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Feasibility of using moulded cellulose packaging when blended into formulations for manufacturing ceramic building materials

Viabilidad en el uso de envases de celulosa moldeada al ser mezclados en formulaciones para fabricar materiales cerámicos de construcción

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ABSTRACT

Keywords:

Clay, Moulded Cellulose Packaging,
Mechanical properties,
Porosity,
Composites.

Millions of tons of solid waste are generated annually worldwide, and in Colombia, according to figures from the Ministry of Environment and Sustainable Development, by 2018, of the more than 12 million tons, only 17% was recycled. In recent years, efforts have been made to find new waste management strategies to minimize this problem. One of these is destined for the construction industry since it allows the inclusion of raw materials that are different from their nature. In this context, the final properties of ceramic formulations used for construction materials when mixed with 5%, 10%, and 15% Molded Cellulose Containers (MCC) and sintered at 900 °C were evaluated using water absorption and compressive strength. The results showed that by increasing the proportion of MCC, the mix requires greater amounts of water, increases porosity, and reduces mechanical properties, causing non-conformity with national masonry standards. However, the results demonstrate in another context technical, economic, and environmental benefits such as lighterweight products, the opportunity to generate new markets by allowing the manufacture of materials with insulating properties and help to increase the degree of rehabilitation in the quarries, reducing the depletion of the clay raw material, reaching a positive environmental impact.

RESUMEN

Palabras clave:
Arcilla, Envases de Celulosa Moldeada,
Propiedades Mecánicas,
Porosidad,
Materiales compuestos.

Anualmente se generan millones de toneladas de residuos sólidos a nivel mundial, y en Colombia, según cifras del Ministerio de Ambiente y Desarrollo Sostenible, para el año 2018, de las más de 12 millones de toneladas, solo se reciclaba el 17%. En los últimos años se están encaminando esfuerzos en buscar nuevas estrategias de gestión de residuos que minimicen esta problemática. Una de estas apunta a la industria de la construcción, ya que permite la inclusión de materias primas ajenas a su naturaleza. En este contexto, se evaluó mediante absorción de agua y resistencia a la compresión, las propiedades finales de formulaciones cerámicas usadas para materiales de construcción cuando se mezclan con Envases de Celulosa Moldeada (ECM) al 5%, 10% y 15% y se sinterizan a 900 °C. Los resultados evidenciaron que al aumentar la proporción de ECM la mezcla requiere mayores cantidades de agua, aumenta la porosidad y reduce las propiedades mecánicas, causando no conformidad con la normatividad nacional para mampostería. Sin embargo, los resultados muestran en otro contexto beneficios técnicos, económicos y ambientales como son, productos más livianos, posibilita la oportunidad de generar nuevos mercados permitiendo fabricar materiales con propiedades aislantes, y ayuda a incrementar el grado de rehabilitación en las canteras, reduciendo el agotamiento de la materia prima arcillosa, llegando a un impacto ambiental positivo.

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Introduction

Millions of tons of waste are generated worldwide yearly, and deposited in dumps. This, together with the indiscriminate use of natural resources, generates an increase in global contamination. Much of this contamination is generated by industries, especially those that fabricate construction materials. Muñoz *et al.* [1] mention that it uses more than 33% of total natural resources, and in particular, the brick industry produces around 1.6 billion bricks annually, leading several governments to limit the use of natural raw materials for construction. Zhao *et al.* [2] commented that China has issued a conservation policy for clay resources and the environment, limiting the amount of soil that can be removed, and trying to control this situation as much as possible.

In Colombia, diverse initiatives have been carried out to raise awareness among industry and consumers about the importance of caring for the environment, since the country, according to information from the Ministry of Environment and Sustainable Development, 2018, of more than 12 million tons of solid waste produced annually, only 17% was recycled, leaving a great amount of this waste at the expense of nature and increasing contamination [3]-[7].

Norte de Santander is no exception to this important problem, as part of its economy is based on brick-making and family microenterprises, including poultry production. Despite not having large infrastructures, it can have around 2,500 laying hens, which generate a few more than 2,000 eggs per day, generating a considerable amount of cardboard containers in which the eggs are packed for distribution and commercialization. This situation increases the amount of agro-industrial waste in the city, since most of these cardboard containers end up on the streets, in landfills, and in municipal dumps, generating a bad visual aspect and causing important environmental damage.

