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Studies on Granite and Marble Sawing Powder Wastes in Industrial Brick Formulations

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Abstract: The main aim of this research is to study the utilization potential of granite and marble sawing powder wastes as alternative raw materials in the production of bricks. To safeguard the environment, efforts are being made for recycling different wastes and utilize them in valuable applications. Granite and marble sawing powder wastes is widespread by-product of industrial processes in India. Generally these wastes pollute and damage the environment due to sawing and polishing processes. Granite and marble wastes were collected from companies located in Salem District, Tamilnadu, India. Local clay and fired industrial brick samples were collected from nearby district namely, Erode, Tamilnadu, India. Mixtures were prepared with amounts of 0, 10, 20, 30, 40 and 50 wt. % of wastes incorporated into the raw clay and then fired at temperatures from 500 to 900°C in steps of 100°C in an electric furnace. Their characterizations were carried out with the determination of particle size, chemical composition, plasticity, XRD, SEM and Mossbauer spectroscopy. The technological properties such as compressive and flexural strengths, water absorption, porosity and bulk density were determined. The results showed that the granite and marble wastes can be added to the clay material with no determinal effect on the properties of the sintered bricks anticipating no costly modifications in the industrial production line.

Key words: Mossbauer spectroscopy, XRD, mechanical properties

INTRODUCTION

The main activity of ornamental stone industries are sawing and polishing process of rock blocks. This activity generates, per unity, a significant amount of mud basically composed of SiO_2, Al_2O_3, Fe_2O_3 and CaO, estimated in 32-40 m^3 day^{-1}. When the rock blocks are air dried, it becomes a very fine material that can be easily inhaled by human being and animals as well and it has been blamed as the reason for severe lung diseases among local people, which can cause serious damages in the environment, such as soil and underground water contamination, if not efficiently treated before disposal. Granite cutting industry produces large amounts of solid wastes world wide, which are expected to increase owing to the fact that the world production of granite industry has been increasing annually at a rate of 6% in the recent years. The wastes of this industrial activity can reach even 20 or 25 wt. % of the raw granite. The international trading is approximately US$ 6 billions per year and around US$ 13 billions, taking into account tools, equipments, etc. (Menezes *et al*., 2005; Torres *et al*., 2004; Saboya *et al*., 2007).

Now-a-days the cost of construction materials is increasing incrementally. In India the cost of cement during 1995 was Rupees 1.25 kg^{-1} and in 2008 the price increased around three times. In case of bricks the price was Rupees.0.66 per brick in 1995 and the present rate is Rupees.3.0 per brick. Similarly, over a period of 13 years from the year 1995, the price of sand has increased four times (Asokan *et al*., 2007). Also, due to high transportation costs of these raw materials, demand,

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environmental restrictions, it is essential to find functional substitutes for conventional building materials in the construction industry. In view of the importance of saving of energy and conservation of resources, efficient recycling of solid wastes is now a global concern requiring extensive R and D work towards exploring newer applications and maximizing use of existing technologies for a sustainable and environmentally sound management.

In the light of recycling, this study investigates the potential profit of incorporation of granite and marble wastes as raw materials in brick industrial formulations, particularly maximization of the use of granite and marble wastes in brick production, which would reduce both the environmental impact and the production cost of bricks.

MATERIALS AND METHODS

A typical clay mixture used in industry and fired brick were collected from Chinnathambipalayam located in Erode District and a dry granite and marble sawing wastes were collected from companies located in Salem District, Tamilnadu, India (Fig. 1). The characterization included particle size distribution (HORIBA LA-910), chemical composition (X-ray fluorescence (XRF), Phillips PW 1400), mineralogical composition, X-ray diffraction (XRD), Rikaku using CuK$_\alpha$ radiation at a wavelength of 1.5405 Å and 2θ range from 20 to 70°, Scanning Electron Microscopy (SEM) and energy dispersive spectrometry (EDS) in a JEOL equipment, model JSM 5610, LV and plasticity measurement.

