

EFFECT OF WASTE GLASS POWDER ON PHYSICO-MECHANICAL PROPERTIES OF CERAMIC TILES

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Abstract

Ceramic tile has excellent physical, mechanical and chemical properties. It was prepared by mixing Bijoypur clay, quartz and different amount of waste glass powder and K-feldspar at different proportions. Samples were fired at 1075 – 1175^oC with soaking time of 1 hr. and the physico-mechanical properties were investigated. Results reveal that the waste glass powder is an efficient fluxing agent when used as an additive in ceramic mixture and can be used as a low cost substitute for feldspar.

Key words: Ceramic tile, water absorption, bulk density, firing shrinkage, modulus of rupture

Introduction

Demands of ceramic tiles are increasing day by day and the researchers are becoming interested in developing lucrative tiles with high mechanical strength for household uses as well as for decorating purposes. Usually ceramic tiles are manufactured by using clay, quartz and feldspar but presently waste materials such as waste glass powder are used to make tiles. This waste glass powder is able to replace the traditional fluxing agents like feldspar without changing the process and quality of the final products. Ceramic products are manufactured using high amounts of fluxing agents like sodium and potassium feldspars, nefeline, talc and ceramics frits (Gennaro *et al.* 2003). Ceramic tiles have various characteristics and it can be used in many different places because of high mechanical resistance and surface hardness (Tucci *et al.* 2004 and Malleucci *et al.* 2002).

The glass is made from sodium oxide, calcium oxide and silicon dioxide which is indicated as a soda-lime-silica glass. In fact soda-lime-glass is already a vitreous silicate. The vitreous silicates are generated during the maturation of clay bodies which act as fluxing agent. The reducing clay body maturation temperature was strong evidence that addition of soda-lime-glass to clay body raw materials could increase the efficiency of clay body firing and therefore be a value-added application for recycled glass fines (CWC-Clean Washington Center 1999).

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This product is a ceramic material with a very compact structure being, impermeable, glazed or not, made up of crystalline phases surrounded in a glassy matrix. It is composed of low amounts of clays and kaoline, high percentage of feldspars as fluxes and some quartz sands (Tucci *et al.* 2004 and Abadir *et al.* 2002).

The objective of the present work is to study the substitution of the feldspar (partial and fully) by waste glass powder and its effect on physico-mechanical properties in ceramic mixtures, for manufacturing of ceramic floors and walls tiles. Waste glass powder when incorporated into a mixture, has a good potential as a new fluxing agent in replacement of traditional feldspar and makes possible to obtain a vitreous microstructure during sintering of ceramic tiles.

Materials and Methods

Chemical analysis of waste glass powder collected from waste broken bottles and other raw materials were carried out using X-ray Fluorescence Spectrometer (XRF) (PANalytical XRF, Model PW- 2404 X-Ray Spectrometer) and results are shown in Table 1. The batch compositions of tile body (Table 2) were formulated using waste glass powder with other materials such as clay, quartz and fluxing materials K-feldspar.

Table 1. Chemical composition of raw materials (in wt%).

Compound	Bijoypur clay	Quartz	K-feldspar	Cullet
SiO ₂	51.11	99.401	63.130	67.902
Al ₂ O ₃	30.84	0.203	15.589	2.300
MgO	-	0.114	0.229	1.943
CaO	0.34	-	0.472	10.744
Fe ₂ O ₃	2.50	-	0.204	0.710
MnO	-	-	-	0.046
TiO ₂	1.38	-	-	0.136
ZrO ₂	-	-	-	0.022
P ₂ O ₅	0.10	-	-	0.0683
LOI	13.20	0.202	0.310	--
Na ₂ O	-	-	1.953	14.226
K ₂ O	0.54	0.079	16.664	0.950
I	-	-	1.449	-

A mixture of 2 kilogram was prepared for each batch compositions. The mixture of raw materials was wet milled for 18 hrs in porcelain pot using agate balls to obtain the desired fineness. The slurry obtained was screened, dried at 110⁰C, powdered to break the agglomerate and granulated for better compaction using 6 - 7% moisture. Test pieces were prepared by applying 8 tons pressure using press machine. The test pieces were fired at 1075 - 1175⁰C in air. The heating rate and soaking time were kept at 5⁰C/min and 1hr., respectively in all cases.

