Effect of the concentration of water acrylic resin in the mechanical, tribological and thermal properties of a composite material based on wastes of the ceramic industry for the elaboration of an eco-brick

Andreina Naranjo-Uribe¹, Gabriel Peña-Rodríguez²*, Héctor Jaime Dulce Moreno³

¹ GIFIMAC, Maestría en Ciencia y Tecnología de Materiales, Facultad de Ciencias Básicas, Universidad Francisco de Paula Santander, Cúcuta- Colombia.
² GIFIMAC, Universidad Francisco de Paula Santander, Cúcuta-Colombia.
³ GIFIMAC, Universidad Francisco de Paula Santander, Cúcuta-Colombia.
* Email: gabrielpr@ufps.edu.co

Abstract. The effect of the concentration of anion–activated resin of acrylic ester copolymer on the mechanical, tribological and thermal behavior of a composite material based on waste powders from the traditional ceramic industry (chamotte) is reported. The samples were formed by uniaxial pressing at 100 kg / cm². The powders of chamotte were sieved through ASTM No. 200 sieve (<75 microns), which were agglutinated using different concentrations of resin. The results of the tribological behavior indicate that the average volume of the footprint (AVF) due to the deep abrasion decreased as the concentration of resin in the samples increased, likewise the modulus of rupture (MR) and the thermal conductivity increased as it increased the concentration of resin. Our results will contribute to the design and development of ecological masonry units.

KEYWORDS: Chamotte, water soluble acrylic resin, mechanical resistance to flexion, wear by deep abrasion, thermal properties.

1. Introduction

The demand for construction materials based on clays (tiles, bricks, blocks), has increased the amount of wasted material (chamotte). This residue is commonly used as a filler, which increases the concern about accumulations of uncontrolled industrial waste [1].

There are reports where bricks and tiles have been used to improve thermal and mechanical behavior in concrete and mortar [2-5]. Nowadays, the chamotte is used in ceramic pastes with high plasticity as well as degreasers in forming processes. On the other hand, when this material is used separately it is necessary to use binders such as, water-soluble acrylic resin, molasses, aloe vera, starches, among others [6-11].

In these studies, it has been shown that the increasing in the proportion of binder enhance the resistance against to the compression, but that does not cause a drastic increase or decrease in tensile and flexural
strength. This is due to the fact that increasing the proportion of binder allows better wetting of the mixture and covers the filling materials causing a strong adhesion. Therefore, the effect of the concentration of water-soluble acrylic resin in the mechanical, morphological and thermal properties of a composite material based on waste from the traditional ceramic industry is reported, with the aim of developing ecological masonry units, which do not need the firing for sintering processes, contributing to the mitigation of the environmental problems generated by the firing in the production of clay bricks.

2. Materials and methods

The residual powders of the traditional ceramic industry, were obtained by grinding materials (bricks, tiles, tablets) made from red clay by the brickmaking industries of the region. For the elaboration of samples of the composite material of chamotte and anion – activated resin of acrylic ester copolymer, with solids percentage of 50%, a viscosity of 40000 CPS and a density of 1.04 g/cm³, according to provider information. The first step was to manually crush the chamotte, subsequently a ball mill is used to obtain homogeneous powders, and to facilitate the sieving process in ASTM No. 200 mesh (<75 μm), which were agglutinated with different concentrations of resin, supplied by INVESA S.A. The concentration of the chamotte in the mixture of the sample was constant (150g), while the resin varied between 40 and 65 g as shown in table 1. The forming process of each sample was made by uniaxial pressing using hydraulic press with a pressure of 100 kg / cm² (98 Bar).

Table 1. Content of resins used in the mixtures to make the samples

| Sample | Resin (g) |
|--------|-----------|
| M1     | 40        |
| M2     | 45        |
| M3     | 50        |
| M4     | 55        |
| M5     | 60        |
| M6     | 65        |

