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Improving the Physical and Thermal Properties of the Fire Clay Refractory Bricks Produced from Bauxite

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Abstract: In this study chemical and mineralogical tests were carried out on the bauxite mineral in order to assess its suitability for the production of refractory bricks. Two types of additives were used, white Kaolinite as a binding material in ratios of 10, 15 and 20% and the chemical additives synthetic alumina in ratios of 10, 20, 30, 40, 50 and 60% for the purpose of improving the general characteristics of the bauxite bricks. Due to the low plasticity of bauxite grains, the semi-dry pressing was used at a forming pressure of 40-45 N m^{-2} with added water ratio of 9-12% in addition to 0.5% of molasses as a binder. The formed samples were of cylindrical shape (50×50 mm) for diameter and height. The drying process of the samples was carried out over a wide range of temperatures up to 110°C with a soaking time of 2 h. The general characteristics for the samples was determined under the optimum conditions achieved in this study. The optimum conditions were: Best forming pressure 45 N mm^{-2}; best firing temperature 1550°C and soaking time 2 h; best bauxite ratio 70% fired bauxite and 10% non fired bauxite; best grain graduation for the bauxite fired as raw material was consisting of 45% coarse, 10% medium and 45% fine. The validity of the samples was tested after firing and the results were compared with the international standards for this type of refractory bricks. It has been proved their suitability for the lining of rotary kilns used in the cement industry.

Key words: Refractory bricks, kaolinite, bauxite, firing temperature, thermal conductivity

INTRODUCTION

Bauxite is defined as a mixture of minerals which consists mainly of aluminum oxide bounded to one or more water molecules with small amounts of impurities such as silica, K_2O, SiO_2, Fe_2O_3, TiO_2, CaO, MgO and Na_2O. The main components of bauxite are listed in Table 1 as published by (Hill and Sheffield, 1979). The word Bauxite is used generally as a name for rocks which contains a suitable amount of mineral aluminum hydroxide. It is distributed worldwide at different deposits and its color varies from ferric brown-black to red-pink depending on its composition. Bauxite is distinguished in its structure that it contains granular crystalline layers (Ullmann, 1982).

During firing of bauxite below 1200°C, its structure is transformed into dense granules contains mainly corandom (α-Al_2O_3). At temperatures within the range 1250-1350°C the mullite phase is formed as a result of the reaction between silica and alumina.

Table 1: Main components of bauxite

Properties	Gibbsite	Boehmite	Diaspore
Chemical formula	$Al_2O_3.3H_2O$ or $Al(OH)_3$	Al_2O_3. H_2O or (AlOOH)	Al_2O_3. H_2O or (AlOOH)
Alumina content (%)	65.4	85	85
Combined water content (%)	34.6	15	15
Crystal system	Monoclinic	Orthorhombic	Orthorhombic
Hardness (mohs 'scale)	3.5-2.3	5-3.5	7-6.5
Sp.density (g cm^{-3})	2.4-2.3	3.06-3.01	3.5-3.3

Mullite is considered as a binding phase in most of refractory bricks and it has a high resistance to melting and minimum thermal expansion as well as low thermal conductivity. Mullite also characterized by its chemical and thermal stability and higher resistance to chemical reactions (Nasr *et al.*, 1982).

In this study, the suitability of bauxite for the production of refractory bricks is studied by applying different chemical, physical and mineral tests of the raw materials. After the production of the refractory bricks from the tested raw materials at different operating conditions, the general properties of the product was tested in terms of granular size, firing temperature, soaking time and forming pressure.

MATERIALS AND METHODS

The raw materials used in the experiments were subjected to different preliminary analysis steps in order to determine to which extent those raw materials are suitable for the preparation of the fire clay refractory bricks. Representative samples of the raw materials

Table 2: The chemical analysis of the raw materials used for the production of the fire clay refractory bricks

Component	Wt. % in bauxite	Wt. % in Kaolinite
SiO_2	28.86	46.84
Al_2O_3	63.02	34.45
Fe_2O_3	1.96	1.09
TiO_2	2.04	1.40
CaO	1.10	0.35
MgO	0.21	0.28

Table 3: Mass ratios of raw materials used to prepare the improved bauxite bricks through the addition of alumina

	Starting materials Wt.%			
Mix.	Calcined bauxite	Unfired bauxite grain size (0.3 mm)	Unfired kaolin grain size (3.5 mm)	Alumina grain size (0.1 mm)
1	70	10	20	-
2	63	9	18	10
3	56	8	16	20
4	49	7	14	40
5	42	6	12	30
6	35	5	15	50
7	28	4	8	60

(Bauxite and Kaolinite) were taken for analysis. The chemical composition of the Bauxite and Kaolinite raw materials is shown in Table 2.

