Densification behavior of floor tiles added with sugarcane bagasse ash waste

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Abstract

In this work, the effect of sugarcane bagasse ash waste on the densification behavior of vitrified floor tiles was investigated. Four tile formulations containing up to 5 wt. % of sugarcane bagasse ash waste as a replacement of quartz were prepared. The floor tile manufacturing route consisted of the following steps: powder preparation by the dry process, uniaxial pressing, and firing at temperatures between 1190 ºC and 1250 ºC using a fast-firing cycle. The densification was measured by three parameters: linear shrinkage, water absorption, and flexural strength. The microstructure was evaluated by XRD and SEM. The experimental results indicated that the densification behavior of floor tile formulations was influenced by both the amount of sugarcane bagasse ash waste and the maximum firing temperature. Microstructural variation occurred during firing. However, the use of sugarcane bagasse ash waste had little effect on phase evolution during the fast-firing cycle. An optimum amount of sugarcane bagasse ash waste (up to 2.5 wt. %) for the replacement of quartz allowed for the highest quality production of floor tile materials.
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Keywords: sugarcane bagasse ash; waste; floor tile; densification; microstructure.

1. Introduction

Industrial activities generate huge amounts of a variety of solid wastes, which cause considerable environmental and economic problems. For this reason, industrial solid waste management in an ecological and economical way has become a matter of high global interest. Currently, the strategies of solid waste management are focused mainly on the reuse instead of elimination or storage of waste. The ceramic industry for building materials is therefore a well-established field for reuse of solid wastes [1]. This approach has environmental and economic advantages because the solid waste is incorporated into ceramic formulations in place of non-renewable natural raw materials.

The sugarcane industry is based on the production of sugar and ethanol, which produces huge amount of sugarcane bagasse ash (SCBA) waste worldwide. SCBA waste is considered a non-biodegradable solid waste material, which has become a negative factor for the sugarcane industry and ecologists. The SCBA waste is mainly disposed of as soil fertilizer [2], but this negatively impacts the environment. SCBA waste has quartz (SiO_2) as a major component, and minor amounts of Al_2O_3, Fe_2O_3, K_2O e CaO [3-5]. Thus, SCBA waste has a high potential to be used in ceramic building materials as replacement of virgin raw materials.

Floor tiles are vitrified ceramic materials with excellent physical, mechanical and tribological properties. These tile materials are primarily formulated from a triaxial mixture with natural raw materials such as kaolin, plastic clay, feldspars, quartz, and feldsphatic sands [6]. On firing, floor tile formulations undergo a series of physical and chemical reactions at different temperature ranges, resulting in a heterogeneous microstructure composed of high-temperature silicate phases embedded in an abundant glassy phase. The tile

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industry in many countries is facing scarcity of viable raw materials in locations close to the plants. This means that there are significant opportunities for research in this field, mainly using solid wastes as a low-cost alternative for raw materials. The literature [7-12] review shows that the use of different solid wastes in the manufacturing of floor tiles has been extensively investigated. However, very little research has been focused on the effects of the SCBA waste addition on the processing properties and sintered microstructure of floor tile formulations [13, 14]. In particular, the densification behavior of floor tile formulation added with SCBA waste is still to be investigated.

The aim of this paper is to report and discuss the densification behavior of floor tiles incorporated with SCBA waste. Special emphasis is given to the tile formulation characteristics, their effects on the densification behavior and microstructural evolution of the fired specimens.

2. Experimental Procedure

Four floor tile compositions were formulated (Table 1) using triaxial mixtures of kaolin, albite, and quartz + SCBA waste. In this study the quartz was replaced with increasing amounts of SCBA waste. The SCBA waste sample was collected from a sugarcane plant located in south-eastern Brazil. The collection of the representative SCBA sample (10 kg) was done during boiler cleaning step of the sugarcane plant. The chemical composition of the SCBA waste has been published elsewhere [4]. Commercial kaolin, albite, and quartz used were supplied by the Armil Mineração do Nordeste Ltda, whose chemical compositions have been published elsewhere [15].

Table 1. Vitrified floor tile formulations (wt.%).

