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Recycling of Granite Waste from Sawing Operation in Clay Brick for Civil Construction

Carlos M. F. Vieira, Bertácia M. da Silva, and Juliana P. R. G. Carvalho

State University of the North Fluminense, Campos dos Goytacazes, RJ, Brazil

Abstract: In This work has as its objective to evaluate the effect of ornamental rock powder waste incorporation in an industrial clayey body used for roofing tiles fabrication. Formulations with 0, 10, 20 and 30 wt.% of waste were prepared by substitution of sand in the industrial clayey body. Specimens were fabricated by 18 MPa uniaxial pressure and then fired from 850 to 1100°C. The specimens were tested to determine the water absorption, linear shrinkage and three points bending flexural strength. The results indicate that the use of 10 wt.% of waste in total substitution of sand increased the mechanical strength of the ceramic fired at 900°C. Incorporation in higher amounts decreased the mechanical strength. On the other hand, the mechanical strength of the industrial clayey body fired at higher temperatures was increased with any amount of the waste incorporated.

Key words: brick, clay, granite waste, recycling

1. Introduction

Ceramic bodies industrially fabricated in Campos (the municipal area of Campos dos Goytacazes, north of the State of Rio de Janeiro, in Brazil) are still empirically elaborated using a mixture of local clays. These clays are predominantly kaolinitic mineral in nature, associated with high plasticity. Due to the excessive plasticity of the ceramic bodies, it is common to observe dimensional defects, which occur after drying and firing. Moreover, the kaolinitic nature and the presence of aluminum hydroxide (gibbsite) confer a refractory behavior to the local clays, which impairs sintering during the firing operation. In the case of clay ceramics for civil construction, this result in greater porosity associated with elevated values of water absorption and low mechanical strength [1, 2]. To avoid these problems, it is necessary to reformulate the ceramic body composition.

The addition of both non-plastic materials to reduce plasticity and fluxes to condition the refractoriness is a possible alternative. One technological method, which is already being tested to decrease porosity, is the incorporation of granite wastes from the sawing process [3-5]. Granite is a rock with large amount of quartz, feldspars and mica. In the initial stages of firing, these minerals act as non-plastic agents which permit the use of lower amount of water in body forming. This makes for an easier drying operation. During firing the quartz generally behaves as an inert material but may also partially dissolve in liquid phases, should they occur. Both the feldspars and mica favor the formation of liquid phases and contribute to lower the porosity of the final ceramic product.

Granite is readily available to the industries in Campos, from the municipal area of Santo Antônio de Pádua State of Rio de Janeiro and 150 km from Campos. Intense industrial activity related to ornamental stones, especially gnaisse, but commercially denoted as granite, is maintained in the region. After mining, the granite flagstone is

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Corresponding author: Carlos M. F. Vieira, Ph.D., Associate Professor, research areas: traditional ceramics, recycling of industrial wastes into heavy clay ceramics and composites. E-mail: vieira@uenf.br.

submitted to sawing to obtain blocks and after a manual operation the final small flagstone. During the sawing operation is generated a sludge composed by water and fine particles of rock. This sludge is commonly referred as granite waste. A monthly production of 1000 tons of granite waste is estimated to be produced at Santo Antônio de Pádua. The final disposal of this waste has brought serious environmental problems. Since most industries do not have an adequate sludge treatment, the granite waste is contaminating the soil and underground waters as well as obstructing rivers and lakes.

Based on the need to give an environmental correct final disposal to the waste together with the necessity to improve the roofing tiles properties of granite waste, the present work had as its main objective to reformulate an industrial heavy clay ceramic body by ornamental rock waste incorporation.

2. Materials and Methods

The basic raw materials used in this investigation were two different local clays from Campos, as well as local sand and granite waste, **GW**, from Santo Antônio de Pádua. The clays are locally called “preta” (Portuguese for black) and “amarela” (Portuguese for yellow). Here they will be referred, respectively, as clays **B** and **Y**. Usually the clays are both mixed with sand to make roofing tiles at small plants, which produce around 50.000 pieces a month. The typical industrial ceramic body made at these plants, here named **MI**, is composed of 70 wt.% of clay **Y**, 20 wt.% of clay **B** and 10 wt.% of sand. Upon receipt, the raw materials were dried at 110°C, manually disintegrated with a crusher and then screened at 20 mesh (0.840 mm).

