academicJournals

Vol. 8(9), pp. 335-342, 9 March, 2013
DOI: 10.5897/IJPS12.563
ISSN 1992-1950 © 2013 Academic Journals
http://www.academicjournals.org/IJPS

International Journal of Physical Sciences

*Full Length Research Paper*

# Utilization of paper sludge in clay bricks industry to obtain lightweight material: Evidence from partial replacement of feldspar by paper sludge

**Bachir Chemani[1]* and Halima Chemani[2]**

[1]Department of Process Engineering, Faculty of Engineering Sciences, University M'Hamed BOUGARA of Boumerdes – 35000 – Algeria.
[2]Department of Materials Engineering, Faculty of Engineering Sciences, University M'Hamed BOUGARA of Boumerdes – 35000 – Algeria.

Accepted 5 March, 2013

**In recent years, scientific issues related to environmental preservation have acquired great importance and the major challenge to be met is the recycling of materials discarded, causing various damages to the environment by technological development through a disposal of waste. This paper offers a possibility of recovery of waste and byproducts of paper manufacturing (paper sludge) and their use as an addition for obtaining a new formulation of ceramic tiles. The raw materials, paper sludge and clay were mixed together in different proportions. The ceramic samples were characterized with respect to water absorption, the porosity, the linear shrinkage and rupture strength. A new formulation was carried using four mixtures (M1, M2, M3 and M4) where feldspar (FT) was substituted by waste sludge (SP). Substituting FT per paper wastes, the rate of FT is reduced by 50%, thereby improving the rheological properties and physic-mechanical ceramic tiles. The influence of the paper sludge on physical-mechanical properties of final fired product has been studied. Paper waste has proven to be a good raw material and the corresponding formulations were shown to be viable and acceptable in the manufacture of ceramic tiles.**

**Key words:** Recycling, sludge, waste materials, tile, ceramic formulation.

## INTRODUCTION

The use of waste sludge in ceramic industry is a field with high potential in the years to come. The majority of waste sludge comes principally from the manufacturing industries and mineral processing. There are classified and selected sludge as from the crushing process of gneiss, sludge from the cutting and polishing process of varvite, sludge from the process of filtration-clarification of potable water and clay as waste, from waste tannery, from glass cullet, etc. The meaningful utilization of industrial waste materials has been acclaimed world over not only as an economic opportunity (Mir, 1982), but also as a step towards solving problems of environmental pollution. For the interest of using these wastes in the ceramic industry, many attempts were made to incorporate sludges into raw materials mixtures in order to produce different ceramic products.

Waste sludge has been considered as low cost resource material for alumino-silicate and many authors

*Corresponding author. E-mail: chemani_ba@yahoo.fr. Tel/Fax: 213 24 81 91 53.

(Marcis et al., 2005) have tried to use waste mill sludge as a partial substitution of clay in the development of ceramic tiles. The fabrication of products from waste is an advantage that may give to the manufacturer a highly competitive position in the market due to economic issues involved and the opportunity of marketing this principle particularly with regard to the ecological aspect (Modesto et al., 2003).

Sustainable development requires a reduction in consumption of natural raw materials that are not renewable. The use of wastes from the beneficiation of coal fly ash and paper mill sludge has been investigated, usually by the addition of up to 70% mass of these wastes into clayish products (Kumar et al., 2001; Olgun et al., 2005). The wastes may be used to replace conventional flux materials, with the advantage of controlling the plasticity and shrinkage of the ceramic body without producing any negative effect on the product properties, and allowing sintering at low temperatures, thus resulting in energy conservation. The paper presents the results of a feasibility study on the use of paper sludge by integrating them into a ceramic product.

The purpose of this study was to evaluate the use of primary sludge from waste paper in ceramic tile manufacturing. For this purpose, it is necessary to control parameters such as the rate of incorporation of sludge in different mixtures and the substitution effect of feldspar (FT) by waste paper sludge as an addition. The rheological and physico-mechanical properties of each mixture were studied and compared to reference formula industry. Incorporation of sludge waste paper into mixture of ceramic tiles mass production has resulted in a new formulation with significantly improved properties.

