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Preparation of mullite whiskers reinforced SiC/Al_2O_3 composites by microwave sintering

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Abstract

Mullite whiskers reinforced SiC/Al_2O_3 composites were prepared by microwave sintering in a microwave chamber with TE_{666} resonant mode. Original SiC particles were coated with SiO_2 using sol-gel processing and mixed with Al_2O_3 particles. Mullite was formed in the reaction between SiO_2 and Al_2O_3. The isostatically pressed cylindrical pellets were sintered from 1350 °C to 1600 °C for 30 min. Physical and chemical responses were investigated by detecting changes in reflected power during the microwave sintering process. XRD was carried out to characterize the samples and showed that mullite could be formed at 1200 °C. Bridging of mullite whiskers between Al_2O_3 and SiC particles was observed by SEM and is due to a so-called local hot spot effect, which was the unique feature for microwave sintering. The optimized microwave sintering temperature was 1500 °C corresponding to the maximum amount of mullite whiskers within SiC/Al_2O_3 composites. The high electro-magnetic field enhanced the decomposition of mullite at higher temperatures above 1550 °C. The mechanical properties of mullite whiskers reinforced SiC/Al_2O_3 composites are much better than the SiC/Al_2O_3 composites without mullite whiskers.

Keywords: *microwave sintering, SiC/Al_2O_3 composites, mullite whiskers, bridging, local hot spot effect*

I. Introduction

Al_2O_3-based composites have been widely used in mechanical, chemical engineering, metallurgy, military and electronic fields because of their excellent refractoriness, low electrical conductivity, oxidation resistance, good chemical stability [1–4]. Most researches have focused on particle-dispersed Al_2O_3 composites to improve mechanical properties [3,5]. SiC, ZrO_2, TiN, TiO_2, Y_2O_3, and some metal particles (Cr) are good candidates as reinforcement for Al_2O_3 composites [6–13]. SiC/Al_2O_3 composites have recently received considerable attention due to their superior properties such as high modulus, high hardness, and good high-temperature strength [2]. Nevertheless, the oxidation of SiC into SiO_2 at high temperatures limits the application of SiC/Al_2O_3 composites. Such problem could be successfully solved by coating SiO_2 on SiC particles which facilitates the reaction between SiO_2 and Al_2O_3 to form mullite at high temperature. Mullite shows comparable thermal expansion coefficient and excellent chemical compatibility with SiC [14,15]. Mullite reinforced SiC/Al_2O_3 composites are promising materials in the field of engineering ceramics due to their excellent high temperature mechanical properties and well resistance toward chemical attack [16–20]. Pressureless sintering or hot pressing are traditionally carried out to prepare mullite reinforced SiC/Al_2O_3 composites [16,19]. The sintering temperature is very high and the mullite formation by the reaction between SiO_2 and Al_2O_3 could not be controlled. Microwave sintering as a new heating method has presently been investigated to prepare oxides and other composites [21–23]. The unique heating behaviour helps to reduce temperature and time for sintering, as well as to improve microstructure of the samples [24–26].

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In this study, microwave sintering is carried out to prepare mullite whiskers reinforced SiC/Al_2O_3 composites. Physical and chemical responses during the microwave sintering process are investigated. Mechanisms on the formation of mullite are analysed.

II. Experimental

α-SiC particles with average diameter of ~10 µm were commercially available from Henan Haixu Abrasives Co. Ltd. α-Al_2O_3 particles (~1 µm, with the purity of 99.7%) were prepared by conventional pyrolysis of aluminium hydroxide precursor at 1300 °C for 30 min. A sol-gel method was carried out to coat SiO_2 on SiC particles through the hydrolysis of tetraethyl orthosilicate, the SiO_2/SiC volume ratio was 1:5. During the coating process, SiC particles were suspended in distilled water and ultrasonicated for 30 min to break down agglomerates. The pH value of the suspension was adjusted at about 2 by HCl. Tetraethyl orthosilicate in alcohol solution was then added to the suspension with stirring for 3 h. Then $NH_3{\cdot}H_2O$ was mixed into the suspension and the pH value was controlled at 10–11. The gel of SiO_2/SiC composite precursor was formed in a few minutes. Then the coated composite particles were mixed with α-Al_2O_3 particles in the volume ratio 1:1. Mullite was expected to form by the reaction between SiO_2 and Al_2O_3 during microwave sintering.

