

## REUSE OF GLASS CULLET AS AGGREGATE FOR RED CERAMIC CLAYS

Silvio Rainho Teixeira  
Francine Bettio Costa  
Agda Eunice de Souza  
Gleyson Tadeu de Almeida Santos  
rainho@fct.unesp.br  
franbettiocosta@gmail.com  
agda\_pb@ig.com.br  
tadeu\_gtas@hotmail.com

Department of Physics, Chemistry and Biology, São Paulo State University (UNESP)

**Abstract:** Numerous silicates based wastes have been produced and considered for recycling and reuse. Among them, glass cullet is one of the most common silicate wastes and has considerable volume in the cities wastes. A big amount of this waste is recycled by the glass industry but another part is dumped in the cities garbage deposits. In the last few decades there has been considerable research on the reuse of glass waste as aggregate to produce glass-ceramic, for mortars, ceramics and for cement and concrete. This work is concerned to study the reuse of one way glass bottles as aggregate to produce bricks and roof tiles (red ceramic). Two kinds of glass powder were prepared by sieving (0.037 to 0.088 mm and 0.088 to 0.125 mm). Prismatic ceramic bodies (CB) were pressed (60 x 20 x ~5 mm) using a ceramic mass with 0, 5, 8 and 10 % of glass cullet powder added and fired at five different temperatures (800 to 1200 °C). The results of the technological tests (flexural strength, water absorption, dimensional changes, density and porosity apparent) show that shrinkage increases with the glass content and all other properties above are improved. These changes are more exhibited at temperatures higher than 900°C and in the higher powder glass concentrations (8 e 10%).

**Keywords:** Glass cullet, Red ceramic, Silicate waste, Reuse, Clay minerals.

### 1. INTRODUCTION

Red ceramic is a very promising alternative for the incorporation of residues due the wide use of pottery and ceramics in all regions of Brazil, and also because a large variety of raw materials can be utilized. The incorporation of residues in controlled quantities can extend the life time of raw materials deposits besides making use of this residue which in general is discarded into the environment, contributing to the economic viability of the recycling of some solid residues. With the growth of the population, the increase in the consumption of disposable and returnable materials has increased the volume of residues produced in cities. The glass consumed by the public, generally in the form of container, is relatively inert and therefore non biodegradable. In 2001, more than 2 million tons of glass were produced in Brazil, of which ~43% of the glass was for packaging (ABC, 2002). Nowadays, approximately 50 % of glass residues are recycled in Brazil. Due to its chemical composition, the incorporation of glass residue in ceramic materials is an option for reuse, and many studies on this theme have been developed in Brazil (Godinho et al., 2004, 2005; Oliveira et al., 2004; Filho & Gibo, 2004; Bragança & Bergmann, 2004; Pereira Filho et al., 2004). In addition, the works published on the recycling of glass in ceramic materials are numerous (Delben et al., 2006; Rawlings et al., 2006; Andreola et al., 2005; Yoon & Yun, 2005; Bernardo et al., 2005; Bragança & Bergmann, 2004; WRAP, 2004). These studies furnish a panorama of the development of research on this theme. The addition of glass to ceramic masses causes an increase in the levels

of fluxing oxides, responsible for the formation of the vitreous phase and densification of the ceramic masses during the firing process, increasing its flexural strength and reducing water absorption. The literature shows that several benefits are obtained by using glass cullet in ceramic materials: reduced electrical energy consumption, enhanced quality of material, saving ceramic raw material, reduction in HF emissions (WRAP, 2004) and less waste disposal problems. The aim of the present work was to determine the effects of the incorporation of glass waste on the ceramic properties of clays used to produce bricks and roof tiles.

