

UTILIZATION OF GRANITE AND MARBLE SAWING POWDER WASTES AS BRICK MATERIALS

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Abstract: The main objective of waste management system is to maximize economic benefits and at the same time protection of the environment. Granite and marble process industry generates a large amount of wastes mainly in the form of powder during sawing and polishing processes, which pollute and damage the environment. Therefore, this work aims to characterize and evaluate the possibilities of using the granite and marble sawing wastes, generated by the process industries from Salem District, Tamilnadu state, India, as alternative raw materials in the production of bricks. Samples of clay material and fired industrial bricks were collected from nearby District namely Namakkal, India. Their characterization was carried out with the determination of chemical composition, mineralogical and petrological analysis, particle size, plasticity, FTIR, and Mössbauer measurements. Secondly, technological tests were conducted on wastes incorporated brick specimens in order to evaluate the suitability of addition of wastes in the production of bricks. The results showed that granite and marble wastes can be added up to 50 wt. % into the raw clay material in the production of bricks.

Key words: Clay material, Granite waste, Recycling, Mechanical analysis, Brick.

1. INTRODUCTION

Recycling waste as useful material is a very important environmental management tool for achieving sustainable development. On the other hand, recycling waste without properly based scientific research and development can result in environmental problems greater than the waste itself. The successful research and development of a new building material or component using waste as raw material, is a very complex and multi disciplinary task having technical, environmental, financial, marketing, legal and social aspects (John & Zordan 2001).

Now-a-days the cost of construction materials is increasing incrementally. In India, the cost of cement during 1995 was Rs.1.25/kg and in 2008 the price increased ~ three times. In case of bricks the price was 0.66 per brick in 1995 and the present rate is Rs.2.5 per brick. Similarly, over a period of 10 years from the year 1995 the price of

sand has increased four times. Also due to high transportation costs of these raw materials, demand, environmental restrictions, it is essential to find functional substitutes for conventional building materials in the construction industry (Pappu et al., 2007).

Growth of population, increasing urbanization, rising standards of living due to technological innovations have contributed to an increase both in the quantity and variety of solid wastes generated by industrial, mining, domestic and agricultural activities.

Granite mining and process industry are one of the most promising business areas of the mining sector, with a mean growth in the world production of approximately 6% per year in the last 10 years. 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).

In India, about 6 million tons of wastes from marble industries are being released from marble cutting, polishing, processing and grinding. In Tamilnadu state, Salem is an area that concentrates a large quantity of granite and marble process industries, which are responsible by the disposing of hundred of tons of wastes in the environment per year. This scene is even more aggravated by the increasing production in the last decade, getting attention from all society with the destination of disposal wastes. The granite and marble dust is usually possesses a major environmental concern. In dry season, the granite and marble powder or dust dangles in the air, flies and deposits on vegetation and crop. All these significantly affect the environment and local ecosystems. The granite and marble dust disposed in the river-bed and around the production facilities causes reduction in porosity and permeability of the topsoil and results in water logging. Further, fine particles results in poor fertility of the soil due to increase in alkalinity.

Use of industrial wastes and by products as an aggregate or raw material is of great practical significance developing building material components as substitutes for materials and providing an alternative or supplementary materials to the housing industry in a cost effective manner and the conservation of natural resources.

Therefore, the objective of this work is to study the possibility to incorporate granite and marble sawing powder wastes in brick products with no major sacrifice of the properties of the final product.

2. MATERIALS AND METHODS

A typical clay mixture used in the brick industry, fired industrial brick were collected from Ponnampalayam located in Namakkal District and a dry granite and marble sawing powder wastes, not beneficiated in any way, collected directly from compares located in Salem District, Tamilnadu, India, were selected and characterized. Figure 1 location map of clay materials and waste investigated. The characterization included chemical analysis, mineralogical and petrological analysis, particle size analysis, plasticity, liquid limit and FTIR analysis (Avator-330 FTIR, Thermo Nicolet) and Mössbauer measurements (M/s WISSEL, Germany). 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 x 2.5x2.5 cm) were prepared.