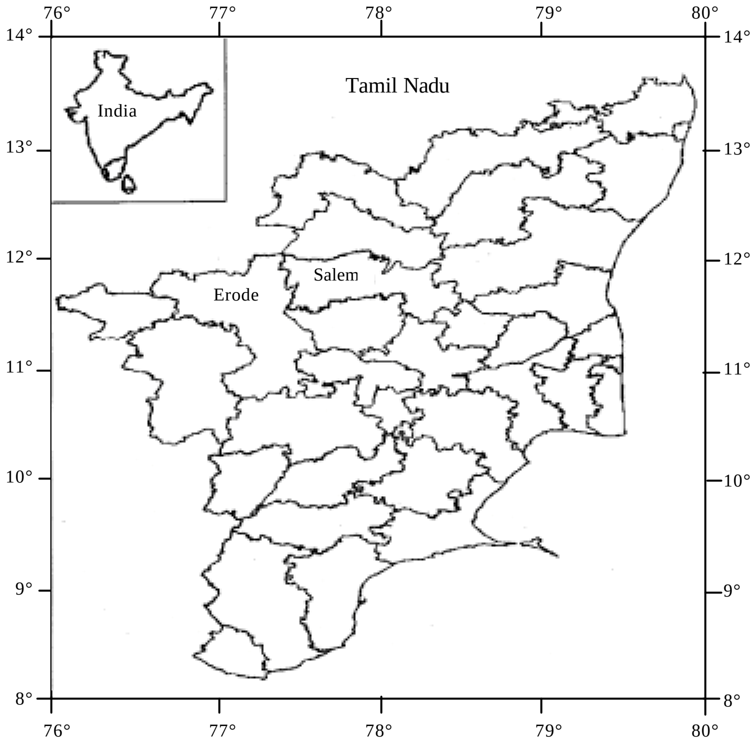


Fig. 1: Location map of clay and wastes collected (clay-Erode, granite and marble waste-Salem)

^{57}Fe Mossbauer spectra were recorded at room temperature (RT) in transmission mode with a conventional constant acceleration spectrometer (M/s WISSEL Germany) equipped with a ^{57}Co (Rh) source. Absorbers were prepared from powdered sample with a thickness of 100±10 m using a PMM compression holder. The spectra were computer fitted to a sum of Lorentzian lines by applying the constraints of equal line width and area for the two peaks of each doublet and equal line width and areas in the ratio 3:2:1:1:2:3 for the six peaks of sextets. Isomershifts (δ) were referred to the centroid of the spectrum of α-Fe at RT. To study the mechanical properties of wastes mixed bricks, the wastes were mixed with raw clay at 0, 10, 20, 30, 40 and 50 wt. % and briquettes samples of size (5.0×2.5×2.5 cm) were prepared. Mixing was made in a planetary mill and minimum of 80 briquettes were manually shaped at workable consistency and the specimens were dried in an oven to 110°C for 24 h. Briquettes specimens were sintered at temperatures between 500 and 900°C for 2 h in an oxidizing atmospheric condition with a heating rate of 10°C min^{-1}. After firing at selected temperatures, the specimens were subjected to several tests in order to verify their technological properties i.e., compressive and flexural strengths, water absorption, porosity and bulk density. The mechanical strength of the sintered specimens were measured with a universal testing machine in a three-point bending test of a constant cross-head speed of 0.5 mm min^{-1}. Water absorption, porosity and bulk density of the respective specimens were determined by using the Archimedes water displacement methods.

RESULTS AND DISCUSSION

Characterizing Tests

It can be observed that the wastes have 80% of slit size and 20% of fine sand, indicating that this material can be classified as a slit clay-like material. These results also point out that the wastes have a granulometry similar to the processed conventional non-plastic raw materials, such as quartz and feldspar, without additional grinding (Fig. 2).

For clay, it can be observed that 45% of the particle size distributions are of clay fraction, whereas 24% are of slit size. The fine sand proportion is about 26%. The coarser fraction of the clay i.e., that with particle size above 200 μm, corresponds to 5% of coarse sand. The intermediate amount of slit fraction helps the development of plasticity of ERE clay.

