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Use of granite sawing wastes in the production of ceramic bricks and tiles

Romualdo R. Menezes[a,*], Heber S. Ferreira[b], Gelmires A. Neves[b],
Helio de L. Lira[b], Heber C. Ferreira[b]

[a] *Departamento de Engenharia de Materiais, Laboratório de Síntese e Processamento de Materiais Cerâmicos,*
LaSP, Universidade Federal de São Carlos, São Carlos 13 565-905, SP, Brazil
[b] *Departamento de Engenharia de Materias, Universidade Federal de Campina Grande, Campina Grande 58 109 970, PB, Brazil*

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Abstract

Granite process industry generates a large amount of wastes, which pollute and damage the environment. This work aims to characterize and evaluate the possibilities of using the granite sawing wastes, generated by the process industries from Paraíba State, Brazil, as alternative ceramic raw materials in the production of ceramic bricks and tiles. Samples of granite sawing wastes were collected from companies located in Paraíba State. Their characterization were carried out with the determination of density, particle size distribution, surface area (BET), chemical composition, and by DTA, TGA, XRD, and SEM. In a second part of the work, tests in ceramic compositions were conducted in order to evaluate the suitability of addition of wastes in ceramic compositions used in the production of ceramic bricks and tiles. The results showed that the granite wastes have physical and mineralogical characteristics that were similar to those of conventional ceramic raw materials. The ceramic bodies produced from reformulated ceramic compositions had technological characteristics in agreement with the Brazilian standardizations for ceramic bricks and tiles.

Keywords: Porosity; Plasticity; Clays; Traditional ceramics; Waste materials; Tiles

1. Introduction

The use of wastes after detecting their potentialities is considered today as an activity that can contribute to the diversification of products, decrease of final costs, besides providing alternative raw materials to a series of industrial sectors.

Recycling of wastes, generated by the industries, as alternative raw materials is not a new thing and has been done successfully in a lot of countries. The reasons that motivate these countries generally are: the exhaustion of the natural resources; the conservation of not renewable resources; improvement of the population health and security;[1] preoccupation with environmental matters; reduction in wastes disposal costs.

The construction industry is the most indicated technological activity sector to absorb solid wastes,[2] such as the wastes from the granite industry, due to the large quantity of raw materials used by the sector as well as by the large volume of final products in construction. The use of wastes as alternative raw materials in the ceramic industry, which embodies part of the construction industry, can contribute to diversify the offer of raw materials in the production of ceramic bricks and tiles and reduce the costs in a building, which is of primary importance in a country with a high deficit of houses, like Brazil.

Granite mining and process industry are one of the most promising business areas of the mining sector, with a mean growth in the world production of approximately 6% per year in the last 10 years. The international trading is approximately US$ 6 billions per year and around US$ 13 billions, taking into account tools, equipments, etc.[3] Although this industrial sector generates a large amount of wastes in the form

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[*] Corresponding author. Tel.: +55 16 260 8552.
E-mail addresses: rrmboca@iris.ufscar.br (R.R. Menezes), heber@paqtc.rpp.br (H.S. Ferreira), gelmires@dema.ufcg.edu.br (G.A. Neves), helio@dema.ufcg.edu.br (H.d.L. Lira), heber@dema.ufcg.edu.br (H.C. Ferreira).

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of a mud, basically composed of SiO_2, Al_2O_3, F_2O_3 and CaO, due to the sawing and polishing processes. Which can cause serious damages in the environment, such as soil and underground water contamination, if not efficiently treated before disposal.[4]

Brazil is one of the biggest granite world producers, either in the form of block or as processed products. Brazilian Northeast Region is an area that concentrates a large quantity of process industries, which are responsible by the disposing of hundred of tons of wastes in the environment per year. This scene is even more aggravated by the increasing production in the last decade, getting attention from all society with the destination of disposal wastes.

The recycling of granite wastes in the ceramic industry has attracted technological attention in the last years due to the possibility of reduction of the production costs, use of residues as a secondary raw material in the production of very stable glassy phases (glass and glass–ceramic industry), and by the opportunity in overcoming some problems in the production of bricks and tiles with the incorporation of granite sawing wastes in their formulations. All these benefits are largely due to the reproducibility of the chemical composition and particle size distribution of these wastes. In this way, some works have been published regarding to the use of granite wastes in the fabrication of glasses and glass–ceramics,[5–8] porcelainized bodies[9] and production of bricks and roof tiles.[10,11]

Therefore, the aim of this work was to characterize and evaluate the suitability of use granite sawing wastes as alternative ceramic raw materials in the production of ceramic bricks and tiles.

2. Experimental procedures

In the development of this research, conventional ceramic raw materials and granite sawing wastes were used. The following conventional raw materials were used:

- red clay, from Santa Rita city, Paraíba State, supplied by *Companhia Industrial Cerâmica—CINCERA*, located in the Industrial Center of Santa Rita;
- ball-clay type clay, from Alhandra city, Rio Grande do Norte State, supplied by *Indústria Armil Minérios*, located in the Industrial Center of Campina Grande city, Paraíba State;
- quartz, from Morro do Careca hill, Parelhas city, Rio Grande do Norte State, supplied by *Indústria Armil Minérios*, located in Parelhas city;
- feldspar, from Parelhas, supplied by *Indústria Armil Minérios*, located at Parelhas;
- calcite, from Boa Vista city, Paraíba State, supplied by *Indústria Armil Minérios*, located in the Industrial Center of Campina Grande.

All these conventional raw materials have already been analyzed and are used regularly as raw material by the ceramic industries of the Northeast Region. Table 1 presents their chemical composition.

