Research paper

Structure, microstructure and mechanical features of ceramic products of clay and non-plastic clay mixtures from Tunisia

Hiba Zouaoui [a,*], Gisele Laure Lecomte-Nana [b], Mohamed Krichen [a], Jamel Bouaziz [a]

[a] LCI: Laboratory of Industrial Chemistry, National School of Engineering, Box 1173, Sfax University, Sfax 3038, Tunisia
[b] ENSCI, SPCTS: Laboratory of Science of Ceramic Processing and Surface Treatments, European Ceramic Center (UMR CNRS 7315), F-87000 Limoges, France

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ABSTRACT

Five clay and non-plastic mineral mixtures (M1, M2, M3, M4 and M5) from Tunisia were investigated for their possible ceramic applications. X-ray diffraction (XRD), thermal analysis (DTA/TG), dilatometry, scanning electron microscopy (SEM) and physical-mechanical analyses were used to assess the phase evolution and microstructure of the mixtures sintered between 900 and 1150 °C. The decrease of water absorption and the increase of the bending strength of the samples were due to the formation of both the anorthite and mullite phases and the glassy phase helping to fill the pores. M1 and M2 had the highest values of bending strength at 1150 °C. According to the European Norm EN 14411, these mixtures are suggested for porcelainized and fully vitrified stoneware thanks to their low water absorption (0.09 and 0.2%, respectively). M3, M4 and M5 mixtures (with respective water absorption of 2.74, 1.68 and 1.74%) are suitable for the production of vitrified stoneware.

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1. Introduction

The clayey materials are exploited worldwide in the production of diversified ceramic products for building construction (Nkoumbou et al., 2009) and have been extensively studied (Jamoussi, 2002). Clays are used in the manufacture of many traditional ceramics. Kaolinite illite is the most extensively used clay mixture in the ceramic industry (Aydin, 2004). A ceramic product is commonly produced from a mixture of raw materials containing clay, flux and a refractory filler (Jamoussi, 2002).

Works on clayey materials are of growing interest in Tunisia and several Tunisian localities have been investigated. Particular attention should be paid to the exploitation of raw materials in Wislatiya region for ceramic products. Mohamed Krichen et al. (Krichen et al., 2008) revealed that these clay raw materials seemed to be relatively poor in phyllosilicates (46 mass%) and rich in quartz (30 mass%), carbonates (10 mass%) and goethite (7 mass%). The study of the sintering behavior of this clay at 1150 °C shows the presence of an important porosity. Because of its lower bulk density value, the clay should be amended with non-plastic materials such as sand and chamotte to obtain the required ceramic product performance.

The aim of the present research was to evaluate the ceramic properties of Tunisian clay from Wislatiya region mixed with non-plastic minerals (sand, waste brick and chamotte) in order to improve their physical and mechanical properties. The possible ceramic applications of five clay and non-plastic minerals mixtures (M1, M2, M3, M4 and M5) from Tunisia were investigated. The relationship between bending strength, bulk density, linear shrinkage and water absorption according to the sintering temperature and the nature and the amount of the non-plastic clay materials is examined in order to optimize the production process and enhance the quality of the final ceramic products.

2. Experimental

2.1. Materials

The studied clay was selected from Wislatiya (Central Tunisia); it mainly contains illite (25 mass%), montmorillonite (13 mass%), kaolinite (8 mass%), quartz (30 mass%), calcite (8 mass%), dolomite (2 mass%) and goethite (7 mass%) (Krichen et al., 2008). The non-plastic constituents are waste brick, chamotte and sand, appointed later as WB, CH and S, respectively. The chemical composition, determined by ICP analysis, is summarized in Table 1. The major elements, assumed as oxides, present in WB are SiO_2, Fe_2O_3 and Al_2O_3. The amounts of CaO and K_2O are higher than those of MgO, Na_2O and P_2O_5 (in relatively minor amounts). The chemical composition of sand shows that the major constituent is SiO2 as expected, while Na2O appears as the main associated oxide. The major elements present in CH are Al_2O_3 and SiO_2.

The XRD diagrams of the non-plastic constituents are represented in Fig. 1. The characteristic peaks of the minerals that have been identified are indicated on the diagram. Waste brick is mainly constituted of anorthite, hematite and quartz. It is seen that all sand diffraction peaks can

Table 1
Chemical composition (mass%) of non-plastic clay.

Oxides	Al$_2$O$_3$	SiO$_2$	K$_2$O	MgO	CaO	Fe$_2$O$_3$	Na$_2$O	SO$_2$	P$_2$O$_5$	L.O.I*	Total
WB	12.79	59.36	4.03	1.15	4.79	12.87	1.41	0.48	1.1	2	99.98
CH	34.65	62.53	1	–	–	–	0.68	0.35	0.32	0.43	99.96
S	0.93	94.58	0.29	–	–	–	3.81	–	–	0.36	99.97

L.O.I*: loss of ignition at 1200 °C.

be identified as a quartz phase as there is no other detectable phase. Chamotte is mainly made up of mullite, cristobalite and quartz.

