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Valorization potential of coffee grounds waste as a renewable pore-forming agent to produce low-cost porous ceramic support

Potencial de valorização de resíduo borra de café como agente formador de poro renovável para produzir suporte cerâmico poroso de baixo custo

Potencial de valorización del residuo de poso de café como agente formador de poro renovable para producir soportes cerámicos porosos de bajo costo

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Felipe Sardinha Maciel
ORCID: https://orcid.org/0000-0002-6739-699X
Universidade Estadual do Norte Fluminense Darcy Ribeiro, Brazil
E-mail: felipesmac@gmail.com
Isabela Oliveira Rangel Areias
ORCID: https://orcid.org/0000-0001-7046-7552
Universidade Estadual do Norte Fluminense Darcy Ribeiro, Brazil
E-mail: oraisabela@gmail.com
José Nilson França de Holanda
ORCID: https://orcid.org/0000-0001-7409-0027
Universidade Estadual do Norte Fluminense Darcy Ribeiro, Brazil
E-mail: holanda@uenf.br

Abstract

Every day expressive amounts of coffee grounds waste generated during making the coffee beverage are produced worldwide. The aim of this work was to investigate the valorization potential of the coffee grounds waste as an alternative pore-forming agent to produce porous kaolin-based ceramic supports. For this purpose, kaolin-based ceramic support formulations containing up to 40 mass % of coffee grounds waste were pressed and fired at temperatures ranging from 1000 to 1200 ºC. The sintering behavior and technical properties (linear shrinkage, mass loss, apparent density, mechanical strength, and apparent porosity) have been investigated. The microstructural evolution has been accompanied by scanning electron microscopy (SEM) and 3D-confocal microscopy. The obtained results demonstrated that the coffee grounds waste could serve as a very effective pore-forming agent to produce kaolin-based ceramic support with highly porous structures. Such ceramic supports incorporated with coffee grounds waste exhibited values of apparent porosity within the range of 41.87 % to 70.96 %. This results suggests that the coffee grounds waste, in the range of 10 - 40 mass %, could be a highly promising renewable porogenic material to be valorized to produce low-cost kaolin-based ceramic support with good porosity properties. This new approach could be an innovative alternative for the sustainable use of coffee grounds waste.

Keywords: Coffee grounds waste; Porogenic material; Kaolin-based support; Porosity; Valorization.

Resumo

Todos os dias quantidades expressivas de borras de café geradas durante o preparo da bebida café são produzidas em todo o mundo. O objetivo deste trabalho foi investigar o potencial de valorização do resíduo borra de café como agente formador de poro alternativo para produzir suportes cerâmicos porosos à base de caulim. Para tanto, formulações de suportes cerâmicos à base de caulim contendo até 40 % em massa de resíduo borra de café foram prensadas e queimadas em temperaturas variando de 1000 a 1200 ºC. O comportamento de sinterização e as propriedades técnicas (retração linear, perda de massa, densidade aparente, resistência mecânica e porosidade aparente) foram investigados. A evolução microestrutural foi acompanhada por microscopia eletrônica de varredura (MEV) e microscopia confocal-3D. Os resultados obtidos demonstraram que o resíduo borra de café pode servir como um agente formador de poros muito eficaz para produzir suporte cerâmico à base de caulim com estruturas altamente porosas. Tais suportes cerâmicos incorporados com resíduo borra de café apresentaram valores de porosidade aparente na faixa de 41,87 % a 70,96 %. Este resultado sugere que o resíduo borra de café, na faixa de 10 – 40 % em massa, pode ser um material porogênico renovável altamente promissor a ser valorizado para produzir suporte cerâmico à base de caulim de baixo custo com boas propriedades de porosidade. Esta nova abordagem pode ser uma alternativa inovadora para o uso sustentável do resíduo borra de café.

Palavras-chave: Resíduo borra de café; Material porogênico; Suporte à base de caulim; Porosidade; Valorização.

Resumen
Todos los días se producen en todo el mundo cantidades expresivas de residuo poso de café generados durante la preparación de la bebida de café. El objetivo de este trabajo fue investigar el potencial de valorización de residuo de poso de café como agente formador de poro alternativo para producir soportes cerámicos porosos a base de caolín. Por lo tanto, las formulaciones de soportes cerámicos a base de caolín que contenían hasta un 40 % en masa de residuo de poso de café fueran prensadas y cocidas a temperaturas que oscilaron entre 1000 y 1200 ºC. Se investigaron el comportamiento de sinterización y las propiedades técnicas (contracción lineal, pérdida de masa, densidad aparente, resistencia mecánica y porosidad aparente). La evolución microestructural se siguió mediante microscopía electrónica de barrido (SEM) y microscopía confocal-3D. Los resultados obtenidos demostraron que el residuo de poso de café puede servir como un agente formador de poros muy efectivo para producir soportes cerámicos a base de caolín con estructuras altamente porosas. Tales soportes cerámicos incorporados con residuo de poso de café presentaron valores de porosidad aparente en el rango de 41,87 % a 70,96 %. Este resultado sugiere que el residuo de poso de café, en el rango de 10 a 40 % en masa, puede ser un material porógeno renovable altamente prometedor que se valorará para producir soportes cerámicos a base de caolín de bajo costo con buenas propiedades de porosidad. Este nuevo enfoque puede ser una alternativa innovadora para el uso sostenible del residuo de poso de café.
Palabras clave: Residuo de poso de café; Material porógeno; Soporte a base de caolín; Porosidad; Valorización.

1. Introduction

In the 21st century, society and industry worldwide are undergoing intense transformations and challenges, one of which is the management of polluting solid wastes including those from biomass wastes. Coffee is one of the most prominent biomass commodities in the worldwide. In fact, the coffee beverage is enjoyed all over the world, and its market value and production volume grow more and more each year. Brazil has held a prominent position as the world's largest coffee producer and exporter for over 100 years. Brazil is also ranked as the second largest consumer in the world of coffee beverage. In 2020, Brazil alone consumed around 1.272 million tonnes of coffee with a per capita consumption of 5.99 kg/inhabitant.year of green coffee and 4.79 kg/inhabitant.year of roasted coffee (ABIC, 2022). The large consumption of the coffee beverage is centralized mainly in the homes and services sector (coffee shops, snack bars, bars, restaurants, hotels, offices, etc.). As a result, a solid waste material known as coffee grounds waste is generated on a large scale in the worldwide every day.