Mixing was made in a planetary mill and minimum of 100 briquettes were manually shaped at workable consistency and the specimens were dried in an oven to 110°C for 24h. Briquette specimens were sintered at temperatures between 500 and 900°C for two hour in an oxidizing atmospheric condition with a heating rate of 10°C/minute. After sintering 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 compressive strength was determined by dividing the maximum load with the applied load area of the brick samples. The flexural strength was measured with a universal testing machine in a three-point bending test of a constant cross-head speed of 0.5 mm/min. Water absorption, porosity and bulk density of the respective specimens were determined by using the Archimedes water displacement method.

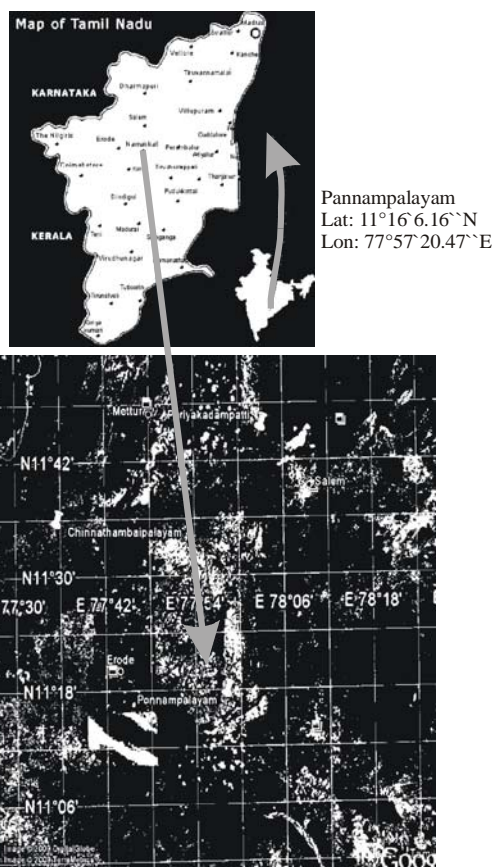


Figure 1. Location map of clay materials and wastes investigated (Namakkal and Salem).

3. RESULTS AND DISCUSSION

3.1. Characteristics of the raw clay material and granite and marble wastes

Table 1 gives the chemical composition of the clay material and the granite and marble sawing powder wastes. The clay material presents a typical composition and are constituted mainly by silica (68.76%) and alumina (10.20%) and minor contents of Ca, Mg, Na, K, Ti and Mn oxides. The significant amount of iron oxide (Fe_2O_3) is responsible for red or dark colouring of fired brick materials (Segadaes, et al. 2005). The loss on ignition (LOI) (7.98%) is within the usual range for clay materials and is most likely associated with volatile components and organic matter. Chemical composition of granite and marble sawing powder wastes consists basically of SiO_2 (66.04%), Al_2O_3 (14.25%) and CaO (4.48%). The loss on ignition is 5.10% results from the decomposition of carbonates (calcite and dolomite). The presence of alkaline earth oxide content (especially CaO and K_2O) in the waste material will act as a fluxing agent during the sintering process (Acchar, et al. 2006).

Table 1. Chemical compositions of the raw materials (%)

	H ₂ O-	SiO ₂	Al ₂ O ₃	TiO ₂	Fe ₂ O ₃	CaO	MgO	MnO	P ₂ O ₅	Na ₂ O	K ₂ O	LOI	Total
Namakkal clay	2.63	68.76	10.20	0.02	2.78	3.36	2.40	0.05	0.03	0.67	1.25	7.98	100.13
Granite and marble wastes	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

The petrographic observation of granite thin section (Photo-1&2) reveals the grains are equigranular, holocrystalline with medium grained. The grains are prismatic, subhedral showing hypidiomorphic texture. The observation reveals the major mineral is feldspar (50-55%) followed by quartz (40-45%). The feldspar grains are dominated by microcline followed by orthoclase and plagioclase. Perthite was also observed. Hypersthene and hornblende presence were noticed. There are few grains of less resistant minerals like biotite and muscovite.

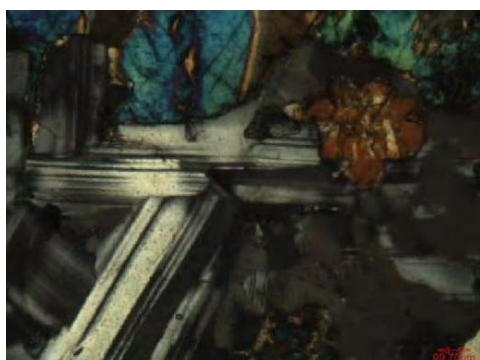


Photo 1



Photo 1

Photo 1 & 2. Thin section image of granite.



