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Research paper

Production of clay ceramics using agricultural wastes: Study of properties, energy savings and environmental indicators

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ABSTRACT

In this study, agro-wastes were used as additive raw materials for the production of fired clay ceramics. The objectives of this study are to evaluate the impact of adding agro-wastes into clay body on the thermal and mechanical properties of ceramic materials, to determine the net energy consumption and to determine gas emissions during firing process. The clay and agro-wastes were characterized by chemical elemental analysis, thermogravimetric and differential thermal analysis (TGA-DTA). The fired clay ceramics were produced with clay and optimal proportions of wheat straw (WS) and olive core flour (OCF). The thermal and mechanical properties were evaluated by measuring thermal conductivity with hot-disk method and bending test respectively. The results showed that for clay incorporated OCF (4, 8 wt%) and WS (3, 7 wt%), thermal conductivity was decreased by 16 to 30%. However, the mechanical strength of the same samples has slightly decreased respectively. TGA-DTA provided an approach to estimate the heat required or released for both clay and agro-wastes thermal decomposition. The addition of agro-wastes into the clay body showed that energy consumption of fired clay ceramics production decreased to above 36% for clay incorporated 4 wt% OCF (C-4wt %OCF). The energy saving during the firing process was a tangible outcome. In order to determine the impact of the agro-wastes addition, the environmental indicators were discussed for the clay incorporated WS and OCF respectively. Total gas yield released were measured by Micro-GC after the combustion of clay incorporated OCF and WS in fixed bed reactor respectively. The analysis of gas emissions are related to the combustion of organic and inorganic compounds of agro-wastes and clay body, respectively. The CO_2 emissions coming from the combustion of agro-wastes reached up to 4.38% for C-8wt%OCF. However, the CO_2 emissions associated with decarbonatation of clay body decreased. Adding agro-wastes into the clay body results to improving thermal properties without negative impact on the mechanical properties of ceramic materials, also to a significant energy saving and decreasing of the inorganic CO_2 emissions related to the decarbonatation of clay body. The relevance of this work, pointed out in the data presented in regards to the state-of-the art is that the paper is focused on fired clay ceramic properties, on energy savings and on the evaluation of environmental indicators in the laboratory scale.

1. Introduction

Several scientific research have been carried out on the study of the physical and chemical properties of ceramic materials in their production process in order to optimize their final properties (Cultrone et al., 2004a; Vassilev et al., 2013). Number of studies have investigated the use of industrial and organic wastes to produce ceramic material such as bricks (Demir, 2006; Muñoz Velasco et al., 2014; Raut et al., 2011) including sewage sludge, fly ash and organic residues from agro-industries paper or wood. (Demir, 2006) mixed Processed Waste Tea (PWT) with clay body to improve the insulation ability of bricks by generating porosity. This author noticed also that the compressive strength of ceramic materials increased significantly by adding PWT into the clay body. Also, the microstructure of clay and additives, granulometry and the contents of additives were studied to develop the optimal formulations (Faria et al., 2012; Madurwar et al., 2013).

The consumption of earth-based materials such as clay and sand in the production of bricks resulted in resources depletion, environmental degradation and energy consumption. The biomass as an agricultural product has been used as an energy source to lower energy consumption in brick manufacturing process. The application of these agro-wastes for the production of ceramic materials is consistent with the concept of improving the efficiency of management and the environmental impact of wastes (Zabalza Bribián et al., 2011). From a

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microstructural point of view, the addition of organic residues in clay raw materials induces the pore formation. This investigation influences the final properties of fired clay bricks (Freyburg and Schwarz, 2007; Gualtieri et al., 2010). On the one hand the thermal properties were determined by the agro-wastes insulating behavior and on the other hand the mechanical properties are strongly related to the particular morphology of the pores induced in the fired clay bricks (Bories et al., 2014; Sutcu and Akkurt, 2009).

It is well known that material production industries have been recognized as one of the largest fuel consuming sectors of the economy (Koroneos and Dompros, 2007). The energy cost for such large scale production in the ceramic industry has become a critical issue. The agro-wastes incorporation within clay bricks can contribute to energy savings by reducing the energy requirements of the brick firing industrial scale. Few studies were focused on the effect of the amount and the type of agro-wastes incorporated in fired clay bricks, their behavior during combustion in the firing stage and their impact on the energy consumption of the process. Very little researches pay attention to develop an eco-friendly fired bricks in an economical and environmental stand point (González et al., 2011; Quijorna et al., 2012).

The relevance of this work, in regards to the state-of-the art is that the paper is focused on mechanical and thermal properties together with the investigation of energy savings and environmental indicators of materials in the laboratory scale. The thermal and mechanical properties were obtained by standards methods and the energy consumption occurring in the firing stage and their relationships with the chemical structure of biomass components (cellulose, hemicellulose and lignin) were discussed. In the other case, the releasing behaviors of CO_2 and total gas emissions were measured using Micro-GC. Regarding CO_2, we have been able to discriminate the fate released from inorganic source such as $CaCO_3$ with the one released from organic source from agro-wastes.