The mixed powders were uniaxially pressed into a cylindrical pellets (ϕ = 30 mm and H = 4 mm) at 10 MPa for 1 min. After isostatically pressing at 100 MPa for 1 min, the prepared green cylindrical pellets were sintered in a microwave chamber with resonant mode of TE_{666} (WXD20S-07, China Nanjing Sanle Microwave equipment). The frequency of microwave was 2.45 GHz with a maximum input power of 10 kW. The temperature was monitored by an infrared radiation thermometer (OI-T6I2-B-1-type, GOID SUN, USA) with initial display temperature of 610 °C. An insulation structure based on a hybrid heating mode was well designed with wall of porous mullite plate, fillings of mullite fibres and aided heaters of SiC rods. Heating rate was arbitrarily controlled by tailoring the input power. Changes in reflected power versus temperature were detected. The selected sintering temperature was 1350 °C, 1400 °C, 1450 °C, 1500 °C, 1550 °C and 1600 °C for 30 min with a heating rate of 15 °C/min, respectively.

Phases of the samples were detected by X-ray diffraction (XRD; XD-3, Beijing Purkinje General Instrument Co. Ltd.). Microstructure of samples was observed by field emission scanning electron microscopy (FESEM; JEOL JSM-6700F, Japan) equipped with energy dispersive spectroscopy analysis (EDS). The samples for SEM analysis were coated with a Pt layer by a vacuum sputtering.

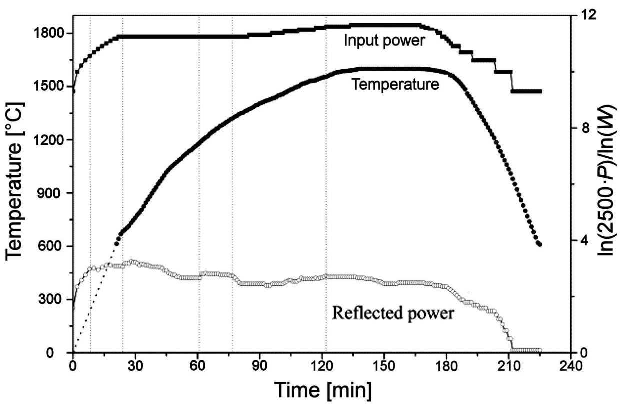

Figure 1. Heating profile for composites preparation and corresponding changes in input and reflected power

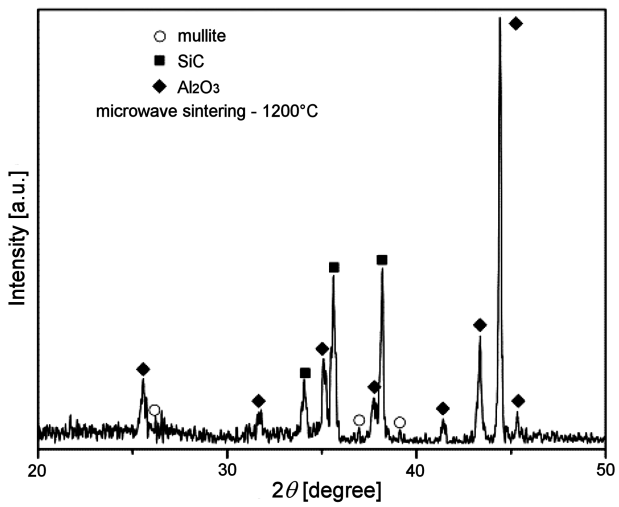

Figure 2. XRD patterns of samples prepared by microwave sintering at 1200 °C for 20 min

III. Results and discussion

Figure 1 illustrates time dependent changes in input power and reflected power together with instantaneous temperature for SiC/Al_2O_3 composites sintered at 1600 °C. Six consecutive heating stages can be distinguished, from which physical and chemical responses during the microwave sintering process could be revealed. At the initial heating stage, microwave is primarily absorbed by SiC aided heaters and SiC particles within SiC/Al_2O_3 samples since the dielectric loss of Al_2O_3 particles is very low at low temperature [27,28]. The heat generated by SiC particles within SiC/Al_2O_3 samples is isolated and gives rise to a so-called local hot spot effect during the microwave sintering [29–32]. The reflected power increases synchronously with input power, as indicated before 8 min (estimated 200 °C). It does not change obviously with the increasing input power from 8 min to 25 min. This indicates the developed microwave absorbing ability due to the increases in dielectric loss of SiC as the temperature rises. At about 25 min (~700 °C), the reflected power increases instantly. It implies that surface oxidation of SiC aided heaters initiates formation of SiO_2, which lowers microwave absorption ability. The reflected power keeps

