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RESEARCH

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Red ceramics produced from mixtures of kaolinite clay and waste glass



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Abstract

Red ceramics were produced at 750°C by mixing reddish yellow kaolinite clay from Marom (West Region of Cameroon) with waste glass (percentage ranging between 0 and 15% mass). Depending on the nature of the materials, kaolinite clay, waste glass and ceramics were characterized by determination of chemical and mineralogical compositions, linear shrinkage, water absorption, flexural strength and variation of color of fired samples. Thermal analysis and Fourier transform infrared spectroscopy were done as well. The final color of ceramics was red, water absorption varied between 17.40 and 13.70%, linear shrinkage ranged between 0.70 and 1.20% and flexural strength was between 5.30 and 8.10 MPa. These results showed that mixing kaolinite clay with waste glass is an interesting process to get red ceramics destined for red bricks or roofing tiles at 750°C.

Keywords: Kaolinite clay, Waste glass, Mixtures, Red ceramics

Background

Since millennia, people have been using clay products in various forms such as earth blocks, fired bricks, roofing tiles for construction and other related uses. These materials are important to our daily life with respect to their properties which enable human beings to construct buildings all around the world according to their mechanical strength, durability, water absorption or chemical resistance (Aubert et al. 2013). Sub-Saharan Africa countries have potential sources of clay deposits for which kaolinite is generally the main mineral associated among other with quartz, goethite, hematite, anatase and alkaline oxides (Yakoubi et al. 2006; Traoré et al. 2007). These last oxides require low temperature to melt and act as binders which link particles of clay during the sintering process. However clays with low alkaline oxide and great amount of iron oxide require high temperature for maturation so as to get suitable ceramic products (Aliprandi 1979; Sei et al. 2004; Elimbi et al. 2014). It is an appeal to reduce energy consumption and to protect our environment for sustainable development (Oti and Kinuthia 2012; Sultana et al. 2015). The world over, treatment and management of wastes is crucial (Suzuki and Tanaka 1997; Sultana et al. 2015). Studies referring to addition of low quantity of certain types of waste to clays in order to manufacture reliable ceramic products such as fired bricks remain of date (Zhang 2013). Waste glass must receive careful



© 2015 Tiffo et al. This article is distributed under the terms of the Creative Commons Attribution 4.0 International License (http:// creativecommons.org/licenses/by/4.0/), which permits unrestricted use, distribution, and reproduction in any medium, provided you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. attention for environmental issues according to their non-biodegradable nature and can be recycling into new items. Waste glass can be utilized as flux to replace common fluxes such as feldspar or other mineral fluxes to save energy in ceramic manufacturing process (Bragança and Bergmann 2004; Rozenstrauha et al. 2006; Raimondo et al. 2007; Rambaldi et al. 2007; Andreola et al. 2008; Dondi et al. 2009; Djangang et al. 2014).

The present study focused on determination of physical characteristics (such as linear shrinkage, water absorption, variation of color) and flexural strength of ceramics obtained at 750°C using waste glass and reddish yellow colored kaolinite clay whose composition contains high mass percentage of iron oxide and low mass percentage of alkaline oxides. However, waste glass is abundant and available especially in urban zones as consequence of daily activities. In spite of this, in many African countries and particularly in Cameroon, there is not always specialized industries in charge of collecting, storing and reusing of waste materials, while it can provide added economical benefits if waste glass can be converted to useful materials and be recycled (Rambaldi et al. 2007; Djangang et al. 2014). Hence this can make waste glass potential secondary input for traditional ceramics production. Such formulations are typically used to manufacture building materials such as fired bricks or roofing tiles. Clays are very common raw materials of such products and glass addition is the opportunity to reuse waste glass to lower maturation temperature in order to reduce the cost of production of these ceramic products in economic plan.