Photo 3

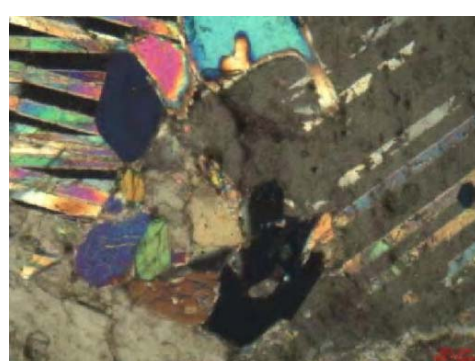


Photo 4

Photo 3 & 4. Thin section image of marble.

The presence of opaque minerals includes magnetite with wedge shaped sphene amounting to 2-3%. The quartz is characterized by anhedral nature with slight yellow interference colour. The plagioclase is characterized by subhedral grains with cleavage and exhibit multiple polysynthetic twinning (albite). The microcline is subhedral and prismatic in nature with cross-hatched twinning whereas the orthoclase in the sample signifies to the simple Carlsbad twinning. Brown to pink pleochroic biotite was observed with prismatic nature with one set of perfect cleavage.

The petrographic examination of the marble section (Photo 3 & 4) signifies the high content of calcite and dolomite (80%), scapolite(10%) with equigranular grains showing granoblastic texture. The remaining includes hornblende and hypersthene with few grains of magnetite. The detailed study under the microscope reveals that calcite grains are having rhombohedral cleavage with twinkling effect and with extinction at 180° giving anomalous interference colour. The hornblende grains are light green to dark green with the presence of cleavage. The grains exhibit 2nd order green interference colour. Hypersthene grains in this section are brown to pink pleochroic, subhedral with prismatic cleavages and straight extinction.

The particle size distribution of clay material as well as wastes are depicted in figure 2, where it can be seen that for the wastes, 80% are of silt size and 20% of fine sand size, indicating, thus, that this material is very fine grained in nature. These results point out that the granite and marble wastes have a granulometry similar to the processed conventional non-plastic raw materials, such as quartz and feldspar without additional grinding.

In the clay material, it can be observed that 49% of the particle size distribution are of clay fraction, where 23% are of silt size. The fine and coarse sand proportion is 24 and 4% respectively. In the present work, the liquid and plasticity parameters of clay material and waste mixed briquette specimens at various wt. % shows the gradual decrease in its value may be due to the presence of higher amount of silt size particles in the wastes compared to that of the clay material. Even though, these parameters do not affect the mechanical properties of the reformulated briquette bars sintering at different temperatures.

Plasticity is an important parameter for the production of constructional clay brick. Because, brick industry mainly uses extrusion shaping and therefore insufficient plasticity creates extrusion failures and heterogeneities in clay body and this cause lower mechanical properties. The plasticity limit, PL, corresponds to the amount of water necessary for the material to reach the plastic consistency, which makes it possible to be formed by extrusion. The liquid limit, LL, corresponds to the amount of

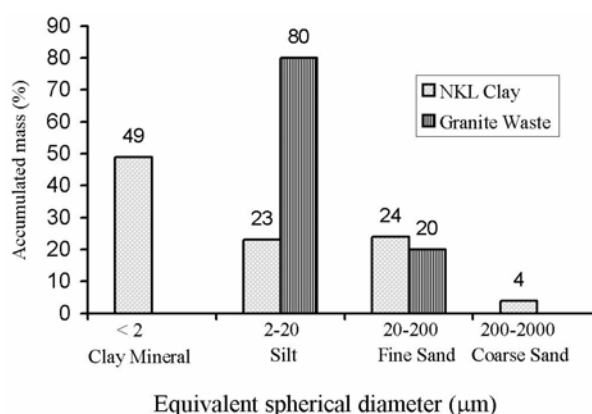


Figure 2. Particle size distribution of the raw materials.

water necessary for the material to loss its plastic consistency. The plasticity index, PI, is associated with the range of plastic consistency. For practical purposes, the plasticity index must be above 10% (Monteiro, et al. 2008). Less than 10% of PI is due to the risk of developing problems during the extrusion process. These problems are related to a possible variation in the amount of extrusion water, causing in appropriate dimensional changes and even cracks in the pieces. The plasticity parameters of the compositions for the production of bricks (clay material and wastes), in terms of the Atterberg limits, are shown in table 2. It can be observed that the values of liquid limit of Namakkal clay material ranged from 32.3 to 46.3%. The plasticity limit ranged from 19.2 to 24.4%. The plasticity index ranged from 13.1 to 21.9%. These results are in agreement with the range defined in the literature and indicated as appropriate to the production of bricks by extrusion (Menezes, et al. 2005).

