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Valorization of Municipal Waterworks Sludge to Produce Ceramic Floor Tiles

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Abstract: In municipal waterworks large amounts of waste in the form of sludge have to be discarded. This investigation focuses on the processing of ceramic floor tiles incorporated with a municipal waterworks sludge. Four floor tile formulations containing up to 10 wt. % of the municipal waterworks sludge were prepared in order to replace the kaolin. The floor tile processing route consisted of dry powder granulation, uniaxial pressing, and firing between 1190 and 1250 °C using a fast-firing cycle (<60 min). The densification behavior and technological properties of the floor tile pieces as function of the sludge addition and firing temperature were determined. The development of the microstructure was followed by XRD and SEM/EDS. The results show that the replacement of kaolin with municipal waterworks sludge, in the range up to 10 wt. %, allows the production of ceramic floor tiles (group BIb and group BIIa, ISO 13006 Standard) at lower firing temperatures. These results suggest a new possibility of valorization of municipal waterworks sludge in order to bring economic and environmental benefits.

Keywords: municipal waterworks sludge; waste; recycling; valorization; floor tiles

1. Introduction

The municipal waterworks are units based on the physical and chemical treatments of raw fresh-water, mainly for human consumption, which produces huge amounts of wastes in form of sludge worldwide [1–6]. Waterworks sludge varies widely in terms of physical and chemical characteristics as a function of the nature of the raw fresh-water and chemical compounds applied during treatments. At present, the non-treated municipal waterworks sludge is disposed in places unsuitable around the world, in many cases near watercourses and landfill sites mainly in non-EU countries, with negative impacts on fauna, flora and human health [7]. Thus, an important issue for the municipal waterworks and ecologists is to find new solutions to the final disposal of this abundant waste material in an economic and ecological way.

Chemically, the municipal waterworks sludge is rich in SiO_2, Al_2O_3, and Fe_2O_3, and, in minor amounts, in K, Na, Mg, and Ca oxides [8]. The waterworks sludge is very similar, in terms of chemical composition, to common clays used to manufacture of clay-based products (Table 1) [8–13]. For this reason, the municipal waterworks sludge attracts high interest in view to prepare clay-based products contributing at the same time to reduce environmental impacts. In fact, previous studies [14–23] have shown promising results for the reuse of municipal waterworks sludge in the production of clay bricks, roofing tiles, and soil-cement bricks. However, the reuse of municipal waterworks sludge in the processing of ceramic floor tiles has not yet been investigated. Floor tiles are ceramic materials widely used in the civil construction sector. They are vitrified materials with heterogeneous microstructure and good technical properties in terms of mechanical strength, water absorption, and durability. The floor tile formulation is composed essentially of a mixture of non-renewable natural raw materials, such as clays, feldspars, and quartz [24]. Floor tile formulation contains large amounts (30–40%) of natural clays (plastic clay and kaolin), which provide favorable processing conditions, green strength, and rigidity of fired product. Thus, the municipal waterworks sludge could be a suitable replacement for natural clays in ceramic floor tile formulation.

Table 1. Chemical compositions of municipal waterworks sludges and common clays (wt. %).

Oxides	MWS	MWS	MWS	Clay	Clay	Clay
	ref. [9]	ref. [8]	ref. [10]	ref. [11]	ref. [12]	ref. [13]
SiO_2	59.70	35.92	52.78	46.42	52.67	41.10
Al_2O_3	10.52	31.71	14.38	27.90	20.20	31.48
Fe_2O_3	4.38	12.79	5.20	9.10	5.69	6.05
K_2O	1.16	0.58	3.62	1.67	2.27	1.77
Na_2O	1.53	0.06	0.97	0.36	0.12	0.62
MgO	2.20	0.37	3.08	0.71	1.33	0.35
CaO	6.01	0.10	4.39	0.22	2.07	0.28
TiO_2	-	1.10	0.61	1.32	0.15	1.49
MnO	-	0.09	0.08	0.11	0.03	-
SO_3	2.85	-	-	-	0.01	-
$^{+}$LoI	11.10	16.93	8.96	11.96	14.48	16.58

$^{+}$LoI—loss on ignition.

