A Tailored Investigation for (Ba,Sr)TiO_3 FGMs

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Abstract

$SrTiO_3$ is usually added as shifters in order to move the T_C of $BaTiO_3$ to lower temperatures because it is well established that the T_C of $BaTiO_3$ decreases linearly with a solid solution of Sr^{+2} in place of Ba^{+2}. It is not fully understood yet, however, how $SrTiO_3$ influences on the peak value of the dielectric constant (ε_{max}) at the T_C of $BaTiO_3$. This research reports the effect of $SrTiO_3$ addition on ε_{max} at the T_C of $BaTiO_3$ ceramics. Based on the chemical composition and the grain size dependence of the dielectric property of $BaTiO_3$ ceramics, functionally graded (Ba,Sr)TiO_3 composites were designed and fabricated. Multi-layered (Ba,Sr)TiO_3 composites with a compositional gradient of $SrTiO_3$ exhibited a low temperature coefficient and high dielectric constant in a wide temperature range.

Keywords : $BaTiO_3$, dielectric constant, FGM

1. Introduction

Pure $BaTiO_3$ shows a paraelectric to ferroelectric phase transition at 120°C, which is accompanied by a sharp peak in the dielectric permittivity. Partial substitution of either Ba ions or Ti ions is often employed to modify the nature and temperature of the paraelectric-ferroelectric transition for particular applications. $SrTiO_3$ is usually added as a shifter in order to move the Tc to lower temperatures because it is well established that the Tc of $BaTiO_3$ decreases linearly with solid solution of Sr^{+2} in place of Ba^{+2}.

It is not fully understood yet, however, how the $SrTiO_3$ concentration effects on the peak value of dielectric constant at the Tc of $Ba_{1-x}Sr_xTiO_3$ solid solutions. Many researchers reported lots of disagreement in the variation of the maximum dielectric constant at the Tc of $Ba_{1-x}Sr_xTiO_3$ solid solutions by increasing Sr content because they did not consider the grain size effect.

Since Kniekamp and Heyang[1] revealed a rather significant dependence of the dielectric permittivity of $BaTiO_3$ ceramics on their grain size, the effect of grain size on the dielectric properties of $BaTiO_3$ has been an active area of research. Nowadays, it is generally accepted that the maximum dielectric permittivity at the Curie temperature decreases with the decrease of the average grain size of $BaTiO_3$. Therefore, when the effects of $SrTiO_3$ content on the dielectric properties is investigated, the grain size effect should be considered. This is the reason why such disagreements occurred because the grain size effect with the change of $SrTiO_3$ content and sintering conditions was not taken into account.

If the Curie point can be changed as a function of position by grading its composition, the transition from the ferroelectric to the paraelectric phase would be broadened with respect to the temperature. Consequently, the temperature coefficient of the dielectric constant could be lowered.

This paper reports the variation of the dielectric constant maximum of $Ba_{1-x}Sr_xTiO_3$ ceramic solid solutions at the Tc in the range of x=0~0.6. Different sintering temperatures were used in order to investigate the influence of the microstructure on the dielectric properties of the materials. Based on this research, multi-layered $BaTiO_3$-$SrTiO_3$ composites were fabricated by using the concept of FGMs in order to lower the temperature coefficient of the dielectric constant.

2. Experimental and Results

Specimens were prepared from commercial $BaTiO_3$ and $SrTiO_3$ (Ferro Corp., Penn Yan, NY, USA) powders. Powder slurries were made by mixing $(BaTiO_3)_{1-x}(SrTiO_3)_x$ (in unit of mol%) powder mixtures (where x = 0.1, 0.2, 0.3, 0.4, 0.5, 0.6) for 24 h in a polyethylene bottle with ethyl alcohol and zirconia balls. The dried slurry was crushed in an agate bowl and sieved to 125 μm. Powders were slightly pressed into disks 10 mm in diameter and 2 mm in thickness and then isostatically pressed again under 150 MPa. The compacts were sintered at 1350°C, 1400°C and 1450°C, respectively, for 1 hour in air. In order to fabricate 21-layered $Ba_{1-x}Sr_xTiO_3$ FGMs, the prepared powders (x = 0 ~ 0.4, step of 0.02) were stacked sequentially with stepwise and pressed into 10×10×10 mm. The compacts were sintered at 1400°C for 1 h in air. The dielectric properties of the specimens with silver electrodes were measured by an impedance/gain-phase analyzer (HP 4194A) at 1kHz from -150°C to 150°C.

