J Electroceram (2008) 21:427–430
DOI 10.1007/s10832-007-9315-z

Dielectric properties of $BaTiO_3$-based ceramics measured up to GHz region

Jianyong Li · Hirofumi Kakemoto · Satoshi Wada · Takaaki Tsurumi

Abstract A new measuring method and analyzing procedure were proposed to determine the complex dielectric constant of materials with relatively high dielectric constant by a lumped impedance measurement using impedance analyzer. Samples used for the measurement were $(Ba_{0.6}Sr_{0.4})TiO_3$ (BST) and $Ba(Zr_{0.25}Ti_{0.75})O_3$ (BZT) ceramics. Micro planar electrodes were formed on the surface of samples by electron beam lithography followed by lift-off method. Complex admittances of these samples were measured up to 3 GHz at different temperatures. Electromagnetic simulations were performed for determining the relative dielectric constant and dielectric loss. The complex dielectric constant vs frequency curves of $Ba(Zr_{0.25}Ti_{0.75})O_3$ showed a broad dielectric relaxation, while that of $(Ba_{0.6}Sr_{0.4})TiO_3$ was almost flat up to 3 GHz on high-temperature side of T_m at which dielectric constant shows maximum value. Dielectric dispersion properties were discussed from the viewpoint of diffuse phase transition in ferroelectrics.

Keywords Relaxor · Barium titanate · Dielectric dispersion · Microwave · Measuring technique

1 Introduction

$Ba_{1-x}Sr_xTiO_3$ (BST), a $BaTiO_3$ (BT)-based solid solution ferroelectric material, has attracted much attention due to its electric-field-dependent dielectric constant. These remarkable properties are currently being used to develop frequency tunable devices in the GHz frequency range [1–4]. $BaZr_xTi_{1-x}O_3$ (BZT) is another type of $BaTiO_3$-based solid-solution material that exhibits high dielectric constants with low temperature-dependence, and therefore, is currently being used as dielectric layer in multilayered ceramic capacitors (MLCCs) with Y5V specification [5]. Both materials with certain compositions show the relaxor behavior but their application fields in the GHz frequency range are completely different. It is therefore important to understand their dielectric spectroscopy, especially, temperature-dependence up to GHz frequency range.

Due to the difficulties in measurement for materials with high dielectric constants and relative high dielectric loss, many studies have only been conducted around the MHz frequency range. A lumped impedance measurement using an RF-impedance analyzer is currently available up to GHz range, which is obviously easy to be used for wide variety of samples. McNeal et al. [6] have used the lumped impedance method to measure the dielectric constant of BT but they pointed out the limit of the frequency was below a few hundreds MHz due to resonance. J. Li et al. [7] proposed the electric length calibration method and succeeded in eliminating the influence of resonance, but they pointed out the frequency range was limited below 1 GHz because the distributed-parameter circuit of sample under measurement could not be accurately converted to a lumped impedance circuit which could be evaluated using an RF-impedance analyzer.

To measure the dielectric properties of materials with high dielectric constant and high dielectric loss in GHz frequency range, we introduced micro planar electrode, microwave probe. The complex admittances (*G*–*B*) of BST and BZT ceramics were measured by RF-impedance analyzer up to 3 GHz in temperature range of from −50 to 100°C. Electromagnetic analysis simulations were employed to

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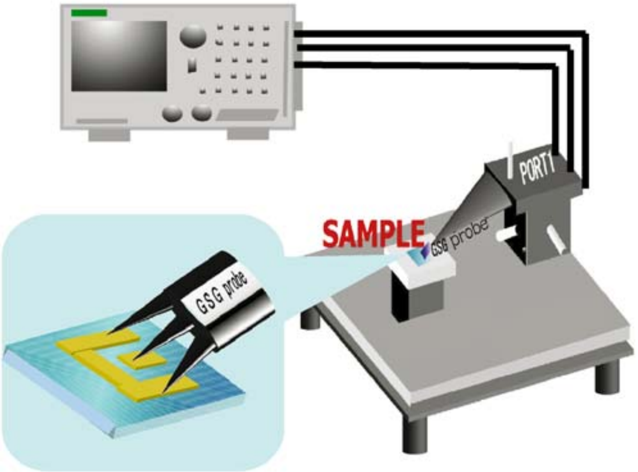