Finally, this work aims to develop new ceramic tiles from waste paper sludge allowing the withdrawal of these residues from the environment and give them a nobler destination.

## MATERIALS AND METHODS

The selected wastes are sludge from the Company Stationery and cardboard Saida "PAPCAS" (Western Algerian region), whose physical properties are:

(i) The natural water content of paper sludge is estimated between 26 and 30%;
(ii) Specific Gravity is approximately 2.36 g/cm$^3$.

The mixtures were prepared with the raw materials, according to predetermined amounts. The study is carried out using combinations of various types clay (yellow clay (YC), gray clay (GC), ordinary clay (OC) hereafter called YC, GC and OC respectively), which are added the FT and the sand (SB) from TIMEZRIT (Eastern Algerian region) and BOU SAÂDA (Southern Algerian region) respectively.

The study is carried on the use of the following raw materials: clays YC, GC and OC, FT, sand. The elementary chemical analyses were carried out with X-ray fluorescence spectrometer, PW2540 type, Vrc, Dy-1189. The mineralogical analyses were carried out in a diffractometer type X'Pert Pro Detector X'celerator, Dy-2233-0525.

Particle size analysis is performed by pipette "Robinson". The method used is a wet process. Raw materials (plastic, not plastic) were ground for 6 h in a moist environment with humidity of 41% in pots to jars "Gerhards" type TPR, using bodies grinding in the form of alumina beads. Particle size analysis is performed by pipette "Robinson." The process used is a process by wet. Raw materials (plastic, not plastic) were ground wet with humidity of 41% in mills jars "Gerhards," type TPR with grinding body into alumina beads with a milling time of 6 h. The research protocol is based on 04 types of mixtures (M1, M2, M3 and M4). The FT was substituted by the paper mill sludge of waste. The content of paper sludges varied in 5, 10, and 15 to 20%. The viscosity is determined using a Ford Cup 4 mm in diameter, by measuring the flow time for 100 ml of the suspension. The slips are dried in an oven for 24 h at 110°C, then ground into powder. The powder obtained was wetted with moisture content in 5 to 6%. The pressing is done using a hydraulic press semi automatic type Gabrielli 262367. The drying of tiles is performed, at a temperature of 110°C, in an oven of laboratory Memmert type. The cooking of ceramic tiles is carried out in a muffle oven model: N200A at quick-cooking, type NR: 55269, according to three cooking temperatures: 105, 1100 and 1150°C, with a temperature landing of 45 min. The mechanical strength is determined by a device force - Model: 424 CRAB Type Gabrielli.

## RESULTS AND DISCUSSION

### Chemical analysis

The chemical analysis of the raw materials was determined by X-ray fluorescence analysis, as shown in Table 1. It can be seen from analysis of the clays body YC, GC and OC, high levels of silica, the absence practically of calcium oxide and the low levels of magnesium oxide. It is observed that the predominant elements in the two clays have a content almost identical among which we mention: $SiO_2$, which must come from different mineral origins (FTs, muscovite and tourmaline) or of free silica. A high concentration of this oxide means that is present in the raw material under these two forms.

It can be observed also that $Al_2O_3$ is present in clays under a low content (<14.1%). The three types of clays are very few refractory. This oxide was correlated to the presence of low clay fractions offering low plasticity for the material. In view of high levels of $Fe_20_3$, these clays are of ferruginous type. Iron plays the role of flow element during firing, forming a eutectic melting at low temperature. It communicates with products ranging in color from red to dark brown. The presence of relatively high CaO in two clays indicates that the mineral contains plagioclase. CaO, derived from carbonates, plays the role as melting element and combines with silicates during firing. Alkali oxides $Na_2O$ and $K_2O$ derived primarily from feldspars, illites, micas and smectites. They favor the formation of vitreous phase in low melting point during cooking, playing the role of energy fluxes. Their associations with iron oxide $Fe_2O_3$ during cooking,

