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Effects of uniaxial stress on dielectric properties of 0.9PMN–0.1PT ceramics

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Abstract This study deals with the influence of uniaxial stress on the dielectric properties of electrostrictive PMN–PT ceramic. The dielectric properties of lead magnesium niobate-lead titanate prepared by a mixed-oxide method with composition ratio 9:1 were measured under uniaxial compressive stress up to 22 MPa. The experimental results revealed that the superimposed compression load significantly reduced both the dielectric constant and the dielectric loss tangent in every measuring frequency. The observations were interpreted in terms of clamping of domain walls and de-poling under the compressive loading. The change of the dielectric properties with stress was attributed to competing influences of the intrinsic contribution of non-polar matrix and the extrinsic contribution of re-polarization and growth of micro-polar regions. In addition, the results reported here also suggested a significant influence of the experimental conditions on the uniaxial stress dependence of dielectric properties of the PMN–PT ceramic.

Keywords Uniaxial stress · Dielectric properties · 0.9PMN–0.1PT

1 Introduction

Ferroelectric lead magnesium niobate, $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN), is widely used in devices such as actuators and transducers because of its good dielectric properties (for instance, at 100 Hz ;ε_r at room temperature ~13,000 and ε_{max} ~16,000) [1–3]. However, the temperature related to the maximum dielectric constant (T_{max}) of PMN is very low (approximately −10 °C) [3]. Thus, lead titanate ($PbTiO_3$ or PT), which has high Curie temperature (T_c ~490 °C, ε_r at room temperature ~300) [3–5] is added to PMN with ratio 9:1 to enhance the dielectric properties of PMN (as well as increasing T_{max}). As a result, at 100 Hz, 0.9PMN–0.1PT ceramic has T_{max}=40 °C and ε_{max}>20,000 [6]. Therefore, 0.9PMN–0.1PT ceramics have been a subject of many investigations and, more importantly, have been widely applied in actuators and transducers [1, 4, 7].

However, when used in devices specified above these ceramics are often subjected to self-induced or external stress, e.g. acoustic transducers [1, 3, 7]. Therefore, it is very important to obtain experimental data, as well as to better understand how these materials behave under stress. Recently, the uniaxial stress dependence of dielectric properties has been studied in materials such as PZT, PMN–PZT [8–12]. For PZT ceramics, the dielectric constant increases with increasing stress in range 0–35 MPa. After reaching a maximum value of dielectric constant at 35 MPa, it gradually decreases with further pre-stress increment [8, 10, 11]. On the other hand, in case of PMN–PZT ceramics, the changes of the dielectric constant with the applied stress can be divided into two different groups. For PMN-rich compositions, the dielectric constant generally decreases with increasing applied stress, while for PZT-rich compositions the dielectric constant rises slightly when the applied stress increases from 0 to 1 MPa, and becomes relatively constant when the applied stress increases further [9]. These results clearly show that the effects of stress on the dielectric properties depend significantly on ceramic compositions and stress levels. Since PMN–PT ceramics are practically very important and there have been previous reports on the electro-mechanical properties of these

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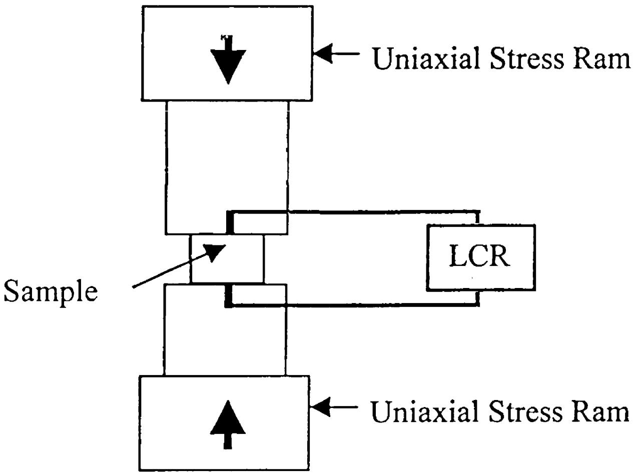

Fig. 1 Schematic diagram of experimental set up

ceramics under various mechanical and electrical loading conditions [13, 14]. However, there has been no systematic study on the influence of an applied stress on their dielectric properties. Therefore, it is the aim of this study to determine the dielectric properties of the 0.9PMN–0.1PT ceramic as a function of uniaxial compressive stress.