The fired tile bodies were subjected to various tests. The water absorption, bulk density and apparent porosity were determined by Archimedes's immersion technique on keeping the specimens in boiling water for 2 hrs. The firing shrinkage was examined by general method. The modulus of rupture (MOR) was studied by Universal Testing Machine (Shimadzu UTM Model Autograph AGS-10 KNG). The phases formed of waste

Table 2. Compositions of various batches (in wt. %) of tile body.

Sample name	Bijoypur Clay	Quartz	K-Feldspar	Cullet
Sample A	40	30	30	-
Sample F	40	30	20	10
Sample G	40	30	-	30

glass powder and tile body composition were identified by using PANalytical, X-ray Diffractometer (XRD) (X-pert PRO XRD PW3040). The microstructure (morphology) of tile body was examined by Scanning Electron Microscope (SEM Hitachi-3400N).

Result and Discussion

In order to better understand the distinctive role of all the raw materials used in processing ceramic tiles, it was first necessary to determine their chemical compositions. Results of chemical analyses of the raw materials are presented in Table 1. It is evident from Table 1 that in case of the clay, loss on ignition, silica and alumina are 13.20, 51.11 and 30.84%, respectively. The percentage of silica present in clay material clearly indicates that it is kaolinitic in nature. On the other hand chemical analysis of quartz shows that its purity is quite reasonable (SiO_2 -99.40%) and K-feldspar is also quartzitic in nature which contains 16.66 and 1.95% of K_2O and Na_2O , respectively. The alkaline oxides act as fluxing agent. From Table 1, it is also seen that waste glass powder contains a relatively large percentage of SiO_2 (67.90), Na_2O (14.22) and CaO (10.74) in addition to MgO (1.94%) and Al_2O_3 (2.30%). The presence of alkaline and alkaline earth oxides in the glass composition will act as fluxing agents helping the sintering process of the ceramic materials with glass powder incorporated (Costa *et al.* 2009). From XRD pattern of waste glass powder (Figure 1), it is observed that the glass powder contains ferrite, aluminum phase which has low intensity and amorphous in nature.

Figure 2 shows the variation of the firing shrinkage as a function of the firing temperature. The firing shrinkage increases with increasing firing temperature (Figure 2). The shrinkage of samples F and G is higher than sample A (containing no waste glass powder). The shrinkage values of partial replacement of K-feldspar with waste glass powder (sample F) is higher than that of total replacement of K-feldspar with waste

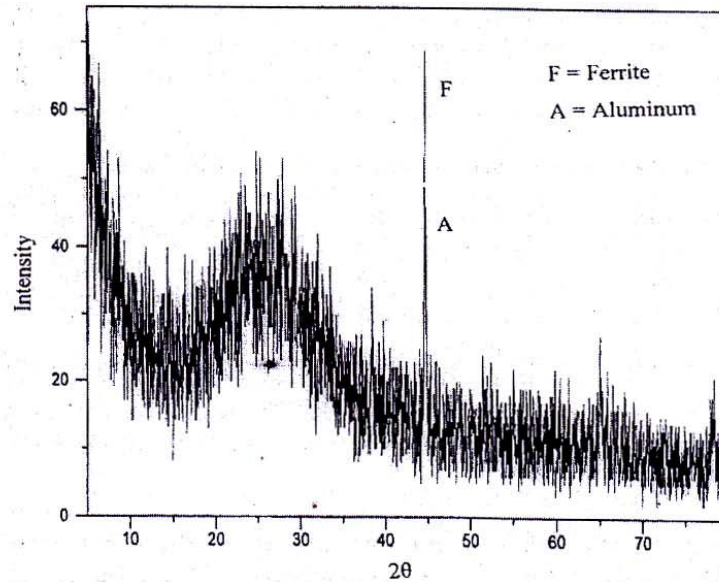


Figure 1. XRD diffractogram of cullet (waste glass powder)

glass powder (sample G) at 1050^o C up to 1150^o C. But at 1025^o C, the shrinkage value of sample F is approximately similar (4.43%, Figure. 2) to the sample G (4.64% from Figure 2). The shrinkage values of sample F increases sharply above 1025^o C and at 1075^o C this value is maximum (13.58%, Figure. 2). After that the value is gradually decreased from 1100^o - 1125^o C and then decreases sharply (Figure 2) in sample F. This phenomenon

