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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 L.V. Korah^{a,b}, P.M. Nigay^{b,c,*}, T. Cutard^c, A. Nzihou^b, S. Thomas^a

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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.

Keywords: Clay ceramic Organic additives Porosity Anisotropy Thermal diffusivity Stress-strain curve behavior

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_2O_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 $180 \times 80 \times 18$ mm³ 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	49.8	6.0	42.0	0.4	0.0			
WS	43.1	5.5	28.5	0.7	0.0			



Fig. 1. SEM micrographs of the particle shape of OSF (a) and WS (b).

of 1100 °C and a grinding to eliminate the porosity. The true density was equal to 2.72 g/cm^3 for the clay ceramic and for the clay ceramic with organic additives. It was also adjusted to the percentage of organic additives for a firing process at temperatures lower than 500 °C.

$$\varepsilon = 1 - \frac{\rho_{\text{Bulk}}}{\rho_{\text{True}}} \tag{1}$$

The microstructure was observed by scanning electron microscopy (SEM) using a Philips XL30 apparatus. The observations were performed along the extrusion plane after a firing process at 900 °C with a 5 °C/min heating rate. The samples were embedded in a solidifying epoxy resin and polished to the finest grain before the observations. The micrographs of the clay ceramic display the clay in grey colours and the porosity in a black colour.

2.4. Determination of the anisotropy and of the thermal properties

The anisotropy of the clay ceramic was determined from the thermal diffusivity in different directions. The thermal diffusivity was measured by flash method using a Profilux Plus 800 (Multiblitz) instrument. The samples of $25 \times 25 \times 5 \text{ mm}^3$ were analyzed along and through the extrusion plane after a firing process at 900 °C with a 5 °C/min heating rate. The anisotropy of the clay ceramic was taken as the along/through ratio of the thermal diffusivity.

2.5. Determination of the mechanical properties

The mechanical properties of the clay ceramic were measured by three point bending after a firing process at 900 °C with a heating rate of 5 °C/min. The samples of $60 \times 30 \times 5 \text{ mm}^3$ were loaded in an Instron 5800R machine with a 40 mm loading span and a 500N load cell at a constant displacement of 1 mm/min.

$$\sigma_{\rm f} = \frac{3FL}{2BH^2} \tag{2}$$

The flexural stress (σ_f) was estimated from Eq. (2) using the force (F), the loading span (L), the breadth (B) and the height (H) of the samples [21]. Furthermore, the flexural strain of the clay ceramic was calculated from the displacement of the crosshead. It was taken as the displacement/height ratio during the test.

3. Results and discussion

3.1. Thermal behavior of the clay ceramic

The thermal behavior of the clay ceramic is displayed in Fig. 2. The clay ceramic with no organic additives experiences a dehydration from 30 °C to 200 °C. The dehydration induces a 3 wt%



Fig. 2. Thermal behavior of the clay ceramic and of the clay ceramic with a 4 wt% and an 8 wt% addition of OSF and WS.

elimination of water remaining in the pores and adsorbed on the clay particles. The firing process also leads to a dehydroxylation of the clay minerals from 400 °C to 600 °C. The release of constitution water induces a weight loss of 2 wt%. Furthermore, the firing process induces a decarbonation of the calcium carbonates from 600 °C to 800 °C. A weight loss of 4 wt% is associated with the release of carbon dioxide. Hence, the clay ceramic presents a weight loss of 9 wt% after a firing process at 900 °C.

The OSF additive experiences a combustion in the clay ceramic from 200 °C to 400 °C. The transformation of OSF into water and carbon dioxide induces a weight loss of 4 wt%. The weight loss of 4 wt% corresponds to the 4 wt% addition in the clay ceramic. Hence, the OSF additive is fully eliminated by the combustion. The elimination is also performed on the free range of temperatures from 200 °C to 400 °C. The thermal behavior of the clay is then not modified by the combustion of OSF additive. The clay ceramic with a 4 wt% addition of OSF presents a weight loss of 13 wt% after a firing process at 900 °C. The WS additive presents a same combustion in the clay ceramic from 200 °C to 400 °C. The transformation of WS into water and carbon dioxide induces a slightly lower weight loss. This is related to the lower organic fraction around 87 wt%. Nevertheless, the clay ceramic with a 4 wt% addition of WS presents a weight loss of almost 13 wt% after a firing process at 900 °C.

