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Evaluation of spent diatomite incorporation in clay based materials for lightweight bricks processing

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HIGHLIGHT

- The use of diatomaceous earth residues in the ceramic industry is a sustainable alternative.

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ABSTRACT

In this study, diatomaceous earth residues from two industrial processes, refining of vegetable oils and brewing, have been used as raw materials for brick making. The aim has been to substitute part of the clay traditionally used for the manufacturing of bricks, between 3 and 10 wt.%, with the aforementioned residues, so obtaining ceramic pieces at three firing temperatures: 850, 950 and 1050 °C. The studied properties were compared with those of conventional materials (100% clay), and the results show that these alternative raw materials can be considered technological "nutrients" whose addition improves the behaviour of the materials in the drying process. The incorporation of these materials also increases the open porosity of the fired pieces and reduces the bulk density by up to 10%. The increase in porosity is greater in materials that incorporate diatomites from oil filtration, reaching the maximum value (37 vol. %). These results are confirmed from the microstructure observed by SEM. With regard to mechanical properties, increasing the content of both residues generally decreases the bending strength to values exceeding 10 MPa, thus resulting admissible for use in construction. Moreover, the energy release from the residues during the firing stage is greater than the energy demand required for drying, while the thermal conductivity values of the final materials decreases with the amount of residue, which confers thermal insulating properties to the ceramic pieces and thus can reduce the energy consumption of buildings.

1. Introduction

Action on climate, the environment and the efficient use of raw materials and resources are important challenges that our society must face. This objective is included within the challenges of the European Union Framework Program for Research and Innovation, Horizon 2020 [23] and, against this backdrop, the building sector must adapt to new materials and systems for environmentally sustainable construction. This requires research lines that reduce energy consumption as much as possible in new buildings and also seek to reduce energy consumption in buildings to be renewed [48,10]. In fact, the energy consumption associated with construction and building materials accounts for about 40% of total energy consumed in Europe (Eco-Innovation) [18]

Moreover, waste management, especially in the industrial and agricultural sectors, is an unresolved problem. The recycling of waste such as building materials can be an interesting solution, not only because it would reduce pollution problems, but also as an economic option in green building design. Different authors have investigated various physical, mechanical and thermal properties of bricks incorporating waste such as agricultural residues, paper, wood, plastic, ash or slag, cutting minerals powder or dry sludge from wastewater treatment plants [8,9,17,20,45,50,51,57]. Many of these works focus on the increase in porosity as a means of obtaining insulating ceramic materials.

Within the use of agricultural residues to obtain lightweight materials, Mekki et al. [41,56] studied the effluents from the production of olive oil in the manufacturing of bricks, ensuring that

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the mechanical properties are maintained within legislative limits using percentages of up to 23 wt.% of waste. Meanwhile, Görhan et al. [32] investigated the use of rice husk residue as a pore forming agent in final sintered materials, while more recently Aouba et al. [4] examined the effect of the addition of organic matter from agricultural residues (olive stone flour and wheat straw residues) in improving the thermal performance of ceramic materials, maintaining the values of compressive strength. These works show that generally there is a significant positive correlation between an increase in organic matter and porosity. However, for the same percentage of incorporated waste, a different total volume of pores is created, depending on the grain size distribution. In conclusion, the pore size distribution of new materials can be controlled depending on the type and granulometry of the incorporated residue.

On the other hand, Kung et al. [36] investigated the addition of sewage sludge from the rice industry, leading to increased porosity in the final fired clay products, and therefore to improved thermal insulation capacity, maintaining the mechanical properties within acceptable limits for the maximum percentage of residue employed (10 wt.%). Herek et al. [34] produced bricks from mixtures of sewage sludge from washing textiles and clay, obtaining satisfactory values for the bending strength and water absorption under Brazilian law. The results showed that this sludge could be incorporated up to a concentration of 20% by weight. Other works that include the valorisation of sewage sludge are those proposed by Cusidó et al. [15] and Qi et al. [49].