Materials and experimental methods

Materials

The studied clay material labeled as K was collected from the area of Marom (West Region of Cameroon) which is located in the central domain of the PanAfrican belt of Cameroon (Nzenti et al. 1998; Ganwa et al. 2008). This area is composed of gneisses and granites partly capped by volcanic rocks of tertiary age from the "Cameroon Volcanic Line". Geology formations of the Maron area are mainly mylonites of granite compositions and clay minerals that were probably developed thanks to the strong mylonitization of the rocks that facilitated circulations of superficial waters and thus the strong alteration of granitic rocks. Once collected, blocks of reddish yellow colored clay sample were first cured at room temperature for 2 weeks then dried at 105°C for 48 h. The dried blocks were crushed and then ground in a ball mill and the resulted powder was sifted using an 80 µm mesh sieve. Colorless waste glass bottles collected from garbage cans, were broken into pieces, washed and dried at 105°C. The resulted pieces were crushed then sieved via an 80 µm mesh sieve to get colorless glass powder which was labeled as V.

Experimental methods

Four types of mixtures denoted as KV_0 , KV_1 , KV_2 and KV_3 were elaborated between powders of K and V according to mass compositions of Additional file 1: Table S1. To get a mixture, powders of K and V were homogenized in distilled water in order to get a slurry which was kept at the ambient atmosphere of the laboratory for 24 h. The mixture was completely oven-dried at 110°C for 72 h then crushed and sieved using an 80 µm sieve. For each type of mixture, two kinds of test samples were produced by extrusion: parallelepiped (82 mm × 42 mm × 9 mm) and cylindrical (13 mm diameter and 9 mm height). The obtained test specimens were cured for 48 h at ambient temperature of laboratory, oven-dried at 110°C for 48 h then fired in a kiln (Nabertherm, model LH 60/40) at 750°C at the rate of 5°C/min for two hours. The firing temperature of 750°C was chosen as a result of preliminary test which showed that the used glass powder started to melt around 700°C. The chemical analysis was carried out by Inductive Coupled Plasma-Atomic Emission Spectrometry via a Perkin Elmer-Optima 7000DV device. The crystalline phases were determined by X-ray diffraction using a Philip PW 3050/60 diffractometer which operated by reflection of $K\alpha_1$ radiation of Copper. Thermal analysis were performed thanks to a NETZSCH STA-449F3 (TG and DTA) operating at the rate of 20°C/min and an Adamel-Lomargy model DM-15 (dilatometry) which operated at the speed of 5°C/min. Fourier transform infrared spectroscopy (FTIR) was performed with the aid of a Bruker Alpha-P, operating in absorbance mode. The variation of color of fired products versus temperature was examined using the Munsell Soil Color Charts (2000). Linear shrinkage was determined on parallelepiped fired test specimens thanks to a caliper (ROCH France, Patented S.G.D.G.) and water absorption was carried out on cylindrical fired test specimens using NF-P-18-554 standard (Norme Française 1979). Flexural strength was performed according to EN-100 standard (Norme Européenne 1982) on parallelepiped fired test specimens using an electro-hydraulic press (M & O, type 11.50, and No 21) operating at an average rate of 3 mm/min.