The coffee grounds waste is a biomass solid waste of organic nature generated during the preparation of the coffee beverage by infusion in hot water (~ 90 ºC). The coffee grounds waste *in natura* has high moisture content (~ 70 %). In the dry state, it appears as a powdery material of brown color. In many parts of the world, it is mainly discarded outdoors in an irregular manner with high negative impacts on the landscape and environment, or else stored in landfills resulting in high cost (Murthy & Naidu, 2012; Arya et al., 2021). The coffee grounds waste has been tested as a soil fertilizer (Cervera-Mata et al., 2019), but this disposal way is considered quite problematic due to its acidity and organic nature that demands a large amount of oxygen to be degraded. In view of this, the coffee grounds waste has also been tested to produce several materials such as briquettes, animal feed, biofuels, absorbent material, activated carbon, cosmetic products, green composites, alumina ceramics, and fired clay bricks (Eliche-Quesada et al., 2011; Murthy & Naidu, 2012; Soares et al., 2015; Laksaci et al., 2017; Kovalcik et al., 2018; Atabani et al., 2019; Hermann et al., 2019; Manni et al., 2019; Banu et al., 2020; Leow et al., 2021; Saberian et al., 2021; Mustafa et al., 2022).

The ceramic membranes are inorganic porous materials chemically inert and stable at high temperature with broad use in separation processes (microfiltration and ultrafiltration), with special emphasis on applications in the chemical, textile, metallurgical, paper, pharmaceutical, food, beverage and biotechnology industries, among others (Ali et al., 2018; Saini et al., 2019; Aissat et al., 2019; Abdullayev et al., 2019). The Ceramic membranes are generally composed of two main components, including porous support and a thin separation layer. In particular, the porous support corresponds to about 99 % of the membrane mass. Such ceramic membranes exhibit competitive advantages over polymeric membranes in terms of pressure resistance, mechanical strength, long life and ease of cleaning. Despite this, commercial ceramic supports are considered expensive due to the two main aspects: i) high cost of the synthetic raw materials used (e.g., alumina, cordeirite, titanium oxide, and mullite); and ii) high firing temperature used to produce the porous support. For these reasons, the development of new porous ceramic supports using cheap raw materials such as kaolin and clays, and the use of lower firing temperatures have been increasingly encouraged (Kouras et al., 2017; Ali et al., 2018; Saini et al., 2019; Aissat et al., 2019; Abdullayev et al., 2019; Elgamouz et al., 2019; Azaman et al., 2021). In addition to this, it should be highlighted that several solid wastes have been used as alternative raw materials to produce porous ceramic supports (Liu et al., 2016; Hossain et al., 2018; Rawat & Bulasara, 2018; Hubadillah et al., 2018a; Abdullayev et al., 2019; Liang et al., 2021). The utilization of solid wastes at the place of traditional raw materials has been verified to be the highly recommended, necessary and cheapest way for economic and ecological management of polluting solid wastes. For our best knowledge, the utilization of coffee grounds waste to produce porous supports for low-cost ceramic membranes has not been investigated yet. This is remarkably important in terms of environmental sustainability due to the coffee grounds waste of an essentially organic nature could be reclaimed as an effective renewable pore-forming material, and also valorized to produce porous supports for potential application in low-cost ceramic membranes.

Therefore, considering the growing concerns related to the sustainable destination of coffee grounds waste, this work is focused on the perspective of valorization of coffee grounds waste as a convenient pore-forming agent to produce porous kaolin-based supports for potential application in low-cost ceramic membrane.

2. Methodology

The study was developed at the Laboratory of Advanced Materials of the Universidade Estadual do Norte Fluminense Darcy Ribeiro, located in Campos dos Goytacazes-RJ, Brazil. Specifically, the study is experimental and developed on a laboratory scale, whose experimental methodology employed allowed the development of the study in a quantitative approach.

The starting raw materials used were kaolin and coffee grounds waste in the form of fine powders, as shown in Figure 1. Commercial kaolin with particle size < 325 mesh (< 44 µm, ASTM) was used. The coffee grounds waste sample was collected from household located in south-eastern Brazil (Campos dos Goytacazes-RJ) after the preparation of the coffee beverage by the hot water infusion and filtration method. In this process, the coffee grounds waste containing high moisture corresponds to the material retained in the filter. The collected sample was subjected to natural drying to decrease the moisture content, and then dried in an oven at 110 ºC for 24 h, disaggregated, and sieved through a 35 mesh **(**< 500 µm, ASTM) sieve.

Figure 1 - Starting raw materials used to produce porous ceramic support.

Source: Authors.

Table 1 presents the kaolin + coffee grounds waste formulations used. In this work, the kaolin was partially replaced by increasing amounts of coffee grounds waste used as a pore-forming agent. The ceramic support formulations were mixed and homogenized in a cylindrical blender for 30 min. Finally, the moisture content was adjusted to 7 %.

The chemical analysis of the raw materials was carried out by X-ray fluorescence. The loss on ignition (LoI) was obtained by calculating the mass % difference between dry sample at 110 °C and calcined sample at 1000 ºC for 2 h. The phase analysis of the raw materials was done by X-ray diffraction (XRD 7000 diffactometer, Shimadzu), by using monochromatic Cu-Kα radiation, scanning speed of 1.5º(2θ)/min, and 2θ ranging from 5° to 80°. JCPDS-ICDD files were used to identify the mineral phases.

Table 1 - The proportions of the mixtures for the formulations (mass %).

Raw materials	**Formulations**				
	FM1	**FM2**	**FM3**	**FM4**	**FM5**
Kaolin	100	90	80	70	60
Coffee grounds waste	0	10	20	30	40

Source: Authors.

The thermal behavior was evaluated in terms of differential thermal analysis (DTA), thermogravimetric analysis (TGA), and dilatometric curves. DTA/TG curves were obtained with a simultaneous thermal analyzer (STA 409E model, Netzsch) under air atmosphere from room temperature (~ 25 ºC) up to 1100 °C at a heating rate of 10 °C/min. The dilatometric tests were made on samples in the green state with a conventional dilatometer (DIL 402C model, Netzsch) within the 25-1050 ºC temperature range under air atmosphere and heating rate of 10 ºC/min.

The ceramic supports in form of disk (25 mm in diameter and 5 mm in height) were prepared by uniaxial pressing at 30 MPa, and then dried at 110 °C for 24 h. The green ceramic supports thus formed were fired at temperatures between 1000 and 1200 ºC at the heating rate of 5 °C/min, and then cooled to room temperature.

