



Coffee husk as substitute material for clay in the manufacturing of ceramic materials in construction in the Metropolitan Area of Cucuta

Cisco de café como posible material sustituto de arcilla en la fabricación de materiales cerámicos de construcción en el Área Metropolitana de Cúcuta

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How to cite: J. Sánchez-Molina, D. Álvarez-Rozo, J.F. Gelves-Díaz and F.A. Corpas-Iglesias, "Coffee husk as substitute material for clay in the manufacturing of ceramic materials in construction in the Metropolitan Area of Cucuta", *Respuestas*, vol. 23, no. 1, pp. 27 - 31, 2018.

Received on July 28, 2017; Approved on November 21, 2017.

ABSTRACT

Keywords:

Ceramic materials
Characterization
Circular economy
Coffee husk

The ceramic industry is a representative sector for the economy of Norte de Santander (Colombia). Despite the recognition of the quality of the products manufactured nationally and internationally, the development of the sector is still in the consolidation phase if it is compared with the major references in the world, including the Spanish and Italian ceramic industry. In the search for alternatives that allow entering this global dynamic of being recognized, the work team has carried out some activities aimed at applying the concept of the circular economy (reuse of waste to production systems) to the regional ceramic process. In this particular case, the reuse of coffee husk (waste from regional agro-industry) as a substitute for clay material in the manufacture of ceramic construction materials has been proposed. The experimental work was developed at the laboratory level, using extrusion as a forming technique. The raw materials were characterized by fluorescence and X-ray diffraction (XRF / XRD), as well as by thermal analysis (TG / DSC). The technological properties of the ceramic evaluated were the linear shrinkage of drying / firing, percentage of water absorption, mechanical resistance to bending and resistance to deep abrasion. To the material with the best ceramic physical behavior, the thermal conductivity was evaluated by means of a transient heat flow method. The results obtained show the potential of the coffee husk to reduce the thermal conductivity of the material and the caloric contribution to the system. The use of coffee husk allows to obtain ceramics for use as coatings and residential type pavement.

RESUMEN

Palabras Clave:

Caracterización
Cisco de café
Economía circular
Materiales cerámicos.

La industria cerámica representa un sector representativo para la economía del Norte de Santander. A pesar del reconocimiento de la calidad de los productos fabricados a nivel nacional e internacional, el desarrollo del sector aún se encuentra en fase de consolidación si se compara con los grandes referentes del mundo, entre estos, la industria cerámica Española e Italiana. En la búsqueda de alternativas que permitan entrar en esta dinámica global, el equipo de investigación realizó algunas actividades encaminadas a aplicar el concepto de la economía circular (reutilización de residuos a los sistemas de producción) al proceso cerámico regional. En este caso particular se ha propuesto la reutilización del cisco de café (residuo de agroindustria regional) como sustituto del material arcilloso en la fabricación de materiales cerámicos de construcción. El trabajo experimental se desarrolló a nivel de laboratorio, usando la extrusión como técnica de conformado. Las materias primas fueron caracterizadas mediante fluorescencia y difracción de rayos X (FRX/DRX), así como por análisis térmico (TG/DSC); las propiedades tecnológicas del cerámico evaluadas fueron: la contracción lineal de secado/-cocción, porcentaje de absorción de agua, resistencia mecánica a la flexión y resistencia a la abrasión profunda. Al material de mejor comportamiento físico cerámico le fue evaluada la conductividad térmica mediante un método transitorio de flujo de calor. Los resultados obtenidos evidencian el potencial del cisco para reducir la conductividad térmica del material y el aporte calórico al sistema. La utilización del cisco de café permite obtener cerámicos para uso como revestimientos y pavimento de tipo residencial.

Introduction

The ceramic industry of Cucuta's metropolitan area is composed of approximately 45 legally constituted companies [1]. The industry has flourished in the area thanks to the abundance of clay materials in the surrounding area, which possesses the optimal physico-chemical properties for the manufacture of ceramic products, such as bricks, blocks, tiles, flagstones, and many others [2]. The circular economy strategy has proved very useful for optimizing processes where the use of other materials is involved, thus reducing the use of raw matter and the production of waste [3].

