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Mullitization, microstructure and physical properties of mechanically activated andalusite sintered by microwave

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Abstract

In this work, the effects of mechanical activation and microwave heating of andalusite on mullite formation have been investigated. XRD results revealed that andalusite peaks disappeared after 60 h of milling and the peaks of alumina were observed. The formation of mullite from activated and as-received andalusite occurred at 800° and 1250 °C, respectively, while mullitization was completed at 1100° and 1400 °C in the former and latter, respectively. Mullite samples prepared from activated andalusite showed better densification with an elongated morphology.

Keywords: Andalusite; Mullitization; Mechanical activation; Microwave sintering

1. Introduction

Mullite and mullite ceramics have specific properties such as high refractoriness, low thermal expansion, good chemical stability and interesting mechanical properties. Having these excellent properties, enable us to use this compound in processing and manufacturing of traditional and advanced ceramics [1–3]. The most traditional method for synthesis of this compound is solid-state sintering at high temperatures. In this method various mixtures of aluminosilicate minerals (with an extra amount of alumina or silica) have been used [3].

Andalusite, as refractory raw materials, is one of the anhydrous aluminosilicates which has been widely used in manufacturing refractory materials containing mullite. Approximate composition of andalusite is 60 wt% Al_2O_3, 38 wt% SiO_2 and 2 wt% other impurity compounds (such as Fe_2O_3 and alkalis) [3–6].

During heating at high temperatures (> 1250 °C), andalusite converts to 80 wt% mullite and 20 wt% silica–glassy phases. The factors affecting this transformation are particle size, sintering process, mixture homogeneity, type and amount of impurities and glassy phase. Among mentioned factors, sintering process and particle size are the most important ones [2,5].

Mechanical activation is the most common method which has been developed for controlling/decreasing the particle size of materials. By introducing the defect to crystal structure of materials during mechanical activation, it is expected that the temperature and time of chemical reaction would be decreased [7–11]. The effect of attrition milling on the decomposition of andalusite in electrical furnace has been studied by Ramirez [11]. The results showed that mechanical activation leads to decrease of mullite formation temperature about 200 °C. Although it was expected that the mullite phase produced from activated andalusite would consists of nano-particles, the results were not satisfying. The reported size of mullite phase in Ramirez's research work was about 1 μm [11]. It seems that long-term heating in electrical furnace has caused the grain growth of the product.

It is predictable that unwanted grain growth could be eliminated from the system, if a faster heating method is employed. One of the fastest heating methods which have been used in materials processing is microwave heating [12–13].

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In microwave processing, energy is directly transferred to the materials through interaction of electromagnetic waves with molecules leading the heating. The main advantages of microwave heating process are: energy/time saving, unique microstructure and improve product yield [14].

In this work, the effect of long-time mechanical activation of andalusite powder on the temperature required for mullite formation, microstructure and sintering properties of fired bodies in microwave furnace was investigated.

Table 1
The chemical analysis of crushed and milled andalusite powder obtained by XRF.

Composition	Al_2O_3	SiO_2	Fe_2O_3	TiO_2	CaO	MgO	K_2O	Na_2O	L.O.I
Weight percentage	44.99	44.63	2.17	0.39	0.14	0.5	2.33	0.52	1.75

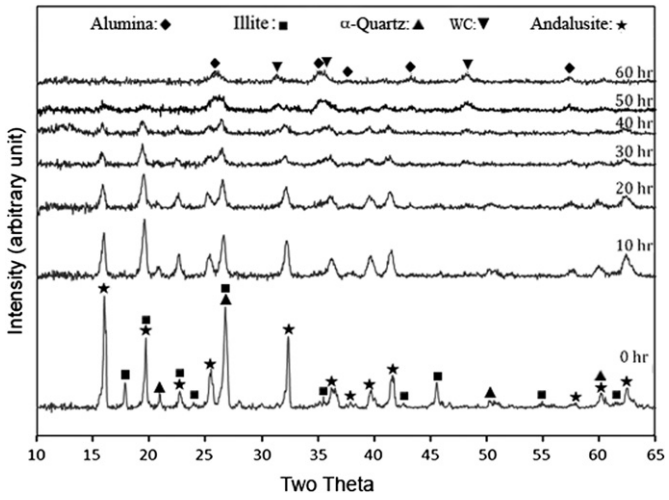

Fig. 1. XRD patterns of as-received (0 h) and milled andalusite powders after mechanical activation for different times.

