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Influence of Packaging Cullet Size on Synterized Properties Produced From Clay Used In the Ceramic Industry

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ABSTRACT: *In the process of producing sintered clay components, solid waste can be incorporated as raw materials. One of the wastes that is widely used for reuse is packaging cullet. There are several works in the literature in which cullet is used as a raw material in the production of clay aggregates and other sintered products, but there are few reports of the influence of their particle size on the sintering process. The present work aimed to investigate the influence of particle size of cullet embedded in a clay that is used in the ceramic industry of Sergipe. The clay was characterized by XRD diffraction, DTA-TG differential thermal analysis. Subsequently clay, cullet and clay-cullet samples were characterized by FRX chemical analysis. The cullet was incorporated into the clay in the proportions (10, 20, 30 and 40% by mass) with two different average cullet particle diameters (450 and 37.5 μm) being characterized by dilatometry. Subsequently sintered were made with the clay-cullet samples burned at 3 different temperatures (1050, 1100 and 1150°C), which were characterized by their linear shrinkage, loss of fire mass, density evolution, water absorption and mechanical resistance. The best result of mechanical resistance was the aggregate containing 30% 37.5 μm cullet, sintered 1100 ° C. Particle size was a key parameter to ensure the efficiency of cullet incorporation into clay, within incorporation and temperature ranges.*

KEYWORDS: *Sintered Clay. Solid Waste. cullet Particles.*

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I. INTRODUCTION

In the process of producing sintered ceramic components as synthetic aggregates, ceramic tiles, porcelain tiles, tiles and blocks, it is mainly used as clays and sands, and it is possible to incorporate solid residues such as calcined sewage sludge, marble dust, among other wastes, in addition to packaging and window glass, known as soda-lime glass [1].

The incorporation of residues aims to reintroduce them in the production scale, and improve the production control of sintered products, such as reducing sintering temperature, water absorption and porosity, increase of specific mass and compressive strength [2].

Packaging cullet is a recurring solid waste in urban centers. In the ceramic industry, cullet waste can be incorporated into clays with the function of forming a liquid phase and guaranteeing a reduction in the sintering temperature of the final product, because the cullet has a low temperature softening point compared to clays [3].

In the literature there are several works in which cullet is used as a raw material in the production of sintered, but there are few reports of the influence of cullet granulometry on sintered clay[1-6].

The present work aimed to investigate the influence of the particle size of cullet embedded in a clay that is used in the ceramic industry, and to evaluate the possibility of reducing energy costs of the production of sintered.

II. MATERIALS AND METHODS

The clay was collected in the production line of a ceramic industry in the state of Sergipe, Brazil, was distorted, ground with hammer mill and sieved in the 150µm opening sieve. Subsequently, it was oven dried at 100° C for 4 hours. A qualified sample was separated for the mineralogical characterization of the clay was made using XRD for phase determination, also made XRD of the cullet. For phase analysis, the MATCH software was used. Additionally, a thermal characterization of the clay was performed using combined DTA-TG techniques, with heating rates of 10 °C / min, in the temperature range between 30 and 1200°C.

The cullet containers were washed, their labels were removed, those of a manual comminution and later the ball mills, as the cullet ones used were those between the opening sieves of 600 and 300 µm (average size 450µm), and as a sieve pass diameter of 75 µm (average diameter 37.5 µm), optical microscopy with 50 times magnification of the two particle sizes used was performed.

Samples were formulated with 0, 10, 20 30 and 40% mass cullet in clay [2-3], with both particle sizes (37.5 and 450µm). clay-culet samples were characterized by chemical analysis and dilatometry, where the bodies were conformed with 8% humidity, pressed at 37.3 MPa, with a diameter of 6 mm and lengths ranging from 9.7 to 15 mm, the heating rate used was 10°C/min in the temperature range between 30 and 1200°C.

