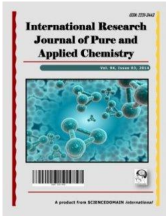

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Use of Borosilicate Glass as Fluxing Agent to Enhance the Physio-mechanical Properties of Bricks

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Authors' contributions

This work was carried out in collaboration between all authors. Author MSI designed the study, performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors USA and KSA managed the analyses of the study. Author KSA managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

The aim of this study is to investigate the effect of borosilicate glass as a flux on the Physio-mechanical properties of the brick. To do this job brick sample was prepared by incorporating borosilicate (BS) glass with red clay in different proportions like 20%, 35%, and 50% and then fired at 850ºC, 950ºC and 1050ºC. The Physio-mechanical properties of the prepared samples fired at different temperatures were examined through different experiments. The crystal phases were studied by XRD and the images of prepared samples at their cross section were taken by SEM. The optimal value of mixing ratio of BS glass with red clay found 35% during sintering at 1050ºC for better Physio-mechanical properties of brick sample. The enhanced Physio-mechanical properties of brick sample like water absorption, bulk density, linear shrinkage and compressive strength were

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7%, 2500 kg/m^3, 1.57%, and 39.26 M Pa, respectively. The outcome of the experiments shows that the BS glass is feasible to be acting as a fluxing agent by improving the Physio-mechanical properties as well as the sustainability of the building bricks.

Keywords: Borosilicate glass, engineering properties, flux, XRD, brick.

1. INTRODUCTION

Borosilicate glass is a boron-containing material where near about 10-14% B_2O_3 is found. Boron-containing materials can be used in glass and ceramic industry as fluxing agent or raw material to amend the overall technological features of the final product by increasing vitrification process and decreasing the melting point during sintering of ceramic body. Kurama et al. [1] found that borax waste increased the vitrification and thereby provided better technological features in ceramic body. Boron minerals used extensively in the glass and ceramics industry as vitrification and flux agents [2]. Boron oxide-bearing minerals can be employed as alternative raw materials in ceramic industry because of its glass forming character and capacity to reduce the melting point of sintered ceramic mass during sintering [3]. So, therefore, borosilicate glass can be used as a fluxing agent and best alternative raw material to make ceramic body. Borosilicate glass is made mainly of silicon oxide and boron oxide where boron acts as a strong flux in low temperature. Ryan describes flux as a material which turns down the fusion temperature of the concoction to which it is added [4]. The boron content of borosilicate glass plays a vital role during sintering of ceramic body. The engineering properties of ceramic body are linked to higher compressive and low water absorption properties. The compressive strength of an engineering brick is located between 75 N/mm^2 to 125 N/mm^2 and water absorption rate is between ≤ 4.5% to ≤ 7%. Engineering brick is mainly used in damped proof courses due to its less porous properties. In this project, borosilicate glass was incorporated in different ratios with clay to make ceramic body. The main ingredient of traditional ceramic body is hydrous aluminosilicate material like clay or kaolinite. The clay contributes enough plasticity and subsequent mechanical strength of final ceramic body through the formation of the mullite during sintering. Flux plays an important role of the filler, which maintains the skeleton of the ceramic body during sintering. The melting of the flux to the liquid occurs during sintering of the green body and the amount of liquid phase depends on the type of flux [5-7] as well as clay. Here, the liquid phase during sintering moves into the alumina silicate ceramic powders by viscous flow, resulting in the bonding of powders and formation of a dense mass due to the deep inter-diffusion of the phases [8]. The remainder of the fluid phase is solidified without active crystallization during cooling and thus resulting in a vitreous ceramic body. To develop engineering properties of ceramic body is the main issue of this project work. To do this job borosilicate glass was incorporated as a fluxing agent in ceramic body. B_2O_3 in borosilicate glass act as a strong flux. So, therefore different test and experiments were carried out to measure where the borosilicate glass is feasible as a fluxing agent to improve the engineering properties of the ceramic body.

2. MATERIALS AND METHODS

2.1 Materials

The BS glass used in the experiments in this study was collected from broken laboratory glassware of IGCRT, BCSIR. The main raw materials of ceramic body are clay which is collected from Mirpur Ceramics Industry, Mirpur-12, Dhaka-1216. The chemical composition of BS glass and Clay are given in Table 1.

