



AN INVESTIGATION ON THE EFFECT OF INCORPORATION OF GRANITE AND MARBLE WASTES IN THE PRODUCTION OF BRICKS

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ABSTRACT

Sawing and polishing process of granite and marble industry generates large amount of wastes, which can seriously pollute and damage the environment. Therefore, this work intends to study the suitability of incorporation of granite and marble wastes in brick products. Samples of clay, industrial brick (fired) and wastes were collected from companies located in Salem District, Tamilnadu, India. Fired industrial brick was characterized by using FTIR and Mossbauer spectroscopic techniques. Secondly, several technological tests were conducted in order to evaluate the suitability of incorporation of wastes in brick production. The results showed that granite and marble waste can be added to an industrial clay mixture, already in use in the production of bricks with no major sacrifice on the properties of the final product.

Keywords: bricks, granite, marble, clay, wastes, suitability, strength.

INTRODUCTION

During different industrial, mining, agriculture and domestic activities, India produces annually about 960 million tonnes of solid wastes as by-products, which pose major environmental and ecological problems besides occupying a large area of land for their storage or disposal. Looking to such huge quantity of wastes as minerals or resources, there is a tremendous scope for setting up secondary industries for recycling and using such solid wastes in construction materials [1].

The use of industrial waste materials as additives in the manufacture of clay-based products like bricks and tiles has been attracting a growing interest from researchers in recent years and is becoming common practice [2, 3]. The reasons for the motivation are, the exhaustion of the natural resources, the conservation of not renewable resources, improvement of the population health and security and preoccupation with environmental matters. The alternative raw materials can also contribute to diversify the offer of raw materials in the production of bricks and reduce the costs in a building. Granite and marble is a natural hard igneous rock formation of visibly crystalline texture formed essentially of SiO_2 , Al_2O_3 , Fe_2O_3 and CaO [4, 5]. It is especially used for building and monuments. In Tamilnadu, granite and marble industry have been increased especially in Salem District in the last years, consequently the volume of residue disposed in the decant lagoons or landfills increased, which results environment problems.

Therefore, the main objective of this work is to characterize and evaluate the effect of incorporation of granite and marble sawing powder wastes up to 50 wt. % into the raw clay material used for brick manufacturing.

MATERIALS AND METHODS

A typical clay material used in the brick industry fired industrial brick and dry granite and marble sawing

powder wastes, not beneficiated in any way, collected directly from the ornamental stone cutting industry from Salem district, Tamilnadu, India, were selected and characterized. The characterization included FTIR analysis (Avatar-330 FTIR, Thermo Nicolet) and Mossbauer 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 2.5 x 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 24h. Briquettes 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 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 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.

RESULTS AND DISCUSSIONS

Determination of firing temperature and firing condition of industrial brick by FTIR and Mossbauer techniques

The present investigation is aimed to determine the original firing temperature, firing condition, type of clay, minerals presence and colour mechanism of industrial brick collected from Salem district. It is also



interested to study any structural changes takes place by mixing granite and marble wastes into raw clay material sintered at different temperatures. Figures 1 and 2 show the room temperature FTIR absorption spectra of clay material in the as received state and at different firing temperatures (100-900°C). Figure-3 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-4 shows the room temperature FTIR absorption spectra of 20 wt. % waste mixed brick sintered at 500°C, 600°C, 700°C, 800°C and 900°C.

The presence of four distinct characteristic bands at 3700, 3669, 3655 and 3620 cm^{-1} are attributed to ordered kaolinite, whereas the disordered kaolinite is characterized by 3700 and 3620 cm^{-1} [6, 7, 8]. On firing the clay in between 450 and 500°C, these bands are disappeared means that kaolinite disappears. At around 600°C, the complete destruction of clay structure (silicate structure) takes place [9]. This can be studied by the disappearance of the bands at 1100, 935 and 915 cm^{-1} are attributed to $[\text{Al-O}(\text{OH})]_6$ and OH deformation moles respectively. Also, the appearance of a broad very strong symmetry band centered at 1030 cm^{-1} for red clay or at 1080 cm^{-1} for white clay is the indication of firing temperature of clay-based materials at around 600°C [10, 11]. For the determination of maximum firing temperature of any clay-based materials, especially the presence / absence or variations in the intensities of the bands at 540 and 580 are focused. These bands are attributed to iron oxides [10]. Generally, the presence of iron components in clay materials is characterized by the presence of characteristic band at 530 cm^{-1} . On firing the clay in air, at around 500°C, Fe replaces Al^{3+} in the octahedral sheet structure and the band shifted to 540 cm^{-1} . Above 500°C, the band position 540 cm^{-1} would be gradually shifted to 580 cm^{-1} which indicates the formation of magnetically ordered and well crystallized hematite, possibly above 750°C.