The behavior of the raw materials when subjected to heating and the corresponding changes during expansion and fusion processes were experimentally measured using the heating microscope.

The softening point of the raw materials was characterized using the standard Sager method. The tests were carried out by manufacturing of molds from the bauxite bricks which have a conical form similar to the Sager cones. The molds then fixed on a soft layer of refractory clay which were then wrapped with Sager cones having the numbers 26, 27, 28, 29, 30, 31, 32 and 33 with the corresponding softening temperatures 1695, 1585, 1605, 1635, 1655, 1665, 1680 and 1610°C, respectively. The clay layer carrying the molds was then put into an electrical furnace. The samples were heated till the temperature in the furnace reached its maximum value at 700°C. The residence time of the samples in the furnace was 2 h. The samples were then cooled and compared with the standard Sager cones.

The Differential Thermal Analysis (DTA) method was implemented to test the thermal behavior resulted from the chemical and physical changes of the raw materials during heating and cooling processes.

The experimentally produced bauxite refractory bricks from the unfired bauxite and unfired kaolin only was characterized by its longitudinal expansions and shrinkages. Such dimensional changes are attributed the reactions taking place between bauxite grains and the silica existing in the clay used as a binding material forming the mullite which has a density less than the density of corandom. This will significantly influence the physical and chemical properties, thus reducing the thermal resistance and life of the bricks (Nasr *et al*., 1982). In order to improve the quality of produced bricks, different amounts of aluminum oxide were added. The refractory bricks were produced with the different mass ratios listed in Table 3 according to the achieved optimum operating conditions through experiments carried out without the addition of aluminum oxide. The operating conditions such as firing temperature soaking time and forming pressure were adjusted to obtain an optimum gradual graining of the bricks.

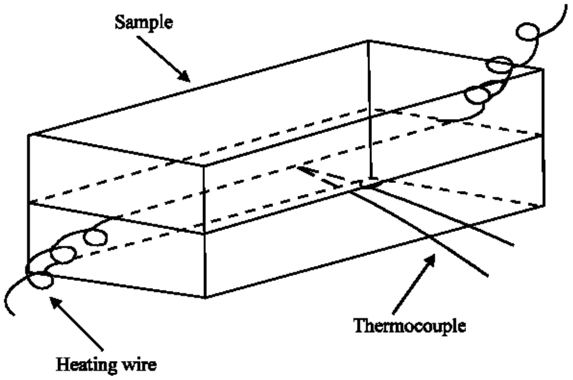

Fig. 1: A schematic diagram of the instrument TC-31 used for the conductivity measurements

Cylindrical shape of bauxite bricks were formed by implementing the semi-dry pressing method. The size of the prepared samples was 50×50 mm and the amount of water added was 9-12% and the amount of mollas added as binding material was 0.5% whereas the forming pressure was varying in the range 40-45 N mm^{-2}. After the shapes has been formed, they were left under atmospheric conditions for 24 h. They were then put in electrical dryer at 60°C. The temperature was then raised to 110°C for 24 h. According to the obtained results of the measurement of behavior of the raw materials using heating microscope and determining its softening points, the samples were heated from room temperature to 700°C with a rate of 2°C min^{-1} and from 700 to 1000°C with a rate of 3°C min^{-1}. The samples were then heated to 1550°C and kept at the same temperature for 2 h as a soaking time in order to increase the sintering time.

After firing of the samples of bauxite bricks, the general properties of the samples were tested. The determined properties are: volumetric density, apparent porosity, water absorption capacity, linear expansion and thermal conductivity. The heat transfer coefficient of the samples after firing at 1550°C was determined by means of an instrument (TC-31) working on the basis of steady state thermal conductance principle as shown in Fig. 1. A metallic tape called the heating wire at the right side of the instrument is inserted between two similar forms of the material to be tested. The heating wire is heated by passing electric current while its temperature change is measured by means of a thermocouple to estimate the thermal conductivity of the sample.

