Dielectric Spectra of $(Ba_{0.6}Sr_{0.4})TiO_3$ and $Ba(Zr_{0.25}Ti_{0.75})O_3$ Ceramics in RF Region Measured by Impedance Analyzers

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Dielectric permittivity of $(Ba_{0.6}Sr_{0.4})TiO_3$ and $Ba(Zr_{0.25}Ti_{0.75})O_3$ ceramics were measured by LF impedance analyzer. Complex impedances and complex reflection coefficients of $(Ba_{0.6}Sr_{0.4})TiO_3$ and $Ba(Zr_{0.25}Ti_{0.75})O_3$ ceramics were measured by RF impedance analyzer. Resonance peaks which was due to the self-inductance of the samples were observed at hundreds MHz in the permittivity *vs.* frequency curves calculated from raw impedance data. Electric length calibration method was proposed to determine dielectric permittivity of high dielectric materials. By using this method, resonance oscillations appeared at hundreds MHz were successfully eliminated, and the dielectric permittivity of these materials were determined up to 1 GHz. The permittivity *vs.* frequency curve of $Ba(Zr_{0.25}Ti_{0.75})O_3$ showed a broad dielectric relaxation above 100 MHz, while that of $(Ba_{0.6}Sr_{0.4})TiO_3$ showed an almost flat dielectric relaxation up to 1 GHz.
Key words: dielectric permittivity, barium titanate, dielectric relaxation, electric length, self-inductance

1. INTRODUCTION

The dielectric permittivity $(Ba_{1-x}Sr_x)TiO_3$ (BST) can be tuned by an applied direct current electric field, and this has led to their use as variable elements in tunable microwave devices, such as tunable microwave devices, phased array antennas, etc.[1-3]. High dielectric permittivity materials $Ba(Zr_xTi_{1-x})O_3$ (BZT) with the same perovskite crystalline as BST, have been intensively studied in recent years for application in multilayered ceramic capacitors (MLCCs), however, not suitable for the application in tunable microwave devices. $BaTiO_3$-based ceramics capacitors have been used in electric circuits operating in GHz region. It is therefore extremely important to measure dielectric permittivity of these materials up to GHz region.

Resonance techniques are frequently used for the characterization of dielectrics at frequencies above 1 GHz, but produce values only at discrete frequencies depending on sample dimensions. The resonance techniques are also limited to low-dielectric-loss materials[4-5]. However, for most high dielectric permittivity materials, the dielectric loss is normally higher than the limit of the resonance techniques[6].

In the view of dielectric relaxation phenomena, dielectric permittivity should be measured at high frequencies as continuous frequency spectra. The time domain reflection (TDR) technique is one of the techniques for measuring dielectric permittivity at high frequencies as continuous frequency spectra. However, TDR spectra need to be transformed to frequency domain using Fourier transformation [7].

A lumped impedance test using an RF impedance analyzer is currently available up to GHz region, 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 permittivity of $BaTiO_3$ but they also pointed out the limit of the frequency was below a few hundreds MHz. Nies *et al.*[8] tried to eliminate the resonance due to the self-inductance using a similar approach. The latter technique gave a reasonable result in comparison with the former but they had to assume a frequency dependence of sample capacitance in the modeling. As mentioned above, some works have been done to measure the permittivity of BT-based materials but the lumped impedance measuring technique for high permittivity and high loss samples has not been established yet.

In this study, the dielectric permittivity of BST and BZT ceramics was measured with impedance analyzers up to 1 GHz. A new correction process of the resonance due to the self-inductance was employed to determine the dielectric spectra of sample. In this process, the calibration of electric length of the system was precisely carried out by the least squares fitting of the reflection coefficients.

2. EXPERIMENTAL PROCEDURE

2.1 Sample Preparation

The $(Ba_{1-x}Sr_x)TiO_3$ ($x=0.4$) and $Ba(Zr_xTi_{1-x})O_3$ ($x=0.25$) ceramics were prepared by a conventional solid solution method. The substitution molar ratio x of BST and BZT was selected to have a maximum of permittivity at the same temperature (about 2°C). Raw powders of these ceramics were derived from a hydrothermal process (Sakai Chem. Ind. Co. Ltd.). Raw powders were formed into a disc shape and isostatically pressed at 100 MPa. The green discs thus obtained were sintered at 1400 - 1500 °C for 4 h. Specimens with high relative densities above 98% were formed to a disc shape with exactly 3.00 mmϕ in diameter and 1.9 mm in length. The surfaces of samples were mirror-like polished using fine Al_2O_3 slurry on the glass plate. Then Au electrodes were deposited by dc-sputtering method.

