



Research paper

Influences of quartz and muscovite on the formation of mullite from kaolinite

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ABSTRACT

A kaolin containing muscovite and quartz (K-SZ) and a pure kaolin (K-SX) with the addition of potassium feldspar, K₂SO₄ and quartz, respectively, were used to investigate the influences of muscovite and quartz on the formation of mullite from kaolinite in the temperature range 1000–1500 °C. In K-SZ formation of mullite began at 1100 °C, and in K-SX at 1000 °C. In K-SZ quartz accelerated the formation of cristobalite and restrained the reaction of mullite and silica. Muscovite in K-SZ acted as a fluxing agent for silica and mullite before 1400 °C and accelerated the formation of cristobalite. The FTIR band at 896.8 cm⁻¹ was used to monitor the formation of orthorhombic mullite.

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1. Introduction

Mullite is an aluminosilicate refractory mineral with excellent high temperature properties including good chemical and thermal stability, low thermal expansion, low dielectric constant and high creep resistance (Schneider et al., 2008). The structural and functional properties of mullite depend on raw materials and synthesis methods. Kaolin is a low-cost starting material to form mullite through thermal treatment, which consists mainly of kaolinite. The phase transformation route of kaolinite is hence of great importance to mullite preparation. However, there is no agreement on the phase transformation routes of kaolinite to mullite. It has been generally accepted that kaolinite loses its hydroxyl at 450–600 °C to form a disordered phase metakaolinite. Metakaolinite transforms to a spinel phase at 980 °C accompanied by crystalline mullite. Crystalline mullite largely forms at temperatures higher than 1250 °C. The mullite initially formed is alumina-rich and progressively incorporates silica to approach 3Al₂O₃ · 2SiO₂ composition at higher temperatures (Brown et al., 1985). Chakraborty (2003) reported two simultaneous paths of mullitization reactions instead of a single solid-state reaction process.

Impurities such as iron oxides, muscovite and quartz affect thermal decomposition behavior of kaolinite and may change the formation route of mullite. The influence of iron oxides on mullite formation from kaolinite has been investigated extensively; it is generally accepted that iron oxides contribute to structural reorganization during mullite nucleation and Fe substitution in octahedral Al (Djemai et al., 2001; Soro et al., 2003).

The role of muscovite on the formation of mullite is not well understood. Lecomte et al. (2007) found that muscovite could enhance the mullite crystallization and grain growth at 1050 °C which was earlier than pure kaolin. Muscovite decomposes to form orthoclase below 1140 °C and orthoclase can partially decompose to leucite and quartz at around 1250 °C. The orthoclase melts incongruently at temperatures higher than 1140 °C and mullite was eventually crystallized from fused orthoclase (Lecomte and Blanchart, 2006; Mackenzie et al., 1987). In contrast, Castelein et al. (2001a) investigated a kaolin containing 17 wt.% muscovite and reported that the fused phase did not contribute to mullite formation. Similarly, quartz has been reported to have minimum influence on the formation of mullite from kaolinite (Brindley and Udagawa, 1960; Tarnvornpanich et al., 2008). The inconsistent conclusions on the mullite formation from kaolin resulted mainly from different impurities in raw materials.

The objective of this research was to investigate systematically the influences of muscovite and quartz on the formation of mullite from kaolinite. A kaolin without detectable impurities and a kaolin containing muscovite and quartz were selected to investigate the decomposition of kaolinite heated in the temperature range 1000–1500 °C. Additives including K-feldspar, K₂SO₄ and quartz were added to investigate the influence of potassium and silica impurities on the phase transformation of kaolinite.

