Characterizations of Semi-Silica Refractory Bricks Produced from Local Iraqi Materials

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ABSTRACT

This research was carried out to investigate the suitability of locally white sand and Dwaikhla clay (Kaolin) for manufacture of semi-silica refractory bricks as thermal insulators for lining the furnaces. Preliminary investigations had been done to determine the physical, chemical, thermal and mechanical properties with mineralogical constitution of raw materials by XRD analysis. Different amounts of kaolinite were added as binder to the white sand (raw materials) of semi-silica bricks. The specimens molded according to DIN standards by using semi-dry pressing method at pressure of (24.516MPa) according to the DIN1068 standards. After drying process at 110°C the specimens fired at firing temperatures (1000, 1100, 1200, 1300 and 1400) °C. The fired specimens have been investigated to determination the physical properties including bulk density, porosity, and the mechanical properties including compressive strength and thermal properties including thermal conductivity. It has been observed from the results that local white sand and kaolinite could be used as possible source raw materials for manufacture semi-silica refractories, mainly required for glass, ceramics factory in Rammadi.

Keywords: semi-silica refractories bricks, Dwaikhla clay (Kaolin, white sand.

خصائص طابوق حراري شبه سليكوني منتج من مواد عراقية محلية

الخلاصة:

تم في هذا البحث اجراء عملية تقييم للمواد الاولية (الرمل الابيض) وطين دويخلة كاؤولين من خلال اجراء الفحوصات الفيزيائية والكيميائية والمعدنية عليها لغرض معرفة مدى صلاحيتها لصناعة الطابوق الناري شبه السيلكوني الذي يستعمل كمواد عازلة في تبطين الافران المختلفة.

تم استعمال الكاؤولينايت كمادة رابطة تضاف الى المادة الاولية لصناعة الطابوق الناري شبه السيلكوني وبنسب مختلفة.

استخدمت طريقة الكبس شبه الجاف لتشكيل نماذج اسطوانية الشكل حسب المواصفات الالمانية (DIN) وكان ضغط التشكيل (24.516 MPa.) حسب المواصفة الألمانية (1068) DIN بعد عملية التجفيف في درجة حرارة 110مئوية , تم حرق النماذج بدرجات (1000, 1100, 1200, و1300 1400) درجة مئوية.

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بعد عملية الحرق تم اجراء الفحوصات لتحديد خواص النماذج المحروقة مثل الكثافة الكلية,المسامية الظاهرية,المقاومة الميكانيكية والموصلية الحرارية.
تشير النتائج الى صلاحية المواد الأولية المستعملة لصناعة الحراريات شبه السليكوني والتي يمكن أستعمالها في تبطين أفران صهر الزجاج ومعمل السيراميك في الرمادي.

INTRODUCTION

Refractory materials may be defined as materials that retain their physical and chemical identity when subjected to high operation temperatures. These materials are non-metallic compounds that can withstand high temperatures fluctuations between (1000 and 1500) °C [1,2]. They are also good thermal and electrical insulators used for metallurgical industry in the construction of furnaces as an internal lining. They are commonly used as a vessels for holding and transporting molten metals and slags, and a flues or stacks through which hot gases are conducted [3].The most common refractory materials are located in the (SiO_2-Al_2O_3) system, and usually called aluminosilicate, they are distinguished by their alumina content [4] .The melting points of SiO_2 and Al_2O_3 are high,1728 and 2050˚C respectively. Phase diagram of silica- alumina is shown in fig.(1). Above 1400˚C there are three stable crystalline compounds in the system: cristabalite a high temperature from of silica, corundum, and mullite. Below eutectic temperature (1580 ˚C), the equilibrium phases present are mullite and silica (cristobalite or tridymite). During refractory application, the presence of a small amount of a liquid phase may be allowable without compromising mechanical integrity. Above 1580°C the fraction of liquid phase present will depend on refractory composition. Upgrading the alumina content will increase the maximum service temperature (i.e. refractoriness), allowing for the formation of a small amount of liquid [5]. Semi-silica bricks are another type of silica refractory bricks. This type of refractory is an intermediate between fire clay and silica refractory bricks. It does not have the large shrinkage of fire clay bricks and the low thermal shock resistance of the silica refractories in certain temperature ranges. Due to the fact that they are self-glazing with a high viscous glass, they are highly resistant to slag, attack. Semi-silica bricks are widely used in open hearth furnaces. The important types of refractories in this diagram are andulusite, silica refractory, semi-silica refractory, fire clay, mullite, high alumina refractory and alumina refractory [7].

