

MANUFACTURE OF REFRACTORY BRICK FROM LOCALLY AVAILABLE RED CLAY BLENDED WITH WHITE PORTLAND CEMENT AND ITS PERFORMANCE EVALUATION

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ABSTRACT: In this paper, refractory bricks were developed by using locally obtainable materials like red clay and glass sand in Bangladesh. The Preliminary investigation was conducted by measuring the compressive strength of bricks produced by adding a range of materials with red clay in different proportions. It was found that the brick containing 30% white Portland cement and 10% glass sand with red clay (60%) demonstrated the highest compressive strength (36.20 mpa). The suitability of refractory bricks was evaluated by examining their physical and thermal properties. The evaluation of these parameters revealed that the properties are suited with refractory requirements such as low water absorption (4.58%), moderate apparent porosity (13.32%), preferable bulk density (2.64 gm/cm³), less linear shrinkage (3.10%), better refractoriness (1408 °C), thermal conductivity (1.3 W/m.K) and thermal shock resistance (15 cycles). The mineralogical analysis carried out by XRD exhibits the presence of Quartz and Bavenite phase. The performance evaluation shows that this brick is able to fit most of the criteria of refractory brick needed for practical purposes. Since local materials were utilized, the proposed method can be used for the manufacture of low-cost refractory brick in Bangladesh.

Keywords: Refractory brick, Local material, red clay, Compressive strength, Optimized composition

1. INTRODUCTION

Due to the rapid industrialization and urbanization, the brick industry is expanding quickly in many parts of Asia including Bangladesh. Refractories are mineral and chemical-based materials with very high heat-resisting properties which make them ideal for the use in construction of oven, furnace walls and ceilings in various industries such as metallurgical, chemical, cement, glass and petrochemical industries [1]. These materials are supposed to possess several qualities such as resistance to heat, exposed to different degrees of mechanical stress as well as strain, corrosion from liquids and gases, and mechanical abrasion at high temperature. The purpose of using refractory bricks in the lining of kilns is to keep the heat inside and prevent its infiltration into the outside air. During the design of the kiln, it has to be kept in mind that the temperature at the outside surface of the kiln does not exceed 40°C whatever the internal temperature might have been. Secondly, the brick should prevent the outside shape of the kiln from melting or folding because it is normally constructed out of steel. Furthermore, it aids in building a suitable working atmosphere for the technician and

engineers to view and observe the processes that are taking place inside the kiln [2,3].

Refractory bricks can be produced from ceramic materials with high temperature kiln firing [4,5]. Refractory materials can be categorized on the basis of the following characteristics such as; size, method of manufacture, shaped products, degree of porosity and type of heat treatment [5]. Refractory materials can also be classified on a chemical basis as basic, neutral and acid products. The more silica is used in a mixture the more acidic are its properties [2,6].

Most of the refractories are made from naturally occurring high melting point oxides- SiO₂, Al₂O₃, MgO, Cr₂O₃, ZrO, and refractory materials include aluminosilicate, dolomite, magnesium oxide, silica, chrome, chrome-magnesite, carbon. One of the best-known refractory material is the fireclay which belongs to the aluminosilicate group of refractory materials [7,8]. Clay is a natural origin, complex mixture, which varies in composition depending upon the geological location [9]. Abundant deposits of clay are available all over the world. Although the composition of clay material can vary based on location, the major components are silica (45-60%), alumina (10-20%), iron oxides (within

10%), CaO (~10%) and other oxides (MgO, Na₂O, K₂O) are in small proportion [10]. Clay mineral groups are kaolin, smectite, palygorskite-sepiolite, illite, chlorite, and mixed-layered clays [11]. The properties of these clays vary in their structure and composition. However, most of the clays are not suitable for the production of refractory materials in as-mined states because the properties of these bricks do not meet the criteria for good refractory bricks. The desired properties of fireclay refractory brick can be attained by partial replacement of clay with other materials such as industrial waste [12], agricultural waste [13], magnesia [14] coal ash [15] and cement [16].

