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Processing of porcelain stoneware tile using sugarcane bagasse ash waste

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Abstract

Large amounts of waste materials are discarded in the sugarcane industry. This work investigates the reuse of sugarcane bagasse ash waste as an alternative raw material for porcelain stoneware tile bodies, replacing natural quartz by up to 5 wt.%. The tile pieces were fired at 1230 °C using a fast-firing cycle (< 60 min). The technological properties of the fired tile pieces (e.g., linear shrinkage, water absorption, apparent density, and flexural strength) were determined. The sintering process was followed by SEM and XRD analyses. The results show that up to 2.5 wt.% sugarcane bagasse ash waste can be used as a partial replacement for quartz in porcelain stoneware tile (group BIa, ISO 13006 standard), providing excellent technical properties. Hence, its application in high-quality ceramic tile for use in civil construction as a low-cost, alternative raw material could be an ideal means of managing sugarcane bagasse ash waste.

Keywords: *traditional ceramics, sugarcane bagasse ash, firing, strength, X-ray diffraction*

I. Introduction

Sugarcane is grown in over 110 countries, with Brazil being the world's largest producer. In 2013–2014, it is estimated that Brazil's sugarcane production will reach 654 million tonnes [1]. In the modern sugarcane industry, however, the production of huge amounts of waste materials, including the solid waste material known as sugarcane bagasse ash (SCBA), is inevitable. Brazil alone produces approximately 4.6 million tonnes of SCBA waste per year, and the levels of such waste are expected to increase continuously. At present, SCBA waste is ultimately disposed of as soil fertiliser in many sugarcane-producing countries [2,3]. However, this option has three disadvantages: i) it causes significant changes in the physico-chemical properties of soils, ii) it contributes to environmental pollution, and iii) it may have a strong negative impact on human health [4]. Thus, SCBA waste production presents a serious waste management problem for the sugarcane industry.

The reuse of SCBA waste by converting it into green material as an alternative raw material to produce building materials is increasingly encouraged by researchers. In fact, several papers have shown promising results for the reuse of SCBA waste in the production of clay bricks, soil blocks, cement, concrete, and mortars [5–12]. The use of SCBA waste for obtaining floor tile has also been recently suggested [13]. However, the reuse of SCBA waste in the manufacturing of porcelain stoneware tile has not yet been investigated. Porcelain stoneware tile is a high-performance ceramic tile and the world tile material market leader, having the most important standard requirements such as low water absorption, high mechanical strength, and good tribological properties [14,15]. However, porcelain stoneware tile formulations contain essentially non-renewable raw materials, such as kaolin, kaolinitic clays, feldspars, quartz, quartz-feldspathic sands, talc, dolomite, and calcite [16,17].

The aim of this study is to investigate the possibility of introducing SCBA waste, a renewable raw material, into porcelain stoneware tile formulation for use in civil construction.

II. Experimental procedure

A typical porcelain stoneware tile formulation and SCBA waste in the form of a fine powder were selected as raw materials. The porcelain stoneware tile formulation used as reference contains 40 wt.% kaolin,

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Table 1. The proportions of the blends for the tile formulations (wt.%)

Formulation	Kaolin	Na-feldspar	Quartz	SCBA waste
MA0	40	47.5	12.5	0
MA1	40	47.5	11.25	1.25
MA2	40	47.5	10	2.5
MA5	40	47.5	7.5	5

47.5 wt.% Na-feldspar, and 12.5 wt.% quartz [18]. The SCBA waste sample was collected from a sugarcane mill located in the southeastern region of Brazil (Campos dos Goytacazes-RJ). The mineralogical analysis of SCBA waste has been published elsewhere [8]. Selected mixtures containing up to 5 wt.% SCBA waste were prepared (Table 1). In this study, quartz was partially replaced by various amounts of SCBA waste. Table 2 gives the chemical composition of the raw materials.

The raw materials were dry milled and mixed using a laboratory grinder, and then passed through a 325-mesh (45-µm ASTM) sieve. The porcelain stoneware tile formulations (Table 1) were mixed and homogenised for 60 min. The tile powders were granulated via dry process.

