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Lightweight dense/porous bi-layered ceramic tiles prepared by double pressing

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ABSTRACT

This work reports the production process of bi-layered ceramics tiles, formed from two layers with different densities – dense and porous – and with adjustable thickness. The novel production method comprises a double pressing action, fast and easy to implement industrially, that ensures the development of a perfect interface bonding between layers.

The bi-layered ceramic tile is formed by an upper layer with density similar to a conventional porcelain stoneware tile, and a porous bottom layer, which promotes weight reduction of the product maintaining suitable mechanical strength. The introduction of porosity is achieved by means of the incorporation of pore forming agent – polypropylene (PP) – into the ceramic formulation, which undergoes complete and non-harmful thermal decomposition during firing. For comparison, polymethyl methacrylate (PMMA) was also tested as porogen. The rapid and complete combustion of PP is suitable for fast-firing ceramic products, such as porcelain stoneware. In addition, the polymer decomposition does not induce defects in the ceramic pieces, and is environmentally acceptable.

The produced tiles are lighter than conventional porcelain stoneware tiles, thus decreasing their transport and distribution costs. Additionally, the thermal attenuation provided by the porous layer could be valuable in innovative applications, such as ventilated facades.

1. Introduction

Interest in ventilated facade systems has grown over recent years, particularly due to the demanding EU directive 2010/31/EU (2010) concerning the energy performance of buildings. Ventilated facades are energy efficient systems that can be implemented both in new or existing buildings. They promote the thermal and acoustic insulation of buildings, and numerous design solutions are available. In ventilated facades, an external facade cladding is installed in front of the common building facade, creating an air space in between. As a rule, the design of ventilated facades involves different layers: a sub-structure anchored to the wall surface of the building, a heat insulating layer, an air gap, and an external skin layer. The thermal performance of the external layer has been somewhat neglected by designers, to the detriment of esthetic criteria; however, the energy efficiency a ventilated facade is augmented when the external layer has low values of thermal conductivity (Patania et al., 2010). Considering these concepts, the possibility of using one single body that assures both functionalities (coating and insulation) could reduce the production costs (material and application) associated with this system of protection/exterior cladding of buildings, and might then constitute an important asset.

Porcelain stoneware tiles show promising potential as material for ventilated facades (Sánchez et al., 2010). They are endowed with high bending strength, virtually null water permeability (<0.5%), excellent frost and chemical resistance and optimal abrasion resistance. These outstanding properties make porcelain stoneware particularly suitable as material for floor and wall tiles in highly demanding environments, such as hospitals, shopping centers and industries. This high performance material is also attractive from an esthetic point of view, and hence, suitable for residential environments. Paradoxically, the high density (2.4 g/cm^3) of the product limits its use in novel applications. However, the production of lightweight porcelain stoneware tiles has been previously investigated. Gárcia-Ten et al. (2012) produced lightweight porcelain stoneware tiles by using silicon carbide as a foaming agent, while Bernardo et al. (2010) used cerium oxide. One common approach to reduce product weight is the incorporation of porosity in the product, which also improves the thermal insulation characteristics. This might be achieved by partial sintering, by using polymeric foam templates, ceramic hollow spheres or sacrificial pore-forming

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agents (see extensive reviews by Studart et al., 2006; Colombo, 2006). This porosity affects the product's mechanical strength, and a balance between all the required properties has to be established. Simultaneously, a porous surface tends to be esthetically unpleasant, and decoration/glazing is more difficult. One possible way to overcome such difficulties is producing bi-layered ceramic bodies, combining a dense top-layer with a porous bottom part. Recently, the preparation of a double-layered glass ceramic using double-pressing was reported by Binhussai et al. (2014), who employed a glass–ceramic frit that formed a dense layer on the surface of a brown-colored porous body derived from natural and waste raw materials. Koh et al. (2006) reported the in situ fabrication of a dense/porous bi-layered nickel–yttria-stabilized zirconia by using freeze casting, while Hong et al. (2011) prepared porous Al_2O_3 ceramics with functionally graded microstructure through a camphene-based freeze-casting process. However, existing production methods for multiple-layered ceramics are not practical for industrial application, since they usually require large solvent quantities and involve several fabrication steps, thus being time consuming and expensive processes. At the same time, the production of bi-layered ceramics is particularly difficult, due to the expected differential shrinkage between the different layers during firing, which induces detachment or deficient adhesion between them. Therefore, the development of novel methods, enabling mass production of macro multiple-layered ceramics, could lead to new and exciting applications.