Results and discussion

Raw materials characterization

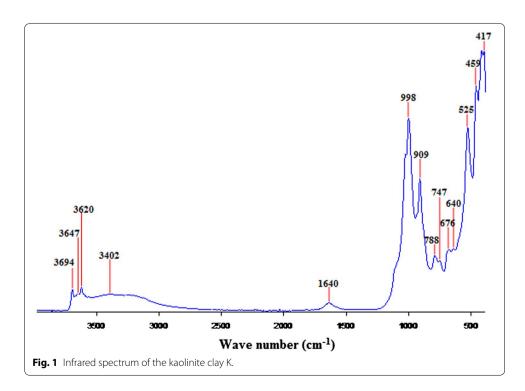
Clay sample

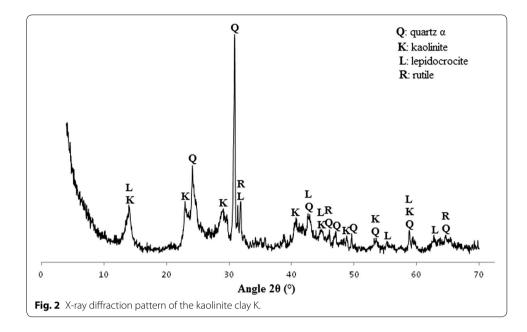
The chemical composition of the K clay is given in Additional file 2: Table S2. It appears that SiO₂ content is 44.70% mass against 18.50% mass for Al₂O₃. The molar ratio of SiO₂/ Al_2O_3 is 4.1 against 2 for pure kaolinite which allows to classify the K clay as a siliceous one (Djangang et al. 2007). The Fe_2O_3 mass percentage of 20.10 is high and this is not favorable to allow for ceramics with high mechanical values (Ergul et al. 2007; Bernhardt et al. 2014). Conversely, this amount of Fe_2O_3 in presence of uncolored waste glass is beneficial to get red colored ceramics (Karaman et al. 2006; Vieira et al. 2008; Sultana et al. 2015). This is an important technological aspect that renders possible the use of K clay for the production of ceramics with red tonality, especially for roofing and rustic floor tiles, then worthwhile for the manufacture of terra cotta. Due to the great amount of Fe_2O_3 in the K clay and presence of low content of $Na_2O + K_2O$ (3.30% mass) and CaO (0.51% mass), oxides which act as fluxes at temperatures greater than 1,000°C, the sintering of this clay material could require high temperature to get reliable ceramics. Hence using the K clay for ceramic production might need adjustment of its chemical composition (Sei et al. 2004; Arib et al. 2007; Elimbi et al. 2014; Djangang et al. 2014). This can be possible through addition of energetic fluxing agents such as sodium or potassium feldspars or waste glass (Bragança and Bergmann 2004; Rozenstrauha et al. 2006; Arib et al. 2007; Raimondo et al. 2007; Rambaldi et al. 2007; Andreola et al. 2008; Dondi et al. 2009; Djangang et al. 2014). The little amount of K_2O (2.93%) might suggest the presence of mica mineral (Vieira et al. 2008; Sultana et al. 2015). Also, the presence of 0.64% mass of TiO_2 allows expecting that the K clay might contain either rutile or anatase. Hence, in addition to the great iron oxide amount, the presence of rutile or anatase will enable to get ceramics with

red color (Chen et al. 2011; Quijorna et al. 2012; Bernhardt et al. 2014). Loss on ignition is 12.69% and this is not very far from the values commonly encountered for kaolinite clay rich materials (14.00%). In fact, the present result correlates well with the high molar ratio of SiO₂/Al₂O₃ (4.10) and the great iron oxide amount (20.10% mass) which may let to predict the presence of iron oxi-hydroxide minerals in the K clay. The FTIR spectrum of K is shown in Fig. 1. The absorption bands at 3,694-3,620 cm⁻¹ express the stretching vibrations of -OH groups of kaolinite network (Kakali et al. 2001; Bich et al. 2009; Hafid and Hajjaji 2015). The bands located at 3,402 and 1,640 $\rm cm^{-1}$ correspond respectively to stretching vibrations of water molecules while those at 998 and 788 cm^{-1} express the vibration of Si–O–Al group of the network (Kakali et al. 2001; Bich et al. 2009; Hafid and Hajjaji 2015). The bands at 909 and at 789 cm^{-1} indicate the stretching vibration of Al-OH with Al in VI coordination (Kakali et al. 2001; Bich et al. 2009; Hafid and Hajjaji 2015). The band at 525 cm⁻¹ indicates the vibration of Si–O–Si and Si–O–Al groups of the network (Kakali et al. 2001; Bich et al. 2009; Hafid and Hajjaji 2015). The crystalline phases found in K via XRD are shown in Fig. 2. The high quantity of iron oxide (20.10% mass) is in accordance with the presence of lepidocrocite. The other minerals present are kaolinite, quartz and rutile. Calculation using X-ray diffraction along with chemical analysis (Bich 2005) enabled to get the quantitative mineralogical composition of K as shown in Additional file 3: Table S3.

