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Dielectric Properties of Low-sintering-temperature $Bi_{12}CeO_{20}$–$BiFeO_3$ Ceramics at Microwave Frequency

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Keywords: low sintering temperature, solid-state method, microwave dielectric properties, $Bi_{12}CeO_{20}$–$BiFeO_3$ ceramic, antenna temperature sensor

In this study, we prepared $(1 - x)Bi_{12}CeO_{20}$–$xBiFeO_3$ ceramics using conventional solid-state methods with various material compositions and sintering temperatures. X-ray diffraction and scanning electron microscopy were employed to analyze the composition of the ceramic microstructures. In experiments, as x increased, the dielectric constant and τ_f value increased, and the $Q \times f$ value decreased. We also investigated the correlation between the relative proportions of two ceramic substances and their microwave dielectric properties. The $Bi_{12}CeO_{20}$–$BiFeO_3$ ceramic system is suitable for use as a substrate in low-temperature cofired ceramic antenna temperature sensors.

1. Introduction

Advances in wireless temperature sensing modules led to them replacing traditional thermocouple sensors with low-temperature cofired ceramic antennas. For example, Zhang *et al.* constructed an antenna sensor from dielectric ceramic materials with a temperature coefficient (τ_f) of 248 ppm/°C.(1) Additionally, Sanders *et al.* used dielectric ceramic materials with $\tau_f = -160$ ppm/°C to construct ceramic antenna sensors.(2) In these dielectric ceramic sensors, the ambient temperature of the sensing antenna can be calculated using the resonant frequency of the sensor antenna, and the τ_f value is high. The resonance of the low-temperature cofired ceramic antenna temperature sensor S_{11} is larger than those of the others.

Bi_2O_3–SiO_2 dielectric materials have a sintering temperature lower than 960 °C and can be fabricated using low-temperature co-firing processes.(3–6) Moreover, Bi_2O_3–SiO_2 has a dielectric constant of approximately 40.(3–6) One study indicated that $Bi_{12}CeO_{20}$ microwave dielectric materials have optimal properties when sintered at 780 ºC for 5 h, with a dielectric constant of 27, a $Q \times f$ value of 10640 GHz, and a τ_f value of −184.4 ppm/°C.(7) In addition, several groups have examined the use of $BiFeO_3$ in communication components.(8–11) In this study, we fabricated a $Bi_{12}CeO_{20}$–$BiFeO_3$ substrate for ceramic

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antenna temperature sensors and evaluated its microwave dielectric properties. We then analyzed the density, X-ray diffraction (XRD) patterns, and microstructures of the ceramics. Finally, we investigated the correlation between the substrate's dielectric properties and the microstructural composition.

2. Materials and Methods

In this study, we employed the following procedure. First, the $(1 - x)Bi_{12}CeO_{20}$–$xBiFeO_3$ microwave dielectric ceramic systems were fabricated per corresponding stoichiometric proportions and synthesized by conventional solid-state methods from individual high-purity (>99.9%) oxide powders of Bi_2O_3, CeO_2, and Fe_2O_3. Second, the powders were ball-mixed in distilled water for 12 h in a plastic bottle with agate balls. Third, all wet mixtures were dried and calcined at 700 °C for 5 to 6 h. Fourth, the calcined powder was pressed into pellets 11 mm in diameter using polyvinyl alcohol as a binder. Fifth, the pellets were sintered at 740–780 ºC for 2 h in open air. Sixth, XRD data on the bulk samples were collected using Cu-Kα radiation in the 2θ range from 20 to 60°. Seventh, microstructural analysis of the sintered surfaces was performed using a scanning electron microscope. Finally, the pellets' dielectric properties at microwave frequencies were measured using a modification of the Hakki–Coleman dielectric resonator method by Courtney.[12,13]

3. Results and Discussion

Figure 1 shows the room-temperature XRD patterns recorded for the $(1 - x)Bi_{12}CeO_{20}$–$xBiFeO_3$ ceramic system sintered at 780 °C for 2 h. XRD patterns revealed a two-phase system containing $Bi_{12}CeO_{20}$ and $BiFeO_3$. Increasing x increased the peak density of $BiFeO_3$, producing a second phase of $Bi_{25}CeO_{40}$. The lattice parameters of $Bi_{12}CeO_{20}$ did not change upon varying x, confirming the existence of a two-phase system.