Formulation	Kaolin	Albite	Quartz	SCBA waste
MS0	40.00	47.50	12.50	0.00
MS1	40.00	47.50	11.25	1.25
MS2	40.00	47.50	10.00	2.50
MS3	40.00	47.50	7.50	5.00

The reference floor tile formulation (MS0) consisted of 40.0 wt.% kaolin, 47.5 wt.% albite, and 12.5 wt.% quartz. Table 2 gives the chemical compositions of the floor tile formulations. The raw materials were dried at 110 ºC, dry ground separately in a laboratory grinder, and then passed through a 325 mesh (45 µm ASTM) sieve. The tile formulations (Table 1) were mixed,

homogenized, and granulated by the dry process. The granulated tile powder was sent to the sieve to eliminate agglomerates coarser than 2 mm.

Table 2. Chemical compositions of the tile formulations (wt.%).

Compounds	Formulations			
	MS0	MS1	MS2	MS3
SiO_2	65.03	64.57	64.11	63.17
Al_2O_3	22.38	22.46	22.53	22.67
Fe_2O_3	0.16	0.25	0.35	0.53
TiO_2	0.02	0.03	0.05	0.08
Na_2O	4.80	4.79	4.78	4.77
K_2O	1.51	1.58	1.66	1.81
CaO	0.24	0.31	0.37	0.49
MgO	0.07	0.08	0.09	0.13
MnO	-	-	0.01	0.01
P_2O_5	-	0.01	0.02	0.05
SO_3	-	0.01	0.01	0.02
LoI[+]	5.79	5.91	6.04	6.27

[+] LoI – loss on ignition

XRD analysis of the tile formulations was performed in a conventional diffractometer (Shimadzu, XRD-7000) using Cu-Kα radiation (λ = 0.154 nm) at a scanning speed of 1.5º (2θ)/min. JCPDS-ICDD cards were used to identify the mineral phases. The grain-size distribution of the tile powders was determined by sieving. The plastic index was determined by the Atterberg method according to NBR 6459 and NBR 7180 standardized procedures. The Hausner ratio was determined as the ratio of tap density to apparent density of the granulated tile powder.

The tile formulations prepared by the dry process were tested at a laboratory scale, simulating the industrial floor tile-making process. The tile powders were moistened (7 wt. % water), uniaxially pressed at 50 MPa into test bars (11.50 cm x 2.54 cm), and then dried in an oven at 110 ºC. Green tile pieces (five test specimens for each composition) were fast-fired between 1190 ºC and 1250 ºC in a laboratory kiln (Fortlab, FQR-1300/3) with a heating rate of 70 ºC/min from room temperature up to 500 ºC, 25 ºC/min between 500 and 600 ºC, and 50 ºC/min up to the maximum firing temperature with 6 min of soaking time. The cooling rate was 120 ºC/min for maximum firing temperature up to 600 ºC, and 25 ºC/min for 600 ºC up to room temperature. The entire fast-firing cycle

lasted less than 60 min, which is typical for floor tile industrial production.

Linear shrinkage was determined by measuring variation in the length of rectangular specimens upon drying and firing according to ASTM C326. Water absorption was determined from weight differences between the as-fired and water saturated tile samples according to ASTM C373. The flexural strength was determined by a three-point bending test at a loading rate of 0.5 mm/min according to ASTM C674, by using a universal mechanical testing machine (Instron, model 5588). The microstructural characterization of fracture surfaces was carried out by scanning electron microscopy (Shimadzu, SSX-550) via secondary electron images, at 15 kV, after gold-coating surfaces. The crystalline phases after fast-firing cycle were identified via XRD analysis.

3. Results and Discussion

3.1. Characteristics of the tile formulations

Fig. 1 shows the typical XRD pattern of the floor tile formulation containing SCBA waste (MS2 formulation). As expected, the MS2 formulation presented as main mineral phases kaolinite ($Al_2O_3.2SiO_2.2H_2O$), albite ($NaAlSi_3O_8$), and quartz (SiO_2). Potassium carbonate (K_2CO_3) and hematite (Fe_2O_3) were also detected as accessory mineral. This is in accordance with the chemical compositions (Table 2). Thus, the incorporation of SCBA waste into a floor tile body modified its mineralogical and chemical compositions.