Waste glass

Powder of V submitted to ICP-AES analysis led to the determination of its chemical composition which is given in Additional file 4: Table S4. The powder contains high amount of silica (68.70% mass) and considerable quantity of CaO (14.30% mass) along with Na₂O (12.60% mass) which enable to classify V as soda-lime glass (Djangang et al.



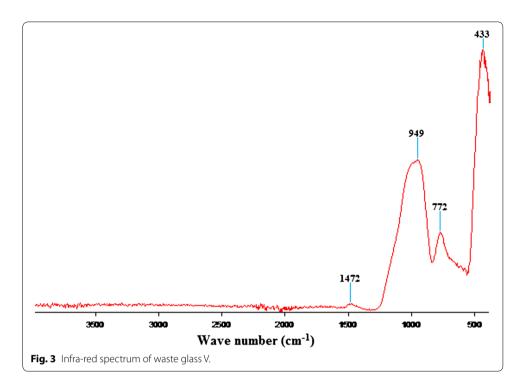


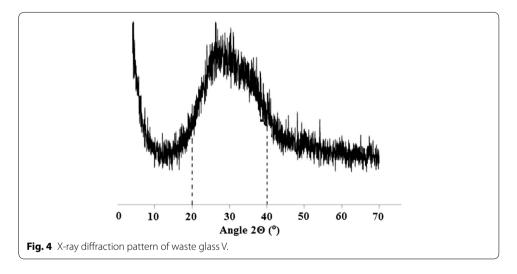
2014). Considerable amount of Na₂O + K₂O (13.35% mass) and CaO + MgO (16.63% mass) could act as energetic fluxing agent when added with the K clay powder to get mixtures enabling to make terra cotta and associated products. Since V has little amount of colorizing oxides, mixing its powder with that of K clay will result in no significant change of color of fired products (Karaman et al. 2006; Djangang et al. 2007). Conversely, the presence of the above oxides in the mixtures will induce viscous flow phase to occur at relatively low temperature, since experimentally V melts around 700°C. According to this, waste glass material can constitute a potential fluxing agent that can substitute feldspar for manufacturing ceramics. The main effect of utilizing waste glass here is then to reduce energy consumption with a view to achieve economic purpose. In addition, waste glass replaces completely feldspar since the latter is expensive and not available everywhere for effective economic consumption while the former is easily found free especially in certain urban zones (Raimondo et al. 2007; Andreola et al. 2008; Dondi et al. 2009). Figure 3 exhibits the FTIR spectrum of V. The bands respectively at 949 and 433 cm^{-1} are fingerprint of quartz (Bich et al. 2009; Tchakouté et al. 2013) while the one at 1,472 cm⁻¹ expresses the vibration of O–C–O (Elimbi et al. 2011; Hafid and Hajjaji 2015) and this could be in accordance with the raw materials commonly used for the making of soda lime-glass. Also the band at 772 $\rm cm^{-1}$ expresses the absorption of quartz inferring mark of crystalline silica within the great amount of amorphous phase of the soda-lime V (Hafid and Hajjaji 2015). This is evidence of high amount content of silica as shown in the chemical composition of V (Additional file 4: Table S4). The XRD pattern of V is shown in Fig. 4 and exhibits mainly a large dome between 20° and 40° (2 θ) which expresses great amorphous silica content (Park and Heo 2002) of V.

Characterization of mixtures and ceramics

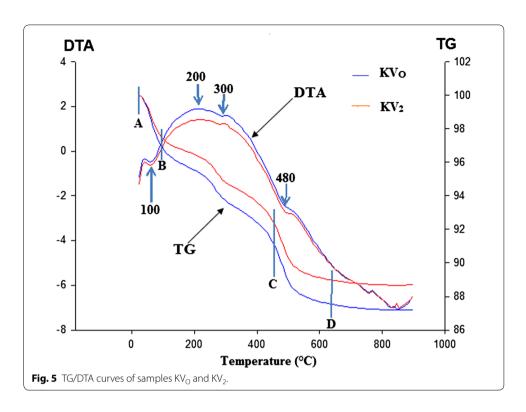
Thermal behavior and color of the mixtures

