Investigative Study of effects of Sawdust and Grog additives on the refractory Properties of Isiagu Clay

1. Emeka Ojukwu
Department of Chemical Engineering, Nnamdi Azikiwe University. Awka, Nigeria
**ve.ojukwu@unizik.edu.ng*
2. Philomena Igbokwe
Department of Chemical Engineering, Nnamdi Azikiwe University. Awka, Nigeria
** pk.igbokwe@uniik.edu.ng*
3. Ifeanyi Ugonabo
Department of Chemical Engineering, Nnamdi Azikiwe University. Awka, Nigeria
** vi.ugonabo@unizik.edu.ng*

Abstract

This research focused on investigating the effects of sawdust and grog additives on the refractory properties of Isiagu Clay. The raw materials were collected locally, processed and analysed using Scanning Electron Microscope/Energy Dispersive X-ray (SEM/EDX). The additives were incorporated into the raw clay in the ratio of 2.5,5.0,7.5 and 10 wt% and fired at temperatures at 900°C and 1100°C, respectively. The various refractory properties were measured namely; linear shrinkage, apparent porosity and bulk density. The result of the SEM/EDX analyses showed that the clay is a fire clay since it contains 61.68% Al_2O_3 and 34.97% SiO_2 while the grog additive contains 65.5% Al_2O_3 and 34.33% SiO_2, respectively. Also, sawdust contains only carbon metal which is due to a large amount of organic matter in it. SEM/EDX morphological analysis results showed that grog contains tiny to coarse particles which makes it suitable for the refractory application while sawdust contains a sheetlike interwoven structure that makes it suitable for the insulating application. The property measurement showed that linear shrinkage had a different variation for the two additives. For sawdust, linear shrinkage increased from 5.635 to 7.628% at 1100°C while grog decreased from 3.383 to 3.509 % at 900°C. The decrement in grog containing samples is due to the high amount of Al_2O_3 and SiO_2 present in the grog which makes it desirable for refractory application. The apparent porosity test showed that the samples containing sawdust are suitable for insulating application since it contains a high amount of organic matter. Bulk density showed that samples containing sawdust were less than the standard for refractory application, however, grog met the standard and hence can be used for refractory application. ANOVA results confirmed that for grog and sawdust additives, temperature and additive percentages were not significant for linear shrinkage measurement.

Keywords: ANOVA, brick, Clay, fireclay, insulating, mixture, and refractory

INTRODUCTION

Heat transfer is of paramount importance in the process industry. Processes like heating, boiling and roasting of metal ores are classical examples of heat transfer operations[1]. In these processes, heat transfer efficiency is the goal and is achieved in heat transfer equipment like ovens, furnaces and boilers by using refractory materials to line the inside to prevent heat loss to the environment and increase the efficiency of the process [2]. In Nigeria, there are about seven clay deposits that have been proven to have refractory properties and hence good feedstock for refractory brick production[3, 4]. According to Olalere, Yaru [5] and Oke, Talabi [4], the Nigerian government is set to resuscitate her moribund steel industries in Kogi and Delta states which would require 68,000 tons of refractories for their operation and this is wholly apart from the refractory demands of other energy industries totalling 300,000 tons as at the year 2000. Until now about $229 million is expended annually on the importation of refractory materials due to the unavailability of refractory materials for use in the kilns and furnaces [6]. There have been concerted efforts towards meeting the growing need for refractories in the country but the challenge is that refractory materials when utilized many times will tend to ware out and become mechanically and thermally weak for further application. These weakness is usually due to chemical attacks and mechanical abrasions in the process of utilizing them, hence bringing their shelf life to as low as 1 to 2 years [7]. Local clays can be used together with additives to improve the various properties of the refractory clay-like; thermal shock resistance, cold crushing strength, bulk density, porosity etc [8, 9]. Some efforts have been made to improve the quality of refractory bricks used in furnaces and boiler lining for efficient operation [1]. The relationship between mechanical properties and thermal conductivity of an insulating fire brick using petroleum coal dust as an additive was studied by [10]. The additive was incorporated in the range of 5wt% to 20w%. It was discovered that the finer coal dust particles of size less than 20µm performed best in terms of the measured mechanical and thermal properties. Amkpa, Ajala [11] worked on Barkin-Ladi clay and determined its suitability for refractory application. It was discovered that it is a good feedstock for refractory application having thermal conductivity of 0.03W/mk, specific heat capacity of 0.07J/g^{0}C, refractoriness of 1665^{0}C, thermal shock resistance of 24 cycles and high energy absorption. In this present study, grog from clay and sawdust were used to improve the refractory properties of Isiagu clay. The Analysis of Variance (ANOVA) was achieved using Excel 2013 and the degree of significance of the factors were determined.