The organic fraction of the additives is fully eliminated by the combustion in case of a larger addition. The 8 wt% addition of OSF and WS induces a weight loss of 8 wt% during the firing process. The thermal behavior of the clay is also conserved in case of a larger addition of organic additives. Hence, the clay ceramic with



Fig. 3. Evolution of the porosity of the clay ceramic and of the clay ceramic with a 4 wt% and an 8 wt% addition of OSF and WS with the firing temperature.

an 8 wt% addition of OSF and WS presents a weight loss of a 17 wt% after a firing process at 900 °C.

3.2. Porosity of the clay ceramic

The evolution of the porosity with the firing temperature of the clay ceramic is displayed in Fig. 3. The clay ceramic with no organic additives presents a porosity of 24 vol% after a firing process at 30 °C. In fact, the air remaining in the mixture is trapped in the clay ceramic as pores during the extrusion process. The porosity of the clay ceramic is also increased by the water release during the drying process. Furthermore, the dehydration of the clay ceramic induces a 2 vol% formation of porosity from 30 °C to 200 °C. The porosity is also increased by 2 vol% during the dehydroxylation from 400 °C to 600 °C. The decarbonation also leads to a 3 vol% formation of porosity from 600 °C. The porosity is only reduced by 2 vol% during the sintering from 800 °C to 1100 °C. Hence, the clay ceramic present a porosity of 30 vol% after a firing process at 900 °C.

The combustion of OSF additive induces a 6 vol% formation of porosity in case of a 4 wt% addition. The porosity formation is performed on the free range of temperatures from 200 °C to 400 °C. Hence, the porosity formations associated with the clay are not modified by the combustion of OSF additive. The porosity generated from 200 °C to 400 °C is also conserved during the sintering. Therefore, the clay ceramic with a 4 wt% addition of OSF presents a porosity of 36 vol% after a firing process at 900 °C. The combustion of WS induces a 10 vol% formation of porosity for a 4 wt% addition. The organic additives in a form of fibers have a lower

density than the organic additives in a form of powder. Hence, the combustion of WS releases a larger volume of porosity than OSF for a same addition. The clay ceramic with a 4 wt% addition of WS present a porosity of 40 vol% after a firing process at 900 °C.

The addition of organic additives induces a linear increase of the porosity. The clay ceramic with an 8 wt% addition of OSF powder presents a porosity of 42 vol% after a firing process at 900 °C. On the other hand, the clay ceramic presents a porosity of 50 vol% in case of an 8 wt% addition of WS fibers. The porosity of the clay ceramic is then related to the percentage and the particle shape of the organic additives.

3.3. Microstructure of the clay ceramic

The SEM micrographs of the clay ceramic after a firing process at 900 °C are displayed in Fig. 4. The clay ceramic with no organic additives presents a microstructure of clay with 75 μ m pores. The pores correspond to the edge of larger porosity sheets orientated along the extrusion plane. In fact, the extrusion process induces a compression of the clay in a form of sheets. The air remaining in the mixture is then compressed between the sheets of clay in a form of porosity sheets. The porosity sheets are also conserved during the sintering of the clay ceramic. Therefore, the clay ceramic present a layered microstructure of clay and porosity sheets after the firing process.

The clay ceramic with an 8 wt% addition of OSF does not contain any porosity sheets. The porosity sheets are filled by the organic additives in a form of powder during the extrusion process. Furthermore, the clay ceramic presents round shape pores with an average size of 50 μ m. These pores correspond to the voids released by the combustion of the round shape particles of OSF. In fact, the pores generated from 200 °C to 400 °C are conserved during the sintering. Hence, the clay ceramic present an isotropic behavior after the firing process in case of a powder addition.

The clay ceramic with an 8 wt% addition of WS contains wide sheets of porosity. The organic additives in a form of fibers are orientated with the air remaining in the mixture during the extrusion process. The combustion of the WS fibers also leads to the formation of orientated pores. The SEM micrograph shows an edge of the newly formed pores orientated along the extrusion plane. In fact, these pores generated from 200 °C to 400 °C are conserved during the sintering of the clay ceramic. Therefore, the clay ceramic present an anisotropic behavior in case of a fibers addition.