**Table 1.** Chemical analysis of materials used.

| Component | $SiO_2$ | $Al_2O_3$ | $Fe_2O_3$ | CaO | MgO | $SO_3$ | $K_2O$ | $Na_2O$ | $TiO_2$ | Loss ignition |
|---|---|---|---|---|---|---|---|---|---|---|
| Clay YC | 47.17 | 11.88 | 5.51 | 14.19 | 1.80 | 0.64 | 1.63 | 0.67 | 1.33 | 15.69 |
| Clay GC | 46.91 | 11.99 | 5.63 | 14.63 | 1.58 | 0.37 | 1.55 | 0.62 | - | 15.90 |
| Clay OC | 53.90 | 14.04 | 5.57 | 8.15 | 1.29 | - | 1.98 | 1.37 | 0.16 | 13.11 |
| Feldspar (FT) | 72.90 | 14.11 | 1.93 | 0.72 | 0.51 | - | 4.98 | 2.16 | 0.73 | 2.00 |
| Sand (SB) | 97.10 | 0.83 | 1.19 | 0.09 | 0.05 | - | 0.09 | 0.11 | 0.11 | 0.50 |

**Table 2.** Mineralogical analysis of the three types of clay.

| Clays (YC + GC) | | Clay OC | |
|---|---|---|---|
| **Mineral** | **Presence** | **Minerals** | **Presence** |
| Montmorillonite | ++ | Montmorillonite | ++ |
| Illite / muscovite | ++ | Illite / muscovite | ++ |
| Chlorite | ++ | Chlorite | ++ |
| Quartz | + | Quartz Quartz | + |
| Calcite / dolomite | ++ | Calcite | + |
| Iron oxide | + | Iron oxide | + |
| Soluble salts | + | Soluble salts | + |

**Table 3.** Mineralogical analysis of feldspar and sand.

| Feldspar (FT) | | | Sand (SB) | |
|---|---|---|---|---|
| **Mineral** | **Presence** | **Contents (%)** | **Minerals** | **Presence** |
| Quartz | +++ | 41.0 | Quartz | +++ |
| Potash feldspar | ++ | 29.5 | | |
| Sodium feldspar | ++ | 18.5 | | |
| Biotite | + | 11 | | |
| Chlorite | + | 11 | | |
| Iron oxide | + | 11 | Iron oxide | + |

Legend: (+) Low percentage ; (+ +) Mean percentage; (+ + +) High percentage.

causes sinterizing reactions which give the products their definitive qualities.

It is noteworthy that the high loss on ignition in two clays (YC and GC) is due specifically to the release of $CO_2$ from the decomposition of carbonates and liberation of $SO_3$ gas present in clays. It is observed that in the OC clay, the $SiO_2$ and $Al_2O_3$ content are more important. Also we notice that the CaO content is lower with no sulfur element which explains a lower loss on ignition. The alkalis content was higher allowing us to deduce that this clay has higher “grésification” (cementation process of grains performed by precipitation and crystallization of the salt dissolved in pore water).

The FT has a $SiO_2$ and alkalis content relatively high compared to other elements. The ignition loss is almost negligible and is correlated with low content CaO and the absence of $SO_3$ gas.

### Mineralogical analysis

The results of mineralogical analyses (Tables 2 and 3) are in accordance with chemical analyses. The results show a predominance of a mineral inter laminate consisting of montmorillonite and illite, the clay fraction consisting. A considerable rate of montmorillonite may be the cause of a high drying shrinkage along with certain sensitivity to drying but could be corrected by adding degreaser. We also detected the presence of muscovite and chlorite iron. The clay fraction is composed of calcite in a high amount and traces of dolomite.

We also note every contents low in soluble salts that are difficultly identifiable without any detrimental effects. Clay OC is without dolomite but has the same minerals as YC and GC. The X-ray diffraction patters of the waste paper sample material under investigation are given in