Although, refractory bricks are produced in many countries using their locally accessible clays with some other materials as a partial replacement, the report on the utilization of such materials in Bangladesh for refractory brick production is scarce. Therefore, most of the local demands of refractory bricks are met through the import from other countries. To ensure sustainable economic growth, Bangladesh must develop refractory products using indigenous raw materials.

Abundant deposits of red clay are available in Bangladesh. Besides, glass sand also exists in the quarries of Bangladesh. Herein, this work was aimed to develop refractory brick using locally sourced red clay and glass sand. In addition, white Portland cement was used as a binder. The suitability of brick as refractory materials was determined by investigating mechanical, physical and thermal properties.

2. MATERIALS AND METHODS

This work was undertaken to produce refractory bricks from red clay as a primary ingredient. Different mixtures were prepared containing additives to produce refractory bricks fired by electrical muffle furnaces. The details of raw materials, mix proportioning, preparation of specimens, firing stages and experimental works are discussed in the sections below.

2.1 Raw Materials

2.1.1 Red clay (RC)

In this work, RC was collected from Belabo upazila under Narsingdi administrative district (24°06'17.1 N and 90°51'09.3 E) of Bangladesh. The composition of RC according to chemical analysis is shown in Table 1. It can be concluded that the major components are silica, calcium oxide and aluminum oxide. Magnesium oxide and other oxides like iron, potassium and sodium are presented in small quantity. The red color is due to the presence of Fe₂O₃ and the intensity of the color is depended on the percentage of Ferric oxide. The low value of loss of ignition indicates the presence of less organic matter. The mineralogical analysis

of red clay performed by XRD is presented in Fig. 1 which exhibits the existence of quartz (COD#96-101-1160 [17]) and chloritoid (COD#96-900-0774 [18]) phase.

Table 1 Chemical composition of red clay and glass sand

Constituent	Red clay (wt.%)	Glass sand (wt.%)
Silica (SiO ₂)	49.98	99.05
Calcium Oxide (CaO)	17.08	0.07
Iron (III) Oxide (Fe ₂ O ₃)	2.85	0.02
Magnesium Oxide(MgO)	3.36	0.13
Aluminum Oxide (Al ₂ O ₃)	16.39	0.22
Potassium Oxide (K ₂ O)	0.33	0.05
Sodium Oxide (Na ₂ O)	0.11	0.11
Loss on Ignition (L.O.I)	5.54	0.15

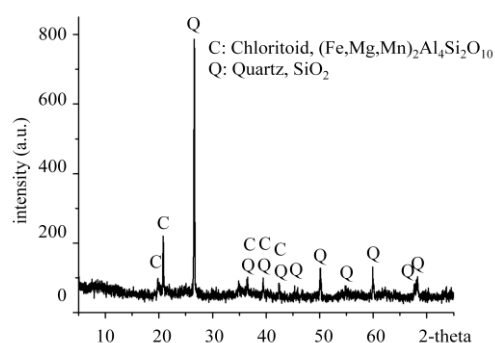


Fig.1 XRD pattern of Red clay showing the existence of a mixture of crystalline two phases-quartz and Chloritoid

2.1.2 Glass sand (GS)

Glass sand is a granular material composed of finely divided rock and mineral particles. GS used in this study was collected from Chaudagram Upazila, Cumilla district (23°13'41.4"N 91°18'51.2"E) of Bangladesh. The composition of glass sand can be varied depending on the local rock sources and conditions; nonetheless, the most common constituent of glass sand is silica (silicon dioxide, SiO₂). The chemical composition of glass sand used in this study is presented in Table 1 which shows that almost the entire portion of sand is silica (99.05%) and other components are presented in very small quantity [19].

2.1.3 White Portland cement (WPC)

WPC was used in combination with red clay and glass sand as a binder to produce refractory brick. White Portland cement was obtained from Siam Bangla Industries located in Khaliapara,

Kaliganj, Gazipur (23°56'48.2"N 90°36'55.8"E) of Bangladesh. The chemical analysis of WPC shows the composition as CaO (61.65%), SiO₂ (19.43%), Al₂O₃ (10.10%), Fe₂O₃ (3.17%) and other oxides are MgO, SO₃, TiO₂, K₂O, Na₂O. The mineralogical analysis performed by XRD depicted in Fig. 2 shows the presence of calcite (COD# 96-210-0993 [20]) and alite (COD# 96-154-0705 [21]) phase.