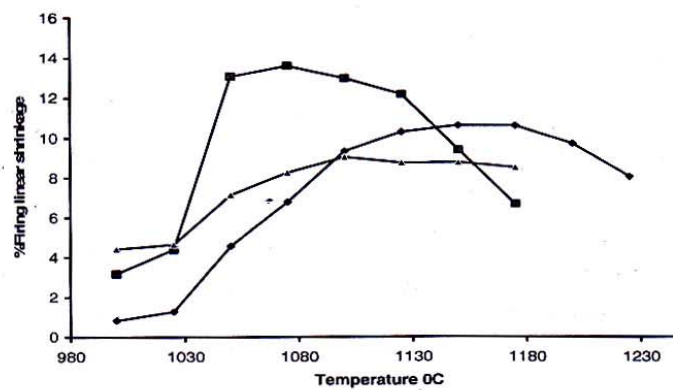


Figure 2. Temperature vs firing shrinkage of ■ sample A, ♦ sample F and ▲ sample G.

indicates an over firing of the tile bodies. It may be suggested that a high content of fluxing oxides, Na_2O , K_2O in the system helps the formation of glassy phases which fill in the pores, thus leading to a shrinkage gain decreasing to firing. The firing shrinkage values of sample G is higher (4.43%, Figure 2) than values of sample F (3.17%, Figure 2) at 1000°C which indicates the sintering mechanism starts at below 1000°C . From Figure 2, it is also observed that the firing shrinkage values of samples G and A gradually increase with increasing firing temperature which reaches maximum values 9.39 and 10.59% from Figure 2 at 1100° and 1175°C , respectively indicating the sintering mechanism is completed in ceramic body. After that the values gradually decrease with temperature in case of samples G and A. Densification is monitored by measuring linear shrinkage and water absorption.

Water absorption is an important parameter in ceramic wall and floor tiles, that defines the class to which the product belongs and according to ASTM standard ceramic or porcelain presents values below 0.5% for floor tiles. Figure 3 shows that both the specimens (samples F and G) present values below 0.5% at 1100°C and the partial replacement of K-feldspar with waste glass powder resulted in a lower firing temperature only for sample F, that reaches a value of 0.15% (Figure 3) at 1100°C .

From Figure 3, it is observed that the water absorption of the tile bodies decreases with increasing firing temperature for all tiles body compositions. It is a natural behavior of ceramic body. With increase in firing temperature, values for water absorption tend to decrease since a greater densification of the sample occurred. The water absorption values of sample F (containing 10% waste glass) is approximately similar (0.22% from

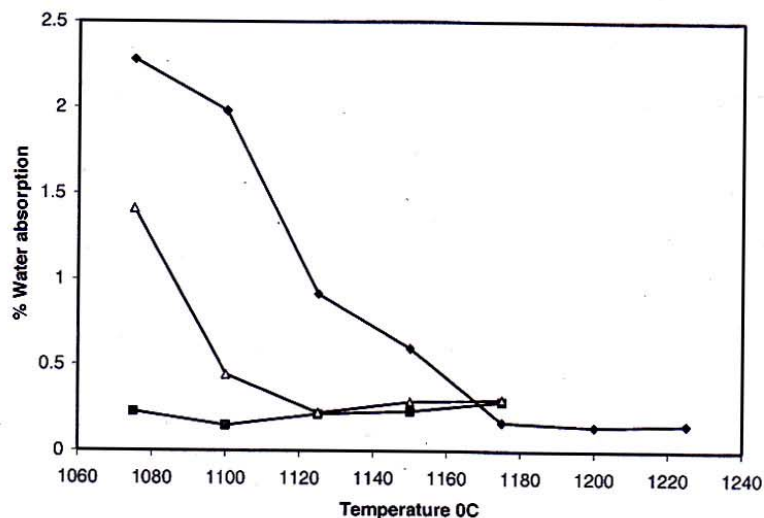


Figure 3. Temperature vs water absorption of ♦ sample A, ■ sample F and ▲ sample G.