2 MATERIALS AND METHODS

2.1 Sourcing and Characterization

The raw fireclay was be collected from Isiagu Awka South Local Government Area, Anambra State following the standard specified by the American Society for Testing and Materials (ASTM) code ASTM-D4700-15 [12]. Awka city is located directly north of Portharcourt, Rivers State, Nigeria. The location of Isiagu as found by google earth lies on Longitude, 6^{o}10'54" N, Latitude, 7^{o}06'54" E and Altitude, 6.94Km (Fig 1). Definite fire clay of precisely grey to light brown colour was collected. The colour may be due to the high amount of Fe_2O_3 in it. All the additives namely groundnut shell, melon shell, palm kernel shell, sawdust, and chicken droppings were sourced in Awka. All the raw materials used were characterized using Scanning Electron Microscope /Energy Dispersive X-ray (SEM/EDX).

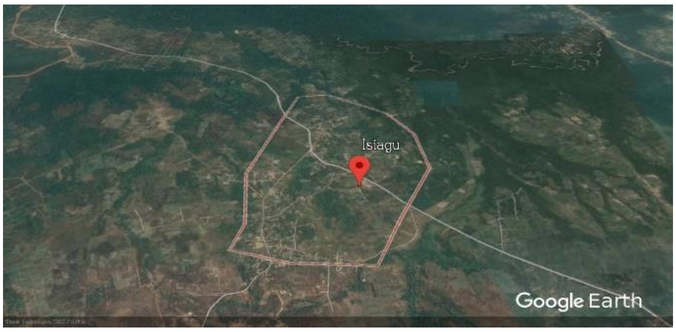


Fig 1. Google Earth Picture of Isiagu Awka South Local Government area

2.2 Material Processing and Moulding of Samples

The raw clay sample was air-dried under shade to allow removal of water and other volatile matter and afterwards crushed to small grain size in a mortar to increase the surface area of the clay sample [3, 13]. To get rid of unwanted particles and plant materials, the slurry was filtered through a 0.425mm mesh sieve [13]. The clay slip obtained was sun-dried for two days and then oven-dried at 100°C and fired at 600°C to remove carbonate and other organic matters [13]. Afterwards, some reasonable part of the clay was fired up to 1000°C for 6 hours to produce grog which was an additive just as did [14]. The processed clay was pulverized and sieved through a 425µm sieve which is recommended by ASTM [15]. The clay sample was mixed proportionally with the additives and moulded into different shapes namely; rectangular, circular, and conical for the respective tests to be carried out.

2.3 Mixing and Moulding

The clay sample was combined individually with the additives namely; grog, groundnut shell, melon shell, palm kernel shell, sawdust and chicken droppings (2.5%wt, 5.0wt%, 7.5wt%, and 10%wt). Due to the addition of water, a stick mass was formed and with the aid of the mould, the samples were formed for different tests to be conducted [13]. The moulds were rubbed with lubricating oil for easy release of the test pieces from the mould after it dries [13]. The formulation of the samples for analysis is shown in table 1.

Table 1. Design of experiment for the production of refractory

Input variables	Ranges
Temperature (^{0}C)	900 – 1100
Additive percentage (wt%)	2.5 – 10