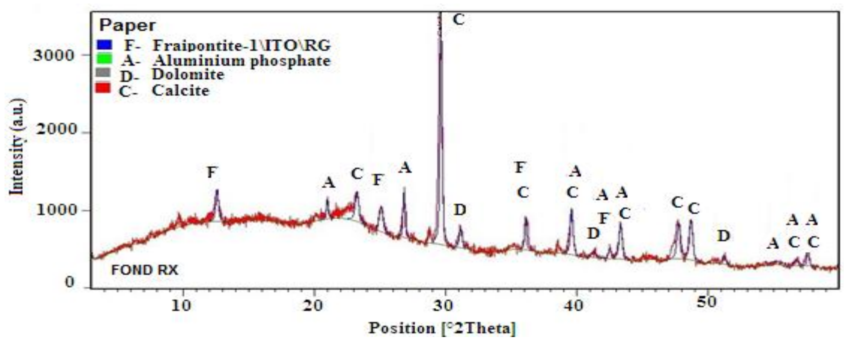


**Figure 1.** X-ray diffraction pattern of the waste paper sample.

Figure 1. In the paper waste the main constituent is Calcite *($CaCo_3$)*, followed by Aluminum Phosphate ($4AlPO_4$), Fraipontite $[(Zn,Al)_3(Si,Al)_2O_5(OH)_4]$ and Dolomite $[CaMg(CO_3)_2]$. These mineralogical data confirm that paper waste is mostly adequate for the mixing with the ceramic body.

## Morphology of raw materials

Figure 2 shows the micrographs of the YC, GC, OC and FT. The micrograph to FT shows a predominance of large aggregates belonging to alumina. The FT is surrounded by spaces inter granular which consist mainly of aggregates belonging to silica which are added to his neighborhood of small particles which includes specific components of FT, namely the potassium and sodium. The empty spaces account for dark areas of the picture. Given the density of large aggregates, the FT requires a grinding that is intense enough during its treatment.

## Technological characteristics

### *Preparation of suspensions*

In order to determine rheological properties, a mixture of 04 preparations M1, M2, M3 and M4 were developed. The grinding is performed in a ball mill for 06 h with a moisture content of 41% suspensions. A residue on the sieve 63 µm determined. The weight of the composition is determined in Table 4. The values of rheological properties are shown in Table 5.

### *Preparation of powders*

A traditional preparation of the powders was applied. This was carried out in two stages:

(i) Drying of slurries in the drying oven,
(ii) Crushing and screening the resulting cake.

The homogenization of the powder obtained was carried out on the choice of a mixture of 04 fractions: fractions (1 - 0.5) = 20%, fractions (0.5 - 0.25) = 45%, fractions (0.25 - 0.15) = 20% and fractions < 0.15 = 15%. A mixture of different particle size classes of particles provides the advantage of reducing the quantity of second phase necessary to the flow and leading to a more compact pile, which reduces the sintering shrinkage (Boch, 2001). After mixing the different fractions, the obtained powder is humidified with 5% humidity and allowed to stand during 24 h before pressing.

### *Confections of tiles*

The pressing is carried out using a semi-automatic hydraulic press, type-Nassetti VIS801. Two pressures were applied on the tiles: One of 9 MPa for desecrating the powder and the second of 22 MPa for compacting. The drying process was carried out into a dryer rolls type NASSITI WHO. The firing is carried out in a roller furnace at temperatures of 1050, 1100 and 1150°C.

### *Characterization of physical-mechanical properties of dry and fired tiles*

The strength values on raw, dried and fired material are shown in Table 6. Considering these values, we notice that the M3 mixture composed of 15% waste paper gives the best resistance to the raw, dried and fired at a temperature of 1100°C (Figure 3). Increasing the temperature to 1150°C, overcooking occurs resulting in a decrease in resistance accompanied by a partial melting of the constituents of the ceramic tile.

Furthermore, considering the mineralogical analysis of waste paper (Figure 1), these contain a rate of dolomite and calcite that are not insignificant. Increasing the concentration of this addition in the mixture led to an appearance of excess liquid phase that in turn led to a reopening of the pores and decreasing in strength.

The values of flexural strength of dry products show a marked increase in the mixture M2. Besides this

