American Journal of Engineering Research (AJER)
e-ISSN: 2320-0847 p-ISSN : 2320-0936
Volume-4, Issue-5, pp-08-15
www.ajer.org
Research Paper Open Access

The Purification and Adaptation of Termite Hill Clay for Furnace Lining by Graphite and Rice Husk Addition

D.O. Folorunso

Department of Metallurgical and Materials Engineering, Federal University of Technology, Akure, Nigeria
Africa Materials Science and Engineering Network (AMSEN): A Carnegie-IAS (RISE) Network.

ABSTRACT: *This work has studied the possibility of purifying termite hill clay hydrometallurgically and subsequently adapting it for furnace lining by the addition of graphite and rice husk granules. A large quantity of termite hill clay was mined from a location on the campus of The Federal University of Technology, Akure (FUTA), Nigeria. The slurry obtained from the clay after washing in water was sundried for three days and again dried in the furnace at 90 °C for 8 hours. Representative samples were characterized via the X-ray diffraction (XRD) and scanning electron microscopy (SEM) in order to ascertain the relative abundance of the constituents of the clay. Subsequently, a substantial quantity of the clay was purified by leaching at a predetermined condition of; 1.6 mol/dm*^{3} *of oxalic acid at 90 °C for 150 min. and 200 rev/min agitations. Cylindrical samples (5cm diameter by 5cm high) containing 5 % - 30 % of graphite and 1% - 5 % rice husk granules were prepared, dried at 110 °C and subsequently fired at 900°C, 1100°C, 1300°C and 1500 °C, at the rate of 4 °C/min and soaked for 2 hrs. These samples were subjected to different refractory tests (bulk density, cold crushing strength, permanent linear change, thermal shock resistance, refractoriness under load and apparent porosity). Results obtained showed that purified termite hill clay admixed with 15 % graphite and 3 % rice husk exhibited the desired properties of a good refractory material suitable for lining a rotary furnace.*

Keywords: *Hydrometallurgy, Characterized, Constituents, Predetermined condition, Refractory tests*

I. INTRODUCTION

The refractory lining is a protective layer installed inside the kiln or furnace to insulate the furnace steel structure from high temperatures. It protects it from chemical attack, thermal shocks and abrasion wears. The lining may be monolithic refractories such as castable, gunning mixes or refractory bricks. The refractory selection depends upon the temperature inside the unit and the chemical nature of the material being processed. The thickness of the lining is generally in the range 80 to 300mm [1]. Furnace lining materials are chosen purposely for their ability to survive protracted exposure to extreme temperatures. Other desirable characteristics include resistance to abrasion, mechanical shock and chemical reactions within the molten metal. The most commonly used furnace lining materials are ceramic compounds and metal/ceramic combinations. Ceramic lining materials are made from a variety of raw materials each with its own particular strengths [2]. Common ceramic furnace lining materials include aluminum oxide, magnesite, silicon carbide, and dolomite. Refractories are chosen according to the conditions they will face [3,4]. Some applications require special refractory materials. Zirconia is used when the material must withstand extremely high temperatures. Silicon carbide and graphite are two other refractory materials used in some very severe temperature conditions, but they cannot be used in contact with oxygen, as they will oxidize and burn [3]. Nigeria has appreciable distribution of industries engaged in metal processing operations but lack adequate raw materials to support their growth [4]. In the light of this, there has been a continuous upsurge of interest in the area of the development of good refractories from vast deposits of clay spread across every region in Nigeria in the last two decades. However, some local clay deposits have been investigated with good results in Benue [5] and Ekiti States to meet local needs [6,7]. It is also on record that clays from chanchanga, Bida, Suleja and Zungeru all in Niger State, have better refractory and physical properties compared with imported ones [8]. Rice husk is an agricultural waste that accounts for 20% of the 649.7 million tons of rice produced each year worldwide [9]. The produced partially burnt husk from the milling plants when used as a fuel also contributes to pollution and efforts are being made to overcome this environmental challenge by applying the material as a supplementary cementing material [10]. The chemical composition of rice husk is found to vary from one sample to another due to the differences in the type of paddy, crop year, climate and geographical conditions [11].