2.3 Measurement and Analysis

a. Linear Shrinkage

This evaluates the linear changes that occur in brick samples when heated. For this purpose, the brick samples will be made in cuboidal shapes. The formula is as given in the work of Adeosun, Akpan [16]. This is calculated based on the original length (Lo) before drying and the final length (Lf) after firing to a certain temperature

$$Percentage\ Total\ Shrinkage = \frac{Lo-Lf}{Lo}\ x\ 100 \quad (2.1)$$

b. Apparent Porosity

Apparent porosity is the percentage relationship between the volume of the open space and the total volume of the material as given by ASTM-C20-00 [17]. This will be determined using the boiling method as was used by Adeniyi, Ajayi [18]. A moulded brick specimen (rectangular) will be used. The brick will be oven-dried at 110^{o}C to constant weight (D). After which it will be transferred to a beaker and boiled with distilled water for 1.5hrs to assist in releasing the trapped air. It will be soaked and the saturated weight free of water (W) will be obtained. Finally, the specimen will be suspended in the water by a rope tied to a spring balance and obtaining the suspended weight (S) when it is completely immersed in water.

$$ApparentPorosity = \%Pa = \frac{W-D}{W-S}\ x\ 100 \quad (2.2)$$

Where:

D = Constant Weight of the dry Sample

S= Weight of the sample suspended in the water

W= Weight of sample in the air including the moisture in its open pores (saturated weight)

c. Bulk Density

The Bulk Density (BD) of a refractory indicates whether the refractory was well fired and thus the degree of densification and will be determined by dividing the test brick mass by the exterior volume and multiplying with the density of water as recommended by ASTM-C20-00 [17]

$$Bulk\ Density = \frac{Dry\ weight\ of\ sample * Density\ of\ water}{saturated\ weight\ in\ air - suspended\ weight\ in\ water} \quad (2.3)$$

d. Refractoriness

This is a thermal property of refractory brick materials that determines the degree of temperature the material can withstand. This will be achieved by making conical brick samples putting them alongside pyrometric sega cones (standard cones with definite deformation temperature) into the kiln and firing to a very high temperature of about 1650^{o}C. After firing, the cones will be examined and the one that bent to the same extent as the test sample is said to have the equivalent temperature of deformation. Alternative, Shuen's formula can be used to roughly estimate the refractoriness of a clay sample [8].

$$Refractoriness\ (K) = \frac{360 + Al_2O_3 - RO}{0.228} \quad (2.4)$$

Where RO is all oxides minus Al_2O_3 and SiO_2

3. RESULTS AND DISCUSSION

3.1 Characterization of Clay and Additives

The results obtained from the characterization of clay and additive samples using SEM/EDX were given in Table 2. SEM/EDX results for the weight composition of elements shows that the clay contains principally, aluminium and silicon elements with a relatively high amount of carbon [19]. The result presented in Table 2 shows that sawdust additive contains only carbon

Weight composition	Mg	Al	P	Si	K	Nb	Ca	Ta	C	O	N
Clay	0.3	17.37		54.01	2.4				24.44	1.47	
Grog	0.25	22.68		76.28						0.78	
Sawdust									100	0	

metal, this is since sawdust is very rich in organic matter compared with coconut shell and rice husk [20]. Grog on the other hand is very high in silicon as expected because it was gotten from a clay sample fired to 900°C where all the carbonaceous materials are removed [14]. The result in Table 3 below was derived from the results in Table 2. The oxide forms of the elements present were obtained and the result showed that clay and grog had almost the same composition by weight of both

Al_2O_3 and SiO_2. Since both samples contain a high amount of Al_2O_3 and SiO_2, they can be referred to as kaolinites, hence suitable for the production of refractory bricks (Chikwelu, et al, 2018; Ajala & Badarulzaman, 2016a).

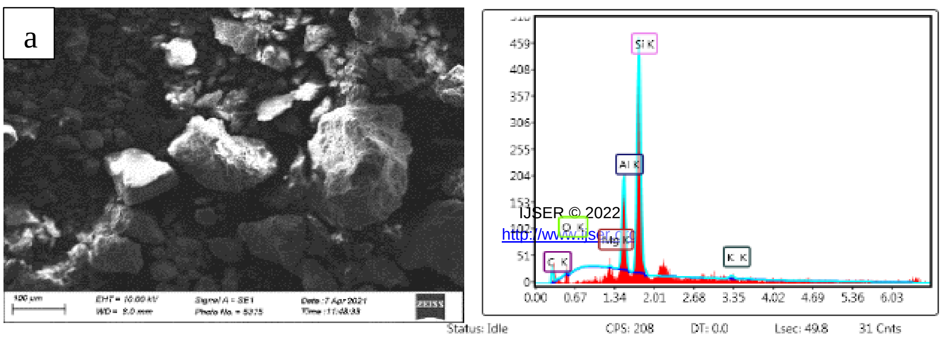
Table 2: SEM/EDX results for the percentage by weight composition of elements in clay and all additives

