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Ceramics International 39 (2013) 1583–1586
CERAMICS INTERNATIONAL
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Effect of Mg doping on microwave dielectric properties of translucent polycrystalline alumina ceramic

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Received 13 July 2012; received in revised form 31 July 2012; accepted 31 July 2012
Available online 9 August 2012

Abstract

In this paper, the microstructure and microwave dielectric properties of translucent polycrystalline alumina (PCA) with various addition amounts of MgO were investigated. Translucent PCA was obtained by adding ~500–2000 ppm MgO. Compared with the undoped PCA, the translucent PCA doped with 500 ppm MgO showed a higher density and a much higher $Q \times f$ value. As the MgO content further increased, the dielectric constants (ε_r) of the translucent PCA samples showed no significant change, while the $Q \times f$ values decreased rapidly. The increased amount of impurities (MgO or spinel) was believed to be the main reason for the lower $Q \times f$ values.
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Keywords: B. Grain size; C. Dielectric properties; D. Al_2O_3; MgO

1. Introduction

In recent years, much attention has been paid to the investigation of microwave dielectric ceramics due to the rapid progress in microwave communication technology [1,2]. As a well-known dielectric ceramic, alumina (Al_2O_3) is widely used in dielectric resonators, as a ceramic substrate and in patch antennas [3,4]. Experiments show that many properties of Al_2O_3, such as thermal conductivity and dielectric constant are excellent and stable over periods of time [5–7]. However, its dielectric loss varies significantly from sample to sample. Although the purity of the powder is an important factor in producing Al_2O_3 with low dielectric loss, experiments also prove that high purity does not guarantee low loss [3–8]. Further investigations show that the major contribution to dielectric loss of Al_2O_3 are the extrinsic losses which are associated with imperfections in crystal structure, e.g., lattice disorder, point defects, dislocations, grain boundaries, random crystalline orientation, impurities, porosity and microcracks [3]. In-depth investigations made by Alford and Penn showed that both the porosity and grain size had major influences on microwave dielectric properties of Al_2O_3 [3]. They also found that by doping with 0.5 wt% TiO_2, the dielectric loss (tan δ) could decrease to 2×10^{-5}. Huang et al. discussed the effect of nano TiO_2 addition on improving the microwave dielectric properties of nano α-Al_2O_3 [4]. A very high $Q \times f$ value of 680,000 GHz could be obtained by adjusting the TiO_2 content to 0.5 wt%. Mollá et al. investigated the effect of Mg doping on the dielectric properties of Al_2O_3 [8]. A very interesting discovery was that MgO induced two different relaxation processes at very high and low frequencies, respectively. For the low-frequency process, there was a clear increase of loss tangent with MgO concentration, while for the high-frequency process there was maximum loss tangent for a concentration value of about 400 ppm. Chen et al. observed that the dielectric loss of Al_2O_3 increased with the Y_2O_3 concentration [9].

Translucent PCA has attracted great attention since it was first developed by Coble in the 1960s [10]. With its high heat resistance and high chemical durability, translucent PCA has been widely used in high-pressure sodium lamps and metal-halide lamps. Many investigations are

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http://dx.doi.org/10.1016/j.ceramint.2012.07.110

focused on further improving the real in-line transmittance and mechanical properties by using new technologies such as the spark plasma sintering (SPS) process. For instance, Kim et al. successfully obtained a high RIT of 47% via the SPS process by controlling the heating rate [11]. After that, they further improved the RIT of PCA up to 64% by applying a high pressure of 500 MPa during the SPS process [12]. Translucent PCA could also be used as a substrate and RF windows in the field of vacuum electronics and microwave circuits because of its high purity, near zero porosity, high surface smoothness, high thermal conductivity and chemical stability. However, research on the microwave dielectric properties of translucent PCA is still limited.

In this paper, translucent PCA was prepared by doping different contents of MgO. The effect of MgO on the microstructure and microwave dielectric properties of translucent PCA was investigated.

