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## RESEARCH ARTICLE

### EFFECT OF GROG WASTE IN CERAMIC BRICK

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#### ABSTRACT

The use of industrial waste materials as additives in the manufacture of ceramic products has been attracting a growing interest from researchers in recent years and is becoming common practice. This work describes the changes in the behaviour of the clay material used in a ceramic industry due to addition of grog, produced in ceramic industry. Mixtures of clay and waste material (0–50 wt. %) were uniaxially pressed and sintered at temperature 1000°C. Results from chemical, mineralogical and morphological analyses (XRF, XRD and SEM), water absorption and compressive strength show that the grog can be added to the clay material with no detrimental effect on the properties of the sintered fire-clay products. The test results indicate that the grog could be used as filler in ceramic bricks, thus enhancing the possibility of its reuse in a safe and sustainable way.

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#### INTRODUCTION

Nowadays industrial activities are responsible for the generation of large amounts of solid wastes (e.g. ceramic industries, marble and granite industries, paper and textile industries, petroleum refinery, urban waste, ashes, Al-rich sludge etc.) (Vieira *et al.*, 2007; Acchar 2006; Ferreira 2004; Luisa Barbieri 2013). Industries have been looking for technological alternative in order to optimize their process, producing less waste material. The ceramic industry, especially the sector devoted to the fabrication of building products is very capable of incorporating and reusing different types of industrial waste materials (Stefanov 1991; Ribeiro 2002; Dondi 1997 a, b). The most used raw materials in the traditional ceramic industries can be basically divided into three categories: plastic components (clays), fluxing components (feldspar) and inert components (quartz and sand). Clay materials used in the red ceramic industry show an extensive range of compositions, which permit the incorporation of a variety of industrial waste materials. Some wastes are very analogous in composition to the raw materials used actually and often contain materials that can also be helpful in the fabrication of ceramic products. One such waste material is the ceramic rejects known as grog produced in growing amounts in the ceramic industry. Grog waste has been considered as replacements for conventional ceramic raw materials that are becoming scarce and will be eventually exhausted. For instance, a reformulated ceramic bricks, in which clay was replaced by grog waste, present lower plasticity that reduces the risk of dimensional defects (Vieira

*et al.*, 2007). According to Vieira *et al.* (2007) grog waste could be used to improve the mechanical properties, workability, and chemical resistance of conventional ceramic brick. In this work, the effect of grog on the physico-mechanical properties of the ceramic bricks was therefore investigated. XRD and morphological characterization (SEM-EDS) of the ceramic bricks are also presented.

#### Experimental procedure

A fire clay material used in the ceramic industry and grog nothing but crushed fired brick waste, collected directly from Government ceramic Institute, Vriddhachalam (southern India) were selected and characterized.

Table 1. The proportions of the mixtures for the formulations (wt. %)

Formulation	Fireclay	grog
C	100	0
C-10G	90	10
C-20G	80	20
C-30G	70	30
C-40G	60	40
C-50G	50	50

The characterization included chemical composition (X-ray fluorescence, Bruker S4- Pioneer), mineralogical composition (X-ray diffraction, Seifert JSODEBYEFLEX 2002) and morphology (SEM EDS, JEOL model, JSE-5610 HV). Mixtures as shown in Table 1 containing 0, 10, 20, 30, 40 and 50 wt. % reject were homogenized for 4 h in a planetary ball mill. The necessary amount of water (10 wt. %) for mixing fire clay and grog was added to obtain ceramic paste and the paste

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was uniaxially pressed into bars (140mm×27mm×24mm) under a load of 30MPa. Subsequently, the bars were sintered at temperature 1000°C for 4 h, with a heating rate of 10 °C /min. Compressive strength was measured for fired samples according to standard procedure BS 6073 (BS 6073, 1981) using a Universal testing machine by dividing the maximum load with the applied load area of the brick samples (Shimadzu Autograph). All shaped samples were tested by applying the load centered in the upper face of the brick with speed lower than 20 MPa/s until fracture. Water absorption values were determined from weight differences between the as-sintered and water saturated pieces (immersed in water for 24 h). The crystalline phases of the raw materials and the sintered sample (C-30G) were identified by X-ray diffraction. The morphological and chemical aspects of the composition C-30G were studied by scanning electron microscopy, SEM, using a JEOL model, JSE-5610 HV equipment coupled with energy dispersive spectroscopy, EDS, facility.

## RESULTS AND DISCUSSIONS

The chemical composition of the fire clay and grog waste is given in Table 2. The fire clay and grog waste present the expected typical composition. The chemical properties of the ceramic brick highly depend on the waste composition (Tudisca *et al.*, 2011). The fire clay and grog are rich in silica (SiO<sub>2</sub>) and alumina (Al<sub>2</sub>O<sub>3</sub>).