Table 1
Chemical composition of the conventional raw materials

Raw materials	Weight percent						
	LOI[a]	SiO_2	Fe_2O_3	Al_2O_3	CaO	Na_2O	K_2O
Red clay	11.03	44.41	13.65	26.45	–	3.12	3.51
Ball clay	12.20	49.34	6.87	20.50	3.50	2.40	3.00
Quartz	0.34	96.76	–	–	–	0.67	0.60
Feldspar	0.51	66.7	–	22.40	–	2.70	6.66
Calcite	39.27	1.55	–	–	53.48	2.70	–

[a] Loss on ignition.

Three samples of granite sawing wastes were selected and collected from process industries located at the Paraíba State. The granite wastes were obtained in the form of a mud, which were dried and used in the reformulation of ceramic compositions. The used wastes were:

- waste from *Poligran, Polimentos de Granitos do Brasil*, located in the Industrial Center of Campina Grande, identified by *Poligran*;
- waste from *Caxambu*, located in the Caxambu Farm, Cabaceiras city, identified by *Caxambu*;
- waste from *Fuji, Mármores e Granitos*, located in the Industrial Center of Campina Grande, identified by *Fuji*.

The wastes identified by Poligran and Fuji were generated in a process using a cutting machine with steel saws, whist the Caxambu was generated in a process using diamond cutting saws.

The wastes were characterized by determination of density, according to the picnometry method, using a picnometer model AccuPyc 1330 from the Micromeritics; particle size distribution, by wet sieving and sedimentation (according to Brazilian standardization[12]); surface area by the method of N_2 adsorption (BET) using He as flowing gas in a Micromeritics device, model ASAP 2370; chemical composition, wet process, differential thermal analyze (DTA) and thermal gravimetric analyze (TGA) using a BP Engenharia equipment, model RB 3000; X-ray diffraction (XRD) using a Siemens/Brucker equipment, model AXD 5005, with radiation Cu Kα (40 kV/40 mA); scanning electronic microscopy (SEM) and energy dispersive spectrometry (EDS) in a Leica equipment, model S 440.

Red clay and wastes (amount of 20, 25, 30, 35, 40, 45, 50, 55 and 60% in weight) were used in the formulation of bricks. The formulation of compositions for production

Table 2
Density of the granite wastes

Waste	Density (g/cm^3)
Poligran	2.69 ± 0.05
Caxambu	2.63 ± 0.05
Fuji	2.70 ± 0.14

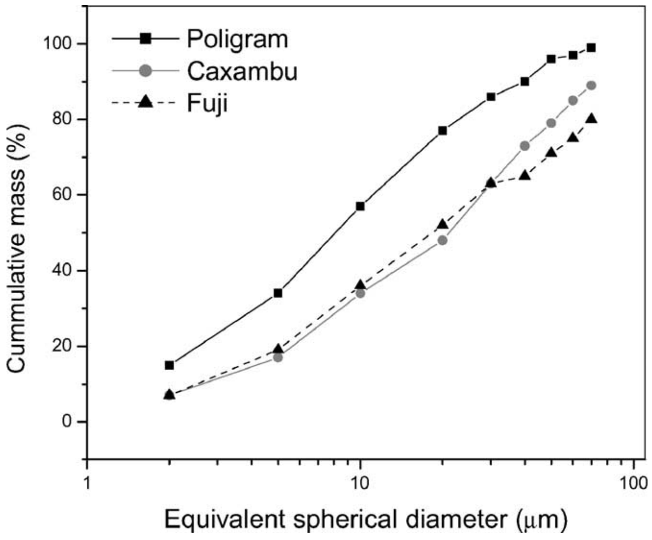

Fig. 1. Particle size distribution of the granite wastes.

Table 3
Surface area of the granite wastes

Waste	Surface area (BET) (m^2/g)
Poligran	11.41
Caxambu	6.90
Fuji	6.16

of tiles was carried out by the REFORMIX 2.0 software, developed by the Materials Engineering Department of the Federal University of São Carlos, Brazil, applying as reference to calculations the chemical composition of a commercial tile. The compositions for production of tiles were composed by ball clay, kaolin, quartz, feldspar, calcite and granite wastes, with the level of wastes ranging from 15 to 40%.

Atterberg limits of compositions constituted by red clay and wastes were determined. In the laboratory-scale studies, samples composed by wastes and red clay, 20 cm × 2.0 cm × 2.0 cm (according to reference[13]), were extruded and fired at 800, 900 and 1000 °C in an oxidizing atmosphere. After firing, the water absorption of the samples was determined and a three-point bend strength test was performed. For the industrial-scale tests, samples of 19 cm × 9.0 cm × 9.0 cm were uniaxially pressed and fired between 750 and 850 °C in a brick industry (high temperature gradient inside the industrial furnace) located at Campina Grande city. After the firing of the ceramic bricks, its compression strength and water absorption were determined according to the Brazilian standardization.[14,15]

Formulations to the production of floor and wall tiles were also submitted to laboratory and industrial-scale tests. In the laboratory tests, ceramic bodies of 12 cm × 6.0 cm × 1.0 cm were uniaxially press at 20 MPa and fired at 1150, 1175 and 1200 °C (fire rate of 9 °C/min and soak time of 30 min). After thermal treatment, the water absorption of the samples was determined and a three-point bend test was performed. Both tests were carried out according to Brazilian standardization.[16] The industrial-scale tests were done in the *Cordeiro do Nordeste* company, located in João Pessoa city, Paraíba State. The samples to the industrial test were produced in the same way as those used in the laboratory tests, although they were fired at 1150 °C and all the thermal

Table 4
Chemical composition of the granite wastes

Waste	Weight percent						
	LOI[a]	SiO_2	Fe_2O_3	Al_2O_3	CaO	Na_2O	K_2O
Poligran	4.44	59.61	5.98	11.77	4.48	2.70	3.63
Caxambu	2.93	88.91	–	6.64	–	0.14	0.06
Fuji	2.57	60.20	6.30	13.80	6.02	3.38	3.63

[a] Loss on ignition.