Fig. 2 shows the grain-size distribution of the granulated tile powders. It was clear that the replacement of quartz with up to 5 wt. % of SCBA waste did not significantly alter the grain-size distribution of the granulated powders. Note that, for all tile powders produced with the dry process, the majority of the microgranulated particles were concentrated in the intermediate size range (150 μm). This size range was typical of a dry process [16], which allows obtaining a high degree of reactivity of the starting components of tile formulations during the fast-firing cycle. All floor tile powders, as shown in Table 3, exhibited a Hausner ratio (Hr = 1.016 – 1.024) around unity, which was in accordance with grain-size distribution data. This means that the microgranulated powders had the required flowability characteristics independently of the added SCBA waste amount. This property is very important from an industrial point of view, because industrial applications involve powder

handling and automatic die filling during floor tile compaction operation. Table 3 also gives the Atterberg consistency limits. UPL and LPL are respectively the upper and lower plastic limits and PI is the plastic index, given by the difference between UPL and LPL. It can be seen that the PI values (12.4 – 13.7 %) presented only slight differences with SCBA waste addition. These values were within the adequate range for floor tile production.

Table3. Atterberg consistency limits and Hausner ratio (Hr) of the tile powders.

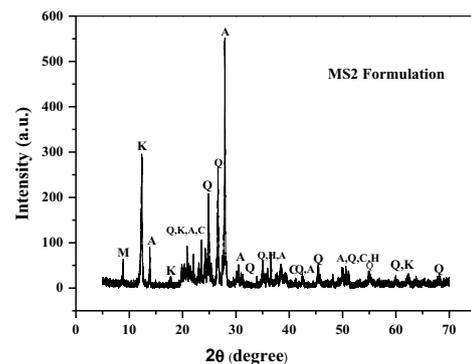| Formulations	UPL, %	LPL, %	PI, %	Hr
MS0	32.3	18.1	18.2	1.024
MS1	30.1	17.1	12.4	1.019
MS2	29.3	15.6	13.7	1.016
MS3	30.5	16.9	12.6	1.023

Fig. 1. XRD pattern for the MS2 formulation. K – kaolinite; M – mica; Q – quartz; A – albite; H – hematite; C – potassium carbonate.
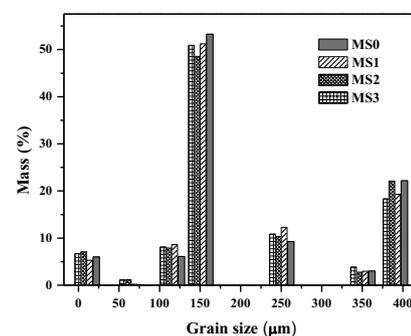

Fig. 2. Grain-size distribution of the granulated floor tile powders.

3.2. Phase analysis and sintered microstructure

XRD patterns of the MS0 formulation fired between 1190 ºC and 1250 ºC are presented in Fig. 3. For all firing temperatures, the main phases detected were

primary mullite ($3Al_2O_3.2SiO_2$), quartz (SiO_2) and the glassy phase. This was in accordance with the Na_2O-Al_2O_3-SiO_2 phase diagram [17]. Albite has been found in small amount as a remaining mineral unreacted during the fast-firing cycle, but disappeared at temperatures above 1210 ºC. Quartz was a residual mineral from the original raw materials. Mullite formed during the fast-firing cycle as a result of the decomposition of kaolinite.
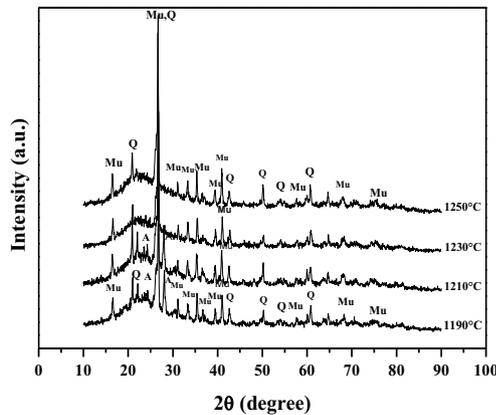

Fig. 3. XRD patterns of the MS0 formulation fired at various temperatures. Mu – mullite; Q – quartz; A – albite.

