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Review

Microwave-assisted sintering of zircon

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Abstract

Zircon samples were subjected to microwave heating (2.45 GHz and 900 W) in a domestic oven between 4 and 60 min and results compared to conventional heating (1300–1600 °C, holding for 120 min). Microwave heating of zircon with susceptor showed a higher temperature for any heating time compared to those heated in the absence of susceptor. To reach almost the same densification, microwave heating reduced the sintering time and temperature (16 min from room temperature to 1280 °C) compared to conventional heating (130 min from room temperature to 1300 °C and holding for 120 min at this temperature). The results also show that a long microwave heating (higher than 30 min) accelerates the decomposition of zircon. The extra phase formed acts a glassy phase during sintering of the samples; however, a lower average particle size (38.5%) was observed for the microwave-sintered samples.

Keywords: Zircon; Microwave; Sintering

Contents

1. Introduction

Microwave sintering of ceramics has been the subject of several studies [1–3]. It is known that ceramic materials can be heaped up at a very rapid rate and at a lower temperature using microwave heating compared to the conventional sintering process [4]. Microwave processing of materials is fundamentally different from traditional techniques and has several advantages over conventional sintering techniques.

In this study, the densification behaviour of zircon prepared through microwave heating was investigated to determine the advantages of microwave over the conventional heating process.

Microwave heating offers the potential of an enhanced sintering process and a decreased densification temperature as well as a shortened processing time [5–7]. Moreover, a smaller grain size can be obtained with microwave processing due to higher heating rate and shorter sintering cycles [8]. In microwave processing, energy is directly transferred to the material through the interaction of electromagnetic waves with molecules leading to heating. A dense material with improved microstructure can be obtained by microwave heating in a much shorter time compared to the conventional processes.

2. Experimental procedures

The raw material was zircon powder (Zircosil 5; Cookson Matthey Ceramics & Materials) with average particle size of 4 μm (91.5% purity). The chemical composition of zircon is reported in Table 1. Zircon powders were pressed at 63.7 MPa in a steel mold with a 20 mm diameter. The thickness of pressed powders was 1.4 mm. Samples were sintered in a domestic microwave (2.45 GHz) and compared with those sintered through the conventional heating. Fig. 1 shows the schematic diagram of microwave system used in the present work. A SiC crucible was used as a susceptor since it efficiently absorbs the microwave energy

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Table 1
Chemical composition (wt%) of zircon

SiO_2	37.81
Al_2O_3	0.44
ZrO_2	61.52
MgO	0.03
Fe_2O_3	0.04
CaO	0.04
TiO_2	0.12

due to its high loss factor [9]. The SiC crucible indirectly heats the samples to a high enough temperature and improves the sinterability of the samples. Microwave heating was carried out in air at 900 W power between 4 and 60 min. All runs were made by fast heating up to a maximum temperature measured using a R-type thermocouple placed in contact with the sample surface quickly (2 s) after turning off the power and samples were cooled down to the room temperature. The maximum error in temperature measurement was between −10 and −15 °C for temperatures lower and higher than 850 °C, respectively. At least five temperature measurements were carried out for each time of heating and measured errors became greater as the temperature went up. The density and porosity of sintered samples were measured by Archimedes method. In the conventional heating, sintering was carried out in a furnace at temperatures between 1300 and 1600 °C at a rate of 10 K/min with soaking at the maximum temperature for a period of 2 h. As-sintered samples were used for X-ray analysis (Siemens D500 with Cu Kα radiation). Scanning electron microscopy (Stereoscan 360, Lecia Cambridge Instrument) was performed on thermally etched (1300 °C, for 20 min) samples. The average grain size of sintered samples was determined from counts of 258 grains.

3. Results and discussion

The microwave absorption efficiency by zircon and susceptor was individually and together determined by the time evolution of temperature (Fig. 2). The temperature of zircon sample with susceptor (curve A) increased rapidly at a rate of about 190 °C/min in region I which can be attributed to the existence of the susceptor as the curve B determines. SiC has a higher tendency to absorb the microwave energy compared to zircon sample (curve C) which is not a good microwave absorber [10]. The heating rate decreases markedly to around 10.5 °C/min for a sample temperature higher than 992 °C (region II). As Fig. 2 reveals, the absorption of microwave energy is limited in region IV and the heating rate is approximately low (4 °C/min). The change of the heating rate of zircon sample with susceptor (curve A) between regions I and II is nearly the same as that of the susceptor (curve B). At the end of region II, the slop of curve A increases rapidly and the temperature of the sample reaches 1330 °C (25.4% increase) after 20 min heating. The thermal changes in susceptor (curve B) is different and remains nearly constant (5.5% increase) and reaches 1020 °C after the same heating regime. The above-mentioned shows the microwave absorbing effect of zircon sample with susceptor after 10 min heating. As Fig. 2 reveals, the increase of temperature after 10 min heating was not observed in zircon and susceptor which were individually heated. The thermal runaway phenomenon is unlikely to occur by the use of hybrid heating since the heating rate slows down at high temperatures, as indicated in Fig. 2. The green density of as-pressed pellets was found to be about 2.57 g/cm^3 (i.e., relative density 55%). In conventional heating, zircon samples sintered at 1400 °C with 120 min showed a relative density of 73.7%. When sintering was performed at 1600 °C for 120 min, relative density increased to 86.3% (Fig. 3). The variation in density as a function of sintering temperature for the microwave process is depicted in Fig. 4. Microwave-sintered samples exhibit enhanced densification at a lower sintering temperature and time (relative density 64.5% for samples heated from room temperature to 1087 °C in 12 min) compared to the conventional-sintered samples (relative density 56.7% for samples sintered at 1100 °C for 120 min). The temperature needed to achieve density of around 69.2% for the microwave-sintered samples (16 min heating) was 20 °C lower than that for the conventional-sintered samples (the conventional sintering was performed at 1300 °C for 120 min). For a