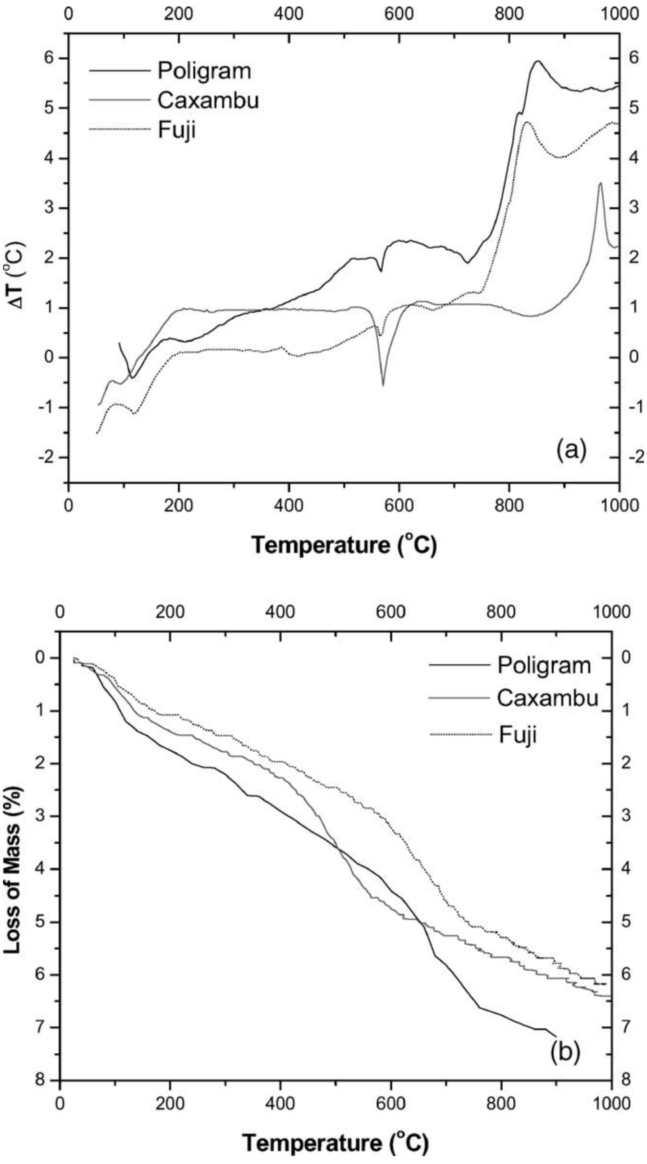

Fig. 2. DTA (a)/TGA (b) analysis of the granite wastes.

cycle was only of 35 min. Afterwards the modulus of rupture and water absorption of the tiles were determined. Both of them according to the Brazilian standardization.[16]

3. Results and discussion

3.1. Characterizing tests

Table 2 presents the densities of the investigated wastes. The values of density are very close, ranging from 2.63 to 2.70 g/cm^3. The results are in agreement with the range of values (2.60–2.75 g/cm^3) observed in literature[13,17] for conventional non-plastic raw materials.

Fig. 1 shows the particle size distribution of the wastes. It can be observed that the wastes have about 80–89% (wt.%) of particles with equivalent diameter lower than 70 μm and from 7 to 15% of particles with equivalent diameter lower than 2 μm. These results point out that the granite wastes have a granulometry similar to the processed conventional non-plastic raw materials, such as quartz and feldspar, without additional grinding. However, the size distribution is large, with a higher fraction of fine particles (equivalent diameter <10 μm).

Table 3 presents the specific surface area of the wastes. The values ranged from 6 to 11 m^2/g and are superior than those values expected for conventional beneficiated non-plastic raw materials (from 1.0 to 2.5 m^2/g). Comparing with the literature,[13,17] it can be verified that the specific surface area of wastes provides values compared to those related to primary kaolin (3.3–19.8 m^2/g).

Table 4 presents the chemical composition of the granite wastes. One can observe that the wastes have high amount of SiO_2, superior than 59%, and levels of Al_2O_3 ranging from 6.40 to 13.80%. The content of Fe_2O_3 was around 6% in the Poligram and Fuji wastes, but it was not observed in the Caxambu waste, due to the use of diamond saw in the cutting process of Caxambu granite. The presence of CaO and Fe_2O_3 in the wastes is primary due to the use of metallic dust and lime as abrasive and lubricant, respectively. The content of fluxing agents (Fe_2O_3, Na_2O and K_2O) ranges from 12 to 13%, with the exception of the Caxambu waste.

DTA/TGA analysis points out that the Poligran and Fuji wastes have peaks related with quartz, mica and calcite. Whilst Caxambu waste presents only peaks associated with kaolinite and mullite (the quartz peak was probably overlapped by the kaolinite band). The TG analysis graphs shows that the loss of mass of the wastes range from 6 to 7%. The Poligran and Fuji wastes exhibit their highest loss of mass in the range of temperatures of decomposition of calcite, while the Caxambu waste has it highest loss of mass in the range