The mechanism of mullite formation is well documented in the literature [18]. The viscous liquid phase that mainly originated from the albite is cooled to a glassy phase, and was characterized by an amorphous hump between $2\theta = 10^o$ and 35°. The amorphous silica originating from the decomposition of metakaolinite was highly reactive, and can also contribute to liquid phase formation. XRD patterns of the sample containing SCBA waste (MS3 formulation) are shown in Fig. 4.
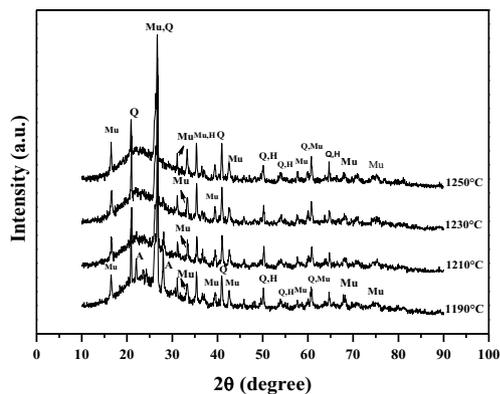

Fig. 4. XRD patterns of the MS3 formulation fired at various temperatures. Mu – mullite; Q – quartz; A – albite; H – hematite.

In addition to mullite and quartz, there was also evidence of the presence of hematite as a residual mineral of the SCBA waste. The potassium carbonate disappeared because this compound melts at 891 ºC [19]. Thus, the replacement of quartz with SCBA waste had little influence on the phase evolution of the traditional floor tile formulation. In fact, as shown in Fig. 5, all tile formulations set forth in this study were located within the primary mullite phase field.
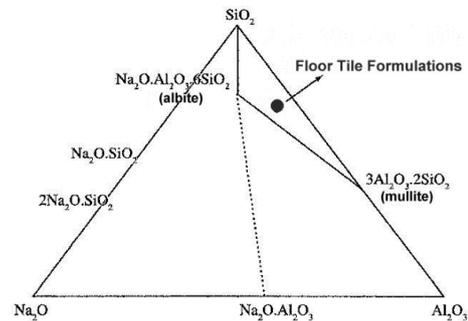

Fig. 5. SiO_2-Al_2O_3-Na_2O phase diagram including the floor tile formulations.

Figures 6-8 show the fracture surface of floor tile samples subjected to mechanical testing fired at various temperatures. SEM micrographs show the typical microstructural evolution of the floor tile formulations. The sintered microstructure for all tile formulations was characterized by high green compact packing. According to XRD (Figs. 3 and 4), the microstructures consisted mainly of quartz and primary mullite crystals embedded in the glassy matrix, independently of the added SCBA waste amount. Fig. 6 shows the microstructural evolution of the MS0 formulation. At 1190 ºC (Fig. 6a), the overall microstructure is marked mainly by rough zones (interconnected pores of different sizes with irregular morphology) connected with dense zones (glassy phase mainly originating from albite). This was typical of the incomplete densification process in floor tile materials. At 1230 ºC (Fig. 6b) and 1250 ºC (Fig. 6c), the open porosity was substantially reduced, in which the floor tile samples were in an advanced sintering stage. The presence of few rounded isolated pores indicated the consistent development of the liquid phase during the fast-firing cycle. The microstructure was highly dense, and transgranular fractures could be observed. The microstructural evolution of MS2 formulation, as shown in Fig. 7, is very similar to that of MS0 formulation. In addition, undissolved quartz particles can be identified by the presence of peripheral cracks (Fig. 7c) around the

particles. According to the literature [18, 20] these cracks are related to microscopic residual stresses, and are associated with the difference between the thermal expansion coefficients of quartz particles and the glassy matrix. Fig. 8 shows that the microstructural evolution of MS3 formulation (with 5 wt.% SCBA waste) differs from that of other formulations. A substantial increase in porosity can be observed. This finding is associated with the gas evolution within the structure, which prevents the pore closure affecting the tile microstructure. Note that, at 1250 ºC (Fig. 8c), a considerable increase in the closed porosity occurred.
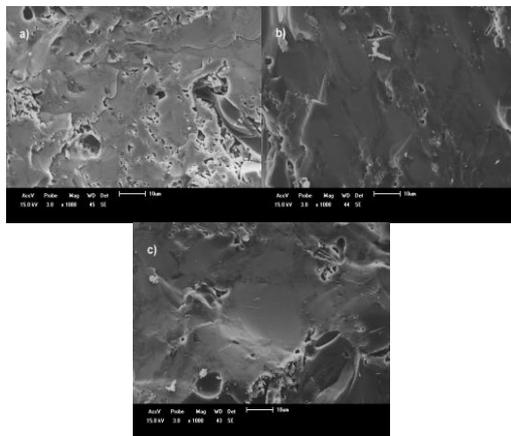