RESULTS AND DISCUSSION

Characterization of the raw materials: As can be noticed from the data of Table 2, the bauxite fired at 1400°C contains an amount of 63.02% alumina which makes it possible to classify it as intermediate level alumina content refractory materials which contains up to 65.60% alumina (Bain, 1986). This means that the produced refractory bricks will have a higher softening and fusion temperatures. The silica content of 28.86% and the presence of 1.96% of Fe_2O_3 will result in improvement of specific gravity and density as well as improvement it is resistance against sudden thermal changes. The Kaolinite used as a softening and binding material, which has the chemical analysis listed in Table 2 has a composition lies within the accepted limits of fire clays suitable as raw material for the production of bauxite bricks.

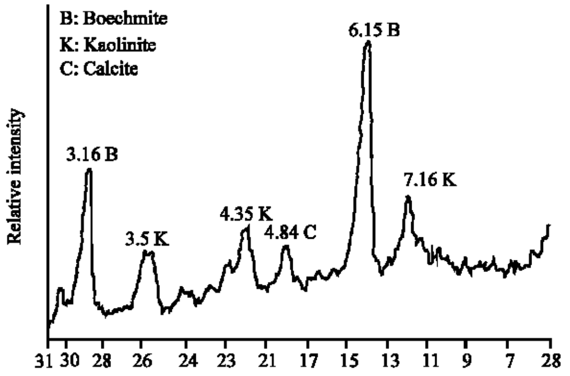

Fig. 2: The mineral composition of the raw bauxite using XRD analysis

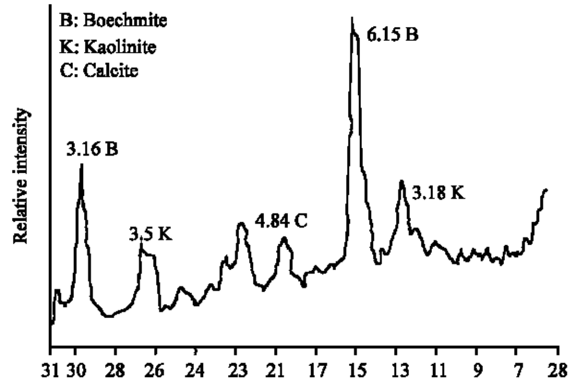

Fig. 3: Mineral composition of white kaolin using XRD analysis

Mineral Analysis of bauxite using X-Ray Diffraction (XRD) method, shown in Fig. 2, indicates that the mullite phase appears dominantly whereas a small amount of corandom phase is existed. On the other hand, the mineral analysis of Kaolinite, presented in Fig. 3, exhibits a higher ratio of Kaolinite and small amounts of quarts minerals.

Heating microscope analysis: The heating microscope analysis was implemented on the raw materials to determine fusion temperatures as well as the changes occurring during heating and the accompanying expansion and shrinkage during the heating at higher temperatures. The results of heating microscope test was used to determine the firing temperatures. The determined temperatures are: 1300, 1400, 1500 and 1550°C. Figure 4 shows the behavior of bauxite during the heating process. The measurements indicate that the softening of bauxite has not been detected up to the maximum temperature of the heating microscope (1500°C). The results of heating microscope tests of Kaolinite indicate that no changes have been detected when the samples are heated up to 1500°C, as displayed in Fig. 5, which means that the Kaolinite has high fusion temperature.