The clay from the termite mound is capable of maintaining a permanent shape after moulding because of its plasticity. It is also less prone to crack when compared with ordinary clay. In addition, it has low thermal conductivity and expectedly reduced solar heat flow and temperature fluctuation within an enclosure [12]
However, most of these previous works did not venture into the use of termite hill clay and the removal of the constituent of the clay (iron oxides) that is responsible for lowering the refractoriness. It was on this background that this work was designed to first remove the iron oxide content by hydrometallurgical purification and again enhance the refractory properties of the purified clay by the addition of graphite and rice husk.

II. MATERIALS AND METHOD

The materials and equipment used were raw termite hill clay, oxalic acid (99.8% purity), graphite powder, pulverized rice husk, atomic absorption spectrometer (AAS) machine (model Spectre AA 220 FS), X-ray fluorescence (XRF) machine (model ARL 8410), scanning electron microscope (SEM) model JEOL 840 and coupled with an EDS analyzer, X-ray diffraction (XRD) machine (model Philips PW 3710 with PW 1752 graphite monocromator), sieve size analyser (Microtrac FLEX 10.5.4), plaster of Paris (P.O.P.) mould, Rawwley Sussex jaw crusher and grinder, Labcon shaking incubator (models 3081U and 5082U), Carbolite furnace and compression testing machine (model Pat 2001)

2.1 Raw clay preparations for analysis

A large quantity of termite hill clay was mined from FUTA campus in Nigeria. It was then washed in water in order to remove the deleterious particles by decantation. Water was then drained from the clay slurry using a P.O.P. mould. The recovered clay was then dried in the sun for three days and again in the Carbolite furnace at 90 °C for 8 hours. The dried clay was finally jaw crushed, ground in a Rawwley Sussex grinder and sieved to 100 µm, being the average sieve size upon the size analysis.

2.2 Clay Characterization

Analyses of the clay were carried out using SEM/EDS, XRD, and XRF according to the standard procedure [13]. The results are presented in Fig. 1, 2 and Table 1 respectively.

2.3 Hydrometallurgical Purification of Termite Hill Clay

The clay was treated hydrometallurgically by putting a large quantity of clay and 500 % by volume of 1.6 mol/dm^3 of prepared oxalic acid solution in a flat-bottom glass container and kept inside the Labcon shaking incubator, models 3081U and 5082U for agitation at 200 rev/min at 90 °C for 150 min [14]. On the expiration of the set time, the container was removed and allowed to cool to room temperature. The filtrate was decanted and the residue washed several times with deionized water, until all the acid content was completely expelled. This was achieved by testing intermittently with a blue litmus paper, until no further change in colour was noticed. The clay residue was then dried in the Carbolite furnace at 90 °C, crushed, ground and sieved to 100µm for subsequent tests.

2.4 Preparation of Samples for Performance Evaluation

Measures (150g each) of the purified clay with varying quantities (5 - 30%) of graphite were formed into cylindrical (50mm diameter x 50 mm high) specimens after mixing with about (10-15)% deionized water. The samples were dried in air for 24 hrs and later in the oven at 110 ºC for 48 hrs. Some selected samples, after drying at 110 ºC were again taken for firing in the furnace at 900 ºC, 1100 ºC, 1300 ºC and 1500 ºC. Samples for porosity test were admixed with rice husk in the varying quantities of 1 – 5%. The fired samples were subsequently tested in accordance to the American Standard for Testing and Materials [15] for the following properties:

2.4.1 Bulk Densities and Cold crushing strengths

Test specimens fired at varying temperatures of 900 ºC, 1100 ºC, 1300 ºC and 1500 ºC were produced. The masses, heights and diameters of the specimens were measured. The bulk densities of these specimens were computed and results are presented in Fig. 3. To determine the cold crushing strength, the specimens were subjected to compressive load until fracture, using an Auto Compression Testing Machine model Pat 2001. The results are presented in Fig. 4.

2.4.2 Permanent Linear Change (PLC)

Permanent linear change tests were carried out in accordance with the [15] standard. The heights of the firebrick test samples were measured with vernier callipers before firing at various temperatures. Three different linear measurements were taken in each case and the average value calculated. The test samples were then fired at temperatures of 900 °C, 1100 °C, 1300 °C and 1500 °C for 2 hours at the rate of 4 °C per min, after which they were slowly cooled to the room temperature. The heights of the fired test specimens were again measured in order to determine the changes in heights. The results are as presented in Fig. 5.