Table 3: SEM/EDX results for the percentage by weight composition of oxides in clay and all additives

Weight composition	MgO	Al_2O_3	P_2O_5	SiO_2	K_2O	Nb_2O_5	CaO	Ta_2O_5
Clay	0.27	34.97		61.68	3.08			
Grog	0.17	34.33		65.5				
Sawdust								

3.2 SEM/EDX Morphological analysis result for Clay, sawdust and grog

SEM/EDX morphological analysis results for various samples are presented in fig 2. The analysis was carried out at an Electron High Tension of 10kV at different wavelengths of 100µm and 20µm, respectively. The SEM/EDX result for the clay and the grog samples indicates the presence of tiny to large coarse particles (fig2a & b), however, Aboutaleb, Safi [21] obtained the same result but with refractory brick waste (RBW). Also, the sawdust is sheet-like and interwoven which indicates that it can impart good strength and toughness to the clay material (fig 2c).



ISSN 2229-5518

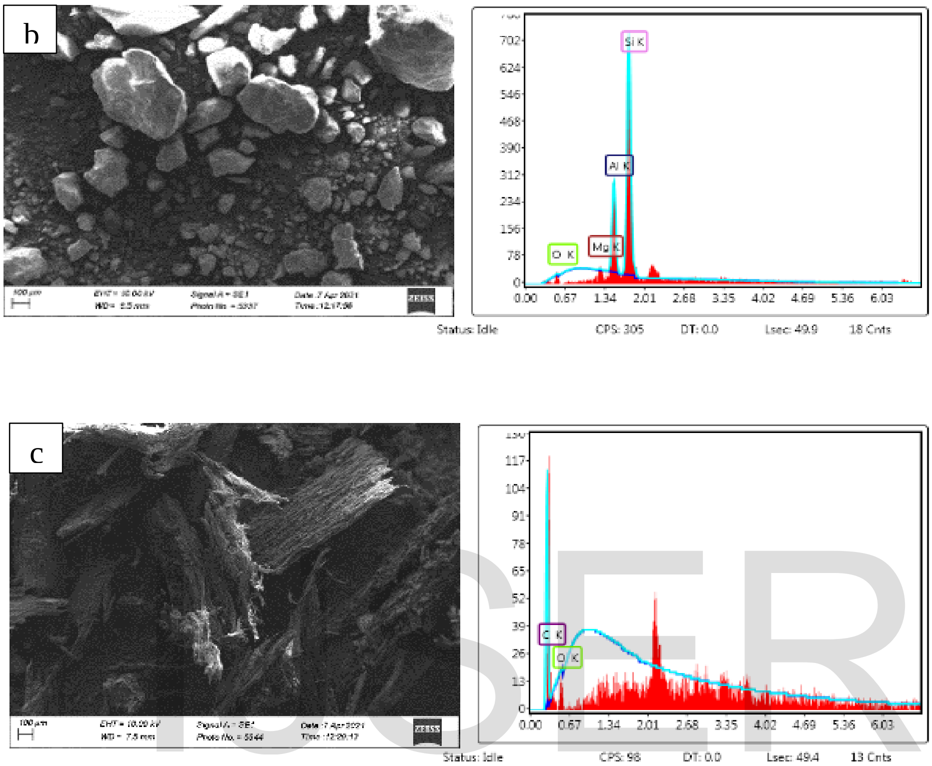


Fig 2. SEM/EDX results at EHT = 10.00kv and wavelength of 100μm for a.) Isiagu Clay sample b.) Grog and c.) Sawdust

3.3Refractory Analysis Results

a. Linear Shrinkage

The results of the linear shrinkage measurement are shown in fig. 3 below. The results show a progressive increase in the linear shrinkage for all samples at (both 900^0C and 1100^0C) and were within the acceptable limit of 7 to 10% [13]. However, the variation of grog was the opposite of this trend and the least among the samples. This agrees with the SEM/EDX results which showed that grog contains more silicon than any other element. The result is shown in Fig 3. shows that grog and sawdust have comparable linear shrinkage at both 900°C and 1100^0C, that is, opposite trends were recorded for the additive percentage.

