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Microwave dielectric response of $(Sr_{0.80}Pb_{0.20})TiO_3$ based ferroelectric composites

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Abstract

This paper reports the results of measurements on the dielectric properties of strontium lead titanate ferroelectric composites (i.e. non-ferroelectric magnesium oxide, MgO, added to the $Sr_{0.80}Pb_{0.20}TiO_3$, SPT, matrix) as a function of temperature over a wide frequency range from 0.05 to 2 GHz. The results showed that SPT–MgO composites have a strong dielectric dispersion (around 500 MHz) for the temperature range of 100–280 K. On the other hand, at room temperature a suppression of such relaxation was observed with low dielectric loss factors $\sim 5 \times 10^{-4}$ over the entire range of frequency from 0.05 to 2 GHz. Such results evidence the potential use of SPT–MgO composites for room temperature tunable microwave applications.
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1. Introduction

For many microwave applications, such as phase shifters, phased array antennas, filters, etc.,[1–4] the microwave component is commonly constructed by using ferrite and some superconductor elements.[5] They present good frequency selectivity and low insertion loss, offering attractive options for commercial and space-based communications.[3] However, they are very costly, large and heavy and the need for suitable materials in frequency and phase agile electronics is still in demand. The desirable characteristics of a dielectric material to be used in phased array applications are low dielectric permittivity (<1500), low dielectric loss factor (<0.01) and high dielectric tunability (change in ε with an applied electric field, >10%).[3,4] Lower dielectric permittivity materials with low loss factor contribute to reduce the overall impedance mismatch and provide lower insertion loss in the device. On the other hand, the degree of the phase shifting ability is directly related to the tunability of the material. Thus higher tunabilities materials are desired. Contrary to the above mentioned materials, ferroelectric components offer the advantage of continuous, quick and low power tunability up to the highest gigahertz frequencies.[6,7] Furthermore, ferroelectric ceramics enable an impressive cost reduction by integration.

Many promising candidates have been reported, including as preference strontium titanate based perovskite ABO_3-type materials ($SrTiO_3$).[1,6–9] The currently most studied solid solutions are barium modified strontium titanate (BST) single phase ceramics, which have been extensively prepared and investigated. However, in spite of possessing high electric field tunability and low losses, which are promising for practical applications, further improvements of the BST performance, such as reproducibility as well as stability of the material, are still desirable. Although many works have been devoted to develop tunable dielectric ceramics and thin films showing low losses, their use in tunable applications at high frequency (gigahertz region) is still in progress.

Recently, significant improvements in reducing the dielectric losses and permittivity have been reported at low frequency by the developing of ferroelectric–non ferroelectric composite materials.[10–12] Among them the MgO modified $(Sr,Pb)TiO_3$ ceramics have received recent attention.[13] However, the reports did not include a detailed study of the high

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frequency dielectric properties (i.e. ∼1 GHz). This article presents a systematic study of the microwave dielectric properties for the strontium lead titanate ferroelectric composites ($Sr_{0.80}Pb_{0.20}TiO_3 + MgO$) in a wide frequency and temperature range (0.05–2 GHz and 100–300 K, respectively).

2. Experimental

The ceramics were prepared by the conventional solid state reaction.[14] The suitable ratio of commercial grade lead oxide (PbO, Aldrich), strontium carbonate ($SrCO_3$, Aldrich), and titanium oxide (TiO_2, Alfa Aesar) (Pb:Sr:Ti = 2:8:10) were ball milled, calcined at 1100 °C for 3 h, and milled again. Then, high purity grade magnesium oxide (MgO, Alfa Aesar) was added to the $(Sr,Pb)TiO_3$ powder, in the weight ratio of 50:50 (SPT:MgO) and the mixture was ball milled again. The composite powder was pressed into pellets by cold uniaxial isostatic pressing and sintered at 1350 °C for 3 h. After polishing, gold electrodes were applied by sputtering to the opposite faces of the samples. The samples were cut into discs with 2.0 mm in diameter and 0.5 mm in thickness.