FTIR spectrum of clay sample in the as received state shows the presence of characteristic very weak bands at 3700 and 3620 cm^{-1} . This indicates that this clay is belonging to disordered kaolinite type. On firing the Salem clay in laboratory in between 400 and 500°C, these bands are getting disappeared. It can be observed that the as received state FTIR spectrum of Salem clay shows the presence of 1100 and 915 cm^{-1} as very weak shoulders centered at around 1030 cm^{-1} . At 600°C, these bands are disappeared and a broad very strong symmetry band is observed centered at 1030 cm^{-1} . This indicates that complete destruction of Salem clay structure and originated from red type clay. FTIR spectra of Salem clay in the as received state, fired at different temperatures, industrial brick in the as received state, refired in laboratory at 900°C and waste mixed brick specimens fired at different temperatures shows the presence of the band 775 coupled with 695 cm^{-1} . This indicates the presence quartz mineral [12, 14]. Generally, the presence of sufficient amount of quartz makes the clay-self tempered [9]. It is interesting to note that FTIR spectrum

recorded for Salem industrial brick in the as received state is well compared with the FTIR spectrum of Salem clay obtained at 800°C. So, it is confirmed that Salem industrial brick must have been fired around 800°C during its manufacturing. The bands observed at 3450 and 1640 cm^{-1} are attributed to OH stretching and H-O-H bending vibrations of adsorbed water molecules. Upon heating, these bands will be diminished and disappeared [15, 16]. But, in the present study, the existence of these bands upto 900°C may be due to absorption of moisture in the atmosphere by spectroscopic grade KBr while recording FTIR spectra.

It is important to note that the FTIR spectra obtained for Salem clay as well as waste mixed brick specimens sintered from 500°C to 900°C are almost identical. This indicates that by mixing granite and marble wastes into raw clay material already in use for the production of bricks have not influenced any major structural changes in octahedral and tetrahedral sheets.

Mossbauer measurement

Figure-5 shows the room temperature Mossbauer spectrum of fired industrial clay brick in the as received state.

Production techniques of any clay-based materials can be assessed by ^{57}Fe Mossbauer spectroscopy [17, 18]. The study based on the fact that iron bearing clay minerals as well as associated minerals including iron oxides undergo characteristic chemical and physical changes during firing. These changes depend on the firing temperature and on the kiln atmosphere. Murad and Wagner [19] reported that when firing the clay-materials in an oxidising atmospheric condition, the quadrupole splitting of octahedrally co-ordinated Fe^{3+} increases abruptly, but reverts to lower values upon the formation of new, better ordered phases at higher temperatures (>750°C). Wagner *et al.* [9] reported that during heating in air a strong increase of the quadrupole splitting of Fe^{3+} species 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 and for hematite formation above 750°C. The iron in clays may be present as structural Fe^{2+} or Fe^{3+} 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 Mossbauer spectra. Well-crystallized iron oxides show the characteristic six-line pattern arising from the magnetic hyperfine interaction.

In the present work, the Mossbauer spectrum at room temperature of the industrial brick in the as received state demonstrate the magnetic six-line pattern i.e., the major phase is $\alpha\text{-Fe}_2\text{O}_3$ (magnetically ordered and well crystallized hematite) with Mossbauer parameters as follows: $\text{IS} = 0.382$ mm/s, $\Delta E_Q = -0.225$ mm/s, $H_{\text{hf}} = 51$ Tesla and area of about 50.5%. The second component is Fe^{3+} species with parameters: $\text{IS} = 0.412$ m/s, $\Delta E_Q = 0.911$



mm/s and area of about 40.78%. The third component is Fe^{2+} species with parameters: $IS = 0.767$ mm/s, $\Delta E_Q = 2.432$ mm/s and area of about 8.73 %. From these results and the informations reported earlier by researchers, it is confirmed that the industrial brick collected from Salem District must have been fired inbetween 800°C and 900°C under oxidising atmospheric condition. The presence of higher amount of magnetically ordered hematite is the reason for red colour of the brick [20].

