively,

D is diameter of the sample (mm),

T_1 and T_2 are temperatures of disk A and disk B (K), respectively,

x is thickness of the sample

t is final time taken to reach a steady temperature (seconds).

Cold Crushing Strength

The samples for the cold crushing strength were simply allowed to dry in air and tested for cold crushing strength, using hydraulic strength testing machine where load was applied axially to the piece until cracks was noticed. The load at which the specimen cracked was noted as the cold crushing strength of the test specimen. Similar method was also done by Iliya and Oluwole, (2023), Shuaib-Babata and Mudiare, (2017) and Folorunso and Bello, (2021).

Refractoriness

The refractoriness of the samples was determined in a kiln using pyrometric cones of size 4 (1209°C) and size 5 (1221°C). The samples were prepared in conical shape in order to determine their refractoriness. They were then dried and taken into the kiln alongside the pyrometric cones. The composite was fired gradually to prevent the residual chemically bonded water from becoming steam inside the clay body, which may cause the clay to burst (Folorunso and Bello, 2021). Temperature of the kiln was measured with the use of pyrometer through a tiny hole created every hour before all openings were sealed off with clay.

Microstructural Characterization

Analysis of the samples containing no additives and those containing 3 and 6 wt. % snail shell were carried out using laboratory stereo- microscope model SMDM-1030. The samples were smoothed out on the surface before being placed on the holder and images were taken.

3. RESULTS

The results obtained in this research is in Table 2; showing the chemical composition of termite Clay used as matrix.

Table 2 XRF of the termite hill clays

Element	Content (%)	Fireclay*
SiO_2	61.40	55-75
Al_2O_3	19.10	25-45
Fe_2O_3	1.86	0.5-2.0
CaO	1.08	--
K_2O	1.24	<2.0
MgO	0.85	<2.0
Na_2O	1.11	--
TiO_2	1.13	--
MnO	0.95	--
LOI	11.28	12-15
Total	100.00	