The aim of this work is the development of lightweight bi-layered ceramics that may be used as floor and wall tiles by using a novel production method. The bi-layered ceramics are formed by layers with distinct density – dense/porous – and with adjustable thicknesses. The method comprises a double pressing action that is easy to implement, and ensures the development of a perfect interface bonding between the two layers. The simplicity of this processing method enables mass and low-cost production. The ceramic tiles show a top-layer in which density is similar to conventional floor pavements materials, and a porous bottom layer. The introduction of porosity in the bottom layer is achieved by means of the incorporation of pore forming agents, also known as porogens, in the spray-dried ceramic powder. Polypropylene (PP) was selected as pore forming agent due to its fast, complete and non-harmful thermal decomposition during firing, thus being suitable for fast-firing products such as porcelain stoneware tiles. The thermal decomposition of PP was investigated by Bockhorn et al. (1999) by using a free reactor with on-line mass spectroscopy. The authors observed that PP decomposes into a large number of aliphatic compounds without a residue.

For comparison purposes, a widely used pore-forming agent for ceramics was also tested—polymethyl methacrylate (PMMA). Thus, the present work also concerns the influence of pore-forming agents, their incorporation, content and particle size distribution on the bi-layered ceramics tiles properties.

2. Experimental

2.1. Materials

The investigation was performed with an industrial spray-dried powder normally used for the production of porcelain tiles.

Two different pore-forming agents were used, namely polypropylene (PP) and polymethyl methacrylate (PMMA). The PP was ICORENE PP CO14RM from ICO POLYMERS®, with a melt flow index of 13 g/10 min (2.16 kg at 190 °C) and 0.9 g/cm^3 density. The PMMA was Altuglas® VSE UVT with a melt flow index of 27 g/10 min (3.8 kg at 230 °C) and 1.18 g/cm^3 density, obtained from Arkema.

2.2. Sample preparation

Bi-layered ceramic discs and tiles were produced by a two-step pressing technique at room temperature, without the addition of binders or plasticizers. The discs were prepared with 25 mm diameter and 8 mm thickness. Initially, the spray-dried powder was placed inside a stainless steel die (diameter: 25 mm) and then was uniaxially pressed at 5 MPa to form an initial compact (green disc top layer). Afterwards a mixture of atomized ceramic powder and pore-forming agent was added to the die and then pressed at 10–12 MPa for 30 s, forming the bottom layer of the disc.

Bi-layered ceramic tiles, 10 × 10 cm, were prepared by uniaxial pressing (Gabbrielli—model L4/250, Italy). First the spray-dried powder was placed inside the mold, and then was uniaxially pressed at 5 MPa for 30 s to form an initial compact. Then a mixture of ceramic powder and pore-forming agent was added to the mold and then pressed at 20 MPa for 120 s, forming the bottom layer of the tile.

The content of pore forming agent in the bottom layer of the disc or tile ranged from 2.5 to 25 wt%.

Prior to pressing, the mixture (ceramic powder and pore-forming agent) was mechanically mixed until uniform distribution was observed.

PP and PMMA particles showed two distinct particle size distributions. Sieves of 250 and 425 μm were used to separate the particles, and the fractions smaller than 250 μm and in the range of 250–425 μm were then used. Hereafter, a sample prepared using PMMA with particles smaller than 250 μm will be coded as "PMMA/250".

The discs were dried in an oven at 110 °C for 12 h, and then fired according to the following cycle: (i) 15 °C/min heating rate up to 500 °C; (ii) 15 min dwell time to burn out the pore-forming agent; (iii) 15 °C/min heating rate up to 1200 °C and 4 min dwell time at this temperature; (iv) cooling (at 10 °C/min) to room temperature.

2.3. Material characterization

Scanning electron microscopy (SEM, Hitachi S4100) was used to investigate the interfacial bonding between the dense (top) and the porous (bottom) layers of the sintered samples.

Optical analysis (Leica EZ4HD microscope) was used to study the porous morphology. Samples were cut from the sintered discs using a Struers Secotom-10 table-top cutting machine. At least five specimens were prepared for each analysis. The images were acquired with a digital camera coupled to the microscope. Infinity Analyze software was used to measure the total and average pore area.

Archimedes method (immersion in ethylene glycol) was employed to measure the specimens' open porosity and water absorption values.

Three-point bending strength measurements were selected to evaluate the mechanical strength. Tests were performed on rectified parallelepiped bars (20 × 10 × 8 mm) of sintered samples (Shimadzu Autograph AG 25TA) using a 0.5 mm/min displacement speed. The specimens were placed with the porous layer facing down. Results were obtained using between eight to ten individual samples.

Thermal conductivity was measured with the C-Therm TCI™ Thermal conductivity analyzer. Tests were performed at room temperature on sintered discs with 25 mm diameter.

Dilatometric analyses (Bahr Thermo Analyse DIL 801L, Germany) were performed on unfired specimens at a constant heating rate of 10 °C/min. Bar testing specimens (5 × 4 × 10 mm) were prepared by uniaxial pressing.