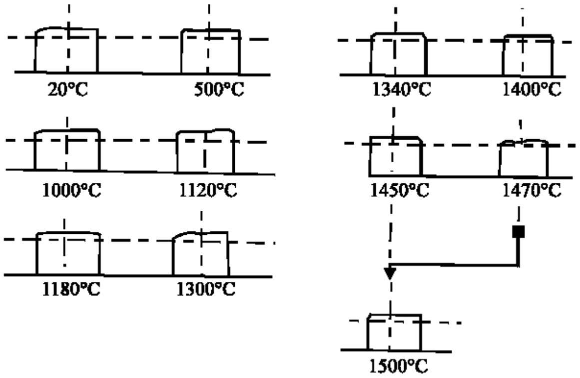

Fig. 4: The behavior of bauxite under thermal microscope

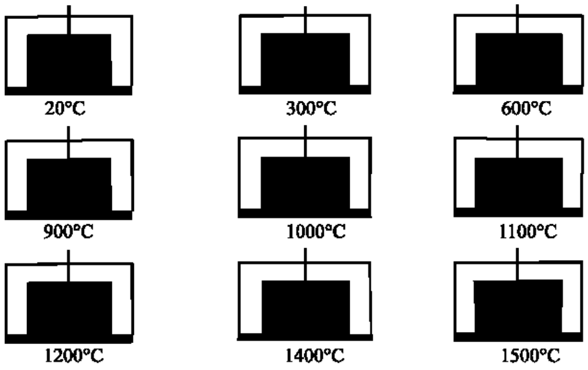

Fig. 5: The measured behavior of Kaolinite under thermal microscope

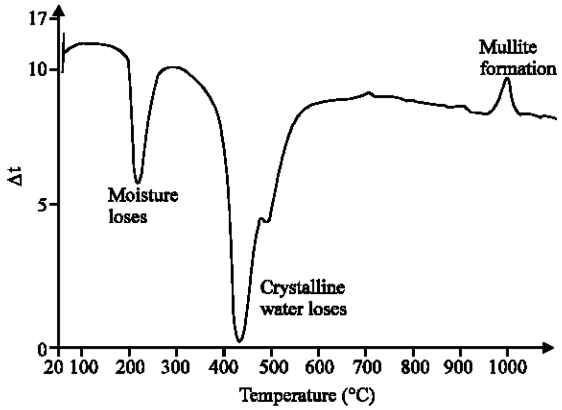

Fig. 6: The measured differential thermal analysis of the white Kaolinite

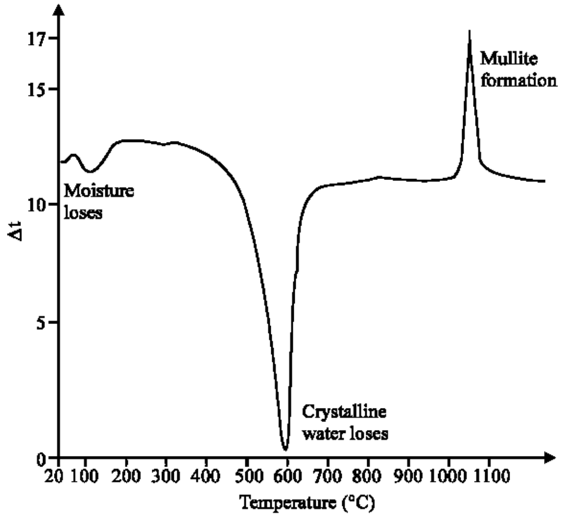

Fig. 7: The measured differential thermal analysis of the raw bauxite

Differential Thermal Analysis (DTA): The measurement of the thermal changes resulted from chemical and physical changes during heating of kaolin and bauxite is presented in Fig. 6 and 7, respectively. Figure 6 indicates that kaolin loses its crystalline water in the temperature range between 566-580°C and the mullite phase is formed at 970°C. Figure 7 shows that the bauxite loses its crystalline water at 450°C whereas mullite phase forms at 912°C.

Characterization of refractory bricks after firing:

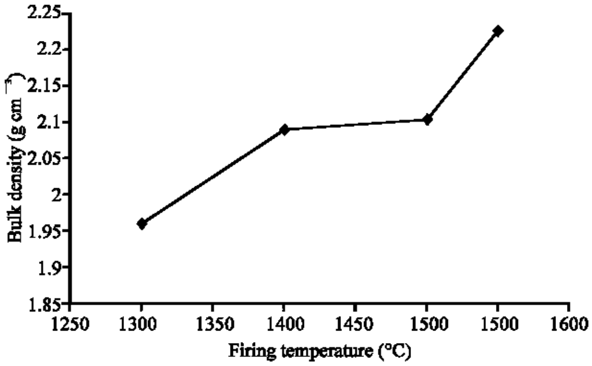

Fig. 8: The influence of the firing temperature on the on the bulk density of the bauxite bricks prepared by the addition of unfired bauxite and Kaolinite

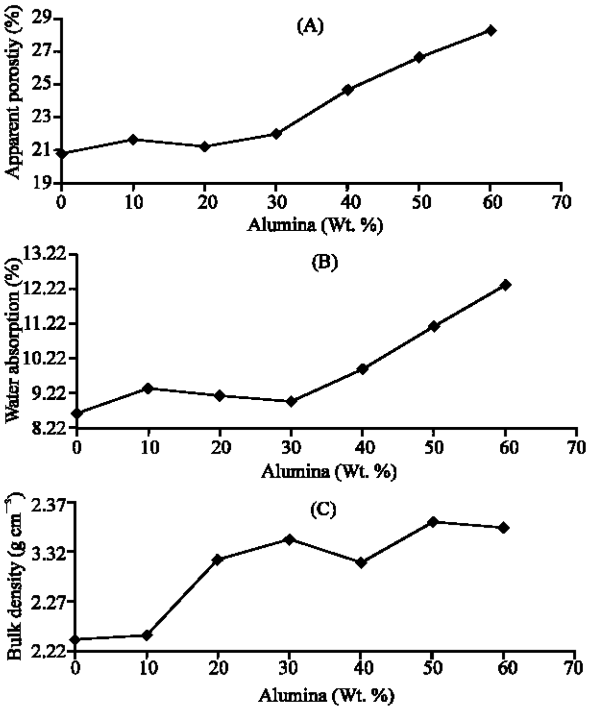

Fig. 9: The influence of the added amount of alumina on: A- Apparent Porosity, B- Water absorption and C- Bulk density of the bauxite fire bricks produced at 1555°C