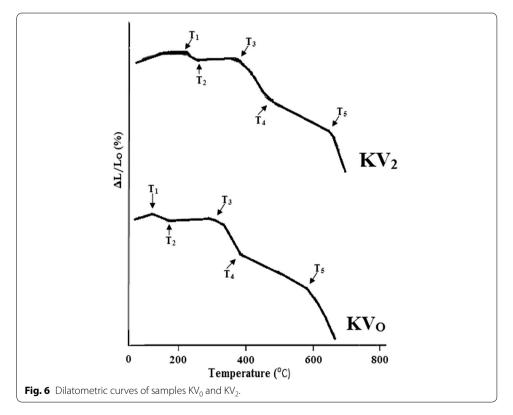
Thermal analysis (DTA, TG and dilatometry) carried out on samples KV_0 and KV_2 are presented in Figs. 5, 6. For each sample, TG curve exhibits three loss of mass (Fig. 5). The





first loss of mass corresponds to hydration water which is expressed by an endothermic peak around 100°C in DTA (Chakraborty 2003; Vieira et al. 2008; Phonphuak and Thiansem 2012). The second loss of mass is combination of exothermic and endothermic phenomena that extend between 150 and 400°C. Thus, the exothermic dome with maximum around 200°C accounts for thermal decomposition of organic matter contained in the clay (Vieira et al. 2008). This exothermic reaction is followed by endothermic phenomenon around 300°C which is linked to deshydroxylation of lepidocrocite and it is well known that its transformation above 500°C leads to hematite (Rollet and Bouaziz 1972; Vieira et al. 2008). The last loss of mass accounts for dehydroxylation of kaolinite which is transformed into metakaolinite (Kakali et al. 2001; Bich et al. 2009; Hafid

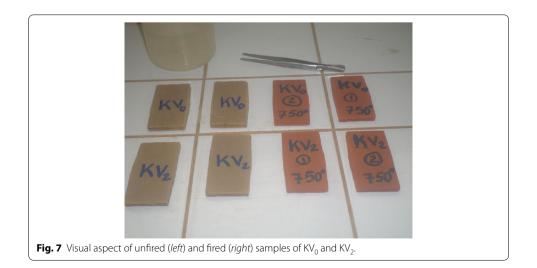




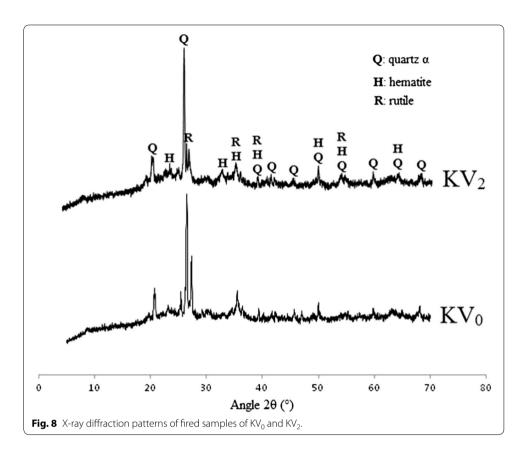
and Hajjaji 2015). The dilatometric curves of KV_0 and KV_2 are shown in Fig. 6 and each exhibits five changes whose temperatures are summarized in Additional file 5: Table S5. According to Fig. 6 and Additional file 5: Table S5, KV_0 exhibits lower temperature changes than KV_2 . This could result to the fact that up to 700°C, the waste glass does not melt but behaves as filler in the mixtures. As a result of this, temperature T_1 (elimination of hydration water) and T_2 and T_3 which may be in relation with the dehydroxylation of lepidocrocite are smaller for KV_0 than their counterpart of KV_2 . This filler behavior of waste glass is also encountered for temperatures T_4 and T_5 which are in relation with dehydroxylation of kaolinite. When unfired, all samples are reddish yellow (Additional file 6: Table S6 and Fig. 7). Fired samples are red (Additional file 6: Table S6 and Fig. 7) and this correlates well with the existence of hematite in presence of rutile in the fired samples (Fig. 8). Thus, addition of 10% mass or more of waste glass with K clay does not hinder to get red colored ceramics destined for the making of fired bricks or roofing tiles.