Mossbauer results are well agreed with the results obtained through FTIR studies.

Mechanical analysis

Measurement of compressive and flexural strengths are essential in order to assure the engineering quality of solidified mass suitable in building construction [21]. The mechanical strength is determined by the stress concentration in structural defects such as pores, voids and microcracks. According to the fundamentals of fracture mechanics, the larger the defect the lower the material's strength [22, 23]. Figures 6,7,8 and 9 show the variation of the compressive and flexural strength as a function of the firing temperature as well as the variations in the average values of the strengths at different wt. %. All reformulated brick bodies increase their compressive and flexural strength with temperature in approximately the same way. It was observed that there is a tendency toward higher average strength value for reformulated brick, which has 20 wt. % of granite and marble waste. It has been reported that 0.65 MPa as a minimum flexural strength for the building materials to be used in structural applications [24]. In the present work, all the reformulated brick bodies at different waste content and the industrial fired brick in the as received state tested for flexural strength satisfy this requirement. This indicates the suitability of incorporation of granite and marble sawing powder wastes in brick production.

Water absorption, porosity and bulk density

Water absorption property of clay fired industrial bricks is another important property, determines the stability and durability of bricks. Less water absorption indicates the presence of more durability in the solidified product. Absorption of more water can increase the leachability of metals due to increase of solubility of metal compounds in the solidified matrix. Figure-10 illustrates water absorption measured for granite and marble waste mixed brick bodies at 0, 10, 20, 30, 40 and 50 wt. % and sintered at temperature between 500 and 900°C . From the results one can understand that water absorption decreases with the increase of heating temperature at each waste wt. %. It has also been observed that when the waste wt. % is increased, the water absorption values decrease may be due to fusion of wastes in the pores. After addition of 50 wt. % wastes, the water absorption becomes 17.12% at 500°C and falling to 15.81% at 900°C , respectively. Even though, it is observed that the average compressive and flexural strength values obtained at 50 waste wt. % is less than that of other lower waste wt. %.

The quantity of absorption of water by heated specimens is closely related to presence of porosity or cavities in the matrix. If cavities or porosity are more in the matrix, specimens will exhibit less density and absorb more water [25]. It has been reported that clay brick normally have a bulk density of $1.8-2.0$ g/cm³ [26].

In the present study, the bulk density of the waste mixed fired specimens is determined by dividing the volume over mass of fired briquettes. The relation between the porosity and bulk density of the fired specimens and firing temperature are shown in Figures 11 and 12. From the results it is observed that the porosity is inversally proportional to temperature of firing, while bulk density is proportional. When firing temperature increases, porosity values at each waste wt. % decreases, whereas bulk density values increase and the values obtained within the range between 1.914 and 2.043 g/cm³.

The decreasing trend of porosity and the increase in the value of bulk density at different waste wt. % as the function of firing temperature indicates the fusion of granite and marble powder wastes in the pores of clay brick.

CONCLUSIONS

FTIR and Mossbauer studies were carried out for fired industrial clay brick collected from Salem, Tamil Nadu, India. The results show that the brick has been fired $\sim 800^\circ\text{C}$ under oxidising atmospheric condition during its manufacturing. The results obtained through technological tests show that granite and marble waste content up to 50 wt. % can be incorporated into clay materials, already in use for brick production, without degrading their mechanical properties. In this work, mechanical strengths observed for conventional brick in the as received state and waste mixed brick at different sintering temperatures are nearly same, resulting in energy saving and waste reduction. Further, it is found that the average strength value of waste mixed brick obtained at 20 wt. % is higher than that of other wt. %. Finally, granite and marble waste as an alternative raw material in brick production will induce a relief on waste disposal concerns.