Three experimental bodies, in wt.%, were formulated replacing sand by **GW**. They are denominated **MA** (70**Y** + 20**B** + 20**GW**), **MB** (65**Y** + 15**B** + 20**GW**) and **MC** (60**Y** + 10**B** + 30**GW**). The plasticity of the ceramic bodies was evaluated by the Atterberg methods [6, 7].

The raw materials were initially characterized in terms of its mineralogical, chemical and physical composition as well as morphological aspects. The qualitative mineralogical phase identification was performed in powder sample by X-ray diffraction (XRD) using a Seifert, model URD 65, diffractometer operating with Cu-K_α radiation and a scanning 2θ angle from 5 to 40°. The chemical composition was determined by X-ray fluorescence (XRF) in a Philips PW 2400 equipment. The particle size distribution was determined by both, sieving and sedimentation methods, following the norm [8]. The morphology of the **GW** powder was evaluated by scanning electron microscopy (SEM) in a Jeol model JSM6460LV equipment with EDS facility.

In order to determine the technological properties of the ceramic bodies, 12.43 cm long rectangular specimens with 1.1 cm × 2.54 cm in cross section, were shaped by uniaxial pressing. Initially the specimens were dried at 110°C for 24 h. Finally, the specimens were fired from 850 to 1100°C in an electric laboratory kiln with a 180 min socket at maximum temperature, using a heating/cooling rate of 2°C/min. The measured technological properties were: dry bulk density, measured by the dimensional method dividing the dry mass by the external volume; drying and firing linear shrinkage; water absorption and mechanical strength, obtained by the flexural rupture strength, using the three points method.

3. Results and Discussion

Fig. 1 shows the X-ray patterns of the raw materials. The patterns indicate that both clays are predominantly kaolinitic with the presence of quartz, muscovite mica and gibbsite. As expected, the sand displays high intensity peaks associated with quartz. It is also observed low intensity peaks associated with feldspars, albite and microcline. In the granite waste, the presence of quartz and feldspars, such as albite, anorthite and microcline is observed. Feldspars are source of alkaline and earth alkaline, materials, such as K_2O, Na_2O and CaO, denoted as flux, which favor the formation of a liquid phase above 700°C [9]. The results from Fig. 1 confirm the tendency of both clays, **B** and **Y**, to have high loss of ignition and refractoriness due to the presence of $Al(OH)_3$ in the form of gibbsite. The fluxing capacity of the granite waste, which is associated with lower porosity after firing, is also confirm by the presence of K_2O, Na_2O and CaO containing feldspar minerals.

Table 1 shows the chemical composition of the raw materials. One can notice, for clay **B**, a relatively low amount of SiO_2 and high amount of Al_2O_3. This indicates and elevated fraction of clay minerals. The black color that gives the name to clay **B** is due to the relatively elevated amount of organic matter in its composition. Clay **Y** displays higher amount of SiO_2 and lower amounts of Al_2O_3 and LoI in comparison with clay **B**, indicating that this clay has lower amount of clay minerals, kaolinite. Moreover, the significant amount of Fe_2O_3 is responsible for the reddish color of the roofing tiles after firing. It should be noted in Table 1 that clays **B** and **Y** have a relatively low content of alkaline oxides (K_2O and Na_2O) and earth alkaline oxides (MgO and CaO), which act as fluxes to improve the sintering mechanisms. Additionally to the high amount of clay minerals, the loss of ignition, **LoI**, is also related to the presence of organic matter and gibbsite.

As far as the sand composition is concerned, only silica is a major constituent. The other oxides are minor components and are probably due to contamination. The main impurity appears to be feldspars, as shown in Fig. 1.

The chemical composition of **GW**, also presented in Table 1, in addition to SiO_2 and Al_2O_3, shows a relatively large presence of alkaline and alkaline earth oxides. This confirms the idea that the granite wastes could produce a fluxing action if added to clay ceramic bodies.