2.2 Methods

First of all, pretreatment processes such as grinding and sieving were performed on raw materials. Three types of mixed materials were prepared by adding BS glass powder with red clay in proportions of 20, 35 and 50wt% respectively. Pressed red clay brick specimens were shaped under a load of 300 kg/cm^2 in a 2" x 2" x 2" cube size mold (350 g materials in weight). Prior to the molding process, 20wt% water was added to the mix to obtain enough plasticity. From each group, 3 pressed cube size brick samples were made. After the molding process, test specimens were dried at room temperature for 2 days, and the drying shrinkage of bricks was measured by slide calipers. As the final step, the specimens were fired in an electrical muffle furnace at their final temperature 850ºC, 950ºC, and 1050ºC respectively. The sintering period for all samples was 180 minutes and soaked at 850ºC,

Table 1. Chemical composition of raw materials

Parameters	BS Glass (wt.%)	Clay (wt.%)
SiO_2	79.81	66.90
Al_2O_3	1.27	22.11
CaO	1.61	0.49
MgO	0.42	0.28
Fe_2O_3	0.52	4.94
Na_2O	4.19	0.54
K_2O	0.82	1.71
B_2O_3	13.59	-
LOI	-	8.14

950ºC and 1050ºC respectively for 30 minutes. After firing they were cooled in the furnace down to room temperature. Brick samples produced at different temperatures were firstly evaluated for the technical performance like compressive strength and then the samples which showed the best compressive strength were subjected to the other series of tests such as linear shrinkage, water absorption, bulk density and apparent porosity.

2.3 Experiments

2.3.1 Compressive strength

In this study, 2"x2"x2" fired brick samples were taken for the compressive strength test. The compressive strength of bricks was measured by a Hydraulic press machine. The distance between supports was measured and then the samples were crushed using the strength machine. The breaking load of the brick was read from the machine. The compressive strength was calculated using the following formula:

$$CS = F/A^2 \quad (1)$$

Where CS is the compressive strength (MPa), F is the breaking load (lbs.), A^2 is the area of prepared sample was tested.

2.3.2 Bulk density and apparent porosity

The bulk density and apparent porosity were measured by the Archimedes method according to ASTM designation C 20-00.

$$\text{Bulk density, } B=D/V \text{ gm/cm}^3 \quad (2)$$

$$\text{Apparent Porosity, } \%P= [(W-D)/(W-S)] \times 1 \quad (3)$$

Where W= water saturated weight, D= dry weight and S= Suspended weight.

2.3.3 Linear shrinkage

The linear shrinkage of samples was determined after drying and firing. The total shrinkage, which is the linear shrinkage from the wet state to the fired state, was also determined. The shrinkage was calculated as follows:

$$LS= Lw-Lf/Lw \times 100 \quad (4)$$

Where LS is the shrinkage percent. L*w* initial length of the body (mm), L*f* is the final length of the body (mm).

Zanelli et al. [9] concluded that the firing shrinkage is not proportional to the amount of liquid phase but rather depends essentially on the viscosity of the liquid phase, which exhibits limited changes in composition with time, seems to be mainly affected by temperature.

2.3.4 Water absorption

The sintered brick samples were dried 110ºC, cooled to room temperature, and then weighted as dry weight, W*d*. Then the samples were immersed in distilled water and boiled for three hours. The heating was stopped and the samples were allowed to remain immersed in the water for 24 h. The samples were taken out and excess water was removed from their surfaces by wiping with a damp cloth and again weighted as soaked weight, W*s*. The water absorption was calculated using the formula:

$$WA (\%)= [(Ws-Wd)/Wd] \times 100 \quad (5)$$

Table No. 1 shows the chemical composition of raw materials which are used to make ceramic body. The table shows that BS glass has 13.59% B_2O_3 and Na_2O_3 which are acting as a fluxing agent during sintering of ceramic body. The high % of $Al_2O_3$and Fe_2O_3 in clay is essential for producing high strength ceramic body.

3. RESULTS AND DISCUSSION

Fig. 1 displays the compressive strength values for blank and BS glass added bodies. The highest value of compressive strength of the blank body was found 13 mpa at 1050ºC. The highest value of compressive strength of the ceramic brick sample was found 37mpa with at 35% BS glass addition sintered at 1050ºC. The compressive strength values were slightly decreased with an increase in the BS glass percentage above 35%. This slight decrease in compressive strength values can be caused by the fact that high fluidity of melting glass which increases the internal voids in ceramic brick.