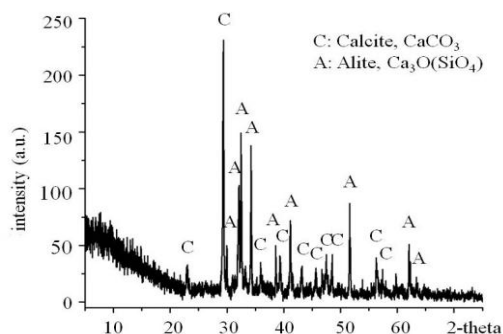


Fig.2 XRD pattern of White Portland Cement showing the existence of a mixture of crystalline two phases-Calcite and Alite

2.1.4 Slaked lime

Slaked lime was added as a ratio of 2% by weight as a mineralizer in refractory bricks specimens. Lime was completely slaked before use and approximately 94.57% of lime was found in slaked lime. In addition, Polyvinyl alcohol (PVA) was used for the resistance to cracking during drying and mechanical resistance of the refractory brick (5%).

2.2 Production of Brick Prototype

After collection, raw materials were washed with water to remove suspended organic matter and then dried. The dried materials were crushed into powder followed by sieve analysis. The raw materials were sieved into three particle size fractions referred to as the coarse, medium and fine fractions. The size of the coarse, medium and fine particles was 2000, 710 and 212 μ m, respectively. Uniform packing density was obtained by optimizing the size gradation as 60% coarse: 10% medium: 30% fine particles [6]. The constituents were then taken according to proportions and mixed properly in a dry state followed by adding the required quantity of water to get proper consistency. Initially, the cement mortar (cement + sand) was prepared by maintaining the water-cement ratio (W/C) of 0.45 and then the red clay was blended with a mortar and slaked lime. Water in slaked lime creates the required consistency for Clay-Mortar mixing. Finally, PVA was added for proper shaping. The

mixture was molded using a hydraulic press where the pressure was maintained at 200 kg/cm². The pressed samples were air-dried, followed by oven-dried at 110 °C for 8 hours and finally fired at 1400 °C in a muffle furnace. The firing was controlled at a steady rate of 5 °C/minute to 1400°C and kept at this temperature for 2 hours. The specimens were then allowed to cool gradually inside the furnace overnight. The brick prototype was made in cube-shaped and the dimensions of test specimens were 2×2×2 inches. The whole process is presented sequentially in Fig. 3.

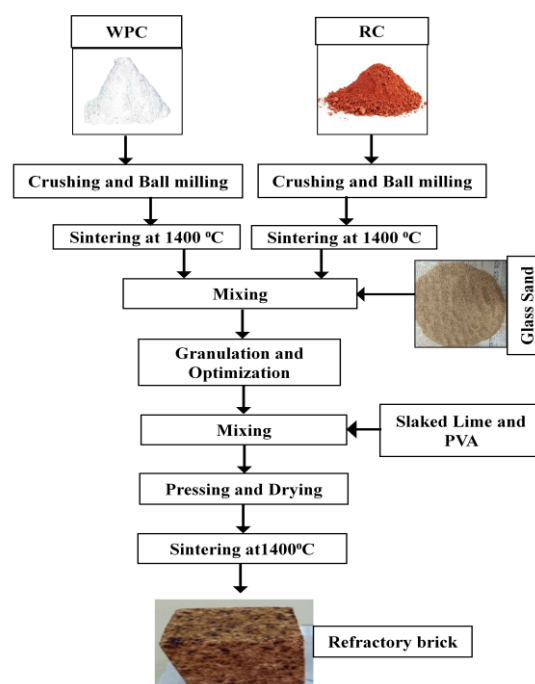


Fig.3 Schematic representation of the experimental program to produce refractory brick

2.3 Characterization

Composition of raw materials and brick was analyzed by using chemical method described in American Society for Testing Materials, ASTM C114 [19]. Loss on ignition (L.O.I.) was measured by heating the sample at 1000 °C until constant weight using a muffle furnace. Size gradation was performed by sieve method. Compressive strength and tensile strength (MOR) were measured according to the method described in ASTM C133-97 (2003) [22] using hydraulic press (Hydraulic press: CARVER laboratory press, model no. C25576, USA) and Hounsfield UTS 10 KN (model: H10KS Hounsfield, UK) respectively. Bulk density and apparent porosity were examined by boiling method according to ASTM C20-00 [23]. Linear shrinkage was measured following the method described in ASTM C179-14 [24].

