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Research paper

Investigation of using bottom or fly pine-olive pruning ash to produce environmental friendly ceramic materials

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ABSTRACT

The present study investigates the feasibility of using fly or bottom biomass ash as a partial replacement in the production of eco-friendly construction materials. The fly and bottom ashes from combustion of biomass (olive pruning and pine pruning) generated in a Spanish Power Plant were used as raw materials to replace clay for the production of fired bricks. First, the mineralogical and chemical composition and thermal behavior of the ashes and clay were determined. Next, ceramic bricks were manufactured by compressing clay and different quantities of fly or bottom biomass (0–30 wt%). Different firing temperatures, 900 and 1000 °C, were studied. The effect of adding fly or bottom biomass ash on the technological behavior of the brick was assessed by studying linear shrinkage, water absorption, bulk density, compressive strength, thermal conductivity and the morphology. The results have shown that the optimum sintering temperature was 1000 °C. The increase in the firing temperature from 900 to 1000 °C, raised the compressive strength and decreased the water absorption. The addition of 30 wt% of bottom or fly biomass ash produced bricks with very high water absorption values and low mechanical properties. Based on the results obtained, the optimum amounts of bottom or fly ash were 20 wt%. These percentages produced bricks whose mechanical properties were suitable and thermal conductivity decreased by 21% respect to the standard bricks. The addition of bottom biomass ash produced bricks with slightly better properties, due to the most appropriate composition of these ashes. The bricks do not present environmental problems according to the leaching study. Therefore, fly and bottom biomass ash from the combustion of olive-pine pruning could be used as secondary raw materials in ceramic brick production.

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1. Introduction

Biomass can be defined as organic matter of animal or vegetal origin, or from any processing thereof, natural or artificial (Hidalgo, 2012). The potential of biomass available in Spain besieges around 88 million tons of primary biomass into green, including remnants of forests, agricultural waste, existing untapped mass and energy crops to implement. In addition to this potential >12 million tons of dry secondary biomass are obtained from forestry industry wastes. The main application of biomass lies in its use as alternative fuel. As a consequence, converting this waste into energy increases the value of these waste materials, reduces the environmental impact of disposal and helps to fight the threat of global warming. In Spain the production of energy from biomass has increased activity, presenting a few good alternatives to growth according to the Plan of promotion of the renewable energies (APPA, 2013). Today the consumption of biomass in Spain supposed 3565 ktep heat consumption. It quantifies a total installed capacity of 533 MW supplied

with waste from agricultural crops and agroforestry industries mainly. In Andalusia (Spain) due to its wide forest and farmland surface, biomass is the renewable energy source that most contributes to the energy system, there are 14 plants with a capacity to produce 66.48 kton of pellets constituted primarily of olives pruning scraps and forest residues (Andalusian Energy Agency, 2016). Likewise, Andalusia with 18 generators of electricity from waste biomass, is the autonomous community that records higher consumption of biomass in Spain and one that has higher production potential.

Biomass is promoted by government in Spain and Europe, because sustainably-produced biomass will reduce the net flow of CO₂ to the atmosphere. Biomass is made up of organic matter and inorganic in different proportions depending on its origin and is transformed during the combustion process resulting in the formation of gases as well as obtaining a solid residue formed by fly ash and slag or bottom ash. These wastes lead to an important environmental and economic problem.

Bottom ash includes the coarse fraction of ash produced in the grill, bottom chamber and the primary combustion chamber. Often it contains mineral impurities present in biomass fuel such as sand, stones

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or mud. Fly ash is the finest fraction of ashes, dragged by exhaust fumes, which is collected and precipitates in filters.

The management of the ashes produced in significant amounts in all over the world is becoming a challenge. There has been an increasing number of studies concerning the characterization and the establishing of suitable processes in which ashes can be efficiently reused. There are different areas of application of fly and bottom ashes, depending on their properties, in sectors such as cement, ceramic, paint, plastic, agriculture, environment and construction (Abd Elrahman and Hillemeier, 2014; Ahmaruzzaman, 2010; Baykal and Döven, 2000; Carrasco et al., 2014; Cultrone and Sebastián, 2009; Donatello and Cheeseman, 2013; Hinojosa et al., 2014; Iyer and Scott, 2001; Kumar et al., 2007; Leiva et al., 2016; Li et al., 2016; Ruiz-Santaquiteria et al., 2013; Sena da Fonseca et al., 2015; Sua-lam and Makul, 2015; Yu Zhuang et al., 2016; Zhang, 2014), but the vast majority of materials generated each year are held in ash dams or similar dumps.

Considering the large quantities of raw materials needed for ceramic production (Zhang, 2014), approximately a medium size ceramic factory uses 500 ton of raw materials per day (Leiva et al., 2016), conventional clay bricks is an appropriate application for the ash usage. Ash presents a composition in oxides mainly in the system $\text{SiO}_2 - \text{Al}_2\text{O}_3 - \text{CaO}$, which can also be accompanied by other metal oxides. It is, therefore, of a composition similar to ceramic raw materials. In addition, due to the growing environmental awareness in the building industry, there is a need to investigate the way of incorporating residuals into traditional construction materials while requirements stipulated in the standards have to be maintained.

A diverse number of studies show the incorporation of ash replacing, in part or completely, traditional construction materials in order to make bricks with different properties. Ash from different sources has been used with satisfactory results, for instance: rice husk ash (Rauta et al., 2013); municipal solid waste incineration fly ash (Zhang et al., 2011); fly ash from coal burning power plants (Naganathan et al., 2015; Çiçek and Çiçin, 2015); sewage sludge incinerator ash (Cheeseman et al., 2003); olive pomace bottom ash (Eliche-Quesada and Leite-Costa, 2016) and sugarcane bagasse ash and rice husk ash (Kazmi et al., 2016).