The ceramic supports produced were tested to determine the following physical and mechanical properties: mass loss, linear shrinkage, apparent density, apparent porosity, and tensile strength. The mass loss (ML) was determined by ML (%) =

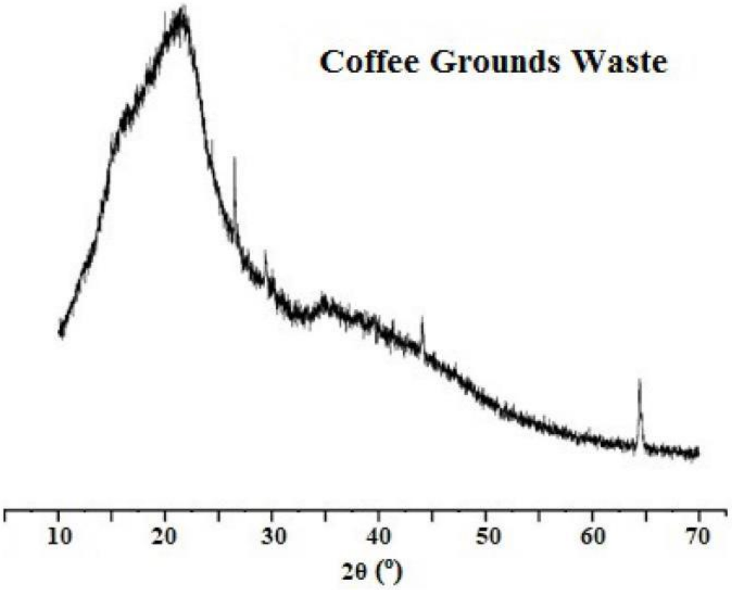
(M1 - M2)/M1 × 100, in which M1 is the mass of the dry support at 110 °C and M2 is the mass of the fired support between 1000 and 1200 ºC, by using a digital balance (± 0.01g). Linear shrinkage values upon drying and firing were determined from the variation of the diameter of the disk supports according to the ASTM C326 standard (American Society for Testing and Materials, 2018a). The apparent density and apparent porosity of the fired ceramic supports were measured according to the ASTM C373 standard (American Society for Testing and Materials, 2018b). Due to the disk geometry of the fired ceramic supports, the mechanical strength was determined in terms of tensile strength by the diametrical compression method (Fett, 1998; Chen et al., 2001). The tensile strength (τ) is given by $\tau = 2P/\pi dh$, where P is the breaking load, d is the support diameter and h is the supports thickness. The value of the breaking load (P) was determined using a universal testing machine (5582 model, Instron) at a loading rate of 0.5 mm/min.

The microstructure of fractured surfaces of the fired ceramic supports was observed by scanning electron microscopy (SEM SSX-550, Shimadzu), and the topography via laser scanning microscopy by using a confocal microscope (3D Measuring Laser Microscope, Lext OLS4000).

3. Results and Discussion

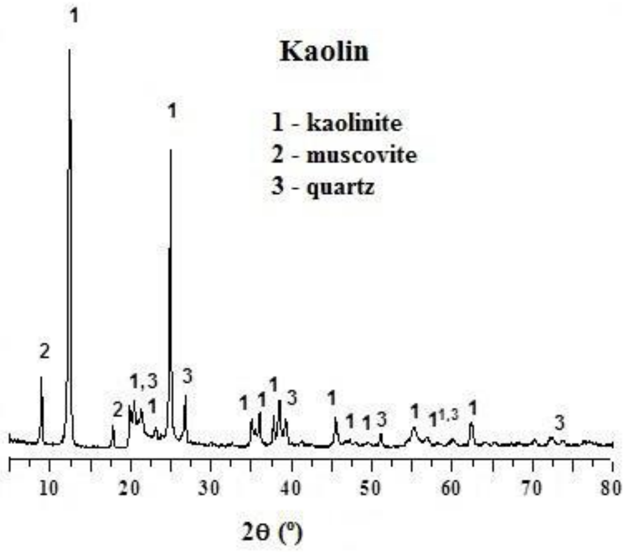
The X-ray diffraction pattern of the coffee grounds waste is shown in Figure 2. A broad diffraction peak at $2\theta \cong 22.5^{\circ}$ and a narrow diffraction peak at $2\theta \cong 35^{\circ}$ were detected, which are characteristic of lignocellulosic components (Rambo et al., 2015). This confirms the essentially organic nature of the coffee grounds waste. The X-ray diffraction pattern of the kaolin sample was dominated by the kaolinite peaks ($2SiO_2.Al_2O_3.2H_2O$; JCPDS-ICDD file: 14-0164), as shown in Figure 3. There are also traces of muscovite ($KAl_2Si_3AlO_{10}(OH)_2$; JCPDS-ICDD file: 7-25) and quartz (SiO_2; JCPDS-ICDD file: 46-1045).

Figure 2 - X-ray diffraction pattern of the coffee grounds waste sample.



Source: Authors.

Figure 3 - X-ray diffraction pattern of the kaolin sample.



Source: Authors.

The chemical analysis and loss on ignition (LoI) of the coffee grounds waste and kaolin used are given in Table 2. Loss on ignition of the coffee grounds waste sample implied very high mass loss of about 98.64 %. This result can be attributed to the decomposition of the coffee grounds waste at high temperature. Traces of Si, Al, Fe, Ti, Mn, Mg, Ca, K, Na, and P oxides were also detected. According to the data in Table 2, the kaolin sample is essentially composed of SiO_2 and Al_2O_3, which correspond to about 85.86 %. In addition, the kaolin used in this work has a SiO_2/Al_2O_3 ratio = 1.21, indicating that it is relatively pure composed essentially of kaolinite (Santos, 1989). This finding is confirmed by the LoI value of kaolin (12.88 %), which is close to the value of pure kaolinite (13.95 %). Such results are in agreement with the XRD pattern (Figure 3).

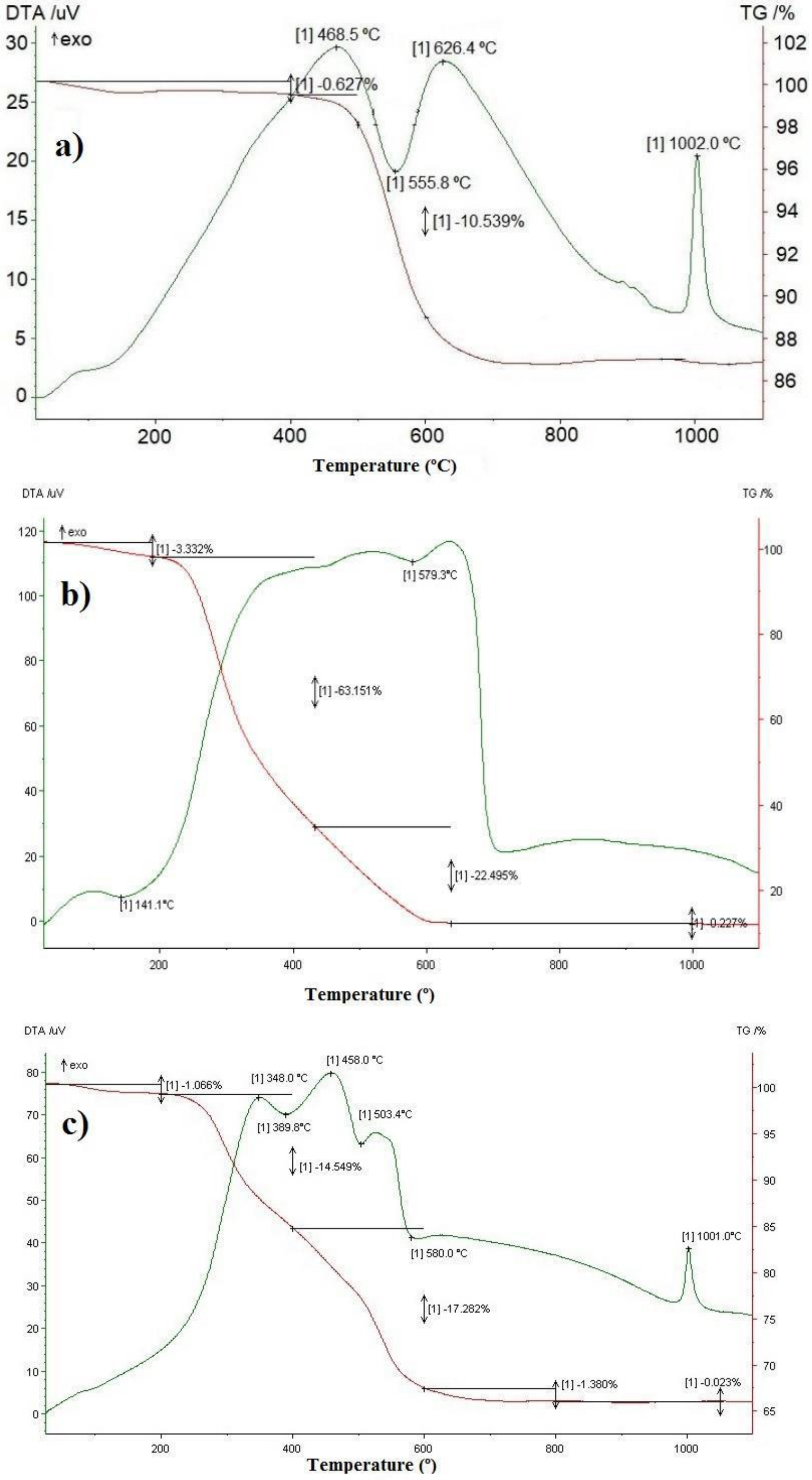
Table 2 - Chemical analysis and loss on ignition of the raw materials (mass %).