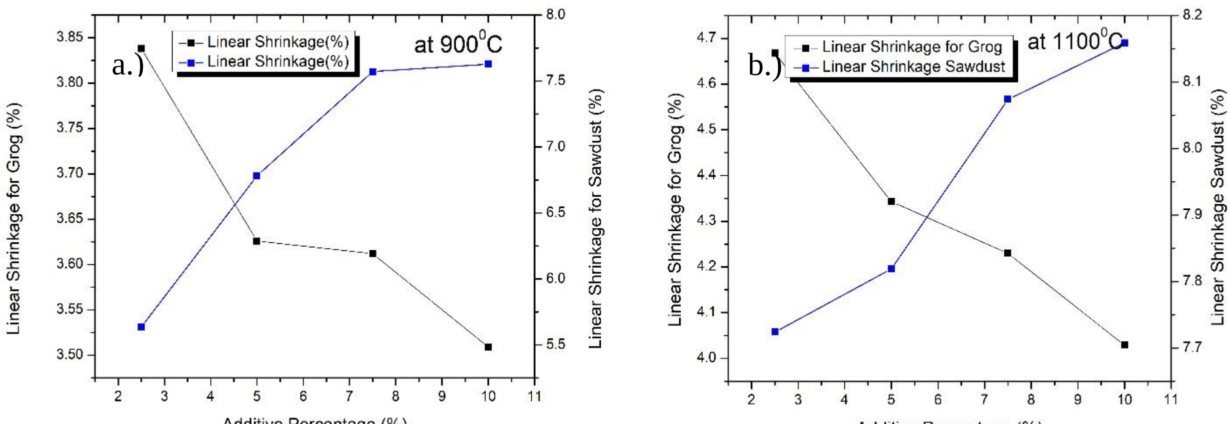


Fig 3. The plot of Linear Shrinkage for Sawdust and Grog at a.) 900°C and b.) 1100°C

b. Apparent Porosity

The apparent porosity increased steadily with the addition of sawdust additive but decreased with grog additives at both 900°C and 1100°C (Fig 4) [22]. Based on the international standard of 2 to 30%, the apparent porosity for sawdust was above the specified limit [13]. This may be due to the presence of carbon only in the sawdust additive as was observed from the SEM/EDX results. This led to large air pockets as observed by Keter [23].

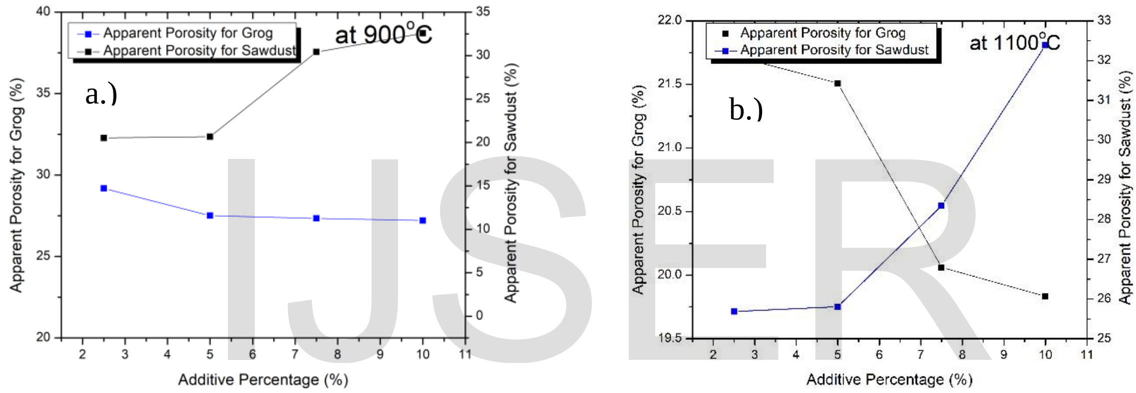


Fig 4. The plot of Linear Shrinkage for Sawdust and Grog at a.) 900°C and b.) 1100°C

c. Bulk Density

There was bulk density decrement according to Fig. 5 is following previous works [10, 13, 24]. A handful of the samples containing grog at 1100°C were within the internationally acceptable range of 1.7 – 2.1g/cm^3 while the samples containing sawdust at 900°C and 1100°C gave values less than the standard [13]