Mineralogical analysis was performed via X-ray diffraction (XRD) using a Shimadzu XRD 7000 instrument with monochromatic Cu-Kα radiation (λ = 0.154056 nm) and a Ni filter. The operating conditions were 40 kV, 40 mA, and a scanning speed of 1.5°/min. Crystalline phases were identified by comparing the intensities and positions of the Bragg peaks to those listed in the JCPDS data files. The grain-size distribution of the tile powder was determined by sieving according to NBR 7181.

Each granulated tile powder was used to form prism-shaped test specimens (11.50 × 2.54 cm^2) by uniaxial pressing at 50 MPa and a moisture content (moisture mass/dry mass) of 7%. The test specimens were dried in an oven at 110 °C. The green tile specimens were fast-fired at 1230 °C in air using a fast-firing laboratory kiln for a total of less than 60 min (cold-to-cold). The fast-firing cycle used in this study was chosen to simulate an actual firing process used in the ceramic tile industry.

The linear shrinkage values upon drying and firing were evaluated from the length variation of the rectangular specimens. Water absorption values were determined from the weight differences between the as-fired and water-saturated samples (immersed in boiling water for 2 h). The apparent density was determined by the Archimedes method. The flexural strength was determined by the three-point bending test using a universal mechanical testing machine (model 1125, Instron) at a loading rate of 0.5 mm/min. The microstructure characterisation of the gold-coated fracture surfaces of fired tile specimens was conducted by scanning electron microscopy (SEM SSX-550; Shimadzu) at 15 kV. The mineral phases after firing were identified via X-ray diffraction analysis.

III. Results and discussion

The XRD pattern of the MA0 formulation (SCBA waste-free formulation) is shown in Fig. 1. As expected, the main crystalline phases identified were kaolinite ($Al_2O_3{\cdot}2\,SiO_2{\cdot}2\,H_2O$), albite ($NaAlSi_3O_8$), quartz ($SiO_2$), and mica muscovite ($KAl_2Si_3AlO_{10}(OH)_2$). In the formulation bearing SCBA waste (MA5 formulation), characteristic peaks of potassium carbonate (K_2CO_3) and hematite (Fe_2O_3) were also identified, as shown in Fig. 2. The identified mineral phases are consistent with the chemical composition data (Table 2). Thus, the incorporation of SCBA waste into a porcelain stoneware tile body modifies its mineralogical composition.

Table 3 gives the grain-size distribution of the granulated powder (MA2 formulation) produced by the dry process. The largest fraction of the granulated tile powder is concentrated in the grain size range of 150 µm, which provides a high degree of sintering during the fast-firing process [19]. In addition, the granulometric behaviour of the tile powder is suitable for ob-

Table 2. Chemical compositions (wt.%) of the raw materials

Oxides	Kaolin	Na-feldspar	Quartz	SCBA waste
SiO_2	49.07	69.55	98.97	61.59
Al_2O_3	33.74	18.61	0.41	5.92
Fe_2O_3	0.22	0.14	0.01	7.36
TiO_2	0.01	0.02	0.02	1.46
Na_2O	0.52	9.63	0.13	-
K_2O	1.97	1.47	0.18	6.22
CaO	0.4	0.17	0.01	5
MgO	0.06	0.09	0.01	1.17
MnO	-	-	-	0.1
P_2O_5	-	-	-	0.98
SO_3	-	-	-	0.42
loss on ignition	14.01	0.32	0.26	9.78

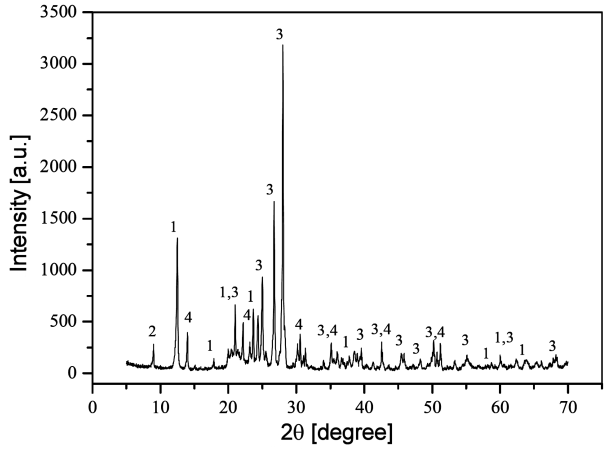


Figure 1. X-ray diffraction pattern of the MA0 formulation (1 – kaolinite; 2 – mica; 3 – quartz; 4 – albite)