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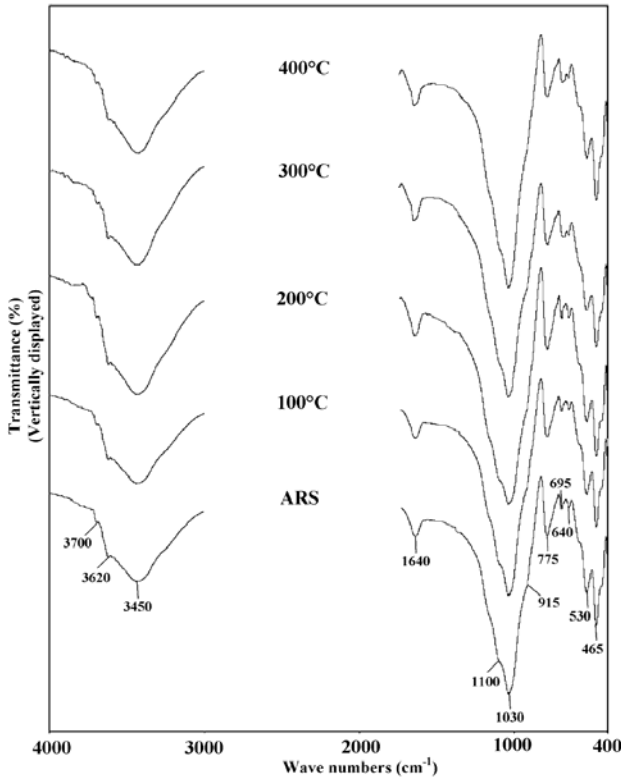


Fig.1. Room Temperature FTIR spectra of Salem Clay in the as received state (ARS) and at 100, 200, 300, 400°C

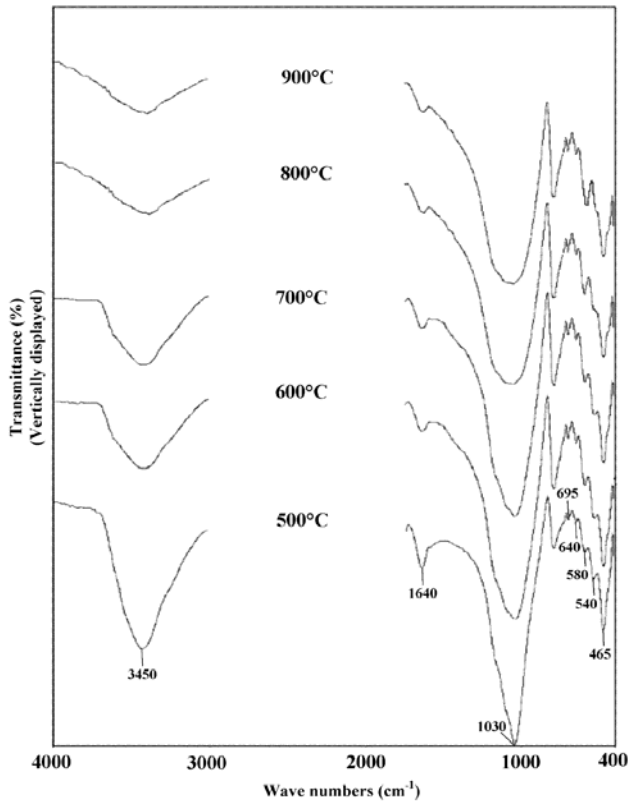


Fig.2. Room Temperature FTIR spectra of Salem Clay at 500, 600, 700, 800 and 900°C

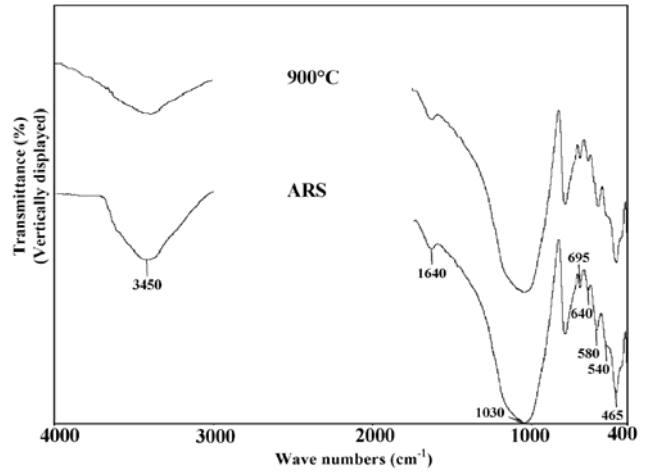


Fig.3. Room Temperature FTIR spectra of fired clay brick in the as received state and refired at 900°C in the laboratory