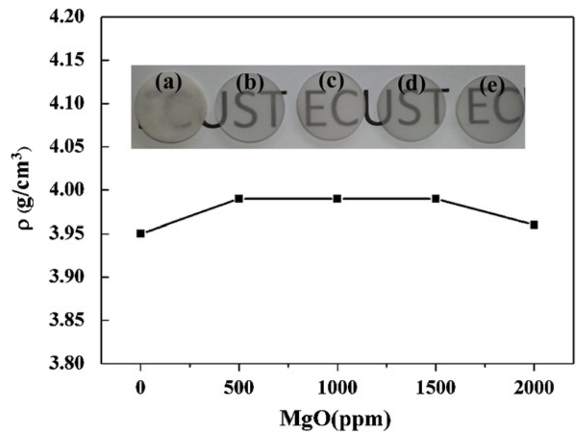

Fig. 1. Densities and photographs of the translucent PCA samples with varying amounts of MgO added (a: 0, b: 500, c: 1000, d: 1500, and e: 2000 ppm). Samples are 0.8 mm thick and polished on both sides. The text has not been retro illuminated.

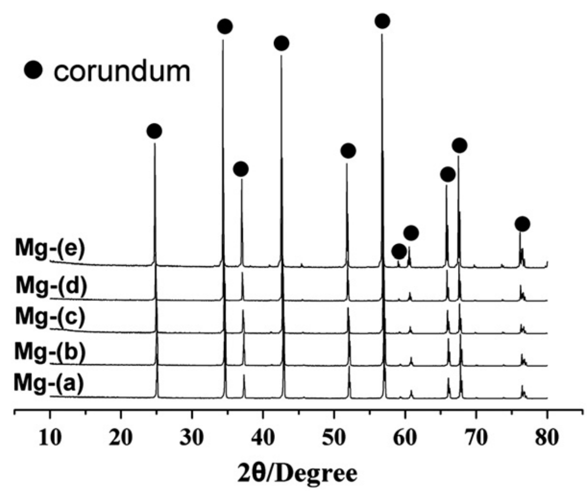

Fig. 2. X-ray diffraction patterns of the PCA with (a) 0, (b) 500, (c) 1000, (d) 1500, and (e) 2000 ppm MgO.

2. Experimental procedure

High-purity (99.99% pure) α-Al_2O_3 powder with a BET specific surface area of 7.24 m^2/g was used as the raw material. Firstly, alumina powder was dispersed into deionized water, and MgO dopants (in the form of nitrate) were introduced into the alumina powder suspension. Then, $NH_3 \cdot H_2O$ was added into the suspension until the pH value of the system reached 9.0. The prepared suspension was dried at 80 °C for 24 h, and then filtered using a 500 μm mesh nylon sieve before being pressed into pellets. The pellets were pre-fired at 1100 °C in air for 4 h to remove the binders and the final sintering was conducted at 1800 °C for 4 h in a H_2 atmosphere. For comparison, undoped specimen was prepared using the same processes but without addition of MgO.

The density of the prepared ceramic was measured by the Archimedes method. The microstructure of the samples was observed under a backscattered electron microscope (Hitachi TM3000, Japan) and the average grain size of sintered alumina pellets was calculated using the lineal intercept method on fracture surfaces [13]; 200–300 intercepts were counted for each sample. X-ray diffraction (XRD) data were collected on a Bruker D8 Advance X-ray diffractometer (Karlsruhe, Germany; Cu*K*α radiation generated at 40 kV and 40 ma), with 2θ ranging from 10° to 80° and a scanning speed of 6°/min. The dielectric constant (ε) and the quality values (Q) at microwave frequency were measured using Hakki and Coleman's dielectric resonator method modified and improved by Courtney. A vector network analyzer (E8362, Agilent Technologies, Loveland, CO, USA) was used for the measurement.

3. Results and discussion

Fig. 1 shows the densities of the prepared PCA as a function of MgO addition ranging from 0 to 2000 ppm. Initially, the density of the PCA increases as MgO is added and then remains almost constant with further addition until the MgO doping reaches 2000 ppm where a decrease is observed. This result is similar to that reported by Mollá et al. [8]. A photograph of the PCA is also shown in Fig. 1. The undoped sample (a) is almost opaque, while samples (b–e) with different amounts of MgO (500–2000 ppm) added are translucent enough that the text can be clearly seen through the samples.