3.4. Anisotropy of the clay ceramic

The evolution of the anisotropy with the percentage of organic additives is displayed in Fig. 5. The clay ceramic with no organic additives presents an anisotropy of 1.38 after a firing process at



Fig. 4. SEM micrographs of the microstructure of the clay ceramic (a) and of the clay ceramic with an 8 wt% addition of OSF (b) and WS (c) after a firing process at 900 °C.



Fig. 5. Anisotropy of the clay ceramic and of the clay ceramic with a 4 wt% and an 8 wt% addition of OSF and WS after a firing process at 900 °C.

900 °C. The anisotropic behavior is related to the layered microstructure of clay and porosity sheets.

The organic additives induce an evolution of the anisotropy. However, the evolution of the anisotropy depends on the particle shape of the organic additives. The organic additives in a form of powder induce a small formation of round shape pores. Hence, the anisotropy of the clay ceramic is decreased by 9% with a 4 wt % addition of OSF. The clay ceramic present an anisotropy of 1.25 after a firing process at 900 °C. Furthermore, the organic additives in a form of fibers induce a large formation of orientated pores. The anisotropy is then increased by 21% with a 4 wt% addition of WS. The clay ceramic present an anisotropy of 1.66 after the firing process at 900 °C.

The addition of organic additives provides a linear evolution of the anisotropy. The clay ceramic with an 8 wt% addition of OSF powder present an anisotropy of 1.11 after a firing process at 900 °C. On the other hand, the anisotropy of the clay ceramic is increased to 1.94 in case of an 8 wt% addition of WS fibers. The anisotropy of the clay ceramic is then related to the percentage and the morphology of the newly formed pores. Hence, the anisotropy can be controlled by the percentage and the particle shape of the organic additives.

3.5. Thermal properties of the clay ceramic

The evolution of the thermal diffusivity with the percentage of organic additives is displayed in Fig. 6. The clay ceramic with no organic additives presents a thermal diffusivity of 0.60 mm²/s along the extrusion plane after a firing process at 900 °C. This value represents a combination between the relatively high thermal diffusivity of the clay and the low thermal diffusivity of air in the porosity. On the other hand, the clay ceramic presents a thermal diffusivity of 0.44 mm²/s through the extrusion plane. The impact of air on the thermal diffusivity is enhanced by the anisotropic behavior of the porosity sheets. The heat diffusion is highly limited through the porosity sheets with no favorable path. Hence, the clay ceramic present a lower thermal diffusivity in this direction of the thermal gradient of the walls.

The 4 wt% addition of OSF induces a 15% decrease of the thermal diffusivity along the extrusion plane. The thermal diffusivity of the clay ceramic is decreased by the low thermal diffusivity of air in the pores resulting from the combustion of OSF. However, the newly formed pores only lead to a 7% decrease of the thermal diffusivity through the extrusion plane. The addition of organic



Fig. 6. Thermal diffusivity of the clay ceramic and of the clay ceramic with a 4 wt% and an 8 wt% addition of OSF and WS after a firing process at 900 °C.

additives in the form of powder reduces the anisotropy of the clay ceramic. Hence, the elimination of the porosity sheets promotes heat conduction through the extrusion plane. The clay ceramic only presents a thermal diffusivity of 0.41 mm²/s in the direction of the thermal gradient of the walls. On the other hand, the organic additives in the form of fibers increase the anisotropy. The heat diffusion is highly limited through the wider sheets of porosity with no favorable path. Hence, the thermal diffusivity is decreased by 23% through the extrusion plane in case of a 4 wt% addition of WS. The clay ceramic presents a lower thermal diffusivity of 0.34 mm²/s in the direction of the thermal gradient of the walls.

The addition of organic additives provides a linear decrease of the thermal diffusivity. The thermal diffusivity is decreased to 0.38 mm²/s with an 8 wt% addition of OSF powder. On the other hand, the clay ceramic with an 8 wt% addition of WS fibers presents a low thermal diffusivity of 0.26 mm²/s. The thermal diffusivity ity in the direction of the thermal gradient of the walls is then related to the anisotropy. Hence, the thermal diffusivity of the clay ceramic can be controlled by the percentage and the particle shape of the organic additives.