Components	kaolin	Coffee grounds waste
SiO_2	46.92	0.07
Al_2O_3	38.94	0.05
Fe_2O_3	0.27	0.05
TiO_2	0.05	0.05
MnO	0.05	0.05
MgO	0.05	0.13
CaO	0.05	0.15
K_2O	0.64	0.47
Na_2O	0.06	0.05
P_2O_5	0.09	0.29
LoI	12.88	98.64

Source: Authors.

The DTA-TG curves for the kaolin, coffee grounds waste, and FM4 formulation are shown in Figure 4. The kaolin sample (Figure 4a) corresponding to the FM1 formulation (coffee grounds waste-free formulation) presented the following thermal events with increasing temperature: 1) a small endothermic event between ~ 25 and 400 ºC associated with the release of moisture water accompanied by mass loss of 0.627 %; 2) relevant endothermic event at 555.8 °C related to dehydroxylation of kaolinite for formation of amorphous metakaolinite, accompanied by mass loss of 10.539 %; and 3) exothermic event at 1002.0 ºC without mass loss related to the formation of primary mullite (Santos, 1989; Coutinho et al., 2022). It may also be noted that the total mass loss obtained from the TG curve of 11.17 % was well correlated with the value of LoI of 12.88 % (Table 2). Such data are in accordance with the XRD pattern analysis, as shown in Figure 3. The thermal behavior of the coffee grounds waste reflects its organic nature, which is a biomass waste material basically made of extractives, hemicellulose, cellulose and lignin (Pujol et al., 2013; Dávila-Guzmán et al., 2013). According to the DTA-TG curves shown in Figure 4b, the observed thermal events can be described as follows: 1) endothermic event at ~ 141.1 ºC accompanied by mass loss of 3.332 % associated with the release of moisture water and decomposition of extractives; 2) a strong exothermic event between ~ 200 and 450 ºC due to the decomposition of hemicellulose and cellulose (i.e., loss of light volatile material) (Cerino-Córdova et al., 2020), accompanied by high mass loss of 63.151 %; 3) an intense exothermic event between ~ 450 and 630 ºC related to the decomposition of lignin (i.e., loss of heavier volatile material) (Cerino-Córdova et al., 2020), accompanied by high mass loss of 22.495 %; and 4) a small weight loss of 0.227 %, probably related to the decomposition of residual lignin, can be observed. The coffee grounds waste presented a total mass loss obtained of TG curve of about 89.21 %. This result is very important for the manufacture of a porous kaolin-based support, as it shows that the coffee grounds waste acts as a pore-forming agent in the fired structure. The thermal behavior of the FM4 formulation (with 30 % of coffee grounds waste) reflects those of the individual raw materials, as shown in Figure 4c. Exothermic events related to the thermal destruction of the coffee grounds waste are evident.

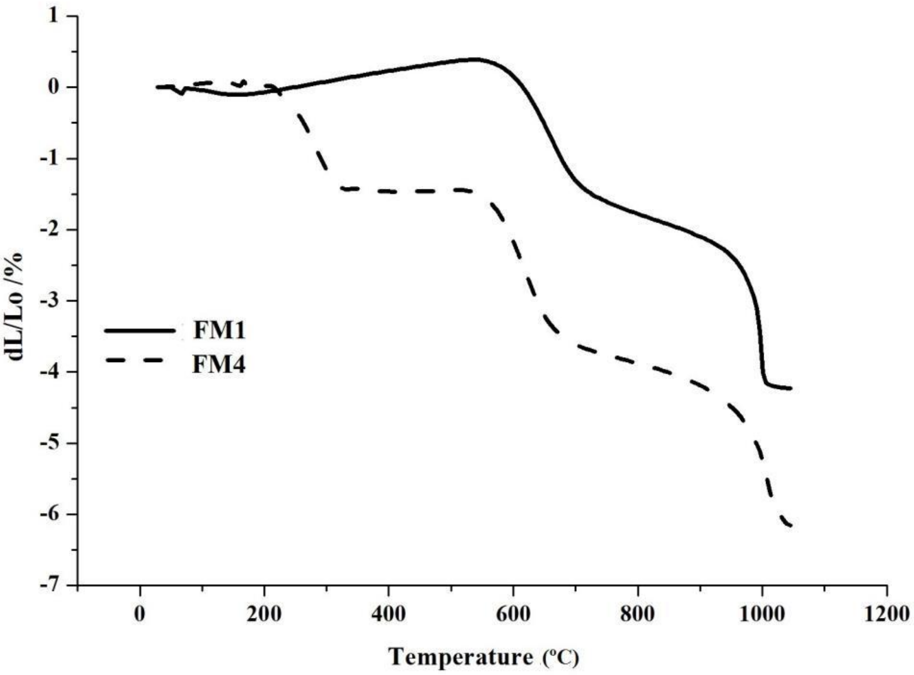
Figure 4 - DTA-TG curves: a) kaolin, b) coffee grounds waste, and c) FM4 formulation.



Source: Authors.