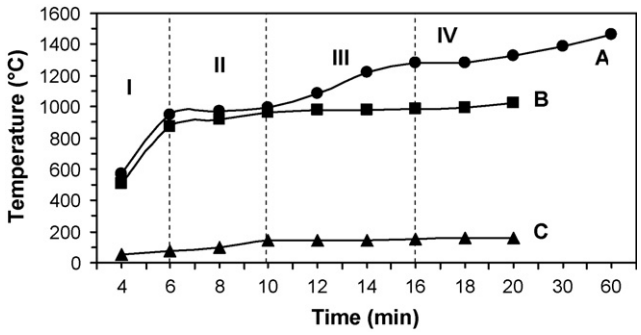

Fig. 2. Microwave absorption efficiency by zircon accompanied by susceptor (A), susceptor (B) and zircon (C).

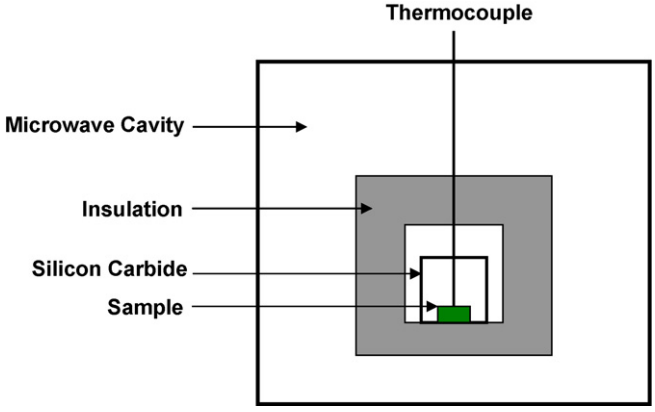

Fig. 1. Schematic diagram of microwave system.

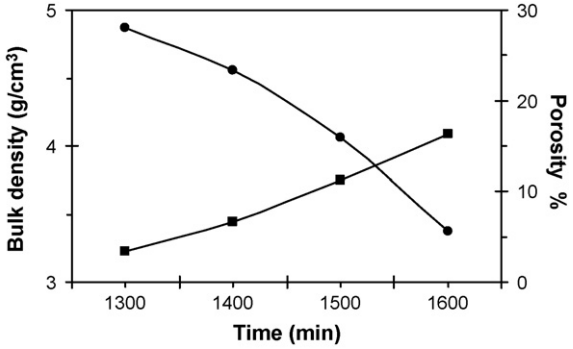

Fig. 3. Densification achieved by conventional process.

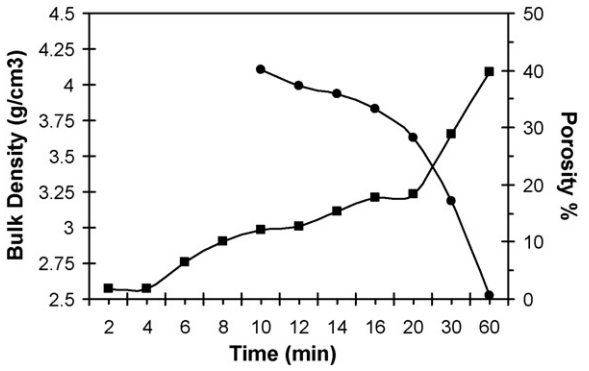

Fig. 4. Densification achieved by microwave process.

relative density of 80%, the temperature difference increased to 110 °C between the microwave-sintered sample (heated from room temperature to 1390 °C in 30 min) and conventional-sintered sample (performed at 1500 °C for 120 min). This implies that the temperature difference between microwave and conventional-sintered samples increases by increasing the time and temperature of heating. As Fig. 5 reveals, the dissociation of zircon ($ZrSiO_4 \rightarrow ZrO_2 + SiO_2$) has been accelerated in the microwave-sintered sample after 60 min heating. A low level of dissociation is observed in the samples sintered conventionally at 1600 °C (with 120 min soaking). Therefore, the microwave process may also effectively assist the dissociation of zircon which may be useful method for the synthesizing of ZrO_2 powder. The dissociation of zircon may also be contributed to the higher density exhibited by samples heated for 60 min (relative density 87.6%, porosity 0.7%) compared to conventional-sintered samples (relative density 86%, porosity 5.56%).