Cylindrical specimens with a diameter of 20 mm and height of 15 mm on average were made, with a dry mass of approximately 9 g, conformed to 8% humidity and 63.7 MPa. The green bodies after drying in an oven at 100°C were baked and burned at a rate of 10°C /min using one hour plateaus at maximum temperature at 1050°C, 1100 °C and 1150 °C [3 and 7]. The synthesized were characterized by water absorption, density evolution, compressive strength using an electric press, and linear shrinkage using a metal caliper.

III. RESULTS AND DISCUSSION

XRF

The chemical analysis of the raw materials by the X-ray fluorescence technique is shown in Table 1, where it can be seen that the clay used is mostly silica and alumina, with high levels of calcium oxide and iron. which are elements associated with clay mineral structures, such as feldspars, typical of traditional ceramic products [8]. Cullet is mainly composed of silica and calcium and sodium oxides, and can be classified as a soda-lime glass. The chemical composition of the caly-cullet samples was calculated based on the proportions of mass incorporation.

SiO_2 content ranged from 52.0 to 58.2%, while Al_2O_3 contents ranged from 11.3 to 18.0%. The ratio (SiO_2 / Al_2O_3) ranged from 2.9 to 5.2, which are higher than the classical values for kaolinite (SiO2 / Al_2O_3: 1.18) or montmorillonite (SiO_2 / Al_2O_3: 2.36) [9], indicating that the higher this coefficient, the higher the percentage of free quartz in relation to accessory minerals present in clays, which can be confirmed by the clay XRD shown in Fig. 1(A). In the specific case Of the clay-cullet sample, SiO_2 is also in the form of amorphous silica due to the incorporation of cullet.

Calcium oxide (CaO) up to 3% has a flux action. Above this value, it is an indication that there are carbonates that are harmful to the densification phenomenon [10]. Fe_2O_2 content was higher than 5% in clay, being responsible for the red color after burning [8].

Table 1: Chemical composition obtained using the X-ray fluorescence technique of Saint Felix Clay, of soda-lime glass and the calculated composition of clay-cullet samples

Oxides	Clay (%)	Soda-lime glass (%)	Formula 10% (%)	Formula 20% (%)	Formula 30% (%)	Formula 40% (%)
SiO_2	52,0	68,6	53,7	55,3	57,0	58,6
Al_2O_3	18,0	1,2	16,3	14,6	12,8	11,3
CaO	13,0	9,7	12,6	12,3	11,0	11,7
Fe_2O_3	7,9	0,1	7,1	6,3	5,5	4,8
K2O	3,7	0,2	3,3	3,0	2,6	2,3
MgO	3,1	2,1	3,0	2,9	2,6	2,7
NaO	1,4	17,9	3,0	4,7	4,6	8,0
TiO_2	1,0	-	0,9	0,8	0,7	0,6
SO_3	-	0,2	0,0	0,0	0,0	0,1
Loss to ignition (%)	14,1	1,0	12,8	11,5	10,1	8,9
Proportion Si/Al	2,9	57,2	3,3	3,8	4,5	5,2

XRD

The phases present in the Saint Felix clay X-ray diffractogram were identified considering the main elements identified in the chemical analysis by the Match! demo version using Inorganic Crystal Structure Database - ICSD crystallographic forms. The result of the identification of phases of Saint Felix clay can be seen in Fig. 1(A), consisting of quartz, muscovite, kaolinite, feldspar, montmorillonite, hematite and calcite, which are common phases for local clays [6 and 11].

Calcite is a recurrent phase of calcium carbonate in sedimentary clays, and at levels above 3%, it is difficult to sinter the synthetic clay aggregate due to the carbon dioxide released during its dissociation, which generates an expansive phenomenon [10]. Fig. 1(B) shows the pattern of the Soda - Cal glass used, which consists of an expected pattern for an amorphous material [2].