2.2 Dielectric Measurement

The complex impedance of the samples was measured at 1kHz-1GHz. In the low-frequency region below 2MHz, the impedance was measured with an LF impedance analyzer (Agilent Tech., 4192A), whereas in the high-frequency region from 10MHz to 1 GHz, it was measured with an RF impedance analyzer (Agilent Tech., 4291B). These impedance analyzers were controlled by a personal computer with a GPIB interface. The temperature was varied from -40°C to 20°C in the low-frequency measurement.

In high-frequency measurement, a coaxial connecter (Amphenol, APC-7) was modified and used as a specimen holder. Each 3mm disc sample was placed on a brass part connecting with the APC-7 of the equipment. The top of the sample was connected to a brass bar connecting to the ground. The fundamental calibrations using open, short, load (50Ω) and low loss capacitor were carried out. Brass discs with the same dimension of the samples (3.0mm×1.9 mm) were prepared for the measurement of the electric length corresponding to the self-inductance (f_L) of samples. In order to determine the electric length corresponding to self-inductance (f_L) of samples, the complex reflection coefficients ($\Gamma = \Gamma_x + j\Gamma_y$) of brass discs were measured as a function of the calibration value (δl) of the electric length. And, in order to determine the electric length corresponding to capacitance of samples, the complex reflection coefficients of samples were measured as a function of the calibration value (δl) of the electric length.

3. RESULTS AND DISCUSSION

3.1 Electric length calibration method

Figure 1 shows dielectric spectra of BST and BZT ceramics calculated from raw data of complex impedance measured as a function of frequency. These curves show clear resonance at the frequency of 400 MHz in BST and 310 MHz in BZT, respectively.

The resonance develops across the sample as the sample size and wavelength of electromagnetic wave approach each other, producing an inductance associated
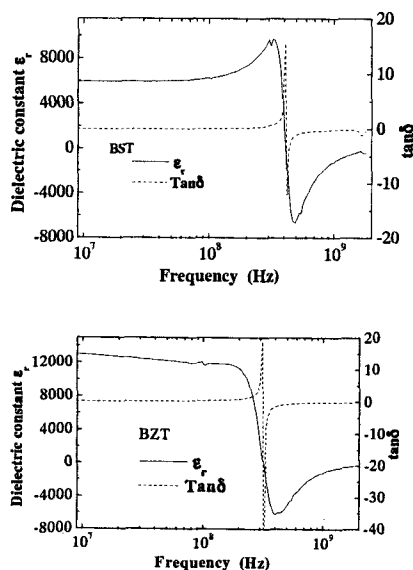

Fig. 1 Dielectric permittivity of (Ba₀.₆Sr₀.₄)TiO₃ and Ba(Zr₀.₂₅Ti₀.₇₅)O₃ ceramics calculated from the raw impedance data measured by RF impedance analyzer.

with the samples. Electric length corresponding to self-inductance (f_L) of sample has been considered as the causation of resonance oscillation. In the low frequency measurement, the electric length corresponding to self-inductance (f_L) can be ignored, and then the impedance measured from standard point which calibrated by open, short, load and low loss capacitor is equal to the load impedance of samples. However, in the high frequency measurement, the electric length corresponding to self-inductance (f_L) approach to the electric length corresponding to capacitance of samples (f_C), and cannot be ignored.