Fig. 2 shows the particle size distribution of the raw materials. One may note that both clays have an

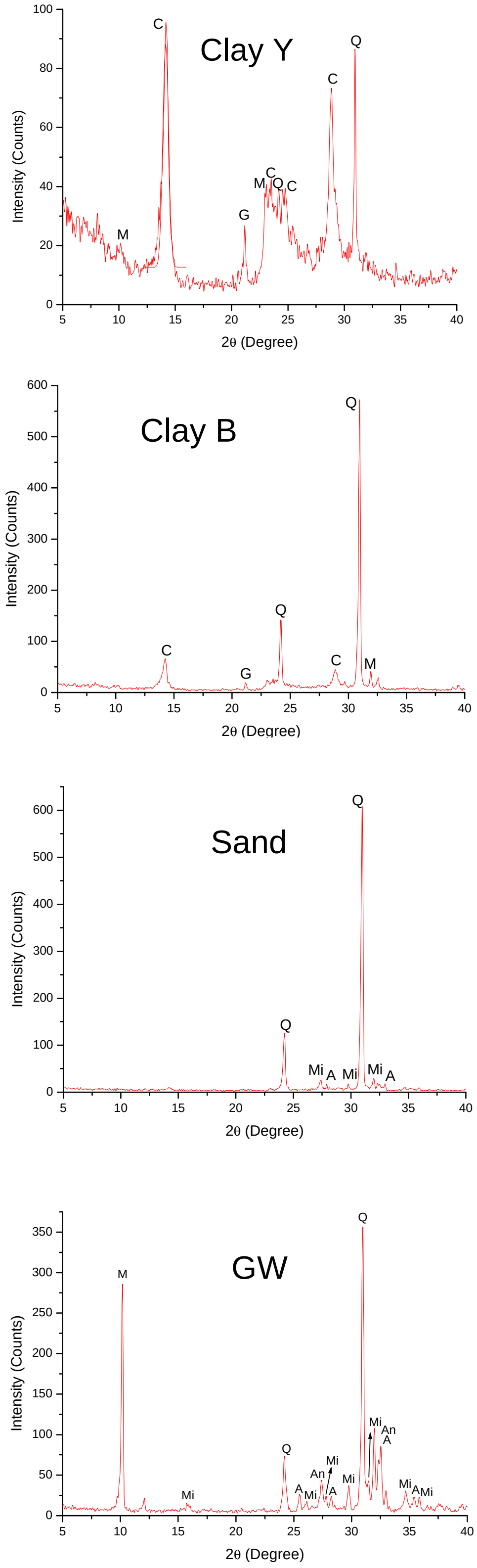

Fig. 1 X-ray diffraction patterns of the raw materials. **A** = albite; **Na** = anorthite; **C** = kaolinite; **G** = gibbsite; **M** = muscovite mica; **Mi** = microcline; **Q** = quartz.

Table 1 Chemical composition of the raw materials (wt.%).

	Clay B	Clay Y	Sand	GW
SiO_2	41.22	69.76	86.66	67.83
Al_2O_3	31.37	18.22	7.53	14.76
Fe_2O_3	1.74	7.61	0,82	4.18
TiO_2	1.16	0.71	0.31	0.71
ZrO_2	-	0.05	0.04	0.09
MnO_2	-	-	-	0.09
K_2O	1.05	1.37	1.90	5.47
Na_2O	-	-	0.67	2.77
CaO	0.15	0.28	0.45	2.08
MgO	0.80	-	-	0.73
P_2O_5	0.21	-	-	0.30
LoI	15.92	7.66	1.34	0.66

elevated amount of clay minerals, considered as that with particle size below 0.002 mm. Clay **P** and clay **Y** present 63% and 44% of clay minerals, respectively. This is important for heavy clay ceramic processing, since the clay minerals development plasticity in mixture with water, being fundamental to the forming stage by extrusion. On the other hand, the sand and the **GW** have only small amount, about 1%, of particles with less than 0.002 mm, that are associated probably with fine particles of mica. Furthermore, both non-plastic sand and **GW** present significant differences. The size of the sand particles is relatively coarse and concentrated in the range from 50 to 500 μm. On the other hand the **GW** has a finer particle size.