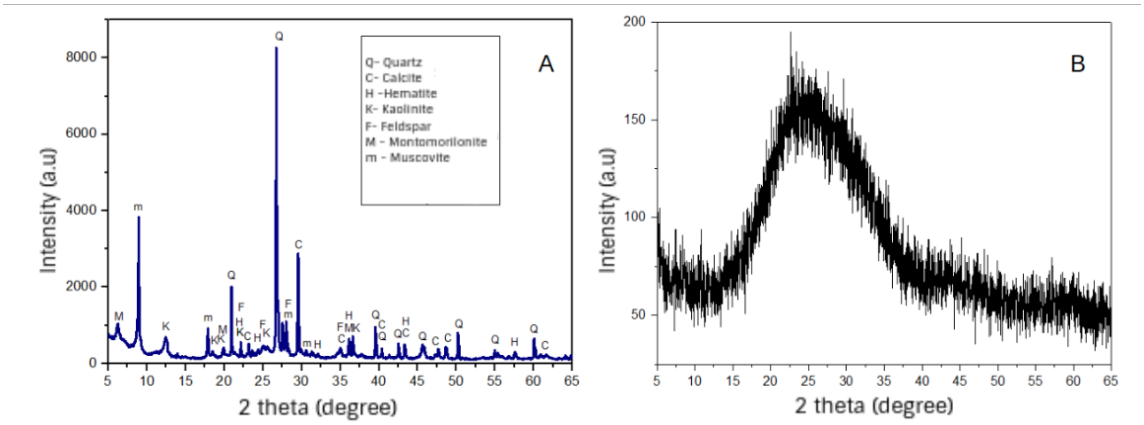

Fig.1. (A) X-ray diffraction pattern of Saint Felix clay and its respective mineral phases according to the ICSD database, (B) X-ray diffraction pattern of Soda - Cal glass used

DTA-TG

Through the mineralogical characterization of Saint Felix clay, it was possible to understand the events that occur in the DTA / TG curves. As DTA / TG curves of Saint Felix clay, shown in Fig. 3, show a mass loss up to 200 ºC, referring to the loss of water adsorbed in the clay (1.25%). It was observed in the temperature range of 500ª 600 the dissociation of organic clay material (4%) [12]. The release of CO_2 due to the decomposition of calcite between 660 and 770 ºC [10], represents more than half of the mass loss during the process (8.1%). The decomposition of kaolinite occurs between 900 and 1060 ºC (0.3%) and between 1070 and 1160 ° C, there is an appearance of the liquid phase [10]. The total mass loss of Saint Felix clay was 14.1%.

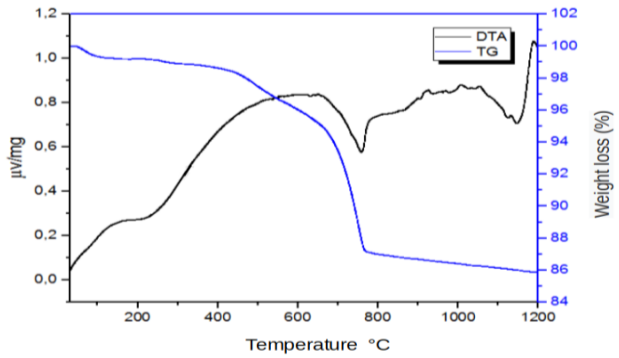

Fig. 3. Graph of thermal analysis obtained by the combined technique DTA / TG of Saint Felix clay

Optical Microscopy

Optical microscopy of the passing cullet particles in the 75 µm aperture screen shows the presence of particles smaller than 1 µm, as shown in Fig. 4(A). Already in the microscopy of the particles that were between the opening sieves 600 and 300 µm, it can be observed that even after grinding, some particles preserved smooth surfaces of their original objects, as shown in Fig. 4(B). Smooth surfaces are less reactive than rough surfaces, which can slow thermal processes such as sintering.