2.4.3 Thermal Shock Resistance (TSR)

Thermal shock resistance tests were performed in accordance to the [15] standard. Cylindrical test specimens were heated in a furnace maintained at 1100°C for 30 minutes and then removed to cool in air for 10 minutes. After this, the test specimen was examined for formation of cracks on the surface. The specimens were then returned into the furnace, heated for 10mins, air-cooled again for 10minutes and surface examined for cracks. This cycle of heating and cooling was repeated until surface cracks were observed. The number of cycles required to produce cracks is regarded as the thermal shock resistance. The results are presented in Fig. 6.

2.4.4 Refractoriness Under Load (RUL)

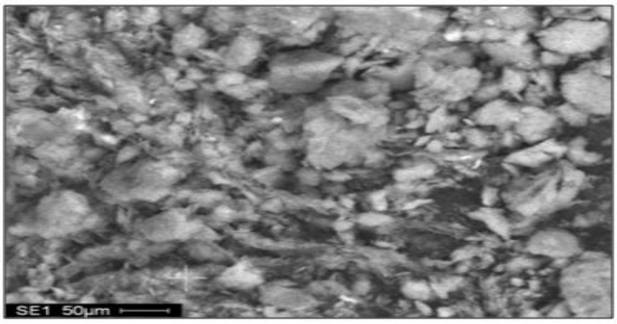
Refractoriness under load test was carried out in accordance to the International Standard Organization and American Standard for Materials and Testing specifications [15]. Samples with dimension 50 mm high by 50 mm diameter were prepared and dried at 110 ºC for 48 hours. They were drilled co-axially with holes of 12.5 mm diameter after cooling to the room temperature. The drilled samples were then set in the RUL furnace one after the other, where the specimens were each subjected to a constant load of 0.2 Nmm^{-2} at increasing temperatures. The heights of the samples were noted at temperature intervals of 100 °C. The change in height was plotted against temperature and the temperature at which the height of each of the samples changed by 0.5 % was taken as the refractoriness of the samples under load. The results are as presented in Fig. 7.

2.4.5 Apparent Porosity

The apparent porosities of the samples were measured using test specimens which had been dried in an oven at 110 °C to a constant weight within 0.1g accuracy and cooled to room temperature. The apparent porosity values were measured in accordance to [15] and the results presented in Fig. 8.

III. RESULTS AND DISCUSSION

3.1 Scanning Electron Microscopy (SEM)



Element	Wt%	At%
C	08.62	14.44
O	44.54	56.00
Mg	01.10	00.91
Al	12.26	09.14
Si	25.00	14.32
K	02.18	01.12
Fe	06.30	04.07
Matrix	Correction	ZAF

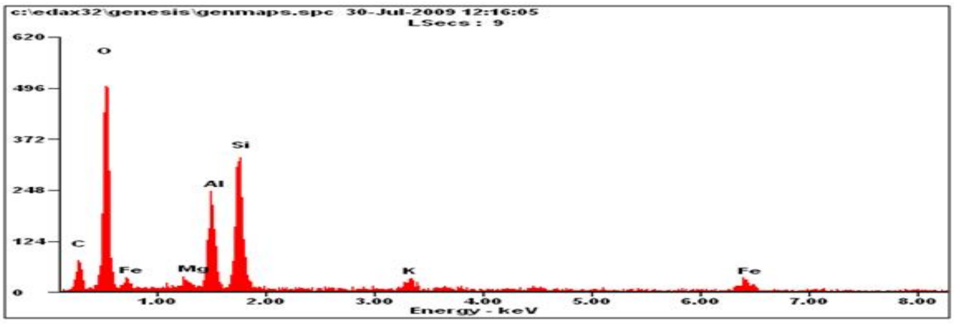


Figure 1: SEM Micrograph (X500) of termite hill clay particle sizes, the EDS pattern and the chemical composition.