Figure 1 shows the dielectric constant versus temperature of $Ba_{1-x}Sr_xTiO_3$ sintered specimens. In the case of the specimen sintered at 1350°C, the Tc and the dielectric constant maximum decreased with increasing $SrTiO_3$ content. When the sintering temperature was increased to 1400°C, however, the dielectric constant maximum increased with an increase in the x value, reaching highest value at around x = 0.4 and then decreased. In the case of the specimens sintered at 1450°C, the dielectric constant maximum monotonically increased with increasing $SrTiO_3$ concentration up to x = 0.6.

Figure 2 shows the relationship between the dielectric constant maximum of $Ba_{1-x}Sr_xTiO_3$ sintered specimens and their average grain size. From this figure, the dielectric constant maximum at the Tc of $Ba_{1-x}Sr_xTiO_3$ ceramics was found to be dependent on $SrTiO_3$ concentration and their average grain size. The $SrTiO_3$ affects contrarily on the peak value of the dielectric constant when it is incorporated into $BaTiO_3$. On the one hand, the intrinsic dielectric constant maximum of $Ba_{1-x}Sr_xTiO_3$ increases with increasing $SrTiO_3$ concentration as Jaffe[2] reported. On the other hand, the addition of $SrTiO_3$ reduces the peak value of the dielectric constant of $Ba_{1-x}Sr_xTiO_3$ ceramics by decreasing their average grain size.

Figure 3 shows the dielectric property of 21-layered $Ba_{1-x}Sr_xTiO_3$ (x = 0 ~ 0.4, step of 0.02) FGM sintered at 1400°C for 1 hour. The linearity of the dielectric constant was remarkably improved in the range between 20° and 120°C.

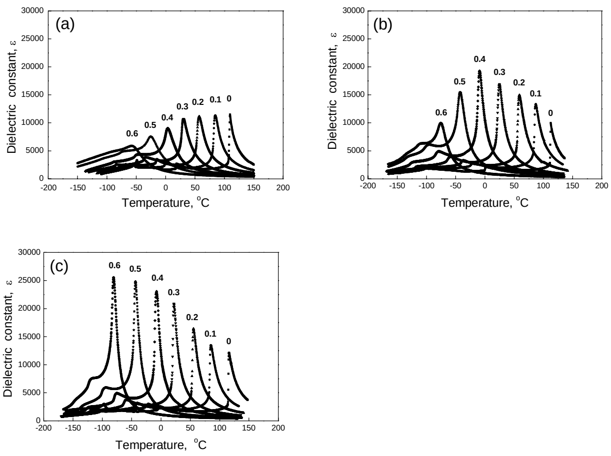

Fig. 1. Dielectric property of $Ba_{1-x}Sr_xTiO_3$ ceramics sintered at (a) 1350°C, (b) 1400°C, and (c) 1450°C for 1 hour.

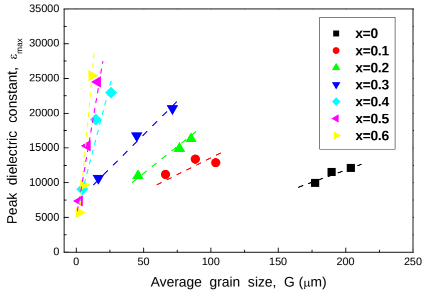

Fig. 2. Relationship between the dielectric constant maximum of $Ba_{1-x}Sr_xTiO_3$ ceramics and their average grain size.

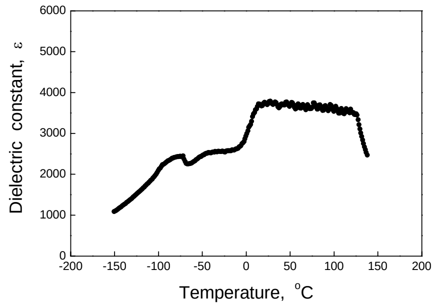

Fig. 3. Dielectric constant of a 21-layered $Ba_{1-x}Sr_xTiO_3$ functionally graded material sintered at 1400°C for 1 hour.

3. Summary

In this study, the dielectric constant maximum at the Tc of $Ba_{1-x}Sr_xTiO_3$ ceramics was found to be dependent on $SrTiO_3$ content and their average grain size. Functionally graded $Ba_{1-x}Sr_xTiO_3$ materials were fabricated by a conventional powder processing. A 21-layered $Ba_{1-x}Sr_xTiO_3$ FGM shows a flat characteristic of the dielectric constant in a wide temperature range.

4. References

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2. Jaffe, B., Cook, W. R. and Jaffe, H., Piezoelectric ceramics, Academic Press, Ohio, p. 91, (1971).