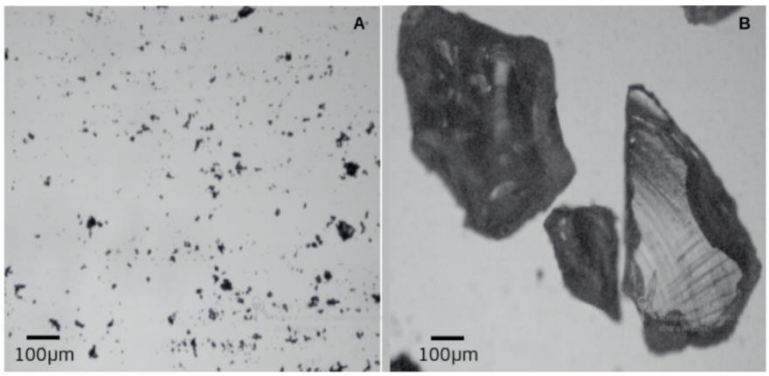

Fig. 3. Image used by optical microscopy in which: (A) Cullet particles that are struck by the 75 µm sieve, with an average diameter of 37.5 µm. (B) Cullet particles that reached the 600 µm sieve and were trapped in the 300 µm sieve, with an average diameter of 450 µm

Dilatometry

Through the DTA / TG curves, it is possible to correlate the physical-chemical events that occur in the clay with the expansion and retraction events that were observed in the dilatometry curves. As Saint Felix clay dilatometry curves and clay-cullet samples, including 4 bulk cullet incorporation contents (10, 20, 30 and 40%) and with two different average cullet exposure diameters (37.5 and 450 µm), are included. in Fig. 4.

The shrinkage up to 250°C gives the loss of adsorbed water in the clay and cullet areas and dehydration of clay minerals. The expansive phenomena observed between 300 and 600ºC should dehydrate by montmorillonite and burn organic materials, or that releases carbon dioxide and monoxide. At 573°C, there is an expansive phenomenon caused by the change from quartz α to quartz β [12].

The expansive phenomena between 660 and 770ºC were generated by the dissociation of calcium and CaO and CO_2[10]. From 800°C onwards, the sintering process begins, which generates a sharp shrinkage to 900°C, where a small expansion occurs between 900 and 1080°C, proving the loss of chemically bound and decomposed water from kaolinite. After 1100ºC, it begins the formation of the liquid phase, or promotes a rearrangement of the catches, contributing to the retraction [13].

A 40% incorporation of cullet was the best result for the 37.5 µm particle size, but it was the worst result for the 450 µm particle size.

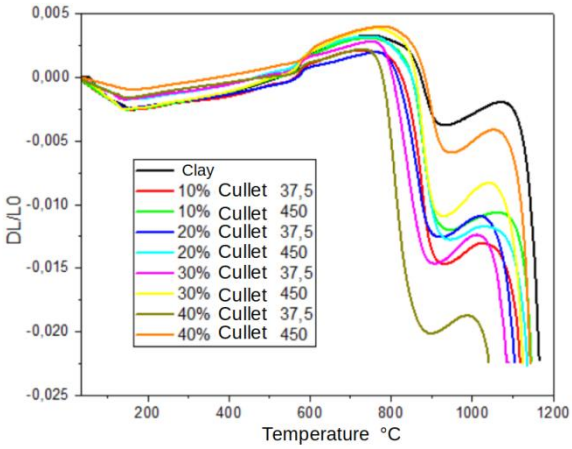

Fig.4. Dilatometric curves of the clay-cullet samples under heating rate of 10ºC / min

Visual Aspect

Fig. 5 shows the visual aspect of the specimens produced at temperatures of 1050ºC, 1100ºC and 1150ºC, with cullet incorporation contents between 0 and 40% by mass and two average cullet particle diameters, 37.5 and 450 µm. The difference between the color of the sintered ones increased as the cullet content and temperature increased, in which it can be noticed that besides the color change there was a gain of vitreous luster, which was more accentuated in the sintered ones with 37.5 particles. µm, the use of smaller cullet particles contributed to the homogeneity of the sintered color distribution.