## 2. MATERIALS AND METHODS

Glass powder was incorporated into a ceramic mass utilized for the fabrication of roof tiles, which was obtained from a ceramic factory in the municipality of Presidente Epitacio, SP – Brazil. The ceramic mass was allowed to stand to dry naturally and then triturated in a blade mill. Part of this sample was assayed for grain-size distribution to determine the concentration of clay, sand and silt, using the pipet method (Klute, 1986). The rest was passed through a set of screens to determine grain-size distribution and help in the preparation of the mixtures used in the ceramic probes. Brown beer bottles, which were the 350 ml long neck type and non-returnable, were broken into pieces and pulverized in a ball mill for 6 h. The powder was passed through sieves of 0.037, 0.088 and 0.125 mm to obtain two different particle sizes: (a) 0.088 to 0.125 mm and (b) 0.037 to 0.088 mm. These particle sizes were chosen considering the grain-size distribution obtained for the ceramic mass. The clay and glass powder were dried in an oven for 24 h at 110°C. The mixture of ceramic mass and glass powder was homogenized in a ball mill for 6 h. Four mixtures were prepared containing 0, 5, 8 and 10% glass powder, for two particle sizes, alpha and beta, which were fired at five different temperatures. Water was added to each sample, 15% by weight, for pressing. Six ceramic probes were made for each sample, with dimensions of 60 × 20 × 5 (mm), utilizing for each one approximately 20g of sample. In each pressing (7 tons), three probes were prepared simultaneously. After pressing, the ceramic probes were cleaned, labeled, measured and weighed.

After making and measuring (length and weight) the ceramic probes, they were placed in an oven at 110°C for 24 h. After the drying period, they were placed in a desiccator until reaching room temperature, and again weighed and measured. The CBs were fired at 800, 900, 1000, 1100 and 1200°C, at a heating rate of 10°C/min up to 110°C, kept at this temperature for 30 min, and then heated up to the firing temperature also at 10°C/min, and kept at this temperature for 2 h. After firing, the furnace was turned off, the ceramic probes were allowed to cool to ~ 60°C and then placed in a desiccator until reaching room temperature. Their dimensions and weight were then determined again. Afterward, the ceramic probes were tested for linear drying shrinkage (LSD), linear firing shrinkage (LSF) and loss on ignition (LOI). Flexural strength (FS) in a three-point flexure test was determined after firing the CBs, with control and automatic data recording, using EMIC instrumentation. Apparent specific weight (ASW), water absorption (WA) and apparent porosity (AP) were determined after firing, using the Archimedes method (hydrostatic balance) (Souza Santos, 1989).

## 3. RESULTS AND DISCUSSION

### 3.1 Grain-Size Distribution

The results for texture, obtained without extraction of organic matter (OM) are presented in Table 1. The determinations were carried out in triplicate, and the values present in the table are the means for the three measurements for each fraction. According to the Winkler diagram (Pracidelli & Melchades, 1997), the ceramic mass for the production of roof tiles should have an ideal concentration of clay between 30 and 40%, of silt between 20 and 50% and of sand between 20

and 40%. Therefore, the amount of clay found in the sample is higher than the recommended range for the production of roof tiles, allowing it to be mixed with non-plastic material.

Table 1: Grain-size distribution.

	<b>Sand</b>	<b>Clay</b>	<b>Silt</b>
<b>Tile mass</b>	24.13 %	58.27 %	17.60 %

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### 3.2 Loss on Ignition (LOI)

As expected, LOI increased with sintering temperature for all the mixtures (Figure 1). The low LOI values (< 8.5%) in the pure sample indicate a low concentration of organic matter. At 1000°C and higher, the loss of mass was constant, indicating that the reactions occurred at the lower temperatures. Kaolitic clays, in general, show a greater loss of mass at temperatures below 600°C, where there is a loss of moisture, structural water (hydroxyls) in clays, hydroxides (mainly of iron and aluminum), and organic matter. Increasing the amount of glass in the mixture reduces loss of mass since it substitutes for the clay. In comparing the effect of particle size of the glass powder, it is seen that there was no substantial difference in loss of mass between the alpha and beta mixtures, although the results show that, principally to 10% of glass, the loss of mass was less with the alpha powder