The purpose of this study is to investigate the possibility of valorization of a municipal waterworks sludge, a renewable raw material, into ceramic floor tile formulation for use in civil construction.

2. Materials and Methods

Four floor tile formulations using triaxial mixtures of kaolin + municipal waterworks sludge, albite, and quartz were prepared (Table 2). A representative municipal waterworks sludge (MWS) sample was collected from a municipal waterworks located in southeastern Brazil (Campos dos Goytacazes city, State of Rio de Janeiro, Brazil). After drying, the MWS sample takes the form of a clay-like powder. Table 3 gives the chemical composition of the MWS sample. From the mineralogical point of view, the MWS sample was mainly composed of kaolinite, with quartz, gibbsite, and goethite as accessory minerals. The standard formulation of floor tile used as a reference consisted of 40.0 wt. % kaolin, 47.5 wt. % albite, and 12.5 wt. % quartz [25]. In this work, kaolin was partially replaced with increasing amounts of municipal waterworks sludge. Each formulation is labeled as follows: ML0 contains 0–wt. % MWS; ML1 contains 2.5 wt. % MWS; ML2 contains 5.0 wt. % MWS; and ML3 contains 10.0 wt. % MWS. The MWS sample was used in moderate amounts (up to 10 wt. %) due to its high amount of organic matter (25.85 wt. %).

Table 2. Compositions of the floor tile formulations (wt. %).

Raw Materials	ML0	ML1	ML2	ML3
Kaolin	40.0	37.5	35.0	30.0
MWS	0.0	2.5	5.0	10.0
Albite	47.5	47.5	47.5	47.5
Quartz	12.5	12.5	12.5	12.5

Table 3. Chemical composition of the MWS sample (wt. %).

SiO_2	Al_2O_3	Fe_2O_3	K_2O	Na_2O	MgO	CaO	TiO_2	MnO	$^{+}$LoI
30.11	31.73	10.39	1.29	-	-	0.35	1.06	0.14	24.93

$^{+}$LoI—loss on ignition.

The raw materials were dry-ground separately using a laboratory grinder, and then passed through a 325-mesh (<45 µm ASTM) sieve. The floor tile formulations were mixed, homogenized, and granulated by the dry process [26].

The chemical compositions of the floor tile formulations were determined by using an energy-dispersive X-ray spectrometer (Shimadzu, Japan, EDX 700). X-ray diffraction analysis was done in a conventional powder diffractometer (Shimadzu, Japan, XRD 7000) by using monochromatic Cu-Kα radiation (λ = 0.154056 nm) at a scanning speed of 1.5 (2θ)/min. Mineral phases were identified by using the JCPDS-ICDD data files. The plastic properties were determined by the Atterberg method according to NBR 6459 and NBR 7180 standardized procedures. The Hausner ratio was obtained as the ratio of the tap density to apparent density of the granulated tile powders. The screen residue (>63 µm) has been also determined [26].

The floor tile powders were moistened with 7 wt. % water, pressed into test bars (11.50 × 2.54 cm^2) under a load of 50 MPa, and then dried at 110 °C. The green tile pieces were fast-fired between 1190 and 1250 °C in air using a laboratory kiln for a fast-firing cycle of less than 60 min, including cooling. The fast-firing cycle used in this investigation was selected to simulate an actual firing process used in the ceramic tile industry [25].

The following as-fired technological properties of the floor tile pieces have been determined in accordance with standardized procedures: linear shrinkage, water absorption, apparent density, and flexural strength. Linear shrinkage values upon drying and firing were evaluated from variation of the main dimension (length) of the pieces [27]. The water absorption values were determined from the weight differences between the as-fired and water-saturated samples (immersed in boiling water for 2 h) [28]. The apparent density was determined using the Arquimedes principle [28]. The flexural strength (average of five specimens for each value) was determined by a three-point bending test using an Instron model 1125 universal mechanical testing machine at a loading rate of 0.5 mm/min. The flexural strength (FS) was calculated by $FS = 3PL/2ab^2$, in which P is the load of rupture, L is the distance between supports, *a* is the specimen width, and *b* its thickness [29].