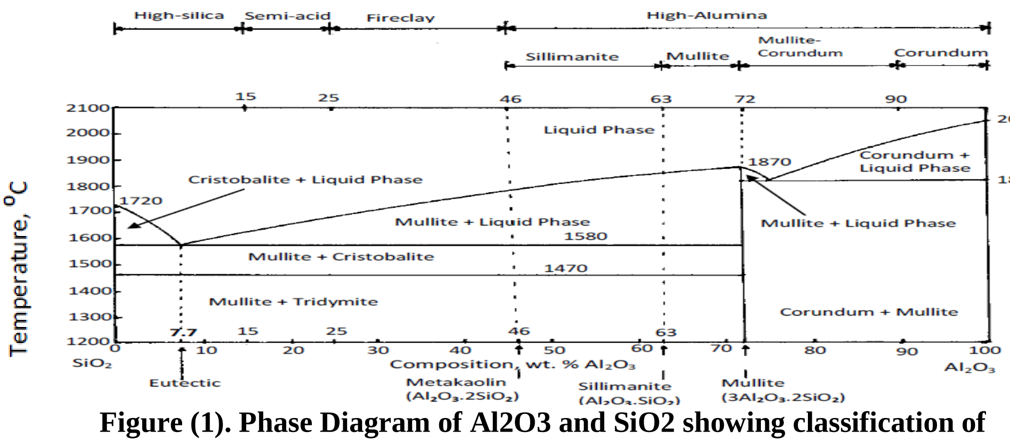

Figure (1). Phase Diagram of Al2O3 and SiO2 showing classification of aluminosilicate refractories [6].

Experimental Part

Raw Materials Investigation

The raw materials (white sand and Dewechla kaolin) are used in this study and obtained from Iraq Geological Survey.

The Chemical Analysis

Table (1) shows the chemical analysis of the raw materials which are used in our development work. It has been indicated that the Dewechla clay was suitable for manufacturing of refractory bricks.

Table (1). The chemical analysis of Kaolin and White sand.

Oxides / Raw type wt%	SiO_2	Al_2O_3	Fe_2O_3	K_2O	Na_2O	MgO	CaO	TiO2	P2O5	Cl	SO3	L.O.I
Kaolin wt %	48.5	34.21	1.33	0.4	0.2	0.23	0.13	1.4	0.01	0.08	0.07	13.54
White sand wt%	97.8	0.56	0.17	0.01	0.02	0.066	0.11	0.08	0.01	0.07	0.07	1.034

This test has been carried out in Iraq Geological Survey.

2.1.2 The Mineralogical Investigation.

All samples obtained from the fired batches were finely grounded, and scanned using X-rays diffraction (type) at room temperature with Cu Kα, and scanning speed of (10deg/min) from 5-45°, and (40)KV as an applied power to estimate the crystallized phases. Charts of raw materials are show in Figs.(2),(3) ,this test was carried out in the Iraq geological survey.

2.2 The Preparation of Specimens

For molding the specimens, the Kaolin rocks must be crushed, grinded and screened. The maximum particle size of milled Kaolin is (- 452)µm, the milled fractions were mixed with white sand in different ratio $\frac{sand}{Kaolinite}$ $(\frac{60}{40}, \frac{65}{35}, \frac{70}{30})$ according to the binary system of SiO_2 and Al_2O_3 diagram. The amount of water (9-12) wt% and Molasses (0.5) wt% were added to the mixture of batches. The green specimens were pressed by the semi-dry method using a hydraulic press and a molding pressure of (24.516) MPa with cylindrical shape (30 mm diameter and 30 mm high) according to DIN 1068 [7]. After molding and drying the specimens were fired at (1000,1100,1200,1300 and 1400)°C. Finally the fired samples were investigated to evaluate its properties such as bulk density, apparent porosity, compressive strength and thermal conductivity.

