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Temperature dependence of electrical and electromechanical properties of $Pb(Mg_{1/3}Nb_{2/3})O_3$–$Pb(Ni_{1/3}Nb_{2/3})O_3$–$Pb(Zr,Ti)O_3$ thin films

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Abstract

The electrical and electromechanical properties of $Pb(Mg_{1/3}Nb_{2/3})O_3$–$Pb(Ni_{1/3}Nb_{2/3})O_3$–$Pb(Zr,Ti)O_3$ (PMN–PNN–PZT, PMN/PNN/PZT = 20/10/70) on Pt/Ti/SiO_2/Si substrates by chemical solution deposition was investigated. The PMN–PNN–PZT films annealed at 650 °C exhibited slim polarization hysteresis curves and a high dielectric constant of 2100 at room temperature. A broad dielectric maximum at approximately 140–170 °C was observed. The field-induced displacement was measured by scanning probe microscopy, the bipolar displacement was not hysteretic, and the effective piezoelectric coefficient (d_{33}) was 66×10^{-12} m/V. The effective d_{33} decreased with temperature, but the value at 100 °C remained 45×10^{-12} m/V.

Keywords: A. Films; C. Ferroelectric properties; C. Piezoelectric properties; E. Actuators

1. Introduction

Recently, electric field-induced strains in thin films have attracted attention due to their potential applications in microelectromechanical systems (MEMS) [1,2]. Thus, research efforts have focused on $Pb(Zr,Ti)O_3$ (PZT) [1–4] due to reasonably good piezoelectric behavior and a relatively low annealing temperature, which is approximately 650 °C. However, the strong hysteretic behavior of the field-induced displacement in PZT is considered to be a problem in the analogue control of an actuator. Recently, relaxor–ferroelectric single crystals such as $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ (PMN–PT) have been of interest due to their excellent piezoelectric properties, which are superior to those of PZT ceramics [5–7]. Compared with PZT [8], the smaller field-induced strain hysteresis of these crystals is also attractive for actuator applications. Trial film preparations of these relaxor–ferroelectric crystals have been reported, but a higher annealing temperature of approximately 800 °C was required, and high quality film preparation has been difficult to achieve [9–13]. Our aim is to modify PZT film properties regarding hysteretic behavior without raising the final annealing temperature. For film preparation, the chemical solution deposition was employed due to its ease of composition control and low temperature deposition, and the $Pb(Mg_{1/3}Nb_{2/3})O_3$–$Pb(Ni_{1/3}Nb_{2/3})O_3$ doped PZT films could be obtained by annealing at 650 °C.

In this paper, the properties of relaxor $Pb(Mg_{1/3}Nb_{2/3})O_3$–$Pb(Ni_{1/3}Nb_{2/3})O_3$ doped PZT films annealing at 650 °C were reported, including a description of their temperature dependence and a comparison with PZT films. In the case of relaxor-doped films, the temperature dependence of the properties is worrisome due to a lower transition temperature. Here, the electrical and electromechanical properties of the films in the temperature range between −250 and 200 °C and the electromechanical properties between −60 and 150 °C were reported and compared with the properties of PZT films.

2. Experimental procedure

PMN–PNN–PZT thin films are fabricated by a state-of-the-art chemical solution deposition from a precursor solution (Inostek Inc.) with PMN/PNN/PZT = 20/10/70. Zr to Ti ratio of PZT is 52/48 [14]. The total film thickness is 500 nm. The final annealing temperature is 650 °C. Thin-film Pt top electrodes were deposited. The polarization hysteresis loops and dielectric

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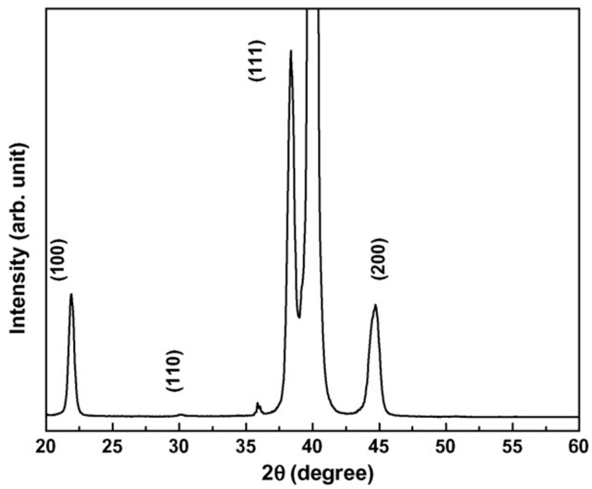

Fig. 1. X-ray diffraction pattern of 20PMN–10PNN–70PZT film.

constant were measured using, respectively, a TF2000 ferroelectric tester and an Agilent Technology impedance analyzer, 4192A. The field-induced strain of the films was measured using scanning probe microscopy. The details of the experimental setup are described elsewhere [4].

3. Results and discussion

Fig. 1 shows the X-ray diffraction pattern of the 20PMN–10PNN–70PZT film annealed at 650 °C. The pattern indicates that the obtained film is pure perovskite and shows a mixed preferred orientation of (1 0 0) and (1 1 1).

