I O U R N A L O F

Ceramic Processing Research

Fireclay bricks produced from lithomargic clay

Anthony Andrews^{a,*}, Simon K. Y. Gawu^b and Peter A. Olubambi^c

^aDepartment of Materials Engineering, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana ^bDepartment of Geological Engineering, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana ^cDepartment of Chemical, Metallurgical and Materials Engineering, Tshwane University of Technology, Pretoria, South Africa

Lithomargic clay underlying Awaso bauxite deposits is non-plastic and therefore cannot be used to produce ceramic bodies. The plasticity of lithomargic clay has been enhanced by addition of 20 wt% of clay binder, moulded, dried and fired at 1350 and 1400 °C, respectively. The dwell time was varied from 30 to 120 minutes. Properties such as bulk density, apparent porosity, linear firing shrinkage, and compressive strength were investigated. Crystalline phases after firing were determined using X-ray diffraction technique. The results show the properties measured depends on both the temperature and dwell time. The optimum compressive strength for such bricks could be achieved either by firing at 1350 °C for 120 minutes or at 1400 °C for 30 minutes.

Key words: Fireclay, Refractory, Lithomargic clay, Compressive strength, Dwell time.

Introduction

Lithomargic clay underlying Awaso bauxite deposits in Ghana result from incomplete bauxitisation process. The chemical and mineralogical characteristics indicate that lithomargic clay consist mainly of kaolinite and gibbsite [1]. The SiO₂ (mainly from the kaolinite) and Al₂O₃ contents varies between 27-52% and 31-45%, respectively. The impurities include Fe₂O₃, TiO₂, K₂O, CaO, MgO and Na₂O [2]. The chemical composition suggests that lithomargic clays could be used as fireclay aluminosilicate refractories [3-6]. Production of refractory bodies from this clay has not been successful due to its low plasticity. The plasticity of this material could be improved by addition of a suitable binder. There are several local clay deposits with high plasticity which could be used as binding material to improve the plasticity of lithomargic clay. Some of the local clays available include Mfensi clay, Fosu clay and Afari clay [7,8]. Densification of refractory bodies depends on firing temperature and dwell time [9]. Dwell time also depends on size of bricks being fired and chemical composition [10]. It is important to determine the optimum firing parameters for efficient utilization of energy. The chemical compositions of lithomargic, Mfensi, Fosu and Afari clays show that the clays are contaminated with impurity oxides [1, 7, 8]. These impurities act as fluxing agents at high temperatures decreasing the eutectic

*Corresponding author:

Fax: +233-32-206-0317

temperature of Al_2O_3 -SiO₂ refractories, thus making it possible to achieve full densification due to mass transport at relatively lower temperatures [3-6]. Nevertheless, the effective use of these fluxing agents is achieved when the liquid phase is minimised to avoid decomposition or deformation during densification [8, 11-13]. At higher temperature and extended dwell time, more liquid phase forms which could promote deformation during densification. The liquid phase could be minimised by controlling the firing temperature as well as the dwell time to obtain desired properties. This work is therefore aimed at selection of suitable clay binder to produce fireclay refractories from lithomargic clay. The effect of dwell time on the properties of fireclay bricks produced is also investigated.

Methodology

Materials

Lithomargic clay was taken from Awaso near Sefwi-Bekwai in the Ashanti region of Ghana. Mfensi, Fosu and Afari clays were obtained from Mfensi, Fosu and Afari towns, respectively and Atterberg limits determined

 Table 1. Atterberg limits of Mfensi, Afari, Fosu and Lithomargic clays.

Clay type	Liquid Limit (LL)	Plastic Limit (PL)	Plasticity Index (PI)		
Mfensi	43.62	21.72	21.90		
Afari	60.17	27.55	32.62		
Fosu	51.76	27.79	23.98		
Lithomargic clay	32.60	24.60	8.00		

Tel:+233-20-046-0447

E-mail: anthonydrews @gmail.com

 Table 2. Chemical composition (wt%) of lithomargic and Mfensi clays.