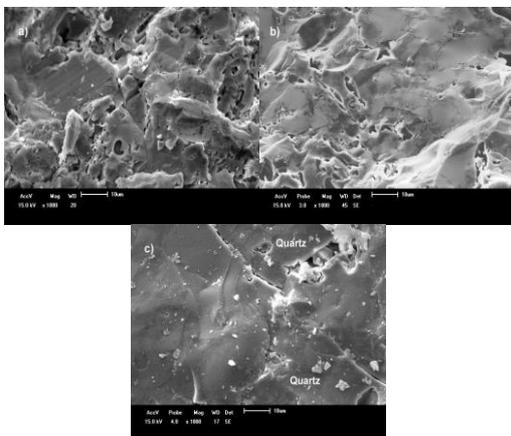Fig. 6. SEM images of MS0 formulation: (a) 1190 ºC; (b) 1230 ºC; and (c) 1250 ºC.

Fig. 7. SEM images of MS2 formulation: (a) 1190 ºC; (b) 1230 ºC; and (c) 1250 ºC.

This detrimental effect could be manly explained by the decomposition of organic compounds (charcoal and organic matter) in the SCBA waste during the fast-firing cycle.
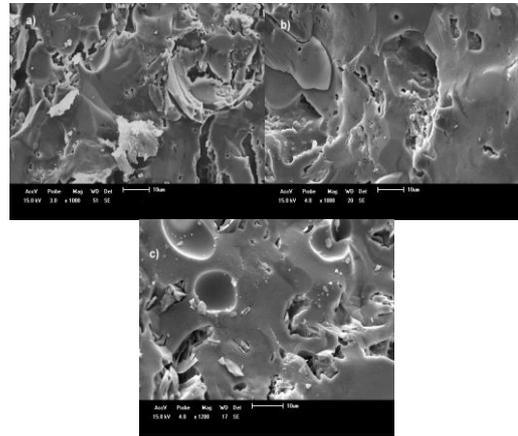

Fig. 8. SEM images of MS3 formulation: (a) 1190 ºC; (b) 1230 ºC; and (c) 1250 ºC.

3.3 Densification behavior

Fig. 9 shows the gresification diagrams of the floor tile formulations. This diagram establishes the optimum vitrification range of floor tile materials during the fast-firing cycle. It is clear that densification behavior of the tile formulations depends substantially on both SCBA waste amount and maximum firing temperature. As expected, the maximum firing temperature raised the linear shrinkage simultaneously with decreases of water absorption (open porosity) of the specimens by means of densification, independently of the amount of added SCBA waste. Hence, densification could be attributed to a viscous flow sintering mechanism closing the open porosity [21]. However, at any given temperature, the linear shrinkage values indicated that the formulations containing SCBA waste shrunk faster than the traditional tile formulation. This behavior was essentially due to the chemical composition of the SCBA waste [4], which contained appreciable amounts of fluxing oxides ($K_2O + CaO + Fe_2O_3$). These oxides tended to increase the liquid phase amount with lower viscosity and accelerate the densification process. The results of Fig. 9 also show that the densification behavior of MS0, MS1, and MS2 formulations presented only slight differences. It is important to note that between 1230 ºC and 1250 ºC, a region of dimensional stability could be observed. In this region, the maximum linear shrinkage occurred at the same time as the minimum water absorption, indicating the end of the sintering process. It is worth mentioning that this vitrification region was the most suitable for production of high quality floor tiles. In fact, these formulations reached water absorption below 0.5 % specified by the ISO 13006 [22] for porcelain