The microstructural characterization of the gold-coated fracture surfaces of fired specimens was examined via secondary electron images using a scanning electron microscopy (Shimadzu, Japan, SSX-550) operating at 15 kV. The mineral phases after fast-firing cycle were identified by X-ray diffraction analysis.

3. Results and Discussion

The chemical compositions of the floor tile formulations are given in Table 4. In terms of chemical composition, the SiO_2, Al_2O_3, and Na_2O were the most abundant components (90.08–92.25 wt. %). Those oxides are responsible for the main chemical and physical activities of the floor tile formulations during the fast-firing cycle. The effect of the incorporation of the MWS into tile formulations was to increase both the amount of iron oxide (Fe_2O_3) and loss on ignition. As shown in Table 3, the MWS sample contains appreciable amount of Fe_2O_3 (10.39 wt. %) and loss on ignition (24.93 wt. %). This can play an important role in the firing behavior and floor tile quality.

Table 4. Chemical compositions of the floor tile formulations (wt. %).

Oxides	ML0	ML1	ML2	ML3
SiO_2	64.98	64.48	64.01	63.07
Al_2O_3	22.47	22.42	22.35	22.27
Fe_2O_3	0.16	0.41	0.67	1.17
TiO_2	0.02	0.04	0.07	0.12
Na_2O	4.80	4.78	4.76	4.74
K_2O	1.51	1.49	1.48	1.44
CaO	0.20	0.21	0.21	0.21
MgO	0.03	0.07	0.07	0.06
MnO	0.04	0.04	0.04	0.05
$^{+}$LoI	5.79	6.06	6.34	6.82

$^{+}$LoI—loss on ignition.

Table 5 gives important physical parameters of the floor tile formulations. It was verified that the IP value (IP = UPL − LPL) tends to lightly increase (11.3 to 12.9%) with the partial replacement of kaolin with MWS. According to these results, all tile formulations presented plastic properties adequate for industrial processing of floor tiles [30]. It can also be seen in Table 5 that the screening residue tends to decrease (higher degree of grinding) with the addition of MWS. Despite this, all floor tile powders exhibited low value of screening residue [31], which was in line with the good comminution of the raw materials. This tends to favor the reactivity of the fine particles during the fast-firing cycle, resulting in higher densification of the floor tile pieces. The tile formulations had Hausner ratio values from 1.29 to 1.57. The effect of the incorporation of MWS was to increase the Hausner ratio, resulting in reduced flowability of the floor tile powders. This effect limits the addition of high amounts of MWS in replacement of kaolin in floor tile formulations.

Table 5. Physical parameters of the floor tile formulations.

Formulation	UPL (%)	LPL (%)	PI (%)	SR (%)	Hr
ML0	31.8	20.5	11.3	3.1	1.29
ML1	32.8	20.5	12.3	0.7	1.57
ML2	34.4	21.5	12.9	1.4	1.42
ML3	35.3	22.8	12.5	1.5	1.57

UPL-upper plastic limit; LPL-lower plastic limit; PI-plastic index; SR-screening residue; Hr-Hausner ratio.

The XRD patterns of the floor tile formulations are presented in Figure 1. The crystalline phases found in ML0 formulation (MWS-free formulation) were kaolinite ($Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$), albite ($NaAlSi_3O_8$), quartz ($SiO_2$), and micaceous mineral. For the MWS containing formulations (ML2 and ML3 formulations), diffraction peaks of gibbsite and goethite were also detected. This indicates that the replacement of kaolin with MWS into tile formulation modifies its mineralogical composition. In addition, the crystalline phases identified via XRD agree with the results of chemical composition (Table 4).