Fig 5. The plot of Bulk density for Sawdust and Grog at a.) 900°C and b.) 1100°C

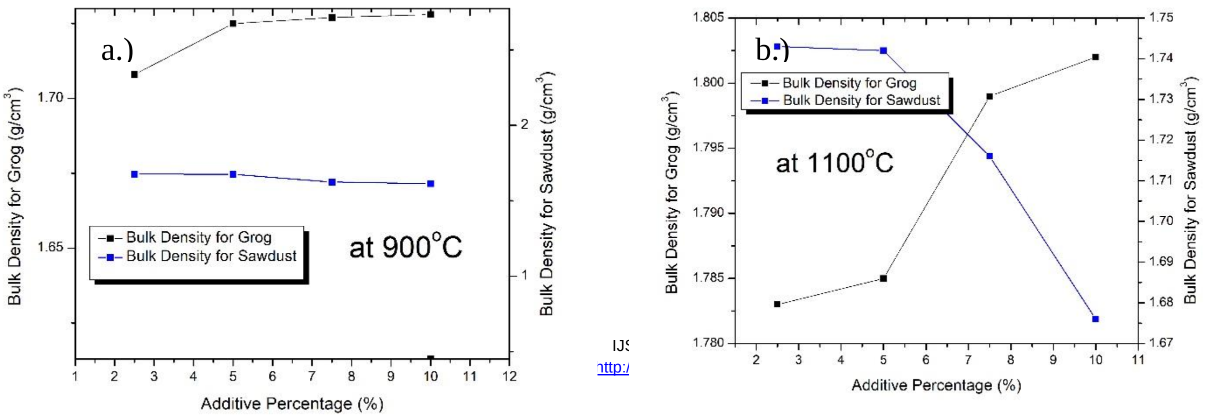


Fig 5. The plot of Bulk Density for Sawdust and Grog at a.) 900°C and b.) 1100°C

d. Refractoriness

This was determined using equation 2.4. The calculation indicated that Isiagu clay is fireclay and hence can withstand temperatures as high as 1447°C. This is very close to that recorded in the work of Harith and Hani [25] where a sintering temperature of 1500°C was observed for 70% kaolin and 30% metakaolin and above 1325°C recorded by Aşkın, Tatar [26] for Cordierite with waste magnesite.

3.4 ANOVA Results for Grog and Sawdust Additives

Analysis of variance was done for all the samples with various additives and all the properties measured.

Grog Only additive

Analysis of variance was carried out for all samples containing grog additives. The result of two-way ANOVA without replication is presented in Table 4 to Table 6. The result shows that the effect of percentage additive for all properties was not significant (P>0.05), while the temperature was not significant only for linear shrinkage measurement [27, 28]. This is because organic matters have been removed from the grog by firing and hence increase in grog per cent had no impact on the properties measured.

Table 4: Linear Shrinkage

ANOVA

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*	*Remark*
Temperature	14.61114	2	7.305569	1.86858	0.233968	5.143253	**Not significant**
Percent Additive	8.061763	3	2.687254	0.687332	0.591915	4.757063	**Not Significant**
Error	23.45814	6	3.90969				
Total	46.13104	11					

Table 5: Apparent Porosity

ANOVA

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*	*Remark*
Temperature	967.1841045	2	483.6	85.06908	3.95E-05	5.143253	**Significant**
Percentage Additives	2.52790825	3	0.843	0.148229	0.927107	4.757063	**Not Significant**
Error	34.1081895	6	5.685				
Total	1003.820202	11					

Table 6: Bulk Density

ANOVA

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*	*Remark*
Temperature	53.83901	2	26.9195	7.794196	0.021468	5.14325285	**Significant**
Percent Additive	10.52781	3	3.509272	1.016064	0.448761	4.757062663	**Not Significant**
Error	20.72273	6	3.453788				
Total	85.08955	11					

Sawdust

Analysis of variance was carried out for all samples containing sawdust additive. The result of two-way ANOVA without replication is presented in Table 7 to Table 9. The result shows that the effect of temperature was significant (P<0.05) for all properties except linear shrinkage [27, 28]. On the other hand, the percentage additive was only significant for two properties (linear shrinkage and bulk density) and was not significant for apparent porosity.