stoneware tiles (BIa group). Fig. 9d shows the gresification diagram of the MS3 formulation (with 5 wt.% SCBA waste). It can be seen that the maximum linear shrinkage (between 1230 ºC and 1250 ºC) did not correspond to the minimum water absorption (between 1210 ºC and 1230 ºC), with no region of dimensional stability. Between 1230 ºC and 1250 ºC, the MS3 formulation also presented a high value of water absorption.
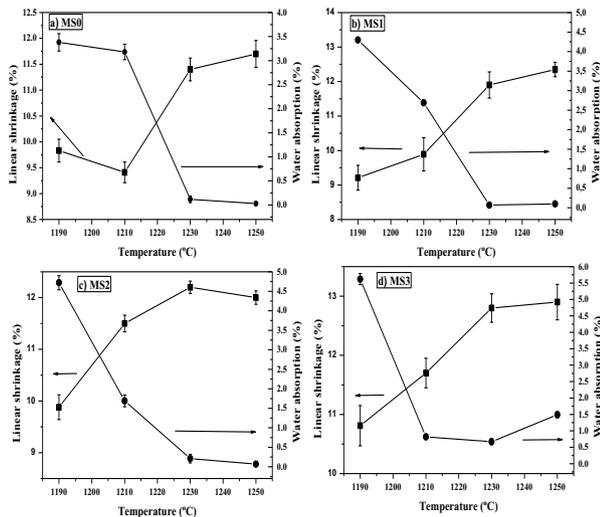

Fig. 9. Gresification diagram of the floor tile formulations: a) MS0; b) MS1; c) MS2; and d) MS3.

This was in accordance with the microstructure (Fig. 8). In fact, the MS3 formulation fired above 1210 ºC presented the coarserming effect the closed pores with a concomitant volume expansion referred to as body bloating. This is mainly related to the decomposition of organic compounds. Black core defects [23] have also been observed. This suggests that only T ≤ 1210 ºC can be used for MS3 formulation resulting in lower quality floor tiles.Fig. 10 shows the flexural strength of the fired tile pieces. The mechanical strength was closely correlated to all the other ceramic properties investigated. It can be seen that the flexural strength of the MS0, MS1, and MS2 formulations increased gradually with the increasing maximum firing temperature. This behavior was related to a high densification during the fast-firing cycle, resulting in a stronger glassy bond associated with a compact microstructure. Additionally, the open porosity was reduced significantly, as shown in Fig. 9. In particular, the MS0, MS1, and MS2 formulations fired between 1230 ºC and 1250 ºC presented high values of flexural strength (40.23 – 52.80 MPa), which are in conformity to porcelain stoneware tiles (> 35 MPa) [22].
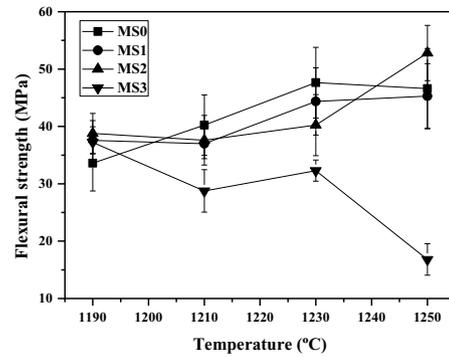

Fig. 10. Flexural strength of the floor tile formulations.

For the MS3 formulation, however, the tile pieces had lower mechanical strength than the other formulations for firing temperatures above 1190 ºC. Indeed, the decrease in mechanical strength is in line with the increase of both open and closed porosities (Fig. 8c). In particular, the closed pores act as microcracks contributing to a significant reduction in the flexural strength of the MS3 sample. This suggests that the replacement of quartz with SCBA waste in floor tile materials was limited. Thus, additions of SCBA waste above 2.5 wt. % should be avoided, because it impairs the mechanical strength of the fired floor tile formulations.

4. Conclusions

In this study, the effect of SCBA waste on the densification behavior of floor tile formulations was investigated. It was found that the replacement of quartz with up to 5 wt.% SCBA waste presented only a slight variation in granulated powder characteristics. In addition, all tile formulations prepared by the dry process presented good flowability, and favorable grain-size distribution to obtain high densification during the fast-firing cycle.

XRD analysis showed that all floor tiles produced were formed primarily by three components: quartz, mullite, and glass. SEM observations indicated that the incorporation of SCBA waste above 2.5 % wt.% should be avoided to prevent significant changes in the sintered microstructure of floor tile materials. It was also verified that the optimum vitrification range (higher densification) for floor tile formulations containing up to 2.5 wt.% is achieved between 1230 and 1250 ºC.

Aside from the environmental and economic benefits of reducing SCBA waste, the replacement of up to 2.5 wt. % of natural quartz with SCBA waste allows for the production of floor tiles fired between 1230 ºC and

1250 ºC with technological properties similar to those presented by commercial porcelain stoneware tiles.

Acknowledgements

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