The SEM/EDS analysis shows the SEM micrographs of the relative sizes of the clay particles at X 500 magnification and the spectra depicting the peaks of the elements present. The EDS shows the actual percentages of the various elements contained in the clay. The clay contained some quantity of iron, principally in the iron oxide mineral. This and other minerals contained in the clay were quantified in the XRF analysis in Table 1. There is however, a close correlation in the quantities of Fe_2O_3 revealed by both SEM/EDS and the XRF analysis, 6.30 % and 5.97 % respectively.

3.2 X-Ray Diffraction (XRD)

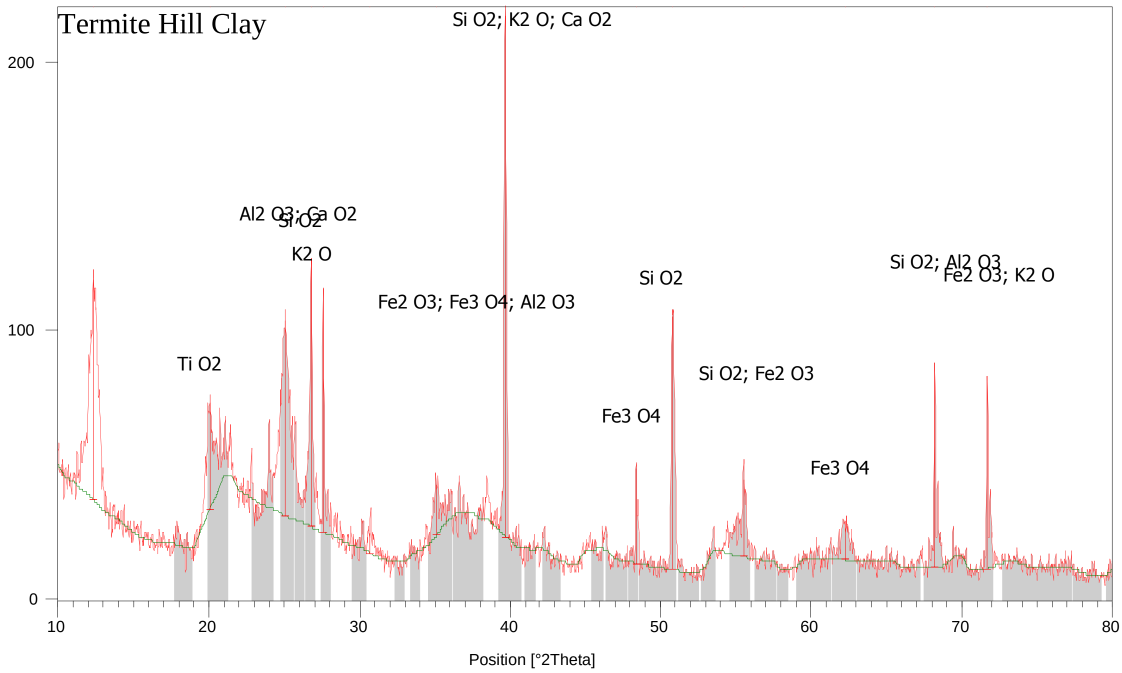


Figure 2: X-ray Diffraction Pattern for Termite Hill Clay

The qualitative analysis of the clay as depicted by the XRD pattern shows sharp high peaks for SiO_2, Al_2O_3, Fe_3O_4 and sharp but shorter peaks for K_2O, MgO, MnO, Na_2O and CaO. Some other oxides were also present but in very negligible proportions.

3.3 XRF of the Raw and Leached Clay Samples

Table 1: XRF of the Raw and purified termite hill clays

Oxides	Raw (Wt. %)	Leached (Wt. %)	Oxides	Raw (Wt. %)	Leached (Wt. %)
Al_2O_3	19.76	24.89	TiO_2	1.11	1.12
SiO_2	62.34	51.84	MnO	0.03	0.03
Fe_2O_3	5.97	2.54	Cr_2O_3	0.02	0.03
CaO	0.63	9.33	P_2O_5	0.03	0.04
K_2O	2.87	1.27	V_2O_5	0.02	0.03
MgO	0.63	0.42	NiO	0.01	0.02
Na_2O	0.85	0.24	ZrO_2	0.05	0.06
LOI(Raw)	6.24		LOI(Leached)		8.13
Total			Total	100.00	99.99