Material	SiO ₂	Al_2O_3	Fe ₂ O ₃	MgO	CaO	MnO	Na ₂ O	K_2O	TiO ₂	LOI
Lithomargic	50.3	42.2	0.4	0.4	0.2	0.3	0.4	0.5	1.4	3.9
Mfensi	65.6	19.5	0.8	0.4	0.2	0.2	0.9	0.9	0.6	10.9



Fig. 1. Mineralogical composition of (a) Mfensi and (b) Lithomargic clays.

for each to select a suitable binder for lithomargic clay. Liquid and plastic limit tests were determined from which the plasticity index (PI) were calculated. Clay samples were sieved using 425 µm sieve size and the undersize used for the tests. Tests were conducted according to ASTM D 4318. The results of the tests are shown in Table 1. An Atterberg limit of lithomargic clay has been added for comparison. The plasticity index (PI), except for lithomargic clay, falls within the range 10-60% recommended for ceramic clays [14]. Severe shrinkage during drying and firing of refractory bricks are not desirable. According to British Standard, BS 1377-7, for low shrinkage property, the PI must be within 10 to 20%. Mfensi clay was therefore selected as the suitable binder for lithomargic clay.

Lithomargic and Mfensi clay samples were dried in an oven at a temperature of 105 °C for 24 hours. The samples were pulverised in a vibratory mill for 30 minutes. The chemical compositions determined using X-ray fluorescence spectroscopy (XRF-Spectro X-LAB 2000) is provided in Table 2. The SiO₂ and Al₂O₃ contents constitute over 70% of the major oxides. The minor constituents are Fe₂O₃, MgO, CaO, MnO, Na₂O, K₂O and TiO₂ which constitute less than 5 wt% and therefore could be used to produce fireclay refractories [4, 6]. Mfensi clay is a semi-acid clay with SiO₂ content of 65.6% with a corresponding Al_2O_3 content of 19.5%. The loss on ignition which constitutes both organic matter and interlayer water hydroxyls of the crystalline structure of the clay minerals was 10.9% for Mfensi clay and 3.9% for lithomargic clay.

Mineralogical compositions were analysed using Xray diffractometer (Siemens D5000), operating at 40 kV and 40 mA and using Cu K α radiation. The samples were scanned at 10-50 ° (2 θ) and step size of 0.02 °. Fig. 1 shows the XRD pattern of the Lithomargic and Mfensi clays. The mineralogical composition of Mfensi clay consists mainly of quartz with minor kaolinite content. The lithomargic clay was composed mainly of kaolinite with minor fractions of gibbsite.

Experimental procedure

Clay samples were prepared by adding 20 wt% Mfensi to 80 wt% litomargic clay by wet mechanical agitation. Fireclay bricks of dimension $5.2 \text{ cm} \times 5.2 \text{ cm} \times 5.2 \text{ cm}$ were produced from this composition by mixing, semidry pressing and drying. Drying was carried out in open air for 48 hours followed by oven drying for 24 hours at 105 °C. The dried bricks were fired in a muffle furnace at 1350 and 1400 °C for 30, 60, 90 and 120 minutes, respectively. Densification parameters were determined from linear firing shrinkage (ASTM C326-09), apparent porosity, and bulk density using the water displacement (Archimedes) method according to ASTM C134. The compressive strengths of the fired bricks were also determined. The phase composition of the fireclay bricks were determined using X-ray diffractometry (XRD). For each dwell time, five bricks were tested and the average value taken.

Results and Discussion

Fig. 2 shows the influence of dwell time on the bulk density and apparent porosity of the bricks fired at 1350 and 1400 °C, respectively. The apparent porosities were calculated from the bulk densities. Slight density variations could be observed for bricks fired at 1350 and 1400 °C. This is because above 1300 °C most of the Al_2O_3/SiO_2 have transformed into mullite therefore densification is minimised. The average bulk density was slightly lower for bricks fired at 1350 °C than 1400 °C except at 120 minutes dwell time where the bulk densities were about the same. At 1350 °C, as the dwell time increased the average bulk density increased from 30 to 60 minutes and remained almost constant thereafter. Improvement in densification is as a result of the formation of mullite phase as observed from the



Fig. 2. Effect of dwell time on the (a) bulk density and (b) apparent porosity.

XRD patterns. Additionally, the presence of oxides could act as fluxing agents enhancing densification [9, 12]. After 60 minutes of dwell time, the average bulk density remained constant indicating little or no further formation of the mullite phase. On the other hand, dwell time did not have significant effect on the average bulk density of bricks fired at 1400 °C. This is due to complete formation of mullite at such higher temperatures; hence as dwell time increased the average bulk density remained the same. It took 120 minutes for bricks fired at 1350 °C to reach the same average bulk density as bricks fired 1400 °C for 30 minutes.