The advantages of the strategy are reflected in preserves in materials acquisition and improvements in the obtained products thanks to the recycling of waste from the process itself, and a favorable image on a social level, associated with the reduction of negative impacts on the environment [3-4]. The traditional ceramic industry is characterized by the use of raw materials of predominantly natural origin (for example minerals such as clay, quartz and limestone), that allow a wide variety of ceramic products to be created, such as road surfaces, coatings and roofing required by construction companies [5].

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Peer review is the responsibility of the Universidad Francisco de Paula Santander.



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One prevalent characteristic of the ceramic industry is the high energy consumption required to complete all its processes, especially in the drying and firing stages [6].

The circular economy has gained support among academics and entrepreneurs in the ceramic sector, with various changes being proposed, such as the recycling of chamotte (the breakage of the firing process in a ceramic kiln); use of inorganic waste such as glass, fly ash, granite waste and foundry sands; or of agro-industrial residues such as sugarcane bagasse, coconut husk or rice husk [7]-[10].

Indeed, there are still many implementation possibilities that may arise depending on the types of businesses and waste generated in the vicinity of the production centers of ceramic construction materials.

Previous studies investigating the use of waste from coffee and clay processing in the manufacture of ceramic products for construction demonstrate that the combustion of this waste creates pores that maximize the properties of thermal insulation and compressive strength.

This paper hopes to further examine the circular economy concept and the benefits that this may bring to the regional ceramic sector of Cucuta's metropolitan area. In this specific case, the study will consider the effects of the coffee husk from the regional agroindustry acting as a substitute material in the ceramic paste, beginning with acknowledging the material's important contents of potassium and organic matter [8].

Materials and Methods

The used methodological aspects are presented below:

Materials Clay material (CM) of the Alejandra sector, municipality of El Zulia Norte de Santander (Colombia) was used as reference raw material. These are clays with a red tone of sedimentary nature, which are geologically part of the "Guaya-bo Group" as described in the literature [11]. The coffee husk (CH) was obtained from the company Almacafe S.A located in the municipality of Cucuta Norte de Santander.

The materials were ground in a laboratory hammer mill (Servitech), through 12 mesh for CM and 80 mesh for CH. The coffee husk was used as a substitute in concentrations of 5%, 10%, 15% and 20%.

Methods The standards used to evaluate technological properties conform to NTC 4321 (using Gabbrielli equipment), ASTM C326-03 and ASTM D5334-00 were also used for thermal conductivity in the latter case, using the system KD2-Pro thermal properties analyzer de Decagon Devices, Inc. The pastes were manually moistened and taken to a New Wave 101 series laboratory extruder for forming the specimens (dimensions 39.6 mm * 10mm * 120mm). The specimens were dried at room temperature for 24 hours and then in a laboratory drying room for a time equal to 110°C. The firing was carried out in a Gabbrielli Technology 50019 electric oven.

The heating in the oven was carried out at a rate of 5°C/min. Firing was carried out up to 900°C, 1000°C, 1100°C, 1150°C and 1200°C for each of the formulated ceramic pastes; once the oven reached the final temperature described above, it remained constant for a time of 120 minutes

For X-ray diffraction (XRD), a powder diffractometer, brand BRUKER model D8 ADVANCE (40 kV and 40 mA, step of 0.02035° (2θ) and sampling time of 0.6 s) was used. Rietveld refinement was carried out using corundum as the standard for the quantitative analysis. The chemical analysis (FRX) was performed on a BRUKER X-ray fluorescence equipment model S8 TIGER.

Thermal analysis was performed on a SDT-600 equipment, using a heating rate of 20°C/min and an air atmosphere with a flow of 100ml/min.

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Results and discussion

Characterization of raw materials: The results of the characterization of raw materials are shown in Tables I, II, and III, and in Figure 1.