The samples, once pressed, were subjected to a drying process first at room temperature during 24 hours, and then using a forced circulation oven (Memmert UF 110) at 100 °C for the same time. The MR was determined according to ISO 10545-4 standard, while the AVF was calculated by the ISO 10545-6 standard, and Gabrielli® equipment. The figure 1 shows a diagram of that process, in which the footprints are marked on the surface of the sample using a steel disc with average dimensions of 200 mm of diameter (d), thickness of 10 mm, and rotates to 75 rpm. F80 grade alumina abrasive powder is added to produce the footprints, which decreases at rate of 1 gram per turn during 2 minutes. Finally, the length (L) of the footprint is measured and the volume (V) in mm³ of the material detached from the sample is calculated using expressions (1) and (2). [12]
To evaluate the thermal properties at room temperature, the transient linear heat flow method was applied through the KD2 Thermal Properties Analyzer system (Decagon Devices, Inc), which use the ASTM D5334-08 norm. The sensor used was the SH-1 (dual-needle) that consists of two needles with diameter of 1.3 mm and length of 3 cm, which are spaced 6 mm from each other. A short heat pulse is sent from one of the needles through the sample, the other needle records the temperature profile as a function of time, from which it is determined the thermal resistivity (R), the thermal conductivity (K), the caloric capacity per unit volume (ρc), the thermal diffusivity (D), the average temperature and the correlation coefficient (r²). To complete the thermal characterization, the thermal effusivity (ε) was calculated using the average value of (k) y (D) through the equation (3).

\[ \varepsilon = \frac{\kappa}{D} \]  

(3)

The study of the crystalline phases of the samples of chamotte powder used in the elaboration of the composite material, was carried out by X-ray diffraction (XRD), for which a BRUKER diffractometer model D8 ADVANCE with DaVinci Geometry was used, voltage 40 (kV); current 40 (mA); Divergence gap 0.6mm; measurement range in 2θ from 3.5 ° - 70 °; CuKα1 radiation; Nickel filter; sampling time of 0.8 seconds.

3. Results and discussion

The table 2 shows the quantification (wt%) of the mineralogical crystalline phases identified by XRD for the chamotte powders. From these it can be seen that the majority phase (73.1%) is low quartz, followed by Mullita, Hematite, Microcline, Muscovite, and in lower concentration Calcite and the anatase phase of TiO₂. The previous results were obtained by Rietveld refinement of the diffraction pattern presented in Figure 2. Comparing the previous results with that reported by F. P. Gouveia et al. [14], Flavia D. Santos et al. [15], for samples of chamotte obtained from the ceramic industry of Brazil, it is seen that they are practically similar to those reported in this work.

The adhesion mechanism of the copolymer resin to the chamotte particles can be summarized in the following phases:

Anion-activated resin of acrylic ester copolymer is dispersed uniformly in the chamotte particles because it has 50% solids and its viscosity of 40000 cps, initiating the process of hydration of the particles of chamotte, which has a water absorption percentage of 52%. During hydration the occupation of the
porosity present in the chamotte particles by the resin is caused due to the mixing and the application of the uniaxial pressing in the forming sample.

Once the water of hydration is consumed, the polymer hardens forming a continuous film that serves as an adhesion between the chamotte particles, forming a monolithic structure by mechanical adhesion between the adherent and the adhesive responsible for the final properties of the sample.

Table 2. XRD crystalline phases of the chamotte. Source. Laboratories of XRD Industrial of Santander University

| PHASE                      | NAME                      | % (wt) |
|----------------------------|---------------------------|--------|
| SiO$_2$                    | Low quartz                | 73.1%  |
| Fe$_2$O$_3$                | α-Hematite                | 6.1%   |
| K$_{0.86}$Al$_{1.94}$ (Al$_{0.965}$Si$_{2.895}$O$_{10})$ | Moscovite-2M1 | 5.2%   |
| ((O H)$_{1.744}$F$_{0.256}$ |                           |        |
| Al$_{4.68}$Si$_{1.32}$O$_6$ | Mullite                   | 8.9%   |
| K Al Si$_3$O$_8$            | Microcline                | 5.8%   |
| Ca (C O$_3$)               | Calcite                   | 0.4%   |
| TiO$_2$                    | Anatase                   | 0.5%   |

Figure 2. X-ray diffraction patterns of chamotte. Source: Laboratories of XRD Industrial of Santander University

Table 3. The MR and the AVF of the samples.