At 1350 °C the average apparent porosity decreased from 3.12 to 2.34% as the dwell time increased (Fig. 2(b)). There was a sharp decrease from 60 to 90 minutes. This is mainly attributed to the expansion that accompanies the quartz to cristobalite polymorphic transformations that occur upon firing at such high temperature and for longer dwell times closing the pores [3-6]. This transformation is observed from the XRD results. The decrease in porosity reduces the volume of the bricks whilst improving the mechanical properties [12]. At 1400 °C, the average apparent porosity decreased from 2.4% to 2.2% after 60 minutes dwell time after which it remained constant. At 120 minutes dwell time, the average apparent porosity was lower (2.19%) than bricks fired at 1350 °C (2.34%). Thus even though the average bulk density were about the same after dwell time of 120 minutes, slight variation



Fig. 3. Effect of dwell time on linear firing shrinkage.



Fig. 4. XRD patterns of bricks fired at (a) 1350 $^{\circ}$ C and (b) 1400 $^{\circ}$ C at various dwell times.

was observed in their average apparent porosity values.

Fig. 3 shows the effect of dwell time on linear firing shrinkage. The linear firing shrinkage could be used as direct measure of extent of densification. The average linear firing shrinkage at 1350 °C increased gradually from 30 to 90 minutes dwell times and remained constant through to 120 minutes. The behaviour was different for bricks fired at 1400 °C; it increased slightly from 30 to 60 minutes after which it remained constant. Thus bricks fired at 1400 °C reach full densification faster than bricks fired at 1350 °C. The average shrinkage values were higher for bricks fired at 1400 °C than 1350 °C becoming almost the same after 120 minutes dwell time. The higher shrinkage values observed at higher firing temperature is due to volume expansion that accompanies quartz to cristobalite phase transformation. At such higher temperatures, nearly full densification would have been achieved at shorter dwell time hence slight changes in shrinkage values observed when dwell time increased [9].

Fig. 4 shows the phase composition of the fired bricks at various temperatures. It is interesting to note



Fig. 5. Effect of dwell time on compressive strength.

that there were no significant differences in crystalline phases present as the dwell time increased for the different firing temperatures. It is observed that at both firing temperatures the presence of mullite is evident in addition to quartz and cristobalite phases. The formation of crystalline mullite starts to form at 1250 °C with complete mullitilisation occurring at 1400 °C. It could be observed that there is an increase in crystallinity of the quartz phases as the dwell increased, especially from 30 to 60 minutes. It is interesting to note that even though the crystalline phases present remained the same at all dwell times, the measured strength varied as the dwell time is increased. This is particularly the case for bricks fired at 1350 °C. Thus the increase in strength as dwell time increased is mainly due to improved densification. At such temperatures complete mullitilisation has occurred which further improves the strength, creep and corrosion resistance [6, 11, 15, 16].

Fig. 5 shows the effect of dwell time on the compressive strength. Whereas the average compressive strength of bricks fired at 1400 °C remained almost constant as dwell time increased, there was a sharp increase in the average compressive strength of bricks fired at 1350 °C from 30 to 60 minutes followed by gradual increase in strength up to 120 minutes dwell time. At 120 minutes, the average compressive strength values at both temperatures are about the same. The results show that the compressive strength is influenced by both firing temperature and dwell time. The compressive strength values at such high temperatures are influenced more by enhanced densification rather than phase composition. This is evident from Figs. 3 and 4. The presence of the highly refractory phases (cristobalite and mullite) and the densification of the bricks due to the presence of flux agents as temperature and dwell time increased are responsible for the improved compressive strength values recorded.

Conclusions

It has been possible to produce fireclay refractories from lithomargic clay using Mfensi clay as a suitable binder. The results show that full densification of fireclay bricks produced from 80 wt% lithomargic-20 wt% Mfensi clay formulations could be achieved by either firing at 1350 °C for 120 minutes or at 1400 °C for 30 minutes. Under these conditions, optimum strength values at both temperatures are about the same. The high strength values at such high temperatures are partly due to crystalline phases present and partly due to enhancement in densification of fired bricks. The linear firing shrinkage values were within limits acceptable for use as refractory material. For practical purposes where access to high temperature furnaces become a constraint, bricks could be fired at relatively low temperature (1350 °C) for longer dwell time (120 minutes) to achieve optimum strength.

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