The sintering behavior of the FM1 formulation (100 % kaolin) and FM4 formulation (with 30 % of coffee grounds waste) was determined via dilatometric curve, as shown in Figure 5. As expected, the dilatometric curve of the FM1 formulation presented a classic behavior corresponding to pure kaolinite (Gomes, 1988). It were found five distinct regions of dimensional changes, described as follows: 1) small shrinkage up to ~ 200 ºC (release of moisture water); 2) small expansion between ~ 200 ºC and 550 ºC (thermal expansion of the solid particles); 3) slight shrinkage between ~ 550 ºC and 700 ºC (attributed to kaolinite dehydroxylation); 4) small shrinkage between ~ 700 ºC and 950 ºC (due to a solid-state sintering process with interparticle neck formation); and 5) sharp shrinkage between ~ 950 ºC and 1050 ºC (connected with the effects of formation of primary mullite and amorphous SiO_2 (Gomes, 1988; Santos, 1989) from a matrix that begins to develop a glassy phase). The dilatometric curve of the FM4 formulation indicated that the thermal destruction of the coffee grounds waste with the release of volatile compounds influenced the sintering behavior. The FM4 formulation exhibits a higher shrinkage during the sintering process.

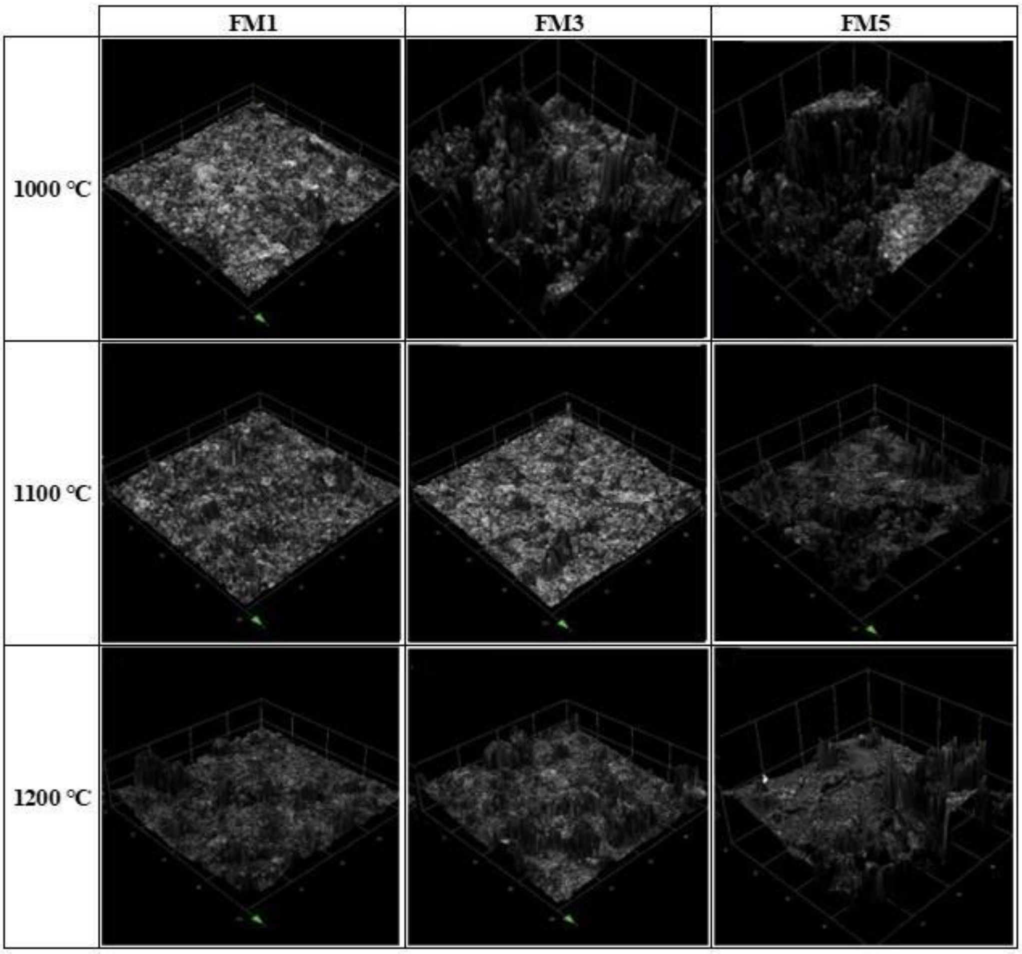
Figure 5 - Dilatometric curves of the FM1 and FM4 formulations.



Source: Authors.

Figure 6 shows 3D confocal images of the fired ceramic supports. For all firing temperatures, the influence of the addition of coffee grounds waste on the surface texture with increasing topography complexity can be observed. This finding can be explained by the thermal destruction of the coffee grounds waste during firing accompanied by the formation of a high amount of interconnected porosity. It can also be observed in the 3D images that the creation of a large amount of open porosity reduced the proportion of solid skeleton of the ceramic support. Such an effect tends to significantly influence the technical properties of the produced kaolin-based supports.

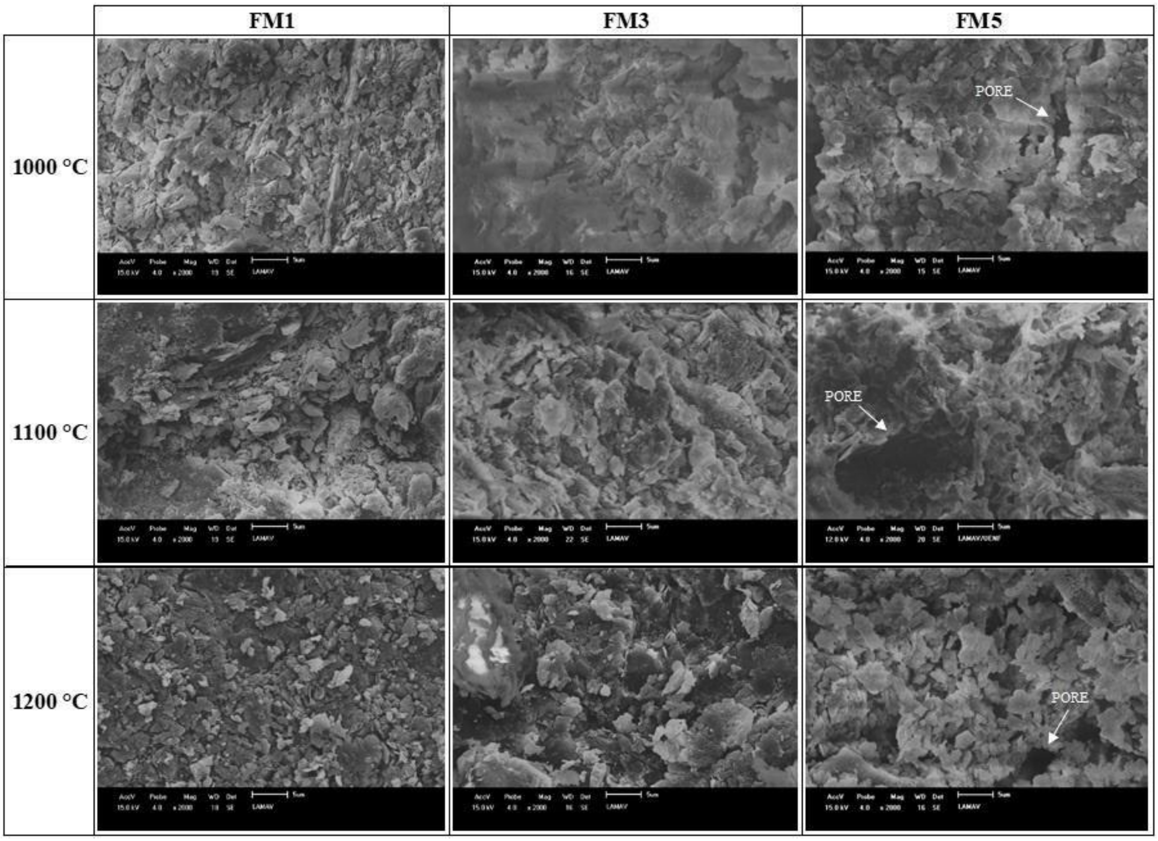
Figure 6 - 3D confocal images (x 2136) of the fired ceramic supports.