2 Experimental method

0.9PMN–0.1PT ceramics were prepared from starting PMN and PT powders by a mixed-oxide method. Perovskite-phase PMN powders were obtained from the columbite method, while PT powders were prepared by a simple mixed-oxide method.

To obtain the perovskite-phase PMN, the magnesium niobate powders were first prepared by mixing MgO (99.0%) and Nb_2O_5 (99.9%) powders and then calcining the mixed powders at 1100 °C for 3 h, to yield a so called columbite powders ($MgNb_2O_6$). After that, the columbite powders were mixed with PbO (99.9%) by ball-milling method and calcined at 800 °C for 1 h to form the perovskite-phase PMN powders. With a simple mixed-oxide route, PT powders were prepared from PbO (99.9%) and TiO_2 (99.9%) starting powders. These powders were ball-milled and calcined at 600 °C for 1 h.

0.9PMN–0.1PT ceramics were prepared from starting PMN and PT powders by the same mixed-oxide method with ratio 9:1. After mixing the powders by ball-milling method and drying process, the mixed powders were pressed hydraulically to form disc-shaped pellets 12 mm in diameter and 1.5 mm thick, with 3 wt.% polyvinyl alcohol as a binder. The pellets were placed on the alumina powder-bed inside alumina crucible and surrounded with atmosphere powders of the same composition. Finally, the pellets were sintered at 1240 °C for 2 h. The phase-formation of the sintered ceramics was studied by XRD technique. The microstructure analyses were undertaken by a scanning electron microscopy (SEM: JEOL Model JSM 840A). The densities of sintered specimens were measured by Archimedes method. The detailed descriptions of the ceramic processing and characterization were presented elsewhere and will not be discussed here [15].

Before studying the dielectric properties under the uniaxial stress, the specimens were lapped to obtain parallel faces. After coating with silver paint as electrode at the faces, the specimens were heated at 750 °C for 12 min to ensure the contact between the electrode and surface of ceramic. To study effects of the uniaxial stress on the dielectric properties of the ceramic, the uniaxial compressometer was constructed [9], as shown schematically in Fig. 1. The specimen was laid between uniaxial stress rams. The dielectric properties were measured by LCR-meter (Instrek LCR-821). The room temperature (27 °C) capacitance and the dielectric loss tangent were obtained at frequency range 1 to 200 kHz under uniaxial compressive pre-stress levels up to 22 MPa. The dielectric constant was then calculated from a parallel-plate capacitor equation, e.g. $\varepsilon_r = Cd/\varepsilon_0 A$, where C is the capacitance of the specimens, d and A are, respectively, the thickness and the area of the electrode, and ε_0 is the dielectric permittivity of vacuum (8.854×10^{-12} F/m).

3 Results and discussion

The dielectric constant at various frequencies of 0.9PMN–0.1PT ceramics as a function of compressive stress during loading and unloading are depicted in Fig. 2. There is a significant change of the dielectric constant of the ceramic with increasing stress from 0 to 22 MPa and returning to stress-free condition. The dielectric constant decreases monotonically with increasing the compressive stress, then increases only slightly when the compressive stress is

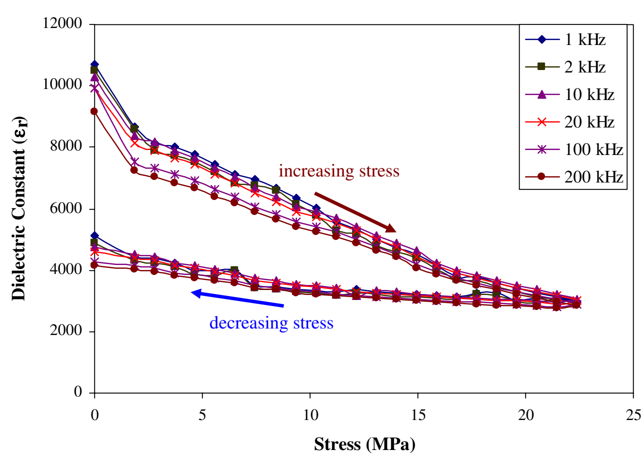

Fig. 2 Uniaxial compressive stress dependence of dielectric constant (ε_r) of 0.9PMN–0.1PT ceramic at various frequencies