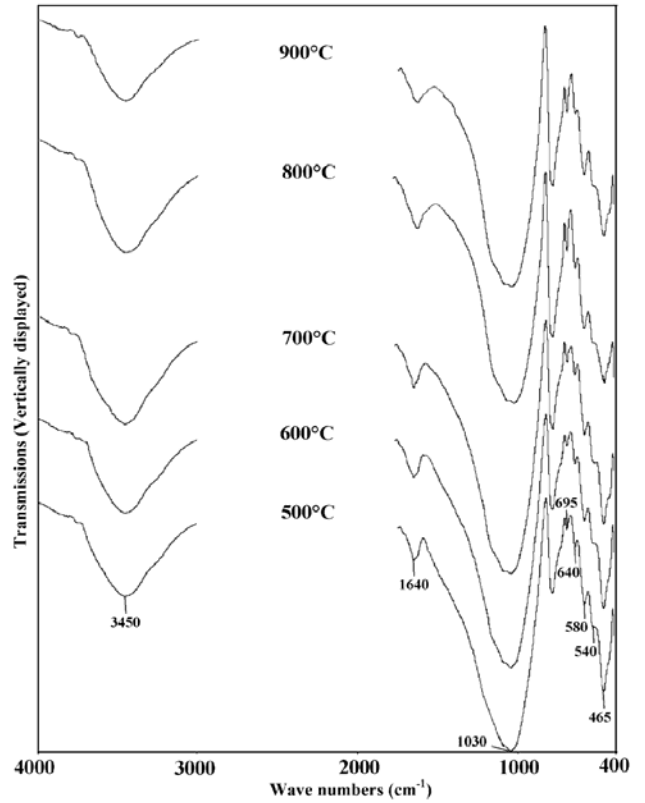


Fig.4. Room Temperature FTIR spectra of granite and marble waste mixed brick sintered at different temperatures

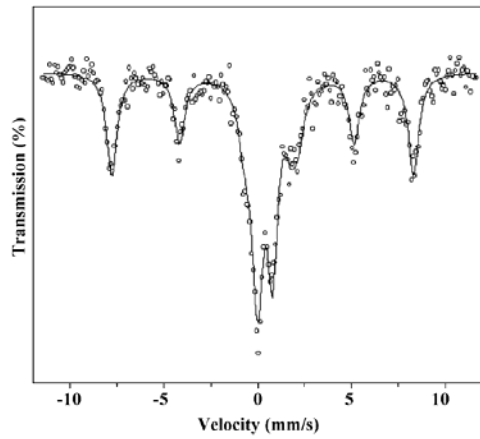


Fig.5. Room temperature Mossbauer spectrum of fired industrial clay brick in the as received state

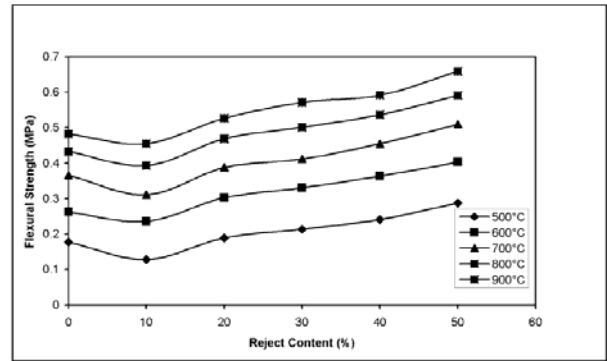


Fig.8. Variation of the Flexural Strength values as a function of reject content and the sintering temperature

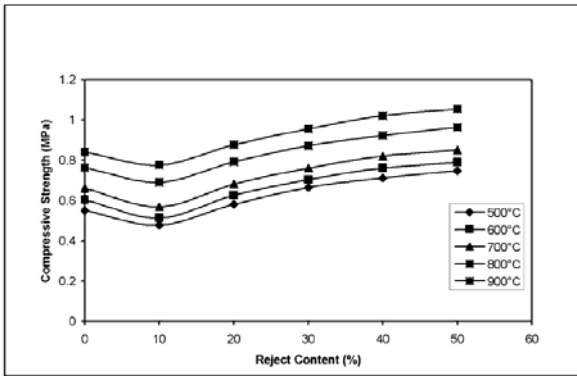


Fig.6. Variation of the Compressive Strength values as a function of reject content and the sintering temperature