Fig. 1 Structure of measuring system

accurately determine the relative dielectric constants (ε') and dielectric loss factor (tanδ) of samples as a function of frequency. Finally, we will discuss the dielectric dispersion properties from the viewpoint of diffuse phase transition in ferroelectrics.

2 Experimental procedures

2.1 Sample preparation

$(Ba_{0.6}Sr_{0.4})TiO_3$ (BST) and $Ba(Zr_{0.25}Ti_{0.75})O_3$ (BZT) ceramics with high relative densities above 97% were prepared by a conventional solid-state reaction method. The surfaces of samples were mirror-like polished and the samples were formed with the dimension of 7.0 mmφ× 0.376 mm. Micro planar electrodes were formed by electron beam lithography followed by lift-off method after Au-sputtering with thickness of about 60 nm.

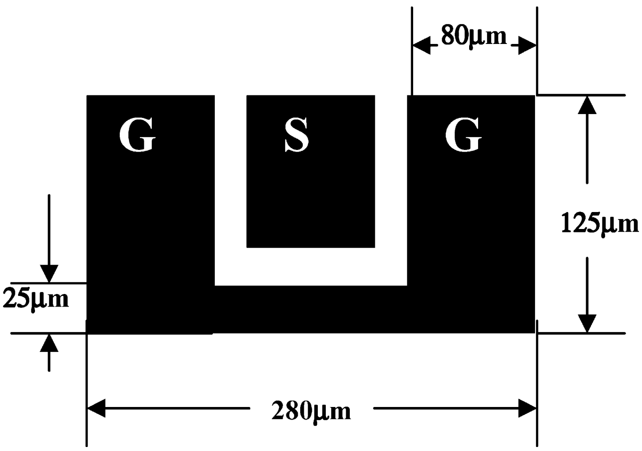

Fig. 2 Size and pattern of micro planar electrode

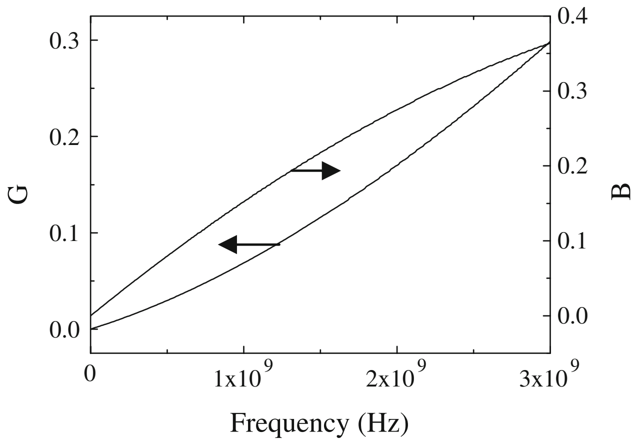

Fig. 3 Complex admittance *G*–*B* simulated by electromagnetic under the condition of ε'=15000 and tanδ=0.5

2.2 Measurement of dielectric properties

Figure 1 shows the measuring system developed in this study which consists of an impedance analyzer (Agilent Tech., E4991A) and a ground–source–ground (GSG) probe (Cascade Microtech). The complex admittances (*G*–*B*) of BST and BZT were measured in the frequency range of 1 MHz to 3 GHz at different temperatures by using a thermostat chamber (Japan High Tech., 10033). Impedance matching at 50 Ω was satisfied to achieve sufficient