To reduce the environmental impact and the consumption of natural resources, the literature reports studies aimed at finding different uses by introducing these wastes as substitutes in traditional ceramic paste formulations, where it is reported that they modify and correct characteristics, obtaining products with inferior technological properties, similar and in some occasions superior to the reference ceramic products. These studies include, for example, the manufacture of bricks with paper and PET matrix, the mixing of clays with different wastes such as sludge from the beer industry, urban wastewater, sugar cane bagasse, waste from the paper industry, coffee husks, rice husks, biomass incinerator ash, fly ash, and others, all oriented mainly to reuse and reduce environmental contamination rates [8]-[14].

Considering the importance of these alternatives to minimize contamination, and to propose an effective and viable solution in the use of these wastes, in addition to contributing to the reduction of environmental impact, this study was developed, where the optimal replacement ratio of cardboard packaging in ceramic formulations to manufacture masonry was investigated through the evaluation of the technological properties under the parameters of the Colombian technical standards, proposing a viable alternative to minimize this problem without generating higher production costs.

Material and Methods

The egg cartons, called from now on Molded Cellulose Containers (MCC), were obtained from poultry farmers in the region and different parts of the city. 15 kg were obtained to make the mixture without considering variables such as color, physical condition, or dirt, so as not to limit the use of this waste in future technological developments at the industrial level in the region. The clayey raw material (RA) was obtained from a quarry in the region located in the municipality of Villa del Rosario in Norte de Santander, Colombia, this clayey soil is used in ceramic pastes by different industries to fabricate construction products such as bricks and roof tiles.

Before mixing, the raw materials were dried in a convection air oven, Pinzuar brand model PG-2004, to obtain the exact mass proportion of the mixture. MCC at 60±5 °C for 24 h. RA was dried at 110±5 °C for 24 h, then manually crushed to a particle size of less than 4.75 mm (No. 4 sieve), then in a hammer mill, Reinaltech brand model MBO HM 230 to a size of less than 2.00 mm (No. 10 sieve), and finally homogenized until a uniform color was obtained and quartered, these processes are traditional in the region [15], [16].

Although the initial moisture content of the raw materials was not considered, it was around 31% in the MCC. Once the MCC were dry, they were cut into pieces and moistened in water for 24 hours. This was necessary to separate the cellulose fibers from the lignin and eliminate the hydrogen bonds that can form between the cellulose molecules during the drying of the paper, facilitating the mixing with the clayey material. The absorbed water was used to control the moisture content and plasticity of the formulations before the extrusion process.

RA was mixed as the matrix, and MCC as the discontinuous phase using a laminar geometry, in the form of honeycomb panels. The four pastes, L (100:0), U (95:5), N (90:10), and A (85:15), were formulated. All were wetted according to plasticity parameters and manually amassed to eliminate occluded air and prevent poor cohesion. Shaper forming was carried out in an extruder with a vacuum pump, Reinaltech brand model 380 V. The molded products were 1:5 scale (6 cm ×2 cm ×4 cm) double hollow horizontal perforation bricks of regional standard dimensions. Samples were initially dried in oven Pinzuar brand model PG-2004 at 30±5 °C for 24 h, then at 60±5 °C for 12 h, and finally heated to 110±5 °C, at a heating rate of 5 °C/min and were maintained for 24 h. The samples did not evidence cracks or geometrical problems after drying. The firing process was developed in a beehive-type inverted flame industrial furnace at 900 °C (frequent temperatures in regional production processes), in an oxidizing atmosphere, occurring simultaneously, that is, all the formulations were fired at the same time [17]-[19].

Analyses

The raw materials were characterized by X-ray diffraction (XRD) and X-ray fluorescence (XRF). The XRD patterns were obtained in a BRUKER powder diffractometer model D8 ADVANCE with DaVinci Geometry, using the Rietveld Method refinement. XRF analysis was performed using the QUANT-EXPRESS method in a 4KW wavelength dispersive X-ray fluorescence sequential spectrophotometer, BRUKER model S8 TIGER.

