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Effect of Poling Conditions on Hysteresis Properties of Lead Magnesium Niobate-Lead Zirconate Titanate Ceramics

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ABSTRACT

xPMN-(1-x)PZT ceramic composites with formula $xPb(Mg_{1/3}Nb_{2/3})O_3$-$(1-x)Pb(Zr_{0.52}Ti_{0.48})O_3$ (when x = 0.0, 0.1, 0.3, 0.5, 0.7, 0.9 and 1.0) were prepared by a conventional mixed-oxide method. The phase formation behavior of these ceramics was studied by an x-ray diffraction method. The hysteresis properties of the ceramics poled at different DC electric fields were measured by a simple Sawyer-Tower circuit. Generally, it was found that poling conditions have little or no effect on hysteresis properties of PMN-riched compositions but show marked effects on those of PZT-riched compositions. For the PMN ceramic, the polarization values, both remanent polarization (P_r) and spontaneous polarization (P_s), increase at the poling field is 10-20 kV/cm and little decrease at 30-40 kV/cm while the coercive field (E_c) remains constant. As for the PZT ceramic, the remanent polarization (P_r), the spontaneous polarization (P_s), and the coercive field (E_c) increase with increasing the poling field.

Keywords: Poling condition, Hysteresis properties, PMN-PZT

INTRODUCTION

PMN is a well-established relaxor-type ferroelectric of perovskite structure, owing to its excellent dielectric properties and its various applications in electronics industry (Wang *et al.*, 2000). PMN has advantages of having broader operating temperature range. In addition, as a result of their unique microstructural features, PMN ceramics exhibit low loss and non-hysteretic characteristics.

PZT is a well known piezoelectric, which is widely employed in a large number of sensing and actuating devices (Wang *et al.,* 2000). PZT ceramics have relatively high electromechanical coupling coefficients, as compared to PMN (Zhao *et al.,* 1999). However, PZT ceramics are fairly lossy as a result of their highly hysteretic behavior. With their complementary features, ceramics in PMN-PZT system are expected to have a combination of excellent properties of both ceramics.

Hysteresis loop is an important characteristic of the ferroelectric ceramics exhibiting the switching mechanism in these ceramics. Poling is the process in which a DC electric field exceeding the coercive field (E_c) is applied to a multidomain ferroelectric to produce a net remanent polarization (P_r). To pole a ceramic, an electric field is usually applied along one of the polar axes. Since coercive field is in general minimum near Curie temperature, common poling practice is to cool through the Curie temperature with a field applied (IEEE Standard

Definitions, 1986). Therefore, this study is undertaken to investigate the effect of poling conditions on the hysteresis properties of the PMN-PZT ceramic composites.

METHODLOGY

The $xPb(Mg_{1/3}Nb_{2/3})O_3$-$(1-x)Pb(Zr_{0.52}Ti_{0.48})O_3$ ceramic composites (when x = 0.0, 0.1, 0.3, 0.5, 0.7, 0.9 and 1.0) were prepared from the starting PMN and PZT powders synthesized by the columbite (Swartz and Shrout, 1982) and conventional mixed-oxide methods, respectively. Initially, the PMN and PZT powders for a given composition were weighed and then ball-milled in ethanol for 24 hours. After drying process, the mixed powders were pressed hydraulically at 1.5 N to form disc-shape pellets 10 mm in diameter and 1 mm thick, with 5 wt.% polyvinyl alcohol (PVA) as a binder. The pellets were stacked in a covered alumina crucible filled with PZ powders to prevent lead loss. Finally, the sintering was carried out at a sintering temperature for 2 hours with 5 min/°C heating and cooling rates. The firing profile includes a 1 hour dwell time at 500 °C for binder burn-out process to complete. For optimization purpose, the sintering temperature was shown in Table 1.