Table 7: Linear Shrinkage

ANOVA

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*	*Remark*
Temperature	1821.949	2	910.9746	1152.313	1.75E-08	5.143253	**Significant**
Percent Additive	90.43702	3	30.14567	38.13197	0.000266	4.757063	**Significant**
Error	4.74337	6	0.790562				
Total	1917.13	11					

ANOVA

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*	*Remark*
Temperature	5.837832	2	2.918916	1.14091	0.380256	5.143253	**Not Significant**
Percent additive	18.62342	3	6.207806	2.42643	0.163722	4.757063	**Not Significant**
Error	15.35047	6	2.558411				
Total	39.81172	11					

Table 8: Apparent Porosity

Table 9: Bulk Density

ANOVA

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*	*Remark*
Temperature	55.62113	2	27.81057	7.860479	0.021077	5.143253	**Significant**
Percent additive	10.02824	3	3.342747	0.944806	0.475959	4.757063	**Not Significant**
Error	21.22815	6	3.538024				

Total	86.87752	11

4. CONCLUSION

The effect of grog and sawdust on the refractory properties of Isiagu clay was studied. Also, the ANOVA was carried out using Excel 2013. The SEM/EDX result showed that sawdust additive contains only carbon metal and implies that samples containing sawdust additive can be useful for insulating applications. Grog additive showed high content of Al_2O_3 and SiO_2 which marks it out for refractory applications. The SEM/EDX morphological results further confirm the class of application of the two additives used in this work. Grog which contains tiny to coarse particles has a strength advantage over sawdust which has a sheetlike interwoven morphology. The reverse trend found in the samples containing grog due to the high content of Al_2O_3 and SiO_2 further confirms its suitability for a refractory application. Bulk density also validated other results showing that samples containing grog can be used in a refractory application. ANOVA results confirmed that for grog and sawdust additives, temperature and the additive percentage was not significant for linear shrinkage measurement.

Acknowledgement

The authors wish to acknowledge the assistance and contribution of members of staff of chemical engineering, management of Production Development Institute (PRODA) and Petroleum Technology Trust Fund for sponsoring this project.

References

1.) Ogunsemi, B., et al., *Investigative study of the effects of certain additives on some selected refractory properties of ant-hill clay for furnace lining.* International Journal of Civil Engineering and Technology (IJCIET), 2018. **12**: p. 761-772.
2.) Hussein, H., *Improve Some Properties of Refractory Mortar Manufactured From Grog Bauxite, Attapulgite, CaO and White Cement by Using Gum Arabic.* International Journal of Civil Engineering and Technology, 2019. **10**(3).
3.) Amkpa, J.B., *A Study on Thermal Conductivity of Six Unexplored Nigerian Clays for possible Refractory and Insulating Materials.* . 2017, Unpublished PhD Dissertation, Faculty of Mechanical and Manufacturing Engineering Universiti Tun Hussein Onn Malaysia.
4.) Oke, S.R., et al., *Production and Characterization of Clay Bonded Carbon Refractory from Ifon Clay and Spent Graphite Electrode.* International Journal of Metallurgical Engineering, 2015. **4**(2): p. 33-39.
5.) Olalere, A., S. Yaru, and O. Dahunsi, *Evaluation of the chemical and thermo-physical properties of locally aggregated kaolin-based refractory materials.* Journal of Mechanical Engineering and Sciences, 2019. **13**(1): p. 4743-4755.
6.) Esezobor, D.E., et al., *Evaluation of cost effectiveness of Onibode Fire-Clay for production of high quality Refractory bricks.* 2015.
7.) Antonovič, V., et al., *Refractory Materials for Biofuel Boilers.* Frontiers in Bioenergy and Biofuels, 2017: p. 443.