The results of the chemical analyses by X-ray Fluorescence are presented in Table1. The Table shows semi-quantitative analysis of the clay, revealing the percentages of the principal minerals and elements present.
The Major minerals contained in the clay are SiO_2 and Al_2O_3, which constituted more than 80%. The other mineral present in some appreciable quantity was Fe_2O_3 (5.97 %). Present in very small quantities were K_2O, MgO, CaO, Na_2O and MnO minerals. The remaining minerals exist in negligible quantities of less than 1%. Their presence has been found not to constitute any threats to the expected performance of the refractory lining at high temperatures [13,14,1].

It is expected that most of the major minerals will enhance the desirable properties. However, the presence of iron oxide (Fe_2O_3) is detrimental and must be reduced to an appreciable level [13,14]. Therefore, for the clay to perform satisfactorily as a lining material, the iron oxide level was reduced by hydrometallurgical purification from 5.97 % to 2.54 %. Purification was further necessitated by the need to justify the following facts as established by [17].

(a) Percentage sum total of K_2O, Na_2O and MgO was more than the required standard of 2 % [17]. Purification now reduced it from 4.35 to 1.93 %,
(b) CaO content was less than 15 % required for good refractory performance [17]. Purification now increased it from 0.63 to 9.33 %.
(c) Silica (SiO_2) content was higher than about 50 % as required [17]. It has now reduced from 62.34 to 51.84% after purification.
(d) Alumina (Al_2O_3) content was lower than about 30 % required of good lining bricks [17]. It has now increased from 19.76 to 24.89.

a. Bulk Density

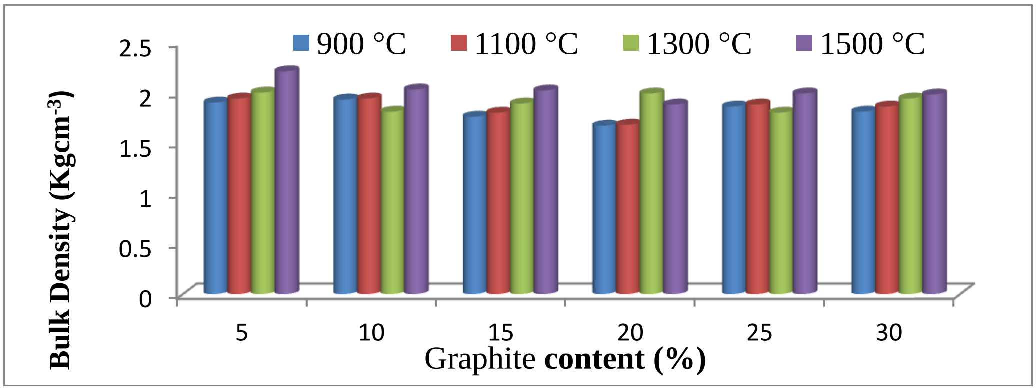


Figure 3: Bulk Densities of Termite hill clay at various graphite contents and firing temperatures

The results of the bulk densities of processed termite hill clay with the addition of different proportions of graphite powder, when fired at different temperatures are contained in Fig. 3. As expected of any good refractory lining material, the bulk density should range between 1.8 and 2.2 gdm^{-3} [17]. This range of values is clearly exhibited by the clay at almost all the percentage graphite addition. Furthermore, the variation in the bulk densities was due to voids formation resulting from the burning off of organic matters from the bricks during firing. These organic matters are usually not uniformly distributed and hence the variation. However, firing at higher temperature could cause voids closure as a result of softening or liquid phase formation which could also cause bulk density variation.