### 3.3 Linear Drying Shrinkage (LSd)

LSd of the ceramic probes, with and without glass powder, is small, varying between 1 and 2%. The incorporation of 5 and 8% glass powder (non plastic material) caused a small increase in LS, and 10% glass powder had practically no effect on LS of the ceramic probes

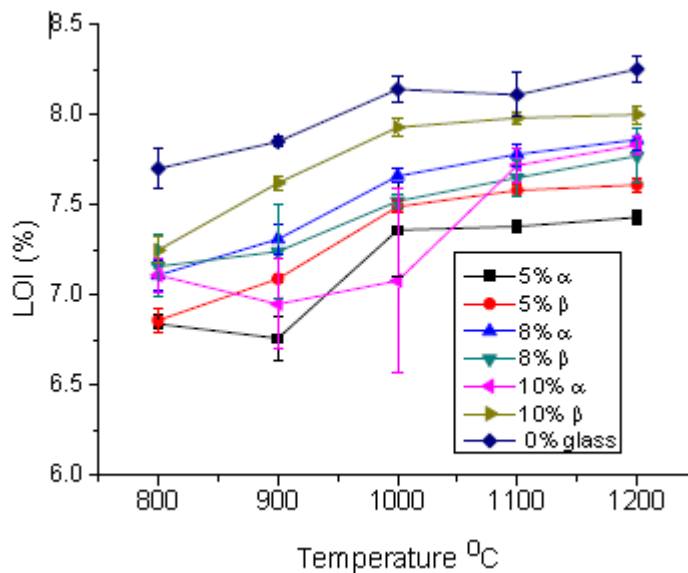


Figure 1: Loss of mass on ignition to ceramic mass with glass powders (a and b).

### 3.4 Linear Firing Shrinkage (LSf)

At 800°C, for 5, 8 and 10% glass powder alpha, LSf was not altered, but for 5% glass powder beta, LSf decreased slightly. At 900 and 1000°C, glass powder alpha (5%) in the ceramic mass decreased LSf. At 1000°C and higher, there was a tendency for linear shrinkage to increase for samples with glass powder (a and b) due to the presence of fluxing oxides (Figures 2). The glass powder concentrations of 8 and 10%, for both powder particle sizes, consistently resulted in a greater linear shrinkage for samples with glass incorporated. The softening warped the samples, mainly those

sintered at 1200oC, impairing the measurements used in the determination of L<sub>Sf</sub>, resulting in errors and a large standard deviation in the measurements.

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Between 950 and 1225°C, vitrification occurs with kaolinitic samples, due to the release of silicon oxide (SiO<sub>2</sub>) which reacts with free oxides, mainly alkaline, alkaline earth and iron oxides, forming glass (Souza Santos, 1989). Some of these oxides are present in clays, some are released in the reaking of the structure of clay minerals, and others, mainly alkaline oxides, are present in glass powder. Even for samples fired at 1200°C, Lsf was below the recommended maximum limit of 6% (Macedo et al., 1996). As seen in Figure 2, particle size had a small influence on Lsf, with the  $\beta$  glass powder (finer) tending to show greater linear shrinkage than  $\alpha$  at temperatures < 1000 °C .

### 3.5 Water Absorption (WA)

The absorption of water (WA) diminished with increase in concentration of glass in the sample and also with increase in firing temperature (Figures 3). As water absorption for ceramic mass was low and since the glass powder tended to reduce this value, all of the samples showed a WA of < 17%. For firing at 1100°C, WA was less than 10%, and at 1200°C, the samples with 10% glass powder ( $\alpha$  and  $\beta$ ) showed a WA of < 7.5%. All these values are below the recommended maximum value for the production of roof tiles, 18%, (ABNT, 1996) and ceramic blocks for structural masonry and for sealing (8% < WA < 22%) (ABNT, 2004) and some values are within the mean limit values established for the production of pressed ceramic plates, BIIb (6 < WA  $\leq$  10%) and BIII (WA > 10%) (ABNT, 1997). It should be noted that for the production of ceramic plates the pressure applied in the making of the pieces is much greater than that used in this work for preparing ceramic probes with clay for bricks and roof tiles. The greater pressure would improve even more the properties of the pieces. The behavior of WA as firing temperature increases was similar for 8 and 10%, for the both types of glass powder ( $\alpha$  and  $\beta$ ).