Thermal conductivity of the samples was measured by Lee's Disc Apparatus. The Pyrometric Cone (Segar cone 30) equivalent (PCE) was used to measure the refractoriness as explained in ASTM C24-89 [25]. Thermal shock (cycles) was measured by ASTM C1525-18 [26]. X-ray powder diffraction (XRD) data was collected by Rigaku Desktop Miniflex II X-Ray Diffract meter equipped with Ni filter and Cu K α radiation ($\lambda=1.54056\text{\AA}$, serial no:HD20972, Rigaku Corporation, Tokyo, Japan) at a scanning rate of $5^\circ/\text{min}$ from 2θ range 05 to 80° . Phases are identified by using program the Match! [27] and Crystallographic Open Database (COD, release March 2020) as reference database.

3. RESULTS AND DISCUSSION

3.1 Compressive Strength (CS)

Compressive strength can be considered as a useful indicator of the efficacy of firing and abrasion resistance according to other properties like bulk density and apparent porosity. Preliminary investigation of refractory brick produced from red clay with other materials such as limestone, glass sand and white Portland cement as a partial replacement of red clay was conducted by measuring compressive strength. The results as shown in Table 2, the compressive strength varies with both the type of materials and its' composition. The addition of glass sand and lime stone with red clay in various ratios did not significantly improve CS. While the combination of white Portland cement and sodium carbonate with red clay was slightly effective, the addition of white Portland cement and glass sand with red clay offered the superior result in comparison to other combinations. Among the various compositions of red clay, glass sand and white Portland cement evaluated, it was demonstrated that red clay (60%), glass sand (10%) and White Portland cement (30%) gave the highest value of CS. The highest value was obtained 36.20 mpa which is well above the minimum CS for refractory materials of 15 mpa [15,28,37]. The presence of silica content in the glass sand which acted as a fluxing agent during sintering may have caused the higher strength of refractory brick [29,30].

The quartz modifications during sintering have relatively closed packed structures. On the other hand, the action of lime (CaO) from WPC act as a mineralizing agent and formation of the strong bond is due to the diffusion of calcium ion on into silica surface during sintering. The phase diagram of lime-silica system shows the considerable amount of lime that may be used to bond quartz in refractory brick without loss of refractoriness.

The permissible compressive strength of refractory brick should be 24.51 mpa at ambient

temperature, should not be less than 34.47 mpa for the arch of the furnace and 16.55 mpa for the bottom of the furnace. Therefore, the laboratory made refractory brick of CS 36.20 mpa is suitable for the above mention purpose.

The tensile strength of blank brick (100% red clay) and refractory brick with optimized composition (RC(60%) + GS(10%) + WPC(30%)) was also measured and it was found that tensile strength is significantly increased in optimized brick (14.91 MPa) compared to blank brick (8.96 MPa).

Table 2 Compressive strength of brick prototypes prepared from various components and compositions

Sl. No	Composition (%)	Compressive strength (mpa)
1	R(100%)	19.31
2	RC(60%) + GS(10%) + LS(30%)	20.00
3	RC(70%) + GS(20%) + LS(10%)	26.72
4	RC(60%) + GS(30%) + LS(10%)	21.72
5	RC(50%) + GS(40%) + LS(10%)	27.23
6	RC(60%) + WPC(40%)	21.37
7	RC(50%) + WPC(50%)	25.86
8	RC(60%) + GS(10%) + WPC(30%)	36.20
9	RC(50%) + GS(20%) + WPC(30%)	34.82

3.2 Water Adsorption and Apparent Porosity

Water absorption test on bricks is performed to determine their durability properties such as degree of burning quality and behavior of bricks in weathering. Less water penetration in bricks is expected as it enhances the durability of bricks and resistance to the natural environment [31]. The water absorption is directly related to the apparent porosity in brick and increases with the increase of pore volume [32]. The water absorption and apparent porosity of prepared bricks were measured varying the proportion of WPC and results are presented in Fig. 4.