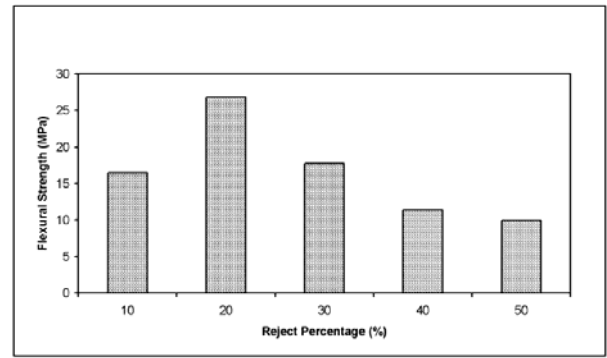


Fig.9. Variation in the average values of the Flexural Strength at different wt. %

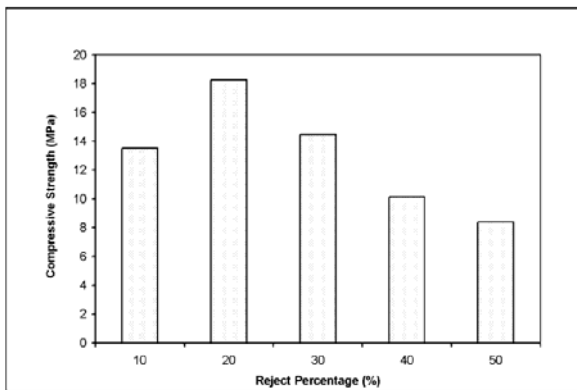


Fig.7. Variation in the average values of the Compressive Strength at different wt. %

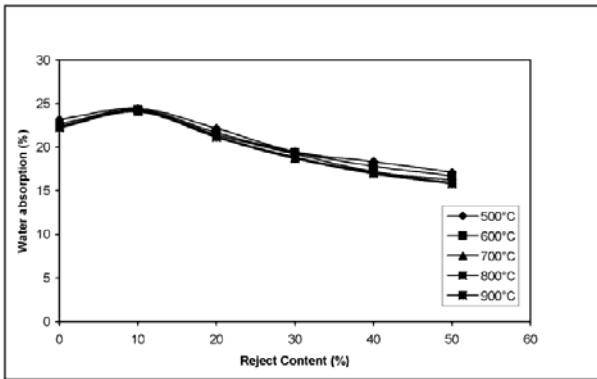


Fig.10. Changes in Water Absorption as a function of the reject content and the sintering temperatures

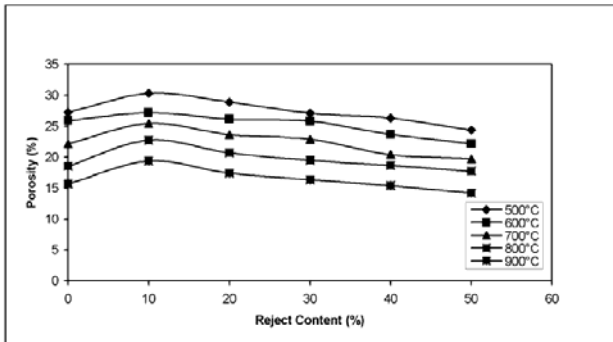


Fig.11. Changes in Porosity as a function of the reject content and the sintering temperatures

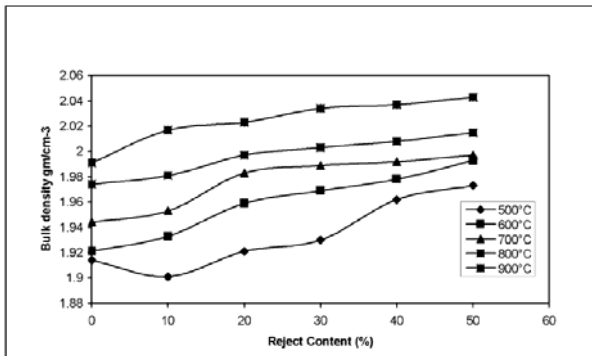


Fig.12. Changes in Bulk Density as a function of the reject content and the sintering temperatures