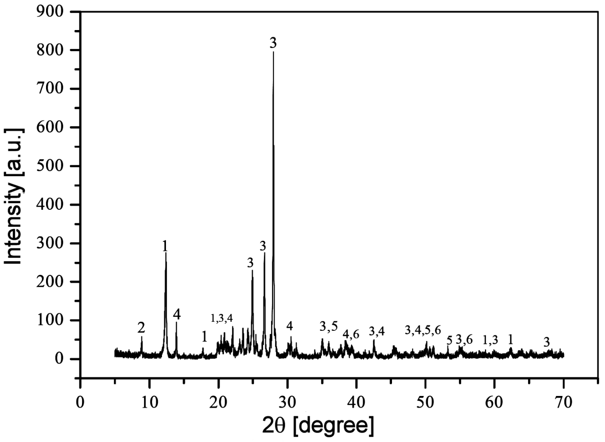


Figure 2. X-ray diffraction pattern of the MA5 formulation (1 – kaolinite; 2 – mica; 3 – quartz; 4 – albite; 5 – hematite; 6 – potassium carbonate)

Table 3. Grain-size distribution of the MA2 formulation

Grain size [μm]	Amount [mas.%]
<75	7.08
75	1.15
105	7.9
150	48.56
250	10.36
350	2.76
355	22.11

taining good flow characteristics and uniformity as the powder fills the mould. This behaviour was observed for all porcelain stoneware tile formulations containing SCBA waste.

The drying process used for the porcelain stoneware tile formulations was monitored through the bulk density and linear shrinkage of the tile pieces dried at 110 °C (Table 4). The results indicated that no significant differences in the drying density (1.91–1.93 g/cm^3) of the green tile pieces were observed regardless of the amount of SCBA waste added. This is very important for comparing the fired properties of the various tile formulations. The results also showed low drying shrinkage of the green tile pieces (0.07–0.12%), which is recommended for the industrial production of stoneware porcelain tiles.