The plasticity parameters of the compositions for the production of bricks (clay and wastes), in terms of the Atterberg limits, are shown in Table 1. Vieira *et al.* (2008) reported that the plasticity index, is associated with the range between plastic and sludge consistency. For practical purposes, the plasticity index must be above 10%. Clays that present values of plasticity index lower than 10% are

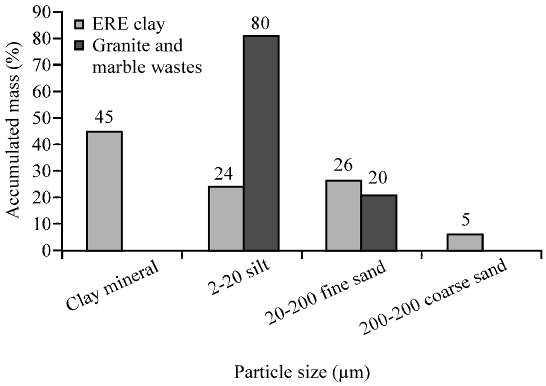


Fig. 2: Particle size distribution of the raw materials

Table 1: Liquid and plasticity limit and plasticity index

	ERE clay (wt. %)					
Parameters	0	10	20	30	40	50
Liquid limit (%)	43.2	40.0	38.0	34.5	36.0	28.6
Plasticity limit (%)	24.1	21.3	20.4	18.6	19.1	17.3
Plasticity index (%)	19.1	18.7	17.6	15.9	16.9	11.3

Table 2: Chemical compositions of the raw materials

Raw materials (%)	H_2O^-	SiO_2	Al_2O_3	TiO_2	Fe_2O_3	CaO	MgO	MnO	P_2O_5	Na_2O	K_2O	LOI	Total
ERE	1.22	70.18	12.92	0.05	2.72	2.80	2.20	0.01	0.03	0.76	1.85	5.24	99.98
WSE	2.13	66.04	14.25	0.02	2.83	4.48	2.42	0.02	0.01	1.08	1.78	5.10	100.16

not appropriate for building-related ceramic production due to the risk of problems during the extrusion process. In the present analysis, It can be observed that the values of liquid limits range from 28.6 to 43.2% which are in agreement with the range define in the literature (30-60%) to composition used for production of bricks. The plasticity limits ranged from 17.3 to 24.1%, in accordance with the values (15-30%) indicated to ceramic bodies used in the production of bricks by extrusion. The plasticity index ranged from 11.3 to 19.1%. It has been reported that the compositions with upto 45% of wastes were classified as high plastic (plasticity index >15%) while the formulations with 50, 55 and 60% of wastes were classified as medium plastic (7% < plasticity index < 15%). According to literature, these results are in agreement with the set of values (10-30%) indicated as appropriate to the production of bricks by extrusion.

Table 2 gives the chemical composition of the raw materials used in this study. The chemical composition of clay is typical of a kaolinite-based material. The clay material presents a typical composition and are constituted mainly by silica and alumina and minor contents of Mg, Ti, Mn, Na and K oxides, whereas the content of calcium oxide in clay material is 2.80% and in waste material slightly higher than clay material as 4.48%. The significant amount of iron oxide (2.72 wt. %) is responsible for the reddish colouring of the sintered samples. Saboya *et al.* (2007) reported that if the clay contains SiO_2 and Al_2O_3 at around 50 and 20% are the indicative of its suitability for producing fired ceramic bodies. The loss on ignition (~5%) is within the usual range for clay mixtures and is most likely associated with volatile components, organic matter burn-off and/or carbonate decomposition (Segadaes *et al.*, 2005). The medium and a minimum content of alkaline earth oxide (particularly CaO and K_2O), present in the reject material will act as a fluxing agent during the sintering process.

Figure 3 and 4 shows the differences between the X-ray diffraction patterns obtained for the raw clay material, industrial brick, granite and marble wastes and 20 wt. % of wastes mixed brick fired at 900°C. The diffraction pattern of the ERE clay and industrial brick have peaks of quartz and plagioclase (albite, anorthite) while in the 20 wt.% of granite and marble waste mixed brick fired at 900°C have the peaks of quartz and plagioclase as the main minerals. Some traces of calcite, dolomite and hematite were also been detected. As to powder granite and marble, the X-ray diffraction analysis shows intense peaks of quartz and plagioclase and discrete presence of calcite and traces of dolomite orthoclase and hematite minerals. Wagner *et al.* (1999) reported that the presence of higher amount of quartz makes the clay-self tempered during firing process. In the present study, clay, industrial brick, 20 wt. % waste mixed brick, fired at 900°C and wastes show the presence of quartz as main mineral. The crystalline phases identified by XRD are in agreement with the results obtained by XRF.

