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Effects of compressive stress on the dielectric properties of PIN–PT ceramics

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Abstract

Lead indium niobate, $Pb(In_{1/2}Nb_{1/2})O_3$ (PIN), is an interesting ferroelectric due to a transition from a disordered to an ordered state by long-time thermal annealing. However, the temperature related to the maximum dielectric constant (T_{max}) of PIN in relaxor phase is low (at 1 kHz, $T_{max} = 66\ ^\circ C$). In this study, lead titanate $PbTiO_3$ (PT) was added to PIN with compositions $(1-x)$PIN–xPT (for $x = 0.1$–0.5) to increase their T_{max}. The influence of stress on dielectric properties of $(1-x)$PIN–PT ceramics was then investigated. The dielectric properties were measured under various uniaxial compressive stresses up to 400 MPa. The results showed the reduction of dielectric constant in 0.9PIN–0.1PT with superimposed compression load. For other compositions, dielectric constants first increased with compressive stress, then decreased when the stress was further increased up to 400 MPa. The loss tangent of all composition was found to decrease with increasing compressive stress.

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Keywords: C. Dielectric properties; D. Niobate; Stress

1. Introduction

Piezoelectric and ferroelectric ceramics are widely used in devices such as actuators and transducers. However, when they are used in devices specified above, these ceramics are often subjected to self-induced or environmental stresses [1–3]. A prior knowledge of the effects of stresses on the material properties is crucial for proper design of a device and for suitable selection of materials for a specific application [4–6]. Therefore, it is very important to obtain experimental data, as well as to understand how these materials behave under stress. Recently, the compressive stress dependence of dielectric properties has been studied in materials, i.e., BT, PZT, PMN, PMN–lead titanate (PT), PZT–BT, and PMN–PZT [7–13]. The results clearly showed that the effects of stress on the dielectric properties depended significantly on ceramic compositions and stress levels.

Recently, there has been a great deal of interest in lead indium niobate, $Pb(In_{1/2}Nb_{1/2})O_3$ or PIN, because it can be obtained in a disordered state and exhibit a transition from a disordered to an ordered state by long-time thermal annealing. In the disordered state, PIN is a relaxor ferroelectric (RFE) with rhombohedral structure. On the other hand, the ordered PIN structure is antiferroelectric (AFE) with orthorhombic structure [14]. However, the temperature related to the maximum dielectric constant (T_{max}) of PIN in RFE phase is low (at 1 kHz, $T_{max} = 66\ ^\circ C$) [15,16]. Thus, to enhance the dielectric properties of PIN (as well as increasing T_{max}), lead titanate is added to PIN with compositions $(1-x)$PIN–xPT (for $x = 0.1$–0.5). Practically, there have been many previous reports on the electrical properties of PIN and PT ceramics, but there has been no systematic study on the influence of an applied stress on the dielectric properties of the PIN–PT ceramics. Therefore, it is the aim of this study to determine the dielectric properties of the $(1-x)$PIN–(x)PT ceramics as a function of compressive stress.

2. Experimental procedure

$(1-x)Pb(In_{1/2}Nb_{1/2})O_3$–$(x)PbTiO_3$ (for $x = 0.1$–0.5) ceramics were prepared from two-stage mixed-oxide method. First, $InNbO_4$ was prepared from oxide powders of Nb_2O_5 and In_2O_3. The powders were mixed via a rapid vibratory mill for 30 min in alcohol. After drying, the mixture was calcined at 1100 °C for 2 h to obtain the intermediate precursor $InNbO_4$ [17]. Next,

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the precursor was mixed with appropriate amount of PbO and TiO_2 via a rapid vibratory mill for 30 min. For optimization purpose, the mixtures were calcined at temperatures between 800 and 900 °C for 2 h. After calcination, the powders with 3 wt.% polyvinyl alcohol as a binder were pressed hydraulically to form disc-shaped pellets with 10 mm in diameter and 1 mm in thickness. Finally, the disc-shaped pellets were sintered at temperatures between 1100 and 1125 °C for 2 h. Detailed preparation process is given elsewhere [18].

Before studying the dielectric properties under the compressive stress, the specimens were lapped to obtain parallel faces. After coating with silver paint as electrode on the two surfaces, the specimens were heated at 750 °C for 12 min to ensure the contact between the electrode and the surface of the ceramic. The ceramics were poled at 150 °C under an electric field of 25 kV/cm. To study the effects of stress on dielectric properties of the ceramic, the compressometer was constructed. The dielectric properties were measured by LCR-meter (Instrek LCR-821). The room temperature (25 °C) capacitance and the loss tangent were determined at a frequency range of 1–200 kHz under compressive pre-stress levels of 0–400 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