Table 2. Liquid and plasticity limit and plasticity index

Namakkal Clay	0%	10 %	20 %	30 %	40 %	50 %
Liquid limit (%)	46.3	43.0	41.2	36.1	38.3	32.3
Plasticity limit (%)	24.4	22.6	21.3	19.1	20.1	19.2
Plasticity index (%)	21.9	20.4	19.9	17.0	18.2	13.1

3.2. FTIR analysis

The present study is aimed to asses the original firing temperature, firing atmosphere, type of clay, minerals presence and coloring mechanism of industrial brick collected from Namakkal District. This study is also intended to monitor any structural changes takes place by mixing granite and marble wastes into raw clay material sintered at different temperatures. Figure 3 and 4 shows the room temperature FTIR absorption spectra of Nammakkal clay material in the as received state and at different sintering temperatures (100-900°C). Figure 5 shows the room temperature FTIR absorption spectrum of industrial brick (fired) in the as received state and refired in the laboratory at 900°C. Figure 6 shows the room temperature FTIR absorption spectra of 20 wt. % waste mixed brick sintered between 500 and 900°C.

Room temperature FTIR spectrum of Nammakkal clay sample in the as received state shows the presence of characteristic very weak hydroxyl bands at 3700 and 3620 cm^{-1} , which indicates that Namakkal clay is belonging to disordered kaolinite type (Ramasamy & Venkatachalapathy 1992). Manoharan et al. (2007) reported that when clay is fired between 300 and 500°C, dehydroxylation of octahedral layers of most clay minerals takes place. In the present study, on firing the Namakkal clay at 500°C, the existence of the above bands are disappeared means that dehydroxylation of octahedral layers takes place. It can also be noticed that the presence of 1100 and 915 cm^{-1} band are as very weak shoulders centered at around 1030 cm^{-1} , which are attributed to O-H deformation and Si-O-Si modes.

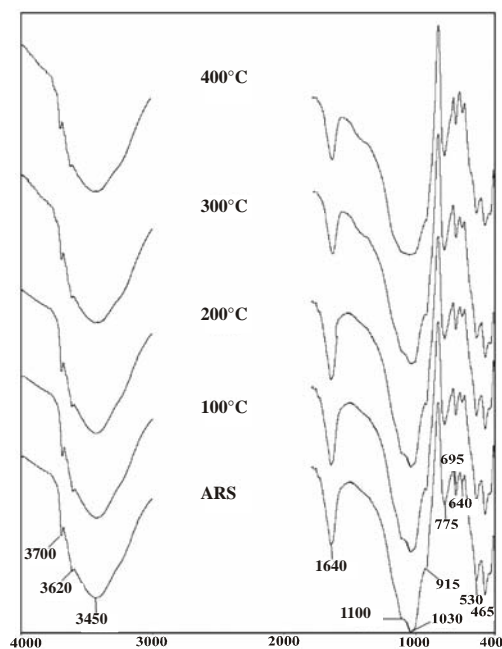


Figure 3. Room temperature FTIR spectra of Namakkal Clay in the as received state (ARS) and at 100, 200, 300, 400°C.

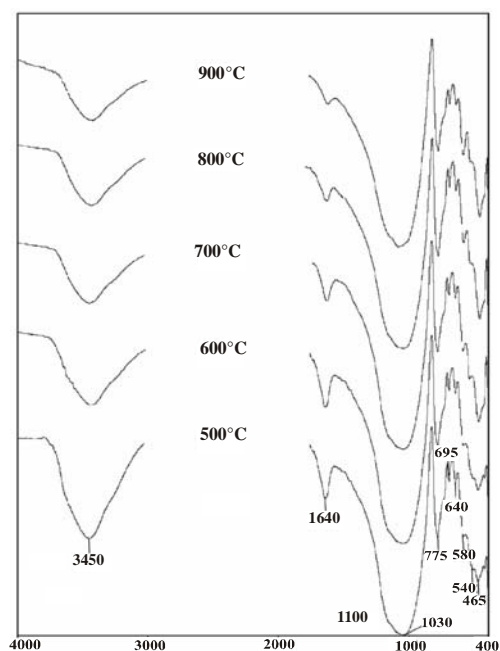


Figure 4. Room temperature FTIR spectra of Namakkal Clay at 500, 600, 700, 800 and 900°C.