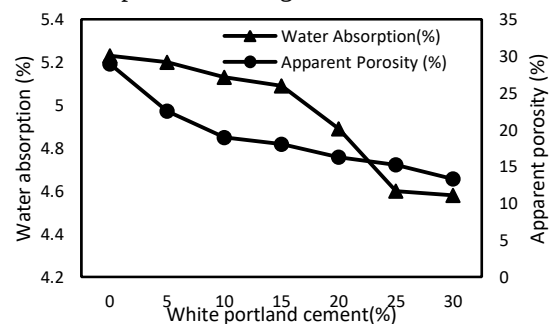


Fig.4 Variation of water absorption and apparent porosity with the proportion of WPC in brick sintered at 1400 °C

It can be pointed out that both the apparent porosity and water absorption were decreased with the increase of WPC percentage. The lowest value of apparent porosity and water absorption were found 13.32% and 4.58%, respectively for the brick specimens where 30%WPC was added. Nonetheless, the highest value of apparent porosity and water absorption were 28.94% and 5.23%, respectively for the prototype with 0% WPC. This value of water absorption with 30%WPC is below the ASTM C 20-00 [23] recommended maximum value ($\leq 18\%$). The obtained apparent porosity is also within the guideline value of 22-26% according to Orumwense and Nwajagu [33]. The WPC and GS act as bonding materials with RC at high temperatures and reduce the surface pores of the brick. The decrease in porosity reduces the volume of brick at the same time improves the mechanical properties [34].

3.3 Linear Shrinkage

Linear shrinkage is the change in linear dimension that has occurred on the brick sample before and after firing. Low shrinkage is preferable for the application of brick in heat treatment furnace. The linear shrinkage of brick produced from RC with GS and varying percentage of WPC (0 to 30%) was investigated and the results are shown in Fig. 5. The results explain that the linear shrinkage of blank sample (100% RC) was 1.11% (not shown) which was increased to 1.71% for 0% WPC, 90%RC and 10%GS and then gradually increased with the rise of WPC percentage. However, the linear shrinkage of brick with 30%WPC was 3.10% which is less than the recommended range of 7% - 9% for refractory bricks [35]. The slight increase of linear shrinkage can be attributed to the fact that when WPC is added, it forms bond strongly during the firing process which causes the removal of pores and inter-granular spaces [15].

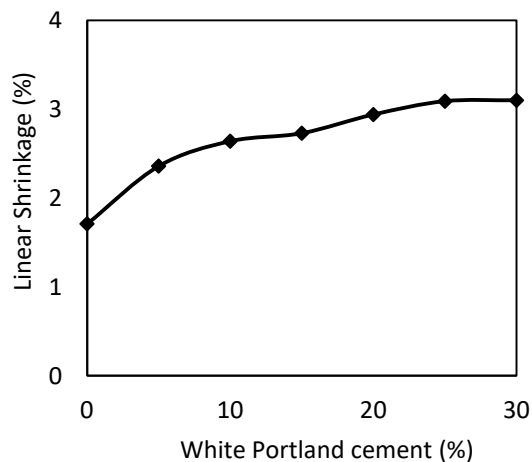


Fig.5 Variation of Linear shrinkage with WPC composition in brick samples sintered at 1400 °C

3.4 Bulk Density

Bulk density measurement was performed for all sintered samples and the results are shown in Fig. 6. It has been observed that the incorporation of WPC by the partial replacement of red clay results an increase in bulk density. The increase in the bulk density with increasing WPC could be due to the fact that during sintering, the calcium oxide of WPC creates a calcium silicate phase by reacting with the active silica of glass sand.