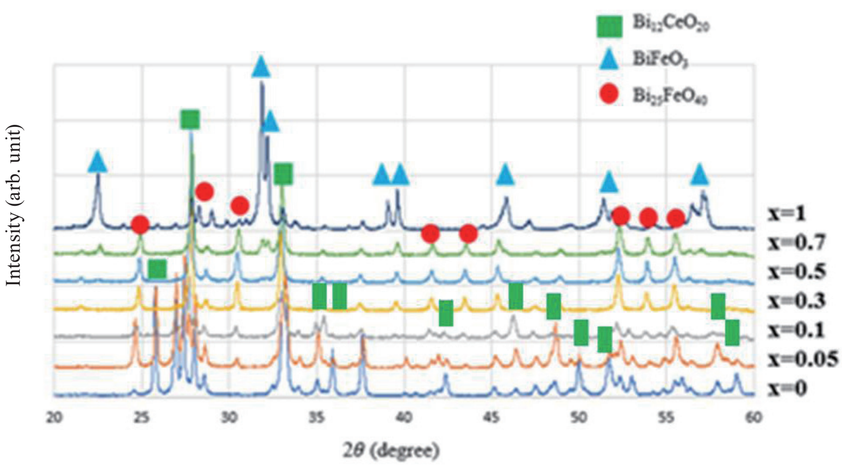

Fig. 1. (Color online) X-ray diffraction patterns of $(1 - x)Bi_{12}CeO_{20}$–$xBiFeO_3$ dielectric ceramics with various values of x.

Figure 2 shows the bulk density of the (1 − *x*)$Bi_{12}CeO_{20}$–*x*$BiFeO_3$ ceramics under different sintering temperatures with various values of *x*. The bulk density increased as *x* increased, owing to the higher theoretical densities of $BiFeO_3$ and $Bi_{25}FeO_{40}$ than that of $Bi_{12}CeO_{20}$.(14) Higher sintering temperatures enhanced the grain growth, and thus, the density. Furthermore, changes in density directly affected the microwave dielectric properties, particularly the dielectric constant.

The dielectric constants of the (1 − *x*)$Bi_{12}CeO_{20}$–*x*$BiFeO_3$ ceramic system under different sintering temperatures with various values of *x* are shown in Fig. 3. Similar to density, the dielectric constant increased as temperature and *x* increased, suggesting that the density and composition of the specimen exerted a strong effect on the dielectric constant. As *x* varied from 0 to 0.5, the dielectric constant increased from 27.9 to 43.8. Additionally, after sintering at a temperature of 780 °C for 2 h, the *εr* value for 0.5$Bi_{12}CeO_{20}$–0.5$BiFeO_3$ was 43.8.

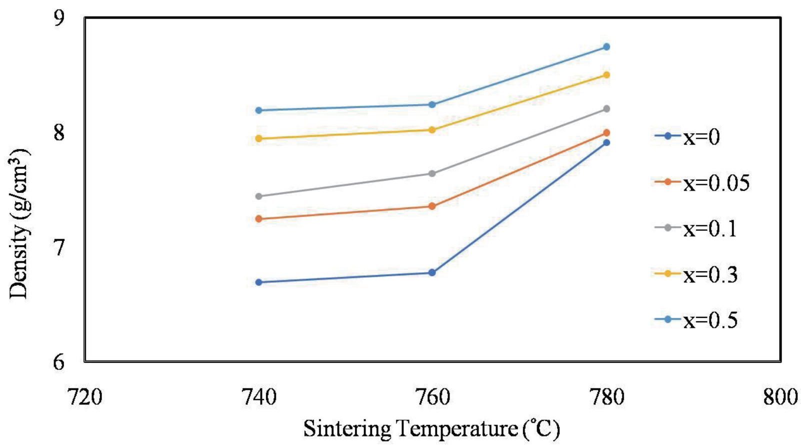

Fig. 2. (Color online) Density of (1 − *x*)$Bi_{12}CeO_{20}$–*x*$BiFeO_3$ thin films at various sintering temperatures and values of *x*.