Figure 5 shows the SEM/EDS analysis of the fracture surface of sintered clay and waste mixed brick samples at 900°C, respectively. It is observed that sintered clay and waste mixed brick still displays large quartz grains which is confirmed by the absorbance of strong quartz peaks in the XRD spectrums. The SEM/EDS analysis fits well with the results of chemical analysis, with the presence of Si, Al, Fe, Ca, K, Mg and Na. Clay sample fired at 900°C, shows the presence of a significant amount of porosity. On the contrary, the samples containing waste additions show clean signs of vitrification and a higher degree of closed porosity.

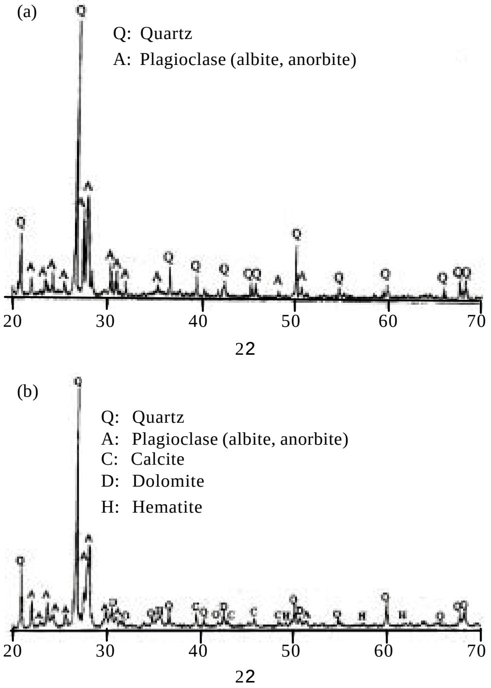


Fig. 3: X-ray diffraction patterns of raw materials. (a) Clay and (b) Granite waste

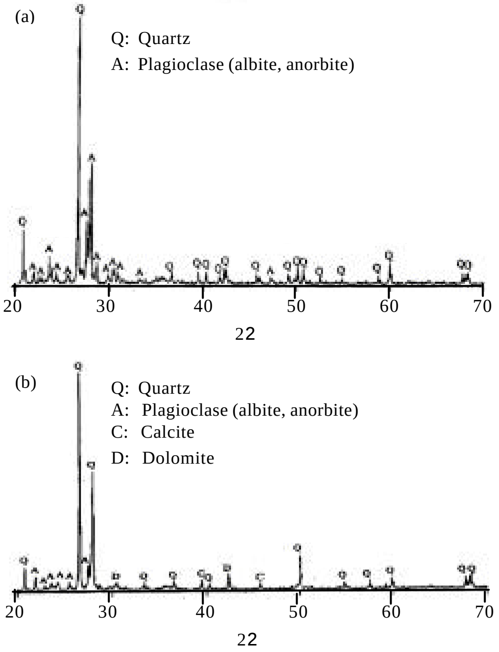
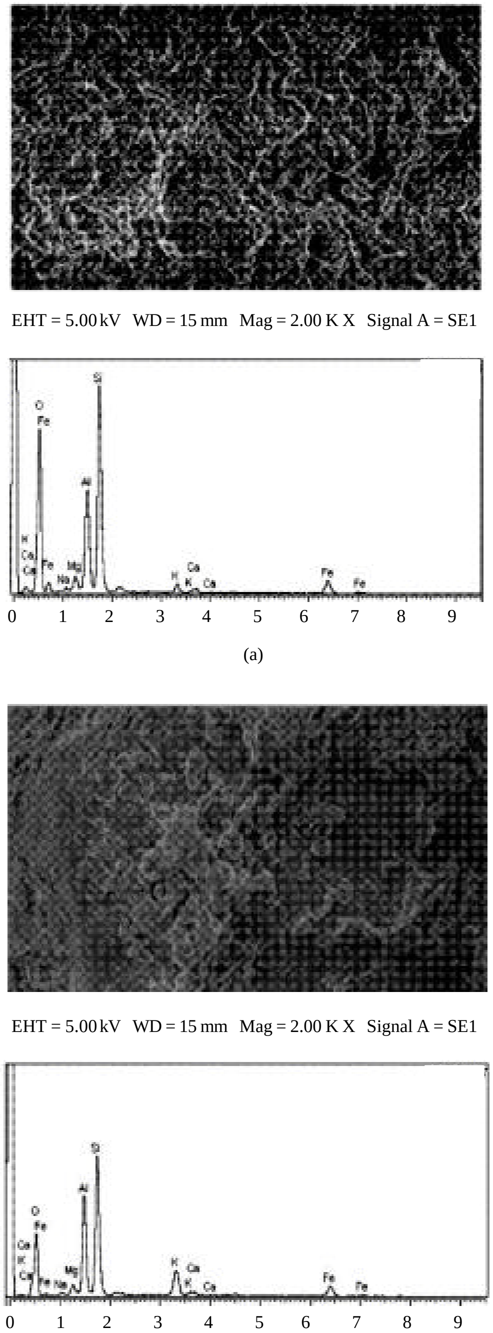