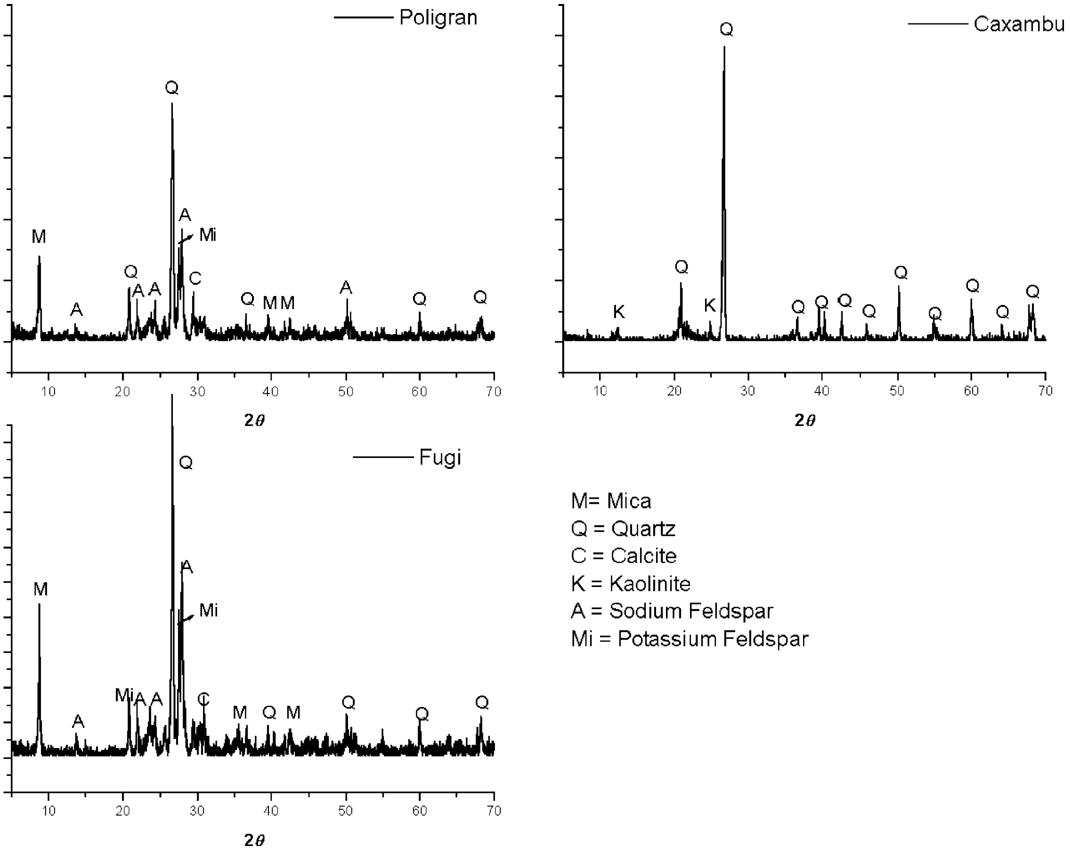

Fig. 3. X-ray diffraction patterns of the granite wastes.

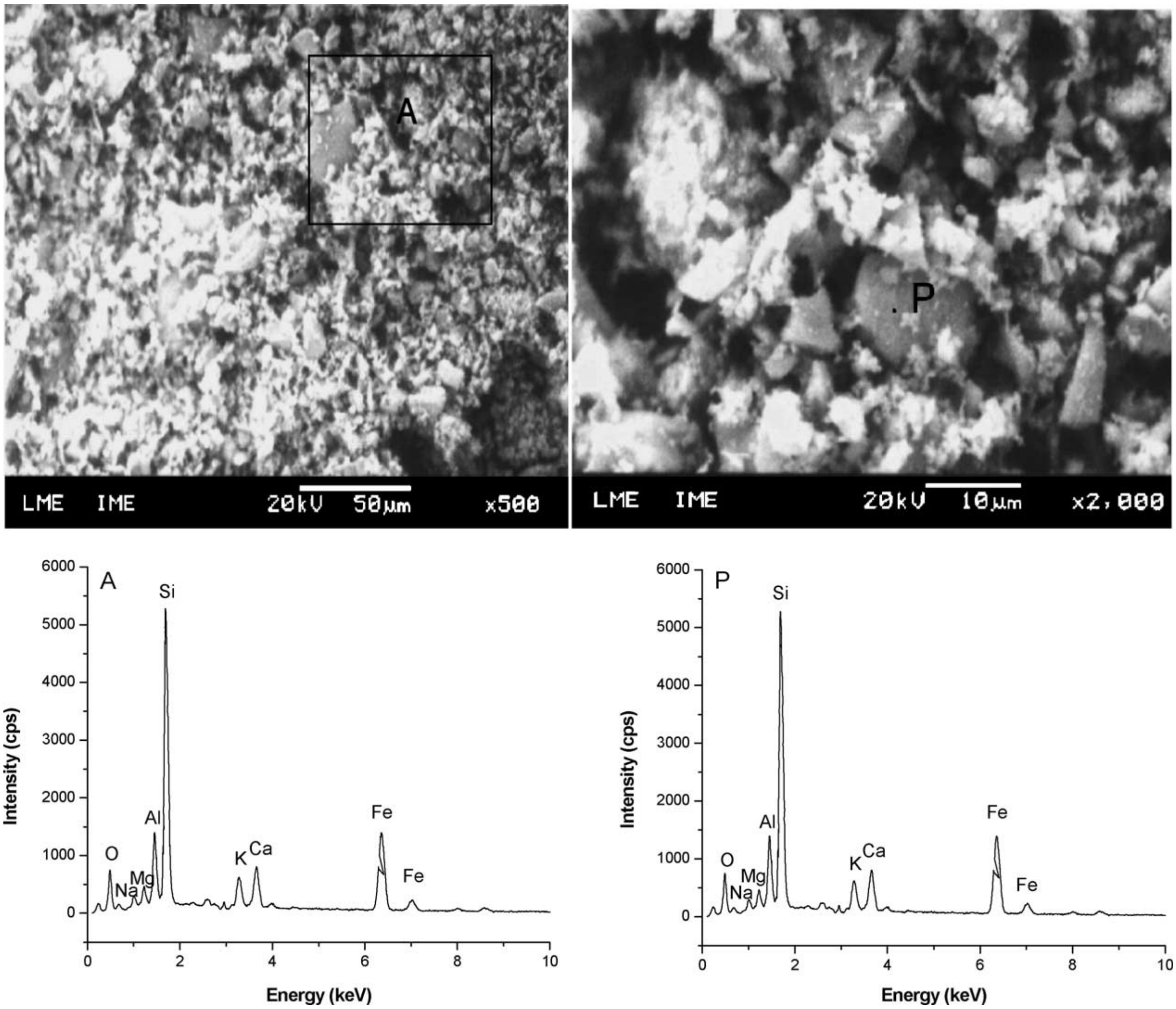

Fig. 4. SEM micrographs and EDS spectrum of Poligran waste.

of temperatures of dehydroxilation of clay minerals. Fig. 2 presents the DTA/TGA graphs of the granite wastes.