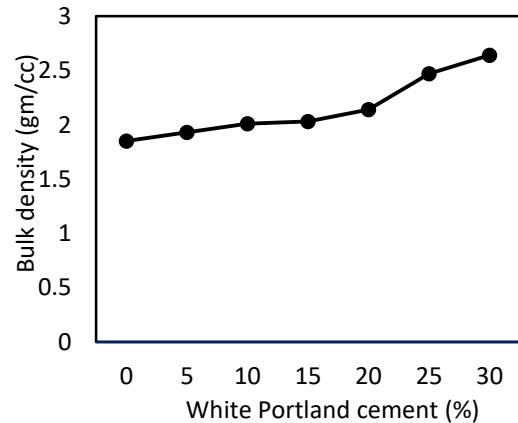


Fig.6 Variation of Bulk density with WPC composition in brick samples sintered at 1400 °C

This active silica has reacted with the surrounding material more effectively which further eliminates the spaces between granules and micropores leading to more packing efficiency and better densification during sintering [13].

3.5 Thermal Properties

Thermal properties like refractoriness, thermal shock resistance and thermal conductivity of bricks prepared from 100%RC and some combination of RC with WPC and GS were explored and the results are presented in Table 3.

Table 3 Thermal properties of prepared refractory bricks produced with different composition

ID	Brick composition	Refractoriness (°C)	Thermal Shock resistance (cycles)	Thermal Conductivity (W/m.K)
A	100% RC	1054	9	2.3
B	RC(60%) + WPC(40%)	991	11	2.7
C	RC(50%) + WPC(50%)	983	10	3.1
D	RC(60%) + GS(10%) + WPC(30%)	1408	15	1.3
E	RC(50%) + GS(20%) + WPC(30%)	1359	13	1.1

3.5.1 Refractoriness

Pyrometric Cone Equivalent (PCE) was employed to test for refractoriness as stated by [26,36]. Each of samples A, B, C, D and E were used to compose a self-made pyrometric cone of standard composition for 1050, 1100, 1200, 1300,

1400 and 1500 °C as described by Chesters [37] of 1.16 cm base diameter. The cones were put in a furnace with a monitoring thermocouple and heated to a high temperature above 1000 °C. Table 3 shows the refractoriness of brick samples with different compositions. It explains that the refractoriness of sample D (RC(60%), GS(10%) and WPC(30%)) was highest (1408°C) which is significantly higher than blank refractory (100%RC).

3.5.2 Thermal shock resistance

The ability of material to withstand repeated cycles of heating and cooling before a crack is observed is termed as thermal shock resistance [38]. The sample was examined to note the presence of cracks and the specimen was returned back to the furnace, heated for 10 mins and cooled again for 10 mins. The procedure was repeated several times until a visible crack was observed on them. The number of cycles before crack was observed and the results are presented in Table 3. It shows that thermal shock resistance of sample D (RC(60%) + GS(10%) + WPC(30%)) was 15 cycles which is considerably higher than brick produced from 100% red clay (9 cycles). Therefore, the 30%WPC blended sample falls within the accepted range of 15+ cycles [42].

3.5.3 Thermal conductivity

The thermal conductivity of refractories depends upon a number of factors such as mineralogical and chemical compositions, percentage of porosity, existence of defects, moisture content as well as temperature at which the refractory is being used [13,14]. Refractory materials should have low thermal conductivity and high mechanical strength to withstand the high temperature condition in the furnace. In this investigation, thermal conductivity was carried out by using (30mm×15mm) cylindrical specimens according to the standard specifications of the instruments using Lee's disk type. As shown in Table 3, thermal conductivity of sample D (RC(60%) + GS(10%) + WPC(30%)) is significantly lower (1.3W/m.K) compared to that of 100% red clay (2.3W/m.K).

3.6 Mineralogical Analysis

X-ray powder diffraction pattern of the brick prototype (RC(60%) + GS(10%) + WPC(30%)) which demonstrates the best performance in terms of mechanical, physical and thermal properties shows (Fig. 7) mainly quartz (COD#96-101-1160 [17] and small amount of Bavenite (COD#96-900-7488 [39]). Bavenite, $\text{Al}_2\text{Be}_2\text{Ca}_4\text{Si}_9\text{H}_3\text{O}_{28}$ is a mixed oxide and the IR spectrum of bavenite remains unchanged up to 900°C [40]. It exhibits good physical, chemical and thermal properties

from room temperature to elevated temperature. The presence of high percentage of quartz phase into the diffractogram ensures that the brick has high silica content and suitable to use as silica refractory brick [43].