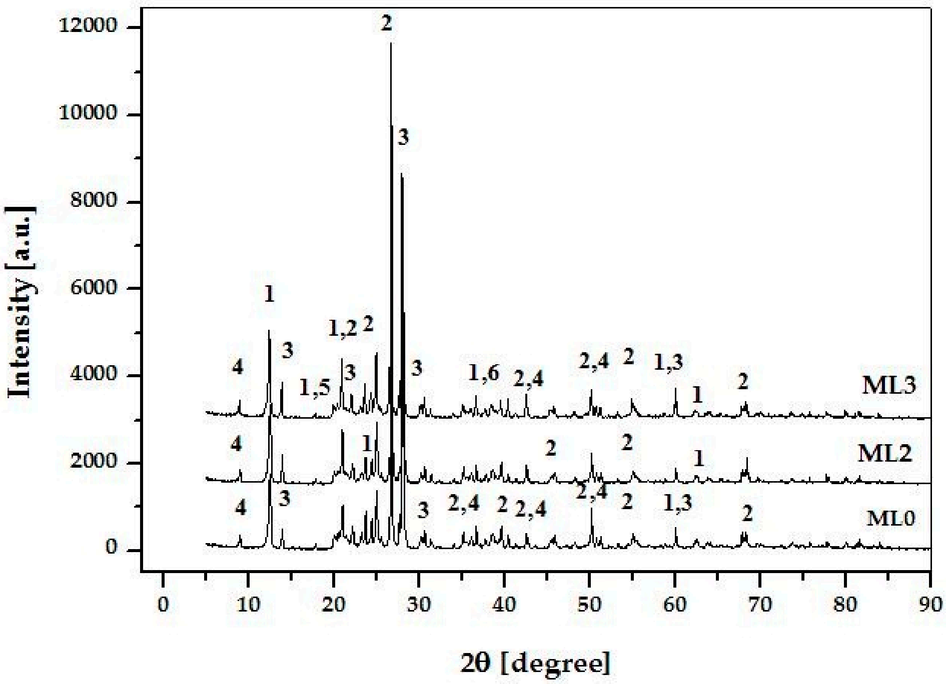


Figure 1. X-ray diffraction patterns of the floor tile formulations (1—kaolinite; 2—quartz; 3—albite; 4—mica; 5—gibbsite; 6—goethite).

Figure 2 shows the typical XRD patterns of the floor tile pieces for the ML3 formulation fired between 1190 and 1250 °C. For all firing temperatures, the main crystalline phases identified were mullite ($3Al_2O_3{\cdot}2SiO_2$) and quartz. This result is in accordance with the Na_2O-Al_2O_3-SiO_2 phase diagram [32]. Mullite found in the fired specimens results from the decomposition of kaolinite, whereas the residual quartz is related to the starting raw materials. Note that albite has been detected in small amount, as a remaining mineral unreacted during the fast-firing cycle (<60 min). In addition, there is evidence of the presence of hematite as a residual iron mineral of the MWS sample. Thus, the partial replacement of kaolin with MWS influenced the phase evolution of the floor tile formulations during the fast-firing cycle employed.