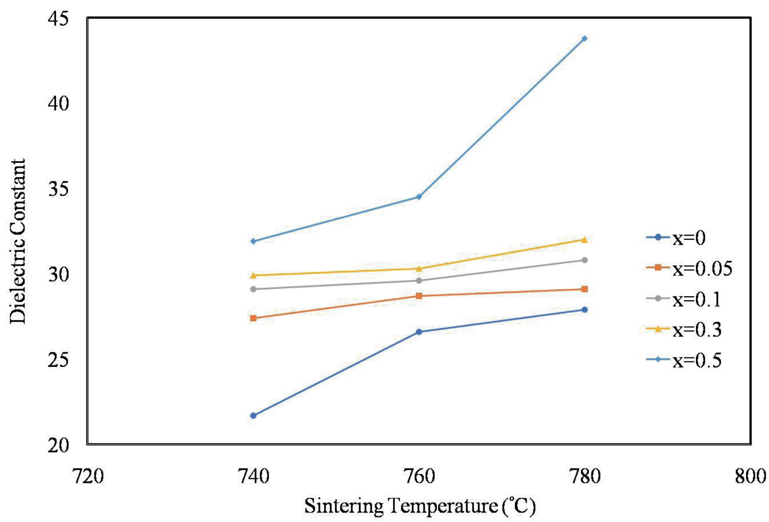

Fig. 3. (Color online) Dielectric constant of (1 − *x*)$Bi_{12}CeO_{20}$–*x*$BiFeO_3$ thin films at various sintering temperatures and values of *x*.

Figure 4 shows the quality factor values ($Q \times f$) of the $(1 - x)Bi_{12}CeO_{20}$–$xBiFeO_3$ ceramic system. Several factors contribute to dielectric loss at microwave frequencies, including density, porosity, second phases, and grain boundaries.(15) The $Q \times f$ value is related to x because $BiFeO_3$ possesses a much lower $Q \times f$ than $Bi_{12}CeO_{20}$. Specifically, as x increased from 0 to 0.5, $Q \times f$ decreased from 11000 to 500 GHz. Moreover, $Q \times f$ increased with sintering temperature for all values of x. Additionally, the decrease in $Q \times f$ at high sintering temperatures was attributable to increased grain growth.(16) Bulk density also increased and decreased with sintering temperature and x, respectively.

Figure 5 shows the temperature coefficients of resonant frequency (τ_f) of the $(1 - x)Bi_{12}CeO_{20}$–$xBiFeO_3$ ceramic system after sintering at various temperatures with values of x for 2 h. The composition of dielectric materials directly affects the τ_f value. Increasing the proportions of $BiFeO_3$ increased the system's overall τ_f value because the τ_f values of $Bi_{12}CeO_{20}$