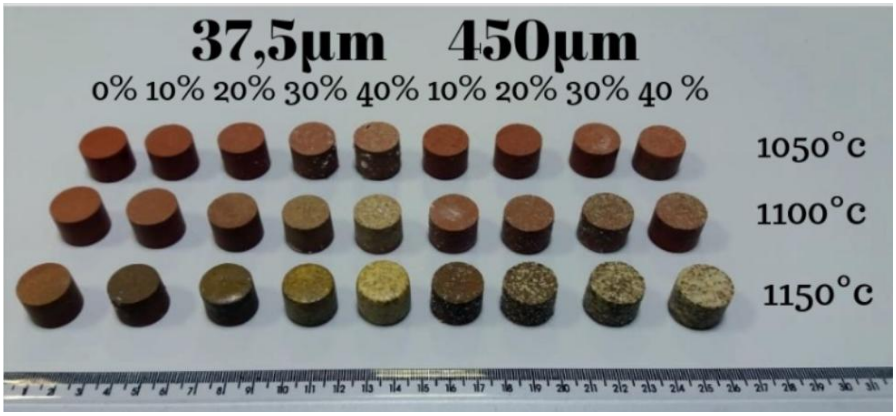

Fig. 5. Visual aspect of the sintered produced, in which columns the cullet incorporation content with different average cullet particle diameters (37.5 µm and 450 µm) is identified, and in the lines the firing temperature

Density

Table 2 shows the evolution of sintered density in relation to different production parameters, the use of smaller cullet particles contributes to the reduction of sintered density as temperature and incorporation content increased, which can be associated with increased gas trapping phenomenon during liquid phase sintering. The sintered ones containing larger cullet particles only showed a density reduction at a temperature of 1150 ºC, which indicates that the gas trapping phenomenon happened late [5].

Table 2: Evolution of sintered density (g / cm^3) as a function of firing temperature, mass glass incorporation content (%), glass particle size (µm)

Samples Temperature	Clay	10%-37.5	20%-37.5	30%-37.5	40%-37.5	10%-450	20%-450	30%-450	40%-450
1050°C	2,40	2,50	2,26	2,25	2,07	2,59	2,52	2,65	2,70
1100°C	2,47	2,47	2,48	2,57	2,36	2,52	2,52	2,61	2,70
1150°C	2,52	2,73	2,40	2,3	1,97	2,61	2,46	2,46	2,43

Compressive Rupture Strength

In Table 3, it is possible to see the evolution of the compressive rupture strength of the sintered in the production functions, noting if the best results are concentrated in the temperature of 1100ºC, showing the ideal burning temperature of the sintered. At a temperature of 1050 ° C, observe that all results with glass obtained results superior to those of pure clay. At 1150 ° C, the sintered ones contain 20% or more glass for the two sizes of use, which are resistant to pressures lower than the sintered ones, clay only. The lowest strength was sintered containing 40% glass, with an average diameter of 37.5 µm, sintered at 1150 ° C, the bodies were the only ones that were generated for designs designed for breaker press.

Table 3: Evolution of compressive rupture strength (MPa) of sintered as a function of firing temperature, mass glass incorporation content (%), and glass particle size (µm)

Samples Temperature	Clay	10%-37,5	20%-37,5	30%-37,5	40%-37,5	10%-450	20%-450	30%-450	40%-450
1050°C	38,6	61,4	49,3	57,3	51,3	54,7	69,8	68,9	61,2
1100°C	72,8	79,4	70,0	135,1	94,4	59,7	65,3	76,6	107,9
1150°C	65,15	79,5	40,0	41,0	32,7	85,1	57,2	52,2	49,4

Water Absorption

Table 4 shows the evolution of sintered water absorption as a function of production parameters. Sintered glass particles with 37.5 µm particle size obtained better results at temperatures of 1100 and 1150ºC. Greater grain size obtained superior absorption results. For both particle sizes the water absorption decreased as

the glass incorporation content and temperature increased.