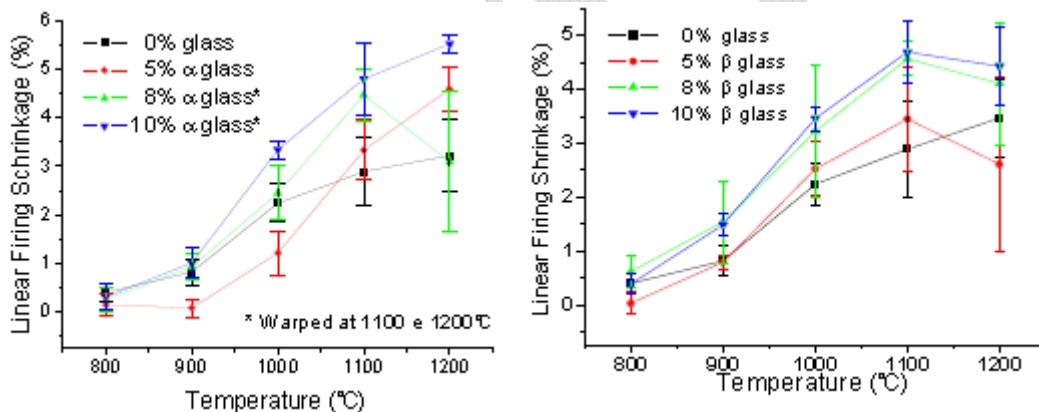


Figure 2: Linear shrinkage as a function of firing temperature for ceramic probes containing 0, 5, 8 and 10% glass powder,  $\alpha$  and  $\beta$ .

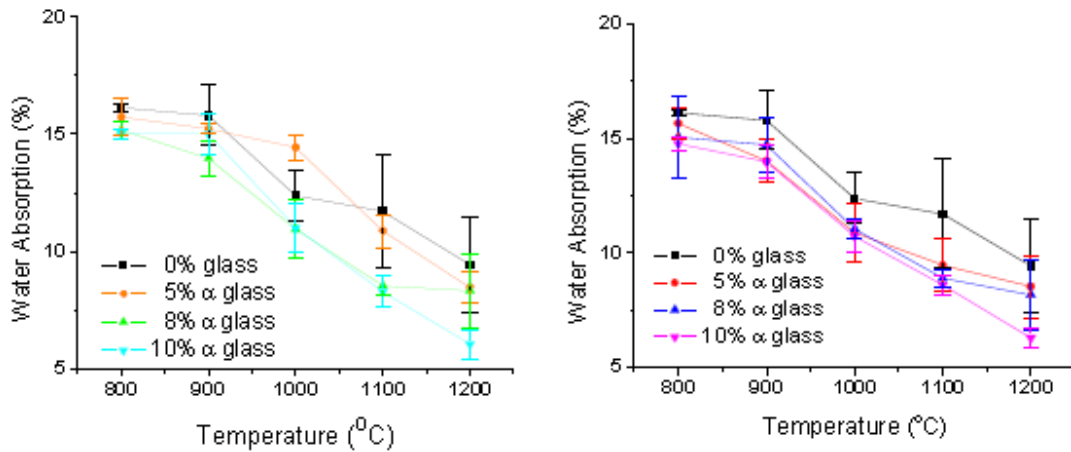


Figure 3: Water absorption (WA) of ceramic mass with 0, 5, 8 and 10% glass powder, alpha and beta, as a function of firing temperature.