Fig. 4: X-ray diffraction patterns of fired materials. (a) Industrial brick and (b) Clay +20 wt. % waste at 900°C



(a)

(b)

Fig. 5: SEM micrographs EDS spectrum of the fracture surfaces of samples sintered at 900°C (a) Clay mixture and (b) Clay +20 wt. % waste

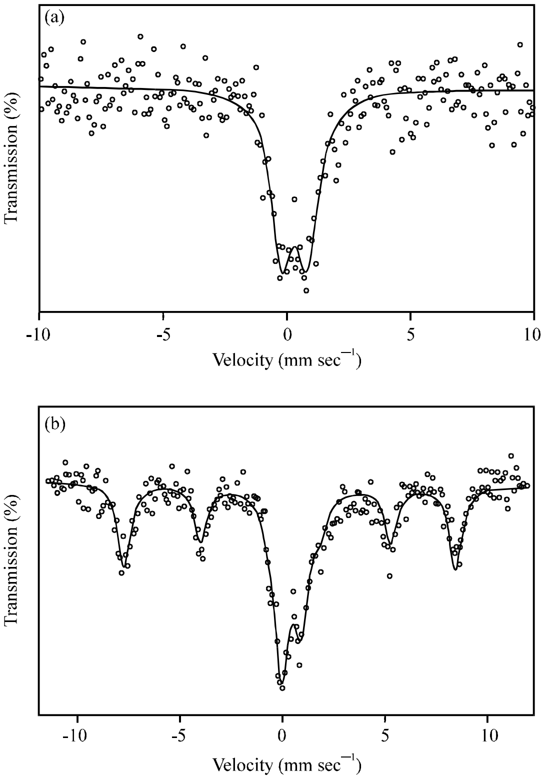


Fig. 6: Room temperature Mossbauer spectrum. (a) Industrial brick and (b) Clay +20 wt. % at 900°C

Iron is generally present in unpurified clays in concentrations of several percent. During firing, the iron undergoes characteristic changes on its chemical and physical state, depending on the Kiln atmosphere and on the maximum firing temperature reached. These changes can be followed by Mossbauer spectroscopy (Wagner *et al.*, 1997, 1998). Room temperature Mossbauer spectra were recorded for industrial brick as well as waste mixed brick fired at 900°C are shown in Fig. 6a, b. Mossbauer spectrum of industrial brick shows only the presence of paramagnetic doublet in the central part of the spectrum is attributed to Fe^{3+} species having an isomer shift of 0.374 mm sec^{-1} and quadrupole splitting of 1.205 mm sec^{-1}. The presence of Fe^{3+} species in industrial brick indicating that brick was fired in an oxidising atmosphere reflect red colour of the brick (Ramasamy *et al.*, 1989). Quadrupole splitting values of the doublet allow to infer a rough estimation of the firing temperatures as below and above 800°C. Wagner *et al.* (1999) reported that during heating in air, there is a strong increase of quadrupole splitting of Fe^{3+} species in the fresh clay from 0.66 to 1.05 mm sec^{-1} on firing at 400°C. Researchers have also been pointed out that the splitting reaches a maximum of 1.35 mm sec^{-1} on firing at 700°C. From these informations and the observed value of quadrupole splitting 1.205 mm sec^{-1}, it is roughly estimated that the industrial brick might have been fired at around 600°C during its manufacturing.