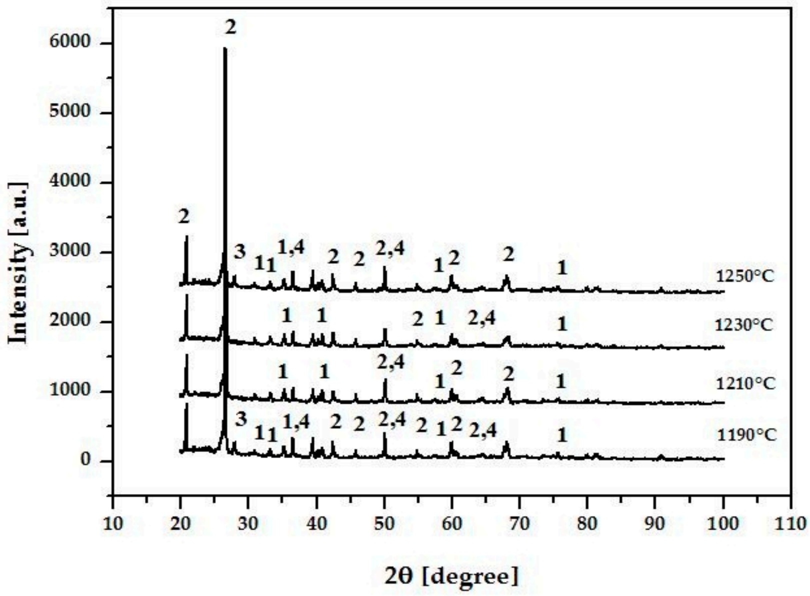
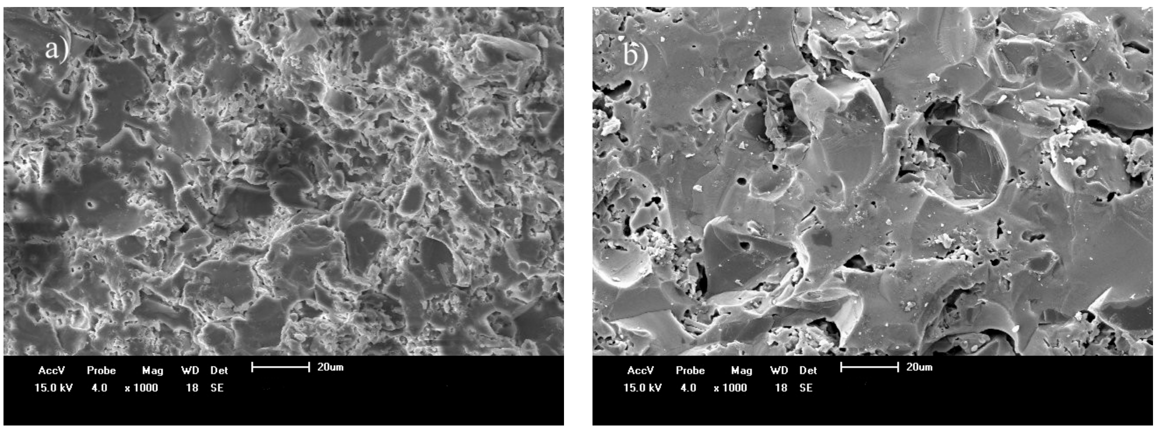


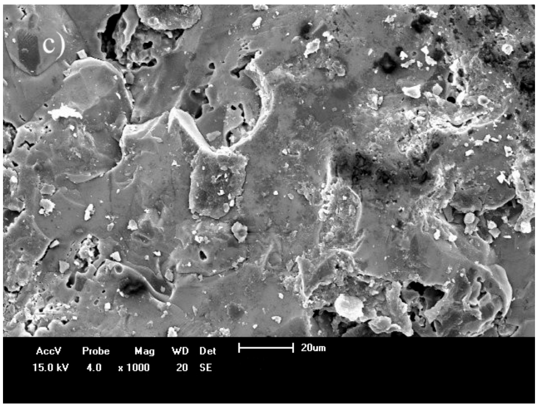
Figure 2. X-ray diffraction patterns of the ML3 formulation fired at various temperatures (1—mullite; 2—quartz; 3—albite; 4—hematite).

Figure 3 shows the fractured surfaces of the sludge bearing floor tile pieces (ML3 formulation) fired between 1190 and 1250 °C. SEM micrographs show the typical sequence of enhanced densification as firing temperature increases.

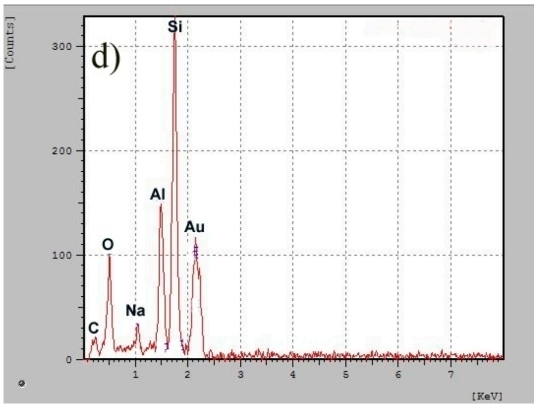


(**a**) (**b**)