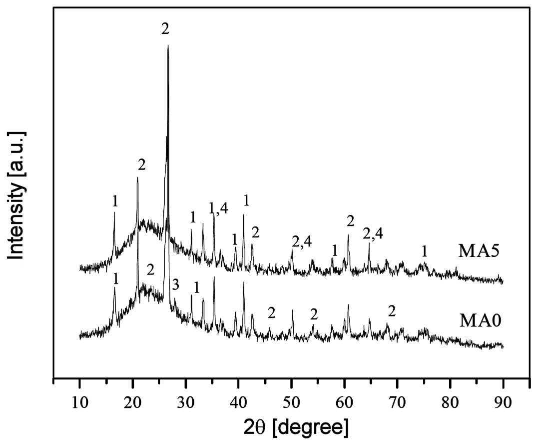


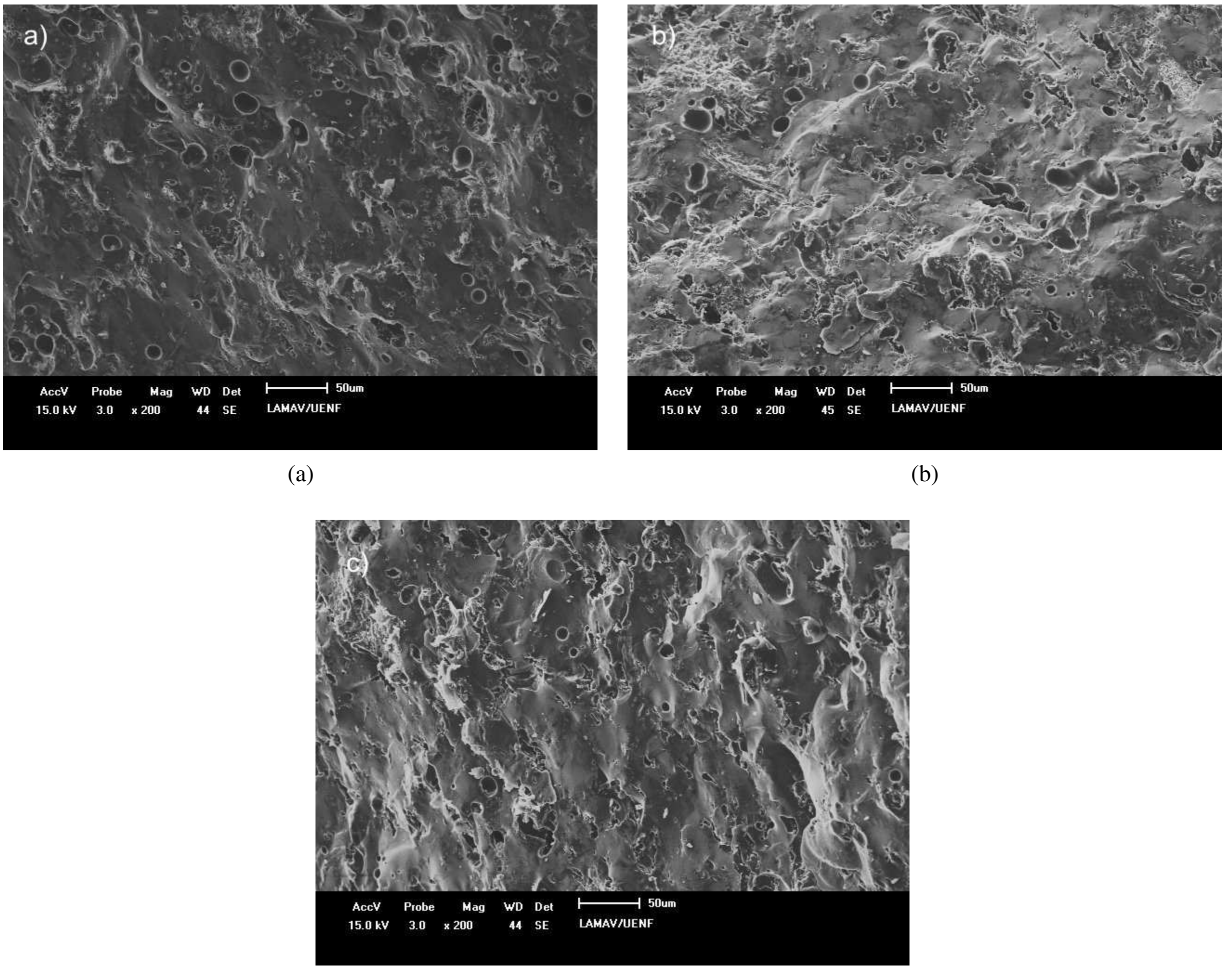
Figure 3. X-ray diffraction patterns of the MA0 and MA5 formulations fired at 1230 °C (1 – mullite; 2 – quartz; 3 – albite; 4 – hematite)

The XRD patterns of the fired tile pieces for the MA0 and MA5 formulations are presented in Fig. 3. The results indicated the presence of mullite ($Al_6Si_2O_{13}$) and quartz (SiO_2) mineral phases for both formulations. In addition, there is evidence of the presence of residual albite unreacted during the fast-firing process used to produce porcelain stoneware tile. Furthermore, the peak intensities changed only slightly. Mullite is formed from metakaolinite by topotactical reaction and can be considered as primary mullite. Quartz is a residual mineral of the original raw materials. The viscous liquid phase is cooled to glass, which is characterised by an amorphous band between 2θ = 10° and 35°. This is typical of porcelain stoneware tile formulations and is in accordance with the Na_2O-Al_2O_3-SiO_2 phase diagram [20]. In the SCBA-containing formulation (MA5 formulation), however, hematite has also been identified. Thus, the partial replacement of quartz with SCBA waste influenced the phase evolution of the porcelain stoneware tile formulations.