Fig. 2 displays the bulk density values for blank and BS glass added ceramic brick at different temperatures. The bulk density level of the blank body was found 1500 kg/m^3. The highest level of bulk density of ceramic brick was found 2500kg/m^3 with at 35% BS glass addition on 1050ºC. A sharp decrease in bulk density was observed with at 50% BS glass addition. The decrease in bulk density level can be caused by the fact that the fluxing features of BS glass lead to the creation of large amounts of liquid phases at 950ºC. The fluidity of glass melts increases due to lower viscosity when 50% BS glass added at 1050ºC.

Fig. 3 displays the effect of borosilicate glass on the linear shrinkage of ceramic brick. The linear shrinkage level of the blank body was 0.4% at 1050ºC. The linear shrinkage level increased with an increase in the BS glass percentage. Both the firing temperature and BS glass addition increase the linear shrinkage of ceramic body but BS glass help to accelerate shrinkage level of ceramic body. The highest level of linear shrinkage of ceramic brick was found 1.7% when 50% BS glass was added fired at 1050ºC.

Fig. 4 displays the effect of BS glass addition on the water absorption level of bricks. From the figure, it was found that the water absorption level of bricks was decreased with the increasing amount of BS glass addition. It was also observed that the water absorption level of ceramic bricks decreases with the increase of sintering temperature. So it is evaluated that the water absorption of ceramic bricks can be decreased by increasing both of the amounts of BS glass and sintering temperature.

The decrease in water absorption level can be caused by the fact that the fluxing features of BS glass that causes an increase in the vitrification of the ceramic body and leads to the creation of large amounts of liquid phases. The vitrified phase gives rise to the filling of open pores of ceramic body which in turn decreases the water absorption [10].

3.1 Role of Borosilicate Glass As Fluxing Agent

BS glass plays a strong flux role in the ceramic body. As fluxing agent borosilicate glass starts melting (or softens) before ceramic body does, therefore it collapses the porosity and bonds the un-melted ceramic particles to each other by making a eutectic system. In sintering, ceramic body does not undergo melting but fluxing agent BS glass melts and binds particles to each other.