2.2.1The Investigations

2.2.1.1Bulk Density (BD) and Apparent Porosity (AP)

DIN 51056 was used to determine the apparent porosity (AP), bulk density (BD) of the samples. The percent of open porosity is calculated as follow,

% Apparent Porosity = [(MW–MD)/(MW–MH)]*100 …. (1)

where, MW is the wet weight (g), MD is the dry weight (g), and finally, MH is the immersed weight (g). According to the DIN 51065 standard, the density of the refractory is calculated as follow,

BD = [(MD) / (MD– MH)]*ρw ….(2)

where BD is the bulk density (g/cm3) and ρw is the density of water($\frac{g}{cm^3}$) [8].

2.2.1.2 Compressive Strength (CS)

The CS of each specimen was determined according to DIN 51067 standard.This is done by using device type gunt HAMBURG model WP300.20 with 20 kN capacity. This test has been measured by placing a specimen of refractory in the flat surface in the device which in cylindrical shapes According to the DIN 51067 standard, the CCS is calculated as follow,

σccs= Fmax/Ao ….(3)

Where:

σccs= cold compressive strength ($\frac{kg}{cm^2}$), Fmax= force at the fracture (kg).

Ao = surface area (cm^2) [8].

2.2.1.3 Thermal Conductivity

It is ability of the refractory to conduct heat.[9,10].Specimens have been prepared with the dimensions(41X10) mm according the standard specifications of the Lee's disk instruments. The value for the thermal conductivity of the specimen is calculated ,as follows:

$$e = \frac{VI}{a_A T_A + a_S (T_A + T_B/2) + a_B T_B + a_C T_C} \quad \text{…. (4)}$$

$$K = \left(\frac{ed}{2\pi r^2 (T_B - T_A)}\right)\left[a_S\left(\frac{T_A+T_B}{2}\right) + 2a_A T_A\right] \quad \text{...(5)}$$

Where :VI:Thermal energy passing through coil after reaching thermal equilibrium, e: Amount of energy passing through material disk at unit area for each second (W/m^2.second), a_A, a_B, a_C, a_S and a_H are the exposed surface areas of A, B, C, the specimen and heater, respectively. Areas a_A and a_C include the flat ends of the disks. T_A, T_B and T_C are the temperatures of the disks A,B and C,d $_{A, B, C, S}$: Disk thickness, (m), r: Disk radius,(m), s: Specimen, *K*: Coefficient of thermal conductivity [9].

2.3 Discussion of Results:

2.3.1 Chemical Analysis:

The suitability of kaolinite for refractory work depends primarily on its sintering and softening point. In this respect the chemical analysis is of great importance. Usually, refractory clays are classified by their alumina content. The TiO2 content in clay used for manufacture of refractory bricks normally ranges between 1 and 4%.The content of iron oxide (Fe_2O_3) which acts as a flux should not exceed 2.5%.The total flux materials alkalis (Na2O+K2O), and alkalines (CaO+MgO) not exceed 6% [10].

2.3.2 Mineralogical Analysis

XRD results of raw materials kaolin and white sand are shown in fig. (2) and (3) that kaolinite contains kaolin and quartz. Whereas quartz is the main phase of white sand as shown from peaks in fig. (3).