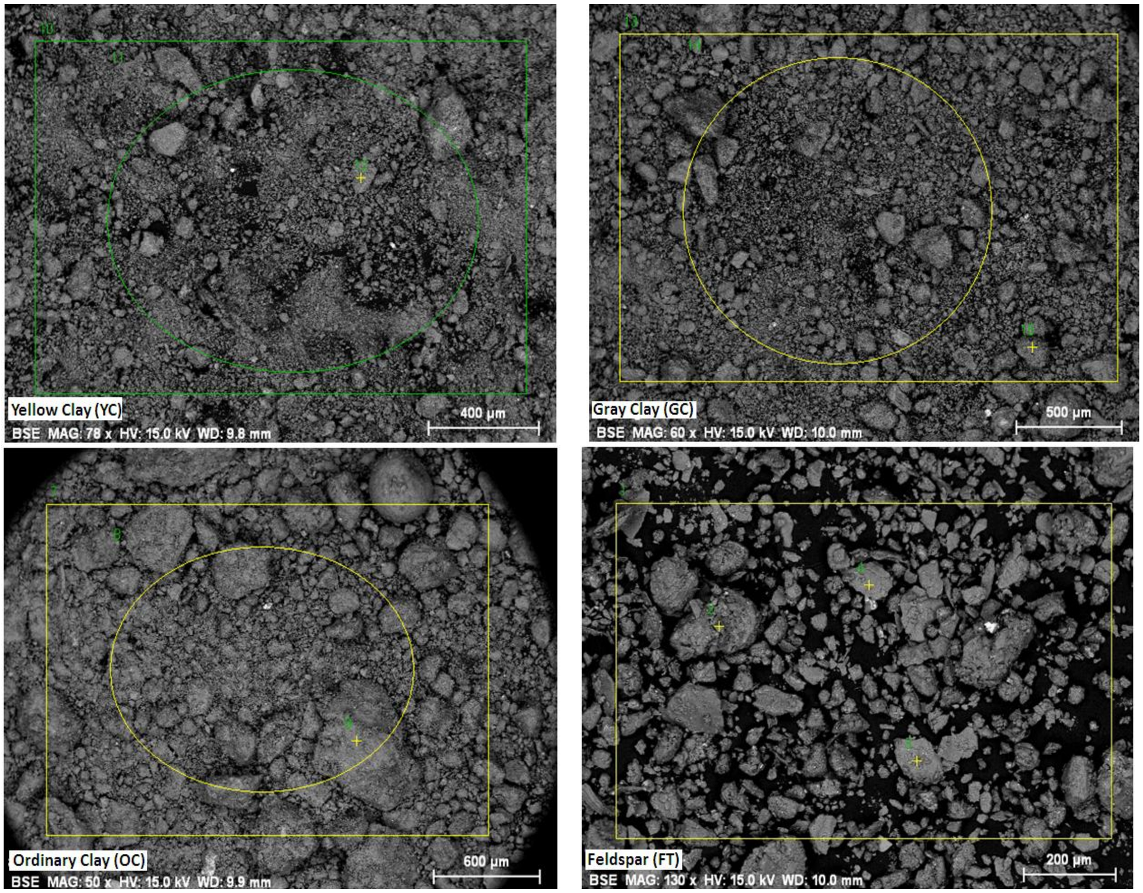


**Figure 2.** SEM micrographs of the surfaces of the yellow, green clays and feldspar.

**Table 4.** Compositions of mass.

| Content (%) | Mixtures | | | | Natural moisture (%) |
|---|---|---|---|---|---|
| | M1 | M2 | M3 | M4 | |
| Clay (YC+GC) | 15 | 15 | 15 | 15 | 13.85 |
| Clay OC | 35 | 35 | 35 | 35 | 9.68 |
| Sand (SB) | 15 | 15 | 15 | 15 | 1.48 |
| Feldspar (FT) | 30 | 25 | 20 | 15 | 5.6 |
| STPP | 0.25 | 0.25 | 0.25 | 0.25 | - |
| Waste paper (SP) | 5 | 10 | 15 | 20 | - |

**Table 5.** Variation of rheological properties depending on the addition of waste paper.

| Mixtures | | M1 | M2 | M3 | M4 |
|---|---|---|---|---|---|
| Refusal (%) | | 8.82 | 6.83 | 5.88 | 3.20 |
| Density ($g/cm^3$) | | 1.65 | 1.594 | 1.594 | 1.584 |
| Viscosity (cp) | | 20.65 | 15.79 | 18.07 | 19.92 |
| Thixotropy | 15 min | 21.78 | 17.20 | 18.29 | 21.02 |
| | 30 min | 22.46 | 18.42 | 18.58 | 22.60 |
| | 45 min | 23.10 | 19.12 | 19.03 | 23.05 |