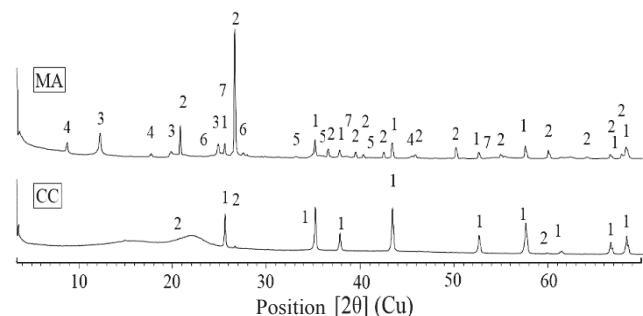


Figure 1. Patrones de difracción de rayos X (DRX) de las materias primas: 1) corindón; 2) cuarzo; 3)caolinita; 4)moscovita; 5) hematita; 6)microclina; 7) anatasa

Table I. Structural composition of raw materials (XRD)

Material	Phase order	Phase	Quantity
Clayey material	Crystalline	Cuarzo	47,4 %
		Moscovita	10,7 %
		Anatasa	0,4 %
		Caolinita	14,4 %
		Calcita	N.C
		Hematita	1,1 %
		Montmorillonita	N.C
		Microclina	0,9 %
	Amorphous	----	74,9 %
			25,1 %
		Total	100,0%
Coffee husk	Crystalline	Cuarzo	00,4%
	Amorphous	----	99,6%
			Total

Table II. Chemical composition of raw materials (FRX)

Oxide	MA	CC
	Concentration (%)	Concentration (%)
SiO ₂	65,66	0,62
Al ₂ O ₃	17,71	0,17
Fe ₂ O ₃	5,74	0,07
K ₂ O	1,60	0,24
TiO ₂	0,91	---
MgO	0,71	0,24
P ₂ O ₅	0,62	0,05
CaO	0,34	0,24
Na ₂ O ₅	0,14	---
BaO	0,05	---
ZrO ₂	0,04	---
V ₂ O ₅	0,04	---
CuO	0,03	---
ZnO	0,03	---
MnO	0,02	---
SrO	0,01	---
Cr ₂ O ₃	0,01	---
SO ₃	---	0,11
PPC	6,28	98,24

From the X-ray diffraction patterns in Figure 1 and the established quantification of phases presented in Table I, approximately 75% of the clay material is crystalline, with quartz as the predominant phase. The presented clay phases are kaolinite, muscovite within the group of micas and traces of montmorillonite.

In addition, minor minerals such as anatase, potassium feldspar and hematite are also present in material M; the hematite likely being accountable for the ceramic’s reddish color.

In relation to the coffee husk, the XRD analysis reveals that this material is mainly amorphous, with small traces of quartz in its composition. The presence of the phases such as the kaolinite and muscovite contribute to the clay material’s plasticity when being shaped and at its glassy phase [4]-[15]

The results of chemical composition (Table II) shows a correlation with the microstructural composition data. For CM it is evident that Si and Al feature strongly. These elements are present in the dominant phases reported in Table I.

Table III. Mass loss events in thermogravimetric analysis

Event	T (°C)	% loss of mass
MA-1	77	1,26
MA-2	154	0,23
MA-3	304	0,13
MA-4	508	4,40
CC-1	86	4,27
CC-2	315	26,94
CC-3	347	32,77
CC-4	415	13,66
CC-4	500	21,08

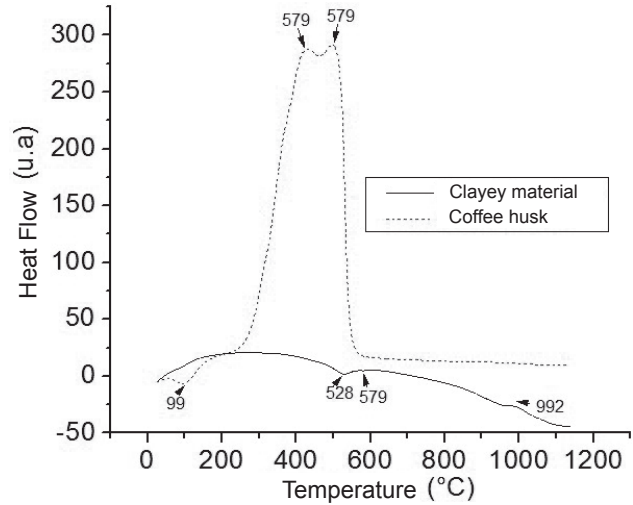


Figure 2. Heat flow profile (HF) of raw materials

From the X-ray diffraction patterns in Figure 1 and the established quantification of phases presented in Table I, approximately 75% of the clay material is crystalline, with quartz as the predominant phase.

