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Dielectric characteristics of $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics at microwave frequencies

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ABSTRACT

Microwave dielectric properties and microstructures of $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics prepared by conventional solid-state route have been studied. The dielectric constant values (ε_r) saturated at 7.1–8.6. The $Q \times f$ values of 2000–95,000 GHz can be obtained when the sintering temperatures are in the range of 1480–1600 °C. The temperature coefficient of resonant frequency τ_f was not sensitive to the sintering temperature. The ε_r value of 8.6, the $Q \times f$ value of 95,000 GHz, and the τ_f value of −52 ppm/°C were obtained for $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics sintering at 1600 °C for 4 h $(Mg_{1/2}Zn_{1/2})Al_2O_4$ is proposed as a suitable material candidate for application in high selective microwave ceramic resonator, filter, and antenna.

1. Introduction

Recent research has been focusing on developing dielectric materials with a high-quality factor ($Q \times f$), a high-dielectric constant (ε_r), and a zero temperature coefficient of resonant frequency (τ_f) for use as dielectric resonators and microwave device substrates. High-dielectric constant materials can effectively reduce the size of resonators as the wavelength (λ) in dielectrics is inversely proportional to $\sqrt{\varepsilon_r}$ ($\lambda = \lambda_o/\sqrt{\varepsilon_r}$ where λ_o is the wavelength in vacuum). The inverse of the dielectric loss ($Q = 1/\tan\delta$) must be high to achieve prominent frequency selectivity and stability in microwave transmitter components. Moreover, a small temperature coefficient of the resonant frequency is needed to ensure the stability of the microwave components at different working temperatures. Several compounds such as $(Zr, Sn)TiO_4$, $Ba(Mg_{1/3}Ta_{1/3})O_3$, and $(Mg, Ca)TiO_3$ have therefore been developed [1–3].

The $A^{2+}Al_2O_4$ (A: Mg, Zn) ceramic family have been reported to have good microwave dielectric properties and have been of great interest as a potential dielectric resonator for microwave applications in the last decade [4,5]. Appropriate substitutions in the A site of the $A^{2+}Al_2O_4$ ceramic family to form a solid solution were investigated to achieve a high $Q \times f$ value, which allows such compounds to adapt to higher frequency applications [6,7]. The $MgAl_2O_4$ ($\varepsilon_r \sim 9$, $Q \times f \sim 69{,}000$ GHz, $\tau_f \sim -50$ ppm/°C) compound exhibits a combination of a high $Q \times f$ and a modest ε_r. In addition, $ZnAl_2O_4$ ($\varepsilon_r \sim 8.5$, $Q \times f \sim 57{,}000$ GHz, $\tau_f \sim -79$ ppm/°C) seems to be a good choice as an end member to form a solid solution with $MgAl_2O_4$ because Zn^{2+} (0.083 nm) not only shows an ionic radius similar to that of Mg^{2+} (0.078 nm) but also has the same crystal structure. Consequently, it should be of interest to conduct a comprehensive investigation of the $(Mg_{1-x}Zn_x)Al_2O_4$ ceramic system [7]. However, past experiments to produce $(Mg_{1-x}Zn_x)Al_2O_4$ ceramics have not discussed the dielectric properties of $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics.

In this paper $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics using start powders of Al_2O_3, MgO, and ZnO were synthesized by solid-state method, and the microwave dielectric properties and the microstructures of $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics were also investigated.

2. Experimental procedure

A sample of $(Mg_{1/2}Zn_{1/2})Al_2O_4$ mixed according to the desired stoichiometry was synthesized by conventional solid-state methods from individual high-purity oxide powders (>99.9%): MgO, ZnO, and Al_2O_3. The powders were ground in distilled water for 12 h in a ball mill with agent balls. All mixtures were dried, forced through a 200-mesh sieve, and calcined at 1250 °C for 2 h. The calcined reagent was ground into a fine powder for 12 h. The fine powder, together with the organic binder, was pressed into pellets with dimensions of 11 mm diameter and 5 mm thickness under a pressure of 2000 kg/cm^2. These pellets were sintered at temperatures of 1480–1600 °C for 2 and 4 h in air. Both the heating rate and the cooling rate were set at 10 °C/min. On other hand, the X-ray diffraction (XRD, Siemens D5000) data of powder and bulk samples were collected using Cu Kα radiation and a graphite monochromator in the 2θ range of 20–60°. The density of the sintered specimens, as a function of sintering temperature, was measured by the liquid Archimedes method using distilled water as the liquid.