3.6. Mechanical properties of the clay ceramic

The stress-strain curves of the clay ceramic after a firing process at 900 °C are displayed in Fig. 7. The clay ceramic with no organic additives experiences a brittle fracture at a flexural stress of 12.1 MPa and a flexural strain of 3.4%. The stress-strain curve behavior of the clay ceramic is related to the layered microstructure of clay and porosity sheets. The clay sheets allow a deformation of the clay ceramic under a flexural stress. However, the large sheets of porosity act as a source of fracture and limit the flexural strength.

The 4 wt% addition of OSF increases the flexural strength of the clay ceramic by 16%. The flexural strength is increased by the elimination of the porosity sheets. However, the lower proportion of clay sheets limits the deformation of the clay ceramic. The flexural strain of the clay ceramic is decreased by 9% in the case of a 4 wt% addition of OSF powder. The clay ceramic present a mechanical strength of 14.0 MPa and a flexural strain of 3.1%. On the other hand, the organic additives in a form of fibers increase the anisotropic behavior of the clay ceramic. The formation of orientated pores induces a 3% decrease of the flexural strength in case of a 4 wt% addition of WS fibers. However, the larger proportion of clay sheets increases the flexural strain by 12%. The clay ceramic with a



Fig. 7. Flexural stress-strain curves of the clay ceramic and of the clay ceramic with a 4 wt% and an 8 wt% addition of OSF and WS after a firing process at 900 $^{\circ}$ C.

4 wt% addition of WS fibers presents a flexural strength of 11.7 MPa and a flexural strain of 3.8%.

The larger addition of organic additives limits the increase of the flexural strength. The porosity sheets acting as a source of fracture are filled by the organic additives in a form of powder. However, the newly formed pores also act as defects in case of a larger addition than 8 wt%. The clay ceramic with an 8 wt% addition of OSF powder presents a flexural strength of 13.7 MPa. Furthermore, the organic additives in a form of fibers induce a larger increase of the anisotropic behavior. The clay ceramic with an 8 wt% addition of WS fibers present a flexural strain of 4.4%. Hence, the mechanical properties of the clay ceramic can be controlled by the percentage and the particle shape of the organic additives.

3.7. Implications

The implications of this research are significant for the recycling of organic waste in fired clay bricks. The addition of organic waste in the clay bricks mixture induces a formation of porosity during the firing process. The clay bricks present higher thermal properties after the firing process. Hence, the better insulation performances of the fired clay bricks provide a reduction of building energy consumption.

However, this work indicates that the insulation performances of the fired clay bricks can be further improved by addition of organic waste in the form of fibers. The fibers induce a formation of orientated pores along the extrusion plane of the fired clay bricks. The impact of the newly formed pores on the thermal properties of the fired clay bricks is also increased by their anisotropic behavior. Hence, the thermal properties were improved by 41% in this study using an 8 wt% addition of fibers. These results could widen the range of organic waste added to the fired clay bricks. In fact, the thermal properties of the fired clay bricks could be improved by many organic waste in case of a suitable shape of the particles.

The control of the anisotropy with the particle shape of the organic additives also provided motivation for further studies. The anisotropy limits the heat transfers through the extrusion plane. However, the heat transfers are highly promoted by the anisotropic behavior along the extrusion plane. The favorable path in the clay ceramic might lead one to think about a fibers addition in clay roofing tiles for a concentration of solar energy.

4. Conclusions

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 of organic additives in the form of powder and fibers. The organic additives added to the clay ceramic experienced a combustion during the firing process. The transformation of the organic additives into water and carbon dioxide resulted in a porosity formation of the clay ceramic. Furthermore, the morphology of the newly formed pores corresponded to the particle shape of the organic additives.

The organic additives in a form of powder induced a formation of round shape pores. The anisotropy of the clay ceramic was decreased to 1.11 for an 8 wt% addition of powder. The decrease of the anisotropy led to a limited 14% improvement of the thermal properties in the direction of the thermal gradient of the walls. However, the mechanical properties of the clay ceramic were improved by 13% after the firing process.

The organic additives in a form of fibers induced a formation of orientated pores along the extrusion plane. The anisotropy of the clay ceramic was increased to 1.94 in case of an 8 wt% addition of fibers. The newly formed pores acting as a source of fracture induced a 26% decrease of the mechanical properties. However, the thermal properties of the clay ceramic were improved by 41% due to the anisotropic behavior. These results 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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Conflict of interest

The authors declare that they have any conflict of interest.

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