Room temperature Mossbauer spectrum of waste mixed brick fired at 900°C, shows a characteristic six-line magnetic pattern (δ = 0.366 mm sec^{-1} and Δ = -0.275 mm sec^{-1}) associated exclusively with a paramagnetic Fe^{3+} doublet having an isomer shift of 0.386 mm sec^{-1} and quadrupole splitting of 0.927 mm sec^{-1}. The observed six-line magnetic pattern is assigned to α-Fe_2O_3 (magnetically ordered and well crystallized hematite) present in the sample (Wagner *et al.*, 1994). In addition, it is confirmed by the obtained effective internal magnetic field of 50 Tesla (Tominaga *et al.*, 1978).

Technological Tests

Compressive and Flexural Strengths

The quality of brick can be studied by examining the compressive and flexural strength of brick. The results of compressive and flexural strength testing of the industrial bricks and waste mixed bricks fired at the different temperatures are shown in Fig. 7a, b. It is observed that the compressive and

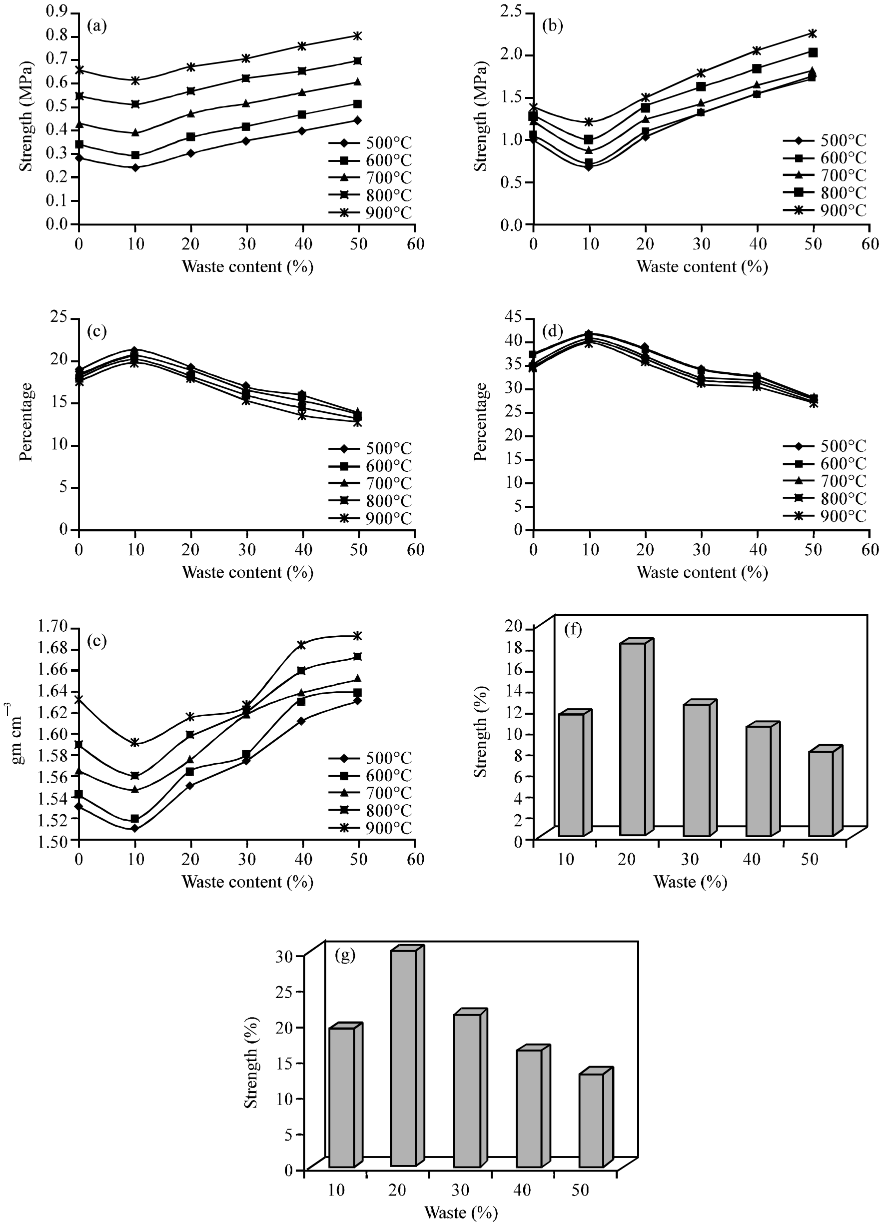


Fig. 7: The most relevant properties of bricks (a) Compressive strength, (b) Flexural strength, (c) Water absorption, (d) Porosity, (e) Bulk density, (f) Compressive strength vs percentage and (g) Flexural strength vs percentage

flexural strength values are directed proportionate to the waste percentage. Both strength gradually increases with an increase in the waste content. It is reported that B 6073 requires 0.65 MPa as a minimum flexural strength for the building materials to be used in structural applications. All the waste mixed brick and as received industrial (fired) brick tested for flexural strength satisfy this requirement. One more point to be noted that the average value of compressive and flexural strength values at 20 wt. % are 19.82 and 30.61 MPa, respectively. These values are higher than that of the average strength values at 10, 30, 40 and 50 wt. %.

Water Absorption and Porosity

The water absorption rate, which refers to the weight of moisture in the pores compared to the sintered specimen's weight, is an effective index for evaluating the brick quality. The less water that infiltrates the brick, the greater its durability and resistance to the natural environment are expected.