Source: Authors.

SEM Micrographs of ceramic supports fired between 1000 ºC and 1200 ºC observed at the fractured surfaces are shown in Figure 7. Such micrographs show the typical sequence of the fired microstructure of ceramic supports with increasing firing temperature and the amount of coffee grounds waste added. Two opposing trends emerged about the behavior of the fired microstructure. From the micrographs, a denser microstructure of the ceramic supports was obtained with increasing firing temperature. Unlike, the addition of coffee grounds waste brought about an important porosity variation in fired microstructure. In fact, it can be noted that the coffee grounds waste acts as a pore-forming agent, which results in highly porous microstructures. This effect is mainly due to the thermal destruction of coffee grounds waste during the firing process with the release of volatiles, according to the TG curves shown in Figures 4b and 4c. It is quite clear that the path taken by the released volatiles during the firing process creates the porous microstructure of the ceramic support. The greater the amount of coffee grounds waste added, the greater the amount of observable pores of different sizes and irregular morphology are produced. This is in line with increasing topographical complexity, as shown in Figure 6. A trend of increasing pore size as the amount of coffee grounds waste is increased can also be observed. However, the presence of macrodefects such as cracks and fissures were not found. Although SEM micrographs did not allow accurate estimation of pore size, it is noted that a significant amount of pores formed was within the size range below 10 µm. This finding is interesting as it indicates that apparently porous kaolin-based supports produced with coffee grounds waste could be applied in microfiltration processes, which require pore sizes between 0.1-10 µm (Habert et al., 2006).

Figure 7 - SEM micrographs of the fired ceramic supports.



Source: Authors.

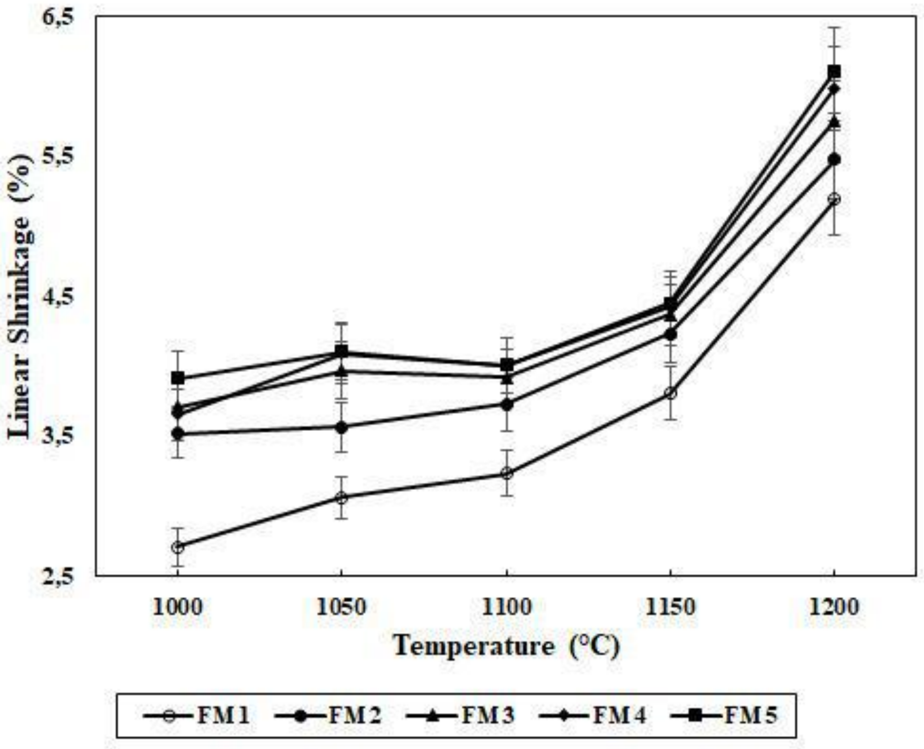
The visual appearance of ceramic supports produced with different amounts of coffee grounds waste is shown in Figure 8. As seen, the addition of coffee grounds waste did not influence the firing color. All porous ceramic supports produced had a white-firing color typical of kaolin-based materials.

Figure 8 - Typical appearance of the produced porous ceramic support.

Source: Authors.

Figure 9 shows the linear shrinkage of porous ceramic supports. The linear shrinkage is a physical property fundamentally related to the sintering degree of the sample during the firing process. Linear shrinkage values within the range of 3.09 to 6.11 % were found. The results demonstrated that both the firing temperature and the amount of coffee grounds waste caused an increase in the total linear shrinkage of the ceramic supports. The effect of the firing temperature was to cause an increase in the vitrification degree that tends to increase the linear shrinkage values, regardless of the coffee grounds waste amount added. It is also verified in Figure 9 that the addition of coffee grounds waste contributes to increase the linear shrinkage of the porous supports. This behavior is in line with the dilatometric curve (Figure 5). The reason for this important finding is that the coffee grounds waste acts as a solid fuel of high calorific power (Jeguirim et al., 2014; Brunerová et al., 2020) during the firing process. In addition to this, the ashes rich in fluxing compounds (Eliche-Quesada et al., 2011) produced during combustion of the coffee grounds waste are incorporated into the ceramic matrix. Both effects tend to aid the sintering of the kaolin-based supports.