Figure 3. *Cont.*



(**c**)

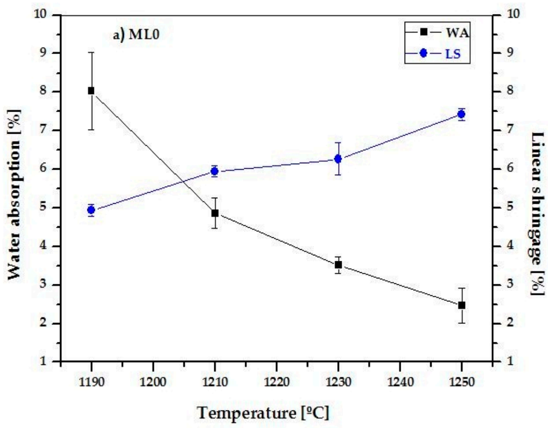


(**d**)

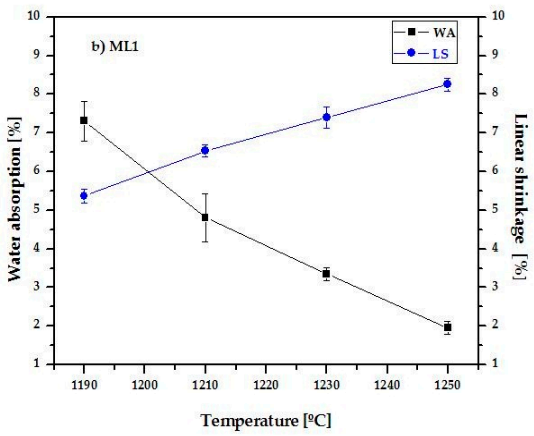
Figure 3. SEM micrographs of the fracture surfaces of the ML3 formulation fired at various temperatures: (**a**) 1190 °C; (**b**) 1230 °C; (**c**) 1250 °C; and (**d**) SEM/EDS spectrum at 1250 °C.

At 1190 °C (Figure 3a), the microstructure revealed a rough fracture surface with presence of substantial open porosity connected with dense regions (glassy phase). At 1230 °C (Figure 3b) and 1250 °C (Figure 3c), however, the open porosity was substantially reduced, an indication that the development of the glassy phase during the fast-firing cycle has continued. At higher firing temperatures, transgranular fractures can be also observed. A line spectrum for the floor tile piece fired at 1250 °C using SEM/EDS is shown in Figure 3d, where Si, Al, and Na were detected. These results are consistent with the chemical composition data (Table 4) and XRD patterns (Figure 2).

The densification behavior of the floor tile pieces during the fast-firing cycle was monitored through the gresification diagram, apparent density, and mechanical strength. The gresification diagrams of the floor tile pieces are shown in Figure 4. This diagram allows evaluating the efficiency of the sintering process during the fast-firing cycle. The densification behavior of the tile pieces was influenced by both firing temperature and added MWS amount. For all formulations, linear shrinkage increases, and water absorption (open porosity) decreases with rising firing temperature (Figure 4). This behavior can be attributed to a dominant viscous flow sintering mechanism during the firing process that closes the open porosity, resulting in higher densification [33].