The measurements were made by the reflectometry method,[15] using an experimental assembly that included a network analyzer (Hewlett-Packard 8719C) and a 50 Ω coaxial line connected to a special sample holder, in which the sample was held under a controlled pressure to avoid poor contacts due to the thermal contraction or expansion of the sample. High frequency dielectric measurements were performed in a temperature and frequency range of 100–300 K and 0.05–2 GHz, respectively. For determining the frequency dependence of the reflection coefficients (Γ', Γ''), careful compensation procedure was carried out to avoid spurious reflections that might result by transmission line discontinuities and also to eliminate the effect of the resistances and capacitances of the sample holder. Three different HP standard terminations (open, short and 50 Ω), with reflection coefficients of 1, −1 and 0, respectively, were used to calibrate the system in the investigated frequency range. The complex dielectric permittivity was determined from the measured complex reflection coefficient.[16] Low frequency measurements were carried out using an Impedance Analyzer HP 4194A.

3. Results and discussion

Fig. 1 shows the low frequency dielectric properties of the studied composite ceramics in the temperature range from 100 up to 450 K at different frequencies. A broad dielectric peak with frequency dispersion is observed at a temperature, which can be associated with the ferroelectric–paraelectric phase transition temperature ($T_m = 209$ K). As can be observed a slight decrease of real dielectric permittivity (ε') occurred with the frequency increasing for temperatures below 250 K. The ε' values, obtained for the composite samples, are notably lower than those obtained for the pure SPT ceramics.[8] This fact clearly shows that the non-ferroelectric magnesium oxide added to the SPT matrix dilutes the dielectric permittivity to the range suitable for a microwave device requirement. On the other hand, the imaginary component (ε'') showed an increase with the frequency evidencing a relaxor-like behavior. The real and imaginary dielectric permittivity values, at room temperature, were 108.5 and 0.05, respectively, leading to a loss factor ($D = \varepsilon''/\varepsilon'$) of about 5×10^{-4}. As observed for temperature values above T_m, both the real and imaginary components of the dielectric permittivity become frequency independent showing a high stability in the imaginary component. The obtained values for the dielectric permittivity and loss factor, if compared with some materials commonly used in high frequency applications, confirm the ability of the studied composites to be used as tunable capacitors.

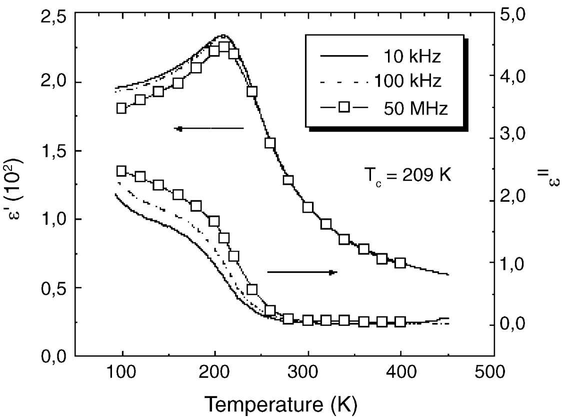

Fig. 1. Temperature dependence of the real and imaginary components of dielectric permittivity, at low frequency, for the based SPT composite.

The microwave frequency dependence of the real and imaginary component of the dielectric permittivity, for several temperatures, is shown in Fig. 2. In the frequency range studied, the results showed a dispersion of the dielectric permittivity that is similar to a Debye type relaxation,[17] presenting a decrease in ε' and a maximum in ε'' in a characteristic frequency (f_R). It can be observed that the dispersion occurs not only in the ferroelectric phase ($T < T_m$) but also towards the expected paraelectric phase ($T > T_m$). At room temperature such dielectric relaxation was completely suppressed and very low dielectric loss values were obtained, which makes this ferroelectric composition a promising material to be used for many microwave applications.