Table 4: Evolution of water absorption (%) of sintered as a function of firing temperature, mass glass incorporation content (%), and glass particle size (µm)

Samples Temperature	Clay	10%-37,5	20%-37,5	30%-37,5	40%-37,5	10%-450	20%-450	30%-450	40%-450
1050°C	6,11	11,89	11,37	9,41	7,98	11,44	8,49	6,91	3,18
1100°C	12,60	14,32	6,87	1,96	0,54	9,34	7,02	3,80	3,68
1150°C	10,51	3,02	0,31	0,12	0,06	3,54	1,60	0,92	0,69

Linear Shrinkage

In Table 5, there is an evolution of the linear shrinkage of sintered heights as a function of production dimensions. The best result was to contain 30% glass with a mass of 37.5 µm, burned at 1050ºC. The worst result was the use of 40% glass in the mass with 37.5 µm reduction, burned at 1150ºC.

Table 5: Evolution of linear shrinkage of height (%) of sintered as a function of firing temperature, glass incorporation content in mass (%) and glass particle size (µm)

Samples Temperature	Clay	10%-37,5	20%-37,5	30%-37,5	40%-37,5	10%-450	20%-450	30%-450	40%-450
1050°C	0,34	0,69	0,00	6,67	0,51	1,28	0,00	0,58	0,58
1100°C	0,12	1,36	2,87	5,31	3,56	-0,71	-1,29	-0,12	1,05
1150°C	1,61	4,69	5,50	1,83	-5,00	0,85	-2,33	-2,50	-3,33

Visual Aspect Of The Fracture Surface And The External Face

In Fig. 6 the visual aspect of the fracture surface and the external face of the sintered at 1150 ° C containing 40% glass with particle sizes of 37.5 and 450 µm can be seen. The sintered ones produced with smaller particles had accentuated the phenomenon of trapping cavities during sintering, and a homogeneous matrix. As for the sintered ones that used larger particles had a heterogeneous matrix divided into two phases, glassy phase and clay, the cavities generated by the gas trapping phenomenon were concentrated in the glassy part. In both cases the surfaces were without pores because the gases that form on the surface exhale into the atmosphere. The pore free surface contributed to the low water absorption of the sintered.

The excess of isolated cavities generated by the gas trapping phenomenon generated by excess liquid phase formation during the sintering process, some up to 1 mm in diameter, compromised the aggregate resistance as the cavities are weak points in the sintered structure [5].

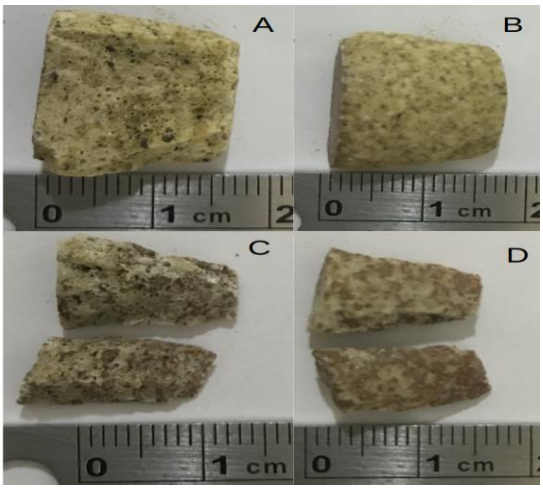

Fig. 6. fragments of sintered bodies at 1150 ° C containing 40% glass in which (A) fracture and (B) external face of the sintered formulated with glass particles of average diameter 37.5 µm. (C) fracture and (D) surface of the sintered formulated particle with an average diameter of 450 µm

In Fig. 7, it is observed that the fracture of the sintered that obtained the best mechanical result, 135 MPa of compressive strength, in the homogeneous and dense matrix, is not observed the presence of cavities generated by gas trapping, which contributed to the good mechanical performance. The sintered in question also had low water absorption (1.96%) and high linear shrinkage (5.31%).

Fig.7. Sintered fracture that had the best apparent mechanical performance, containing 30% glass embedded with 37.5 µm particle diameter, sintered at 1100°C

IV. CONCLUSION

All cullet incorporations obtained linear shrinkage results superior to Saint Felix clay in the dilatometry technique. In the case of cullet particle incorporations with an average diameter of 450µm, the best incorporation results were 30%, followed by 20, 10 and 40%. It is noted that in this case there was a strip in which the cullet could be used.