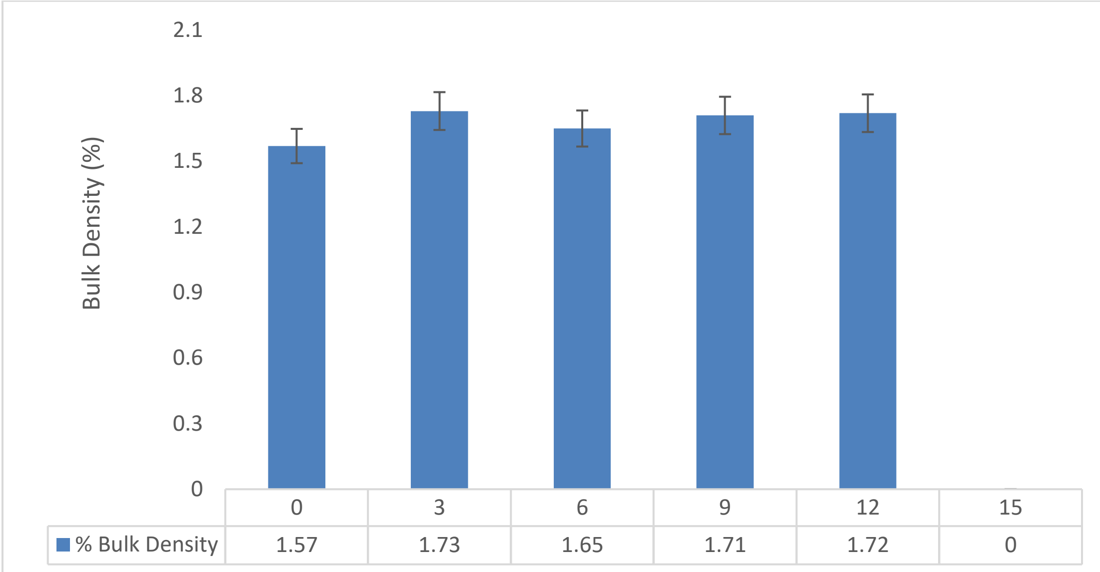

Figure 1 Variation of bulk density with % snail shell

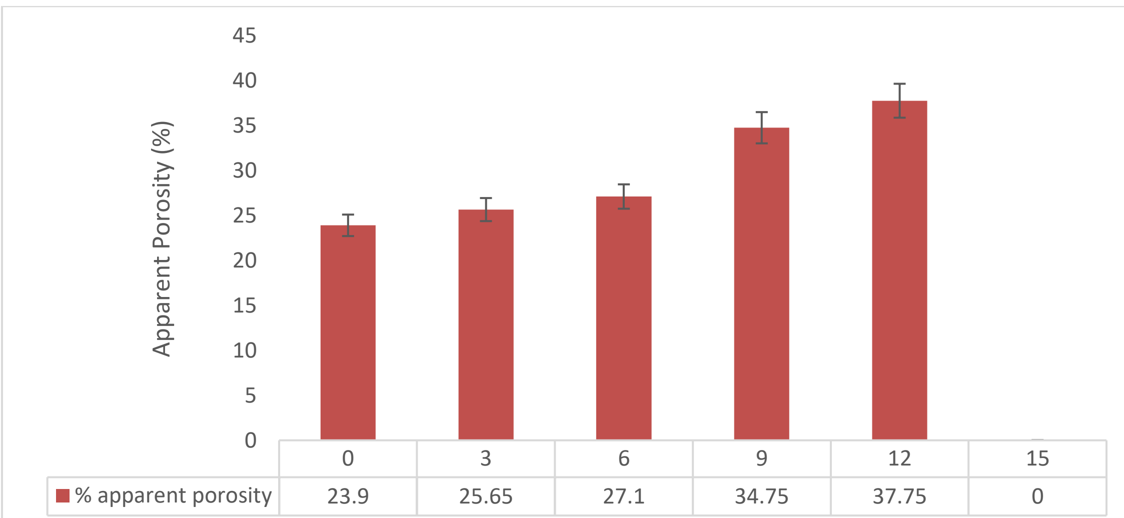

Figure 2 Variation of apparent porosity with % snail shell

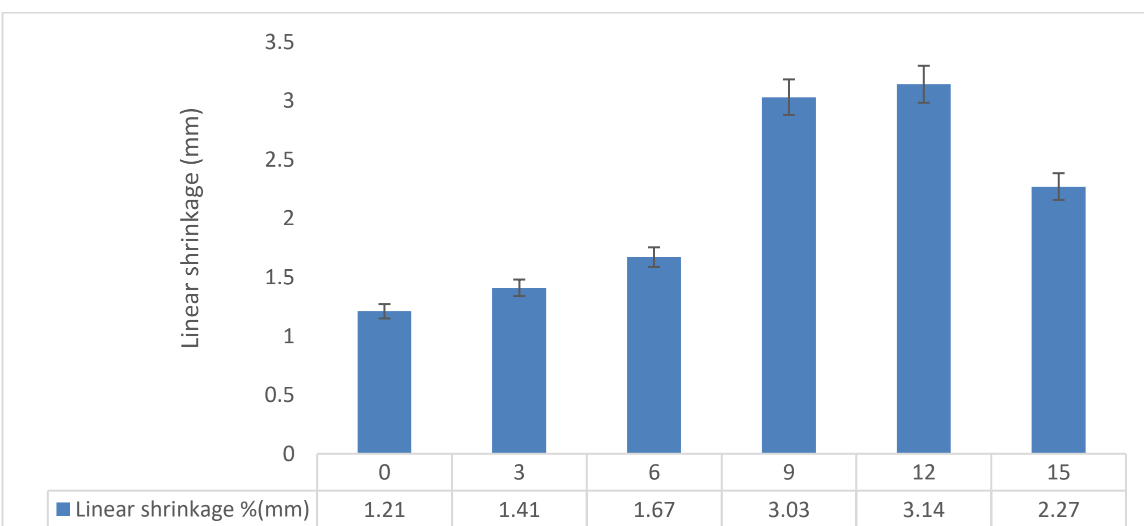