Tite, 1981). This favoured the formation of anorthitic plagioclase during firing (Kreimeyer, 1987) and also prevented shrinkage of raw brick. The fire clay and grog waste also contain a reasonable amount of iron oxide (Fe<sub>2</sub>O<sub>3</sub>). This oxide Fe<sub>2</sub>O<sub>3</sub> in addition to K<sub>2</sub>O, Na<sub>2</sub>O, CaO and MgO are considered as fluxes. They can influence the densification behavior of the ceramic building materials during firing (Segadaes, 2006). Fig. 1 shows the X-ray diffraction patterns of the raw materials. It can be noted that the composition is constituted by quartz and kaolinite and minor amounts of muscovite, anorthite, calcite, and hematite. The crystalline phases identified are in agreement with the results observed by XRF (Table 2). Morphological aspects of the sample C-30G fired at 1000°C are outlined in Fig. 2. The scanning electron micrographs of C-30G show the typical irregular angular-shaped particles of quartz and/or cristobalite. In addition, a wide particle size range is also observed in the micrographs. The line spectrum for C-30G sample determined by using EDS (Fig. 2) indicated the presence of C, O, Fe, Al, Si, Ti and Cu. These results are consistent with the chemical composition data (Table 2), except Cu. Fig. 3 shows the XRD pattern of composition C-30G fired at 1000 °C. It is observed that the grog addition does not change the major crystalline phases of the fired composition C-30G, such as quartz, mullite, hematite and anorthite formed at high temperatures (Carty 1998). The main difference observed in Fig. 3 is the increase of the hematite intensity peaks and formation of mullite.

Table 2. Chemical composition of the clay and grog waste (wt. %)

Oxides	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	CaO	MgO	MnO	K <sub>2</sub> O	Na <sub>2</sub> O
Clay	59.60	18.62	16.48	2.81	0.67	1.41	0.27	0.03	0.11
Grog	59.64	30.48	4.21	1.14	0.52	0.70	0.03	3.05	0.22

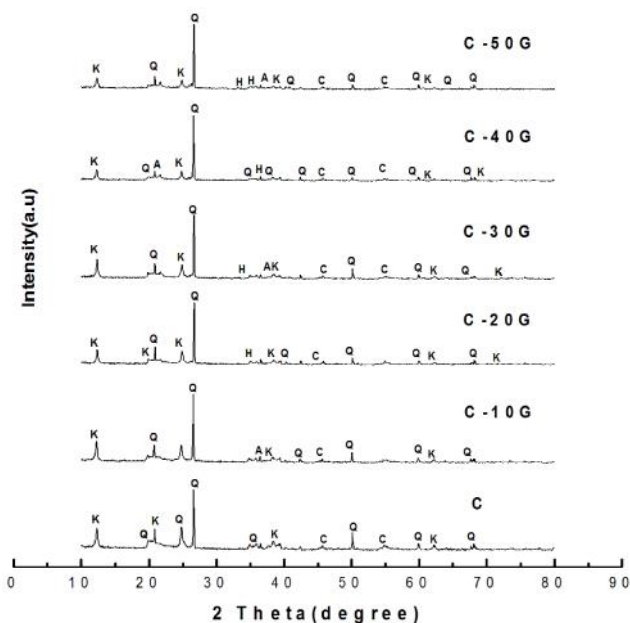
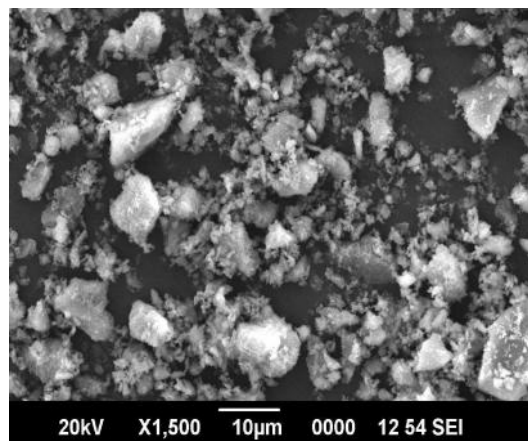
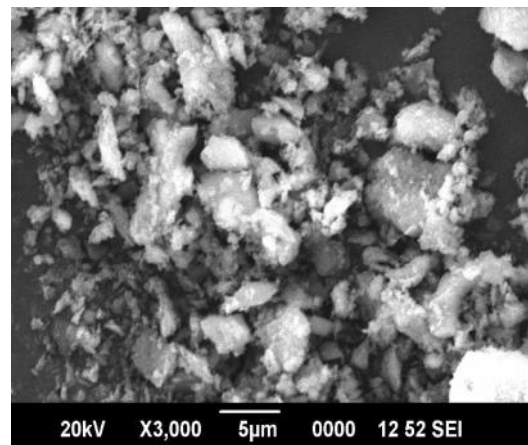