3.5 Cold Crushing Strength

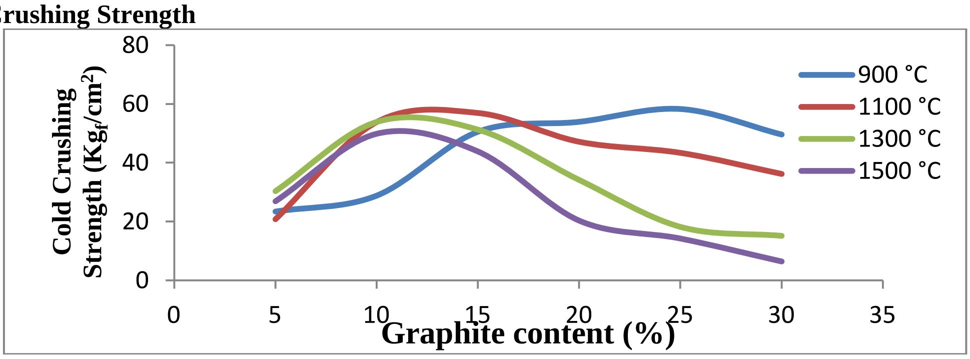


Figure 4: Variation of cold crushing Strength with graphite Contents for purified termite clay fired at different temperatures.

Fig. 4 shows the variation of cold crushing strengths of firebricks containing 5% to 30% graphite and fired at temperatures between 900 °C and 1500 °C. It was observed that at all the firing temperatures, the strengths increased to the maximum at about 10-15 % graphite and then decreased at higher graphite contents. Also, the lower the quantity of graphite, the higher the firing temperature at which the maximum strength was obtained. This trend can be attributed to the fact that the low melting compounds burn off completely at higher firing temperatures thereby reducing the cohesiveness of the clay grains and consequently the adhesion of the grains to graphite as well. The reduction in the cohesive and adhesive forces leads to the reduction in the crushing strength.

The trend was more evident at 1500 ºC and graphite content above 15 % when the crushing strength dropped drastically. Even though the sample fired at 900 ºC exhibited a reasonably high crushing strength, the fact still remains that melting in the furnace is usually at higher firing temperatures. Therefore it is more desirable to consider the behaviour of the samples at higher temperatures and, above all, the graphite content at which the high strength occurred (25 %) was grossly uneconomical for industrial production. It therefore becomes imperative to choose 15 % graphite as the optimum, which is in agreement with [17,18]

3.6 Permanent Linear Change

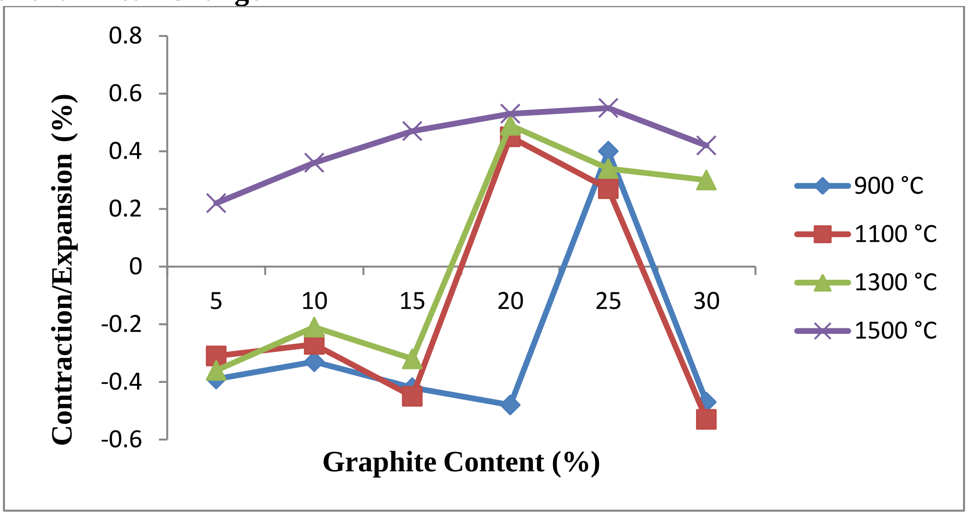


Figure 5: Permanent Linear Changes at different firing Temperatures

The results of the variation of permanent linear changes with graphite contents and firing temperatures are shown in and Fig. 5. It was observed that firebricks produced from the processed termite hill clay underwent linear contractions for firing temperatures of 900 ºC, 1100 ºC and 1300 ºC for low additions of graphite. For graphite contents between 20% and 30% the firebricks suffered linear expansion after which they showed linear contraction again. This is indicative of variations in the clay-graphite reactions at different graphite contents. Firing at 1500 ºC caused the firebricks to undergo linear expansion at all levels of graphite addition without suffering dimensional instability. This therefore, confirms that the bricks so produced will function satisfactorily at a temperature as high as 1500 ºC.