Figure 3 Variation of linear shrinkage with % snail shell content

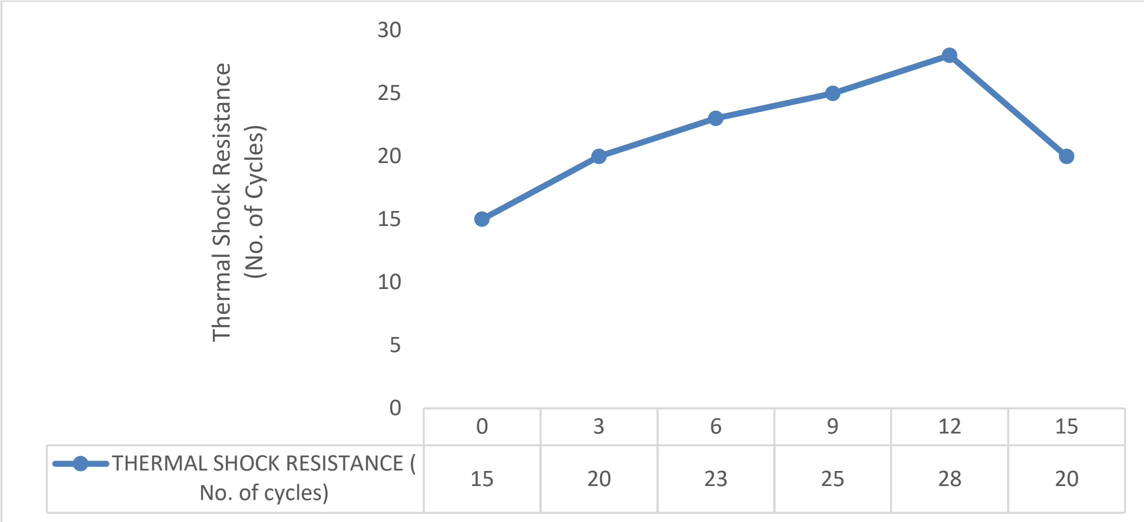

Figure 4 Variation of Thermal Shock (TSR) with % Snail Shell Content

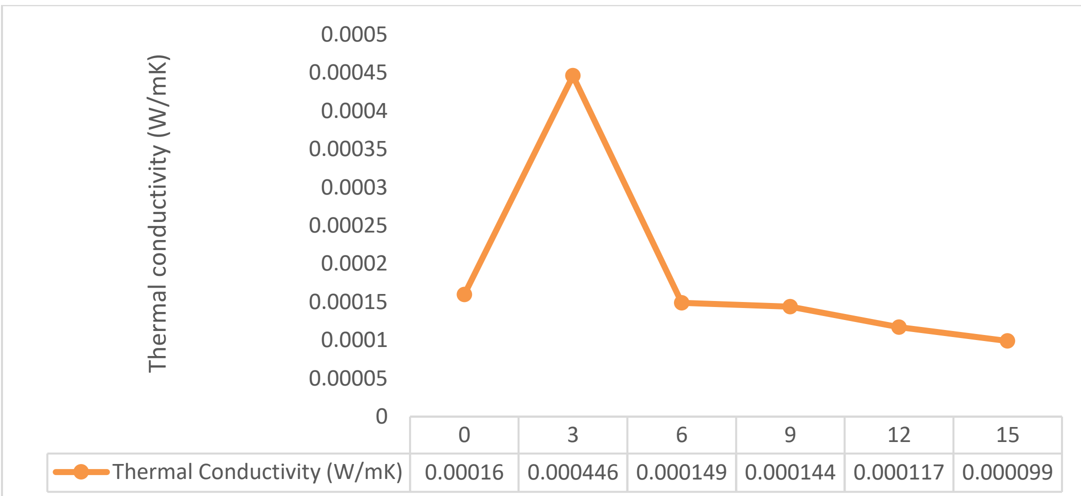

Figure 5 Variation of thermal conductivity with % Snail Shell Content