The polarization hysteresis loop of the film at room temperature is shown in Fig. 2. The polarization hysteresis loop was slim and nonlinear and, compared with PZT, the hysteretic behavior was smaller. The small-signal dielectric constant (ε_r) and loss tangent of the films at room temperature (20 °C) were 2100 and 0.04, respectively. The polarization hysteresis loops at −200, 0, and 150 °C are shown in Fig. 3. The

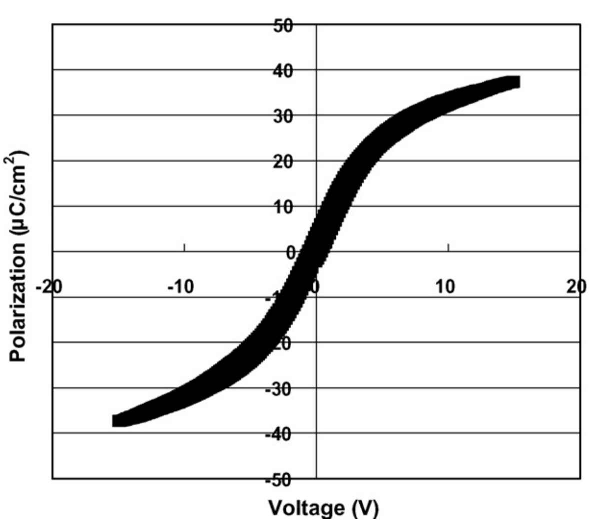

Fig. 2. Polarization hysteresis loop of 20PMN–10PNN–70PZT film at room temperature.

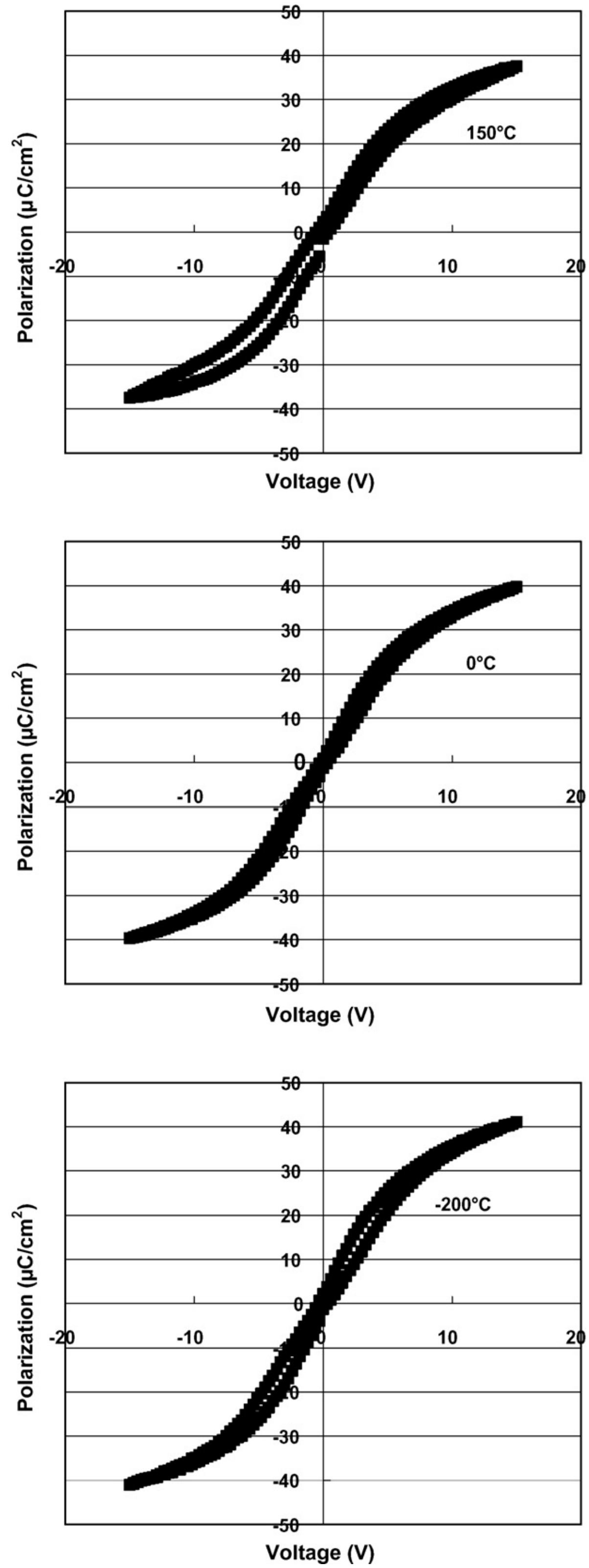

Fig. 3. Polarization hysteresis loops of 20PMN–10PNN–70PZT film at various temperatures.

polarization hysteresis loops were almost unchanged between temperatures in the range from −200 to 150 °C. The roundness of shape at 150 °C is probably due to increased leakage components in the films; a slight decrease in polarization is