2.3.3 Bulk Density (BD)

Information about bulk density is essential for the calculation of the heat storage capacity and for the evaluation of porosity. Usually, bulk density is related to chemical composition but is mainly a function of the method of manufacture. According to the DIN 51053, the bulk density of wet pressed refractory bricks should vary between 1.9 and 2.0 $\frac{gm}{cm^3}$ semi-dry pressed bricks have a bulk density of more 2.0. In our work, considerable variations in density were observed depending on the firing temperature and the silica/kaolin ratio.Bricks made from silica/kaolinite $\frac{70}{30}$ and fired at 1400°C had a comparatively low bulk density (2.006$\frac{g}{cm^3}$). The highest bulk density was achieved on bricks with silica/kaolin $\frac{60}{40}$ and firing temperature of 1400°C (2.274$\frac{g}{cm^3}$). Table (2).This can be explained by the presence of Al_2O_3 in the kaolin which was added to the silica sand. During firing, kaolin clay is converted to mullite and silica

$$3Al2O3+4SiO2 \xrightarrow{1470°C} 3Al2O3.2SiO2+2SiO2.$$

The bulk density of produced bricks increased with increasing quantity of mullite and indirectly with the content of Al_2O_3in the kaolinite. The last mentioned bricks meets the requirement according to the DIN standard as far as bulk density is concerned. A relationship was found between bulk density and firing temperature, Fig (4). From the results the values of bulk density to all samples increased with increased fired temperatures [11], this due to continuously reaction to phase crystal formation specially mullite crystal and this interpenetrate with each other due to liquid sintering mullite in high temperatures cause to reduce the porosity of the refractory production and that increased the values of bulk density, the increases in density of a refractory lead to decreases of the porosity and vice versa as shown in fig. (4). And the increased porosity will lower the bulk density [12].

2.3.4 Apparent Porosity (AP)

Porosity is of great influence on cold compressive strength, gas permeability and resistance against slags-metals and glass melting. Compressive strength can be increased substantially by increasing the sintering temperature. Dense bricks have lower gas permeability than less dense ones and high porosity normally leads to high values of gas permeability. Infiltration of foreign material increases with size of the pore space. Therefore, the DIN standard required that dense bricks have a porosity of between 15-25%.The data presented in table (2) indicated that the bricks tested are in line with the requirements of this standard. Fig. (5) shows that the porosity in reduced substantially with increasing temperature [13].

2.3.5 Compressive Strength

The compressive strength of refractory bricks at room temperature is of great importance. These properties are indicated for the quality of the load and the possible existence of structural faults. Bricks produced in our research work with silica/kaolin ratio $\frac{70}{30}$, and firing temperature of 1400°C had the lowest compressive strength, table(2). The compressive strength of bricks produced from this mix was only (27.007) MPa. On other hand, the highest compressing strength was reached with mix sand/kaolinite ratio $\frac{60}{40}$ firing at 1400°C (29.879) MPa. In most cases compressive strength drops with increasing porosity. Chemical reactions between refractory and furnace charge, slag and molten glass and metals as well as with dust and vapors generated in the kiln are the main reasons for wear on refractory linings. This is frequently combined with infiltration of liquid phase into the refractory with results that the mechanical properties in particular the resistance against temperature change are altered. The strength of bricks produced from this mix is well above the minimum requirements of the standard. In general, increasing firing temperature up to 1400°C are beneficial for the compressive strength on the material investigated may be obtained by further increasing firing temperature. It has been observed the values of cold compressive strength for all specimens ratio increased with the raising fired temperatures [14]. Fig.(6).

2.3.6Thermal Conductivity

As mentioned the thermal conductivity of solid bricks can be lowered by the introduction of air space (pores).The lower coefficient of heat transfer is due to the fact that the conductivity of air is only one hundredth (0.02$\frac{Kcal}{m.h.c^{\circ}}$ of the conductivity of the solid phase(2.0$\frac{Kcal}{m.h.c^{\circ}}$).The conductivi- ty of refractory bricks must be expected to be between these two extremes, depending on the volume fraction of air in the materials. The reasons standing behind this increase in thermal conductivity are the increase of the density and the corresponding reduction in apparent porosity as the temperature was increased this phenomena resulted from the increase in sintering process and the crystal growth due to crystallization [15].

CONCLUSIONS

1-The chemical analysis indicated that the Dwaikhla clay and white sand are suitable for manufacture of semi-silica bricks.

2-The clay is used as a binder to facilitate the molding process of the semi-silica bricks, because the sand is non plastic materials.

3-The most suitable process for molding of this type of refractory bricks is the semi-dry pressing.