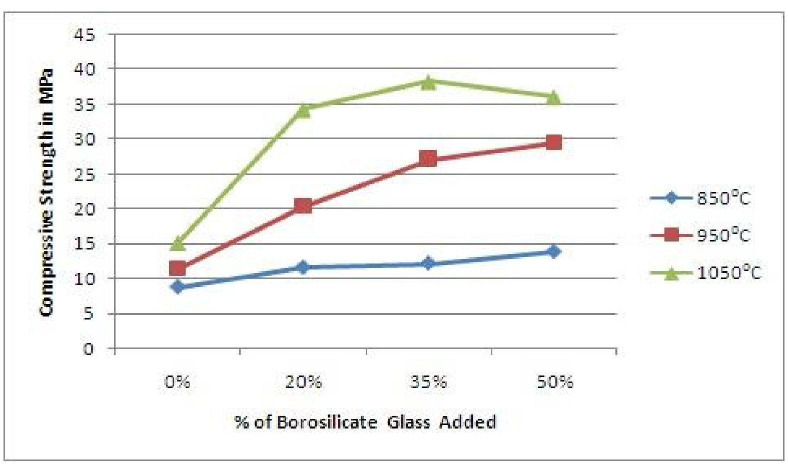

Fig. 1. Effect of BS glass on the compressive strength of ceramic brick

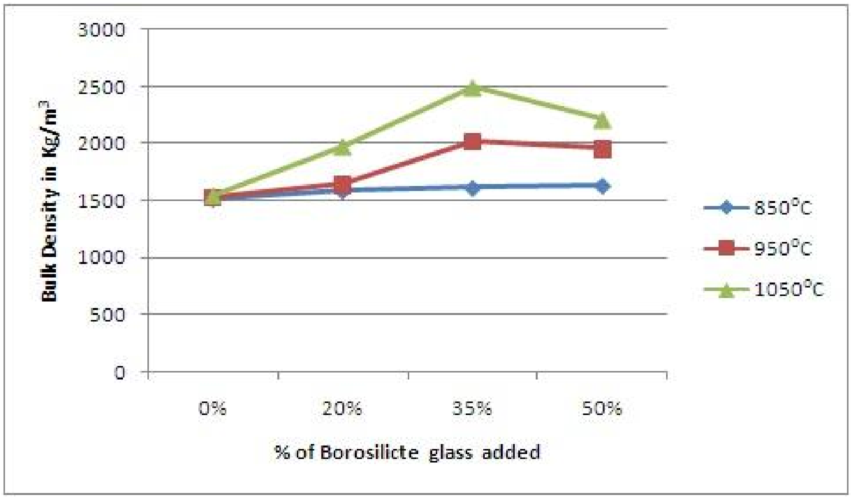

Fig. 2. Effect of BS glass on bulk density of ceramic brick

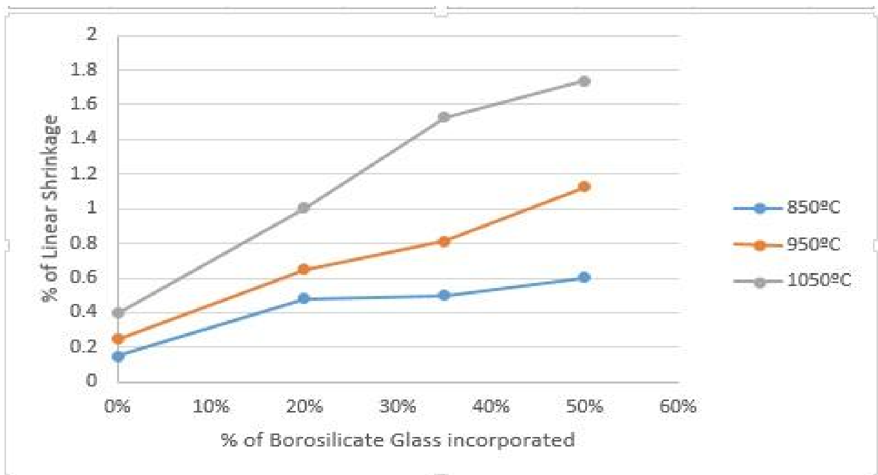

Fig. 3. Effect of BS glass on linear shrinkage of ceramic brick

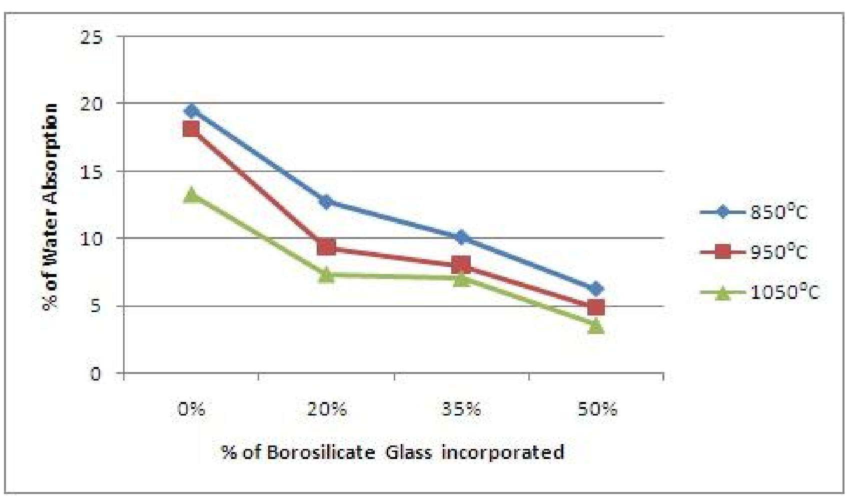

Fig. 4. Effect of BS glass on water absorption of ceramic brick

Fig. 5 shows SEM images of ceramic bodies with and without loaded fluxing agents and it clearly shows that BS glass plays a strong flux role in the ceramic body. As fluxing agent borosilicate glass starts melting (or softens) before ceramic body does, therefore it collapses the porosity and bonds the un-melted ceramic particles to each other by making a eutectic system. In sintering, ceramic body does not undergo melting (from Fig. 5c and 5d) but fluxing agent BS glass melts (Fig. 5a and 5b) and binds particles to each other.

Micro structural Characterization: An addition of the amount of borosilicate glass in the mixture led to the formation of mullite at 1050ºC. The microcline phase disappears with an addition of the sintering temperature because these phases were melted at a low eutectic temperature [11]. Mullite peaks increased in intensity with the firing temperature, whereas quartz peaks decreased slightly associated with its partial dissolution [12].

An increase of the amount of borosilicate glass waste in the mixture led to the formation of mullite at 1050ºC. In addition, the intensity of the quartz peaks decreased with an increase in the BS glass.

Thus, an increase of the temperature and BS glass is intended to increase the formation of mullite. Mullite is the only stable phase in the Al_2O_3-SiO_2 system at atmospheric pressure. The mullite phase is thought to act as a substantial part of the maturation of traditional and advanced ceramics [13]. The quartz content decreases with an addition of the sintering temperature due to its gradual disintegration into the glassy phase [14]. Besides, part of the quartz combined with Al_2O_3 to give mullite.