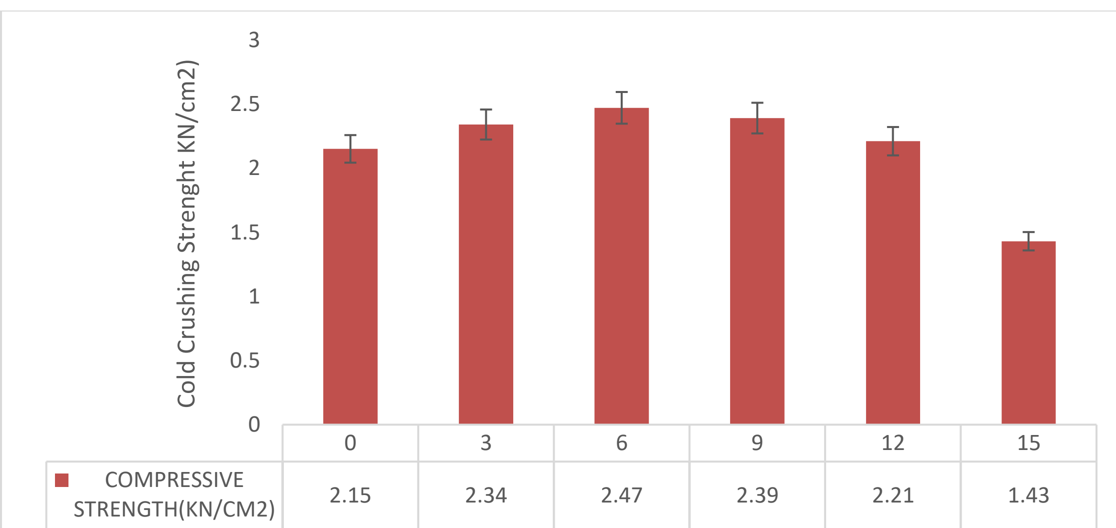

Figure 6 Variation of cold crushing strength with % snail shell content

Plate 1 Pyrometric Cones after Firing

Plate 2 Clay Samples after firing (0-15 wt. % snail shell)