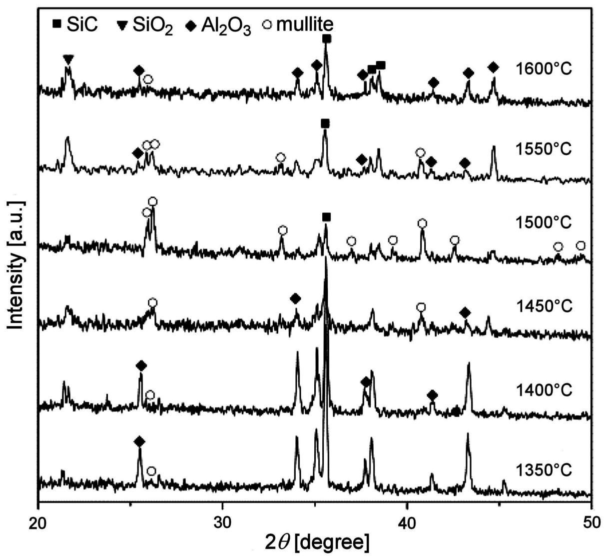

Figure 3. XRD patterns of SiC/Al_2O_3 composites sintered at different temperature for 30 min

increasing until ~30 min, indicating that the oxidation finishes after 30 min. The subsequent decrease in the reflected power appears due to the improved conductivity of SiC aided heaters and SiC particles within SiC/Al_2O_3 greens, as well as the increased dielectric loss of Al_2O_3 at higher temperature. The reflected power rises after 62 min (~1200 °C), implying the reaction between SiO_2 and Al_2O_3 to form mullite. Mullite has very low dielectric loss and does not absorb microwave energy. Such a result can be well proved from the XRD detection shown in Fig. 2. It is obvious that mullite phase appears in the samples sintered at 1200 °C for 20 min. The reflected power decreases at about 78 min (~1330 °C). This might be attributed to local fusion of SiO_2. The successive decrease in reflected power after 123 min (~1550 °C) indicates the decomposition of mullite into Al_2O_3 and SiO_2 [33].

Figure 3 shows XRD patterns of the SiC/Al_2O_3 samples prepared at different temperatures. It is obvious that SiC, SiO_2, Al_2O_3 and mullite co-exist below 1450 °C. As the sintering temperature rises, local hot spot effect is enhanced. More mullite is formed around SiC particles. The peak height referring to mullite increases versus sintering temperature. Maximum amount of mullite appears at 1500 °C. At higher temperature, mullite tends to decompose readily due to the high electro-magnetic field. As a result, the height of peaks referring to mullite decreases at 1550 °C. Further heating of the SiC/Al_2O_3 composites at 1600 °C, most of mullite peaks disappear and the peaks of Al_2O_3 phase increase significantly. Such a result is well in accordance with Fig. 1.

Figure 4 shows the SEM images of the fracture surface of the SiC/Al_2O_3 composites prepared by microwave sintering at different temperature. Needle-like mullite is observed on the surface of SiC particles at 1350 °C, as shown in Fig. 4a, which proves the abovementioned local hot spot effect. The local hot spot effect refers to that only SiC particles absorb microwave at initial heating stage and become hot. Surface temperature of SiC particles with SiO_2 coating is much higher than that of Al_2O_3 particles. Therefore, reaction between SiO_2 and Al_2O_3 only occurs locally near the surface of SiC particles. That is why needle-like mullite is formed. As the temperature increases, the local hot spot effect is enhanced. More mullite phase is generated