Figure 4 shows the fracture surface of the MA0, MA2, and MA5 formulations fired at 1230 °C. The SEM micrographs show the evolution of the densification behaviour of porcelain stoneware tile body with varying SCBA waste contents. For all formulations, the mi-

Table 4. Technological properties of the tile pieces in the dried state at 110 °C

Properties	MA0	MA1	MA2	MA5
Bulk density [g/cm^3]	1.92 ± 0.02	1.91 ± 0.02	1.92 ± 0.03	1.93 ± 0.02
Linear shrinkage [%]	0.11 ± 0.07	0.12 ± 0.08	0.07 ± 0.08	0.07 ± 0.05



(a) (b)

(c)

Figure 4. SEM micrographs of the fracture surfaces of the tile pieces fired at 1230 °C: a) MA0; b) MA2 and c) MA5

crostructure shows a compact matrix containing isolated rounded pores. A transgranular fracture characterised by strong interfaces and glassy bonds between grains is evident. This indicates that the tile formulations reached the final sintering stage with concomitant vitrification. However, Fig. 4c shows that the fracture surface of the formulation MA5 is more porous. This effect suggests the escape of trapped gases in the vitrified matrix of the tile pieces [21] for higher SCBA waste contents. In fact, the increase in porosity could be explained by the high concentration of organic compounds (charcoal and organic matter) in the SCBA waste sample [11].

The linear shrinkage of the fired pieces is shown in Fig. 5. Firing shrinkage values between 11.30–12.80% were found for the tile pieces, indicative of acceptable dimensional control. Note that, at any given percentage of waste, the firing shrinkage of the SCBA waste-added tile pieces is higher than that of the waste-free pieces (MA0 formulation). This behaviour can be explained by the chemical composition of the SCBA waste sample (Table 2), which tends to favour the formation of a more abundant liquid phase with lower viscosity. In fact, the SCBA waste-added formulations tend to be richer in fluxing oxides (K_2O, CaO, and Fe_2O_3).

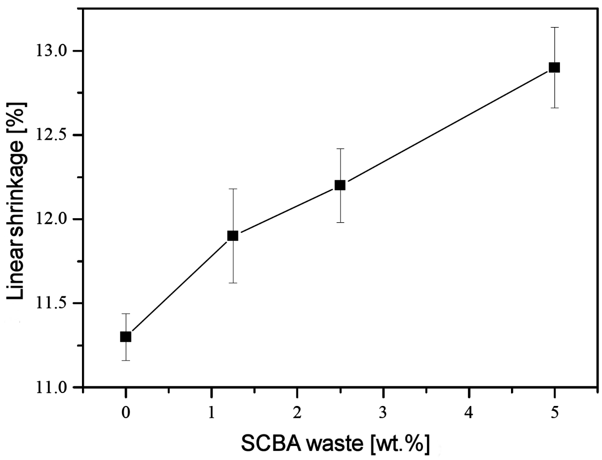


Figure 5. Linear shrinkage of the tile pieces as a function of waste content

The apparent density of the tile pieces is shown in Fig. 6. It was observed that the apparent density (2.02–2.04 g/cm^3) was essentially unaltered upon the addition of the SCBA waste. This behaviour can be explained by the combined inverse effects of sintering and weight loss. The incorporation of greater amounts of