Table 1 Sintering temperature of xPMN-(1-x)PZT ceramics

Ceramics	Sintering temperature (°C)
PMN	1225
0.1PMN-0.9PZT	1275
0.3PMN-0.7PZT	1275
0.5PMN-0.5PZT	1275
0.7PMN-0.3PZT	1275
0.9PMN-0.1PZT	1250
PZT	1250

The phase formations of the sintered specimens were studied by an x-ray diffractometer (SIEMENS, D500) with CuK_α radiation ($\lambda = 0.15405$ nm) at room temperature. Before the properties measurements, the pellets were lapped to obtain parallel faces, and the faces were then coated with silver paint as electrodes. The samples were heat-treated at 750 °C for 12 minute. The samples are then poled in a silicone oil bath at a temperature of 110 °C with an applied DC field varying between 10 kV/cm and 40 kV/cm for 30 min and field-cooled to room temperature (Koval *et al.*, 2003)

The ferroelectric hysteresis (P-E) loops were measured by using a Sawyer-Tower circuit as shown in Figure 1. A sinusoidal field of 2200 V_{rms} and 50 Hz was applied to the sample with fixed R_1 R_2 and C_0. The voltage across the ceramic (C_s) was recorded on the horizontal axis (X-axis) of the oscilloscope, while the voltage across the capacitor (C_0), which is proportional to the polarization on the sample C_s, is recorded on the vertical axis (Y-axis) of the oscilloscope. The measurement was carried out at room temperature.

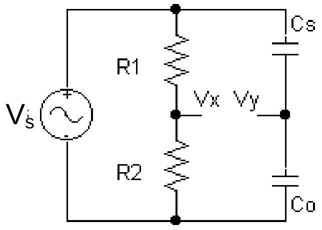


Figure 1 Sawyer-Tower circuit used for measuring hysteresis loop (Wongsaenmai *et al.,* 2003) ($R_1 = 6.8$ MΩ, $R_2 = 10$ kΩ, $C_0 = 0.1$ μF, C_s = Sample, $C_0 >> C_s$)

RESULTS AND DISCUSSION

The x-ray patterns, shown in Figure 2, show that the sintered ceramics are mainly in perovskite phase. From the XRD pattern, PZT ceramic is identified as a single-phase material with a perovskite structure having tetragonal symmetry as 2θ is 30.9, 31.4 and 55.7, while PMN ceramic is a perovskite material with a cubic symmetry as 2θ is 31.1, 44.5 and 55.4 . The PMN-PZT composites can be identified as an XRD single-phase material with a perovskite structure having the symmetry varying between tetragonal and cubic types.

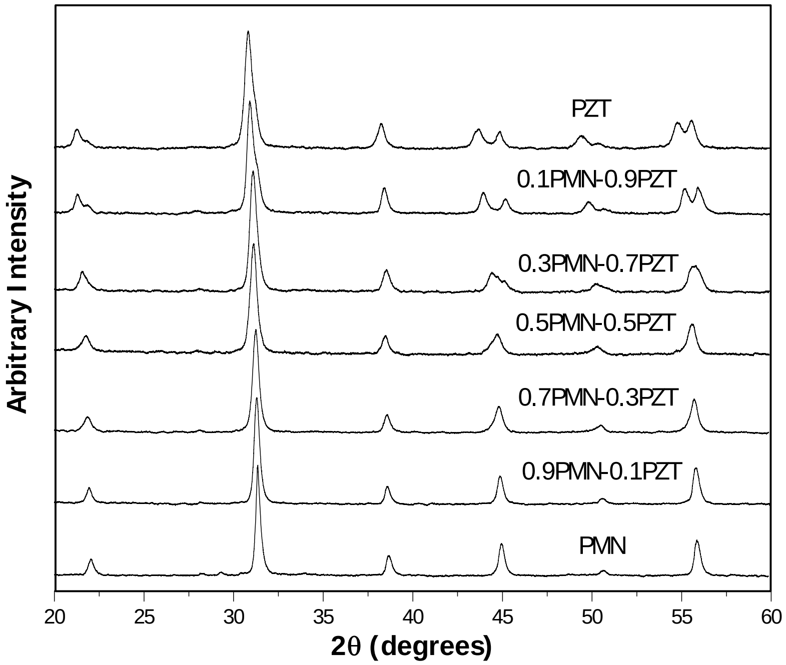
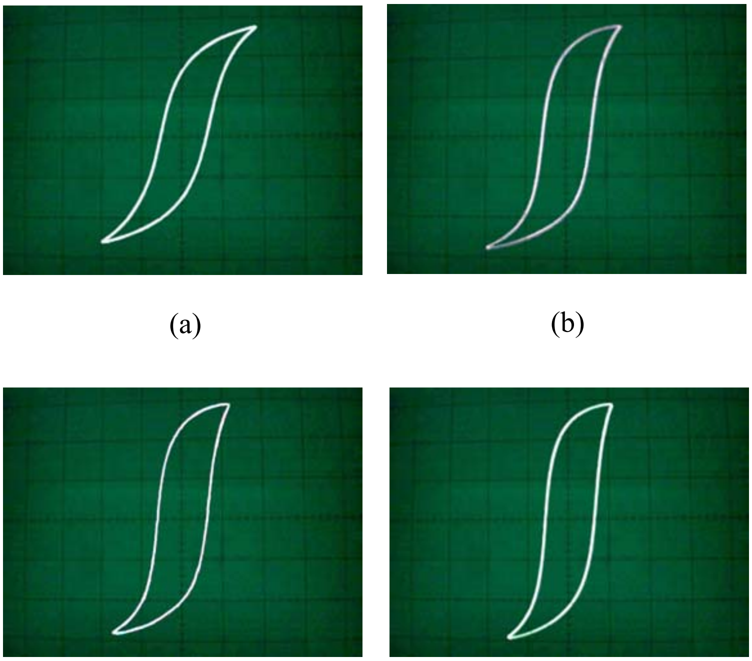