In order to determine the dielectric permittivity of samples, the complex reflection coefficients of samples were measured at a fixed frequency as a function of the calibration value of electric length (δl). From distributed constant circuit theory, the equivalent circuit model of measuring circuit is showed in Figure 2.
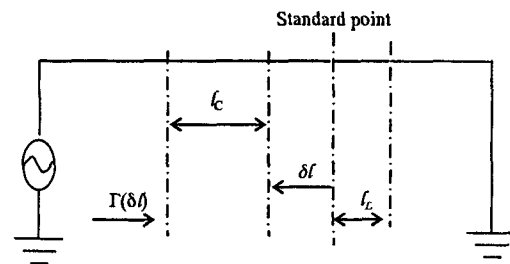

Fig. 2 Electric calibration method model

Electric length corresponding to the self-inductance (f_L) and that to the capacitance of sample (f_C) are given as follows:

$$\omega L = Z_C \tan \beta \, f_L \qquad (1)$$

$$-1/\omega C = Z_C \tan (-\beta \, f_C) \qquad (2)$$

where ω is the angular frequency, L is the self-inductance of sample, β (= $2\pi/\lambda$, λ: wavelength of electromagnetic wave) is the phase constant, C is the capacitance of sample. C can be determined from f_C at an angular frequency ω. In this model, complex coefficients (Γ(δl)), electric length corresponding to self-inductance (f_L) and that to capacitance of samples (f_C), and the calibration value of electric length (δl) have the relation as follow:

$$\Gamma(\delta l) = \Gamma(0)\exp(-2\gamma(f_C + f_L + \delta l)) \qquad (3)$$

where Γ(0) is equal to $(Z_L - Z_c)/(Z_L + Z_c)$. Resonance occurs when $f_L + f_C + \delta l = n\lambda/2$, anti-resonance occurs when $f_L + f_C + \delta l = (2n-1)\lambda/4$, where n is an integer. Figure 3 shows the results of BST measured at 1 GHz. The resonance occurred when the calibration value of electric length (δl) equal to about 5 mm + $n\lambda/2$. The electric length corresponding to capacitance of sample f_C can be determined from the resonance condition showed in equation (3), and then the capacitance of sample C can be determined from equation (3). However, the loss tangent of samples was hardly determined within certain accuracy by this procedure.
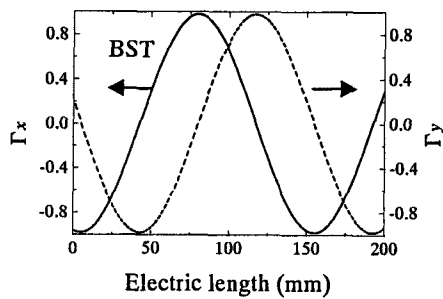
Fig.3 Reflection coefficient ($\Gamma = \Gamma_x + j\Gamma_y$) measured for ($Ba_{0.6}Sr_{0.4})TiO_3$ ceramic at 1 GHz as a function of the calibration value of the electric length (δl).

In order to determine the electric length corresponding to the self-inductance (l_L), the reflection coefficient ($\Gamma = \Gamma_x + j\Gamma_y$) of brass disc with the sample dimension was measured as a function of the calibration value of electric length (δl). In case of this, the electric length corresponding to capacitance (l_C) should be 0, and the electric length corresponding to the self-inductance (l_L) can be determined easily from the resonance condition mentioned above. Figure 4 shows the relation between the reflection coefficients (Γ) vs. the electric length (δl) measured for BST at 1GHz. The solid lines are
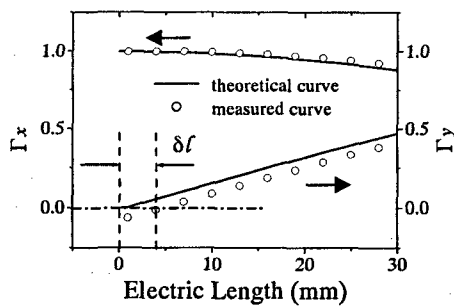

Fig. 4 Reflection coefficient measured for BST ceramic at 1 GHz as a function of calibration value of electric length (δl).

experimental data, and dashed lines are theoretical data calculated in case of $l_C = 0$. The electric length corresponding to the self-inductance (l_L) was determined by a least squares method to fit the observation to the calculation. The observed and calculated data agreed very well at $l_L = 5.85$ mm.
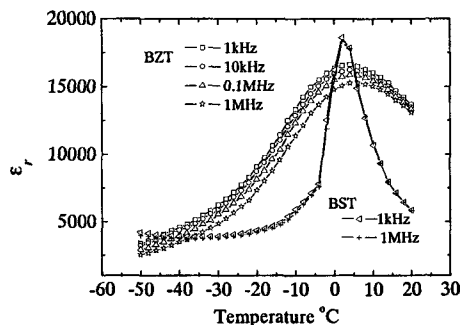

Fig. 5. Relative permittivity vs temperature curves of ($Ba_{0.6}Sr_{0.4})TiO_3$ and $Ba(Zr_{0.25}Ti_{0.75})O_3$ ceramics.