Plate 3 Clay Sample with 15wt. % of Snail Shell

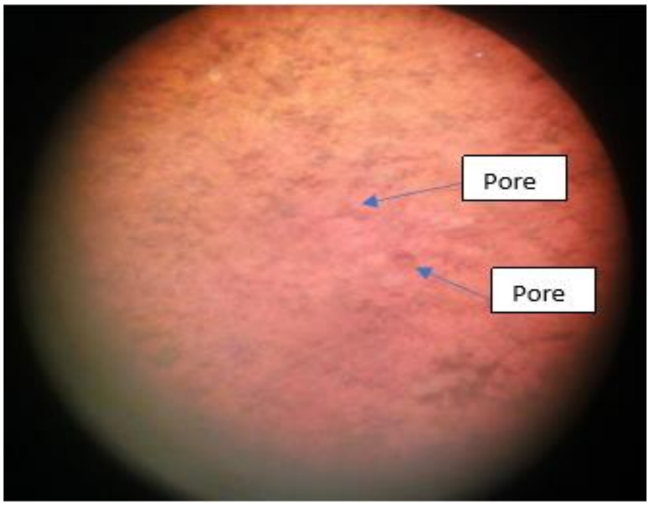

Plate 4 Brick without Additives (snail shell) fired at 900°C

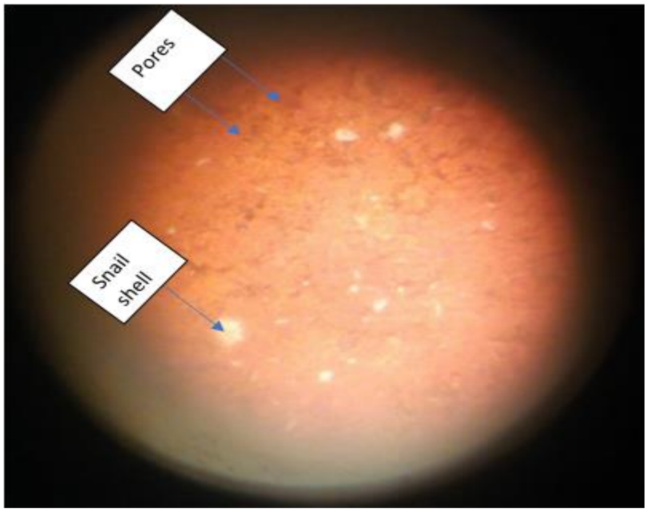

Plate 5 Brick with 3wt. % of Snail Shell fired at 900°C

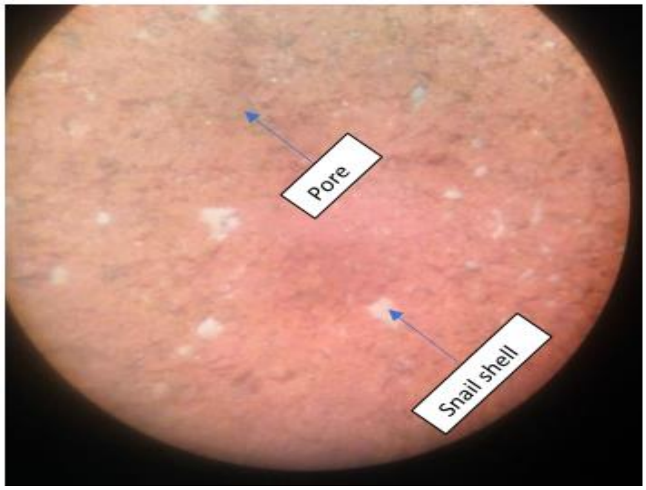

Plate 6 Brick with 6wt. % of Snail Shell Fired at 900^{o}C

4. DISCUSSION

Clay Characterization

An XRF result of the termite clay was as in (Table 2). As per ASTM C27-98, (2018), aluminum and silicon compounds were not within the standard range for the classification of fired clay bricks. The small amount of iron-bearing compound (1.86%) is desirable as it does not support thermal conductivity in clay (Folorunso and Bello, 2021) and enhances the thermal insulation capacity of clay, as the quality of clay for high-temperature applications is measured in terms of its low iron content. The thermal, physical, and mechanical properties of fired clay bricks were not significantly affected by other elements present in small quantities (Folorunso and Bello, 2021; Folorunso, 2015).

Bulk Density and Porosity

The results of the bulk densities of processed termite hill clay with the addition of different proportions of snail shell are contained in (Figure 1). As expected of any good refractory lining material, the bulk density should range between 1.6 and 2.0 gdm^{3} (Folorunso and Bello, 2021; Chester, 1973). This range of values is clearly exhibited by the clay at almost all the percentages of snail shell addition. Samples with 3, 6, 9, and 12% snail shell addition fall within the range because fine particles of clay promote the sintering kinetics, promoting the particles cohesion (Folorunso and Bello, 2021), while samples with 0% were slightly closer to the range, and for 15% snail shell addition, the sample crumbled during the test. Furthermore, the variation in the bulk densities might be due to void formation resulting from the burning off of organic matters (snail shell) from the bricks during firing, this organic matter is usually not uniformly distributed, hence the variation occurs. Also, the forming pressure, particle size can also be a factor (Folorunso and Bello, 2021; Folorunso, 2015; Folorunso and Akinwande, 2021).

The results in Figure 2 demonstrate that the samples exhibited different apparent porosity values depending on the amount of additives. It has been discovered that sample porosity rises as the proportion of additives or reinforcement added to the clay increases. This might be explained by the possibility that the additive will burn off at the high firing temperature, leaving pores for the free passage of the generated gases. The porosity increases as the amount of snail shell added increases because more pores are formed (Folorunso and Bello, 2021; Folorunso, 2015).

It was also observed from the result that the highest porosity was composition with 12% snail shell addition, and the lowest porosity of 23.90% with no additives and for 15 wt. % the apparent porosity was 0% because the sample crumbled during the test. The apparent porosity between 0% and 6% of snail shell were within the acceptable range (10-30%) as suggested for refractory bricks while the apparent porosity at 9% and 12% of snail shell were outside the recommended range (Chester, 1973). And for 15% snail shell addition the sample could not be moulded. Similar results were observed by Amaakaven et al., (2019) in a research done on utilization of clay around the banks of river Benue in Makurdi for use as a medium duty refractory material and Mokwa et al., (2019) on characterization and evaluation of selected kaolin clay deposits in Nigeria for furnace lining application.