**Table 6.** Variation of the flexural strength as a function of the addition of waste paper and temperature.

| Mixture | | M1 | M2 | M3 | M4 |
|---|---|---|---|---|---|
| Bending strength on raw (MPa) | | 0.8 | 0.82 | 0.97 | 0.93 |
| Bending strength on dry (MPa) | | 1.93 | 3.23 | 2.84 | 1.66 |
| | 1050°C | 11.75 | 13.62 | 16.18 | 12.52 |
| Bending strength on fired (MPa) | 1100°C | 12.81 | 14.55 | 16.27 | 14.04 |
| | 1150°C | 12.01 | 13.87 | 13.08 | 12.24 |

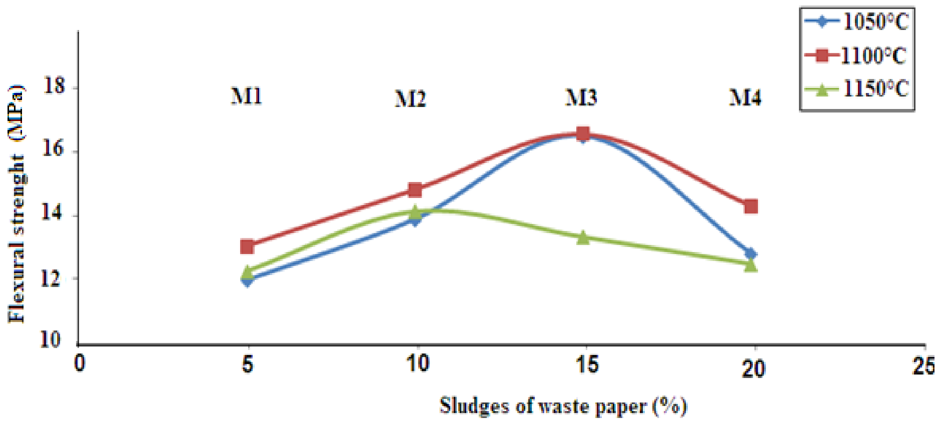


**Figure 3.** Flexural strength of the various mixtures versus in wastes paper sludge content and the firing temperature.

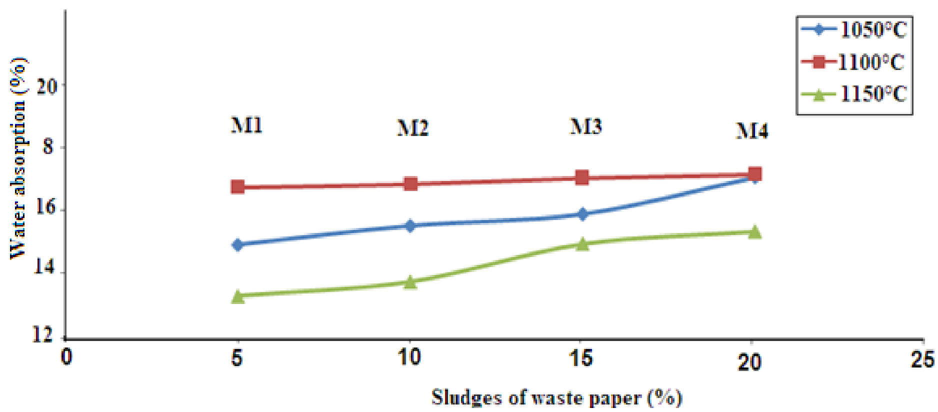


**Figure 4.** Water absorption of the various mixtures versus in wastes paper sludge content and the firing temperature.

concentration the resistance decreases due to the high evaporation of the physically bound water of the water in the clay suspension (mixture) and one contained in the waste paper. For fired products the mechanical properties increase in the range temperature 1050 - 1100°C. Over 1100°C, these properties are strongly dependent on the addition of waste which provides an excess of liquid phase which decreases mechanical properties. The diagrams of "Grésification" of the various compositions were prepared in terms of water absorption and density, porosity open, closed and total. The dependence of these parameters as a function of firing temperature is shown in Figures 3 to 6.