Phase formation was investigated throughout the $(1-x)$PIN–xPT composition ranging between $x = 0.1$ and 0.5 by X-ray diffraction method (not shown here). In general, pseudo-cubic symmetry is observed at low PT contents ($x = 0.1$, 0.2 and 0.3), in agreement with other researchers [19,20]. However, with the influence of PT, the tetragonal symmetry has developed in the $(1-x)$PIN–xPT compositions with $x = 0.4$ and 0.5. The dielectric constants (ε_r) of all compositions were also measured as functions of both temperature and frequency, as shown in Fig. 1. At the $(1-x)$PIN–xPT compositions with $x = 0.4$ and 0.5, the dielectric properties were nearly independent of frequency, except in the vicinity of phase transformation temperature, a typical characteristic of normal ferroelectrics [19,20]. At low PT contents, the dielectric behavior of the compositions was similar to that of relaxor materials. The dielectric properties of the $(1-x)$PIN–xPT compositions with $x \leq 0.3$ exhibited a diffuse phase transition with dielectric peak broadening [20]. As listed in Table 1, the high value of dielectric constant under stress-free condition at room temperature was obtained in the $(1-x)$PIN–xPT composition with $x = 0.4$ because of its vicinity to the MPB of PIN–PT system, as reported earlier [14].

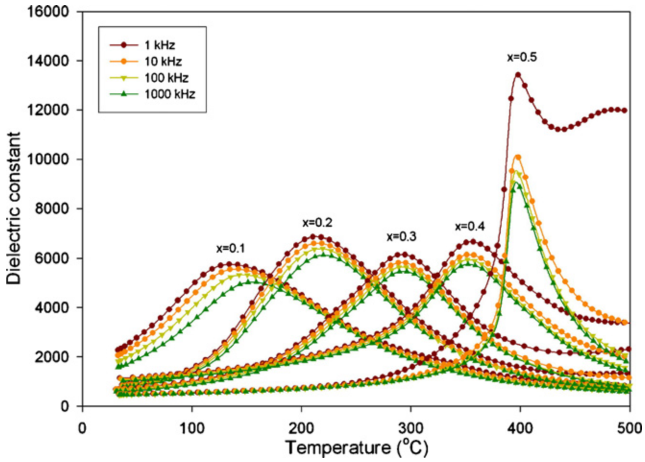

Fig. 1. Temperature and frequency dependence of dielectric constant of $(1-x)$PIN–(x)PT ceramics.

Table 1
Characteristics of PIN–PT ceramics with optimized processing conditions

Ceramic	Sintering temperature (°C)	Density (g/cm^3)	T_C (°C)	Stress-free dielectric properties	
				$\varepsilon_{r,\ room}$	$\varepsilon_{r,\ max}$
0.9PIN–0.1PT	1100	7.78	134	2275	5756
0.8PIN–0.2PT	1125	7.85	213	863	6863
0.7PIN–0.3PT	1125	7.81	292	689	6154
0.6PIN–0.4PT	1125	7.85	355	1132	6674
0.5PIN–0.5PT	1125	7.80	398	483	13422

The dielectric properties at 100 kHz of $(1-x)$PIN–(x)PT ceramics as a function of compressive stress are depicted in Figs. 2 and 3. For better comparison, the dielectric properties of each composition under stress are normalized to the stress-free values. It is clearly seen that a significant change of both dielectric constant and loss tangent occur when the ceramics subjected to an applied stress from 0 to 400 MPa and return to stress-free condition. As depicted in Fig. 2, the changes of dielectric constant with an applied stress can be divided into two groups. For pseudo-cubic compositions ($x = 0.1$–0.3), the dielectric constant changes drastically with the applied stress. It alters more than 5–35% when the stress reaches 400 MPa. The change of the dielectric constant during loading and unloading does not follow the same path. The dielectric constants of these

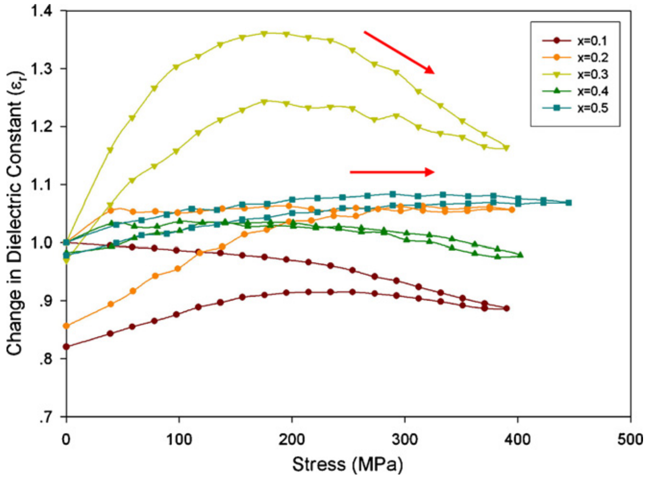

Fig. 2. Relative changes of dielectric constant (ε_r) with compressive stress for $(1-x)$PIN–(x)PT ceramics (measured at 25 °C and 100 kHz; solid arrows indicate loading direction).