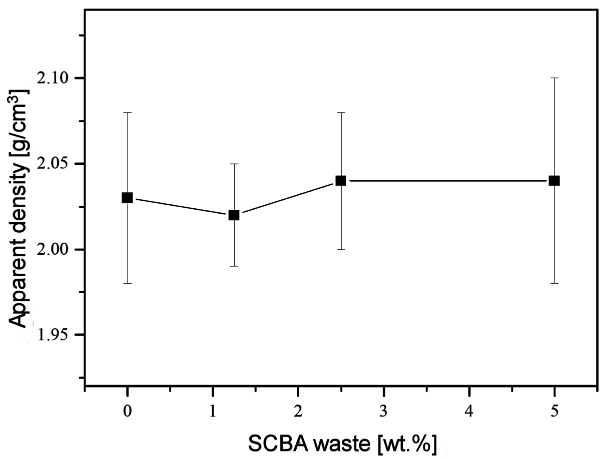


Figure 6. Apparent density of the tile pieces as a function of waste content

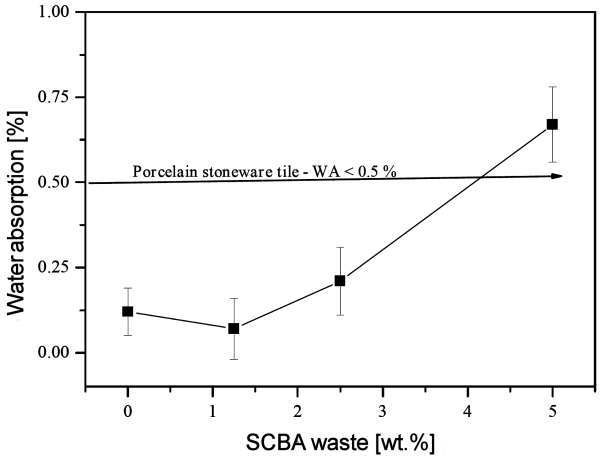


Figure 7. Water absorption of the tile pieces as a function of waste content

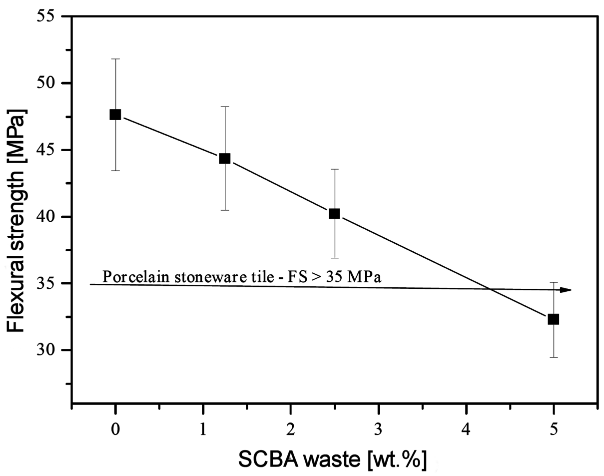


Figure 8. Flexural strength of the tile pieces as a function of waste content

SCBA waste tends to decrease the densification rate of the porcelain stoneware tile pieces during the fast-firing process. This is in accordance with the observation of the microstructure (Fig. 4).

Figure 7 shows the level of open porosity evaluated by the water absorption of the fired pieces. The measured values of water absorption were in the range of 0.07–0.67 %. The water absorption varied only slightly with the addition of SCBA waste, except for the MA5 formulation (with 5 wt.% SCBA waste). This finding is related to the incorporation of organic compounds of SCBA waste into the tile formulations. The decomposition of organic compounds from such waste generates pores in the fired microstructure. This means that there is a limit to the replacement of quartz with SCBA waste in the production of porcelain stoneware tile.

The mechanical strength of the fired tile pieces was determined in terms of flexural strength (Fig. 8). It can be seen that the flexural strength gradually decreased with the addition of SCBA waste. This behaviour may be related to the higher porosity of the tile pieces. This finding was expected because the gas evolution resulting from the decomposition of organic compounds prevents pore closure.

The specified values of water absorption (WA) and flexural strength (FS) for porcelain stoneware tile (group BIa, ISO 13006 standard) are WA $\leq$ 0.5% and FS $\geq$ 35 MPa [22]. As shown in Figs. 7 and 8, this porcelain stoneware tile specification was satisfied for the following formulations: MA0, MA1, and MA2. These results show that the SCBA waste is particularly well suited for porcelain stoneware tile production when used in small amounts (up to 2.5 wt.%) as a partial replacement for natural quartz. Thus, its use in the ceramic tile industry is a potential mean for valorisation of SCBA waste.

IV. Conclusions

The following conclusions may be drawn from the results and discussion presented herein.

- The SCBA waste used in this study is a zero-cost raw material that can replace natural quartz in processing of porcelain stoneware tile formulations.
- The incorporation of SCBA waste influenced the technical properties and sintered microstructure of the tile pieces. However, the replacement of natural quartz with SCBA waste to extents up to 2.5 wt.% allows the production of porcelain stoneware tile with good technical properties.
- The management of SCBA waste by incorporation into porcelain stoneware formulations is rather promising and has the potential to minimise the negative impact of the sugarcane industry on the environment.

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