2. Experimental procedures

The chemical analysis of andalusite after crushing and milling to reduce the size of particles lower than 44 μm is shown in Table 1. Fig. 1 presents X-ray diffraction pattern of as-received andalusite (0 h). Besides andalusite, the peaks of illite and quartz are observable. Activation of andalusite powder was carried out using a Retsch PM400 type planetary ball milling system. A 400 ml tungsten carbide vial and tungsten carbide balls with diameter of 20 mm were used as a milling medium at a milling speed of 300 rpm, with a ball-to-powder weight ratio of about 20:1. Stearic acid (0.1 wt%) was used as a deflocculating agent. Milled powders were pressed at 100 MP in a steel die with diameter 1 cm and thickness 0.3 cm. The pressed samples were sintered in a multimode microwave furnace (2.45 GHz and 900 W) between 800 °C and 1500 °C without holding. A SiC crucible was used as a susceptor. All runs were made by fast heating up to a maximum temperatures (heating rate 45 °C/min) using an optical pyrometer (model: RAYR312MSCL2G) [14].

Density and porosity of sintered samples were measured by Archimedes method. Phase identification of powders and samples before and after milling and heating was carried out using X-ray diffraction (Siemens D500). A microstructural observation was done with a scanning electron microscope (SEM, Vega-Tescan MV2300).

3. Results and discussion

Fig. 1 shows X-ray diffraction patterns of andalusite powders after milling for different times. As observed, the peak intensities of andalusite and quartz phases are decreased by increasing milling time. After 10, 40 and 60 h of milling, illite, quartz and andalusite completely disappeared, respectively. Alumina and WC peaks are also observed which can be attributed to the dissociation of

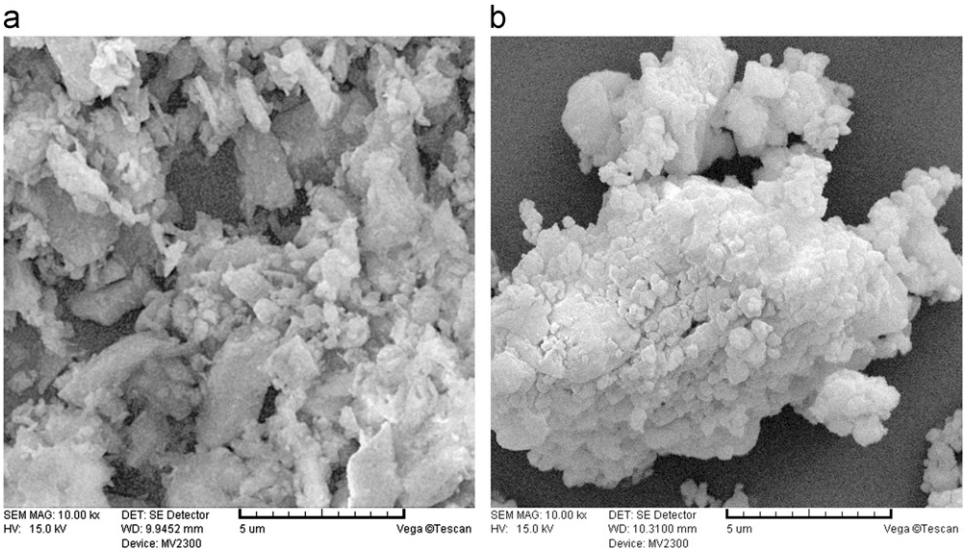

Fig. 2. SEM micrograph of andalusite powder after (a) as-received (0 h) and (b) milled for 60 h.

andalusite and wearing of WC balls after mechanical activation. The morphology of powders before and after milling is shown in Fig. 2. Before milling (Fig. 2a), the morphology of particles is different and most are angular with particle size less than 44 μm, while after milling (Fig. 2b), the agglomerated powders with fine particles are obtained. As Fig. 2 and X-ray diffraction patterns of milled powders for 60 h (Fig. 1) reveal, the effect of milling on these powders is the reduction of particle size, shape changes and loss of crystallinity.