3.2 Dielectric permittivity of BST and BZT

Figure 5 shows the dielectric permittivity vs temperature curves of BST and BZT at different frequencies measured using an LF impedance analyzer. The BST ceramic shows a sharp maximum of permittivity while the BZT ceramic shows a broad maximum. The permittivity of BST is higher than that of BZT. These results are basically consistent with those reported in the literature[9].

Figure 6 shows the relation between relative permittivity vs. frequency of the BST and BZT ceramics. The permittivity of samples was calculated from the capacitance determined by the procedure mentioned in the previous section. For two samples, the resonance peaks observed in Figure 3 are successfully eliminated.
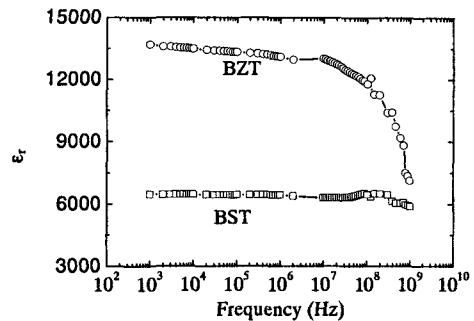

Fig. 6 Relative dielectric permittivity of ($Ba_{0.6}Sr_{0.4})TiO_3$ and $Ba(Zr_{0.25}Ti_{0.75})O_3$ ceramics as functions of frequency.

The permittivity curves are continuous from the low-frequency to the high-frequency region. This indicates the accuracy of theanalysis procedure in determining the capacitance of the samples in the high-frequency region. It is interesting that BZT shows a broad dielectric relaxation from 100 MHz, whereas the permittivity of BST is almost flat up to 1 GHz. BST is used for high frequency applications such as tunable microwave devices because the dielectric relaxation accompanied with the increase in dielectric loss does not occur up to the GHz region, while BZT is used for MLCCs because it shows high permittivity as well as temperature stability of the permittivity as shown in Figature 9. Microscopic origins of the difference between the two materials need to be understood in a future study.

4. CONCLUSION

The dielectric permittivity of ($Ba_{0.6}Sr_{0.4})TiO_3$ and $Ba(Zr_{0.25}Ti_{0.75})O_3$ ceramics was determined as a function of frequency up to 1 GHz by a lumped impedance measurement using a impedance analyzers. Resonance peaks were observed at hundreds MHz in the permittivity vs. frequency curves calculated from raw impedance data, which was due to the self-inductance of samples.

The reflection coefficients of samples were measured as a function of the calibration value of the electric length (δl) at fixed frequencies. The capacitance of samples was determined from the electric length corresponding to the capacitance (l_C) in the measuring circuit. This electric length could be obtained from the electric length corresponding to the self-inductance (l_L) and that at the resonance where the relation, $\delta l + l_L + l_C = n\lambda/2$, was

satisfied.

The electric length corresponding to the self-inductance (f_L) was determined from the curves of the reflection coefficient *vs.* the calibration value of the electric length measured for a brass disc with the sample dimension. A least squares fitting method was employed to exactly determine the electric length due to the self-inductance.

The resonance peaks were successfully eliminated by the procedure proposed in this study. The permittivity *vs.* frequency curve of $Ba(Zr_{0.25}Ti_{0.75})O_3$ showed a broad dielectric relaxation above 100 MHz, while that of $(Ba_{0.6}Sr_{0.4})TiO_3$ showed an almost flat dielectric relaxation up to 1 GHz. The low-frequency measurement indicated that the permittivity of BZT was high permittivity in a relatively wide temperature range. From these results, it is considered that BST is suitable for tunable microwave devices applications, whereas BZT is for MLCCs.

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