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Research paper

Influence of mechanical treatment on thermophysical processes in illitic clay during firing

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ABSTRACT

The influence of mechanical treatment on thermophysical properties of illitic clay was studied during firing using the acoustic emission technique (AE), scanning electron microscopy (SEM), X-ray diffracion analysis (XRD) and thermal analyses (DTA, TGA, DIL). The milling process promotes the formation of agglomerates created from the illite crystals damaged by milling. Due to the newly formed crystal defects, the course of dehydroxylation as well as the high temperature processes were shifted to lower temperatures owing to rising milling time. The AE activity during heating increases with increasing the milling time. Thus, longer milling time contributes to the crack formation during heating. However, during cooling, the low AE activity indicates that the crack formation was suppressed. The density of the samples increases with increasing milling time, thus a denser, more homogeneous microstructure is formed.

1. Introduction

Sintering of clay materials, particularly the maximum firing/sintering temperature, is significantly influenced by the particle size. It is known that finer particles can improve sintering conditions (Rahaman, 2003). As was reported in Nakahara et al. (1999) that smaller particles shift the temperature of high-temperature reactions to lower values and the structure after firing becomes denser. The simplest way of the particle-size reduction is a mechanical milling. Numerous studies have been focused on mechanical treatment of kaolinite (see e.g. Hamzaoui et al., 2015; Mitrović and Zdujić, 2013; Ptáček et al., 2013; Sánchez-Soto et al., 2000; Vágvölgyi et al., 2008; Vdovic et al., 2010; Vizcayno et al., 2010). The milling process decreases the structural order of kaolinite and leads to the reduction of the activation energy of dehydroxylation (Ptáček et al., 2013). Thus, the dehydroxylation process occurs at lower temperatures (Sánchez-Soto et al., 2000; Vágvölgyi et al., 2008). A limit in the kaolinite crystals size, reached after milling, strongly depends on process parameters, such as type of grinding mill and milling duration. It was reported in Elmas et al. (2013) that milling also increases the quantity of mullite in porcelain and reduces metakaolinite-mullite transformation temperature (Koç et al., 2011). Moreover, with respect to X-ray diffraction (XRD) measurements (Hamzaoui et al., 2015; Leonel et al., 2014; Mitrović and Zdujić, 2013; Vágvölgyi et al., 2008), kaolinite undergoes a structural decomposition during milling.

In contrast to kaolinite, the mechanical treatment of other phyllosilicates, which are used in ceramic manufacture, were investigated not so often. It has long been known that milling of minerals damages their crystal structures. The intensity of X-ray patterns gradually weaken and original sharp reflexions become more diffused by milling and after 8 h of milling no sharp reflections can be found. The increase in moisture content with grinding was also clearly indicated (Mackenzie and Milne, 1953). Milosevic et al. (1992) found that milling can result in extensive changes in the crystal structure of kaolinite and illite. The changes led to amorphization of the clay crystals, as was supported by XRD, IR and DTA. With prolonged milling time, the hydroxyl groups became less stable and the dehydroxylation peaks were shifted toward lower temperatures. XRD and IR results indicated that the dry-milling process caused nearly complete destruction of the crystal structures of the clay crystals, whereas the structures of quartz and feldspar were affected only at the surfaces of the grains. Similar conclusions are in Sánchez-Soto et al., 1994 where was also found for different kaolinite-illite-pyrophyllite mixtures that the milling process increases the exothermic process at 985 °C due to a presence of pyrophyllite in the clays. Milling also produces agglomerates as detected by SEM. Juhász (1998) deals with some clay minerals (mainly kaolinite) in his review

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Table 1
The chemical composition of illite from Füzérradvány, Hungary (in wt%).

SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	K_2O	Na_2O	LOI
58.0	24.0	0.6	0.05	0.38	1.70	7.85	0.10	7.3

of mechanical and mechanochemical activation. Juhász and Somogyi (1984) studied influence of milling of the illitic clay from Hungary on its crystal structure and chemical reactivity. They found that dry milling causes a comminution of particles and deformation of the crystal structure. Yang et al. (2005) reported that milling of illite increases the pore size, and reduces the specific surface area and the total pore volume. In addition, observed illite crystals have reached their size limit after 2 h of milling. Illite is transformed into an amorphous structure during the early stages of the milling and after 8 h-milling is completely destroyed. The DC electrical conductivity up to 1100 °C was investigated for milled illite samples in Csáki et al. (2015). It was found that the conductivity increases with the milling time.

As follows from the cited articles, the most often object of investigation was alteration of the structure and microstructure by the milling.

Mechanical properties of ceramic products are crucial for their use. These properties are significantly influenced by crack formation during a thermal treatment. Such investigation of illite, fired at different maximum temperatures, using the acoustic emission (AE) technique and dynamical thermomechanical analysis was performed in Knapek et al. (2016). The authors reported a detectable crack formation during cooling when the firing temperature exceeds 900 °C.

In this study, a raw illitic clay which contains approximately 80 wt% of illite was used. The aim of our work is to evaluate the influence of dry milling on the microstructure development and crack formation during both heating and cooling stage of firing. Since the cracks influence mechanical strength significantly, the crack formation study under different conditions is valuable for ceramic industry. To the authors` knowledge, this type of study has never been done before.