The 152 H hydrometer technique was used for particle size measurement since it measures particle size distributions from 5 µm to 4.75 mm and identifies the ceramic aptitude of the materials [20]. Plasticity was evaluated using the Atterberg method, using Casagrande apparatus Pinzuar brand model PS-11 [21], [22]. Technological aspects such as density, shrinkage, mass loss, and dry mechanical strength were evaluated by measuring the mass and dimensions of the specimens using gauge digital Mitotoyo brand model CD-

8" ASX and a balance Ohaus brand model SPX6201 [18]. The structural property was evaluated by water absorption by 24 h immersion. The mechanical property was evaluated for compressive strength, using a systematized double range machine Pinzuar brand model PC-107D, according to the NTC 4017:2018 standard [23]. The possible functionality of the finished product was analyzed to establish the use and application of the product [24]-[26].

Results and Discussion

Mineralogical and chemical composition

The RA geologically belongs to the Guayabo Formation. Table 1 presents that quartz, kaolinite, and muscovite are the mineralogical phases with the highest presence, in addition, there are minority phases of cristobalite, hematite, and anatase. The results are consistent with the regional literature about clayey materials of the Guayabo group, the difference in the concentrations can be attributed to the location where the clay was extracted, which was in Villa del Rosario [27], [28]. However, Álvarez Rozo et al. [29] evaluated clays from the same area and reported concentrations similar to those reported in Table I. The presence of quartz and phyllosilicates such as kaolinite, although not high, generated a slight heat resistance. Hematite and anatase developed an orange hue in the product, as evidenced in the fired products. Muscovite helped the plastic behavior and the shaping of the formulations.

Table I. Mineralogy of raw materials research.

RA		MCC	
MINERAL PHASE	%	MINERAL PHASE	%
Quartz	49,5	Calcite, Mg	4,1
Kaolinite	17,2	Kaolinite	3,3
Hematite	1,0	Cellulose	92,6
Anatase	1,1		
Muscovite	23,5		
Cristobalite	7,6		

Source. X-Ray Laboratory of the Universidad Industrial de Santander - Headquarters UIS Guatiguará

The mineralogical phases of the MCC evidences calcite as an inorganic component and cellulose as an organic component, the most common mineral phase in vegetal polymers such as cardboard, the principal component of these materials. XRD evidence kaolinite as a minority inorganic phase in this waste, being these results consistent with tests performed by other researchers where they report the presence of kaolinite, talc, or quartz, as accompanying materials of low proportion in this type of waste [30], [31].

Table II presents the chemical composition of the RA, indicating SiO_2 and Al_2O_3 as the predominant oxides, followed by Fe_2O_3. SiO_2 is close to the maximum range for the production of construction materials, possibly dangerous during firing due to the increase in volume during the allotropic change of quartz. This percentage is confirmed with 3.04 in the SiO_2/Al_2O_3 ratio, which is higher than the value of 1.18 for pure kaolinite. RA can be classified as a non-calcareous clay by its low CaO content; and together with the low percentages of Na_2O, K_2O, MgO, and Al_2O_3 content, it can be considered appropriate for the production of ceramics, and also for refractory purposes. Fe_2O_3 and TiO_2, confirm the XRD observations of orange color in the specimens.

Table II. Chemical composition of research raw materials.

RA		MCC	
COMPOSITE	%	COMPOSITE	%
SiO2	62,79	CaO	2,75
Al2O3	20,67	SiO2	1,26
Fe2O3	4,00	Al2O3	1,01
K2O	1,43	Na2O	0,25
TiO2	0,92	MgO	0,17
MgO	0,70	Fe2O3	0,14
Na2O	0,20	Cl	0,09
CaO	0,14	SO3	0,19
Otros	0,30	TiO2	0,11
L.O.I	8,82	K2O	0,04

Source. X-Ray Laboratory of the Universidad Industrial de Santander - Headquarters UIS Guatiguará

The chemical composition of the MCC indicates CaO, a porosity generator, which must be controlled. Furthermore, it presents a minimum amount of SiO_2 and Al_2O_3. The presence of SiO_2 generated compatibility with RA at the moment of mixing and firing, since it contributed to the dimensional control of the specimens, fundamentally in the drying operation. Al_2O_3 enhances heat resistance during firing.

Physical characterization and ceramic aptitude

As can be seen in Figure 1, there is variability in the use that formulations can have depending on the proportions of MCC. Two formulations are presented within the zones of influence, and two are situated outside the zones defined by Helmut Winkler for the development of structural ceramics [32], [33].