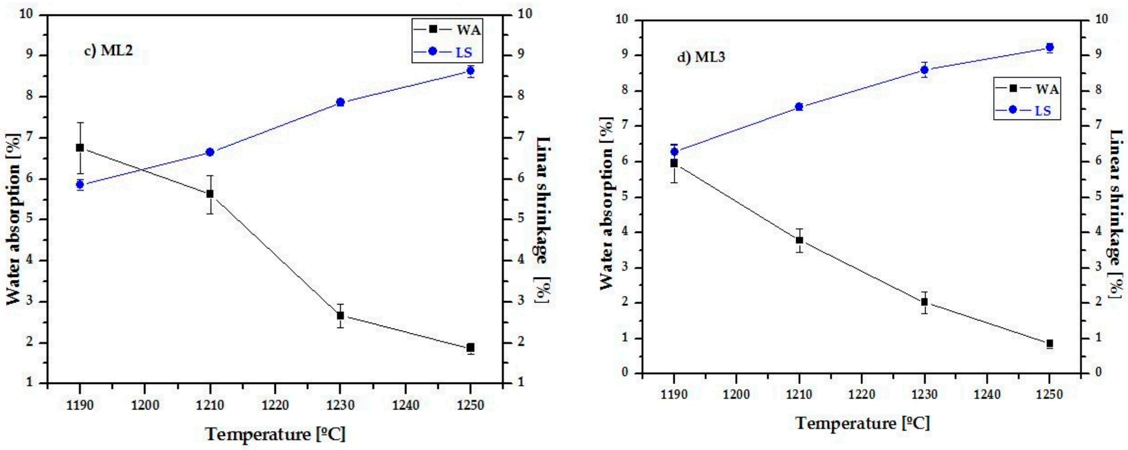


(**a**)



(**b**)

Figure 4. *Cont.*



(**c**) (**d**)

Figure 4. Gresification diagrams of the floor tile formulations: (**a**) ML0; (**b**) ML1; (**c**) ML2; and (**d**) ML3.

Linear shrinkage values between 4.94–9.23% were obtained for the floor tile pieces, indicative of good dimensional control. Note also that, at any firing temperature, the MWS-added tile pieces presented higher linear shrinkage and lower water absorption than those of the MWS-free pieces (ML0 formulation). This finding was mainly due to the presence of iron oxide (Fe_2O_3) in the MWS sample (Table 3), which acts as an auxiliary fluxing agent that lowers the viscosity of the liquid phase on firing [34,35].

The apparent density of the floor tile pieces is shown in Figure 5. The results showed that the apparent density depends on both the added MWS amount and the firing temperature. Note that the MWS-added tile pieces had higher apparent density. This increase in apparent density is in line with the chemical and physical characteristics of the MWS sample, leading to accelerated densification of the floor tile pieces. As expected, the effect of the firing temperature was to improve the densification of the tile pieces due to vitrification, independently of the added MWS amount. These results are in accordance with the gresification diagrams (Figure 4a–d).

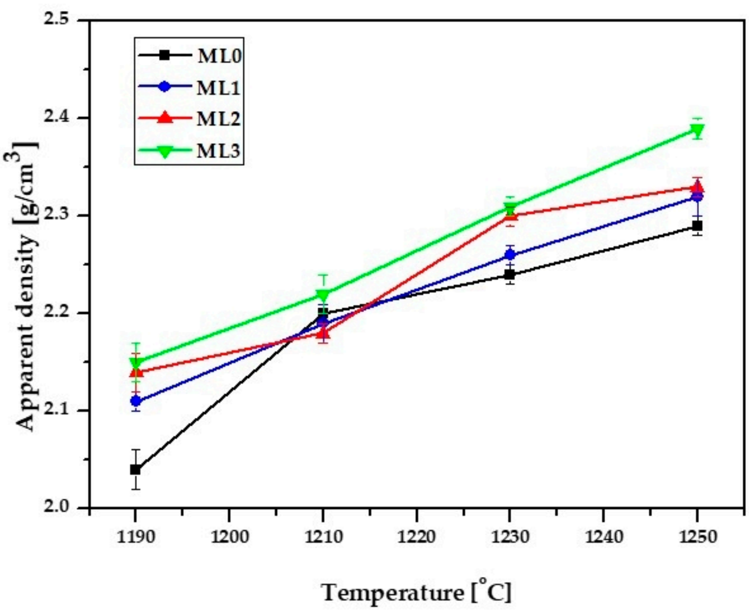


Figure 5. Apparent density as a function of MWS waste amount and firing temperature.

The flexural strength of the floor tile pieces is shown in Figure 6. The mechanical behavior is quite correlated with all the other technological properties and sintered microstructure investigated. Such data indicate a tendency to higher flexural strength with higher MWS amount. This finding was expected, given that MWS addition leads to an increase of the densification degree during the fast-firing cycle.