Figure 7c shows the results of the water absorption tests for waste mixed bricks fired at five different temperatures. It can be observed that as the heating temperature increases, the amount of water absorption in the brick decreases. It is to be noted that when the waste content was increased the water absorption of the bricks increased. The smaller water absorption rate that occurred after heating of the higher temperatures (900°C), suggests that local liquid-phase sintering occurred, which contributed to a decrease in the pore volume and thus the water absorption rate.

The porosity test was carried out for waste mixed brick at 10, 20, 30, 40 and 50 wt.% at five different temperatures. From the Fig. 7d it can be noticed that the porosity values of wastes mixed brick at different wt.% is inversely proportional to the temperature of firing. Porosity value of the brick decreases as the temperature increases. The water absorption and porosity values observed in the present study are in good agreement with the maximum allowed value of water absorption and porosity for commercial use of the bricks (Saboya *et al.*, 2007).

Bulk Density

During sintering, open and closed pores are usually formed. The minimum density corresponds to the maximum volume of closed pores in the sample densification is a pore filling process that occurs during the liquid-phase flow and by pore shrinkage (Nowok *et al.*, 1990). The measurements of bulk density of samples with different proportions of wastes and fired at five different temperatures are shown in Fig. 7e. Clay brick normally have a bulk density of 1.8-2.0 g cm^{-3} (Lin, 2006). The results indicate that increasing the temperature results in an increase in the bulk density of the bricks as the wastes content increased.

CONCLUSIONS

The characterization and technological tests of incorporation of granite and marble sawing powder wastes in the production of bricks led the following conclusions:

- Recycling of granite and marble wastes in brick production anticipates safe for the health and environment
- The results obtained in this study show that 20 wt. % of granite and marble reject can be added to an industrial clay mixture already in use in the production of bricks with no major sacrifice of the properties of the final product
- The incorporation of the wastes up to 10 wt. % into clay slightly increases the water absorption and decreases the mechanical strength after the firing stage
- Finally, granite and marble sawing powder wastes may be recycled into brick manufacturing possibly in low percentage to avoid the damage and quality of bricks

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