Wang et al. (2000) have shown that if sintering duration

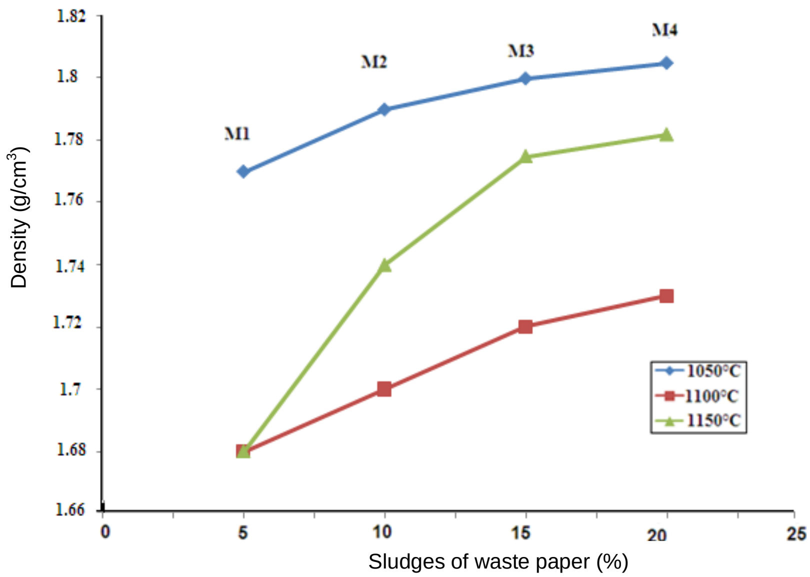


**Figure 5.** Density of the various mixtures versus in wastes paper sludge content and the firing temperature.

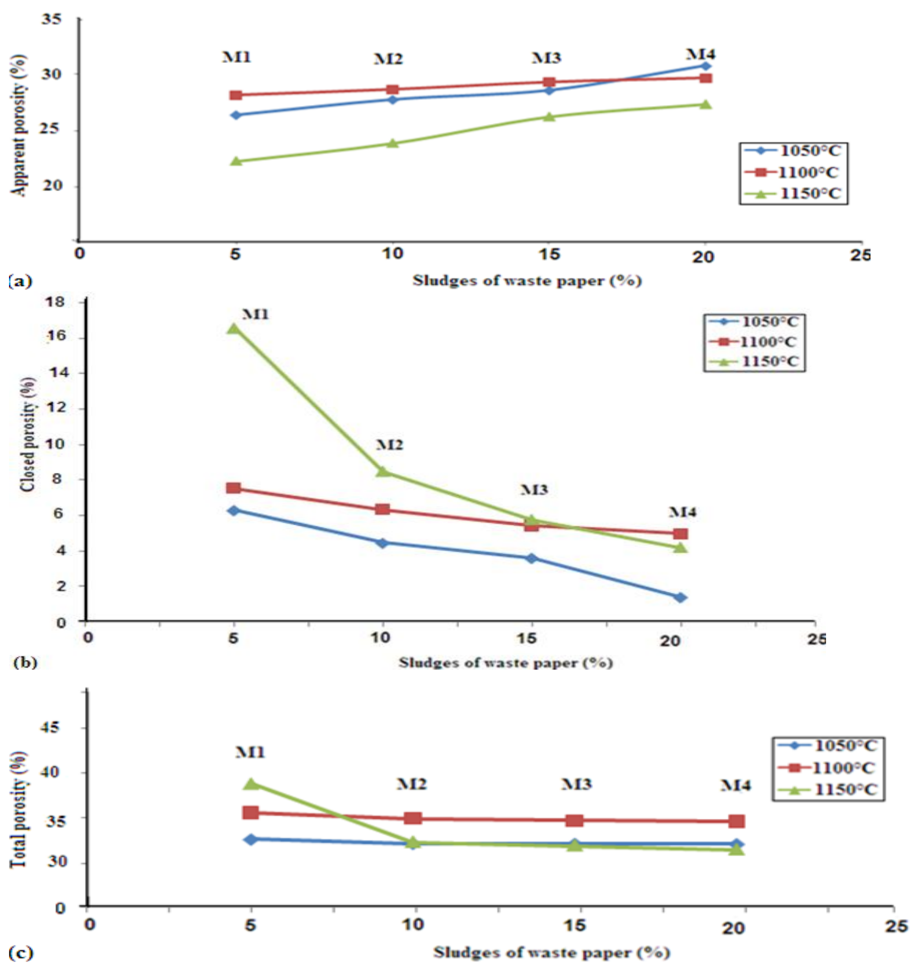


**Figure 6.** (a) Apparent porosity, (b) Closed porosity, (c) Total porosity of the various mixtures versus in wastes paper sludge content and the firing temperature.