2.2. Preparation of mixtures and ceramic products

Initially, the raw materials were crushed and ground to pass through a 63 μm-diameter sieve. The obtained powders were heated for two hours at 350 °C for the organic complete combustion. According to the great amount of quartz already present in the starting clay, the following composition has been investigated: 70 mass% clay with 30 mass% of non-plastic mineral materials. Table 2 summarizes the composition of the various samples. After weighting, the mixtures were homogenized by humid way according to the following experimental protocol:

- The powders were dispersed in distilled water by mechanical agitation for 5 min;
- The water of the got suspension was evaporated progressively under continuous agitation at 110 °C until getting a dough;
- The dough was dried at 110 °C during 24 h;
- The homogeneous mixture clods were finally ground using a mortar.

The obtained powders were dried pressed at 150 MPa. Cylindrical pellets of 20 mm diameter and 5 mm thickness were produced with dimensions suitable for the Brazilian test. The specimens were then fired in an electric furnace between 900 and 1150 °C using heating and cooling rates of 10 °C/min and 12 min soaking time at maximum temperature.

2.3. Methods

A Perkin Elner 8300 DV ICP-AES plasma torch was used to measure the major and trace elements in the sample. The sample was previously dissolved within an acidic solution using a microwave device under appropriate pressure and temperature cycles. X-ray diffraction (XRD) diagrams were obtained on powdered samples with a "D8 Bruker Advance" powder diffractometer using Kα1 radiation of Cu (λ = 0.15406 nm). XDR experiments were achieved in step-scan mode from 15° to 55° (2θ). Quantification of the different phases was carried out using the computer program X′-pert High Score. The differential thermal (DTA) and thermogravimetric (TG) analyses were performed
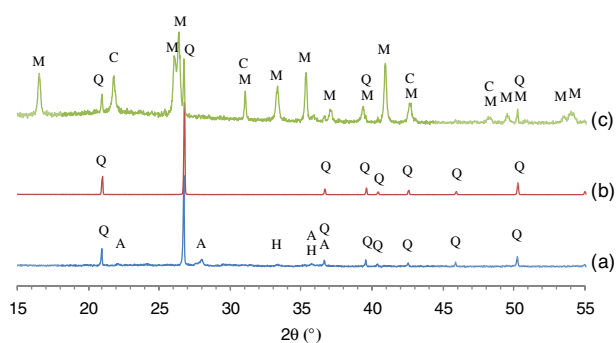
using a SETARAM Setsys 24 apparatus, operating under a constant dry air flow at a heating rate of 10 °C/min between ambient temperature and 1200 °C. Previously fired alumina was used as the reference material for all experiments. Each analysis was performed using an aliquot of 64 mg of the relevant powdered sample. Linear shrinkage was determined with dilatometry using a SETARAM Setsoft 2000 apparatus at a heating rate of 10 °C/min between ambient temperature and 1150 °C, under dry air atmosphere of samples (6 mm diameter and 5 mm thickness) pressed (25 MPa) from the powders (fraction < 63 μm). The SEM was performed on the flat polished surface to ensure investigation on the real bulk structure of the products. The microstructure was analyzed using the CAMBRIDGE S260 Scanning Electron Microscopy (SEM) working with 15 kV accelerating voltage. The samples were coated with a gold layer for more electronic conductivity.

The mechanical properties of the sintered samples were assessed using a compressive test. The Brazilian test was used to measure the bending strength (or stress to strength) of the sintered compacts (ISRM, 1978; ASTM, 1984). Brazilian tests or splitting tests consist of crushing a bedded cylindrical specimen between two crushing plates (ISRM, 1978; ASTM, 1984). The usual way of evaluating the bending strength (MPa) from the diametrical compression test is through the following equation (Elliott, 1994):

$$\sigma_r = (2 * P)/(\pi * D * t)$$

where P is the maximum applied load (N), D the diameter (mm) and t the thickness of the sample (mm).

The bending strength is calculated on the assumption that failure is initiated at the point of maximum tensile stress (Fairhurst, 1964). The influence of the sample diameter, D, and the thickness, t, on the failure stress was studied for compacted powder (Dorémus et al., 2001). According to this study, the ratio t/D ≤ 0.25 ensures plane stress conditions. The bending strength was determined (4 pellets for each experimental point) using Lloyd EZ50 apparatus.

3. Results and discussion

3.1. Thermal behavior and phase evolution

Fig. 2 illustrates the DTA-TG analysis curves of M1, M2, M3, M4 and M5. The interpretation of these DTA-TG curves leads to the following results: the endothermic peaks near 100 °C and 160 °C are related to the removal of adsorbed and interlayer water (Baran et al., 2001). This is confirmed by the first mass loss event (~1 mass%). The minor endothermic effect which occurred at 300 °C is associated with goethite (Rollet and Bouaziz, 1972) as already mentioned. The dehydroxylation of kaolinite, illite (first step) (Gualtieri and Ferrari, 2006) and montmorillonite

Fig. 1. XRD diffractograms of the waste brick (a), the sand (b) and the chamotte (c). Legend: Q: quartz, A: anorthite, H: hematite, M: mullite and C: cristobalite.

Table 2
Sample composition (mass%).

Sample label	Clay	WB	S	CH
M1	70	30	0	0
M2	70	15	15	0
M3	70	0	30	0
M4	70	0	0	30
M5	70	0	15	15