8.) Iyasara, A.C., et al., *Influence of grog size on the performance of NSU clay-based dense refractory bricks.* American Journal of Materials Science and Engineering, 2016. **4**(1): p. 7-12.
9.) Ahmad, S., Y. Iqbal, and R. Muhammad, *Effects of coal and wheat husk additives on the physical, thermal and mechanical properties of clay bricks.* Boletín de la Sociedad Española de Cerámica y Vidrio, 2017. **56**(3): p. 131-138.
10.) Rahman, M.H., et al., *Study of thermal conductivity and mechanical property of insulating firebrick produced by local clay and petroleum coal dust as raw materials.* Procedia Engineering, 2015. **105**: p. 121-128.
11.) Amkpa, J.A., J. Ajala, and N.A. Badarulzaman, *Thermal Conductivity of Barkin-ladi Fireclay Brick as Refractory Lining.* IOSR Journal of Mechanical and Civil Engineering, 2017. **14**(2): p. 1-5.
12.) ASTM-D4700-15, *Standard Guide for Soil Sampling from the Vadose Zone*, W.C. ASTM International, PA, www.atms.org, Editor. 2015.
13.) Chima, O., C. Nwoye, and E. Nnuka, *Enhancement of refractory properties of blended clay with groundnut shell and rice husk additives.* American Journal of Engineering Research (AJER), 2017. **6**(6): p. 218-226.
14.) Hassan, A.M., H. Moselhy, and M.F. Abadir, *The use of bagasse in the preparation of fireclay insulating bricks.* International Journal of Applied Ceramic Technology, 2019. **16**(1): p. 418-425.
15.) Ajala, A.J. and N.A. Badarulzaman, *Thermal conductivity of aloji fireclay as refractory material.* International Journal of Integrated Engineering, 2016. **8**(2).
16.) Adeosun, S., et al., *Refractory behaviors of magnetite–kaolin bricks.* JOM, 2016. **68**(11): p. 2824-2831.
17.) ASTM-C20-00, *Standard test method for apparent porosity, water absorption, specific gravity and bulk density*, A. international, Editor. 2015.
18.) Adeniyi, S.S., O.J. Ajayi, and C.A. Umeh, *The Effect of Grog Sizing on the Performance of Ire Ekiti Fire Clay Refractory Bricks.* , in *Department of Glass and Ceramic Technology*. 2018, Federal Polytechnic Ado Ekiti, P.M.B 5351, Ado Ekiti, Nigeria.
19.) Elakhame, Z., et al., *Characterization of Ujogba Clay Deposits in Edo State, Nigeria for Refractory Applications.* Kathmandu University Journal of Science, Engineering and Technology, 2016. **12**(2): p. 71-82.
20.) Obidiegwu, E.O., et al., *Physiochemical and thermal properties of insulating refractory bricks developed from Ikorodu and Osiele Fireclays and Agroforestry wastes*. nd, UNILAG journal of medicine, science and technology.
21.) Aboutaleb, D., et al., *Use of refractory bricks as sand replacement in self-compacting mortar.* Cogent Engineering, 2017. **4**(1): p. 1360235.
22.) Babaso, P.N. and H. Sharanagouda, *Rice husk and its applications: Review.* International journal of current microbiology and applied sciences, 2017. **6**(10): p. 1144-1156.
23.) Keter, C.K., *Development and testing of composite refractory bricks (a case study of selected Kenyan clays)*. 2016, Egerton University.
24.) Velasco, P.M., et al., *Development of sustainable fired clay bricks by adding kindling from vine shoot: Study of thermal and mechanical properties.* Applied Clay Science, 2015. **107**: p. 156-164.
25.) Harith, I.J. and M.H. Hani, *Study the physical properties of refractory mortar contains kaolin-metakaolin fire brick powder SiC.* International Journal of ChemTech Research, 2017. **10**(9): p. 790 - 799.

26.) Aşkın, A., et al., *The utilization of waste magnesite in the production of the cordierite ceramic.* Energy Procedia, 2017. **107**: p. 137-143.

27.) Li, J., et al., *Application of response surface methodology (RMS) for optimization of sintering process for the preparation of magnesia partially stabilized zirconia (Mg-PSZ) using natural baddeleyite as starting material.* . Ceramics International, 2013. **39**: p. 197-202.

28.) Shehu, M.S., S.I. Lamido, and T. Nyam, *Utilization of response surface methodology for the optimization of refractory bricks production.* International Journal of Engineering Development and Research., 2018. **6**(4): p. 301-307.