The presented clay phases are kaolinite, muscovite within the group of micas and traces of montmorillonite. In addition, minor minerals such as anatase, potassium feldspar and hematite are also present in material M; the hematite likely being accountable for the ceramic’s reddish color. In relation to the coffee husk, the XRD analysis reveals that this material is mainly amorphous, with small traces of quartz in its composition. The presence of the phases such as the kaolinite and muscovite contribute to the clay material’s plasticity when being shaped and at its glassy phase [4]-[15]

The results of chemical composition (Table II) shows a correlation with the microstructural composition data. For CM it is evident that Si and Al feature strongly. These elements are present in the dominant phases reported in Table I.

A feature to take note of is the iron concentration – the value obtained in Table II does not correspond to the concentration of hematite in Table I; meaning that iron is present in more than one phase, like in the muscovite and the amorphous fraction, for instance.

The presence of fluxes such as sodium (Na) and potassium (K) is not particularly prevalent in the ceramic material MA. The featuring of potassium in this material (as shown in Table II) could correspond to the muscovite and montmorillonite phases. In relation to the coffee husk CH, the information in Table II (lost due to calcination) prove that CH is not a great carrier of inorganic materials, even though this sample of inorganic residue (which makes up less than 2% of the total mass) has a high content of potassium, which could in turn have an impact on the ceramic process itself.

The amount of Sulphur in the CH in its natural state is low, however upon evaluation of the only the inorganic component (the ash), it is clear, that if this material is used as a substitute, one should take measures to avoid the formation of soluble salts in the proposed ceramics.

Finally, the thermal analysis results (Figure 1 and Table III), facilitate the discrimination of the source of the calcination losses reported in Table II. In the case of CM, there were 4 events of mass loss, which correspond to the physical absorption of water (at 77°C), montmorillonite dehydration (at 154°C), dihydroxylation of iron hydroxides (at 304°C) and kaolinite (at 508°C) [12]. In the case of CH, the first identified event (at 88°C) is associated with water absorption, the other events correlate to the oxidation of organic matter (presence of hemicellulose, cellulose and lignin reported for this material) [13].

The heat flow profile in Figure 2 shows how the CH material may be characterized by the existence of exothermic events (peaks upwards), whereas in CM endothermic reactions hold sway. The heat input by the CH when being added to the ceramic paste is certainly an interesting aspect.

Physical description of the ceramics The results of the physical characterization are presented below in Figures 2 to 5. Additionally, Table IV shows the thermal conductivity results for the pastes with the best performance.