Figure 3) to sample G (containing 30% waste glass) at 1125°C. The water absorption value of sample A (no waste glass powder content) is the lowest (0.14%, Figure 3) at 1200°C.

The partial and fully replacement of K-feldspar with waste glass powder in a ceramic tile mix changes the amounts of the different alkalis in the mixture (Tucci *et al.* 2004 and Malleucci *et al.* 2002). Increasing the amounts of waste glass powder in the mixture, the amount of calcium, magnesium and sodium is increased, but the amount of alumina decreases and its presence decreases the refractoriness. The firing behaviors shown by the mixture can be attributed to the changes in composition resulting from the replacement of total and partial K-feldspar with waste glass powder which led to differences in the viscosity of the liquid formed at the firing temperatures used to produce the porcelain stoneware tile (Malleucci *et al.* 2002, Marghussian *et al.* 1999, Cheeseman *et al.* 2003 and Duran *et al.* 2003). The water absorption values of samples F and G are lower than sample A (Figure 3).

Figure 4 shows the behaviors of ceramic tile bodies in terms of bulk density as a function of firing temperature. It can be seen that samples F and G present a decrease in bulk density with increasing firing temperature which may be due to the presence of waste glass powder. But sample A presents a increase in bulk density with increasing firing. It is a natural behavior of ceramic body.

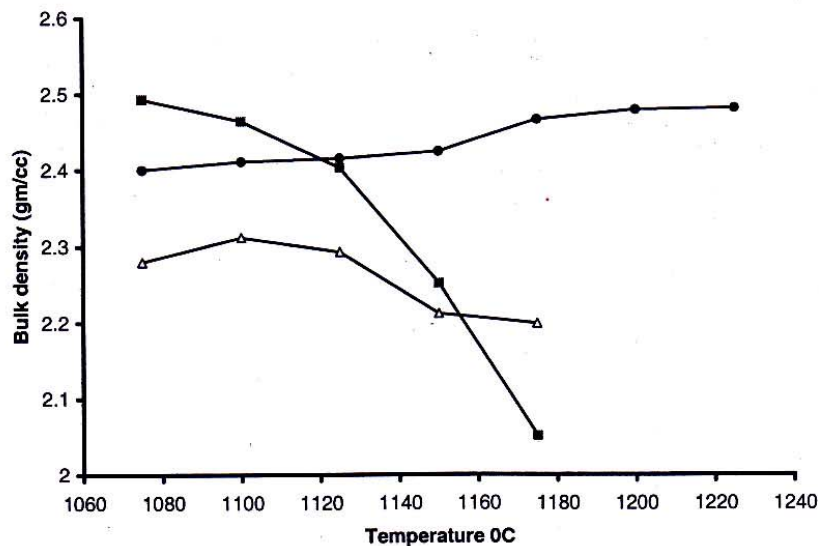


Figure 4. Temperature vs bulk density of ● sample A, ◆ sample F and ▲ sample G.

Modulus of rupture is important factor for ceramic tile. Figure 5 shows the variation of modulus of rupture as a function of firing temperature. Modulus of rupture depends on the materials composition and dimension and morphology of the flaws. The mechanical behavior of the specimens can be explained taking into account at different microstructures developed during firing (Leonelli *et al.* 2001, Esposito *et al.* 2005 and Hernandez-Creso *et al.* 2001). The combination of waste glass powder and K-feldspar sample F presents a higher modulus of rupture than sample G without feldspar content but lower than sample A (Figure 5).

At 1150°C the average modulus of rupture values of samples F and G are between 64.97 and 25.07 Mpa, respectively, the value (64.97 Mpa) is in accordance with ASTM standard (≤ 35 Mpa for floor tiles and ≤ 25 Mpa for wall tiles) that requires values higher than 35 Mpa and 25 Mpa. According to ASTM standard, the samples F and G can be used as floor and wall tiles body, respectively. The presence of K- feldspar and waste glass powder are favoring the developing of a more compact microstructure and lower glassy matrix (Figure 6b) with lower water absorption (Figure 3) at 1150°C. As a result the modulus of rupture in sample F is higher than sample G. The modulus of rupture reaches the maximum value (64.97 Mpa, Figure 5) for sample F characterized by the presence of pores with narrow size (Figure 6b). High glassy form is present in sample G (Figure 6c) as well as modulus of rupture is low (Figure 5).