Figure 2 XRD patterns of the sintered (x)PMN-(1-x)PZT ceramics

A sequence of polarization-electric field (P-E) hysteresis loops for the (0.1)PMN-(0.9)PZT ceramics with various poling conditions is illustrated in Figure 3. It is evident that the shapes of P-E loops do not change with a poling field. The loops are more of a square form than of a slim form found in typical relaxor materials. This form of the loop is stipulated by abruptly switching of a domain structure in an electrical field. From the hysteresis loops, the remanent polarization (P_r), the spontanous polarization (P_s) and the coercive field (E_c) were determined and were listed in Table 2. For PMN ceramic the polarization values P_r and P_s increase at the poling field is 10-20 kV/cm and little decrease at 30-40 kV/cm, while the coercive field (E_c) remains constant. On the other hand, PZT ceramic shows an increase in the values of E_c and the polarizations (P_r and P_s) with poling field. This shows that when a DC field exceeding the coercive field is used to pole the PMN-PZT ceramics, a net remanent polarization and switching can be produced. Furthermore, it can be seen that a poling field strength does not have an influence on the hysteresis properties of PMN-riched compositions while it shows a marked effect on ceramics with PZT-riched compositions. This is a result of a characteristic of PZT ceramics with compositions near the morphotropic phase boundary (MPB) between the rhombohedral and the tetragonal phases in which the poling may draw upon 14 orientation states leading to exception polability. Therefore, they are easier to pole than PMN ceramic (Cross, 1996).



(a) (b)

(c) (d)

Figure 3 Hysteresis loops of (0.1)PMN-(0.9)PZT ceramic composites poled at (a) 10 kV/cm (b) 20 kV/cm (c) 30 kv/cm (d) 40 kV/cm

Table 2 Remanent polarization (P_r), spontanous polarization (P_s) and coercive field (E_c) of xPMN-(1-x)PZT ceramics.

Ceramics	**Poling field**	**E_c (kV/cm)**	**P_r ($\mu C/cm^2$)**	**P_s ($\mu C/cm^2$)**
PZT	10 kV/cm	0.79	0.82	1.68
	20 kV/cm	4.77	0.00	2.53
	30 kV/cm	7.95	10.10	23.58
	40 kV/cm	Break down	Break down	Break down
0.1PMN-0.9PZT	10 kV/cm	5.48	13.16	18.95
	20 kV/cm	5.48	18.09	22.21
	30 kV/cm	5.48	18.91	23.03
	40 kV/cm	5.48	19.74	23.03
0.3PMN-0.7PZT	10 kV/cm	4.29	21.26	22.96
	20 kV/cm	4.29	22.11	23.66
	30 kV/cm	4.29	22.11	23.81
	40 kV/cm	4.29	22.11	23.81

Table 2 (ext.)

