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The impact of the particle shape of organic additives on the anisotropy of a clay ceramic and its thermal and mechanical properties



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HIGHLIGHTS

- The clay ceramic with no organic additives presented an anisotropy of 1.38.
- The powder additive decreased the anisotropy to 1.11 by the formation of round shape pores.
- The fibers additive increased the anisotropy to 1.94 by the formation of orientated pores.
- The 8 wt% addition of powder improved the flexural strength of the clay ceramic by 13%.
- The 8 wt% addition of fibers improved the thermal diffusivity of the clay ceramic by 41%.

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ABSTRACT

This research investigated the impact of the particle shape of organic additives on the anisotropy of a clay ceramic and its thermal and mechanical properties. The impact of the particle shape was elucidated by addition in the clay ceramic mixture of Olive Stone Flour (OSF) in the form of powder and Wheat Straw (WS) in the form of fibers. The OSF powder reduced the clay ceramic anisotropy with a formation of round shape pores during the firing process. The flexural strength of the clay ceramic was improved by 13% in case of an 8 wt% addition of OSF. On the other hand, the WS fibers increased the anisotropy by the formation of orientated pores along the extrusion plane. The thermal diffusivity of the clay ceramic was improved by 41% in the direction of the thermal gradient of the walls in case of an 8 wt% addition of WS. Hence, this study proved the particle shape of the organic additives as a useful parameter to control the anisotropy of clay ceramics and improve their thermal and mechanical properties.

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1. Introduction

The energy efficiency of buildings is ever expected to grow in order to reduce our energy consumption [1]. Buildings are responsible at the present time for more than 40% of energy consumption in the European Union [2]. Therefore, the building materials such as fired clay bricks have a key role to play in this challenge [3]. The fired clay bricks can reduce energy losses of buildings with higher insulation performances. Hence, the improvement of their thermal properties has become an important issue. The investigations have focused in recent years on the recycling of organic waste in fired clay bricks [4,5].

The waste added to the composition of fired clay bricks comes from the wood, paper or oil industries... These organic additives are used for an important organic fraction higher than 60% [6]. The addition of organic additives is ranging from 1 wt% to 30 wt %, depending on the organic fraction [7]. Furthermore, the particle size of the organic additives is commonly not higher than 4 mm in order to keep the process unchanged [8]. The clay with organic additives and water is shaped in a form of bricks using an extrusion process. Afterwards, the water is removed by means of a drying process at temperatures up to 100 °C. The clay bricks are also subjected to a firing process at around 1000 °C. The firing process of the clay bricks induces a thermal degradation of the organic additives [9]. The fired clay bricks present high thermal properties as a result of the thermal degradation of the organic additives [10]. However, the mechanical properties of the fired clay bricks are relatively limited compared to the fired clay bricks with no organic additives [11].

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The trade-off between the thermal and mechanical properties of fired clay bricks becomes an issue since they are used in building applications for insulation and resistance purposes at the same time. Many studies have attributed this trade-off to the microstructure of the fired clay bricks. In fact, the thermal degradation of the organic additives induces a porosity formation during the firing process [12]. The clay bricks present a higher percentage of porosity after the firing process [13]. Hence, the low thermal conductivity of air in the newly formed pores improves the thermal properties of the fired clay bricks [14]. Some studies have shown that pores resulting from the thermal degradation of residues from oil industry can improve the thermal properties up to 20% [15]. However, a recent study has also shown that the thermal properties of fired clay bricks are enhanced by the anisotropy [16]. The heat conduction is highly limited through their extrusion plane. The fired clay bricks present higher thermal properties in the direction of the thermal gradient of the walls. Hence, the morphology of the newly formed pores could provide a higher improvement of the thermal properties.

According to the literature, the improvement of the thermal properties is associated with a decrease of the mechanical properties [17]. The pores resulting from the thermal degradation of the organic additives act as defects in the microstructure of the fired clay bricks [18]. Some studies have shown that the mechanical properties of fired clay bricks were reduced by 39% for a 10% addition of residues from kraft pulp production [19]. However, some other studies indicated that an addition of processed waste tea in a form of fibers could even improve the mechanical properties [20]. This means that the morphology of the pores resulting from the thermal degradation of the organic additives could have an impact on the thermal and on the mechanical properties of fired clay bricks. The morphology of pores could also be related to the particle shape of the organic additives.

The purpose of this study is to investigate the impact of the particle shape of organic additives on the anisotropy of a clay ceramic and its thermal and mechanical properties. Therefore, the study focuses on an addition of olive stone flour, in a form of powder, and on an addition of wheat straw, in a form of fibers. The particle shape of the organic additives is related to the morphology of the newly formed pores. The morphology of these pores is also related to the anisotropy of the clay ceramic. The impact of the anisotropy on the thermal and on the mechanical properties of the clay ceramic is finally displayed. Hence, this study provides an insight to control the anisotropy of fired clay bricks and improve their mechanical and thermal properties.