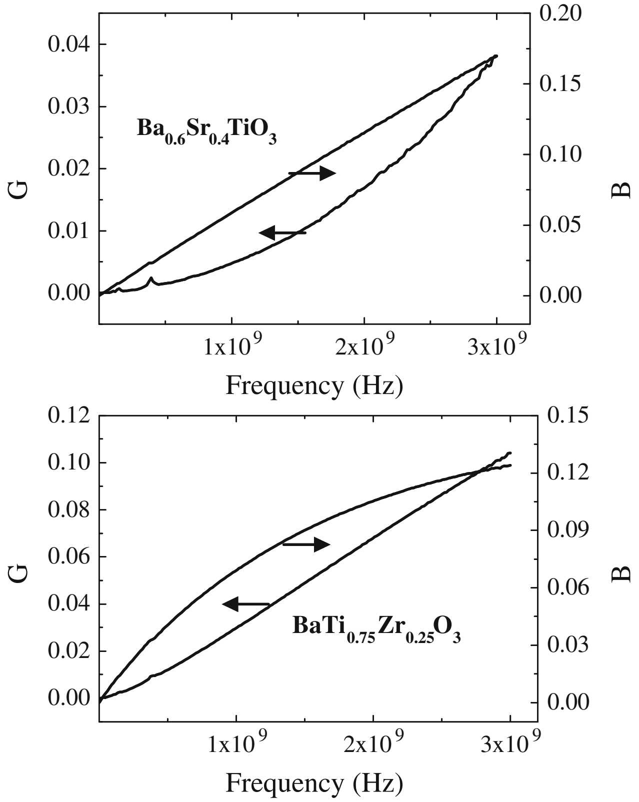

Fig. 4 Complex admittance *G*–*B* of BST and BZT measured at room temperature

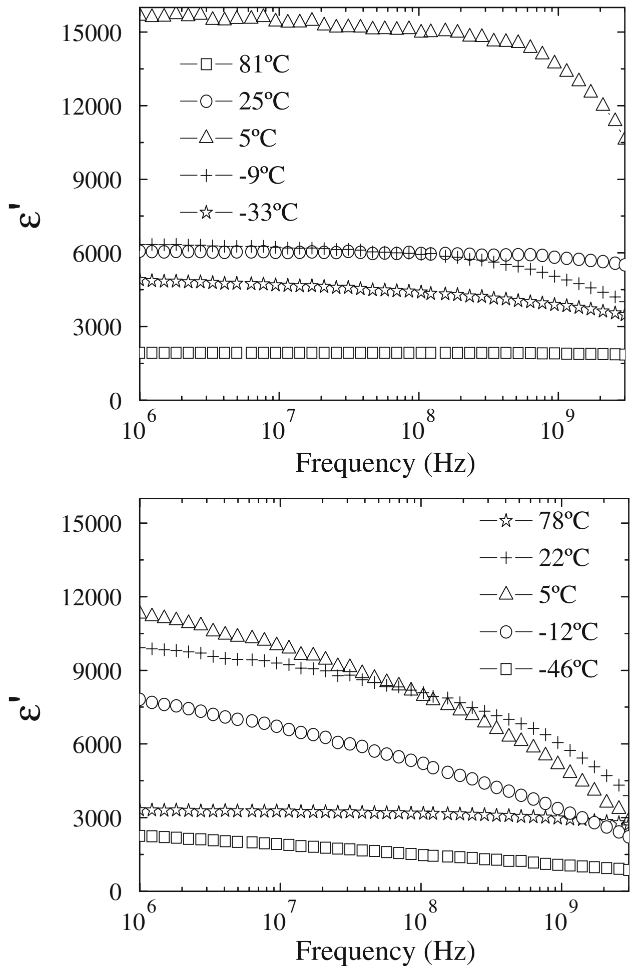

Fig. 5 Frequency dependence of dielectric constants of BST and BZT

accuracy in this measurement. The fundamental calibrations using open, short and load were performed by using impedance standard terminals. The resistivity of the planar electrodes was measured by dc four-probe method using a RESTEST8200 (Toyo Crop., Japan).