3.7 Thermal Shock Resistance

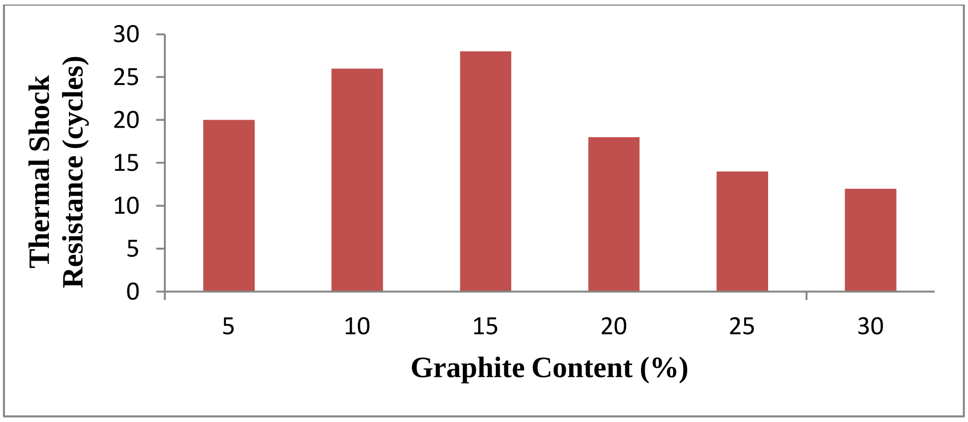


Figure 6: Thermal shock resistance at various graphite contents

Figure 6 shows the thermal shock resistance of the firebricks produced from the processed termite hill clay at different graphite contents. The results represent the outcome of the tests carried out as described in section 2.4.3 above. They revealed that thermal shock resistance increased from 20 cycles at 5% graphite addition to 28 cycles at 15% addition. Thereafter, further graphite additions caused sharp reductions in the thermal shock resistance.

[19] in his previous work adopted the principle below in determining thermal shock resistance: TSR above 30 cycles were classified as "excellent", those in the range 25-30 cycles as "good", 20-25 cycles as "fair", 15-20 cycles as "acceptable", 10-15 cycles as "poor", while less than 10 cycles were classified as "very poor". It is therefore concluded that the addition of; between 10 % and 15 % graphite will produce good lining material for refractory works, 5 % graphite is quite fair while 20 % graphite is acceptable but 25 -30 % graphite is poor for the purpose.