The vitreous phase contains more Na_2O and B_2O_3 with lower amounts of Al_2O_3 and K_2O in the BS glass. These oxides bring about a lower viscosity of the liquid phase at sintering temperatures in the materials containing waste glass [15]. The oxide compositions of the samples are given in Table 1. The amounts of Na_2O and B_2O_3 increase with an increase in the amount of BS glass, while the amounts

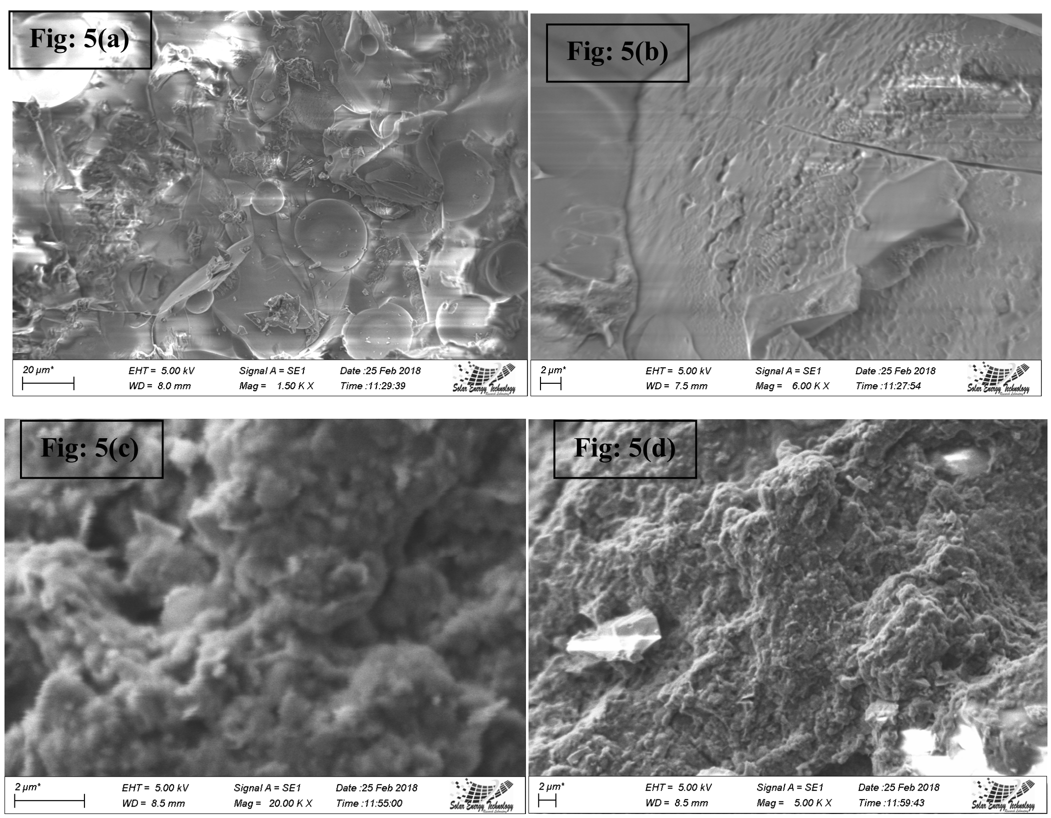

Fig. 5. SEM images of fluxing agent (BS glass) added ceramic body (fig 5a and fig 5b)and without any fluxing agent added ceramic body (fig 5c and fig 5d)