Fig. 3 shows the X-ray diffraction patterns of the wastes. The diffraction patterns of the Poligran and Fuji wastes have peaks of quartz (SiO_2), potassium feldspar ($KAlSi_3O_8$), sodium feldspar ($NaAlSi_3O_8$), mica and calcite ($CaCO_3$), while in the Caxambu waste only the presence of quartz and kaolinite was observed. The results of the X-ray diffraction analysis are in agreement with the thermal analysis results.

Figs. 4 and 5 shows the SEM micrographs of the Poligran and Caxambu wastes. It can be observed that the Poligran waste has particles with irregular forms and round surface as well as some others show angular corners. The SEM/EDS analysis fits well with the results of the wet chemical analysis, with the presence of Si, Al, Fe, Ca, K and Na. The Fuji waste displays a microstructure and EDS spectrum similar to the Poligran waste. The Caxambu waste exhibits particles with irregular forms and round surface and some others with angular corners, although the EDS spectrum revealed to be very different from the Poligran waste EDS spectrum, because only Si and Al were present in the early case.

Based on physical and mineralogical characterization of the wastes, it can be observed that they have characteristics similar to the ones of the conventional non-plastic ceramic raw materials (quartz and feldspar). The wastes have particle size distribution very close to the conventional materials, with a slightly higher level of fine particles. In this way, the use of granite wastes instead of conventional non-plastic raw materials yields a reduction in the energetic costs of the ceramic process due to the elimination of grinding steps. The Poligran and Fuji wastes have chemical and mineralogical similarities with the feldspar, with a high level of fluxing agents, while the Caxambu waste has a composition closer to sands, due to the high level of SiO_2 and low content of fluxing agents.

3.2. Technological tests

The plasticity parameters of the compositions for the production of bricks (red clay and wastes), in terms of the Atterberg limits, are shown in Table 5. It can be observed that the values of the liquid limits range from 34.6 to

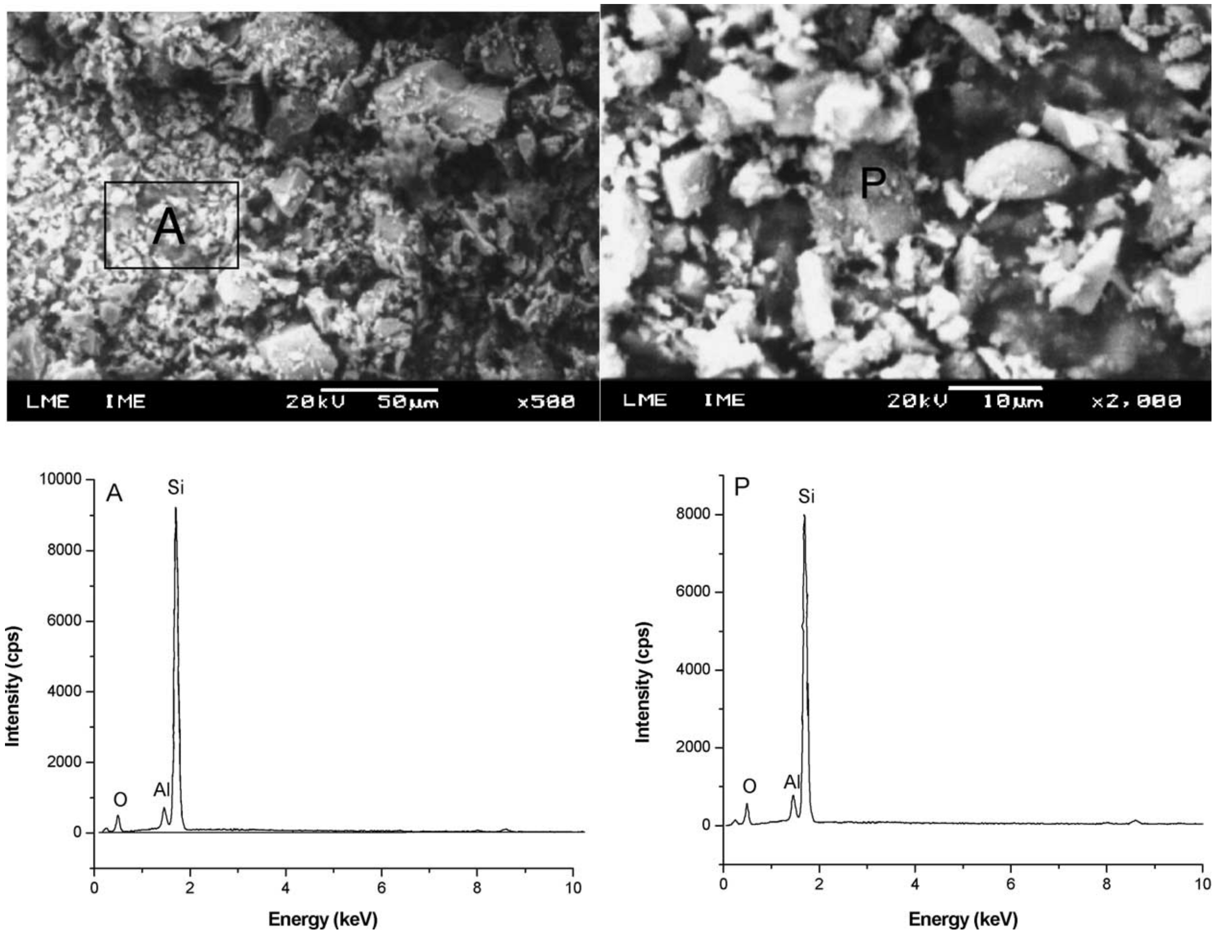

Fig. 5. SEM micrographs and EDS spectrum of Caxambu waste.