3.8 Refractoriness Under Load

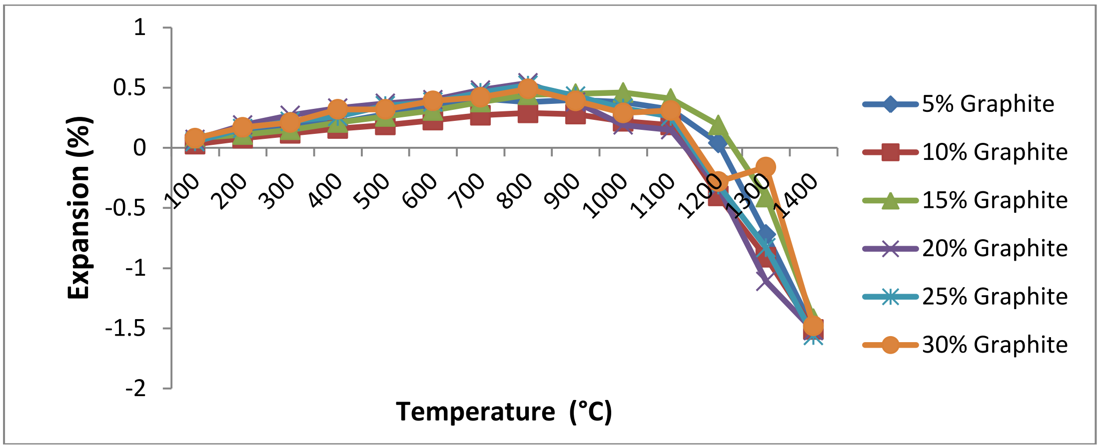


Figure 7: Refractoriness Under Load (RUL) at different graphite contents

Fig. 7 shows the results for the refractoriness under load tests for specimens with 5% – 30 % graphite contents under a constant load of 0.2 N mm^{-2} and increasing temperature up to 1300 ºC, in accordance to [15] standard. The RUL value of about 1190 ºC, which was the temperature at which the height of the sample reduced by 0.5 % under the 0.2 N mm^{-2} load, was obtained for graphite content of 15%, which is the highest and therefore the recommended value for the production of refractory lining firebricks from termite hill clay.

3.9 Apparent Porosity

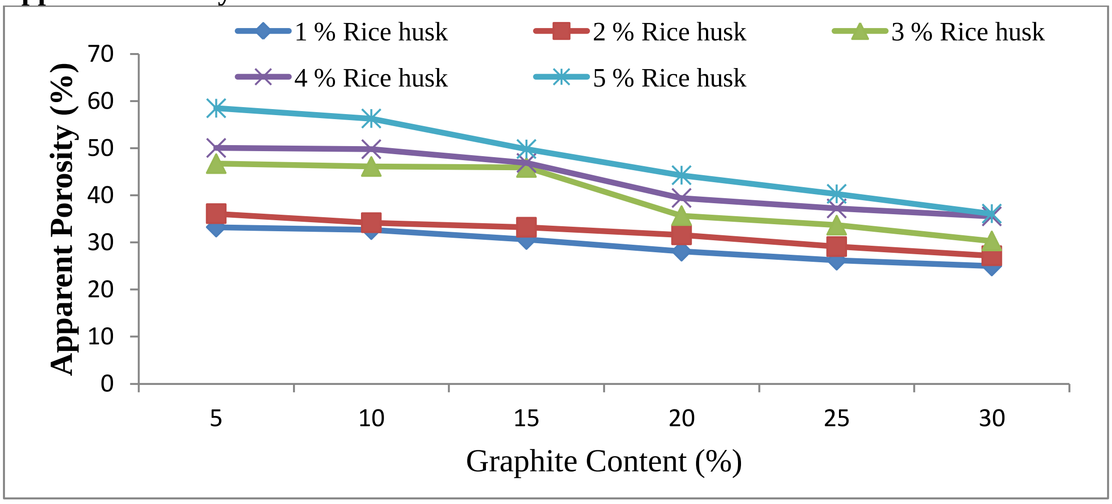


Figure 8: Variation of apparent porosities with graphite and rice husk content

The apparent porosities were enhanced by the addition of rice husk in the range of 1% - 5 %. The results are as presented in Fig. 8. The apparent porosity increased as the percentage rice husk increased at all levels of graphite content because the rice husk burnt off at the elevated temperature of 1500 ºC. The more the rice husk added, the more the pores created. Despite this trend, the porosity still reduced with graphite at each rice husk content. The reduction is attributable to the increased binding effect of graphite in the samples, which binds the grains together, thereby reducing the interparticle distances and hence the reduction in the porosity.

The results obtained show that 3 % rice husk exhibited the desired expectations; ≥ 45 % porosity [20]. Strength, however, is inversely proportional to porosity because the larger and more numerous the pores, the thinner the enclosing wall of solid material and the lower the strength [17,18]. Sample with 15 % graphite and 3 % rice husk was therefore chosen as the most promising of the samples because it possesses apparent porosity of 45.91 %.

IV. CONCLUSION

The research has shown that termite hill clay will serve as a good material for furnace lining if purified hydrometallurgically in order to remove one of its constituents (Fe_2O_3) that is responsible for lowering the refractoriness. The addition of graphite has greatly improved the refractory performance of the termite hill clay, especially when added in the appropriate quantity of 15 %. The porosity of refractory lining material increases with the addition of rice husk. However, the optimum value of porosity is obtained when the proportion of rice husk in the mix is 3 %. The refractory mix, therefore, that contains 82 % of hydrometallurgically purified termite hill clay, 3 % rice husk and 15 % graphite will perform satisfactorily as a good material for lining a rotary furnace.

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