The present work aims at providing further knowledge in the properties and behavior of bricks made of clay and biomass combustion bottom or fly pine-olive pruning ash (BBA and FBA). For this purpose, raw materials were characterized and clay-ash mixtures with 0–30 wt% of BBA or FBA were conformed. Green samples were fired at 900 °C and 1000 °C and the effect of sintering temperature and type and amount of ash were studied in terms of crystalline phases as well as physical, mechanical, thermal and environmental properties.

2. Experimental

2.1. Preparation of the samples

Clay was supplied by a clay pit located in Bailen, Jaen (Spain). It was obtained by mixing three types of raw clay in the following percentages: 30 wt% red, 30 wt% yellow and 40 wt% black clay. To obtain uniform particle size, the clay was crushed and ground to yield a powder with a particle size suitable to pass through a 500 μm sieve. The fly and bottom biomass ash were supplied by the plant Aldebarán Energía del Guadalquivir S.L. located in Andújar (Jaen, Spain). This company generates renewable energy using as fuel biomass of the pruning of olive groves and forest pruning (pine).

Fly or bottom biomass ash were added to the clay in different amounts (10–30 wt%) and mixed to obtain good homogenization. To enable comparative results, ten samples per series were prepared for the tests. The necessary amount of water (7–10 wt% moisture) was added to obtain an adequate plasticity and assess the absence of defects, mainly cracks, during the semi-dry compression moulding stage under 54.5 MPa of pressure, using a uniaxial laboratory-type pressing Mega

KCK-30 A. Waste-free mixtures were also made as a reference. Solid bricks with 30×10 mm cross-sections and a length of 60 mm were obtained. Samples were fired in a laboratory furnace at a rate of 3 °C/min up to 900 and 1000 °C for 4 h. Samples were then cooled to room temperature by natural convection inside the furnace. The shaped samples were designated as TC for the bricks without waste and TC-xW for the mixtures, where T indicates the firing temperature (900 or 1000 °C), x denotes the ash content (%) in the clay matrix and W the waste incorporated (W:FBA (fly biomass ash) and W:BBA (bottom biomass ash)).

2.2. Characterization of raw materials

A laser diffractometer Malvern Mastersizer 2000 was used to measure particle size distribution of different ash particles. Chemical composition was determined by X-ray fluorescence (XRF) using the Philips Magix Pro (PW-2440). Attenuated total reflection FTIR (FTIR-ATR) spectra of the materials were obtained on a Vertex70 FT/IR spectrophotometer from BRUKER by using a Golden Gate Single Reflection Diamond ATR System. It was studied the 500–4000 cm^{-1} region, with a resolution of 4 cm^{-1} . Crystalline phases were evaluated by powder X-ray diffraction with a X'Pert Pro MPD automated diffractometer (PANalytical) equipped with Ge (111) primary monochromator, using monochromatic Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$) and an X'Celerator detector. To better analyze the mineralogical composition of the starting raw clay, a granulometric separation by decantation was carried out to separate the different fractions: sand, lime and clay (<2 μm). Oriented aggregates of the clay fraction were prepared after deposition of a clay suspension on a flat glass sample holder and liquid evaporation at room temperature. After that, the sample was saturated with ethylene glycol (EG) or dimethyl sulfoxide (DMO) for 48 °C at 60 °C and finally calcined at 550 °C (1 h). A semiquantitative analysis of the phases present in the fired samples was performed by using X'pert HighScore Plus (3.0d) software.

Thermal behavior was determined by thermogravimetric and differential thermal analysis (TGA-DTA) with a Mettler Toledo 851e device in oxygen. The total content of carbon, hydrogen, nitrogen, and sulphur was determined by combustion of samples in O_2 atmosphere using the CHNS-O Thermo Finnigan Elementary Analyzer Flash EA 1112. The organic content was measured according to ASTM D-2974, Standard Test Method for Moisture, Ash, Organic Matter of Peat and Other Organic Soils (ASTM D-2974, 1987). The ignition temperature was 440 °C. Carbonate content (expressed as calcium carbonate) was determined by calcimetry in a Bernard calcimeter.

2.3. Characterization of clay-ash bricks

Linear shrinkage was obtained by measuring the length of samples before and after the firing stage, using a caliper with a precision of ± 0.01 mm, according to ASTM standard C326 (ASTM C326, 1997). Water absorption values were determined from weight difference between the fired and water-saturated samples (immersed in boiling water for 2 h), according to ASTM standard C373 (ASTM C373, 1994a). Open porosity (in vol.%) were determined from weight difference between saturated mass and dry mass with respect to exterior volume; and closed porosity (in vol.%) was calculated from weight difference between dry mass and suspended mass in water with respect to exterior volume according to ASTM standard C373 (ASTM C373, 1994a). Bulk density was determined by the Archimedes method (ASTM C373, 1994a). Water suction of a brick is the volume of water absorbed during a short partial immersion. Tests to determine water suction was implemented according to standard procedure UNE-EN 772-11 (UNE-EN 772-11, 2011). An efflorescence study was carried out. The bricks were immersed in water for 24 h and after dry in shade. After this treatment the presence of soluble salts in the bricks was determined.

Compressive strength of bricks is their bulk unit charge against breakage under axial compressive strength. For this trial, six fired

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