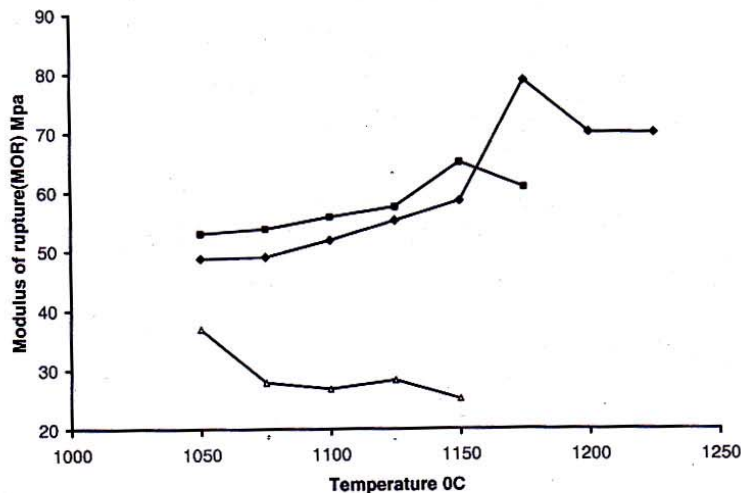


Figure 5. Temperature vs modulus of rupture of ♦ sample A, ■ sample F and ▲ sample G.

The microstructure of the fractured surface of the samples fixed at 1150°C is shown in Figure 6(a-c). It is observed from the micrographs that crystalline materials are embedded in a glassy matrix and microstructures of the tile bodies are not thoroughly homogeneous. Compared to sample G (containing 30% waste glass powder), the fracture

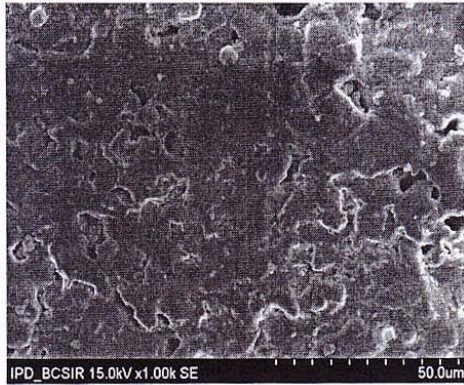


Figure 6a. SEM of sample A at 1150⁰C.

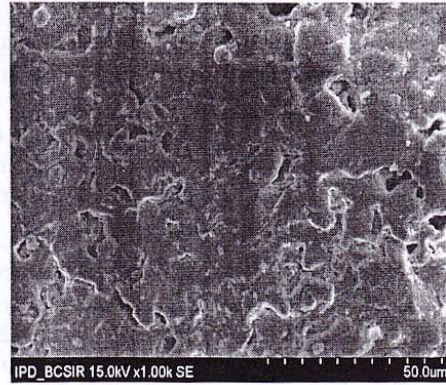


Figure 6b. SEM of sample F at 1150⁰C.

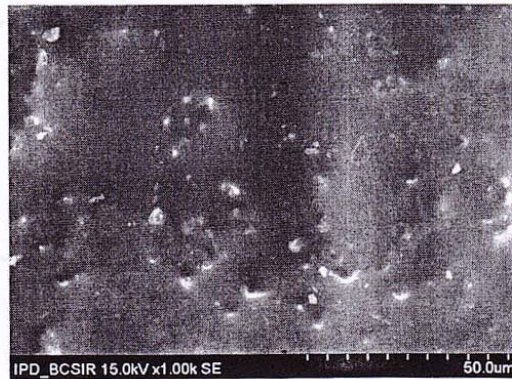


Figure 6c. SEM of sample G at 1150⁰C.

surface of the control sample F shows a more dense well-sintered microstructure with uniform distribution of pores (Figure 6b) which is due to the presence of feldspar and waste glass powder. The fracture surface of the sample G is very different from others (samples A and F, Figure 6a-c) with more dense well-sintered microstructure associated with the formation of elongated cavities. More glassy matrix is formed in sample G (Figure 6c) than sample F (Figure 6b) and sample A (Figure 6a) at 1150⁰C which indicates the formation of glass. From these observations, it is suggested that the dense microstructure is responsible for better mechanical properties of the ceramic tiles (Dondi *et al.* 1999).