Formulations L and A are situated outside the zones, while formulations U and N are situated in zones 3 and 4, respectively. Therefore, the U formulation is appropriate for producing masonry, tiles, and roof tiles; and the N formulation is appropriate for producing structural masonry [34]. Accordingly, the MCC in percentages of at least 10% adjusts the formulations to the reality of the ceramic sector in the region. Furthermore, in terms of environmental impact, it represents a considerable reduction in the demand for natural resources, considering that the RA used in the regional production processes will be substituted by MCC [27], [35].

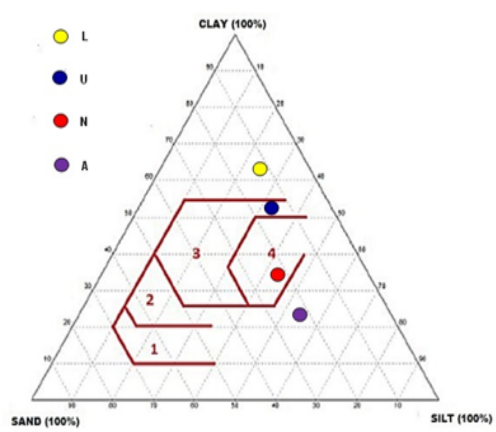

Figure 1. Ceramic suitability of formulations
Source. Authors.

Because the extrusion process was developed in an extruder, the Atterberg limits were evaluated using Casagrande's method [21], [22]. Figure 2 presents that MCC reduces plasticity, generating an increase in the amount of water required to develop adequate workability in the formulations. Formulation L has a difficult extrusion prognosis, it is important to mention that the use of MCC improves workability until 5%, modifying its prognosis towards an acceptable extrusion, allowing for improvement in the manufacturing processes, an important characteristic considering that extrusion is the more commonly used shaping method in the region. The opposite occurred with formulations N and A, which were situated in the difficult extrusion zone, possibly caused by the reduction in plasticity and the high water absorption developed by the cellulose fibers, which reduced the water available for the clay particles, increasing the proportion necessary to obtain the plastic limit required to extrude and confirming that mentioned by Kizinievic et.al. [30] and ratifying Muñoz et.al [1].

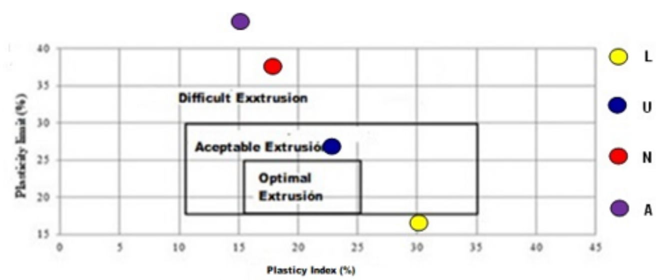

Figure 2. Extrusion prediction of the formulations
Source. Authors.

Figure 3 presents an increase in linear dry shrinkage (LDS) between 0.55% and 2.61% caused by the increase in the amount of water, increasing the interstitial water and reaffirming the results of plasticity. However, the literature reports higher values; for example, Demir et al. present increases of 4.55% and 18.18% by adding 5% and 10% of Kraft pulp production residues, and Muñoz et al. [1] citing Demir [36] reports increase from 16% with the addition of paper pulp residues. Besides, these increases did not exceed 8%, the maximum range to prevent problems such as cracks or fissures during drying, demonstrating favorable behavior during this process.

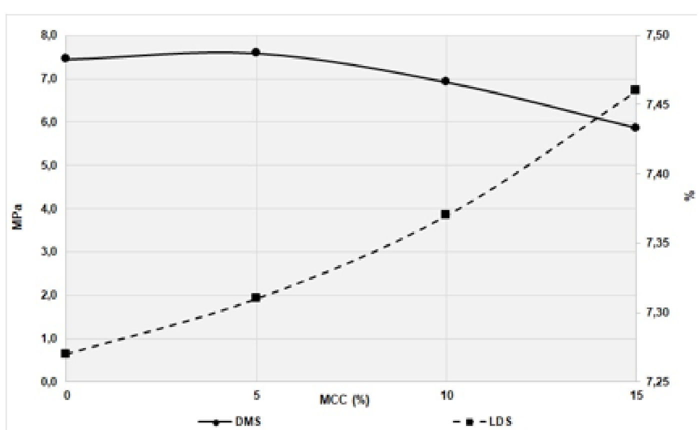

Figure 3. DMS and LDS as a function of additive percentage
Source. Authors.