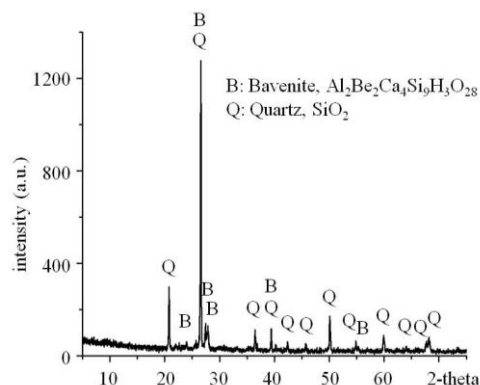


Fig. 7 XRD pattern of Refractory brick showing the existence of a mixture of crystalline two phases-Quartz and Bavenite

3.7 Chemical Analysis of Brick

Chemical properties of laboratory made refractory brick (RC(60%) + GS(10%) + WPC(30%)) which demonstrates the best performance is given in Table 4.

Table 4 Chemical composition of brick sample

Constituent	Content (wt.%)
Silica (SiO_2)	79.03
Calcium Oxide (CaO)	12.40
Iron (III) Oxide (Fe_2O_3)	1.68
Magnesium Oxide (MgO)	0.81
Aluminum Oxide (Al_2O_3)	6.00
Potassium Oxide (K_2O)	0.08
Sodium Oxide (Na_2O)	0.32
Loss on Ignition (L.O.I)	0.04

The table shows that the brick contains highest concentration of silica about 80% and the percentage of other major constituents like Calcium oxide and Alumina are about 12% and 6%, respectively. According to XRD data, SiO_2 is also a major phase along the mineral bavenite as a minor phase (Fig. 7). The content of BeO in bavenite is only 6-7%. On the other hand, Be is a very light element. Therefore, determination of BeO might be beyond the detection limit of ASTM method. The value of Loss on Ignition (LOI) is better to be low. So, it can be explained as the high silica content of the brick indicates the brick can be used as silica refractory brick and the presence

of lime and alumina indicates the refractoriness of the brick [43].

3.8 Comparison with Conventional Refractory Brick

There is only one industry in Bangladesh named “Savar Refractories Company” that produces refractory brick. The main raw material of this industry is fire clay which is imported from abroad over the years. A comparative study of the performance of laboratory made refractory brick with the conventionally used refractory brick was carried out to evaluate the suitability of our brick for the practical application [35,41]. The results are illustrated in Table 5 which confirms that the various parameters of brick produced in our laboratory are in good agreement with that of conventional refractory bricks.

Table 5 Comparison with conventional refractory brick

Parameter	Lab refr. Bricks	Commercial refr. Bricks
Compr. Strength (MPa)	36.20	20-70
Water absorption (%)	4.58	4.5%
Linear shrinkage (%)	3.10	7-9
Porosity (%)	13.32	10-15
Bulk density (gm/cm ³)	2.64	1.98
Refractoriness (°C)	1408	1400
Thermal conductivity (W/m.K)	1.3	0.01-1.1 (Fireclay refr. bricks)

4. CONCLUSION

This work describes the production and performance assessment of refractory brick from red clay of local origin. Physical, mechanical and thermal properties investigated in this work undergone significant improvement due to the combination with glass sand and white Portland cement compared to the brick produced from red clay only. The properties of produced refractory brick are in good match with the refractory requirements. The compressive strength of the brick meets the required standard for refractory brick and is in line with the properties found in similar products in the market. Results showed that the refractoriness of laboratory made refractory brick is 1408 °C which could be used as a refractory lining material for furnaces, kilns, incinerators and reactors. Based on the performance evaluation testing results, it can be concluded that the proposed local raw materials are suitable for the manufacture of refractory bricks.

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