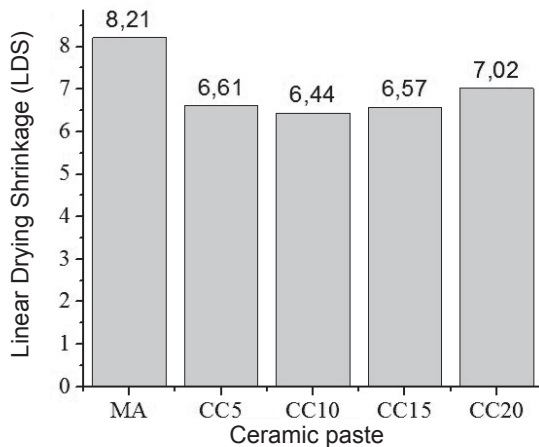


Figure 3. Drying contraction of the ceramic pastes after setting

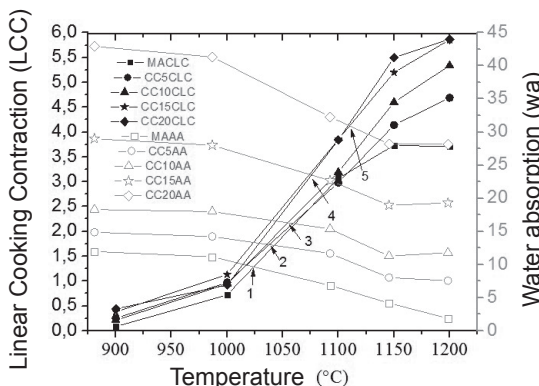


Figure 4. Firing contraction vs percentage of water absorption

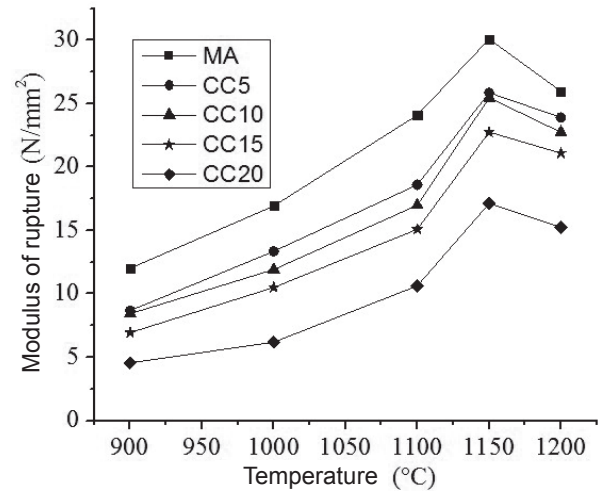


Figure 5. Mechanical resistance to bending of the obtained ceramics

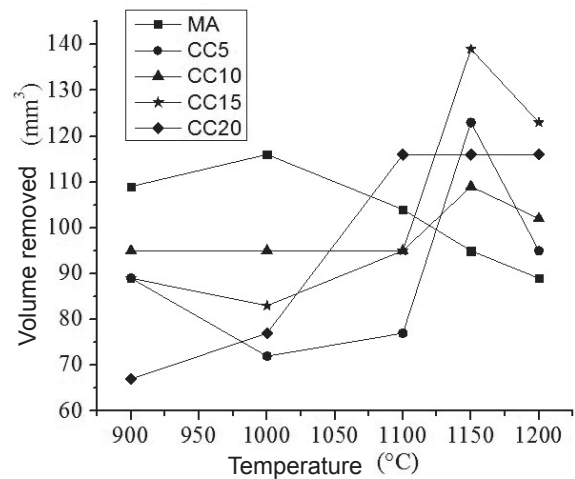


Figure 6. Mechanical resistance to deep abrasion

Table IV. Thermal conductivity of ceramics made with CM and CH5

Temperature (°C)	Thermal conductivity (W/m.K)	
	MA100	CC5
900	0,336	0,257
1000	0,407	0,335
1100	0,691	0,395
1150	0,777	0,403

The first aspect to be highlighted from the physical analysis is the reduction of the drying contraction (Figure 2) due to the presence of the coffee husks; in such circumstances, there are no significant differences between using a concentration of 5% or 15%. The information in Figure 3 enables the identification of the vitrification point in the ceramic pastes. It is evident that there is a directly proportional link between the husk concentration in the paste and the gresification point. What's more, when the firing contraction was raised above 1050°C, important changes can be seen. High husk concentrations produce extremely porous ceramics, due to the high calcination losses previously identified in section 3.1.

In relation to the mechanical resistance tests (Figure 5 and 6), two important features can be identified: the first is associated

with the presence of a limit point between 1100°C and 1150°C, where the recrystallization processes potentially affect the material's resistance [14]; the second, when coffee husk concentration is increased, there is evidence of a resistance decrease. The high porosity, a result of the lack of the organic phase in the coffee husk, as well as a smaller presence of kaolinite and muscovite (by substituting the CM for CH) which act as generating agents of the glassy phase, could explain the loss of mechanical resistance to bending and deep abrasion.

Despite the obtained results, the mechanical resistance data to bending exceeds the breaking load of 400 N, which is the minimum limit for use in coatings, and with a rise of temperature it exceeds 900 N, this being the minimum load for residential and commercial paving [15].

Finally, the results in Table IV show how the presence of coffee husk in the paste reduces the ceramic's thermal conductivity. This is a favorable result since ceramics of this type would make house interiors cooler and thus make any air conditioning systems run more effectively, especially in hot climate areas, like those in Cúcuta's metropolitan area.

Conclusions

The results reveal that the use of coffee husk as a substitute material creates ceramics especially advantageous as a coating or paving. The increased porosity improves the thermal insulation capacity of the ceramic, which highlights the potential of this waste material within the circular economy concept. The TG/DSC analysis shows that the use of husk brings heat to the system, thus favoring fuel savings.