The X-ray diffraction patterns of the specimens with different amounts of MgO added are shown in Fig. 2. Only the single-phase of corundum (PDF # 10-0173) exists and no peaks of second phase can be observed. However, as has been previously reported, the solid solution limitation of MgO in the Al_2O_3 is very low ($<$ 500 ppm) at this high sintering temperature [14], which renders the formation of second phase unavoidable. The reason corundum can only be detected in Fig. 2 is probably that the content of the second phase is lower than the detection limit of XRD. In fact, the presence of the second phase has been identified by SEM observation, which will be discussed in the next section.

Fig. 3(a–e) shows the SEM images of the PCA samples. All the samples show the classic equiaxed morphology. In sample (a), a significant amount of sealed pores can be observed in the matrix, while in samples (b)–(e) no pores can be detected. This is consistent with the measured lowest density of sample (a). In Fig. 3(b–e) white zones can be detected in the grain boundaries sporadically. Although the composition of the second phase is unclear and needs further investigation, this second phase can be deduced to consist of either MgO aggregates or $MgAl_2O_4$ precipitates which may form during the sintering process, as many investigators have previously pointed out [15,16].

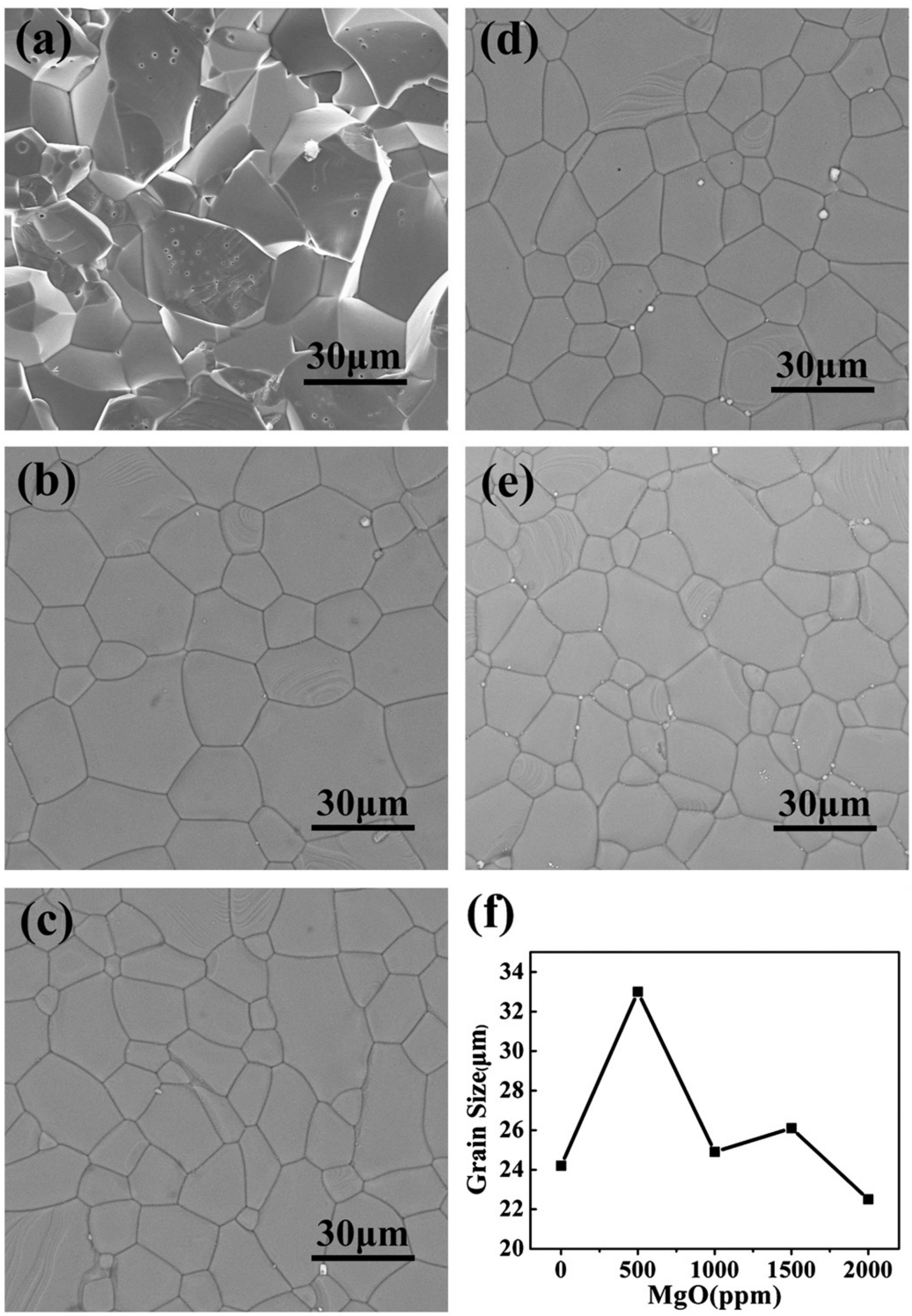

Fig. 3. Backscattered electron image of the surfaces of PCA samples doped with different amounts of MgO ((a) 0 ppm, fracture surface, (b) 500, (c)1000, (d) 1500, and (e) 2000 ppm), and average grain size (f).

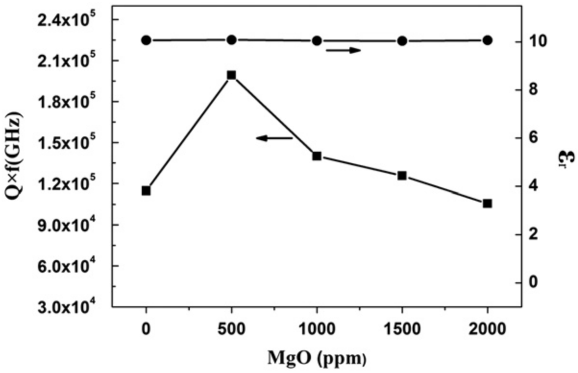

Fig. 4. Dielectric constant and quality factor values of the TPCA with different amounts of MgO.

Fig. 3(f) illustrates the dependence of the average grain size on the content of MgO addition. The maximum grain size of 33 μm can be obtained by 500 ppm MgO addition. Then the average grain size decreases as MgO content increases, which is in agreement with the results by Mollá et al. [8].