The best result was the incorporation of cullet in 40% by mass, with an average diameter of 37.5µm, being evident by the anticipation of the temperature in 125 ° C in linear shrinkage compared to pure clay, which shows the economy of energy that can be achieved.

The use of smaller particles of cullet in sintered resulted in better water absorption results, but it accentuated the expansive phenomena that cullet generates in sintered by cavity formation by gas trapping at 1150 ° C.

Within an ideal range of manufacturing parameters (temperature and incorporation content), cullet provides the possibility to obtain sintered mechanical strength, linear shrinkage, water absorption and density results, much higher than those of pure clay sintered.

The best result of water absorption was from the aggregates that used 40% of 37.5µm mass cullet, sintered at 1150 ° C, reaching almost zero absorption. However, they had the worst mechanical performance, the highest expansion, the lowest density and the ones that showed the largest volume of cavities due to gas trapping.

The best mechanical result was the aggregates containing 30% 37.5µm mass cullet, sintered at 1100 ° C, twice the strength of aggregates with the same cullet content, and burned at the same temperature as using particulate matter. 450 µm cullet.

Cullet particle size is a parameter that interferes with the efficiency of cullet incorporation in clay, as it ensures better results in the sintering process at lower temperatures, due to the higher reactivity of the smaller cullet particles, and the greater homogenization of the cullet distribution, same in clay.

REFERENCES

[1]. Islam, Md Sagirul, et al. "Effect of soda lime silica glass waste on the basic properties of clay aggregate." Int. J. Sci. Eng. Res 7.4 (2016): 149-154.
[2]. Njindam, O. R., et al. "Effect of glass powder on the technological properties and microstructure of clay mixture for porcelain stoneware tiles manufacture." Construction and Building Materials 170 (2018): 512-519.
[3]. Abuh, M. A., et al. "Implications and characterization of waste glass cullet–kaolinite clay ceramics." Journal of Applied Sciences and Environmental Management 23.3 (2019): 513-518.
[4]. Canakci, Hanifi, A. L. Aram, and Fatih Celik. "Stabilization of clay with waste soda lime glass powder." Procedia engineering 161 (2016): 600-605.
[5]. Ji, Ru, et al. "Utilization of mineral wool waste and waste glass for synthesis of foam glass at low temperature." Construction and Building Materials 215 (2019): 623-632.
[6]. Oliveira, Herbet Alves de, et al. "Produção de agregado sintético de argila com reaproveitamento de resíduo de vidro." Matéria (Rio de Janeiro) 24.1 (2019).

[7]. Mustafi, Sonjida, et al. "Effect of waste glass powder on physico-mechanical properties of ceramic tiles." Bangladesh Journal of Scientific Research 24.2 (2011): 169-180.
[8]. Mahmoudi, Salah, et al. "Characterization, firing behavior and ceramic application of clays from the Gabes region in South Tunisia." Applied Clay Science 135 (2017): 215-225.
[9]. Boussen, Slim, et al. "Characteristics and industrial application of the Lower Cretaceous clay deposits (Bouhedma Formation), Southeast Tunisia: Potential use for the manufacturing of ceramic tiles and bricks." Applied Clay Science 123 (2016): 210-221.
[10]. Soares, R. A. L., R. J. S. Castro, and R. M. Nascimento. "Estudo da potencialidade da aplicação de uma argila contaminada com calcário na produção de placas cerâmicas." Cerâmica 58 (2012): 475-480.
[11]. Santos, C. P., et al. "Caracterização de argilas calcárias utilizadas na produção de revestimentos cerâmicos no Estado de Sergipe-Brasil." (2016).
[12]. BROWN, Michael Ewart. Introduction to thermal analysis: techniques and applications. Springer Science & Business Media, 2001.
[13]. Chinelatto, A. L., and DPF de SOUZA. "Porcelanas elétricas aluminosas: Parte II–Desenvolvimento da microestrutura e sua influência no módulo de ruptura." Cerâmica 50.315 (2004): 172-184.

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