4-Optimuim performance was obtained when using a mix with a sand/kaolinite ratio of 70/30 and water content between 9-12%. Suitable compaction was achieved by a molding pressure of 24.516MPa.

5-Optimuim firing temperature was 1400°C.

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Table (2). Results of investigations for fired specimens at firing temperature 1000-1400 °C.

The Raito of samples	The result of Bulk density at 1000-1400°C firing temperature				
	1000°C	1100°C	1200°C	1300°C	1400°C
70%M1+30%S	1.896	1.951	1.976	1.93	2.006
65%M2+35%S	1.943	1.969	1.988	2.014	2.125
60%M3+40%S	1.978	1.991	2.167	2.256	2.274
	The result of Apparent porosity at 1000-1400°C firing temperature				
70%M1+30%S	24.246	22.964	22.233	20.435	18.759
65%M2+35%S	22.818	21.725	19.594	17.663	15.961
60%M3+40%S	21.734	20.502	18.959	16.961	14.898
	The result of CCS in MPa. at 1000-1400°C firing temperature				
70%M1+30%S	12.745	13.578	14.818	17.595	27.007
65%M2+35%S	13.116	14.189	15.634	17.871	28.583
60%M3+40%S	15.672	16.274	16.757	18.632	29.879
	The result of thermal conductivity at 1000-1400°C firing temperature				
70%M1+30%S	0.614	0.639	0.648	0.655	0.717
65%M2+35%S	0.628	0.633	0.657	0.661	0.735
60%M3+40%S	0.689	0.697	0.735	0.752	0.766

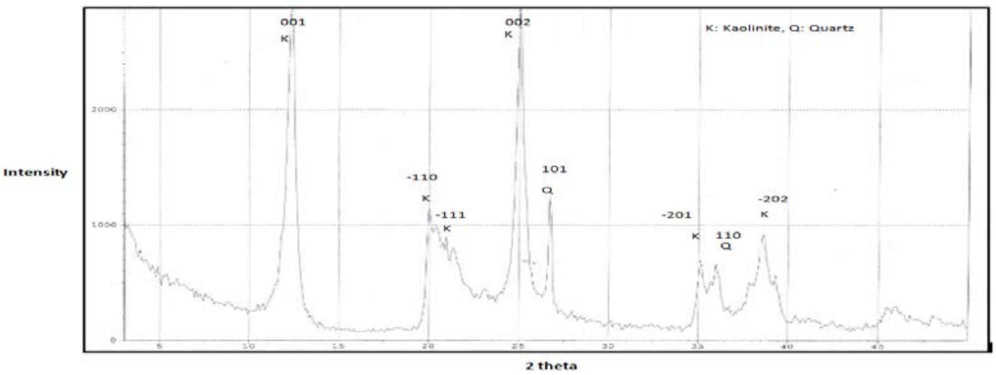

Figure (2). XRD of kaolinite raw material.

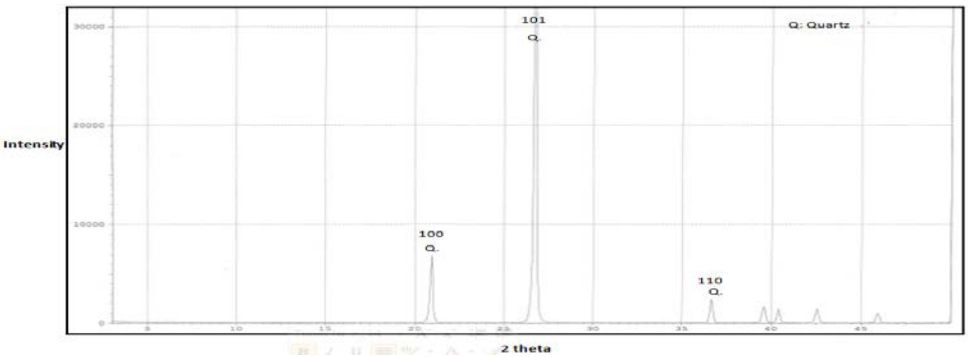

Figure (3). XRD of white sand raw material.