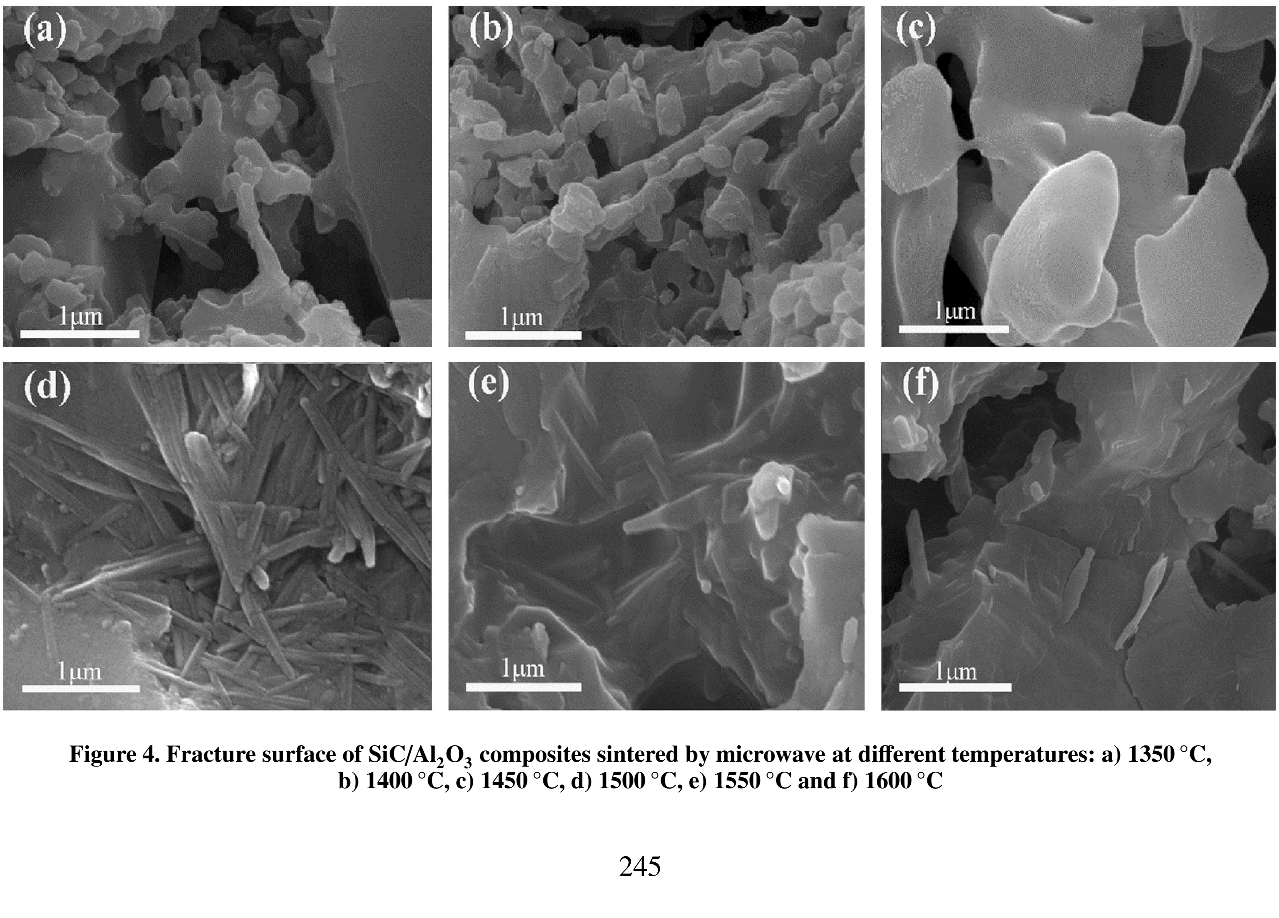

Figure 4. Fracture surface of SiC/Al_2O_3 composites sintered by microwave at different temperatures: a) 1350 °C, b) 1400 °C, c) 1450 °C, d) 1500 °C, e) 1550 °C and f) 1600 °C

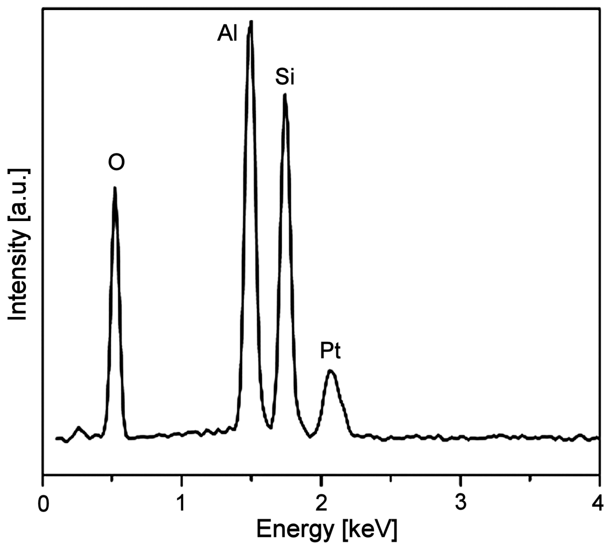

Figure 5. EDS spectrum of SiC/Al_2O_3 composites sintered by microwave at 1450 °C (presented in Fig. 4c)