Fig. 1. X-ray diffraction patterns of the raw materials

The presence of quartz prevented cracking, shrinking and warping of raw bricks and also provided uniform shape to the bricks (Rajput, 2004). The durability of bricks depended on the proper proportion of quartz in the waste. In the present study, the relatively low CaO content of the samples (less than 6%) is indicative of non-calcareous type of sample (Maniatis and



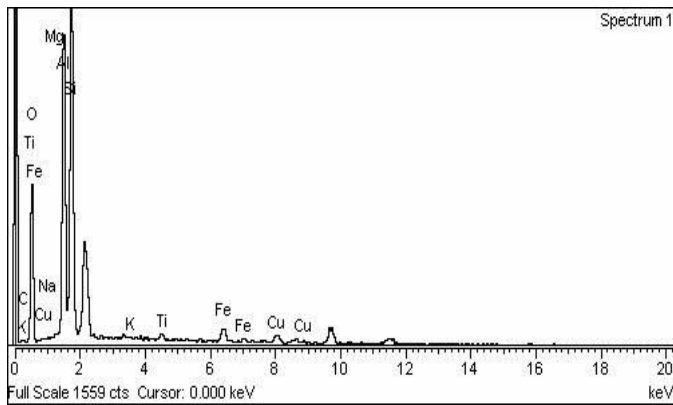


Fig. 2. SEM micrographs and EDS spectrum of the ceramic brick formulation C-30G

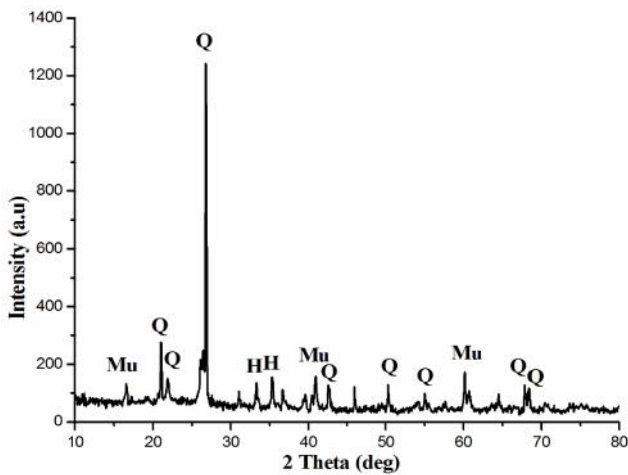


Fig. 3. X-ray diffraction pattern of the ceramic brick formulation C-30G fired at 1000 °C

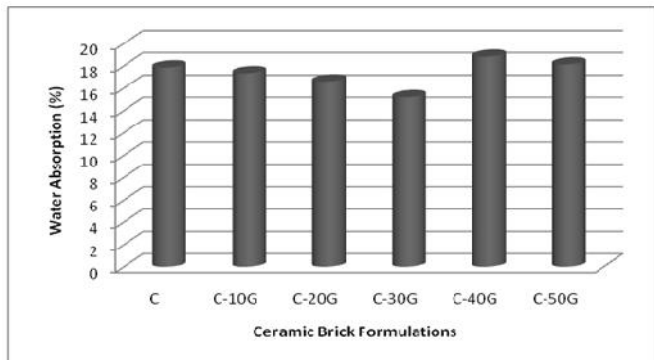


Fig. 4. Water absorption of the ceramic bricks versus waste content

The hematite observed in the composition C-30G is formed from the iron hydroxide (goethite) dehydration. The presence of high temperature minerals (quartz, mullite, hematite and anorthite) contribute for the reinforcement of ceramic bricks. Water absorption is a measure of openness in the bricks as well as a quality parameter. The lower the water absorption, the greater the life of the ceramic piece is and the greater its resistance to external weather conditions. Incorporating waste in the clay body decreased the water absorption. Fig. 4 depicts the water absorption values for the ceramic brick. This physical property is very important, because it is related to the open porosity of the fired products. As expected, the waste containing pieces show lower water absorption values than the

reference formulation C. The decline in water absorption values suggests that the replacement of clay by grog improves densification. This is probably due to the reduction of the viscosity of the glassy phase, which accelerates the sintering process (Segadaes, 2006).  $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$  and  $\text{K}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$  phase diagrams (Levin *et al.*, 1964; Kingery *et al.*, 1976) provide important information on the formation of viscous liquid at ternary eutectic temperatures. Hence, the glassy phase formed during firing fills the pores, and decreases the open porosity level of the ceramic bricks. This is consistent with the apparent density values (Fig. 5).