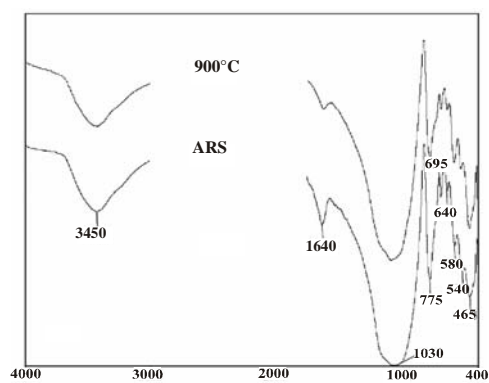


Figure 5. Room temperature FTIR spectra of Namakkal fired clay brick in the as received state and refired at 900°C in the laboratory.

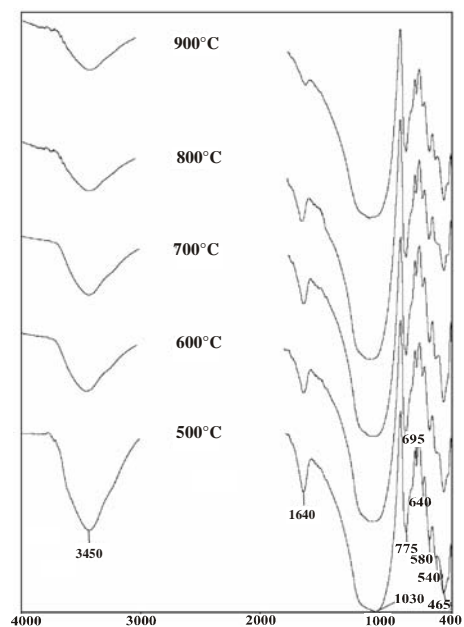


Figure 6. Room temperature FTIR spectra of granite marble waste mixed brick sintered at different temperatures.

Wagner et al. (1999) reported that during heating the clay, kaolinite disappears at 400-450°C. At around 600°C, the expandable layer silicates collapse are resulting the disappearance of the bands 1100, 915 cm^{-1} and appearance of a very strong and a broad symmetry band centered at around 1030 cm^{-1} for red clay and 1080 cm^{-1} for white clay (Ghosh 1978). In the present work, a broad symmetry band observed at around 1030 cm^{-1} of the Namakkal clay indicates that red clay out of which conventional bricks were produced. In order to assess the maximum original firing temperature (>600°C) of clay-based materials, the presence / absence or increase / decrease in intensities or shifting of the band position from 540 to 580 cm^{-1} are playing a vital role, which are attributed to iron oxides. In the present investigation, as received state FTIR spectrum of Namakkal clay shows the presence of a strong band at 540 cm^{-1} . During firing at different temperatures up to 900°C in steps of 100°C, it can be observed that particularly at 500°C and above, intensity of the band 540 cm^{-1} is gradually reduced and shifted to 580 cm^{-1} . At 800 and 900°C, relative intensity of characteristic band 580 cm^{-1} was observed as medium whereas 540 cm^{-1} as very weak shoulder. Manoharan et al. (2008) have reported that the band 580 cm^{-1} is attributed to magnetically ordered and well crystallized hematite present in the sample and is possible above 750°C under oxidizing atmospheric conditions, which can also be confirmed by subjecting the same sample to Mössbauer spectroscopic technique (Wagner, et al. 1992).

The presence of a sharp and medium band at 775 cm^{-1} coupled with weak band at 695 cm^{-1} is due to the presence of quartz (Si-O) (Russel 1987). Generally sufficient amount of quartz and feldspar are often present in clays and which makes the clay to be self-tempered during heating (Wagner, et al. 1999). The band occurring at 465 cm^{-1} is assigned to mixed Si-O-Si bending mode. These bands are free from any temperature effects (Venkatachalapathy et al., 2004).