| Samples | Mechanical Resistance to Flexion. Modulus of Rupture (MR) (MPa) | Average volume of the footprint (AVF) Wear by deep abrasion. (mm$^3$) |
|---------|---------------------------------------------------------------|---------------------------------------------------------------|
| M1      | 2.30 ± 0.26                                                   | 174.39 ± 8.71                                                |
| M2      | 2.69 ± 0.14                                                   | 149.11 ± 7.80                                                |
| M3      | 3.54 ± 0.20                                                   | 123.36 ± 7.16                                                |
| M4      | 2.99 ± 0.18                                                   | 138.29 ± 7.10                                                |
| M5      | 3.58 ± 0.30                                                   | 154.51 ± 5.90                                                |
| M6      | 4.41 ± 0.23                                                   | 147.46 ± 7.83                                                |
The results of the MR and the AVF of the samples are reported in table 3. From these it is observed that as the concentration of resin in the sample increases, the modulus of rupture increases, reporting values ranging from 2.3 MPa to 4.41 MPa, the results that affirm an increase of 91.70% in the mechanical resistance to flexion due to the increase of the resin. Otherwise, it is appreciated that the AVF decreases up to 30% as the concentration of resin in the sample increases, this indicates that the effect of the concentration of the additional resin to act as a binder for the chamotte particles, it also protects the material against abrasion or wear. When comparing the result of the AVF for the sample M1 (AVF = 174.39 mm³), with the reported one (AVF = 193.50 mm³) by Marilse Araque et al. [16], and G. Peña et al. [17] for ceramic tiles based on red clays sintered at 1100 °C, an approximate difference of 10% is observed in this parameter, then from the point of view of the cost-benefit ratio, the composite material offers significant economic and environmental advantages, since it does not require firing for the sintering. The Figure 3 shows the behavior of the MR and the AVF at function of the concentration of resin added to the composite.

![Figure 3. a) Modulus of rupture (MR) and b) average volume of the footprint by deep abrasion (AVF) at function of the concentration of resin of the sample.](image)

The results of the thermal properties at room temperature (~ 23 °C) of the samples are reported in table 4. It is shown that both the $k$, the $\rho c$, and $\varepsilon$ increase by increasing the concentration of resin in the sample. While the speed of temperature per unit length ($D$) in the sample practically does not change.

| Samples | D (m²/s) x 10⁻⁶ | k (W/mK) | $\rho c$ (J/m³ K) x 10⁶ | $E$ W⁰⁵/² m⁻² K⁻¹ |
|---------|-----------------|---------|------------------------|-----------------|
| M1      | 0.29±0.02       | 0.47±0.04 | 1.62±0.01              | 872.77          |
| M2      | 0.35±0.01       | 0.59±0.04 | 1.68±0.07              | 997.28          |
| M3      | 0.35±0.01       | 0.78±0.02 | 2.23±0.01              | 1318.44         |
| M4      | 0.41±0.02       | 0.84±0.02 | 2.05±0.02              | 1311.86         |
| M5      | 0.39±0.01       | 0.88±0.02 | 2.25±0.01              | 1409.13         |
| M6      | 0.41±0.02       | 0.88±0.01 | 2.15±0.01              | 1374.33         |
According to Pape Moussa Toure et al [18], they found values of (k) between 0.69 and 0.81 W/m K, and of (ρc) in the range of 1.65x10^6 and 2.0x10^6 J/m^3 K, for pressed mud bricks, that when compared with those reported in table 4, it is evident that they are practically similar. Likewise, it is confirmed that these materials, from the point of view of their thermophysical properties, are ideal to be used in the manufacture of masonry units that provide thermal comfort to buildings, similarly, when observing the value of thermal effusivity, it is inferred that from the bioclimatic point of view, samples with a higher concentration of resin are ideal for this purpose.

4. Conclusions

This study has demonstrated that the use of anion-activated aqueous dispersion resin of acrylic ester copolymer are effective to be used as binders and reinforcement material in the processing by uniaxial pressing of composite materials based on chamotte powders.

It is evident that the resin as a binder serves as an adhesive between the chamotte particles propitiating a mechanical adhesion due to the porosity present in the powder of the chamotte particles.

Both the flexural strength (modulus of rupture), as the resistance to deep wear and thermophysical properties, increases due to the increasing resin concentration, this is due to the cohesion and adhesion of the particles of chamotte by to the wettability of the resin in the porosity of the particles, increasing the density, forming an optimal material to be used in the design and manufacture of ecological masonry units, which contributes to mitigate the environmental problems of air pollution due to the firing processes in clay bricks, as well as in the contribution to the use of solid waste recycling produced by these industries.

Acknowledgements

Special thanks to FINU-UFPS 035-2017 and 028-2017 for the financial support.