Apparent density: There are several parameters influencing the apparent density such as the non-bounded materials, the firing temperature, granular size, additives and the forming pressure (Nasr *et al.*, 1988). Figure 8 presents that the density of the bauxite fire bricks increases with increasing the firing temperature resulted from the sintering process which is affected significantly by the firing temperature. Also the occurrence of the liquid phase at higher temperatures resulted from the chemical reactions taking place between the components of the bricks among them MgO, CaO, Fe_2O_3, Al_2O_3 and SiO_2. Those components contribute in formation of chemical compounds of low fusion temperature which

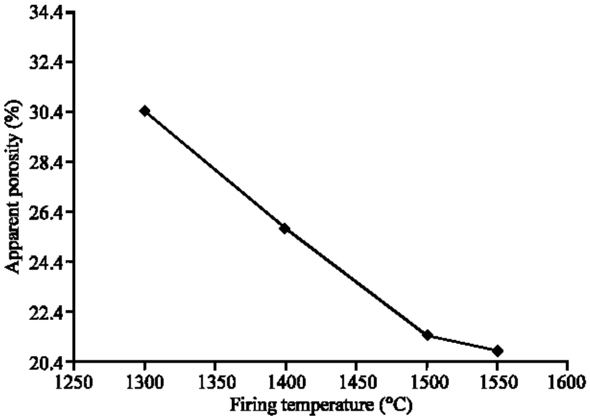

Fig. 10: The change of the apparent porosity of the bauxite bricks as a function of the firing temperature

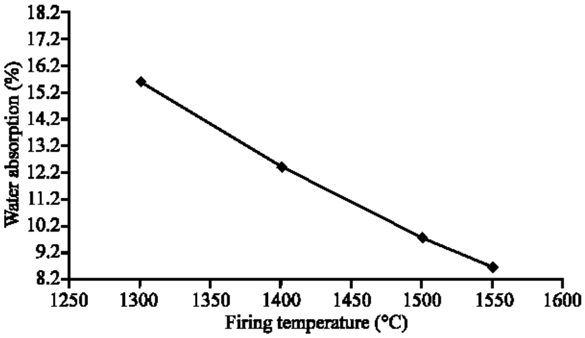

Fig. 11: The effect of the firing temperature on the property of water absorption for bauxite bricks obtained by the addition of different amounts of unfired bauxite and Kaolinite

flow easily into the into the pores resulted in an increase in the density of the bricks. Moreover, the surface tension of this liquid phase cause a compacting of the grains together causing a reduction in the dimensions of the form. The optimum density achieved which has no influence on the other properties is 2.31 and 2.33 g cm^{-3} at a temperature of 1550°C with the amount of the added aluminum oxide is 30.20% as shown in Fig. 9.

Apparent porosity and water permeability: This property is dependant on two important factors. The first factor is the influence of the sintering process while the second factor is the formation of channels and pores within the body of the brick due to the liberation f gases. The two factors have a counter effect because the reaction products resulted from the firing of the clays form a glass phase which fills the pores inside the body of the bricks. This results in reduction of the pores ratio (Junse, 1994).

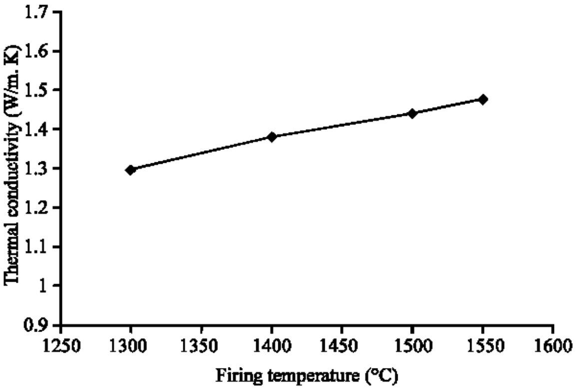

Fig. 12: Influence of the firing temperature on the thermal conductivity of the bauxite fire bricks