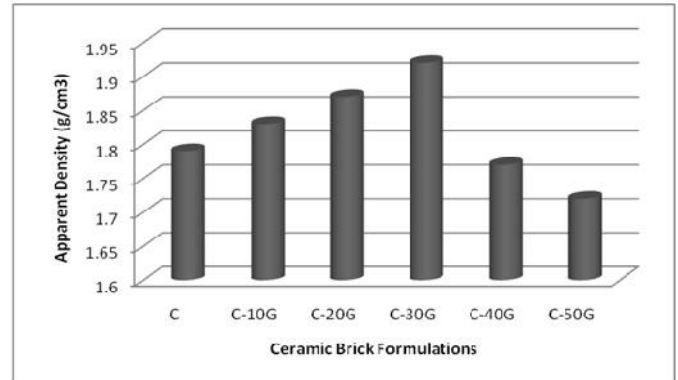


Fig. 5. Apparent density of the ceramic bricks versus waste content

Water absorption is used to estimate the pore ratio of brick specimens. High water absorption in brick is characterized by a high pore ratio. Milheiro *et al.* (2005) used the Brazilian specifications to limit the acceptable values of water absorption for bricks (WA 25%) and for roofing tiles (WA 20%). From this specification it is clear that all the ceramic brick samples satisfy Brazilian standard which can be seen in Fig 4.

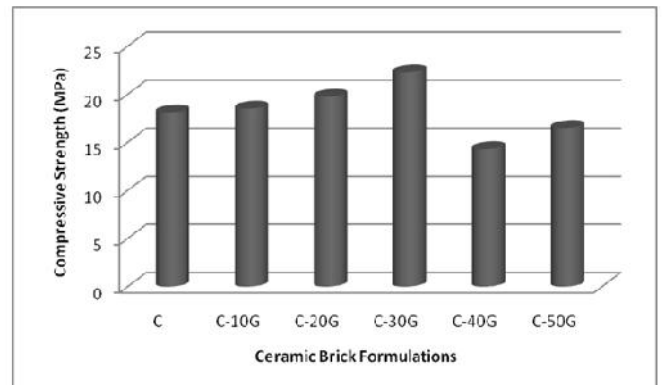


Fig. 6. Compressive strength of the ceramic bricks versus waste content

The compressive test is the most important test for assuring the engineering quality of a building material. The results (Fig.6) indicate that the strength is greatly dependent on the amount of grog in the brick excluding the samples C-40G and C-50G. Grog residue addition increases the compressive strength of the clay samples up to 30 wt. % ascribed to the enhancement of density. The densification of the samples is influenced by the sources of fluxing agents such as  $\text{K}_2\text{O}$ ,  $\text{Na}_2\text{O}$ , and  $\text{Fe}_2\text{O}_3$ , which favour the formation of a vitreous phase. The densification is caused also by the formation of mullite evidenced by XRD analysis which hence as poreless phases

substitute for porous metakaolinite at elevated temperatures (Baccour *et al.*, 2008; Milheiro *et al.*, 2005). The highest strength of the ceramic brick (C-30G) is associated with the lowest water absorption and highest bulk density as indicated in Figs. 4, 5 and 6. The achieved brick strength lies from 22.26 MPa to 14.28 MPa. According to BS 6073 (British Standards Institution, BS 6073, 1981) standards, compressive strength of bricks must be 15 MPa for a first-class brick and 10 MPa for a second-class brick. Therefore it is possible to incorporate up to 50 wt. % of grog into fireclay.

## Conclusions

In this study we demonstrated that it is possible to utilize grog as alternative raw material resources for the production of the ceramic brick. On the basis of the results reported in the present investigation, the following conclusions can be drawn.

- The predominant oxides present in fire clay and grog are  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$  which influence the refractoriness and strength of the final product.
- An increase in the content of the waste addition leads to (a) an increase in the density (b) decrease in water absorption. The result shows that the compressive strength is directly related to the grog content in the ceramic brick, which increases as the grog content of the ceramic brick compositions is increased. Compressive strength of the grog added brick is higher than the grog free reference brick. The obtained results showed that an incorporation of grog waste until a concentration of 30% (mass basis) leads to good quality bricks. In general, the mechanical properties of the obtained ceramic bricks were quite satisfactory under BS 6073 standard. The fluxing agents such as  $\text{K}_2\text{O}$ ,  $\text{Na}_2\text{O}$ , and  $\text{Fe}_2\text{O}_3$ , evidenced by chemical analysis and mullite formation according to XRD analysis are responsible for the densification process.
- The recycling of grog waste in ceramic bricks shows highly positive results in terms of environmental protection, waste management practices, and saving of raw materials.
- Further studies are needed to expand the results by using brick size as specimens in order to draw conclusion for manufacturing commercial ceramic bricks.

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