53.0%, which are in agreement with the range defined in the literature (30–60%) to compositions used for production of bricks.[17–20] The plasticity limits ranged from 20.0 to 26.5%, in accordance with the values (15–30%) indicated to ceramic bodies used in the production of bricks by extrusion.[18]

Based on Table 5, it is verified that the values of the plasticity index (PI) range from 12.4 to 27.0. The compositions with up to 45% of wastes were classified as high plastic (PI > 15%) while the formulations with 50, 55 and 60% of waste were classified as median plastic (7% < PI < 15%). According to literature,[17–20] these results are in agreement

Table 5
Liquid and plasticity limit and plasticity index

Wastes	Weight percent								
	20	25	30	35	40	45	50	55	60
Liquid limit (%)									
Poligran	45.0	43.0	30.0	39.5	39.0	37.8	36.5	35.5	34.6
Caxambu	53.0	52.0	50.0	45.0	46.0	44.0	40.0	40.0	38.0
Fuji	52.0	50.0	49.0	49.0	39.0	43.0	36.0	41.0	36.0
Plasticity limit (%)									
Poligran	24.2	23.0	22.4	23.0	24.0	23.4	23.2	22.8	22.5
Caxambu	26.0	27.0	25.8	24.0	25.0	23.0	25.0	24.0	23.0
Fuji	26.5	26.2	25.0	25.0	20.0	27.0	20.0	26.0	21.0
Plasticity index (%)									
Poligran	20.8	20.0	18.9	16.5	15.4	15.5	12.9	12.7	12.4
Caxambu	27.0	25.0	25.0	21.0	21.0	21.0	15.0	16.0	15.0
Fuji	25.5	23.8	24.0	24.0	19.0	18.0	16.0	15.0	14.0

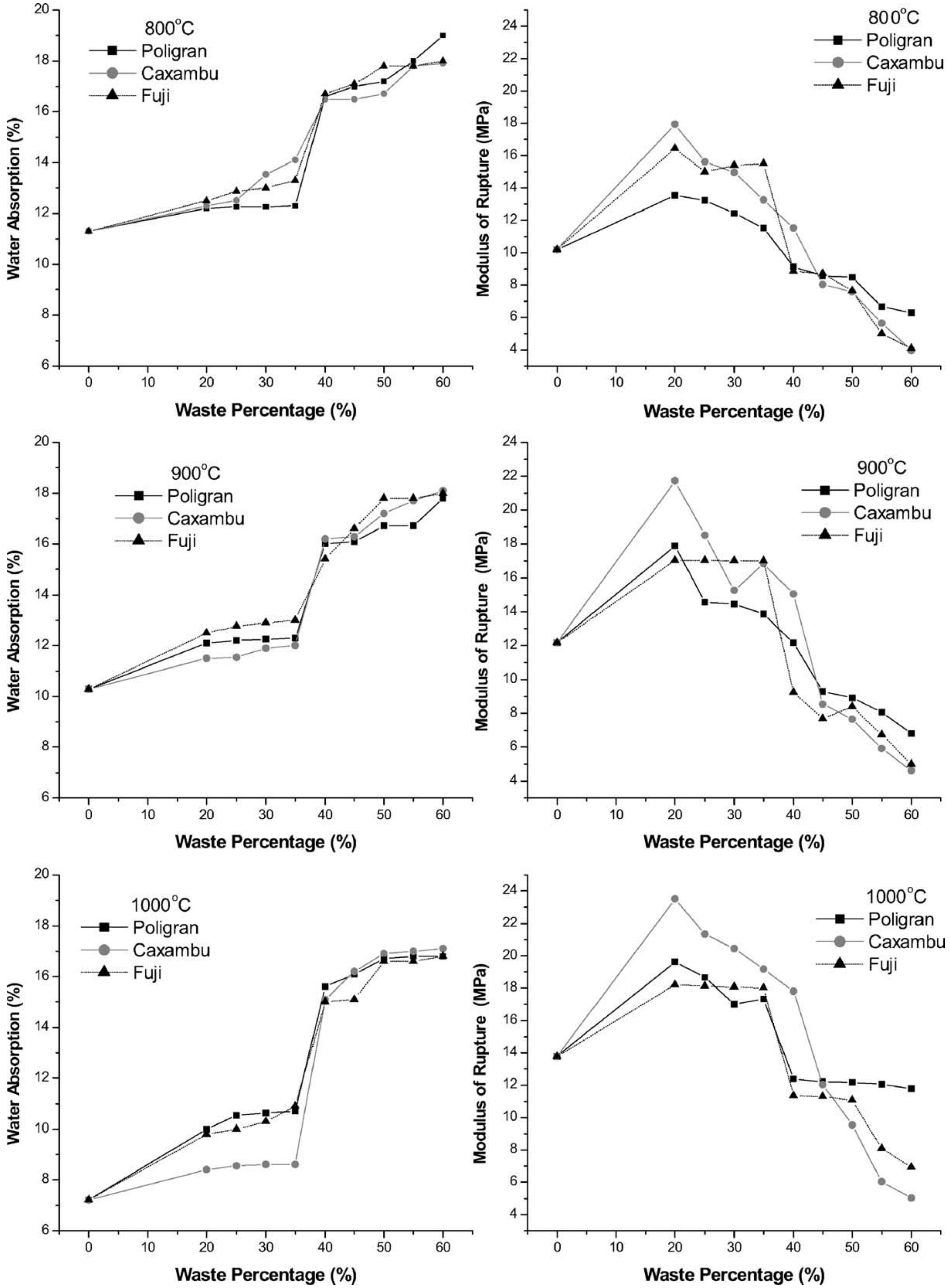

Fig. 6. Water absorption and modulus of rupture of laboratory-tested ceramic bodies fired at 800, 900 and 1000 °C.

with the set of values (10–30%) indicated as appropriate to the production of bricks by extrusion.