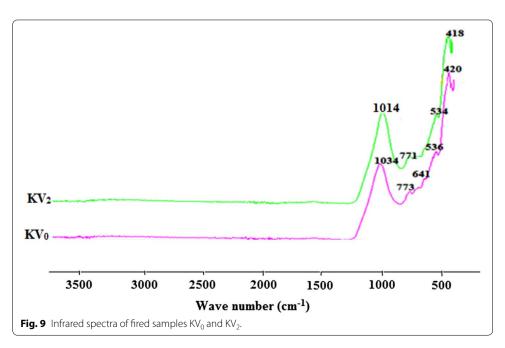
XRD and FTIR analysis

Figure 8 presents the X-ray patterns of fired products of samples KV_0 and KV_2 which provide the mineralogical phases such as quartz α , rutile and hematite. This last crystalline phase is probably a neomineral resulting from thermal deshydroxylation of lepidocrocite (Rollet and Bouaziz 1972; Vieira et al. 2008). Both rutile and hematite are the main minerals enabling to obtain red ceramics (Karaman et al. 2006; Vieira et al. 2008). Quartz is the mineral initially present in the K clay and the intensities of its main peaks (3.34 and 4.27 Å) are lower for KV_2 than for KV_0 because the incorporation of powder glass (amorphous material) in the clay diminishes its amount of sand and silt. Also, there is total transformation of kaolinite at 750°C (Elimbi et al. 2011) and this may expect to generate low swelling ceramics when put in contact with liquids such as water, hence to allow more sustainability. The absence of kaolinite in the fired products is also confirmed by FTIR spectra (Fig. 9). In fact in the fired products, the absorption band at 3,694–3,620 cm⁻¹ which expresses the stretching vibrations of –OH groups of kaolinite network (Elimbi et al. 2011) (Fig. 1) is absent (Fig. 9). Also, except for the decreasing



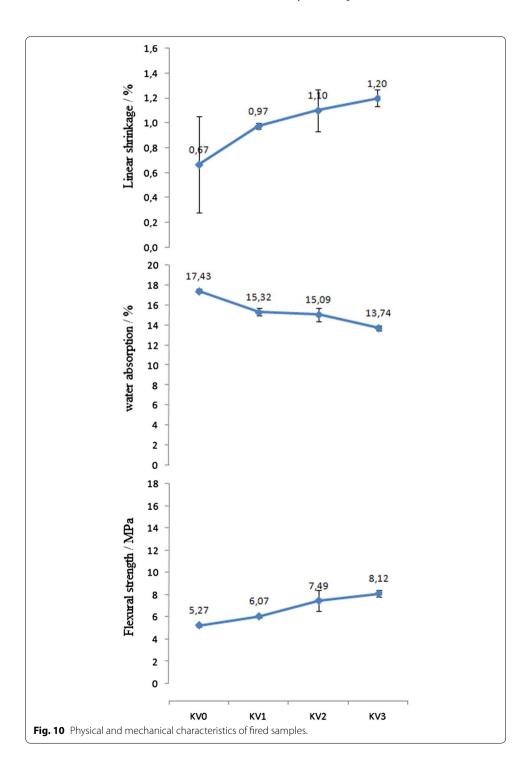
of quartz peaks, the aspects of X-ray patterns or FTIR spectra of fired products do not change significantly when mixing up to 10% of waste glass powder with the K clay (Figs. 8, 9).