Room temperature FTIR spectrum obtained for the as received state of Namakkal industrial brick (fired final product) is well compared with the FTIR spectrum of Namakkal clay obtained by firing at 800°C in laboratory condition (oxidizing atmosphere). So, it is estimated that Namakkal industrial brick must have been fired around 800°C during its manufacturing. Further, this estimation is confirmed by the absence of the bands at 3700, 3620, 1100, 915 cm^{-1} and the presence of a medium band at 580 cm^{-1} in the as received state Namakkal industrial brick. The medium band observed at 580 cm^{-1} is due to the firing nature of Namakkal brick under oxidizing atmosphere which reflects red colour of the material (Venkatachalapathy et al., 2003; 2004).

The characteristic bands observed at 3450 cm^{-1} and 1640 cm^{-1} are attributed to OH stretching and H-O-H bending vibrations of adsorbed water molecules. During heating the clay based materials, these bands will be diminished and disappeared (Wolff, 1963). In the present study, the existence of these bands up to 900°C might have been due to the absorption of moisture in the atmosphere by spectroscopic grade KBr while recording FTIR spectra.

It is interesting to note that FTIR spectra of granite and marble waste mixed brick obtained at 500, 600, 700, 800 and 900°C are almost identical as the FTIR spectra obtained for Namakkal clay material at the same temperatures. Eventhough, the reason for a sharp band at 580 cm^{-1} at 500°C on waste mixed brick is due to the presence of higher amount of iron oxides in the wastes.

From the above results, it is found that by mixing granite and marble wastes into raw clay materials, no major structural changes are observed at higher temperatures (500-900°C).

3.3. Mössbauer measurement

The main aim of this analysis is to confirm the results obtained through FTIR spectroscopic technique.

Manufacturing procedures of any clay-based materials can be assessed by ^{57}Fe Mössbauer spectroscopic technique. The study based on the fact that iron bearing clay minerals as well as associated minerals including iron oxides undergoes characteristic chemical and physical changes during firing. These changes depend on the firing temperature and on the Kiln atmosphere.

The wide applicability of ^{57}Fe Mössbauer spectroscopy for investigations of clay-based materials arises from the fact that practically all clays contain iron, usually in concentrations between 1 and 10 wt. %. Natural iron contains only about 2% of the ^{57}Fe isotope used in Mössbauer spectroscopy, but this is sufficient for measuring good Mössbauer spectra of most clay-based materials (Wagner, et al. 2000). Generally, iron in clays may be present as structural Fe^{2+} or Fe^{3+} species in the silicate structures of the clay minerals, in other silicates, and in particles of iron oxides or oxyhydroxides adhering to the clay mineral particles. The structural iron in silicates is paramagnetic at room temperature; therefore, it usually gives rise to electric quadrupole doublets in the Mössbauer spectra. Well-crystallized iron oxides show the characteristic six-line pattern arising from the magnetic hyperfine interactions. Murad & Wagner (1989) and Wagner et al. (1999) reported that when firing the clay-based materials in an oxidizing atmosphere, the quadrupole splitting of octahedrally co-ordinated Fe^{3+} species increases abruptly from 0.66 mm/s in fresh clay to 1.05 mm/s on firing at 400°C takes place. The splitting reaches a maximum of 1.35 mm/s on firing at 700°C, but reverts to lower values upon the formation of new and better ordered phases (hematite formation-six line pattern) at higher temperatures (>750°C).

In the present investigation, room temperature Mössbauer spectrum of Namakkal industrial brick in the as received state shows the presence of a

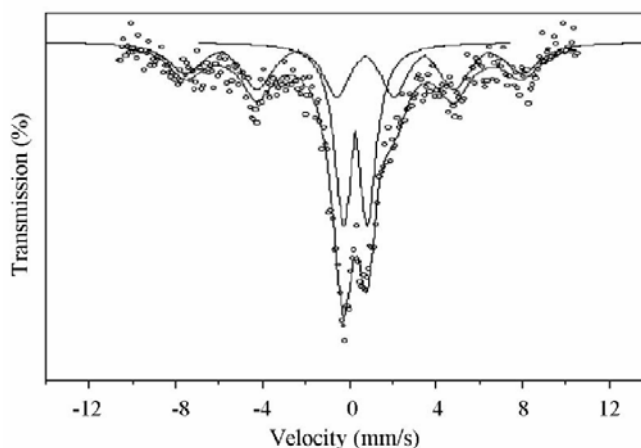


Figure 7. Room temperature Mössbauer spectrum of fired industrial clay brick in the as received state.

