

Effect of Er_2O_3 and Pr_6O_{11} on non-isothermal kinetics of mullite formation from kaolinite

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Abstract: The non-isothermal kinetics of mullite formation from both non-mixed and mixed with different rare earth oxides (Pr_6O_{11} and Er_2O_3) kaolinite were studied by comprehensive thermal analysis technologies and X-ray diffraction (XRD). Meanwhile, Kissinger equation, Ozawa equation and JMA modified equation (I) and (II) were employed for analysis of the effects of Pr_6O_{11} and Er_2O_3 on the kinetics of kaolinite in phase transformation at high temperatures. The results showed that the addition of two kinds of rare earth oxides influenced the crystallization kinetic parameters of kaolinite. Crystallization activation energies and frequency factors of the kaolinite mixed with Pr_6O_{11} were obviously decreased compared with the kaolinite with none, but the kaolinite mixed with Er_2O_3 weakly decreased. Crystallization behaviors were not changed, belonging to volume crystallization. Mullite formation from kaolinite was suppressed while generation of cristobalite was promoted by Pr_6O_{11} , and effect of Er_2O_3 on mullite formation from kaolinite under high temperature was not evident.

Keywords: kaolinite; mullite; Pr_6O_{11} ; Er_2O_3 ; phase transformation kinetics; rare earths

Mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) is a vital porcelain phase in traditional ceramics (such as tableware, architectural ceramics and refractories), advanced hyperthermal structural material, heat exchange material catalytic material, filter material, optical element and electronic packaging material^[1]. Whereas low breaking tenacity restricts the application of mullite in industry. Traditional synthetic route of mullite depends on solid-phase reaction between aluminum oxide and silicon oxide, in which the process was mainly diffusion controlled^[2] and the temperature mullite formation needed was relatively high ($>1500^\circ\text{C}$). Temperature and morphology of mullitization chiefly depended on particle size of raw material and preparation process of precursor before sintering^[3].

Kaolinite is a layered silicate that consists of Al-O octahedral layers and Si-O tetrahedral layers. Such asymmetrical structures result in connections between adjacent layers through hydrogen bonds along with large binding energy. Kaolinite has been widely used in ceramic industry for many years^[4,5]. Mullite with chemistry of 3:2 is the only stable crystalline in thermodynamics^[6,7]. Kaolinite is calcined at a suitable temperature to form mullite. SiO_2 in kaolinite is more than that in mullite. Moreover, when temperature is higher than 1000°C , redundant SiO_2 in kaolinite will give rise to glass phase and cristobalite phase accompanied with mullite forming

process^[8]. When temperature is higher than 1500°C , cristobalite will transform into glass phase. Therefore, the appearance of plenty of glass phase during mullite producing process with kaolinite is harmful to the mechanical properties of mullite.

Non-isothermal dynamics method arises extensive attention in research on the process of mullite formation from kaolinite, and a series of achievements have been acquired^[9-14]. Phase transformation and temperature sensing behaviors of rare earth doped solid phases are highly promising and attract more and more attention from researchers^[15,16]. Using the experience of predecessors for reference, in this paper we mixed Er_2O_3 and Pr_6O_{11} into kaolinite. By means of comprehensive thermoanalysis technologies, DTA parameters of kaolinite under conditions of different heating rates were obtained. Kissinger equation, Ozawa equation and JMA equation (I) and (II) were employed for kinetic analysis of the processes to research the effects of Er_2O_3 and Pr_6O_{11} on kinetics of mullite formation from kaolinite.

1 Experimental

The kaolinite employed in tests was from China Kaolin Corporation in Suzhou. The rare earth oxides were Er_2O_3 and Pr_6O_{11} (analytically pure) from Qiandong Rare

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Earth Company Limited by Shares from Ganzhou, and distilled water were adopted for the preparation course of thermoanalysis samples. Since Er_2O_3 and Pr_6O_{11} are insoluble in water, planetary mill was introduced for stirring and blending. The Er_2O_3 and Pr_6O_{11} amount required were calculated according to the molar weight of 100:1 between Al in kaolinite and respective rare earth element, and then these two rare earth oxides were mixed into a planetary mill to be stirred for 10 min. In order to exclude the effects of planetary mill, kaolinite equal in quality with none was placed in the planetary mill to be preprocessed at the same time. The homogeneous mixed samples were dried off in an oven at 373.15 K, and then a part of which was taken for comprehensive thermoanalysis experiment after levigation. Moreover, each sample was taken a little part separately in a muffle for two-hour heat preservation of 1200 °C at a heating rate of 10 °C/min, and then cooled to room temperature. The above calcined samples were taken for XRD testing.