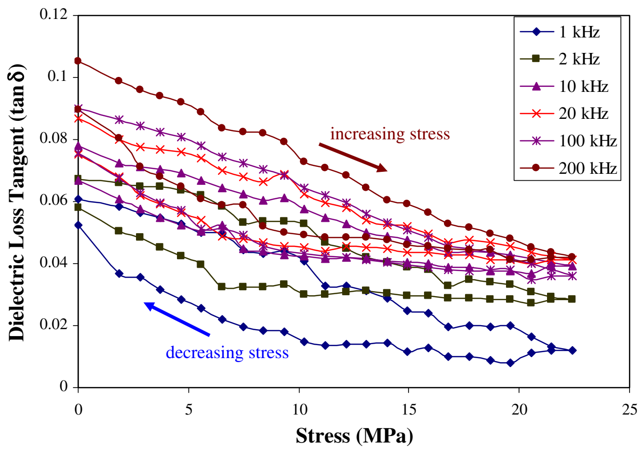

Fig. 3 Uniaxial compressive stress dependence of dielectric loss tangent (tan δ) of 0.9PMN–0.1PT ceramic at various frequencies

gradually removed. The changing of the dielectric constant with increasing and decreasing the applied stress does not follow the same path. In every frequency, the dielectric constant with increasing the compressive stress is larger in value than that with decreasing stress. It is also of interest to see that the stress-free dielectric constant value decreases significantly after a stress cycle. Furthermore, as shown in Fig. 2, the dielectric constant at a constant stress also changes with frequency. The dielectric constant decreases significantly when the measuring frequency increases from 1 to 200 kHz. The trend is opposite for the dielectric loss tangent (as shown in Fig. 3). This is a typical characteristic of a relaxor ferroelectric, in which below the dielectric maximum temperature, the dielectric constant decreases and the dielectric loss tangent increases with increasing frequency [1–4].

As displayed in Fig. 3, the results of the uniaxial compressive stress dependence of the dielectric loss tangent show a similar tendency to those of dielectric constant. At each measuring frequency, the dielectric loss tangent deceases with increasing the compressive stress and then slightly increases when the compressive stress is removed (shown in Fig. 3). The dielectric loss tangent is also found to decrease significantly after a stress cycle. Since the dielectric properties change with the applied stress in very similar trend for every measuring frequency, the data at 10 kHz are selected as representatives for better comparison in Fig. 4 for the fractional changes of the dielectric properties with the compressive stress. It can be seen very clearly that the dielectric constant decreases as much as 70% at the maximum applied stress and only returns to slightly less than 50% of its original value when the stress is removed. Though following the same trend, the change in the dielectric loss tangent value is less significant, as it only decreases about 50% at the maximum stress and almost returns on its original value after a stress cycle. It is also noticed that the results of this study are in parts similar to the experimental results for PMN–PZT system in earlier investigation [9]. For 0.9PMN–0.1PZT ceramic, the dielectric constant generally decreases with increasing applied stress. However, the trend for change of the dielectric loss tangent is different. The dielectric loss tangent of 0.9PMN–0.1PZT is found to first increase when the applied stress is raised from 0 to 1 MPa, and then decrease with further increasing stress [9]. It is interesting to observe that a mixture of different normal and relaxor ferroelectrics responds to the applied stress in a similar manner.

To understand these experimental results, various effects have to be considered. Normally, the properties of ferroelectric materials are derives from both the intrinsic and extrinsic contribution [9–11]. When a compressive stress is applied to the ferroelectric materials, the domain structure in the material will change to maintain the domain energy at a minimum; during this process some of the domains engulf other domains or change shape irreversibly. Under a uniaxial stress, the domain structure of ferroelectric ceramics may undergo domain switching through non-180° domain walls, de-aging, de poling and clamping of domain walls [11, 14].