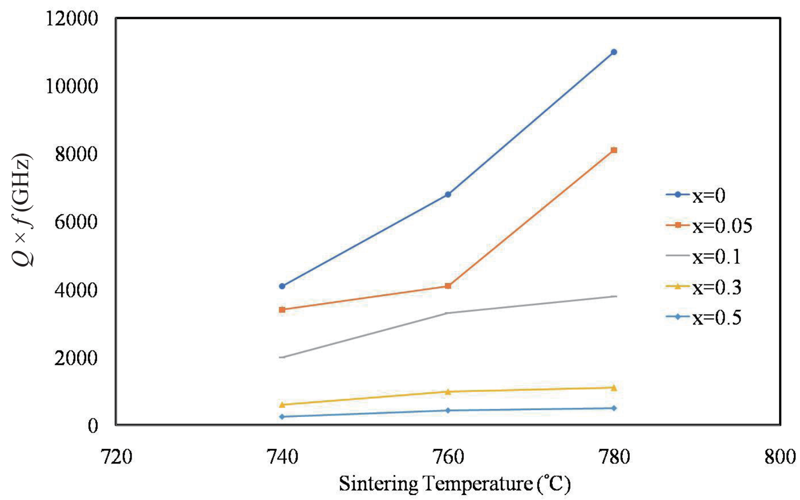

Fig. 4. (Color online) $Q \times f$ value of $(1 - x)Bi_{12}CeO_{20}$–$xBiFeO_3$ thin films at various sintering temperatures and values of x.

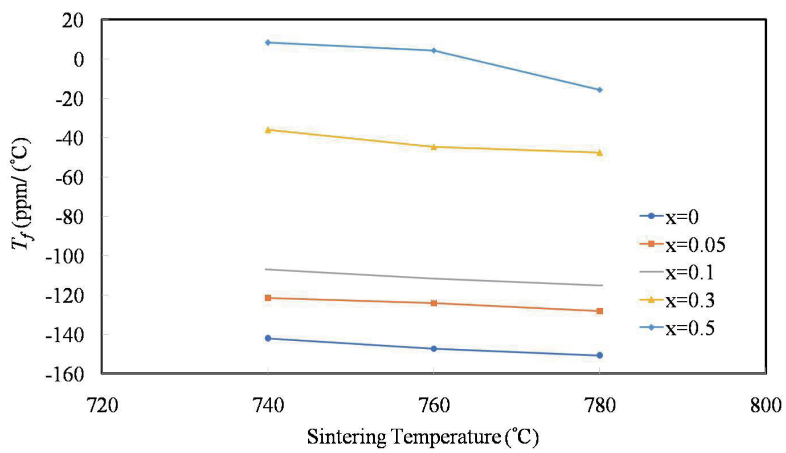

Fig. 5. (Color online) τ_f value of $(1 - x)Bi_{12}CeO_{20}$–$xBiFeO_3$ dielectric ceramics at various sintering temperatures and values of x.

and $BiFeO_3$ are negative and positive, respectively. The τ_f values of the $(1-x)Bi_{12}CeO_{20}$–$xBiFeO_3$ ceramic system varied from −151 to +8.3 ppm/°C under various sintering temperatures. According to a previous study,(15) this variation in the τ_f value is attributable to factors that are extrinsic and intrinsic to the $(1-x)Bi_{12}CeO_{20}$–$xBiFeO_3$ ceramics. This variation in τ_f values produced a wide process window, indicating that the $(1-x)Bi_{12}CeO_{20}$–$xBiFeO_3$ ceramic system was suitable for use as a substrate in antenna temperature sensors.

4. Conclusions

In this study, we investigated the microwave dielectric properties of $(1-x)Bi_{12}CeO_{20}$–$xBiFeO_3$ ceramics. X-ray analysis revealed a $Bi_{25}CeO_{40}$ secondary phase with x from 0.05 to 0.5. The $Bi_{25}CeO_{40}$ secondary phase affects the ceramics' microwave dielectric properties, increasing the dielectric constant and τ_f value but decreasing the $Q \times f$ value as the proportion of $BiFeO_3$ increases. Under all tested values of x in $(1-x)Bi_{12}CeO_{20}$–$xBiFeO_3$, the density, dielectric constant, and the $Q \times f$ value increased as the sintering temperature increased. $Bi_{12}CeO_{20}$–$BiFeO_3$ ceramics are thus suitable substrates in a diverse range of applications requiring antenna temperature sensors made from low-temperature cofired ceramics.

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