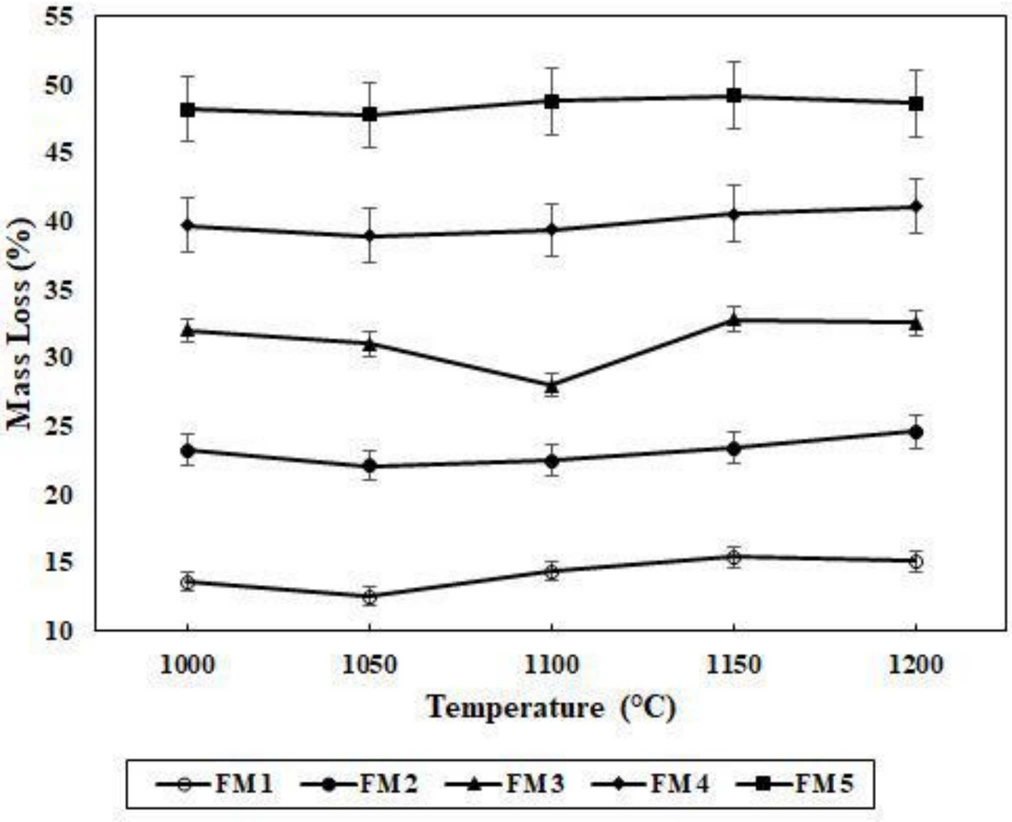
Figure 9 - Linear shrinkage of the fired ceramic supports.



Source: Authors.

The mass loss of the ceramic supports during the firing process is shown in Figure 10. The mass loss is a key parameter in order to obtain a porous support. As expected, the ceramic support produced only with kaolin (FM1 formulation) had lower mass loss (12.53 – 15.41 %), which is close to the LoI value of kaolin. However, it was found that the ceramic supports incorporated with coffee grounds waste presented high mass loss values (22.08 – 49.21 %). Such mass loss values are essentially connected with the thermal destruction of the coffee grounds waste (i.e., decomposition of organic matter), and is in accordance with the TG curve shown in Figure 4b. It was also observed that, at any firing temperature, the effect of the coffee grounds waste was to substantially increase the mass loss of the kaolin-based ceramic supports.

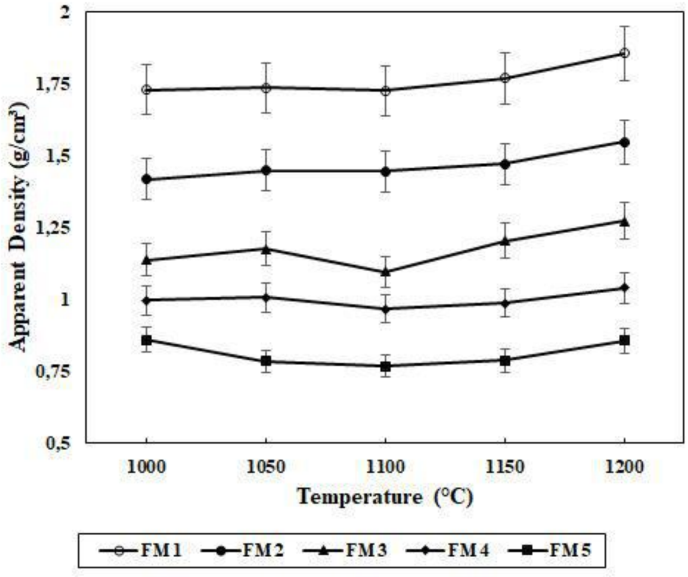
Figure 10 - Mass loss of the fired ceramic supports.



Source: Authors.

The apparent density of the ceramic supports is shown in Figure 11. As may be observed, the ceramic supports produced exhibited values of apparent density ranging over a wide range from 0.77 g/cm^3 to 1.86 g/cm^3. The densification behavior of the ceramic supports containing coffee grounds waste is quite intricate due to the simultaneous opposite effects, which occur as the firing temperature increases. These effects can be described as: 1) sintering; and 2) degassing related mainly to the release of volatiles from the combustion of coffee grounds waste, as shown in Figure 10. The coffee grounds waste impacts the sinterability of the ceramic supports, as seen in the dilatoletric curves (Figure 5) and linear shrinkage (Figure 9). It can also be seen in Figure 11 that the apparent density values of the ceramic supports changed very little when the temperature was increased up to 1100 ºC, indicating predominance of solid state sintering. Above 1100 °C, however, the apparent density tends to increase. This finding indicates that the ceramic supports sintered prevalently by viscous flow (Milheiro et al., 2005). However, at any firing temperature, the effect of the high mass loss is dominant on the densification behavior. Thus, the thermal destruction of the coffee grounds waste during the firing process strongly accounts to delaying densification and, as such, more porous ceramic supports are produced. This is in accordance with the microstructural and topographical features, as shown in Figures 6 and 7.

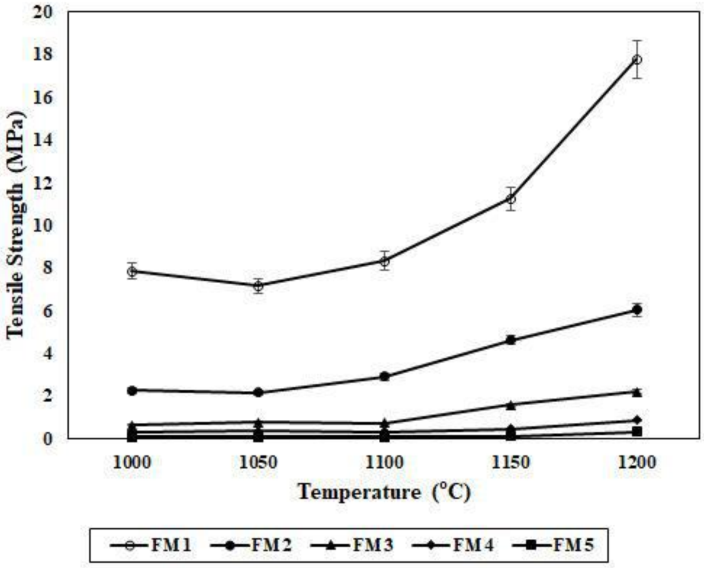
Figure 11 - Apparent density of the fired ceramic supports.