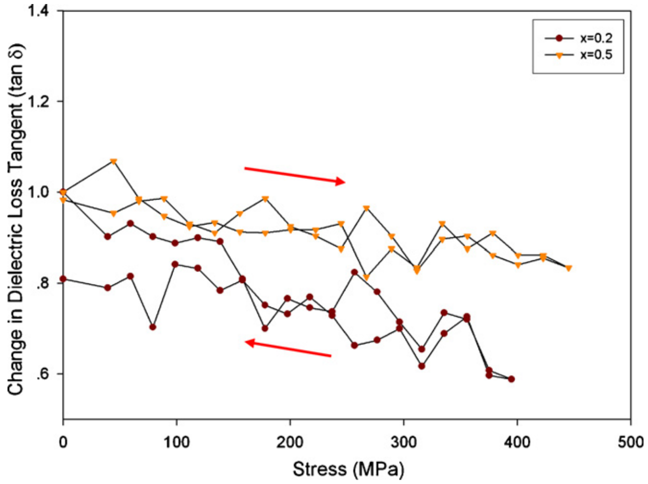

Fig. 3. Relative changes of dielectric loss tangent (tan δ) with compressive stress for (0.8)PIN–(0.2)PT and (0.5)PIN–(0.5)PT ceramics (measured at 25 °C and 100 kHz; solid arrows indicate loading direction).

compositions either initially increase then decrease until maximum stress, or only decrease with increasing stress. On the other hand, for the tetragonal compositions, with x values of 0.4 and 0.5, the dielectric constant shows a very slight change with stress. In addition, the compressive stress dependence of the loss tangent ($\tan\delta$) is observed, as shown in Fig. 3. Since all the compositions show very similar behavior with stress, only compositions with $x = 0.2$ and 0.5 are shown in the graph. It can be seen that the loss tangents during loading and unloading are insignificantly different. It is also noticed that the changes in the dielectric properties with the compressive stress obtained in this study are in parts similar to those for PMN–PZT and PMN–PT systems in earlier investigations [10,11].

To understand these experimental results, at least qualitatively, various effects have to be considered. When a compressive stress is applied to the ferroelectric materials in the direction parallel to the poling direction, the domain structure in the material will alter itself to maintain the minimum level of domain energy. Because the stress will move some of the polarization away from the poling direction; during this process some of the domains engulf other domains or change the shape irreversibly. Under a 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 [10].

The experimental observations, both the drastic change case and the very slight change one, can be attributed to competing influences, in an opposite way, of the intrinsic contribution of domains and the extrinsic contribution of re-polarization and growth of micro-polar regions. Under the applied compressive stress, the non-180° domain wall density increases. Hence the increase of the dielectric constant is observed. The de-aging mechanism, which also increases the dielectric constant [9–11], is also expected to play a role here. Therefore, a combination of the domain switching and the de-aging mechanisms is believed to be a reason for the increase of the dielectric constant during low-stress application, i.e. PIN–PT with $x = 0.2$, 0.3, 0.4 and 0.5. With a further increase in the stress, the stress clamping of domain walls, which results in a decrease of domain wall mobility, and the stress-induced decrease in a switchable part of spontaneous polarization are expected to play a role in the decrease of the dielectric constant [9,10]. In addition, in case of 0.9PIN–0.1PT the continuous decrease in the dielectric constant can be attributed to the switching of 90° domains, which causes the significant decrease in the dielectric constant, as seen in Fig. 2. The cause of the stress dependence of the dielectric loss tangent is more straightforward. As depicted in Fig. 3, the clamping of the domain walls under the compressive stress results in a decrease of domain wall mobility and reduces the loss tangent [21]. This is a reversible effect with the domain wall mobility returning to near the original values when the applied stress is removed, as seen in Fig. 3 that the dielectric loss tangents return to near their original values after a stress cycle. In addition, a significant decrease in the dielectric constant after a full cycle of stress application has been observed and attributed to the stress-induced decrease in switchable part of spontaneous polarization at high stress and the irreversible 90° domain switching [10,21].

4. Conclusions

The dielectric properties of $(1-x)\mathrm{Pb}(\mathrm{In}_{1/2}\mathrm{Nb}_{1/2})\mathrm{O}_3$–$(x)\mathrm{PbTiO}_3$ (for $x = 0.1$–0.5) PIN–PT ceramics prepared by a two-stage mixed-oxide method were measured under compressive stress from 0 to 400 MPa. The results clearly show that the superimposed compression stress has pronounced effects on both the dielectric constant and the dielectric loss tangent of PIN–PT ceramics. The observations are mainly interpreted in terms of competing influences of the intrinsic contribution, and the extrinsic contribution from domain switching through non-180° domain walls, clamping of domain walls, de-aging, and the stress-induced decrease in a switchable part of spontaneous polarization.

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