Physical and mechanical properties

Linear shrinkage and water absorption and flexural strength of samples initially fired at 750°C are shown in Fig. 10. Addition of waste glass up to 15% mass leads to increase linear shrinkage from 0.70 to 1.20%. This increase correlates with the chemical transformations which take place during the heating of the samples. Among these are elimination of structural water of kaolinite, formation of new crystalline phase (hematite), dissolution



of quartz via the vitreous phase generated by the fusion of waste glass (Fig. 8). Anyhow, increase of linear shrinkage of fired products is an expression of their reactivity and this is emphasized when the additive (V) increases up to 15% mass. The water absorption percentage decreases from 17.40 to 13.70 when the mass percentage of V increases up to 15 (Fig. 10). This behavior is attributed to the vitreous phase brought about by waste glass. In fact the amount of this vitreous phase increases with increase of waste glass percentage and gradually, this viscous phase wraps up particles of the samples. Hence progressive decrease of voids within the samples leads to the decrease of water absorption percentage (Vieira et al. 2008; Phonphuak and Thiansem 2012; Pérez-Villarejo et al. 2015; Sultana et al. 2015). Flexural strength is a characteristic which is strongly dependent of voids: the lower there are voids, the higher are the values of flexural strength of fired products. As the mass percentage of V increases up to 15%, there is a noticeable tendency of increase of flexural strength of fired samples between 5.30 and 8.10 MPa. As previously mentioned, these results are in accordance with the variation of water absorption as consequence of viscous flow mechanism, inducing densification through consolidation of particles of fired samples. Vitrification then causes the reduction of porosity and therefore samples experience increase of mechanical strength (Chen et al. 2011; Emrullahoglu 2014; Bernhardt et al. 2014; Hafid and Hajjaji 2015). Thus, with respect to above mentioned, this process can be technologically applied for the production of fired bricks, roofing tiles, rustic floor or wall tiles and related materials which could be used for masonry purpose and sustainable development.

Conclusion

Red ceramics were produced by heating (750°C) samples of mixtures of reddish yellow colored kaolinite clay with waste glass (percentage ranging between 0 and 15% mass). The chemical analysis of the clay revealed both low content of alkaline oxides and rich amount of ferrous oxide. The mineralogical characterization of the clay revealed the presence of minerals such as kaolinite, quartz α , rutile, lepidocrocite and others unconfirmed phases attributed particularly to organic matter as confirmed by thermal analysis. Waste glass can be considered as potential fluxing additive that can substitute feldspars in terra cotta for low heating temperature production. Hence increasing the mass percentage of waste glass in mixtures with kaolinite clay leads to fired products whose linear shrinkage and flexural strength increase while there is decrease of water absorption. This is attributed to the densification of samples via the viscous flow mechanism induced by a vitreous phase brought about by the fusion of waste glass. The obtained results show that mixing waste glass with poorly alkaline oxide and abundant ferrous oxide kaolinite clay content is an interesting process to get red ceramics destined for the making of bricks or roofing tiles at 750°C.

Additional files

Additional file 1: Table S1. Composition of the mixtures of K and V expressed as mass percentage.
Additional file 2: Table S2. Chemical compositions of the kaolinite clay K (LOI = Loss On Ignition).
Additional file 3: Table S3. Mineralogical composition of the kaolinite clay K.
Additional file 4: Table S4. Chemical composition of waste glass V (LOI = Loss On Ignition).
Additional file 5: Table S5. Temperature changes on the dilatometry curves of KV₀ and KV₂.
Additional file 6: Table S6. Variation of color of samples with firing temperature.

Abbreviations

DTA: differential thermal analysis; EN: European norm; FTIR: fourier transform infrared spectroscopy; ICP-AES: inductive coupled plasma-atomic emission spectrometry; K: kaolinite clay; KV: mixture of Kaolinite clay and glass powder; NF: Norme Française; S.G.D.G: Sans Garantie Du Gouvernement; TG: thermogravimetry; V: glass powder; XRD: X-ray diffraction.

Authors' contributions

ET and AE participated in the research process. JDM helped for the determination of different crystalline phases on the DRX patterns and for grinding and sieving the raw materials (both clay and waste glass). ABT helped to collect the raw materials and to achieve thermal analysis. All authors read and approved the final manuscript.

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Compliance with ethical guidelines

Competing interests

The authors declare that they have no competing interests.

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