characteristic initial formation magnetic six-line pattern is attributed to Fe_2O_3 (hematite) having isomer shift of 0.43mm/s and quadrupole splitting of 0.13mm/s with internal magnetic hyperfine field value of 502 kOe. Tominaga et al. (1978) characterized Ancient Japanese roofing tiles by ^{57}Fe Mössbauer spectroscopy and reported that reddish clay-based materials will have the effective field value of 502 to 507KG, slightly smaller than the value (517KG) for the commercially available hematite ($\alpha\text{-Fe}_2\text{O}_3$). The spectrum also shows the presence of second component as Fe^{3+} species having isomer shift of 0.30mm/s and quadrupole splitting of 1.07 mm/s. The fractional area of the sextet and doublet were calculated as 55 and 44%.

Formation of Fe_2O_3 (hematite), revert quadrupole splitting value 1.07mm/s (Fe^{3+} species) and internal magnetic hyperfine field value 502kOe are indicates that Namakkal brick must have been fired between 750 and 800°C under oxidizing atmospheric condition which reflects reddish colour of the brick. Mössbauer results are well agreed with the results obtained through FTIR studies.

3.4. Technological Properties

The quality of bricks can be examined by measuring the compressive and flexural strengths. Figure 8 and 9 shows the results of the compressive and flexural strength values obtained from the tests. The compressive strength is determined by dividing the maximum load with the applied load area of the brick samples. The flexural strength is determined by the three point bending test of a constant cross-head speed of 0.5mm/min. From the results, one should notice that granite and marble wastes additions tend to increase compressive and flexural strengths of reformulated brick as the sintering temperature increases. It is also observed that the average compressive and flexural strength values obtained at 20 wt. % is higher than that of other wt. %.

Figure 11 shows the bulk density of the elaborated compositions.

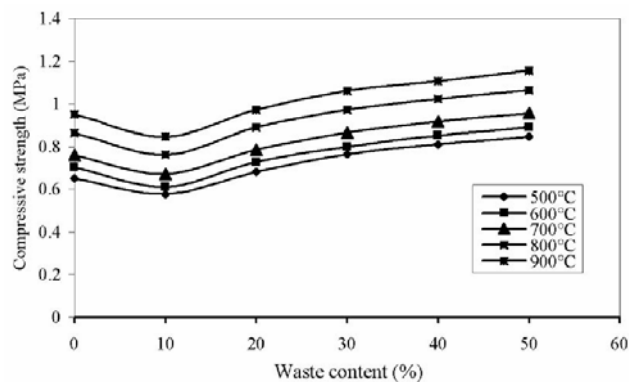


Figure 8. Variation of compressive strength with waste content.

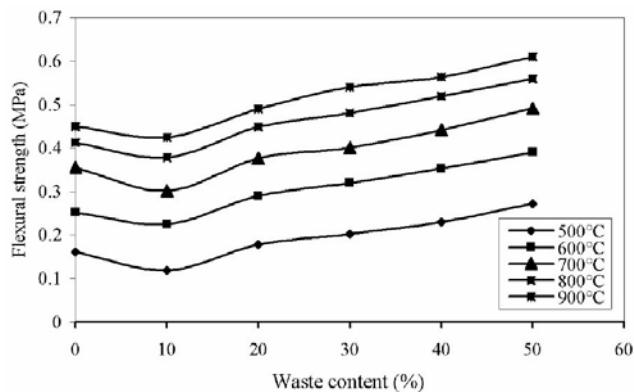


Figure 9. Variation of flexural strength with waste content.

From the results, one should observe that wastes additions tend to increase the bulk density of the reformulated bricks as the sintering temperature increases. This behavior is due to the silt and fine sand particle size of the granite and marble waste as compared with the clay, which favors the packing of the system as well as the higher real density of the waste. These are the reasons in the reduction of porosity values of reformulated bricks at various wastes wt. % as the sintering temperatures increases (Fig. 12).