3 Results and discussion

3.1 Design of micro planar electrode

Commercial software (Sonnet, Em) was used to design the pattern of micro planar electrodes. The software performs the electromagnetic analysis for high-frequency microstrip circuits, and calculates the complex admittance of micro planar electrodes formed on sample surface. In the electromagnetic analysis, the moment method was used to solve the Maxwell equations of measuring circuits. Micro planar electrodes were designed by a trial-and-error process in order to satisfy the following conditions: 1) no resonance in the measuring frequency range, 2) impedance matching at 50Ω, 3) matching of the electrode size to that of the probe. Figure 2 shows the pattern of micro planer electrode finally designed for BST and BZT samples.

No resonance was observed in electric property simulation for this electrode pattern in wide range of dielectric constant (~18,000) and loss factor (~1.0). Figure 3 shows the complex admittance (*G*–*B*) obtained by a simulation with $\varepsilon'=15000$ and $\tan\delta=0.5$. The *B* value correlates with the dielectric constant while the *G* value correlates with the dielectric loss factor. Almost linear relation was obtained between *B* vs frequency when the dielectric constant was assumed independent from the frequency and it indicated that the result was free from the resonance and the electrode pattern could be used for the real measurement.

3.2 Dielectric properties of BST and BZT

Figure 4 shows the complex admittance (*G*–*B*) vs frequency curves of BST and BZT ceramics measured at room temperature. The *G*, *B* vs frequency curves of BST was similar to those in Fig. 3, indicating that the permittivity of BST was almost constant in the measuring frequency range. However in the case of BZT, the imaginary part *B* of complex admittance show a non-linear relation with