Linear Shrinkage

A range of 4 - 10% shrinkage was recommended for fireclays (Folorunso and Akinwande, 2021; Amaakaven et al., 2019; Mokwa et al., 2019; Omowumi, 2001; Dansarai et al., 2020), and range of 7-10% was recommended for refractory brick (Chester, 1973; Iyasara et al., 2016). The samples have a comparatively lowest linear shrinkage value of 1.21% and highest value of 3.14% which fall out of the recommended range for both fireclay and refractory brick. However, lower values are more desirable as this means the clay is less susceptible to volume change (Legese et al., 2021; Folorunso and Akinwande, 2021; Ogunsemi et al., 2018; Abolarin et al., 2004). This shows that the termite clay can withstand high temperature without deformation (high dimensional stability).

Therefore, the selected termite clays can be classed based on this range, as refractory clays. It is evident from Figure 3 that the percentage of linear shrinkage increased slightly with an increase in the amount of snail shell addition. The increase in linear shrinkage may be due to particle bonding at high temperatures, resulting in a decrease in inter-particle separation, or it could be due to an increase in organic (combustible) materials that burned off during firing (Obidiegwu et al., 2015; Iloabacie et al., 2020).

Thermal Shock Resistance

The result obtained generally showed that the thermal shock resistance of the materials increases with increasing percentage of additive. The result in Figure 4 indicates that snail shell of 12 wt. percent has up to 28 cycles, which was the highest among the samples tested. Previous study Folorunso, (2015) used the following principle to determine thermal shock resistance: TSR greater than 30 cycles was rated as "outstanding", 25-30 cycles as "good", 20-25 cycles as "fair", 15-20 cycles as "acceptable", 10-15 cycles as "poor", and less than 10 cycles as "extremely poor". As a result, it is concluded that the inclusion of snail shell (between 9% and 12%) will generate a good lining material for refractory works that is acceptable. Samples with 3%, 6%, and 15% snail shell addition are all viable. However, 0% snail shell is unsuitable for the purpose. Also as quoted by Ugwuoke and Amalu, (2017), samples from 3% to 15 % snail shell fall within the acceptable standard of 20-30 cycles, while that of 0 percent snail shell was out of range.

Thermal Conductivity

The test results in Figure 5 revealed that thermal conductivity declines as the percentage of snail shell in termite clay increases, with the exception of sample with 3 wt. %, which shows a progressive increase in thermal conductivity. When a good fire clay brick is burnt, the pore former (volatile matter in both snail shell and clay) burn off, thereby creating pores in the microstructure. These pores contain air, which is a poor conductor of heat, reducing its thermal conductivity and extending its lifespan. As a result, it is believed that the porosity is responsible for the burned bricks decreased thermal conductivity. Because the thermal conductivity of the gas phase that fills the pore is much lower than that of solid materials, porosity reduces the thermal conductivity of ceramic materials. In small air cells, thermal conduction via gaseous convection is almost negligible (Folorunso and Bello, 2021). For refractory fireclay bricks, a lower value of is desired.

Cold Crushing Strength

The essence of undertaking this test is to determine the ability of the samples to withstand compressive stresses in service. Figure 6 shows the observed variation in mechanical strength of clay bonded brick with addition of snail shell in varying percentages. The results indicated that the strength of test specimens depend on the percentage weight (wt. %) of additives. The observed compressive strength indicated that the compressive strength of test specimens fired at 900^{0}C decreased with an increase in the quantity of snail shell addition. The highest compressive strength was in sample containing 3% of snail shell. This trend can be explained by the fact that at higher firing temperatures, the low melting compounds burn off completely, diminishing the cohesiveness of the clay grains and the grains' adhesion to the snail shell. The crushing strength is reduced when the cohesive and adhesive forces are decreased (Folorunso, 2015).

Generally, in clay based ceramic systems, strength decreases with an increase in porosity. The values obtained for the samples were between 1.43 and 2.47KN/cm^2 respectively. These values fall between the standard for fireclay; 1-15 KN/Cm^2 (Oyetunji et al., 2018) which is very satisfactory.