Ceramics	**Poling field**	**E_c (kV/cm)**	**P_r (μC/cm^2)**	**P_s (μC/cm^2)**
0.5PMN-0.5PZT	10 kV/cm	4.34	18.10	21.76
	20 kV/cm	4.34	19.26	22.61
	30 kV/cm	4.34	19.26	22.61
	40 kV/cm	4.34	19.26	22.61
0.7PMN-0.3PZT	10 kV/cm	2.38	10.38	18.83
	20 kV/cm	2.38	11.98	20.54
	30 kV/cm	2.38	11.98	20.54
	40 kV/cm	2.38	11.98	20.54
0.9PMN-0.1PZT	10 kV/cm	0.96	4.23	16.07
	20 kV/cm	0.96	5.08	16.46
	30 kV/cm	0.96	5.08	16.92
	40 kV/cm	0.96	5.08	16.92
PMN	10 kV/cm	0.78	1.27	4.88
	20 kV/cm	0.78	1.63	9.76
	30 kV/cm	0.78	1.63	8.13
	40 kV/cm	0.78	1.63	10.57

In addition, by comparing between the ceramics poled at the same electric field, for instance at 30 kV/cm, it can be seen (Figure 4) that the hysteresis parameters (E_c, P_r and P_s) decrease with increasing the content of PMN into the xPMN-(1-x)PZT composites (when $x \geq 0.3$). This indicates that the hysteresis parameters are lowered as a result of PMN addition. This can be understood from the fact that PMN itself is transforming into a paraelectric (non-hysteresis) phase at temperature above –8 °C, which is its Curie temperature.

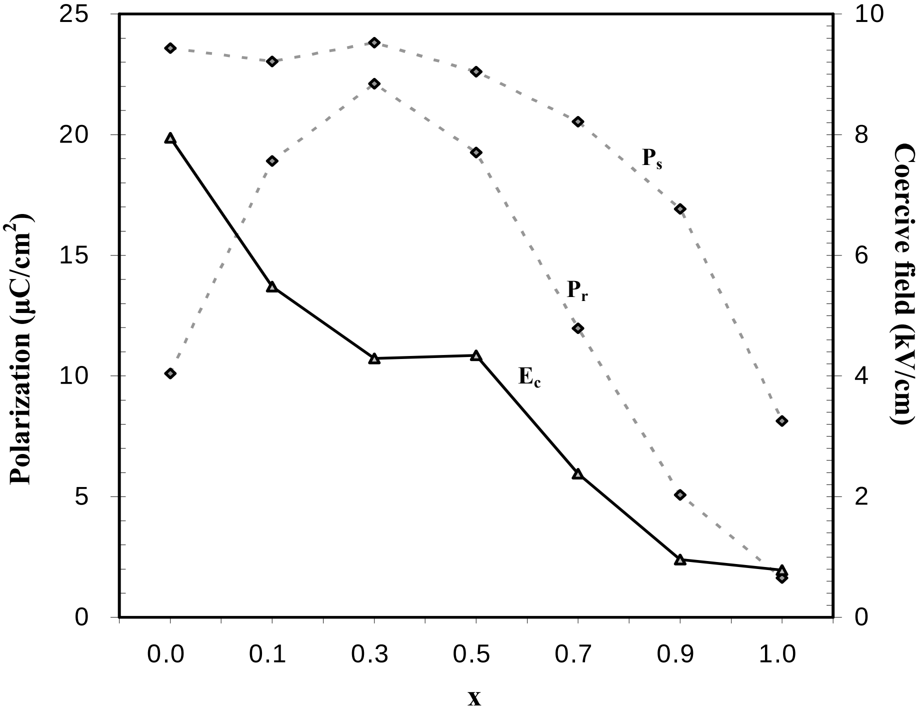


Figure 4 The P_r, P_s and E_c of xPMN-(1-x)PZT ceramics poled at 30 kV/cm

CONCLUSION

In this study, the ceramic composites with formula $xPb(Mg_{1/3}Nb_{2/3})O_3$-$(1-x)Pb(Zr_{0.52}Ti_{0.48})O_3$ (when x = 0.0, 0.1, 0.3, 0.5, 0.7, 0.9 and 1.0) are successfully prepared by a conventional mixed-oxide method. XRD studies indicate that these ceramics are single-phase materials with a perovskite structure. The hysteresis loops of the ceramics are of typical square loop. The measurement of hysteresis loops has revealed that poling conditions show no effect on the hysteresis properties of the PMN ceramics, while poling shows marked effect on the hysteresis properties of PZT ceramics. For the PMN ceramic, the polarization values, both remanent polarization (P_r) and spontaneous polarization (P_s), increase at the poling field is 10-20 kV/cm and little decrease at 30-40 kV/cm while the coercive field (E_c) remains constant. As for the PZT ceramic, the remanent polarization (P_r), the spontaneous polarization (P_s), and the coercive field (E_c) increase with increasing the poling field.

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