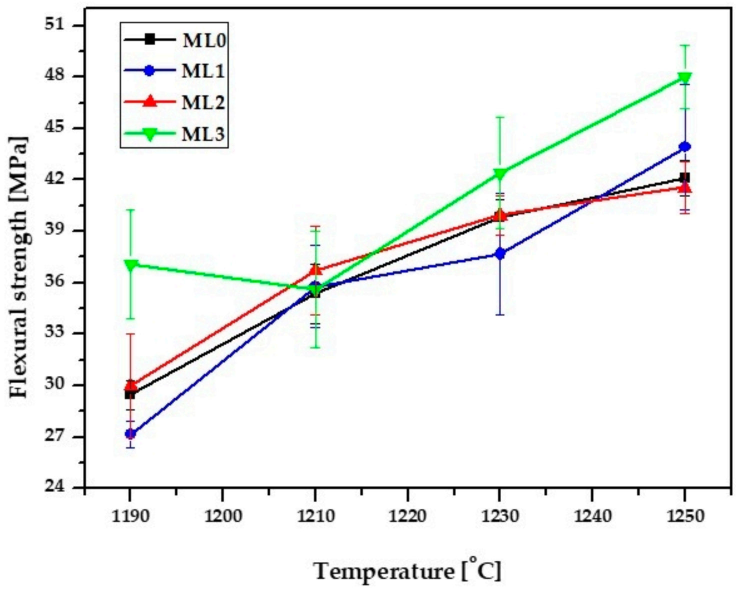


Figure 6. Flexural strength as a function of MWS waste amount and firing temperature.

From the standpoint of the ceramic industrial processing, it is very important to evaluate the feasibility of the use of MWS in the production of ceramic floor tiles. Water absorption (WA) and flexural strength (FS) are technological properties which, according to ISO 13006 standard [36], define floor tile product specification. In this study different ceramic floor tiles were obtained depending on the MWS amount and firing temperature (Table 6). It can be observed that the partial replacement of kaolin with MWS produced important changes in the floor tile quality. For example, low water absorption floor tile (ISO 13006 standard Group BIb $-$ 0.5% $<$ WA $\leq$ 3.0% and FS $\geq$ 30 MPa) could be obtained with ML2 and ML3 formulations fired at 1230 °C, while medium water absorption floor tile (ISO 13006 standard Group BIIa $-$ 3.0% $<$ WA $\leq$ 6.0% and FS $\geq$ 22 MPa) could be obtained with ML3 formulation fired at 1190 °C. These results can have significant economic and environmental benefits because of the possibility of using a shorter firing cycle (energy savings) and MWS as a renewable raw material (zero-cost or low-cost) in the processing of ceramic floor tiles.

Table 6. Classification of the ceramic floor tiles containing MWS waste according to the ISO standard 13006.

Formulation	1190 °C	1210 °C	1230 °C	1250 °C
ML0	BIIb	BIIa	BIIa	BIb
ML1	BIIb	BIIa	BIIa	BIb
ML2	BIIb	BIIa	BIb	BIb
ML3	BIIa	BIIa	BIb	BIb

4. Conclusions

The following conclusions may be drawn from the experimental results and their discussion.

- It was found that the municipal waterworks sludge used in this investigation is a renewable raw material that could partially replace natural kaolin in ceramic floor tile formulations.

- The incorporation of municipal waterworks sludge positively influenced the densification behavior and technological properties of the floor tile pieces during the fast-firing cycle. It was demonstrated that the replacement of kaolin with up to 10 wt. % of municipal waterworks sludge allowed important effects in the tile quality, such as the production of group BIb (low water absorption floor tile) and group BIIa (medium water absorption floor tile) of ISO 13006 standard at lower temperatures (energy savings).
- The processing of ceramic floor tiles could be an interesting technological solution for the valorization and final disposal of municipal waterworks sludge.
- The fact that the tile materials obtained can be considered of good technical quality opens the possibility of using municipal waterworks sludge as an alternative raw material in the production of vitrified floor tiles with a commercial value.

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Conflicts of Interest: The authors declare no conflict of interest.

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