The dielectric constants (ε_r) and $Q \times f$ values at microwave frequencies were measured using the Hakki–Coleman dielectric resonator method, as modified and improved by Courtney [8,9]. The dielectric resonator was positioned between two brass plates to form a cavity-like structure. The test cavity is placed over a thermostat and the temperature range used is +25 to +80 °C in which the heating rates were 1 °C/min for heating and the residence time is 10 min at each time. The τ_f (ppm/°C)

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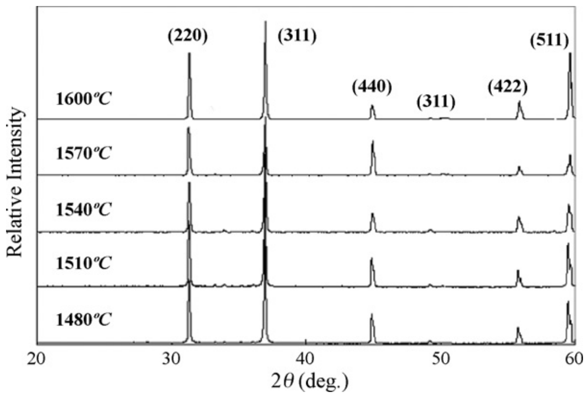

Fig. 1. XRD patterns of $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics at different sintering temperatures for 4 h.

is calculated by noting the change in resonant frequency as,

$$\tau_f = \frac{f_2 - f_1}{f_1\,(T_2 - T_1)} \qquad (1)$$

where f_1 is resonant frequency at T_1 and f_2 is the resonant frequency at T_2.

3. Results and discussions

Fig. 1 shows the XRD patterns of the $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics at different sintering temperatures for 4 h. It was observed that $(Mg_{1/2}Zn_{1/2})Al_2O_4$ exhibited a spinel crystal structure. Similar results had been reported as phases only existed for the $(Mg_{1/2}Zn_{1/2})Al_2O_4$ sintered [7]. Similar XRD patterns were detected for the $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics with various sintering temperatures. The second phase was not observed at different sintering temperatures because detection of a minor phase by XRD is extremely difficult. In addition, identical XRD patterns were observed for the ceramics irrespective of the sintering temperature.

The plot of bulk density of the $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics versus the sintering temperature and time is illustrated in Fig. 2. The density increased with increasing sintering temperature. This was because such a high sintering temperature would cause grain growth resulting in an increase in density. The increase in density may directly affect the microwave dielectric properties.

Fig. 3 demonstrates the dielectric constant of the $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics as a function of its sintering temperature and time. The dielectric constant revealed the same trend with the density as higher density means lower porosity that results in higher ε_r value. It increased with the increase of sintering temperature. In this experiment, a maximum dielectric constant of 8.6 was obtained for $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics sintered at 1600 °C for 4 h.

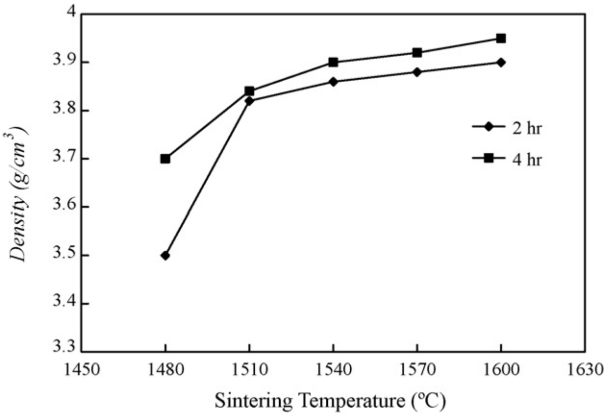

Fig. 2. Dependence of sintering condition of $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics on relative density.

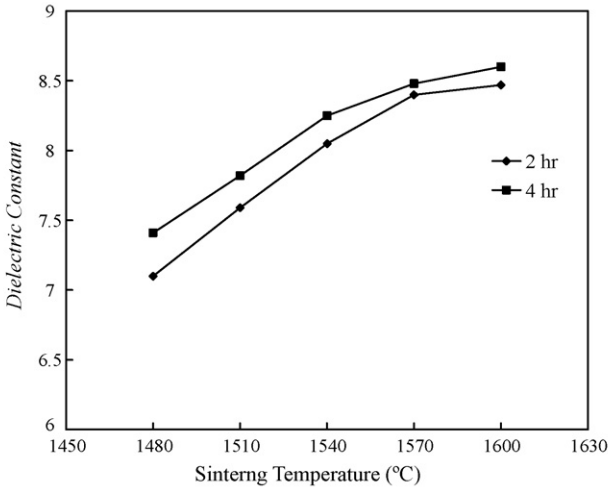

Fig. 3. Dependence of sintering condition of $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics on dielectric constant.