Moreover, some mineral residues are considered suitable for making ceramics [54,12,38]; due to their low cost and their ability to cause a decrease in the temperature required for the sintering process. One of these mineral wastes is diatomaceous earth from the treatment of industrial effluents. Diatomite, diatomaceous earth or kieselgur, is a siliceous sedimentary rock composed of the fossilized skeletons of frustules of diatomites, single-celled marine algae, which accumulate by sedimentation to form large deposits. Diatomaceous earth is very light, due to its high porosity, and is chemically inert. It also has a low thermal conductivity, a high melting point, a soft abrasive capacity, a high surface area and a good absorption capacity [5,35,40,33]. All these properties mean that products with a high porous structure can be obtained, and thus 61% of the world production of diatomite is used as a filter aid, mainly in filtering sugar syrup, beer, whiskey, wine, fruit juice, water, vegetable or mineral oils and pharmaceuticals [19]. The remaining 39% is mainly applied as a filler agent in different industries, in particular paint and plastic production, but also used as an absorbent for industrial residues, bedding for pets, abrasive in polishing metal, silica additive in the manufacture of Portland cement and as raw material for the manufacture of certain thermal and acoustic insulating materials [16]; [14]. The world's largest reserves of diatomite are in the United States [14], although large deposits have also been reported in China [39]. World production in 2013 was 2.3 Mt, with the United States of America holding 33% of total world production, followed by China with 18%, Denmark 14% and Peru 5.3%. Small amounts of diatomaceous earth are further obtained in 25 countries [14].

There are different studies on the use and improvement of the physical, mechanical and thermal properties of building materials made from natural diatomaceous earth. Some authors have studied the kinetics and microstructural development of ceramics prepared with diatomite characterizing the porosity of the final product [58]. There are also studies on improving the mechanical properties of concrete made with an addition of diatomaceous earth [22]. Other authors have studied the improvement of mechanical and thermal properties and the chemical stability in porous ceramics prepared with the addition of natural diatomites [40,2,3,33,46,1].

However, there are few investigations studying the use of spent diatomites from filtering processes. Mymrine et al. [43] studied the use of diatomaceous earth from oil filtration and galvanic waste in the manufacture of red ceramic, obtaining products that respect the environment, with high bending strength, low levels of leaching and production costs lower than those of traditional materials. Lin and Lan [37] investigated the retention capacity of porous ceramic adsorbents prepared with the addition of diatomaceous waste and fly ash from thermal power plants, obtaining materials with optimized adsorption and retention properties and improved thermal properties due to the porous structure formed.

Eliche-Quesada and Corpas-Iglesias [21] studied the effects of adding spent filtration earth and spent bleaching earth from the oil refinery industry to structural ceramic materials. They proved that these residues can be used to obtain bricks with good insulating properties without losing mechanical strength and determined that the optimal percentage for both residues is 10%.

The present study shows technological, thermal and microstructural properties of structural ceramics for construction, which in their formulation use diatomites from the refining vegetable oils and beer brewing industries as an alternative raw material to traditional clay. The research paper points specifically to the shaping of pieces by paste extrusion instead of dry pressing or with a low water content generally described in literature. Thus, the present work includes, according to the authors' knowledge, two original aspects. First, the study of the drying process of ceramic pieces, which can be crucial to the viability of brick making from the energetic and technological properties standpoint. Second, the firing temperature range evaluated covers thermal treatment below 950 °C (minimum operating temperature usually reported in the literature) in order to stress the application of the developed materials as common bricks and to improve the energy efficiency of the proposed valorisation process.

2. Materials and methods

2.1. Materials

A clay mixture from the geographical area of Bailén (Jaén, Spain) used routinely by the ceramic industry for the production of bricks [27,31] has been used as a reference ceramic material. Increasing quantities of weight, 3%, 7% and 10% of two types of diatomaceous earth used as filter media, one from the refining vegetable oil industry, OD, and another from the brewing industry, BD, were added to the reference clay material, R, in order to formulate the mixtures of clay and residues encodes ROD3, ROD10, D7 and RBD10.

2.2. Characterization methods

2.2.1. Raw materials

The chemical composition of the reference material, R, has been determined by energy dispersive XRF (PANalitical, Axios PW4400, Netherlands), with scintillation and flow detectors and Rh anode, while for the semi-quantitative determination of the mineralogical composition, X-ray diffraction equipment was used (Bruker, D8I-90, USA) with a Bragg-Brentano configuration and θ:θ geometry, a Cu target and a standard scan rate of 2° 2θ/min at 30 mA and 40 kV, using the methods of disoriented powder for the total sample, and oriented aggregates for the fraction <2 μm.

The chemical composition of the spent diatomaceous earth, OD and BD, has been determined by wavelength dispersive X-ray fluorescence WDXRF (PANanalytical, Axios Advanced) using a Rhodium (Rh) tube with voltage 4 kW. The qualitative diffractometric analysis, XRD, of both diatomites was carried out using a Bruker D8