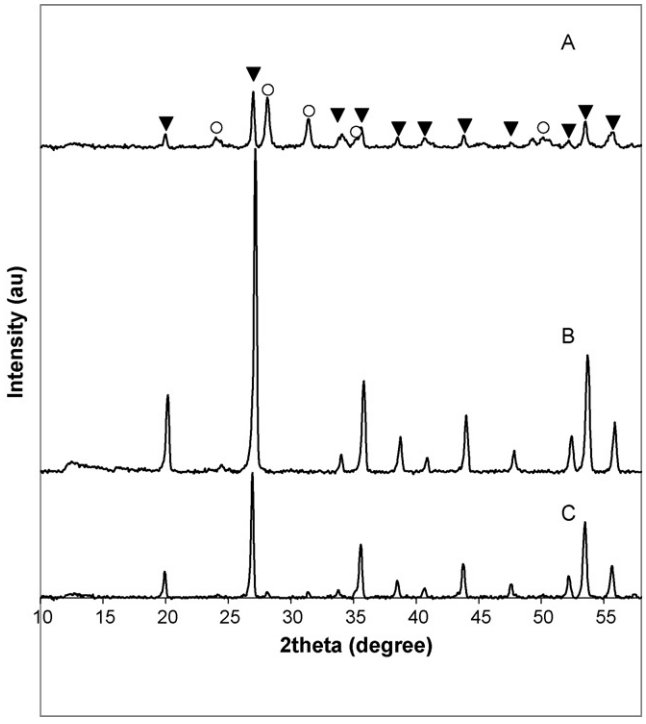

Fig. 5. X-ray diffraction patterns of microwave heated samples for 60 min (A), 30 min (B) and conventional sample sintered at 1600 °C for 2 h holding time (C) (zircon: ▼ and zirconia: ◯).

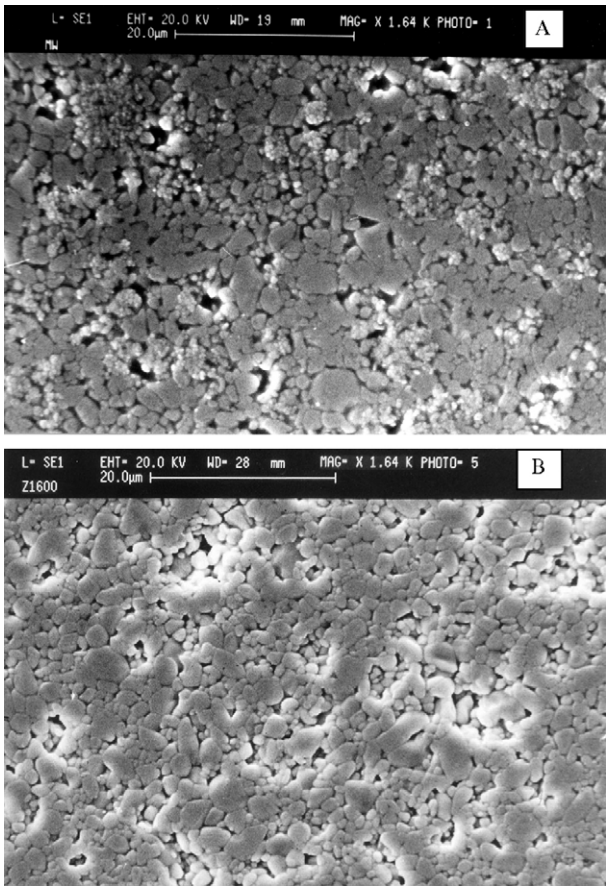

Fig. 6. Electron microscope images of samples sintered by microwave at 1460 °C (A) and conventional process at 1600 °C (B).

Scanning electron microscope images of samples sintered via microwave and conventional heating at 1460 and 1600 °C are given in Fig. 6. The rapidity of microwave method also avoids undesirable grain growth and provides the finer microstructure in spite of the existence of a glassy phase due to the dissociation of zircon (Fig. 6A). The average grain size of zircon particles was 0.8 ± 0.5 μm for the microwave-sintered sample which is lower than that for the conventional-sintered sample (1.3 ± 0.8 μm).

4. Conclusions

The results reveal that the use of 2.45 GHz microwave as a heating source improves densification kinetics of zircon materials. Microwave heating of zircon with a susceptor showed a higher temperature for any heating time compared to samples heated in the absence of the susceptor. Long microwave heating (60 min) accelerates the dissociation of zircon while gives a lower average particle size (38.5%) compared to conventional-sintered samples.

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