2. Materials and methods

2.1. Characterization of brick raw materials

The agro-wastes release heat during combustion which depends on its composition. Elemental analysis was carried out using CHNS of respectively olive core flour (OCF) presenting spherical particles of 50 µm and wheat straw (WS) presenting a lamellar structure with 1 mm particle sizes. In fact, a flash combustion of samples followed by chromatographic separation provides the amount of carbon, hydrogen, nitrogen and sulfur respectively.

The high heating value (HHV) of agro-wastes was determined by using a calorimeter bomb C500 CONTROL IKA. The combustion process was realized under pressure of 30 bars of oxygen at 25 °C using 1 g of samples. According to the standard NF EN 14918 (NF EN 14 908, 2010), the calorimeter bomb used had an internal volume of 250 mL. In order to avoid any acid gas released during combustion, 10 mL of 0.05 mol/L Na_2CO_3 (aq) was added to the combustion bomb. The HHV was then determined at reference temperature of 25 °C.

The raw clay used in this study was supplied by TERREAL (Castelnaudary, France). The chemical analysis of the raw clay was considered. The final properties of fired bricks are strongly influenced by the chemical and mineralogical compositions of raw clay. The chemical composition was determined by Inductively Coupled Plasma – Optical Emission Spectrometry (ICP-OES). The clay samples were immersed in a hydrofluoric acid solution (48 wt%) and perchloric acid (64 wt%), in respective proportions of 75 and 25%.vol, then heated up to a temperature of 80 °C for 30 min.

The qualitative determination of major crystalline mineralogical phases present in the clay was achieved by X-ray diffractometer using the PanAlytical X'Pert Philips (45 mA, 40 kV). The determination of crystalline phases of clay was realized at 600 °C, 700 °C, 800 °C and 900 °C using a copper anticathode (λcopper = 1,54 Å) for 2 θ angles ranging from 4 to 75° in steps of 0.017° and every 29 s. The patterns were exploited by using software X'Pert HighScore based on ICDD database (International Centre for Diffraction Data) to identify the peaks corresponding to each component. The main databases used were PDF2 (Predicted Powder Diffraction Database) and COD (Crystallography Open Database). The XRD analysis was completed by a semi-quantitative study using a calibration method namely Reference Intensity Ratio (RIR) method. The PDF2 and COD database were used also to quantify the crystalline phases.

To understand the decomposition of OCF, WS and raw clay respectively, the thermal behavior was determined by thermogravimetric and differential thermal analyses (TGA-DTA) using a TG-ATD 92 by SETARAM equipment under air atmosphere. Samples of about 100–200 mg were placed in a platinum crucible, and heated up at 5 °C/min from the room temperature to 1000 °C. The energy of transformation associated with the decomposition of OCF, WS and clay body was determined respectively by the enthalpies estimation method.

2.2. Samples preparation

Olive core flour (OCF) and wheat straw (WS) presenting respectively particles of 50 µm and 1 mm, were incorporated in clay body of 100 µm. The raw clay, sand, WS and OCF were then mixed together by a laboratory-type blender. A quantity of water (up to 22 wt% depending on the formula) was added to the mixtures Clay/OCF or WS to obtain 7.5–8.0 bars pressure of extrusion. Four formulations were prepared using 3, 7 wt% of WS and 4, 8 wt% of OCF respectively.

The samples were then extruded and shaped as molds of 180 mm × 80 mm × 18 mm. In order to avoid some defaults such as cracks or problems of efflorescence, the drying process were carried out at a high relative humidity (RH) of 25% at 105 °C for 24 h and terminated with a relative humidity of 5%. The shaped samples without waste (20 wt% sand + 80 wt% raw clay) namely clay body were designed as C and for the mixtures C-X wt%OCF or C-X wt%WS, where X indicates the amount in the clay body.

2.3. Characterization of the bricks

A thermal property, such as the thermal conductivity (λ) was measured by hot-disk method with TPS 2500 apparatus. The samples of bricks were cut from a single green body extruded to the dimensions of 30 mm × 30 mm × 5 mm. The measurements were done after a firing process at 920 °C with a constant heating rate of 5 °C/min. For each fired sample, the parallel faces were polished to have a performant contact with the Transient plane source (TPS) sensor.

The bending test was performed to investigate the load bearing capacity of fired clay brick using agro-wastes. The extruded samples were cut to the dimensions of 60 mm × 15 mm × 5 mm. The samples were heated up to 920 °C (final kiln temperature) with a constant heating rate of 5 °C/min and then cooled at room temperature respectively. A three point bending test was performed on fired samples using the mechanical test machine INSTRON 500 N with a constant rate of 1 mm/min until the failure of samples. The fired-bending stress was obtained with the Eq. ((1):

$$\sigma = (3 \cdot Fmax \cdot L)/(2 \cdot e\hat{}2 \cdot l) \quad (1)$$

With F_{max} is the force, L is the loading span, l is the breadth and e is the height of the samples.

The thermal behavior was determined by thermogravimetric and differential thermal analyses (TGA-DTA) using a SETARAM equipment under air atmosphere. Samples of about 100–200 mg were placed in a platinum crucible, and heated up to 1000 °C with a heating rate of 5 °C/min.

The released gas from fired samples in the fixed bed reactor named Carbolite was measured using Micro-GC. On the one hand the gas emissions related to decomposition of WS and OCF were measured