### 3.6 Apparent Porosity (AP)

The apparent porosity (AP) versus firing temperature curves show a behavior similar to that of water absorption (WA). The data to calculate WA and PA are the same (Teixeira, 2006). With increase in firing temperature, values for WA and AP tended to decrease since a greater densification of the sample occurred. The apparent porosity of the ceramic mass (CM) was approximately 30%, at 800 and 900°C. At these temperatures, the incorporation of the two glass powders showed a tendency for a small decrease in the AP value (values below those for CM). At 1000°C and higher, there was a continuous fall in AP for the two glass powders, up to 1200°C, where there appears to be difference between percentage of glass incorporated into the sample. At 1200°C, a greater decrease in AP ( $AP < 15\%$ ) was observed for 10% glass powder (alpha and beta). The AP values for the two glass powders and all the samples were less than the maximum limit value (35%) established for the production of perforated bricks.

### 3.7 Apparent Specific Weight (ASW)

All of the samples showed ASW values greater than the recommended minimum value (1.7g/cm<sup>3</sup>). Figures 4, shows that there was an increase in ASW as the firing temperature was increased.

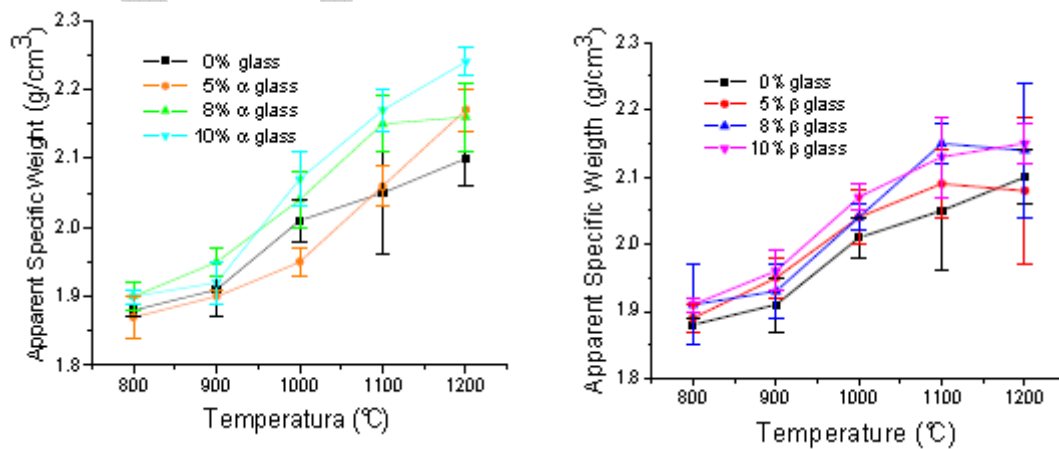


Figure 4: Apparent specific weight (ASW) of the ceramic mass containing 0, 5, 8 and 10% glass powder, alpha and beta, as a function of firing temperature.

### 3.9 Vitrification Curves

The vitrification graphs (Figures 6) show a variation in WA and LS as a function of firing temperature. The two curves WA and LS are governed by the porosity of ceramic bodies. At 900oC and higher, there is marked decrease in water absorption and increase in LS indicating the beginning of the formation of the liquid phase. At 1200oC, there is an indication of expansion of the ceramic probes, meanwhile some of them became twisted, impairing measurements that determine the LS. The plots show that the incorporation of glass powder causes a decrease in WA of approximately 10%, at temperatures above 1000oC, and that LS is less than 4% at temperatures lower than 1000oC, which are close to those used in kilns for ceramics. Although the firing temperature was diminished with the incorporation of glass, WA and LS values are more sensitive to small variations in temperature