The dry mechanical strength (DMS) presents a moderate increase of 1.88% in formulation U; and a reduction in formulations N and A, of 7.1% and 21.3%, respectively. This increase is attributed to the strengthening of the cellulose fibers which, upon drying, generate stability in the structure of the ceramic body. Although the decrease of DMS is usual, up to percentages of 10% these residues do not present inconveniences in the behavior, since the results are in the permissible ranges for manufacturing construction products since the literature mentions that the minimum requirements for masonry in Colombia should be between 2.94 MPa (30 kg/cm^2) to 6.87 MPa (70 kg/cm^2); achieving satisfactory values until in proportions of 10%, since the handling of the dry material is high, especially in regional factories [37], [38].

Technological characterization

Thermal processes modify the properties of a material; a ceramic raw material until 450 °C undergoes expansion, and when mixed with non-clay bodies such as MCC, it generates combustion and excess gases and develops in its structure voids and capillaries of different diameters increasing porosity and reducing density and mechanical properties, because the glassy phase developed during firing is not able to fill these pores [39].

Figure 4 presents evidence that the higher the amount of MCC, the higher the porosity and the reduced the density. This increase can be attributed to the decarbonization of $CaCO_3$ present in the MCC, affecting the physical and mechanical properties of the specimens, as will be discussed later in this paper. This is of no benefit for a structural product because it does not conform in terms of the water absorption required by the Colombian standard NTC 4205:2009 [24]-[26]. However, high porosity values can be used for superior specification products with better-added value that are not manufactured in the region, for example, insulating bricks or slabs, if combined with clay with the most refractory characteristics [40].

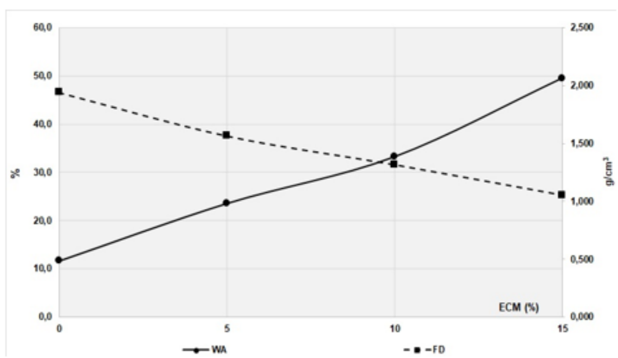

Figure 4. FD and WA as a function of additive percentage
Source. Authors.

However, a difference with similar studies, the results indicate better behavior. Shibib [39] reports a reduction in density of 21% for 5% paper residue at 900 °C, while MCC presents a maximum of 19%. Sutcu and Akkurt [41] reported firing density values of 1.65 g/cm^3 with 10% of paper residue at 1100 °C, and this research obtained a density of 1.31 g/cm^3, this value is not distant if it is considered that this research was developed at less temperature and using extrusion, where the cohesion of the particles is lower in comparison with that obtained in pressing, which is the method used by Sutcu and Akkurt.

Kÿosek-Wawrzyn et al. [42] and Souza et al. [43] report that the linear firing shrinkage (LFS) permitted for these processes should be between 1.0% and 3.5%. Figure 5 presents that LFS increases from 0.28% to 0.72%, obtaining an important technical feature to control the manufacturing dimensions of the products, and explains the control developed during the firing process to avoid deformation and cracking of the specimens. This is a consequence of the combustion of the cellulose fibers of the MCC and the voids that are generated during combustion, voids that the liquid phase does not fill, causing the clay particles to not approach, reducing its shrinkage [8], [39]. In addition, some aspects on the surface of the specimens are detailed, no black craters were observed indicating that the residue was completely burned and the generated gas eliminated; but small white spots and a lighter shade in red were observed, indicating that $CaCO_3$ reacted, confirming the XRD results and what was previously reported.