Refractoriness

The refractoriness of the samples was determined using the pyrometric cones 4 and 5. Cone 4 deformed after firing to a temperature of about 1209°C while cone 5 deformed when the temperature rose to 1221°C as in (Plate 1). The highest refractoriness of the termite clay was found out to be 1300^{o}C. Samples with 15% snail shell addition exhibited the lowest refractoriness of about 1200^{o}C before fusing as in (Plate 3); this low refractoriness might have been due to appreciable amount of snail shell (15%) in the clay. In addition, all the clay samples refractoriness did not exceed 1300°C but were above the deformed temperature of cone 4 and cone 5.

These show poor refractoriness, this low value of refractoriness is as a result of the high silica content of the clay and low alumina content according to the mineralogical characterization. Fireclay should have refractoriness in the range of 1500°C- 1700°C as reported by Shuaib-Babata and Mudiare, (2017). Similar result was also observed in Abubakar and Okoye, (2016) and Irabor and Okunkpolor, (2020). It is therefore concluded that the material can be used for refractory applications not exceeding temperature of about 1300^{o}C.

Microstructural Examination

The stereo microscope image in Plate 4 shows a more uniform structure in the termite clay than in the structure containing snail shell (3wt. % and 6wt. %), which had some aggregation of composite material with larger pore diameters (Plate 5, 6). White and black spots on the plates represent the snail shell and the pores, respectively. Increased pore diameters and increased amount of pores result in higher apparent porosity, as seen in the results. This phase is responsible for refractoriness as well as other thermal and mechanical properties (Chima et al., 2017)

5. CONCLUSION

Based on the results obtained in this research, the refractory properties of the clay such as refractoriness, compressive strength, thermal conductivity, thermal shock resistance and bulk density were effectively improved by the addition of 9 – 12 wt. % particulate snail shell to the termite clay. It was concluded that;

The porosity increased with increasing amount of snail shell up to 12%. The increase in the porosity was as a result of voids created during the burning of volatile content on firing. However, the apparent porosity between 0% - 6% of snail shell were within the acceptable range (10-30%) as suggested for refractory clay.

The bulk densities of the samples increased with the amount of snail shell but showed a little variation between 3 wt. % - 12 wt. %. However, they were in the range as expected of any good refractory lining material. The bulk density should range between 1.6 and 2.0 gdm^{3}.

Thermal shock resistance results showed significance increase in the thermal shock resistance as the addition of snail shell increases up to 12 wt.% while the thermal conductivities of the samples reduce with the exception of 3 wt.% which showed a progressive increase in the thermal conductivity. For a good fireclay lower value of thermal conductivity is needed thereby prolonging the lifespan of the firebricks in service.

The cold crushing strength increased up to 6% and slightly decreased with increasing amount of snail shell addition from 9% - 15%, this decrease in strength was as a result of increasing porosity. However, the compressive strength lies within permissible limit for refractory specification according to which is very satisfactory.

The refractoriness of the termite clay brick found out to be 1300^{o}C except for 15% which was lower than 1300^{o}C. Hence, it can be used as refractory lining for non-ferrous extraction whose melting point is below 1300^{o}C.

Hence, the clay can be used as refractory material in furnace lining. It can find extensive application in lining of soaking pits and reheating furnaces, safety lining of steel ladles and kilns in cement industry due to its refractory properties.

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Author's Contribution

Davies Oludayo Folorunso: Review of manuscript and approval of the version to be published.

Isiaka Oluwole Oladele: Review of manuscript and approval of the version to be published.

Samuel Olumide Falana: Acquisition and analyzing of data, drafting the Manuscript

Gabriel Omotuyi Mogbojuri: Substantial contribution to acquisition of data and analyzing of data

Linus Nnabuike Onuh: Substantial contribution to drafting the manuscript

Ethical issues

Not applicable.

Informed consent

Not applicable.

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Conflict of Interest
The author declares that there are no conflicts of interests.

Data and materials availability
All data associated with this study are present in the paper.

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