increases, the density final product increases. In the case where the sintering stage decreases the opposite case occurs (Won et al., 2002). Developments specific structural characteristics and mechanical properties of clay products are attributed to their chemical and mineralogical composition (El Yakoubi et al., 2006). Waste paper contains a significant concentration of calcite and dolomite involved in the formation of the glassy phase. From the values of absorption and open porosity of mixtures M2 and M3, we can say that in the mixture M2, the tiles are less absorbent and less porous and their mechanical strength is lower. As mentioned above, the mixture contains more waste M3 acting as flows that produce a quantity of liquid phase due to their higher content (15%). This is what gives M3 mixture a greater mechanical strength, and be chosen as optimal formulation for the ceramic tiles manufacture.

## Conclusion

The use of industrial wastes and by-products is broadly recognized as major privileged options for achieving sustainable development. Environment policies in effect require the recovery of waste instead abandoning them to the landfill. In view of the considerable rate of waste thrown in the nature by paper industry, it becomes important to recycle them and use them in other industry sectors. In this study we are interested in recovering sludge from the treatment of paper mill effluents and their use as additions into the mass for ceramic tiles manufacturing. The mineralogical composition of the sludge shows the presence of significant levels of alkaline - earth elements type dolomite and calcite. These chemical compounds have played a role of flux in the masses for ceramic products elaboration. The ceramics industry has always used the FTs as main source of flux elements. Because FT is a non-plastic material that is very tough, its mechanical treatment is often the cause of damage of equipments which requires a good crushing and grinding of the flux. Moreover, the replacement of 50% of FT by waste paper (PS) has allowed the improvement of the properties of ceramic tiles and obtaining a new formulation.

## REFERENCES

Boch P (2001). Frittage et microstructure des céramiques. In : BOCH P. Ed. Matériaux et Processus Céramiques, Paris. Hermès Sci. Pub. pp. 75-112.

El Yakoubi N, Aberkan M, Ouadia M (2006). Potentialité d'utilisation d'argiles marocaines de Jbel Kharrou dans l'industrie céramique. C R Geosci. 338(10):693-702.

Kumar S, Singh KK, Ramachandrarao P (2001). Effects of fly ash additions on the mechanical and other properties of porcelanaised stoneware tiles. J. Mater. Sci. 36:5917-5922.

Marcis C, Minichelli D, Bruckner S, Bachiorrini A, Maschio S (2005). Production of monolithic ceramics from incinerated municipal sewage sludge, paper mill sludge an steelworks slag. Ind. Ceram. 25(2):89-95.

Mir AA (1982). Utilization of industrial wastes - An economic opportunity. Ind. Waste. Proc. 14th Mid-Atlantic Conf. Eds. James E. Alleman and Joseph T. Kavanagh, Am. Arbor Science Publishers, Anu Arbor, Michigan. 2:8-12.

Modesto C, Bristot V, Menegali G, De Brida M, Mazzucco M, Mazona A, Borba G, Virtuoso J, Gastaldon M, Novaes de Oliveira A (2003). Collection and Characterization of Ceramic Materials from Industrial Solid Wastes. Ind. Ceram. 8(4):14-18.

Olgun A, Erdogan Y, Ayhan Y, Zeybek B (2005). Development of ceramic tiles from coal fly ash and tincalore waste. Ceram. Int. 31:153-158.

Wang SW, Chen LD, Hirai T (2000). Densification of $Al_2O_3$ powder using spark plasma sintering. J. Mater. Res. 15(4):982-987.

Won JH, Kim KH, Chae JH, Shim KB (2002). Sintering of attrition-milled TiN powders using a spark plasma sintering technique. J. Ceram. Process. Res. 3(3):166-170.