Mechanical strength

Figure 5 presents the mechanical strength in compression, indicating that the addition of MCC reduces it, this is a habitual technical aspect when this type of residue is used. However, the relative rates of reduction are not considered problematic, since in the same working conditions (10% and 900 °C) Demir et al. [36] report a reduction of 38.7%, Muñoz et al. [1] presents a reduction of up to 36.0% and Shibib [39] reports decreases of up to 35.3%; while with the MCC maximum values of 35.1% were reached; generating important uses with the MCC in comparison with other researches. Analyzing the mechanical strength values, that define the minimum values of conformity and classification for the use in the masonry, a better behavior is also evidenced, presenting values of 11,92 MPa and 9,22 MPa for proportions of 5% and 10%, that are above the 9,84 MPa and 8,15 MPa reported by Abdel Hafez et.al. [44], at the same temperature.

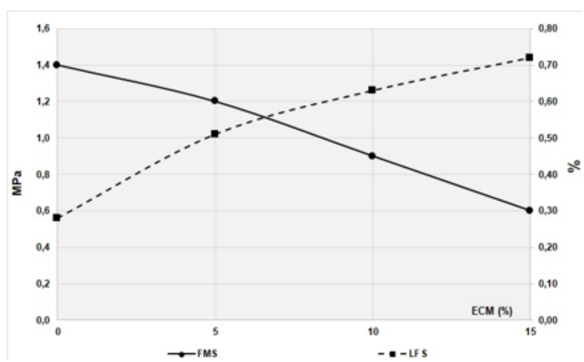

Figure 5. FMS and LFS as a function of additive percentage
Source. Authors.

In this respect, the resistance values are under the minimum values required by NTC 4205:2009, standard used for specifications, classification, and use in the construction of masonry. However, it is important to mention two significant aspects; first, sample L does not reach the minimum requirement, conditioning the normative non-conformity. Second, possibly by modifying the working conditions with additions less than 5%, or using superior temperatures, it is possible to reach the minimum requirement for Non-Structural Horizontal Drilling masonry in Light Units, which requirement should be superior to 2.0 MPa, this is assumed due to the close values reported. These products are widely commercialized in the region since they are used for dividing or enclosing walls without elevated loads; evidencing that, with certain controlled working conditions, it is viable to use MCC in formulations with clay-type soils to manufacture light construction materials and facade materials for enclosures [24]-[26].

Conclusions

The mineralogical composition indicated the clay, Quartz, Muscovite, and Kaolinite minerals as mineralogical phases of greater proportion, contributing to the dimensional control and plasticity of the formulations; additionally, Hematite, Anatase, and Cristobalite were found as minor phases, generating the predominant orange color of the specimens. The MCC evidenced Calcite and Kaolinite as inorganic components, and also Cellulose as an organic component, this mineral phase is common in materials based on vegetable polymers such as the cardboard from which these containers are made, increasing the mechanical resistance when drying.

The chemical composition of the clay presented principal oxides SiO_2, Al_2O_3, and Fe_2O_3 in combination with TiO_2, developed the orange color in the specimens, and also presented low contents of fluxing oxides; although SiO_2 was around the permissible range, it was not dangerous, but it is recommended that control be exercised during firing. The MCC reported CaO, generating the porosity, SiO_2, and minor concentrations of Al_2O_3, Na_2O, MgO, Fe_2O_3, TiO_2 K_2O, and others that contributed to the compatibility of the formulations.

Adding MCC increased the amount of water required for plasticity, however, until percentages of 5% enhance extrusion shaping, and no structural defects are observed.

MCC modify the ceramic aptitude towards products that meet the needs of the ceramic sector in the region; and the fibrous nature of the residue develops an increase of up to 1.88% in mechanical strength, contributing to a reduction in drying defects and controlling shrinkage to values under 8%, preventing problems associated with cracks or fissures.

MCC are pore formers that modify the microstructure of the clay body, developing porosity and limiting the final use of the products, but providing the opportunity to open new markets for products with insulating properties and a greater added value.

Normatively, results in percentages higher than 5% are not very promising, However, in less demanding conditions, it is possible to obtain lightweight bricks with minor structural dead load and less transportation costs; furthermore, it protects the environment, reducing the pollution generated by the MCC and the proportion of clay in the productive process.

Finally, the conditions for the use of molded cellulose containers were demonstrated, the results evidenced that the proportion in the mixture and the firing temperature are two important factors that affect the structural and mechanical properties of the ceramic products, but controlling the conditions of work, additions until 5% are economically, technically and environmentally feasible.

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