In this study, since 0.9PMN–0.1PT is a relaxor ferroelectric with T_m ~39–40 °C and the experiment was carried out at room temperature (~27 °C) which is slightly below the T_m, the experimental observation, which shows decreases in both dielectric constant and dielectric loss tangent with increasing stress, can be attributed to competing influences of the intrinsic contribution of non-polar matrix and the extrinsic contribution of re-polarization and growth of micro-polar regions. Since the behavior of 0.9PMN–0.1PT depends on the ratio between the micro-polar region and the non-polar matrix, in this case the micro-polar regions dominate [9–12]. Hence, the dielectric responses of the 0.9PMN–0.1PT ceramic are observed to

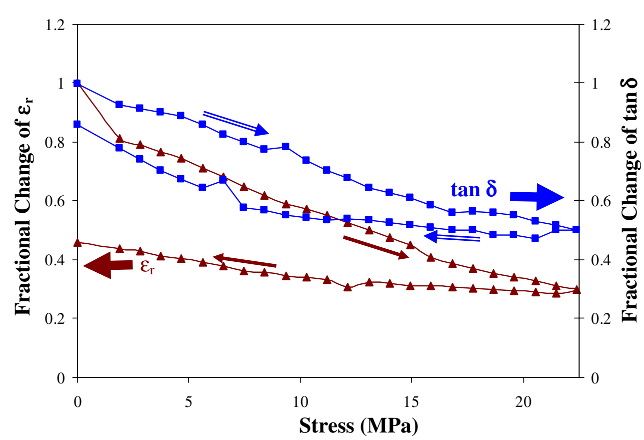

Fig. 4 Fractional changes of dielectric constant (ε_r) and dielectric loss tangent (tan δ) with uniaxial compressive stress of 0.9PMN–0.1PT ceramic (measured at 10 kHz)

decrease with increasing the compressive stress, as seen in Figs. 2, 3 and 4.

In addition, the de_poling mechanism also plays a role in the experimental results. The decrease of both the dielectric constant and the dielectric loss tangent after a stress cycle is believed to be the direct contribution of the partial de-poling effect under the compressive stress cycle [9–11].

More interestingly, earlier work by Viehland et al. [13] reported the large-signal polarization–electric field (P–E) relation under uniaxial stress of 0.9PMN–0.1PT ceramic. For comparison, one would be able to estimate from the P–E loops presented in that study the dielectric permittivity by using Eq. 1, which gives

$$\varepsilon = \frac{\Delta P}{\Delta E} \tag{1}$$

where ΔP is the polarization difference between +2.5 and 0 kV/cm. The calculated dielectric permittivity can be called *differential permittivity*, which includes the reversible (intrinsic dielectric property) and irreversible (extrinsic domain switching related property) contributions of the materials [8, 16]. The calculation shows that the differential permittivity decreases approximately 13% when the applied stress is increased from 0 to 30 MPa. This change is significantly lower than that observed in our low-field study (a decrease of more than 70% over the same stress range). Even though the reasons for the difference are still not clearly known, this observation clearly signifies the importance of the experimental conditions used to determine the dielectric properties, as well as the stress-dependence dielectric properties. More detailed study is underway and further results will be presented in future publications. Finally, the information obtained in this current work is practically very useful for design and calculation consideration of devices in low-field applications.

4 Conclusions

In this study, the dielectric properties of 0.9PMN–0.1PT ceramics prepared by a conventional mixed-oxide method are measured under the compressive stress from 0 to 22 MPa. The results clearly show that the dielectric constants and the dielectric loss tangent of the 0.9PMN–0.1PT ceramic decrease with increasing the compressive stress. The change of the dielectric properties with stress is attributed to competing influences of the intrinsic contribution of non-polar matrix and the extrinsic contribution of re-polarization and growth of micro-polar regions. In addition, the difference in the uniaxial stress dependence of the dielectric properties measured under low- and high-field conditions is also apparent. Finally, this study undoubtedly shows that the applied stress has significant influences on the dielectric properties of the 0.9PMN–0.1PT ceramics.

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