2. Experimental section

Sample K-SX is a kaolin without detectable impurities collected from Shanxi province in China and K-SZ is a kaolin from Suzhou, Jiangsu province containing muscovite and quartz (Fig. 1). The chemical compositions of samples K-SX and K-SZ determined using an

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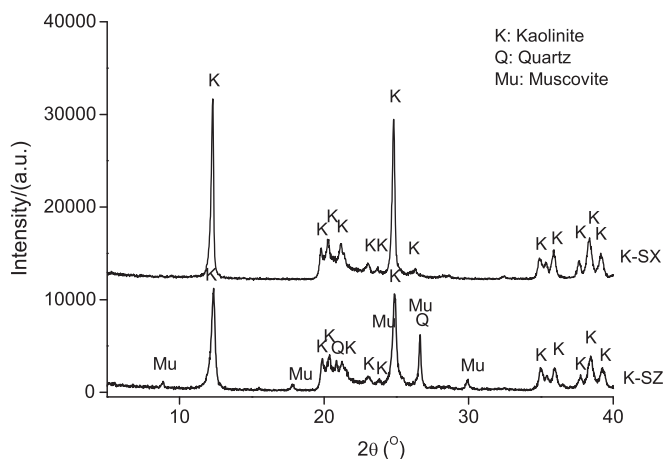


Fig. 1. X-ray diffraction traces of samples K-SX and K-SZ.

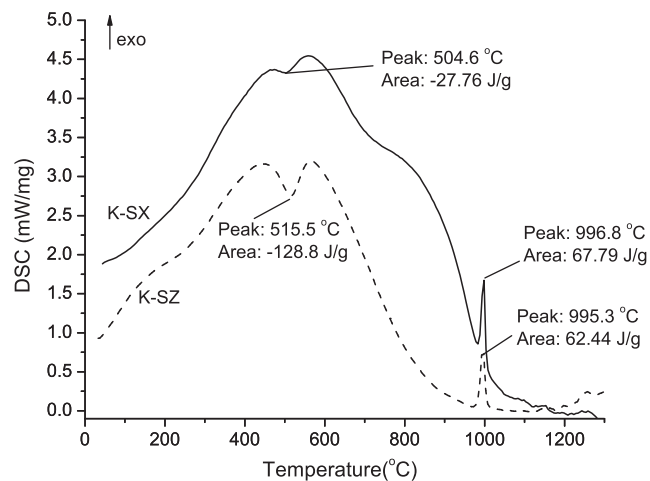


Fig. 2. DSC curves of samples K-SX and K-SZ.

X-ray fluorescence (XRF) spectrometry (Advant 3600) are listed in Table 1. Both K-SX and K-SZ were wet milled in an agate tank to pass through a 10 μm sieve and then pressed to form 10 \times 3 mm cylindrical specimens. The prepared specimens were oven-dried at 105 $^{\circ}\text{C}$ for 24 h and were then calcined at 1000, 1100, 1200, 1300, 1400 and 1500 $^{\circ}\text{C}$ for an hour. In order to investigate the influence of quartz on the formation of mullite from kaolinite, AR quartz was added to K-SX to render SiO_2 content in K-SX the same as that of K-SZ. K-SX with quartz is denoted as K-SX1 and was calcined at 1400 $^{\circ}\text{C}$ and 1500 $^{\circ}\text{C}$ for an hour. A commercially available K-feldspar ore with 71.27 wt.% KAlSi_3O_8 and AR K_2SO_4 chemical were added to K-SX to render the K_2O content in K-SX the same as that of K-SZ. K-SX with AR K_2SO_4 is denoted as K-SX2 and K-SX with K-feldspar is denoted as K-SX3. Both K-SX2 and K-SX3 were calcined at 1300 $^{\circ}\text{C}$ and 1500 $^{\circ}\text{C}$ for an hour. The treatment of sample “X-XXX” at “YYYY $^{\circ}\text{C}$ ” has been denoted as “YYYY-X-XXX” in the following text.

The mineralogical compositions of samples were identified by a D/max2550 VB/PC X-ray diffractometer (Rigaku D/MAX 2550 VB, Cu target, run at a step of $0.02^{\circ}2\theta$). DSC (Netzsch STA 449F3, 10 $^{\circ}\text{C}/\text{min}$, air atmosphere) and FTIR (Nicolet 6700, KBr disk) test methods were also used to determine sample characteristics.

3. Results and discussion

3.1. Characterization of the K-SX and K-SZ

K-SX could be considered as a pure kaolinite sample and K-SZ was a mixture of kaolinite, muscovite and quartz (Fig. 1). The weight $\text{Al}_2\text{O}_3/\text{SiO}_2$ (A/S) ratio of K-SX is 0.854 i.e. very close to the theoretical value 0.85 of kaolinite. The A/S ratio of K-SZ is 0.796. If the K_2O content in K-SZ was due only to muscovite and the Al_2O_3 in K-SZ was from kaolinite and muscovite, the weight percent of kaolinite, muscovite and quartz in K-SZ would be 88.03, 6.27 and 2.90, respectively.