Source: Authors.

The diametral tensile strength of the fired ceramic supports is shown in Figure 12. Due to the cylindrical disk geometry of the ceramic supports, the diametral tensile strength data should only be compared with each other. As seen, a significant variation in the diametral tensile strength values (30 - 6590 kPa) as a function of the firing temperature and amount of coffee grounds waste occurred. However, firing temperature and coffee grounds waste had opposite effects on the mechanical strength. The effect of the firing temperature above 1100 ºC is to progressively increase the tensile strength due to the greater sinterability of the ceramic supports. By contrast, a trend of decreasing tensile strength with the increase of the amount of coffee grounds waste was well established. These behaviors were expected, given that the combustion of the coffee grounds waste lead to high mass loss (Figure 10) with concomitant decrease in the level of densification (Figure 11). In particular, it was also found that additions of high amounts of coffee grounds waste implied a strong decrease in the mechanical strength of the fired ceramic supports.

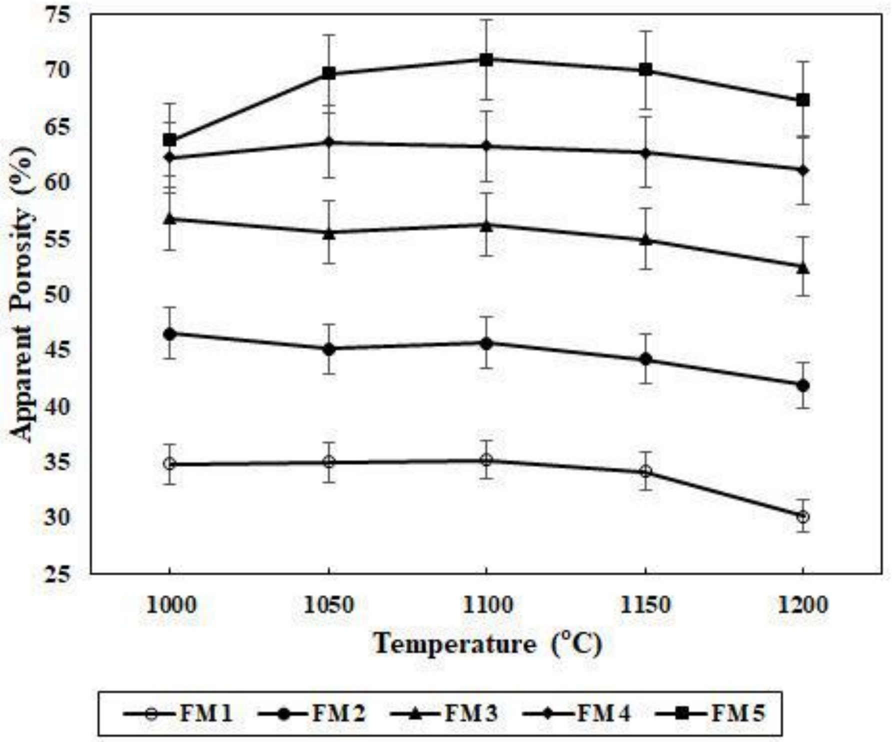
Figure 12 - Tensile strength of the fired ceramic supports.



Source: Authors.

The apparent porosity of the ceramic supports is shown in Figure 13. Apparent porosity is a very important physical property, as it provides the probable amount of open pores of the ceramic support in relation to its total volume. For all firing temperature, the apparent porosity values were found to increase in the ceramic support structure, as the coffee grounds waste amount increased up to 40 %. Specifically, the thermal destruction of the coffee grounds waste leaves a large fraction of pores within the sintered kaolin-based support. For this reason, the coffee grounds waste could be remarked as an efficient pore-forming agent during the firing process. Such a result is in good agreements with the fired microstructures (Figures 6 and 7), mass loss (Figure 10), and apparent density (Figure 11). This is very relevant for practical application in low-cost ceramic membranes due to the possibility of increasing the permeability of the porous support. It should be highlighted that the mechanism of formation of open pores in this new kaolin-based ceramic support is directly related to the processes of thermal destruction of the coffee grounds waste, as described in the DTA-TG curves (Figure 4b).

Figure 13 - Apparent porosity of the fired ceramic supports.



Source: Authors.

In this work, the kaolin-based ceramic supports incorporated with coffee grounds waste showed apparent porosity values in the range from 41.87 % to 70.96 %. Such data are in line with the porosity level required for the production of porous supports applied to low-cost ceramic membranes. In fact, the obtained results in this work are in good agreement with those reported by Kumar et al. (2015), Kouras et al. (2017), Lima et al. (2018), Hubadillah et al. (2018b), Abdullayev et al. (2019) and Jiang et al. (2019). Thus, the coffee grounds waste utilized in this work had a very positive influence on the creation of open pores and, as such, it may be quite promising to be valorized to produce porous kaolin-based support. From a practical application point of view, however, this work will continue in the future, including accurate determination of the pore characteristics, membrane assembly, and permeation flux tests in order to determine the efficiency of the filtration properties of the porous kaolin-based support produced with coffee grounds waste.

4. Conclusion

This work demonstrated in a very promising way that the coffee grounds waste have the potential to be valorized as a low-cost renewable biomass raw material for the production of porous kaolin-based ceramic support. The obtained results proved that the coffee grounds waste behaved as an excellent pore-forming agent, which facilitated the creation of high open porosity in the structure of the kaolin-based ceramic support when fired at different temperatures.

The sintering behavior, microstructure, and technical properties of kaolin-based ceramic supports are strongly dependent on the amount of coffee grounds waste added. The greater the incorporation of coffee grounds waste, the stronger the effects were observed. In addition, microstructural analyzes by SEM and 3D-confocal microscopy found that the coffee grounds waste led to the obtaining kaolin-based ceramic supports with highly porous structures. Such characteristics are relevant for the production of porous kaolin-based ceramic supports of good quality.

It was found that high porosity kaolin-based supports could be produced using varying amounts of coffee grounds waste (between 10 and 40 %) as a partial replacement for natural kaolin, fired between 1000 and 1200 ºC, with good porosity properties for possible application in low-cost ceramic membranes. Such new approach can be considered as a viable option to minimize the harmful effects caused by improper disposal of coffee grounds waste, but it also suggests a suitable way to produce low-cost porous kaolin-based ceramic support.

Finally, the pore characteristics, membrane assembly, and permeation flux tests related to the efficiency of the filtration properties of the porous kaolin-based support produced in this study will be the focus of future researches and articles.

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