Fig. 6 shows the results of water absorption and modulus of rupture of ceramic bodies composed by red clay and wastes obtained in the laboratory tests. It can be observed that the increase in the waste content give rise to an increase in the water absorption, independently of the firing temperature. The modulus of rupture increases followed by a decrease with the growth of the waste content, despite of the firing temperature.

Based on Fig. 6, one can observe that the firing temperatures are not sufficient to cause the fusion of the granite wastes, with them acting merely as inert non-plastic materials in these range of temperatures, similar to the quartz. In this way, the presence of granite wastes reduced the firing shrinkage with regard to the pure clay with consequent increase in the open porosity (estimated by the water absorption). The increase followed by the decrease of the modulus of rupture can be explained by an increase in the permeability of the green body which allowed the easier escape of the water during the drying (higher permeability) without generation of internal stresses and micro-cracks in the ceramic body, yielding specimens without such defects and consequently higher modulus of rupture. Macro-cracks in some bodies composed only by red clay was observed after their drying, and is the primary indication of the generation of high internal stresses during the drying.

According to the Fig. 6, it was verified that all samples with wastes up to 50% have values in agreement to literature,[13] even for the water absorption—lower than the maximum values of 25% to bricks and of 20% to roof tiles—as well as for the modulus of rupture—minimum values of 5.5 MPa to bricks and of 6.5 MPa to roof tiles. Based on this preliminary analysis, industrial-scale tests were conducted with compositions with up to 50% of wastes. Fig. 7 shows the results of water absorption and compression strength of bricks after firing at the industry. It can be observed that all the compositions present values of compression strength in the range normalized by the Brazilian standards.[14,15]

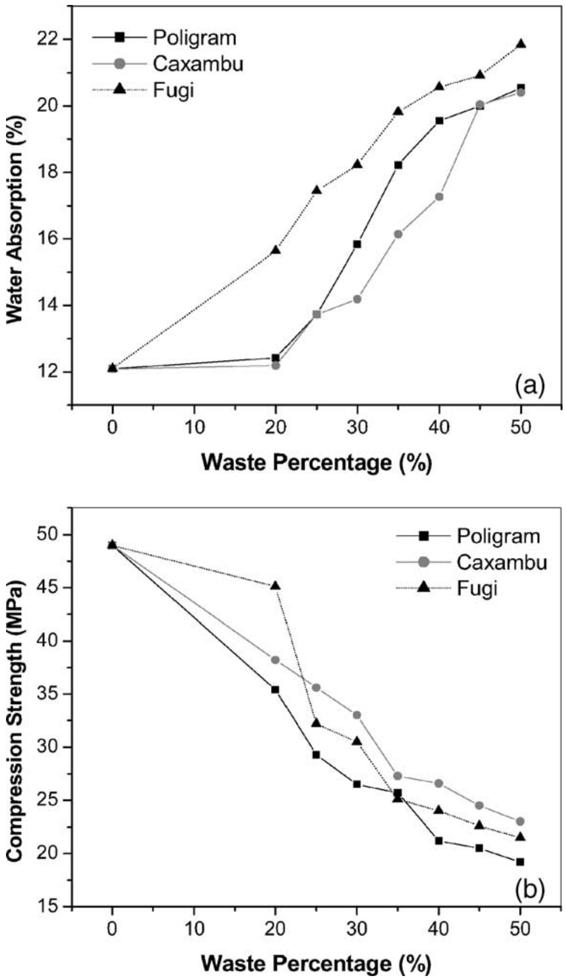

Fig. 7. Water absorption and compression strength of bricks fired at industry.

The values of water absorption and modulus of rupture of some tile formulations after firing at 1150, 1175 and 1200 °C, in laboratory tests, are shown in Fig. 8. Table 6 presents the formulation of these ceramic compositions. According to Fig. 8, it can be observed a decrease in the water absorption and an increase in the modulus of rupture with the elevation of the firing temperature, which is related with the fusion and vitrification of the granite wastes, that acted as fluxes in the studied temperatures. According to the Brazilian standardization,[16] it can be observed that, after firing at 1150 °C, only the ceramic bodies with Polligran waste (MPL4) and Fugi waste (MFG2 and 3) can be used as wall and floor tiles. The ceramic bodies with Caxambu waste do not have physical characteristics that make them usable as ceramic tiles in that temperature. Firing at 1175 and 1200 °C, all the formulations can be used as floor tiles. At 1200 °C, it can be observed that the formulation MPL4 can be used to produce ceramics with water absorption lower than 3%.

Table 6
Batch composition of formulations to tile production

Mass	Raw materials (wt.%)					
	Ball clay	Kaolin	Waste	Feldspar	Calcite	Quartz
MPL1[a]	46	10	25[a]	–	7	12
MPL4[a]	45	12	38[a]	–	2	3
MCA1[b]	32	12	30[b]	23	2	–
MCA3[b]	32	10	21[b]	31	5	–
MFG2[c]	35	15	35[c]	–	5	10
MFG3[c]	41	13	30[c]	–	4	12

[a] Poligran waste.
[b] Caxambu waste.
[c] Fugi waste.