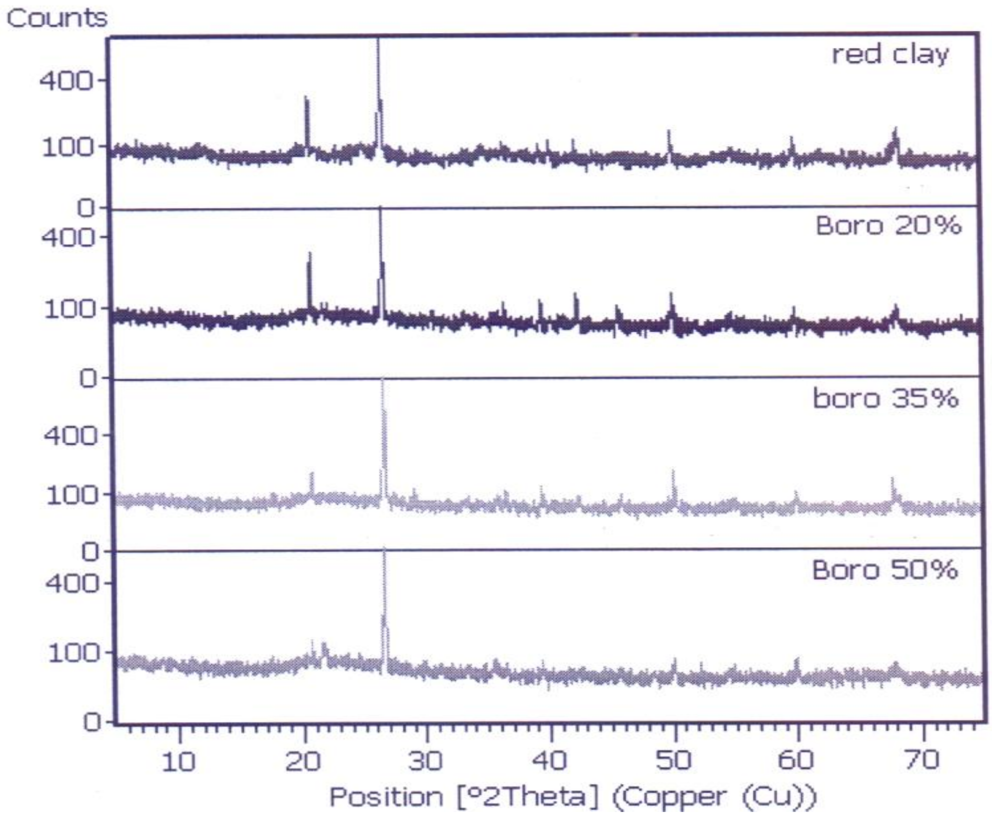

Fig. 6. X-ray Diffraction pattern of ceramic brick using BS glass in different ratios during sintering temperature 1050ºC

of Al_2O_3 and K_2O decrease. The glassy phase produced from these oxides has a lower viscosity and flows easily to fill open pores. Therefore, an increased compressive strength was found by increasing the addition of waste glass at 950ºC to 1050ºC (as seen in Fig.1). However, as borosilicate waste glass is added, the bulk density of samples dramatically decreased at sintering temperatures of 950ºC to1050ºC (as seen in Fig. 2).

During sintering, glass waste accelerates the densification process, with some positive effects (lower open porosity) as well as negative effects (higher values of shrinkage and closed porosity with lower values of bulk density (as seen in Fig. 2 and Fig. 3).

Densification at higher temperatures is rate-controlled by the strong dependence of melt viscosity on temperature and by the solubility of solids in the liquid phase, mainly free quartz. On the other hand, it can be said that quartz dissolution is the slowest process during firing of such products, and it helps to maintain a higher melt viscosity, even with an increase in the temperature, which prevents pyro-plastic deformation. A maximum firing temperature can be defined by the coarsening and solubility of gases filling the closed pores accompanied by a more or less pronounced expansion called bloating [16].

In particular, the sintering rate of ceramic body depends on the properties of the liquid phase at high temperature (i.e. surface tension/viscosity ratio) [17]. Increasing the sintering time and temperature can reduce the viscosity of the glassy phase contained in the matrix, resulting in an increase in viscous flow and densification of the ceramic body by lowering the activation energy [18]. This is also associated with a high rate of sintering due to the increased BS glass addition and glassy phase formation, leading to a decline in the volume of the overall porosity and an improvement in the densification resulting in easier diffusion and a reduced activation energy.

The content of B_2O_3 is very high in borosilicate glass and this oxide is responsible to decrease the melting temperature of the powder mixture and facilitate fusibility, leading to liquid phase sintering [19].

4. CONCLUSION

Uslu and Arol [20] found that additions up to 30wt. % of this Boron Waste enhances the glassy phase and improves the overall brick quality. Higher additions reduce the compressive strength and make rough the surface of the bodies. The starting temperature for sintering was reduced by the additional fluxing oxides such as B_2O_3 and Na_2O which eased the vitrification by reducing the viscosity of the liquid phase during sintering. The BS glass powder shows to be an efficient fluxing agent when it is used as an additive in ceramic mixture. During firing glass powder accelerates the densification process, with some positive effects (lower open porosity, water absorption) combined with negative ones (higher values of shrinkage and high closed porosity). From the overall discussion, the finding is that 35% BS glass addition with clay at 1050°C firing temperature is an optimal value for enhancing the engineering properties of bricks.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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