The $Q \times f$ value of the $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics as a function of its sintering temperature and time is illustrated in Fig. 4. The $Q \times f$ value increased from 2000 to 95,000 GHz as the sintering temperature increased from 1480 to 1600 °C. Many factors are believed to affect the microwave dielectric loss and can be divided into two categories, that is, intrinsic loss and extrinsic loss [10]. Intrinsic losses are mainly caused by lattice vibration modes, while extrinsic losses are dominated by a second phase, oxygen vacancies, grain sizes, and densification or porosity. Because the variation of the $Q \times f$ value was consistent with the density, the degradation of the $Q \times f$ value was attributed to a decrease in density. As the $Q \times f$ value revealed the same trend with the density, it implies that the variation of $Q \times f$ value was dominated by the change of density.

Fig. 5 shows the temperature coefficient of resonant frequency (τ_f) of the $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics as a function of its sintering temperature. The temperature coefficient of resonant frequency (τ_f) is well known to be related to the composition and the secondary phase of a material. Since the composition remained unchanged and no secondary phase was detected, no significant change in the τ_f value was observed as expected. The τ_f value varied from −41 to −58 ppm/°C for the various sintering temperatures and times. The temperature coefficient of resonant frequency (τ_f) is well known to be related to the composition and the second phase

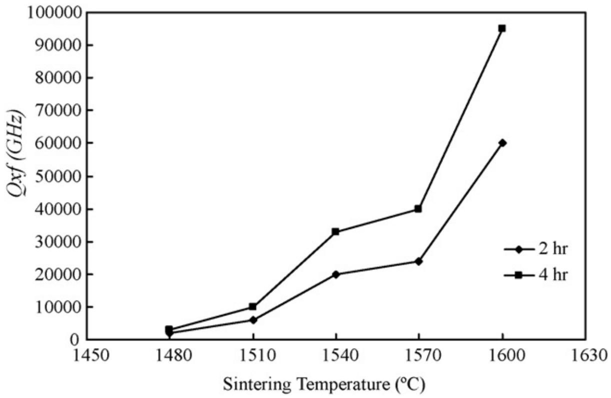

Fig. 4. Dependence of sintering condition of $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics on quality factor ($Q \times f$).

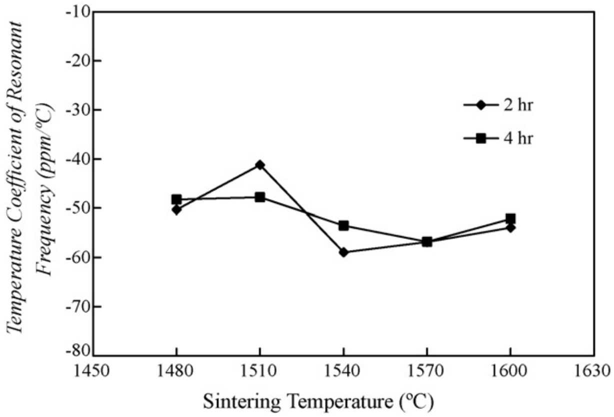

Fig. 5. Dependence of sintering condition of $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics on τ_f.

of a material. At 1600 °C, a τ_f value of −52 ppm/°C was measured for $(Mg_{1/2}Zn_{1/2})Al_2O_4$ sintered for 4 h.

4. Conclusion

The microwave dielectric properties of $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics were investigated. Compared to previous reports, a significant improvement in the dielectric properties has been accomplished. Excellent microwave dielectric properties ($\varepsilon_r \sim 8.6$, $Q \times f$ value ~ 95,000 at 14 GHz and τ_f value ~ −52 ppm/°C) can be obtained for $(Mg_{1/2}Zn_{1/2})Al_2O_4$ sintered at 1600 °C for 4 h. Comparing to past results of $(Mg_{1-x}Zn_x)Al_2O_4$ which possesses $\varepsilon_r \sim 8.56$, $Q \times f \sim 106{,}000$ GHz, and $\tau_f \sim -63$ ppm/°C at $x = 1.0$ with sintering temperature 1650 °C for 3 h, new $(Mg_{1/2}Zn_{1/2})Al_2O_4$ ceramics also can find applications in microwave devices requiring low sintering temperatures and excellent microwave dielectric properties

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