and growth of mullite is developed, as shown in Fig. 4b–d. Thus, higher amount of mullite appears within the samples prepared at 1400 °C. A mullite whisker bridge is observed between Al_2O_3 and SiC particles at 1450 °C. The corresponding EDS spectrum, as shown in Fig. 5, indicates that the whiskers are composed of Al, Si and O elements. The distinctive bridging phenomenon of mullite whisker is the unique feature for microwave sintering related to the local hot spot effect. Massive well-grown mullite whiskers exist within the samples prepared at 1500 °C. Less mullite is observed within the samples prepared at 1550 °C due to its decomposition, as shown in Fig. 4e. Decomposition is enhanced at even higher temperature. As a result, most of mullite whiskers disappear within the samples prepared at 1600 °C.

Figure 6 shows the density and fracture toughness of SiC/Al_2O_3 composites sintered by microwave at different temperatures. It can be seen that, the density and fracture toughness increased with the increase of sintering temperature from 1350 °C to 1500 °C, the maximum density is 2.13 g/cm^3, and the highest fracture toughness value of the SiC/Al_2O_3 composites is 2.36 MPa·m$^{1/2}$. Combined with the XRD and SEM analysis, we can find that the mechanical properties of mullite whiskers reinforced SiC/Al_2O_3 composites are much better than the SiC/Al_2O_3 composites without mullite whiskers.

IV. Conclusions

Mullite whiskers reinforced SiC/Al_2O_3 composites were prepared by hybrid microwave sintering. The heating rate was well controlled by adjusting the input power. Mullite whiskers could be formed during microwave sintering. The local hot spot effect is the unique heating feature for microwave sintering which facilitates the directional growth of mullite whiskers bridging between SiC and Al_2O_3 particles. The optimized microwave sintering temperature was 1500 °C corresponding to the maximum amount of mullite within SiC/Al_2O_3 composites. Mullite decomposes to SiO_2 and Al_2O_3 due to the high electro-magnetic field at higher temperatures, above 1550 °C. The mechanical properties of the mullite whiskers reinforced SiC/Al_2O_3 composites are much better than the SiC/Al_2O_3 composites without mullite whiskers.

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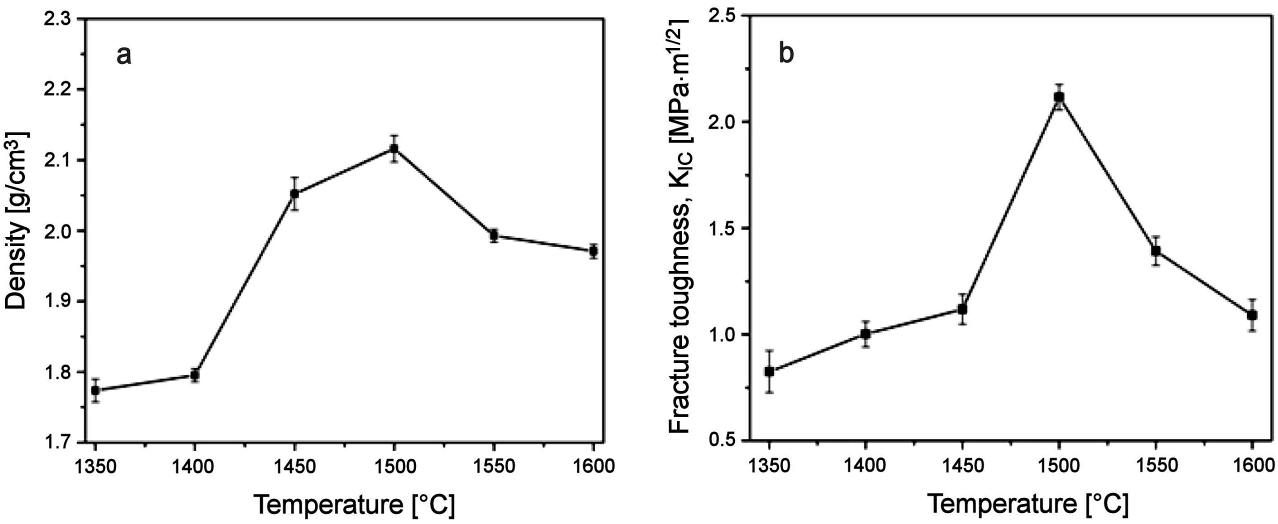

Figure 6. Density (a) and fracture toughness (b) of SiC/Al_2O_3 composites

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