Fig. 3 shows SEM micrographs of GW powder. It is observed submicrometric grains, agglomerates and subrounded grains with a compact morphology, indicated with red arrow, probably associated with quartz particles. It is also observed that the size of the particles is in accordance with Fig. 2.

Fig. 4 shows a characteristic map of the extrusion prognostic [10] with the location of the investigated formulations. This figure indicates the so called Atterberg limits for clay bodies based on the combined positions of the plastic limit and the plastic index. The indicated rectangular areas in the figure can be associated with optimum extrusion, acceptable extrusion and non-recommended extrusion. In this figure it is worth worth observing that the industrial body, **MI**, owing to it excessive plasticity, is located in the acceptable extrusion area. It is also observed that the formulation **MA**, with 10 wt.% of **GW** replacing sand, slightly increased the plastic index and decreased the plastic limit in comparison with the **MI**, however without change the area. The **MB**, although still remain in the acceptable extrusion area, it is nearest the optimal extrusion area. Finally, formulation **MC** is located inside the optimal extrusion area, indicating the **GW** acts as a non-plastic material and it is advantageous to the forming stage by extrusion.

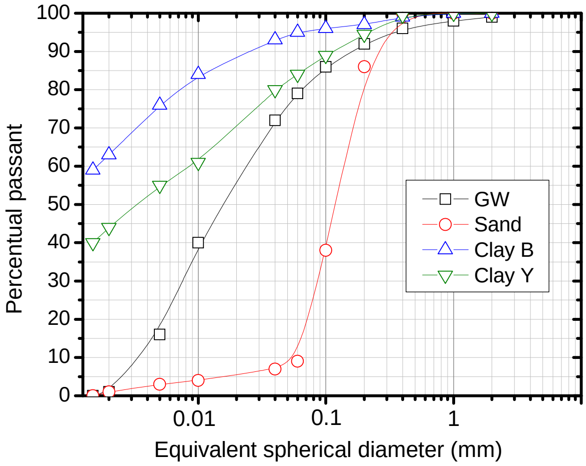

Fig. 2 Particle size distribution curves of the raw materials.

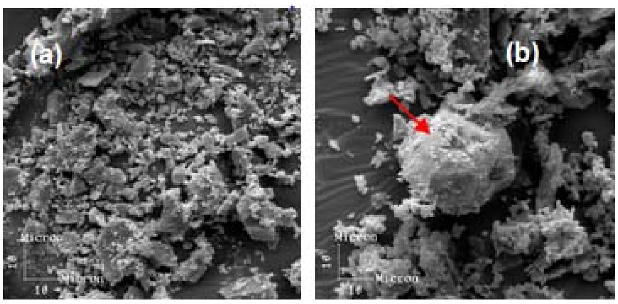

Fig. 3 (a) and (b) SEM micrographs of the GW powder.

Fig. 5 shows the gresification diagram, water absorption and linear shrinkage as a function the firing temperature, of a typical industrial clayey body for roofing tiles fabrication, **MI**, from Campos dos Goytacazes. It can be seen that from 850 up to 1000°C, both water absorption and linear shrinkage slightly change. This behavior is associated to a refractory