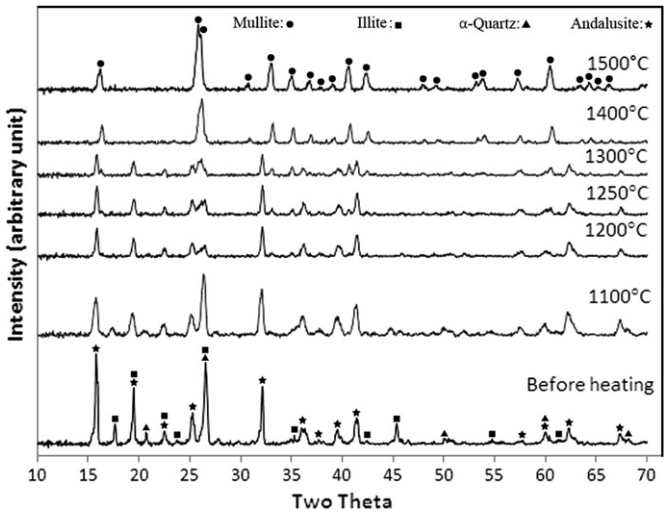

Fig. 3. XRD patterns of samples prepared from as-received powder sintered in microwave at different temperatures.

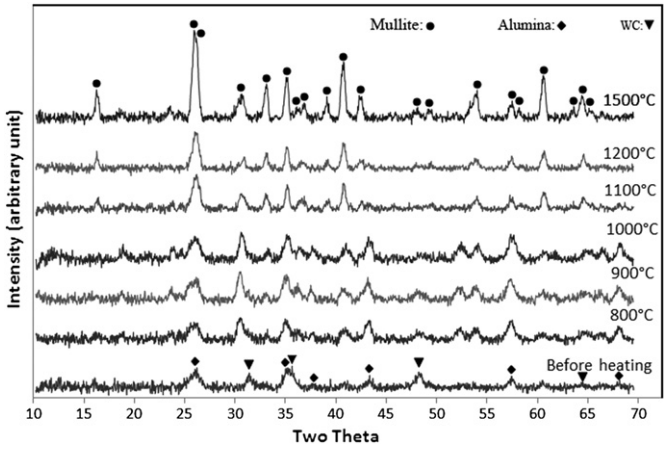

Fig. 4. XRD patterns of samples prepared from milled powder (60 h) sintered in microwave at different temperatures.

The effect of sintering temperature on mullitization behavior of as-received andalusite was carried and the results are shown in Fig. 3. As this figure reveals, andalusite, illite and quartz are phases in as-received andalusite heated at 1100 °C, these phases are observed in andalusite powder before any processing (Fig. 1). Increasing sintering temperature to 1200 °C provides no mullite phase but the complete and partially dissociation of illite and andalusite, respectively. Mullite peaks are observed in sample sintered at 1250 °C and with increasing sintering temperature to 1400 °C, mullite is found to be the only crystalline phase.

In order to study the effect of milling on phase evolution, milled powders for 60 h were selected. X-ray diffraction patterns of heated samples at different temperatures were obtained and the results are shown in Fig. 4. At 800 °C, the broad peaks of alumina and small peaks of mullite are observed. Further heating causes the increase of mullite and decrease of alumina peaks intensities. The formation of mullite is completed at 1100 °C without holding at maximum temperature.