The thermal weight loss of kaolinite or muscovite is attributed to the dehydroxylation process. According to the mineralogical and chemical compositions (Table 1, Fig. 1) the calculated weight losses of K-SX and K-SZ were 13.53% and 12.37%, respectively. Higher LOIs of K-SX and K-SZ in Table 1 result from the elimination of organic

carbon, as the whiteness of calcined K-SX and K-SZ increases after calcination. The DSC results showed an endothermic peak at 515.5 $^{\circ}\text{C}$ in K-SZ, related with the dehydroxylation of kaolinite (Fig. 2). The corresponding endothermic peak of K-SX, however, was at 504.6 $^{\circ}\text{C}$ and showed lower decomposition energy. A shoulder like endothermic peak also appeared at around 700 $^{\circ}\text{C}$ in K-SX. This is related with incomplete decomposition of better crystalline kaolinite in K-SX at 504.6 $^{\circ}\text{C}$ under the heating rate of 10 $^{\circ}\text{C}/\text{min}$ (Fig. 1). An exothermic peak at the temperature of 980 $^{\circ}\text{C}$ is generally associated with the formation of mullite during thermal treatments, although some researchers assign this exothermic peak to the crystalline phase transformation of $\gamma\text{-Al}_2\text{O}_3$, Al-Si spinel or the compounds of Al-Si spinel, mullite and amorphous SiO_2 (Chakraborty and Ghosh, 1978; Mazumdar and Mukherjee, 1983; Roy et al., 1955). The K-SX used in this research was a highly crystalline kaolinite without impurity; hence it could be regarded as a standard for decomposition and phase transformation of kaolinite. Mullite and spinel formed in 1000-K-SX (Fig. 3). Therefore, the exothermic peak observed at 996.8 $^{\circ}\text{C}$ corresponds to the formation of mullite and spinel. With the presence of muscovite in K-SZ it shifted to 995.3 $^{\circ}\text{C}$ which represents only the formation of spinel (Fig. 3).

3.2. Thermal decomposition characteristics of the K-SX and K-SZ

The crystalline phases in calcined K-SX are shown in Fig. 4. The amount of mullite kept increasing from 1000 $^{\circ}\text{C}$ to 1500 $^{\circ}\text{C}$ (Figs. 3, 4). Spinel formed at 1000 $^{\circ}\text{C}$, however, its concentration decreased from 1100 $^{\circ}\text{C}$ and disappeared at 1300 $^{\circ}\text{C}$. A hump between 2θ and $25^{\circ}2\theta$ was observed in the XRD traces of the samples calcined between 1000 $^{\circ}\text{C}$ and 1300 $^{\circ}\text{C}$, which gradually decreased with the temperature and disappeared at 1400 $^{\circ}\text{C}$, and cristobalite formed meanwhile. Therefore, the amorphous aluminosilicate and/or silica phase can react with spinel produced from decomposition of kaolinite before 1300 $^{\circ}\text{C}$ to form mullite. After calcination at 1500 $^{\circ}\text{C}$ the crystalline phases present were mullite and cristobalite.

During calcination spinel and quartz formed in 1000-K-SZ (Fig. 3). Mullite formed in 1100-K-SZ accompanied by the decrease in the amount of spinel (Fig. 5). However, Castelein et al. (2001b) reported

Table 1
Chemical composition of K-SX and K-SZ (wt.%).

Sample	Al_2O_3	SiO_2	TiO_2	Fe_2O_3	CaO	K_2O	Na_2O	MgO	SO_3	P_2O_5	LOI	Total
K-SX	38.32	44.86	0.42	0.19	0.07	0.05	0.03	0.13	/	/	15.84	99.91
K-SZ	37.18	46.68	0.42	0.42	0.11	0.74	0.07	0.21	0.10	0.17	13.68	99.85

LOI: loss on ignition.

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