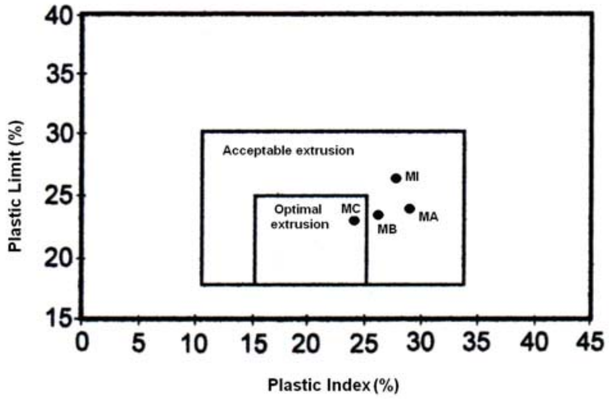

Fig. 4 **Extrusion prognosis through the plastic limit and plastic index.**

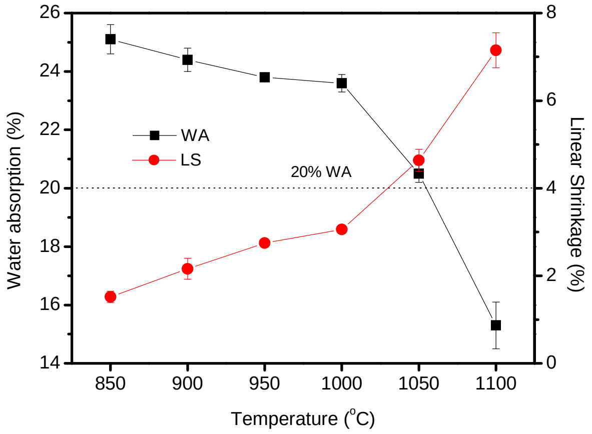

Fig. 5 **Gresification diagram of the MI.**

behavior of a kaolinitc clayey body, with low amounts of flux oxides and high amount of alumina, that difficult the closure of porosity. At temperatures above 1000°C, it is observed an abrupt decrease in water absorption and increase in linear shrinkage. This behavior is associated with the sintering mechanisms, diffusion in the solid state and liquid phase formation, which act effectively in the Campos dos Goytacazes clays from 1000°C. It is also observed that the level of 20% water absorption, maximum acceptable value for the type Roman roofing tiles, according to technical standard [11], is only reached at 1053°C. However, in this temperature, the variation of the water absorption and the linear shrinkage is very significant. This shows that variations of 50°C in an industrial kiln can cause dimensional problems and water absorption outside the specifications. Fig. 5 further shows that, in order to avoid, this kind of problem, the maximum kiln temperature should not surpass 1000°C. On the other hand, this temperature does not allow reaching the water absorption values required by technical standard. Therefore, one possible solution would be to fire roofing tiles in only one horizontal layer of kiln or attempt to increase the degree of dry packing of the specimens and also to reduce the loss on ignition — **LoI** to enable the reduction curve in the temperature range from 850 to 1000°C.

Figs. 6-8 show respectively the water absorption, linear shrinkage and flexural rupture strength of the clayey body as a function the firing temperature, respectively. It is possible to observe, in Fig. 6, that at the temperatures ranging from 850 to 1000°C the reformulated bodies, **MA**, **MB** and **MC**, have similar behaviour in comparison with **MI**, i.e., with slightly variation. By contrary, at higher temperatures all formulations present a significant decrease in water absorption, mainly **MB** and **MC**. These formulations have higher amounts of **GW**, and consequently elevated amount of flux oxides and lower amount of alumina, facilitating liquid phase formation. Finally, it is also observed the temperatures that the formulations reached 20% of water absorption, maximum value according to Brazilian norm [11]. These temperatures are 1028, 1032, 1051 and 1053°C for **MC**, **MB**, **MA** and **MI**, respectively.

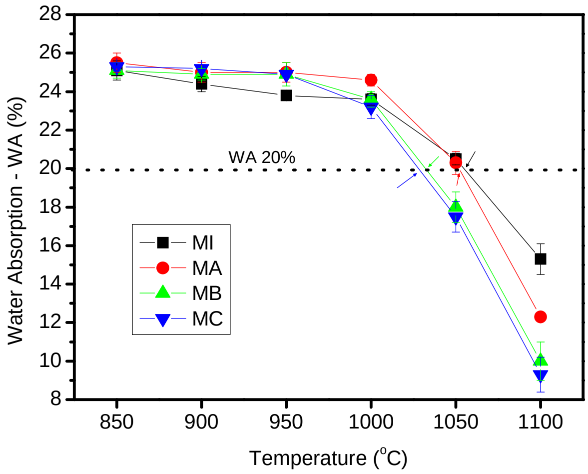

Fig. 6 **Water absorption of the formulations as a function the firing temperature.**

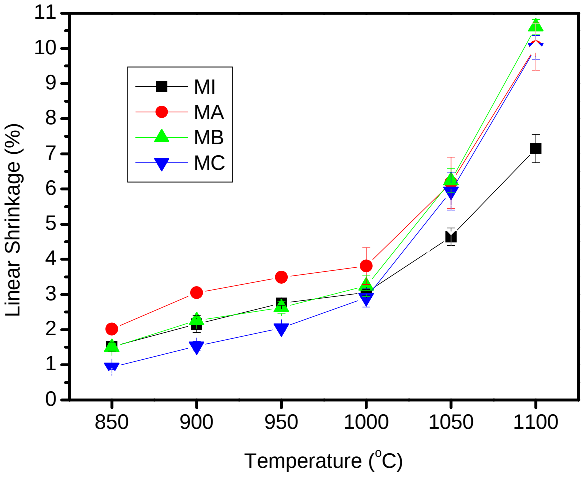

Fig. 7 Linear shrinkage of the formulations as a function the firing temperature.