The use of coffee husk ash instead of only husk is of interest to future work, which could help reduce the firing temperature of the ceramic, due to the high content of potassium in the ash.

Acknowledgments

The authors thank the personnel of the Ceramic Materials Research Center of the Francisco de Paula Santander University (CIMAC) for their support provided in the development of this research.

References

- [1] I. Díaz, Informe de resultados Análisis comparativo de las propiedades del conjunto de arcillas del proyecto: Fortalecimiento a 10 empresas del Clúster de la cerámica de Norte de Santander a través de la estandarización de sus principales productos a la Norma Técnica Colombiana, 2015.
- [2] J. Sánchez y P. Ramírez, *El cluster de la cerámica del Área Metropolitana de Cúcuta*. Cúcuta: Editorial Universidad Francisco de Paula Santander, 2013.
- [3] P. Ghisellini, C. Cialani and S. Ulgiati, "A review on circular economy: the expected transition to a balanced interplay of environmental and economic systems", *Journal of Cleaner Production*, vol. 114, pp. 11-32, 2016.
- [4] M.S. Andersen, "An introductory note on the environmental economics of the circular economy", *Sustainability Science*, vol. 2, no. 1, pp. 133-140, 2007.
- [5] E. Sánchez, J. García-Ten and M. Regueiro, "Materias para la industria cerámica española. Situación actual y perspectivas", *Boletín de Sociedad Española de Cerámica y Vidrio*, vol. 45, no. 1, pp. 1-12, 2006.
- [6] N. Tikul and P. Srichandr, "Assessing the environmental impact of ceramic tile production in Thailand", *Journal of the Ceramic Society of Japan*, vol. 118, no. 1382, pp. 887-894, 2010.
- [7] M.V. Madurwar, R.V. Ralegaonkar and S.A. Mandavgane, "Application of agro-waste for sustainable construction materials: A review", *Construction and Building Materials*, vol. 38, pp. 872-878, 2013.
- [8] N.V. Boltakova, G.R. Faseeva, R.R. Kabirov, R.M. Nafikov, and Y.A. Zakharov, "Utilization of inorganic industrial wastes in producing construction ceramics. Review of Russian experience for the years 2000-2015", *Waste Management*, vol. 60, pp. 230-246, 2017.
- [9] H.F. Dorado Guerra, "Uso de subproductos de la industria de café y caña de azúcar como fundente y formador de poros en la manufactura de ladrillos de arcilla roja", Trabajo de fin de grado, Universidad del Valle, 2016
- [10] R.K. Chinnam, A.A. Francis, J. Will, E. Bernardo and A.R. BoCHaCHini, "Functional glasses and glass-ceramics derived from iron rich waste and combination of industrial residues", *Journal of Non-Crystalline Solids*, vol. 365, pp. 63-74, 2013.
- [11] A. Pedroza, "Formación León y Grupo Guayabo: Fuentes de provisionadoras de arcillas en el área de Cúcuta, Norte de Santander". Texto conferencia, (Cúcuta), pp. 4-8, 1996.
- [12] E. Manals-Cutiño, M. Penedo-Medina y G. Giralt-Ortega, "Análisis termogravimétrico y térmico diferencial de diferentes biomásas vegetales", *Tecnología Química*, vol. 31, no. 2, pp. 36-43, 2011.
- [13] I. Viczian, "Földvári, Mária: Handbook of the thermogravimetric system of minerals and its use in geological practice: OCHasional Papers of the Geological Institute of Hungary", *Central European Geology*, vol. 56, no. 4, pp. 397-400, 2013.
- [14] D.A. Rozo, J. S. Molina y J.F. Gelves, "Influencia de las materias primas y de la técnica de moldeo en la fabricación de productos cerámicos tipo gres", *Ingeniería y Competitividad*, vol. 19, no. 2, pp. 89-101, 2017.
- [15] Enrique José. "Requisitos técnicos de baldosas cerámicas para usos concretos". Qualicer 94. [En línea]. Disponible en <<http://www.qualicer.org/recopilatorio/ponencias/pdf/9411060s.pdf>>. [Accedido: 29-nov-2016]