Figure 10 shows the changes in water absorption of sintered specimens as a function of the sintering temperatures and waste content, respectively. It is observed that the water absorption values decreases significantly at 800 and 900°C. This behavior is related to the lower viscosity of the liquid phase and the consequently improvement on the densification process produced at these temperatures. From figure 12, it can be observed a decreasing trend of the values of porosity of reformulated bricks as the sintering temperature increases, which is related with the fusion and vitrification of the granite and marble wastes that acted as fluxes in the studied temperatures.

The above properties such as compressive and flexural strength, water absorption, porosity and bulk density measured for the as received state Namakkal industrial brick is almost same as exhibited by the reformulated bricks at different sintering temperatures especially between 800 and 900°C.

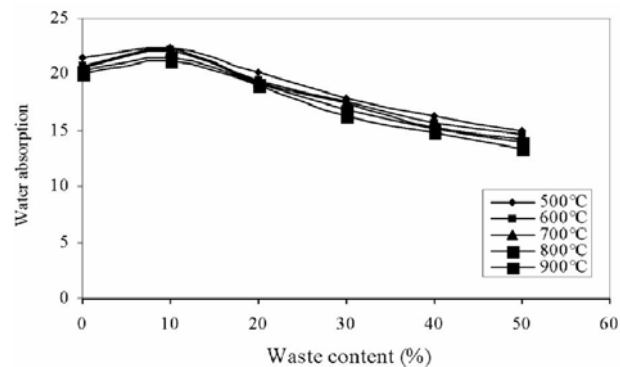


Figure 10. Variation of water absorption with waste content.

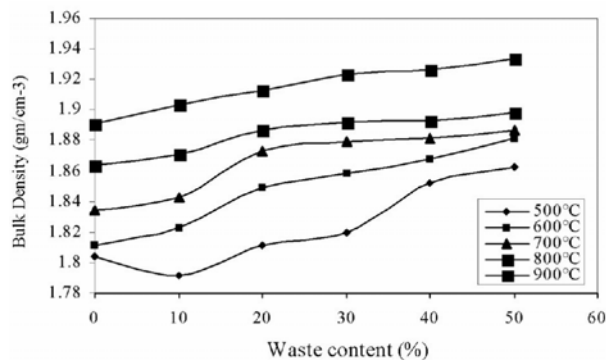


Figure 11. Variation of bulk density with waste content.

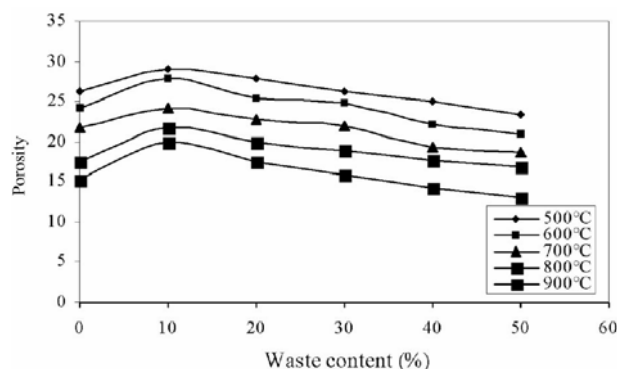


Figure 12. Variation of porosity with waste content.

4. CONCLUSIONS

The characterization and tests of Namakkal industrial brick, granite and marble wastes and its incorporation into clay material used for brick manufacturing, led to the following conclusions.

- It is assessed that Namakkal industrial bricks were fired at around 800°C during its manufacturing under oxidizing atmospheric condition.
- Red type clay out of which this brick has been made in industry.
- The physical property studies, records that the addition of granite and marble waste mixture imparts physical strength to the bricks when they are kilned at higher temperature. More specifically, bulk density, compressive strength, flexural strength was found to increase due to the addition of the above mixtures. This is because of the fact that the addition of the mineral matter especially quartz and feldspar to the clay, act as flux when they are kilned at higher temperature as evidenced by the physical test of the bricks.
- From the results of technological tests, it is suggested that granite and marble wastes can be incorporated up to 50 wt. % into clay materials for the production of bricks.
- The incorporation of granite and marble wastes has negligible effect on the mechanical properties during the entire process, anticipating no costly modifications in the industrial production line.
- The possibility to use the granite and marble wastes as an alternative raw material in the production of clay-based products will also induce a relief on waste disposal concerns.

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