2. Materials and methods

2.1. Materials

The clay used in this study was extracted from a clay quarry in Toulouse Area (France). The clay was ground at the laboratory with a rolling mill at 3 mm. The elemental composition of the clay was measured with an ICP-AES instrument (Jobin Yvon Ultima 2). The clay was dissolved in a mixture of perchloric and hydrofluoric acid heated at 80 °C for 30 min. The solutions were diluted 10 times in purified water before the analysis. The results in Table 1 indicate a

Table 1

Elemental composition of the clay with the concentrations in silicon, aluminum, calcium, iron, potassium and magnesium oxides.

| Sample | Concentration (wt%) | | | | | | | |
|--------|---------------------|-----------|-----|--------------------------------|------------------|-----|--|--|
| | SiO ₂ | Al_2O_3 | CaO | Fe ₂ O ₃ | K ₂ O | MgO | | |
| Clay | 44.8 | 16.8 | 9.4 | 9.9 | 3.8 | 1.9 | | |

predominance of silica and alumina with smaller amounts of calcium, iron and potassium.

The organic additives consisted of Olive Stone Flour (OSF) and Wheat Straw (WS). The OSF additive obtained from BARDON Company was used in the given form. On the other hand, the WS additive obtained from ARTERRIS Cooperative was ground at the laboratory with a knife mill at 1 mm (Pulverisette 15, Fritsch). The composition measured by organic elemental analysis (Flash 2000, Thermo Fisher Scientific) is given in Table 2. The OSF additive is composed of carbon, oxygen, hydrogen and nitrogen elements for more than 98 wt%. The WS additive is composed of the same elements than OSF additive with smaller concentrations. It indicates that the WS additive contains a smaller organic fraction of around 87 wt%.

The organic additives were observed by scanning electron microscopy (SEM) using a Philips XL30 apparatus. The SEM micrographs show the particle shape of the organic additives in Fig. 1. The OSF additive is composed of round shape and angular particles with an average size of 50 μ m. On the other hand, the fibers of the WS additive present an average breadth of 100 μ m and an average length of 1 mm.

2.2. Clay ceramic

The clay ceramic investigated in this study was made of different mixtures of clay, organic additives and water. The mixtures were prepared in a kneading bowl by mixing 10 kg of clay with 4 wt% and 8 wt% of organic additives. Afterwards, some water was added to the mixtures until an 8 bar pressure of extrusion was obtained. The mixtures were extruded as clay ceramics of $180 \times 80 \times 18 \text{ mm}^3$ for a water amount of 17 wt%. The clay ceramics were subsequently dried at 25 °C, 65 °C and 105 °C for 24 h at each temperature in an electrical oven. The samples were prepared from the dried ceramics by polishing with P80, P120, P180 and P280 SiC abrasive papers (CarbiMet, Buehler). Finally, some samples were subjected to a firing process in an electrical furnace (Nabertherm Controller P320) at temperatures given in the next sections.

2.3. Characterization of the microstructure

The thermal behavior of the clay ceramic was investigated by differential thermal analysis (DTA). The dried ceramics were analyzed as 200 mg cylinders with a 5 mm diameter using a Setaram 92 instrument. Data were collected in air atmosphere from 30 °C to 1100 °C with a 5 °C/min heating rate.

The porosity of the clay ceramic (ϵ) was determined by Eq. (1) from the bulk density of the clay ceramic (ρ_{Bulk}) and the true density of the particles (ρ_{True}). The bulk density of the clay ceramic was estimated by the weight/volume ratio. The weight and the volume of the samples were measured at room temperature after a firing process at 30 °C, 100 °C, 200 °C, 300 °C, 400 °C, 500 °C, 600 °C, 700 °C, 800 °C, 900 °C, 1000 °C and 1100 °C using a 5 °C/min heating rate. The true density of the particles was measured by helium pycnometry (Accupyc 1330, Micromeritics) after a firing process of the clay ceramic at the maximum temperature

Table 2

Elemental composition of the organic additives with the concentrations in carbon, hydrogen, oxygen, nitrogen and sulfur elements.

| Sample | Concentration (wt%) | | | | | | |
|-----------|---------------------|------------|--------------|------------|------------|--|--|
| | С | Н | 0 | Ν | S | | |
| OSF WS | 49.8 43.1 | 6.0 5.5 | 42.0 28.5 | 0.4 0.7 | 0.0 0.0 | | |

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