Based on the laboratory tests only the compositions formulated with Poligram and Fugi wastes were used in the industrial-scale tests. Table 7 presents the values of water absorption and modulus of rupture of ceramic bodies fired at industrial furnace. According to Brazilian standards[16] it can be observed that, in the industrial conditions the ceramic

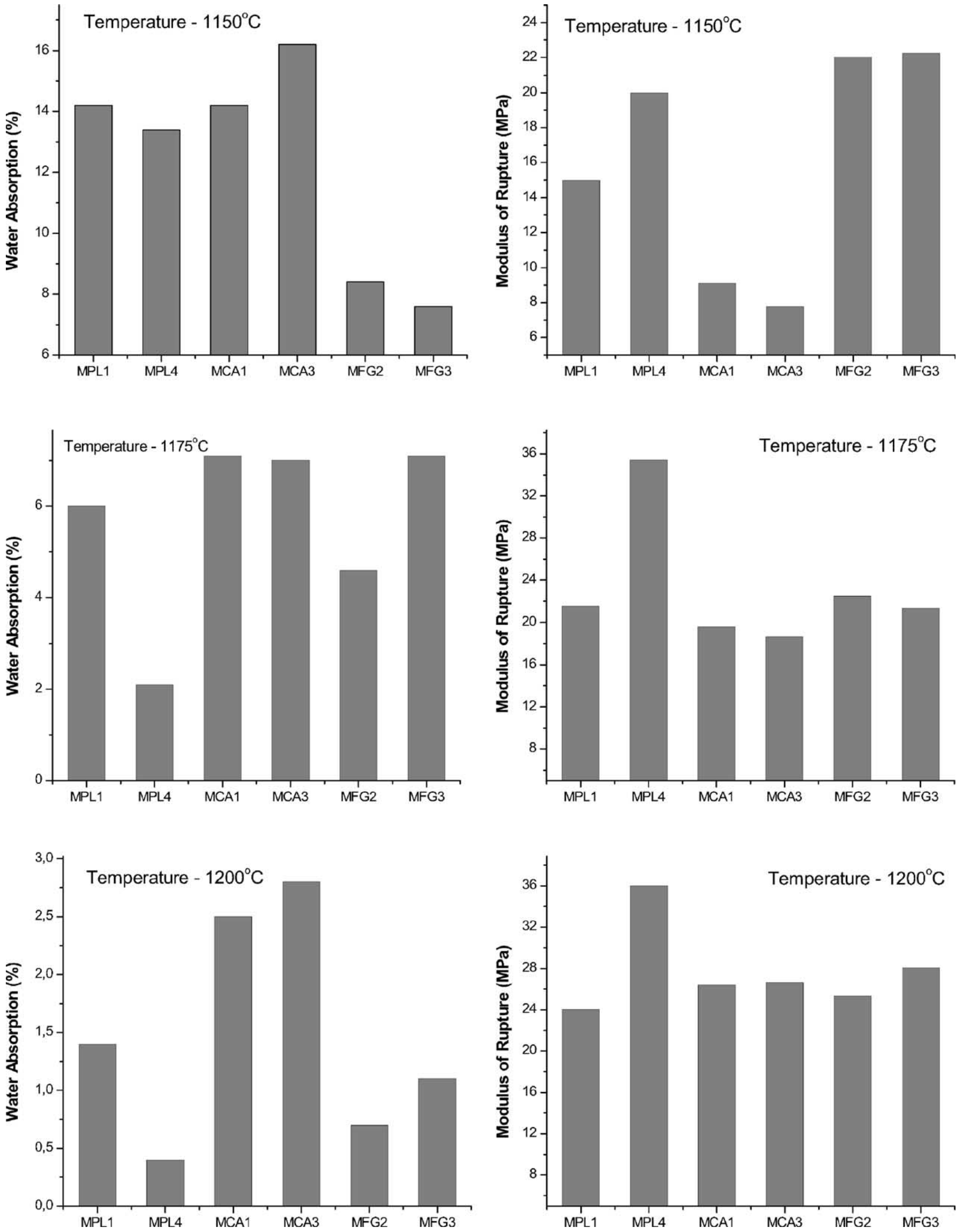

Fig. 8. Water absorption and modulus of rupture of laboratory-tested ceramic tiles fired at 1150, 1175 and 1200 °C.

Table 7
Water absorption and modulus of rupture of industrial fired ceramic tiles

Mass	Water absorption (wt.%)	Modulus of rupture (MPa)
MPL1[a]	13.2	15.00
MPL4[a]	8.9	22.67
MFG2[c]	13.3	19.73
MFG3[c]	12.7	19.00

[a]Poligran waste, [c]Fugi waste.

bodies produced using the MPL4, MFG2, and MFG3 compositions can be used as ceramic floor and wall tiles.

4. Conclusions

The characterization and analysis of the feasibility for the use of granite sawing wastes, produced by the granite processing industry from Brazilian Northeast Region, as alternative ceramic raw materials for production of bricks and tiles, yielded the following conclusions: (a) the wastes were essentially composed by quartz, feldspar, calcite and mica, with one of them basically composed by quartz and kaolinite; (b) the density, particle size distribution and specific surface area of the wastes were very similar to those of the conventional ceramic raw materials, placed between the typical values of the plastic and non-plastic raw materials; (c) the addition of wastes in ceramic compositions for production of bricks, up to 35% in weight, caused a slight increase in the water absorption, but an increase in the modulus of rupture was observed otherwise; (d) ceramic compositions with additions of wastes can be used to produce wall and floor tiles, and firing at 1200 °C they can be used to produce ceramics with water absorption lower than 3%.

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