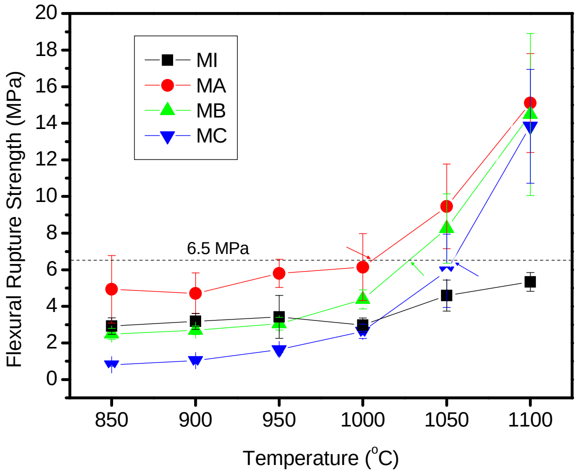

Fig. 8 Flexural rupture strength of the formulations as a function the firing temperature.

The linear shrinkage, Fig. 7, follows the behavior of water absorption, however, with the opposite effect. From 1000°C, all formulations display an abruptly increase in linear shrinkage, mainly the reformulated one **MA**, **MB** and **MC**. This behavior is a result of sintering reactions that enable an approximation of the particles.

In Fig. 8, it is observed the flexural rupture strength of the formulations. It is observed that from 850 to 1000°C, all formulations present a slightly variation in the mechanical strength. The recommended value of 6.5 MPa [12] was not overcome. The worse value for mechanical strength of **MC** can be attributed to the higher amount of quartz particles that act as a stress concentrator. Although the **GW** contributes supplying flux oxides, quartz particles are also increased with their addition. By contrary, **MA** displays higher amounts of flexural rupture strength since in this formulation sand was replacement by the same amount of **GW** that has a finer particle size, minimizing the stress concentrator action of quartz.

At temperatures above 1000°C the formulations **MA**, **MB** and **MC** present a significant increase in mechanical strength, as consequence of liquid phase formation with partial dissolution of quartz particles. It is important to mention that the flexural rupture strength of **MI** still presents a slightly variation**.**

Finally, it is also observed the temperatures that the formulations reached 6.5 MPa of flexural rupture strength, minimum recommended value. These temperatures are 1006, 1028, 1055°C for **MA**, **MB** and **MC**, respectively. According to Fig. 8, **MI** does not reach this value, proving its refractory behavior.

As final remarks, the preliminary characterization of a granite waste generated from sawing operation apparently revealed favorable conditions for addition into a heavy clay body in terms of compatible chemical composition, as well as a contribution to sintering with fluxing agents and adjustment of the clay extrusion forming process. The particle size distribution of the waste is also appropriate to the clayey ceramic processing.

4. Conclusion

The study of the effect of ornamental rock powder waste on the processing and properties of an industrial clayey body used for roofing tiles fabrication led to the following results:

- The investigated ornamental rock waste from sawing operations is composed of feldspars (microcline, albite, anorthite), quartz and micaceous mineral. This waste can be consider as a flux materials due to the amount of alkaline and earth alkaline oxides and also to the fine particle size.

- The waste contributed to the reduction of the plasticity of the industrial clayey body. In an industrial process, the consequence will be the decreasing in the amount of extrusion forming water, facilitating the drying stage and the dimensional control of the pieces.
- With respect to the fired properties, the waste contributed effectively to the reduction of water absorption and to increase the mechanical strength of the reference formulation, industrial clayey body, MI. The best formulation was MB, with 20wt.% of waste fully replacing sand and partially replacing clays, that attended both recommended values for water absorption and flexural rupture strength at 1032°C.

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