The comprehensive thermoanalysis tests were conducted on a Diamond TG/DTA 6300 integrated thermal analyzer from Perkin Elmer Company in America. The samples were placed in corundum crucibles. The tests were processed at heating rates of 10, 15, 20, 25 and 30 K/min, respectively, with a warming range from 298.15 to 1373.15 K. Al_2O_3 powder calcined at 1823.15 K was chosen as reference.

Phase analysis of each sample was conducted by a DX-2700 X-ray diffractometer produced from Dandong Fangyuan Instrument Co., Ltd. Target was Cu, tube voltage was 35 kV, tube current was 25 mA, scanning mode was step scan, scan step width was 0.02°, and scanned area was from 5° to 80°.

2 Results and discussion

2.1 Thermoanalysis experimental results

TG-DTG-DTA curve graphs of different samples at a heating rate of 20 K/min are shown in Fig. 1. It is evident that endothermic peak appears approximately at 783 K in DTG curve of kaolinite, in the meantime, evident weightlessness arises in TG curve. These manifested that dehydroxylation took place from about 673.15 to 973.15 K and the kaolinite transformed into metakaolinite. With the increasing of temperature, exothermic peak appears at 1296.8 K in DTA curve due to transformation from kaolinite into mullite under high temperature condition. Devitrification exothermic peaks of kaolinite mixed with Er_2O_3 and Pr_6O_{11} , respectively, appear at 1296.4 and 1295.2 K, respectively, in DTA curve. The above indicated that the effects of incorporation of Er_2O_3 and Pr_6O_{11} on devitrification exothermic peaks of kaolinite at a heating rate of 20 K/min were not very great.

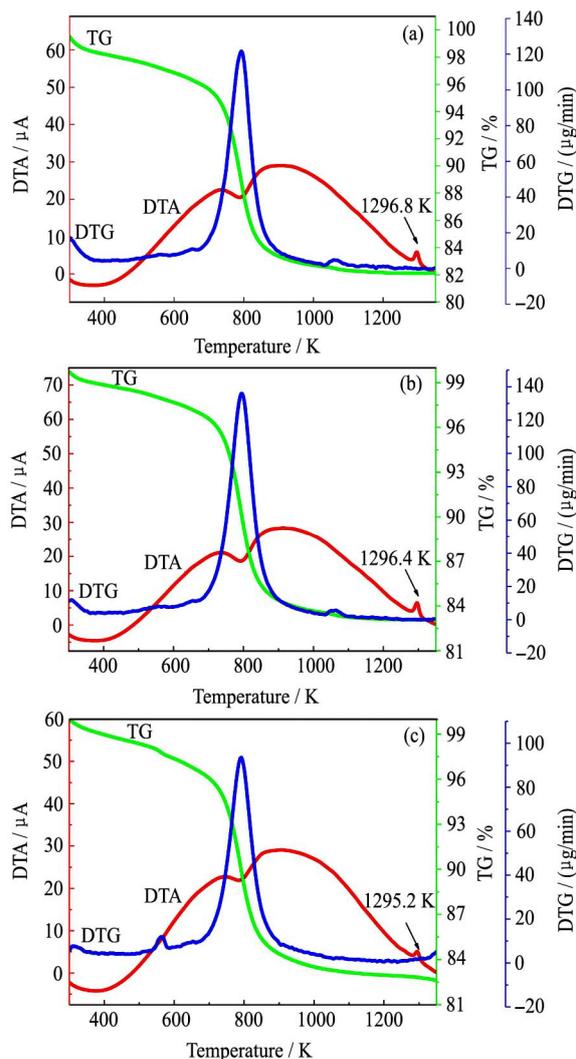


Fig. 1 TG-DTG-DTA curves of each sample at the heating rate of 20 K/min

(a) Kaolinite; (b) Kaolinite mixed with Er_2O_3 ; (c) Kaolinite mixed with Pr_6O_{11}

2.2 Effects of rare earth oxides on devitrification exothermic peaks in DTA curves

Fig. 2 displays the DTA curve phase transition of each sample at different heating rates. As shown in the figure, with the increasing of heating rate, devitrification exothermic peak of kaolinite gradually shifts toward high-temperature region. Table 1 instructs the DTA curve characteristic parameter values of devitrification exothermic peaks of each sample at different heating rates. The temperatures T_p where devitrification exothermic peaks in the DTA curves appeared of each sample at different heating rates all increased as each corresponding heating rate increased, meanwhile, kaolinite mixed with Er_2O_3 and Pr_6O_{11} slightly but not obviously decreased T_p of each heating rate. The peak widths and half-widths of devitrification exothermic peaks of each sample tended to increase on the whole followed by the increases of heating rates.

2.3 Kinetic analysis

2.3.1 Kissinger equation analysis

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