The characteristic frequency (f_R) and the maximal variation of the dielectric permittivity ($\Delta\varepsilon$) were calculated from the Cole–Cole's model relations.[18] The temperature dependence of f_R and $\Delta\varepsilon$ for the SPT composite is shown in Fig. 3. It is noticed that the characteristic frequency goes through a minimum and $\Delta\varepsilon$ presents a maximum close to the temperature at which ε' (measured at low frequencies) reach its maximal value. The value of f_R (∼540 MHz) obtained at T_m suggests that the dielectric dispersion observed might be related to the domain walls motion mechanism. Furthermore, it is interesting to notice that the values of f_R are in the order of the

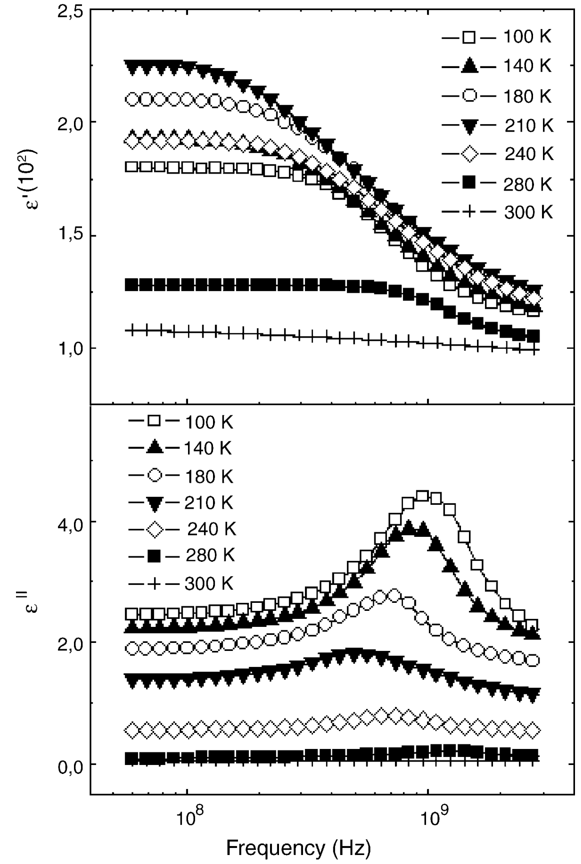

Fig. 2. Frequency dependence of the real and imaginary components of dielectric permittivity, in the microwave region, at various temperatures, for the based SPT composite.

those reported for other ferroelectric materials. Since the $\Delta\varepsilon$ variations in the microwave region have been attributed to the existence of polar regions (ferroelectric domains and/or polar clusters for normal and relaxor ferroelectrics, respectively),[19] it would be expected that the obtained behavior could be associated to the size/interaction of the polar regions. Investigations to better understand the responsible mechanism for the observed dielectric dispersion are in progress.

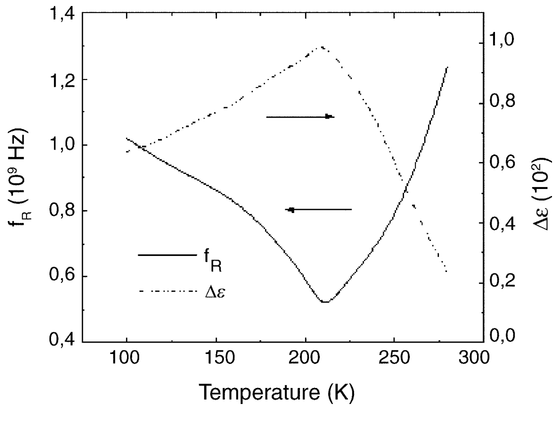

Fig. 3. Temperature dependence of the characteristic frequency (f_R) and maximal dielectric permittivity change ($\Delta\varepsilon$) for the based SPT composite.

4. Conclusions

Based SPT ferroelectric composites has been successfully characterized in the high frequency region, and the results may promote further detailed studies in this and other similar materials that have a significant position in the electro-electronic industry for many practical applications such as wireless communication and dielectric resonators.

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