5. References

[1] R. Robayo-Salazar, R. Mejia de Gutierrez, and A. Mulford-Carvaljal, “Producción de elementos constructivos a partir de residuos de ladrillo activados alcalinamente,” Rev. Fac. Ing., vol. 25, no. 43, pp. 21–30, 2016.
[2] R. Harbi, R. Derabla, and Z. Nafa, “Improvement of the properties of a mortar with 5% of kaolin fillers in sand combined with metakaolin, brick waste and glass powder in cement”, Constr. Build. Mater., vol. 152, pp. 632–641, 2017
[3] R. A. Robayo-Salazar, J. M. Mejía-Arcila, and R. Mejía de Gutiérrez, “Eco-efficient alkali-activated cement based on red clay brick wastes suitable for the manufacturing of building materials,” J. Clean. Prod., vol. 166, pp. 242–252, 2017
[4] N. M. Ibrahim, S. Salehuddin, R. C. Amat, N. L. Rahim, and T. N. T. Izhar, “Performance of Lightweight Foamed Concrete with Waste Clay Brick as Coarse Aggregate,” APCBEE Procedia, vol. 5, pp. 497–501, 2013.
[5] Cheng, “Reuse Research Progress on Waste Clay Brick,” Procedia Environ. Sci., vol. 31, pp. 218–226, 2016.
[6] H. A. Bulut and R. Şahin, “A study on mechanical properties of polymer concrete containing electronic plastic waste,” Compos. Struct., vol. 178, pp. 50–62, 2017.
[7] Y. Zare, M. Fasihi, and K. Y. Rhee, “Efficiency of stress transfer between polymer matrix and nanoplatelets in clay/polymer nanocomposites,” Appl. Clay Sci., vol. 143, no. March, pp. 265–272, 2017.

[8] R. S. Chen, S. Ahmad, and S. Gan, “Characterization of recycled thermoplastics-based nanocomposites: Polymer-clay compatibility, blending procedure, processing condition, and clay content effects,” Compos. Part B Eng., vol. 131, pp. 91–99, 2017.

[9] C. A. Anagnostopoulos, “Cement-clay grouts modified with acrylic resin or methyl methacrylate ester: Physical and mechanical properties,” Constr. Build. Mater., vol. 21, no. 2, pp. 252–257, 2007.

[10] W. Lin, C. A. Wang, B. Long, and Y. Huang, “Preparation of acrylic anodic electrophoretic resin/clay nanocomposite films by water-based electrodeposition,” Compos. Sci. Technol., vol. 68, no. 3–4, pp. 880–887, 2008

[11] M. Ayeldeen and M. Kitazume, “Using fiber and liquid polymer to improve the behaviour of cement-stabilized soft clay,” Geotext. Geomembranes, vol. 45, no. 6, pp. 592–602, 2017.

[12] E. La, Caracterización físico-químicas Resistencia a la abrasión profunda, Instituto de promoción cerámica, pp 1-3, Castellón, España, 2013.

[13] V. Useche Julio, G. Peña Rodríguez, and J. Dulcé Moreno, “Efecto de la presión de compactación en las propiedades termofísicas de polvos de arcilla roja elaboradas por atomización,” Respuestas, vol. 2, no. 2, pp. 25–33, 2010

[14] F. P. Gouveia, R. M. Sposto. “Grog incorporation in ceramic mass for the manufacture of bricks. A study of the physical-mechanical properties” Cerámica Vol. 55 pp. 415-419, 2009

[15] Flavia D. Santos, Leyvison Rafael V. da Conceição, Annie Ceron, Heizir F. de Castro, “Chamotte clay as potential low cost adsorbent to be used in the palm kernel biodiesel purification”. Applied Clay Science Vol. 149. Pp. 41-50, 2017

[16] M. Araque-Pabón, G. Peña-Rodríguez, and F. Vargas-Galvis, “Desempeño mecánico y tribológico de baldosas cerámicas de arcilla roja recubiertas por proyección térmica a partir de alúmina,” Tecno- Lógicas, vol. 18, no. 35, pp. 125–135, 2015.

[17] G. Peña-Rodríguez, H. J. Dulce-Moreno and F. Vargas-Galvis. “TiO2 coatings on red clay substrates by thermal spraying”. Revista Latinoamericana de Metalurgia y Materiales. vol. 38, no. 1, pp. 5–9, 2018

[18] Pape Moussa Toure, Vincent Sambou, Mactar Faye, Ababcar Thiam. “Mechanical and thermal characterization of stabilized earth bricks”. Energy Procedia Vol. 139, Pp. 676–681, 2017.