The phase formation of ceramic tiles was identified by XRD analyses. From Figure 7(a-c), it is observed that the samples A, F and G contain quartz and mullite phases. Quartz is a residual mineral phase which is observed from the original raw material (sand) and mullite is formed during firing.

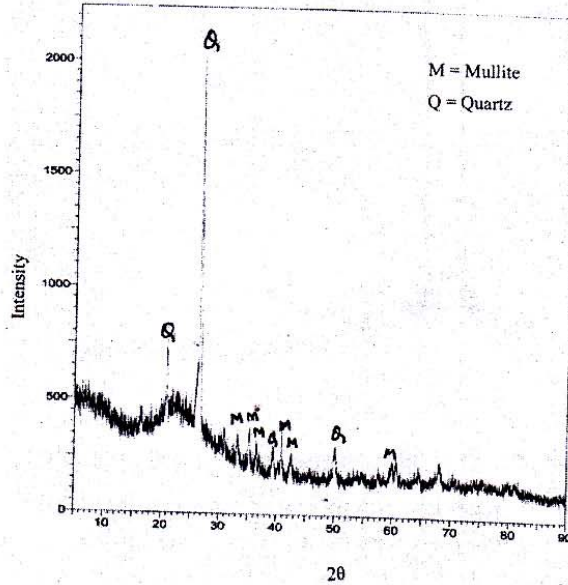


Figure 7a. XRD diffractogram of sample A at 1150°C.

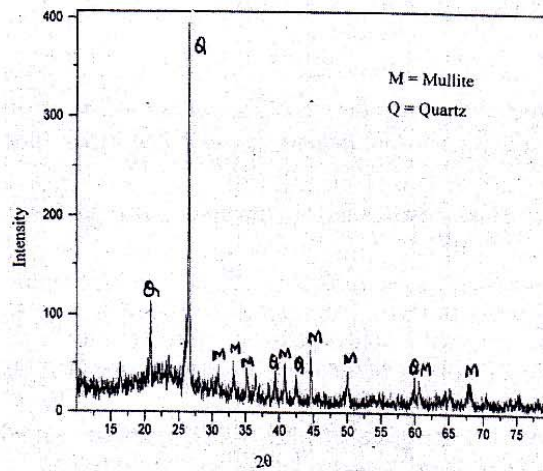


Figure 7b. XRD diffractogram of sample F at 1150°C.

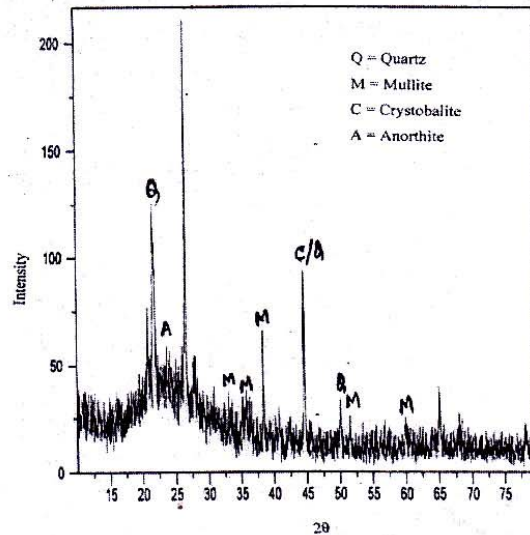


Figure 7c. XRD diffractogram of sample G at 1150°C.

Ceramic tile bodies generally contain a single mullite phase, ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$), evolution pathway: the dehydroxylated kaolin, metakaolin, transform into a non-equilibrium unstable spinel type structure, which converts to mullite above 1075°C (Marghussian *et al.* 1999 and Duran *et al.* 2003). For the compositions containing waste glass powder in intensity and position of the peaks in the X-ray diffraction patterns indicate the presence of anorthite.