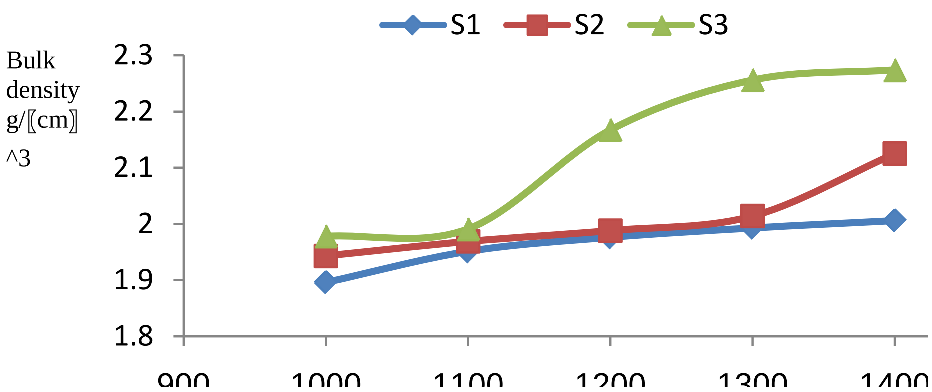

Figure (4). Relationship between bulk density and firing temperature.

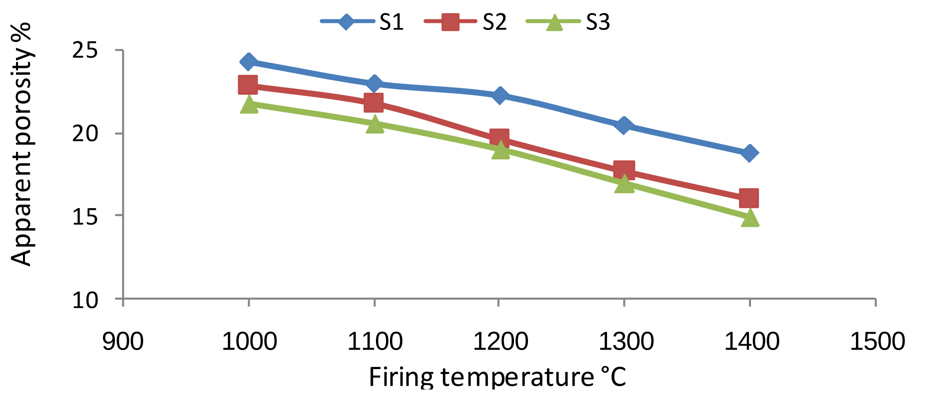

Figure (5). Relationship between Apparent porosity % and firing temperature.

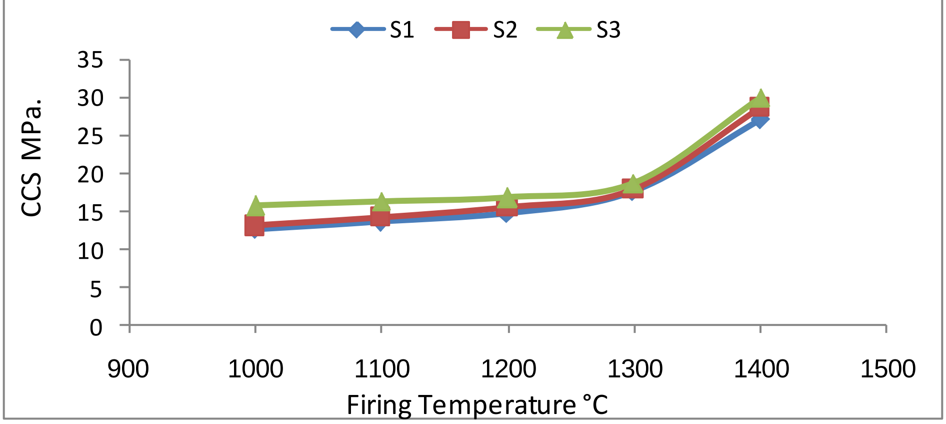

Figure (6). Relationship between CCS and firing temperature.