Fig. 4 shows the microwave dielectric properties of the translucent PCA as a function of different amounts of MgO added, ranging from 0–2000 ppm. The dielectric constant of the translucent PCA varies in a very narrow range of ~10.13–10.80, which is consistent with many other reports [5,8,9]. However, the variation of the $Q \times f$ value is completely different compared to previous results. First, the $Q \times f$ value increases rapidly from 115,000 to 199,231 GHz when 500 ppm MgO is added. Then, as the MgO content further increases, the $Q \times f$ value decreases sharply. Finally, with 2000 ppm MgO doped, the $Q \times f$ value becomes as low as 105,600 GHz. This result can be explained by the structural difference between the samples. As has been shown, the presence of intragranular pores could be a big contributor in the dielectric loss [17]. Therefore, sample (a) with many pores left in the grains shows a high loss and low $Q \times f$ value. Then, with 500 ppm MgO doped, the pores in the grain are effectively removed and correspondingly the $Q \times f$ value increases sharply. However, considering the very low solubility of Mg^{2+} in Al_2O_3, as the MgO content increases, more and more Mg will aggregate or become $MgAl_2O_4$ precipitates at the grain boundary, which may explain the decreased $Q \times f$ value.

4. Conclusion

(1) Translucent PCA with different grain sizes could be obtained by 500–2000 ppm MgO addition.
(2) The translucent PCA containing 500 ppm MgO yielded a significantly improved $Q \times f$ value of 199,231 GHz, which was significantly higher than that of the undoped PCA. The increase in density was believed to be the main reason for this major increase in $Q \times f$ value.
(3) Higher content of MgO could lower the $Q \times f$ value due to the existence of a second phase of MgO or $MgAl_2O_4$ at the grain boundaries.

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