Conclusions

Results presented were demonstrates that it is possible to utilize waste glass powder as alternative raw materials for the production of wall and floor tiles. It is, therefore, concluded that:

- (i) The use of waste glass powder in the standard wall and floor tiles composition increases the firing shrinkage.
- (ii) The waste glass powder can be used as an efficient fluxing agent when it is used as an additive in ceramic mixture. During firing waste glass powder accelerates the densification process, with some positive effect (lower open porosity, water absorption) combined with negative ones (high values of shrinkage and high closed porosity).
- (iii) The combination of waste glass powder and K-feldspar (sample F)), lowers the water absorption value than standard tile body (sample A) and fully replacement of feldspar with waste glass powder (sample G).

- (iv) The partial replacement of feldspar with waste glass powder (sample F) showed improved mechanical and physical properties which are important parameters that represent the quality of the product.

References

- Abadir, M. F., E. H. Sallam and I. M. Bakr. 2002. Preparation of porcelain tiles from Egyptian raw materials. *Ceram. Int.* **28**(3): 303-310.
- Cheeseman, C. R., C. J. Sollars and S. McEntee. 2003. Properties, microstructure and leaching of sintered sewage sludge ash. *Res. Conserv. Recycl.* **40**: 13-25.
- Costa, F. B., S. R. Teixeira, A. E. Souza and G. T. A. Santos. 2009. Recycling of glass cullet as aggregate for clays used to produce roof tiles. *Materia (Rio de Janeiro), Materia (Rio J.)* vol. **14** no. 4, Rio de Janeiro.
- CWC-Clean Washington Center. 1999. Glass Clay Body Flux, Final Report, May. [http://www.p2pays.org/ref/04/03905, htm](http://www.p2pays.org/ref/04/03905.htm). Accessed in January 2009.
- Dondi, M., G. Ercolani, C. Melandri, C. Mingazzini and M. Marsigli. 1999. The chemical composition of porcelain stoneware tiles and its influence on microstructural and mechanical properties. *Interceram.* **28**(2): 75-82.
- Duran, P., J. Tarjat and C. Moure. 2003. Sintering behavior and microstructural evolution of agglomerated spherical particles of high-purity barium titanate. *Ceram. Int.* **29**: 419-425.
- Esposito, L., A. Salam, A. Tucci, A. Gualtieri and S. H. Jazayeri. 2005. The use of nepheline-syenite in a body mix for porcelain stoneware tiles. *Ceram. Int.* **31**(2): 233-240.
- Gennaro, R., P. Cappelletti, G. Cerri, M. Gennaro, M. Dondi, G. Guarini, A. Langella and D. Naimo. 2003. Influence of zeolites on sintering and technological properties of porcelain stoneware tiles. *J. Eur. Ceram. Soc.* **23**(13): 2237-2245.
- Hernandez-Creso, M. S. and J. Ma. Rincon. 2001. New porcelainized stoneware materials obtained by recycling of MSW incinerator fly ashes and granite sawing residues. *Ceram. Int.* **27**: 713-720.
- Leonelli, C., F. Bondioli, P. Veronesi, M. Romagnoli, T. Manfredini, G. C. Pellacani and V. Cannillo. 2001. Enhancing the mechanical properties of porcelain stoneware tiles, a microstructural approach. *J. Eur. Ceram. Soc.* **21**(6): 785-793.
- Malleucci, F., M. Dondi and G. Guarini. 2002. Effect of soda-lime glass on sintering and technological properties of porcelain stoneware tiles. *Ceram. Int.* **28**(8): 873-880.
- Marghussian, V. K. and A. Maghsoodipoor. 1999. Fabrication of unglazed floor tiles containing Iranian copper slags, *Ceram. Int.* **25**: 617-622.
- Tucci, A., L. Esposito, E. Rastelli, C. Palmonari and E. Rambaldi. 2004. Use of soda-lime scrap-glass as a fluxing agent in a porcelain stoneware tile mix. *J. Eur. Ceram. Soc.* **24**(1): 83-92.

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