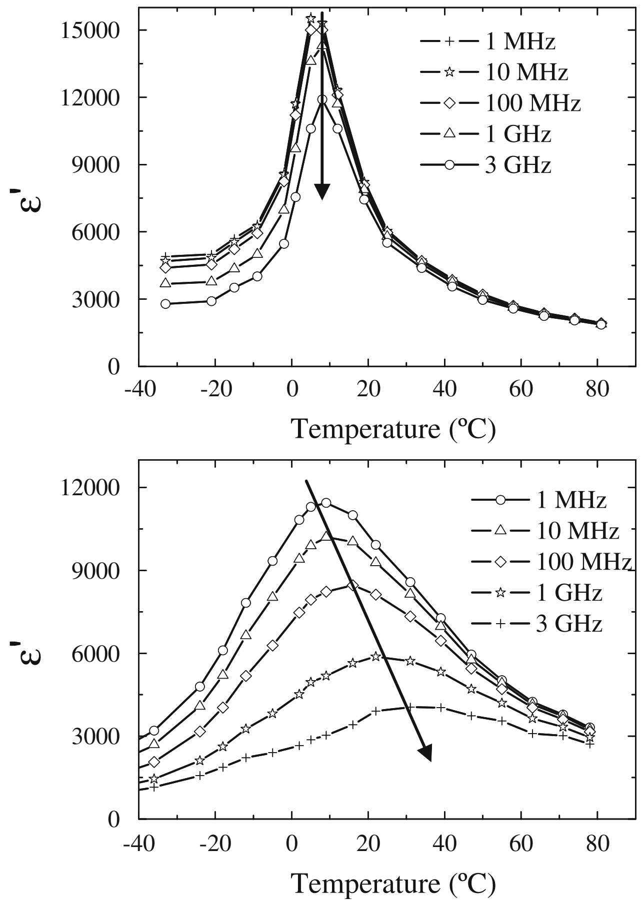

Fig. 6 Temperature dependence of dielectric constants of BST and BZT

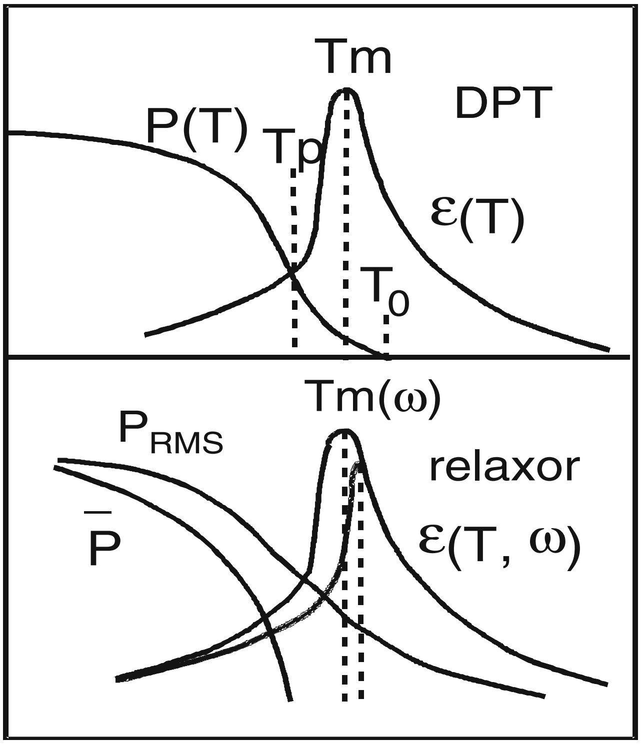

Fig. 7 Characteristic properties of DPT and relaxor ferroelectrics

measuring frequency and it implied the dielectric dispersion in BZT.

An electromagnetic analysis software (Sonnet Em) was employed to determine the relative dielectric constants and dielectric losses of BST and BZT ceramics from raw *G–B* date. Simulations were carried out continually with changing the dielectric constant and dielectric loss of samples until the simulation data were consistent with the measured data, and then, the relative dielectric constants and dielectric losses of BST and BZT were determined. Figure 5 shows the frequency dependence of relative dielectric constants of BST and BZT measured at different temperatures, respectively.

Both BST and BZT showed the maximum dielectric constant at the temperature (T_m) of about 5°C. A larger dielectric dispersion was observed in BZT while the dielectric constants of BST were almost constant values up to 3 GHz on the high-temperature side of T_m. The temperature dependence of dielectric constants of BST and BZT measured at different frequencies were shown in Fig. 6. BST showed relative sharp peaks compared with that of BZT, and no shift of T_m was observed. On the other hand, in BZT, Compared with the high-temperature side of T_m, a larger dielectric dispersion was observed in BZT on the low-temperature side of T_m, and the T_m shifted to higher temperature side observably with the increasing of measuring frequency.

Characteristic properties of diffuse phase transition ferroelectrics (DPT) and relaxor ferroelectrics are shown in Fig. 7 [8]. Differing form ferroelectrics with a definite phase transition temperature (T_c), it is considered that the T_c of DPT is distributed and with non-frequency dependence. Relaxor shows a broad peak of dielectric constant and the T_m shift to high-temperature side with increasing of measuring frequency. The temperature dependence of dielectric constants of BZT was consistent with well-known behavior of relaxor ferroelectrics and that of BST can be considered as the behavior of DPT. It is interesting that BST and BZT show different dielectric properties although have the similar perovskite crystal structure. Microscopic region of the difference between the two materials need to be understood in a future study.

4 Conclusions

Adaptive micro planar electrode was designed for high frequency dielectric measurement by using electromagnetic simulation and formed by electron beam lithography followed by lift-off method.

The complex admittances of BST and BZT ceramics were measured by a lumped impedance analyzer.

Dielectric spectra of BST and BZT ceramics were determined by electromagnetic simulation up to 3 GHz. Temperature dependences of dielectric constants of BST and BZT were estimated.

BST shows a flat dielectric dispersion up to 3 GHz on high-temperature side of T_m, while the BZT shows a broad dielectric relaxation in the range of measuring frequency.

The temperature dependence of dielectric constants of BZT was consistent with well-known behavior of relaxor ferroelectrics and that of BST can be considered as the behavior of DPT. From these results, it is considered that BST is suitable for high frequency applications, whereas BZT is for MLCCs.

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