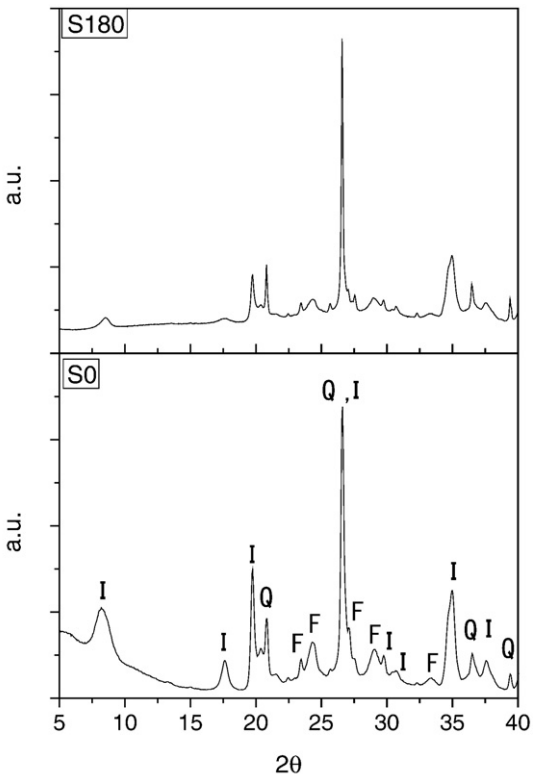

Fig. 1. XRD patterns of the raw samples S0 and S180. I – illite, Q – quartz, F – K-feldspar.

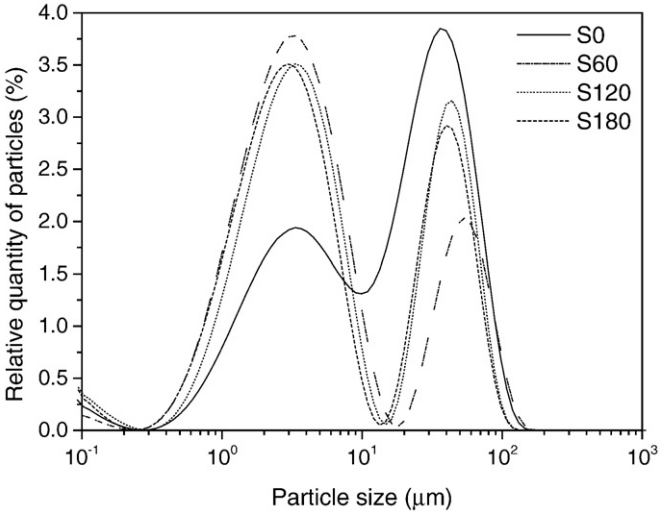

Fig. 2. Particle size distribution in samples S0, S60, S120 and S180.

In the heating stage of firing, the samples were studied using in-situ techniques: acoustic emission (AE), thermodilatometric analysis (DIL), differential thermal analysis (DTA) and thermogravimetry (TGA). In the cooling stage of firing, when crack formation can take place, AE and DIL measurements were used to determine the influence of milling on this process. The results are supported by microstructure observations of samples using scanning electron microscopy (SEM).

2. Materials and methods

The illitic clay from Füzérradvány (Hungary) consists of illite (80 wt%), montmorillonite (4 wt%), quartz (12 wt%) and orthoclase (4 wt%). Its chemical composition can be found in Table 1.

The raw material was crushed, dried and sieved to obtain a powder with a particle size below 100 μm. The as-prepared powder was then milled for various times (0, 60, 120 and 180 min; samples made from these powders are labeled as S0, S60, S120 and S180, respectively) using the planetary laboratory mill Retsch PM100. Dry milling of 95 g of powder was performed in an alumina vessel (250 ml) with 12 alumina milling balls having a diameter of 20 mm at an angular speed of 350 rpm in 5 min cycles. After a 30 min of milling, a 10 min pause was applied in order to prevent the clay to reach temperature higher than 100 °C. The plastic mass for samples was prepared by mixing the powder with distilled water (clay:water ratio was 5:3). The rectangular samples having a cross-section of 11 × 11 mm were manufactured by pressing this wet plastic mass into a gypsum forms and dried in them for 4 days in the open air.

A computer-controlled DAKEL-XEDO-3 AE system (DAKEL ZD, Rpety, Czech Republic) was used to record the AE activity. Dry sample (11 × 11 × 80 mm) was placed into the furnace. The alumina rod with a diameter of 3 mm, which served as a waveguide, was fixed to the sample. The other end of the rod was glued to the piezoelectric AE transducer MST8S (DAKEL ZD Rpety, Czech Republic) with a diameter of 3 mm and flat response in a frequency band from 100 to 600 kHz. A preamplifier with a gain of 35 dB was used. The total gain was 91 dB, the signal

Table 2
The volume fractions of particles (in vol%).

	Sample			
Particle size	S0	S60	S120	S180
p.s. < 3 μm	21.8	43.2	37.5	39.4
3 μm < p.s. < 40 μm	55.7	43.7	42.5	45.0