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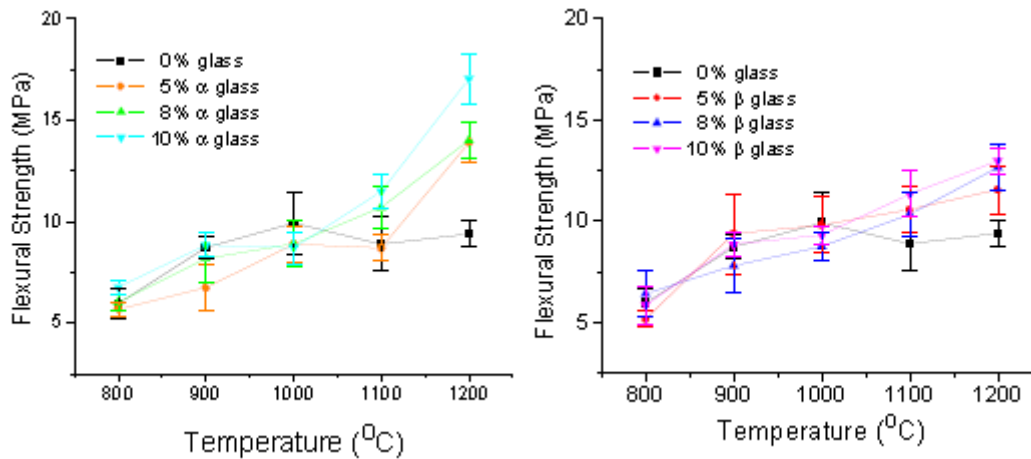


Figure 5: Flexural strength (FS) of ceramic mass with 0, 5, 8 and 10% glass powder, alpha and beta, as a function of firing temperature.

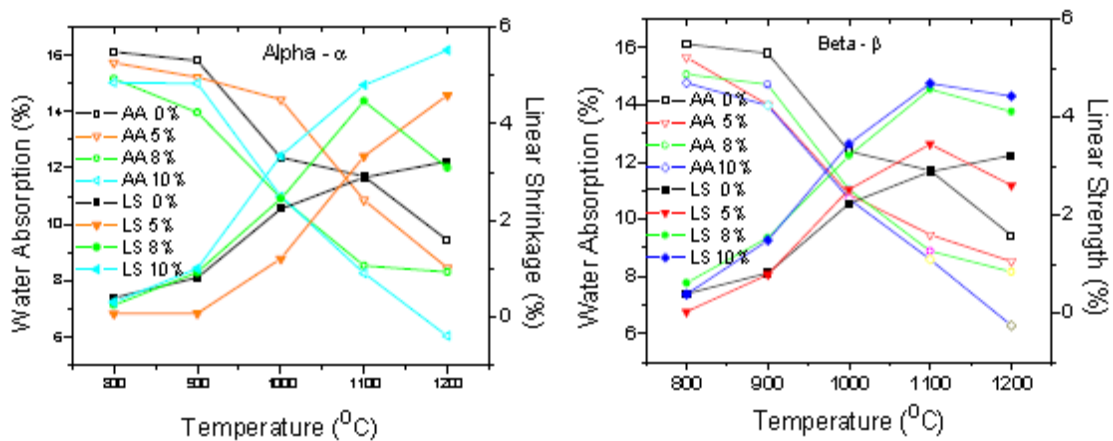


Figure 6: Vitrification curves for the ceramic probes with glass powder incorporated (0, 5, 8 and 10%), glass alpha and glass beta.

### 3.10 CONCLUSIONS

The results show that the incorporation of glass powder tended to improve all of the technological properties of clay for firing temperature higher than 1000°C. Therefore, glass powder can be used for improving the technological properties of red ceramic clays. These changes in properties depend on the amount of glass powder incorporated into the clay, firing temperature and particle size of the glass powder. On the other hand, these samples showed greater linear shrinkage and many of them warped, mainly at 1200°C, indicating the formation of a liquid phase at lower temperatures for the samples with glass powder. Some samples warped at 1100°C. The effect of the incorporation of glass powder was marked in some cases for the glass powder alpha which has a grain size closer to that of the clay used. Flexural strength was significantly altered at firing temperatures of 1100 and 1200°C, due to the formation of liquid phase attributed to the glass powder. This effect was more pronounced for the coarser powder (alpha). The vitrification curves show that the incorporation of glass powder lowered the firing temperature of ceramic bodies. The incorporation 10 and 8% glass powder, in general, showed better results compared to 5%.

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