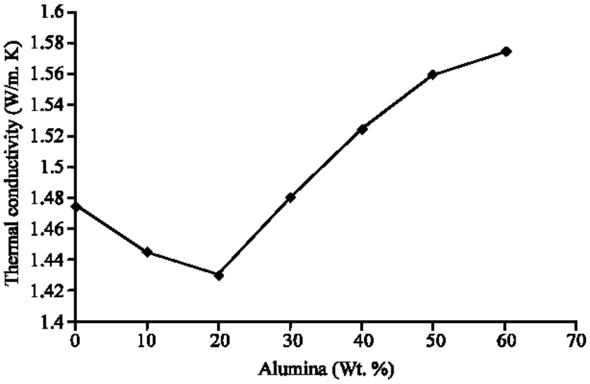

Fig. 13: The effect of the content of the added Alumina on the thermal conductivity of the bauxite fire bricks

The obtained experimental results indicate that at a firing temperature of 1300°C the porosity is reduced to (34.05-30.65%) accompanies with a reduction in the water permeability of (15.41-19.70%) Fig. 10. At a firing temperature of 1550°C, the porosity is reduced to (20.80-26.72%) and the water permeability is reduced to (8.55-14.05%). It was concluded from the experiments that the optimum properties obtained in terms of porosity and water permeability when the raw materials are composed of 70% fired bauxite, 10% unfired bauxite and 20% kaolin. At this composition when the firing temperature is raised from 1300 to 1550°C, the porosity is reduced from 30.65 to 20.80% as shown in Fig. 10 while the water permeability is reduced from 15.41 to 8.55% as presented in Fig. 11.

The influence of aluminum oxide: It was experimentally found that the amount of aluminum oxide added to the raw materials mixture influences the properties of the refractory bricks. When the aluminum oxide is added within the range (10-60%) the ratio of the liquid phase formed during firing is reduced as the amount of aluminum oxide increases. This is resulted from the decrease in the amount of melted materials in one side and the high fusion temperature (2050°C) of aluminum oxide on the other side. This property makes the aluminum oxide little influenced by high temperatures and thus will not flow within the bores. However, aluminum oxide contributes in the reaction with silicon oxide to form the mullite which improves the density of the refractory bricks.

Thermal conductivity: Thermal conductivity is an important property of the refractory bricks which determines its capability for thermal isolation especially in industrial processes operating under elevated temperatures such as rotary kiln furnaces and the furnaces used for melting of metals. The thermal conductivity of the refractory bricks plays also an important role when designing glass and ceramic furnaces.

It was experimentally observed that the thermal conductivity of the refractory bricks is significantly influenced by the chemical composition of the raw materials and the granular size as well as the method and pressure of forming (Junse, 1994). Figure 12 shows that the value of the thermal conductivity of the firing bricks increases with increasing the firing temperature. the experimental measurements depicted in Fig. 12 presents the value of the thermal conductivity was increased from 1.295 to 1.475 W/mK as the firing temperature was changed from 1300 to 1550°C. The reasons standing behind this increase in thermal conductivity are the increase of the apparent density from 1.97 to 2.231 g cm^{-3} and the corresponding reduction in the apparent porosity from 30.65 to 20.80% as the temperature was increased from 1300 to 1550°C. This phenomena resulted from the increase in sintering process and the crystal growth due to crystallization.

It was experimentally obtained that the addition of different amounts of industrial alumina influences significantly the value of the thermal conductivity as presented in Fig. 13. It can be shown in Fig. 13 that the thermal conductivity decreases up to 20 Wt% alumina due to the formation of mullite with few amounts of Corandom which has a low thermal conductivity property. After 20% Alumina in the raw materials mixture, the addition of alumina results in an increase in the thermal conductivity.

CONCLUSIONS

The suitability of bauxite as a raw material for the production of refractory bricks was tested. It was found out that the properties of the refractory bricks produced are suitable in terms of its general properties to be used for the construction of rotary kilns. The optimum operating conditions were determined based on the properties of the bricks produced after firing of bauxite. The results obtained proved that the type of bauxite used in this study is has the advantage to be used as a raw material for the production of bauxite refractory bricks since it contains low amounts of crystalline water, thus reducing shrinkage during the firing process. On the hand the bauxite used was of white color indicating higher amount of alumina and low amount of iron oxides which makes it more suitable for the production of refractory bricks with improved physical and thermal properties.

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