Fig. 5 shows density and porosity of sintered samples before and after milling. As observed, in all samples the milling of andalusite resulted in an increase in density and a decrease in porosity. As Fig. 5 further reveals, the effect of milling on density and porosity of sintered samples is more recognizable at low heating temperatures between 900 °C and 1200 °C. The possible reason for this behavior can be explained from X-ray diffraction results (Figs. 3 and 4). The weak peaks of mullite are observed in as-received samples sintered at 1250 °C while the strong peaks of andalusite still remain at 1200 °C and no peaks of mullite are detectable in sintered samples at lower temperatures. In milled samples, the weak peaks of mullite are detected even at low heating temperature of 900 °C and after heating over 1000 °C, mullite are the only crystalline phase. From above results, it can be assumed that in milled samples more energy of heating consumes for sintering and

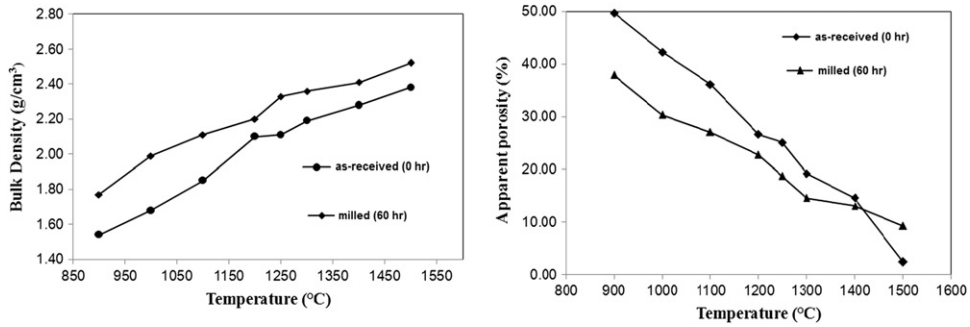

Fig. 5. Bulk density (a) and porosity (b) variations in samples prepared from as-received (0 h) and milled (60 h) powders at different temperatures.

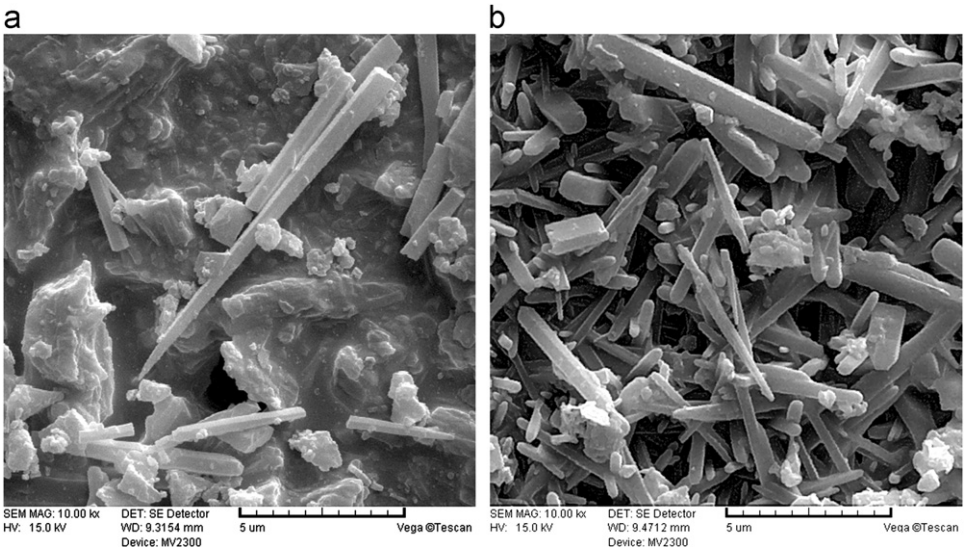

Fig. 6. SEM micrographs of sample prepared from (a) as-received (0 h) and (b) milled (60 h) powders sintered at 1500 °C.

improving density since andalusite was dissociated during milling and mullite was formed readily. In as-received samples a part of energy of heating consumes for dissociation of andalusite and crystallization of mullite and subsequently densification process is postponed.

The microstructure of as-received sample sintered at 1500 °C (Fig. 6a) contains some elongated mullite grains which are impeded in a glassy like mullite matrix. Mullite grains in milled sample sintered at 1500 °C (Fig. 6b) are found most elongated with a large aspect ratio.

4. Conclusion

The results of the present work reveal that the combination of mechanical activation and microwave heating of andalusite powder can reduce the mullitization temperature accompanied by better densification. Meanwhile, the formed mullite exhibits a microstructure of elongated grains with a large aspect ratio.

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