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The studies of single crystal PMN-PT68/32/polymer 1–3 composites

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Abstract

The single crystal PMN-PT68/32 is used to fabricate single crystal/polymer 1–3 composites with different PMN-PT volume fractions. These composites exhibit good properties in dielectric constant, piezoelectric constant and electromechanical coupling coefficient. The results are useful for underwater acoustic transducer applications.

Keywords: PMN-PT single crystal; Polymer; 1–3 Composites

1. Introduction

PMN-PT ferroelectric single crystal has been known to possess high piezoelectric coefficient ($d_{33} > 2000\,\text{pC/N}$), electromechanical coupling factor ($k_{33} \sim 0.9$) and electrically induced strain ($x_{33} \sim 1\%$) [1,2]. The single crystal has the potential for wider bandwidth due to an enhanced electromechanical coupling factor and the potential for higher projected acoustical energy density due to their bigger strain for ultrasonic and underwater acoustic transducer applications. However, for these applications, the relatively high acoustic impedance of the single crystal (approximately 30M rayls) results in a problem of acoustic impedance mismatch with that of human tissue (1.5M rayls) and water. In order to resolve this problem, the single crystal is combined into a passive polymer matrix to form 1–3 composites. The so-called 1–3 composites consist of parallel rods of PMN-PT single crystal embedded in a passive polymer matrix. The PMN-PT rods inside the composites are relatively free to vibrate. So, the 1–3 composites show good properties, such as higher thickness electromechanical coupling coefficient, intermediate dielectric constant, lower acoustic impedance, lower mechanical quality compared to PMN-PT single crystal and PZT ceramics. Someone studied the single crystal PMN-PT/epoxy 1–3 composites for ultrasonic transducer applications [3]. This work is researched the preparation and properties of the single crystal PMN-PT68/32/polymer 1–3 composites. The experimental data of the dielectric and piezoelectric properties and figure of merit dependence of hydrostatic pressure for the 1–3 composites are presented. The results are useful to underwater acoustic transducer applications.

2. Experimental procedure

Reagent grade PbO, MgO, Nb_2O_5, TiO_2 powders were used as raw materials. The single crystal of the composition PMN-PT68/32 was grown by an accelerated crucible rotation technique (ACRT) and Bridgman method [2]. The single crystal was oriented along pseudocubic [0 0 1] direction by means of XRD. The poled crystal samples were cut in [0 0 1] thickness directions into 1 mm × 1 mm × 3 mm rods. Four samples of single crystal/polymer 1–3 composites with PMN-PT 68/32 volume fraction of 0.30, 0.27, 0.24 and 0.22 (Sp A, Sp B, Sp C and Sp D for short) were made by the arrangement-and-cast technique. These crystal rods were arranged regularly in the reticulation with the crevice of 0.89, 0.99, 1.12 and 1.22 mm among the crystal rods, respectively. Polymer, epoxy E44 with a hardener 650 (in 1:1 ratio), was cast in the crevice among the crystal rods. The composites were allowed to cure at 80 °C in an

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oven for 12 h in order to harden and gain optimum properties. The composites were cut into the rectangle samples in different size after hardening. The samples were ground to smooth and subsequently electroded with gold for an hour. The samples were repoled in oil again with about electric field 2 kV/mm.

To measure the dielectric and piezoelectric properties of the composites with increasing pressure at room temperatures, the samples were amounted into the pressure vessel and the experimental date were simultaneously recorded by a HP4274 LCR meter at 0.4, 1, 10, and 100 kHz frequency. The pressure was generated in the pressure apparatus using transformer oil as the pressure transmitting media, the pressure measurements were calculated from the resistance of a manganin wire stress gauge in the pressure cavity using a computer controlled Keithley 2000 multimeter with the accuracy better than 0.2%. The electrical impedance Z versus frequency for the samples were measured with an HP4294 impedance analyzer.

3. Results and discussions

The measured properties of the crystal and four composite samples at room temperature and 1 bar are given in the following table.

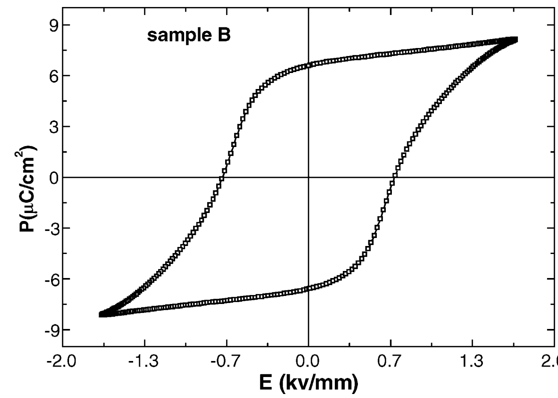

Fig. 1. P–E hysteresis loop for sample B.

constant ε', acoustic impedance Z decreased with increasing epoxy volume fraction compared to single crystal. The reason may be because the epoxy exerts a certain degree of lateral clamping on the PMN-PT crystal rods and modify its behavior as a totally free rod. In general, the soft polymer matrix can reduce the clamping to the single crystal and increase the electromechanical coupling properties, and decrease dielectric constant and acoustic impedance [3].

Figs. 1–6 are the measured properties for the sample B at different electric field, hydrostatic pressure and frequency.

	Crystal	Sp A	Sp B	Sp C	Sp D
(1) Size (mm)	16 × 10 × 3	10 × 8 × 1.5	11 × 9 × 1.5	10 × 9 × 1.5	11 × 9 × 1.5
(2) PMN-PT volume fraction (%)	1	0.30	0.27	0.24	0.22
(3) Density (g/cm^3)	8.02	3.19	3.004	2.80	2.64
(4) Dielectric constant ε'	4436	340	324	309	295
(5) Dielectric loss $\tan\delta$	0.015	0.013	0.024	0.02	0.022
(6) Series resonant frequency f_s (kHz)	630	707	630	707	630
(7) Parallel resonant frequency f_p (kHz)	793	1120	1120	1257	1120
(8) Frequency constant Nt (Hz m)	2380	1771	1748	1836	1703
(9) Acoustic impedance Z (M rayl)	38	11.2	10.5	10.3	9.16
(10) Electromechanical coupling factor k_t (%)	64.6	80.5	85.15	85.15	85.16
(11) Piezoelectric strain constant d_{33} (pC/N)	2000	584	521	473	424
(12) Hydrostatic piezoelectric strain constant d_h (pC/N)	80	111	102	93	81
(13) Hydrostatic piezoelectric voltage constant g_h (mV m/N)	2	37	36	34	31
(14) Figure of merit $d_h \times g_h$ (fm^2/N)	163	4115	3665	3188	2528

It is seen from the above data that the properties for 1–3 composites can be changed much more by using PMN-PT crystal rods embedded in the passive epoxy matrix. Some properties, such as electromechanical coupling factor k_t, hydrostatic piezoelectric voltage constant g_h, figure of merit $d_h \times g_h$ enhanced greatly, other properties, such as dielectric

Fig. 1 is typical of ferroelectric hysteresis loop, It is shown that the composite has clearly ferroelectric properties. Fig. 2 is the curve of the polarization dependence of the hydrostatic pressure, the curve slope is the hydrostatic piezoelectric strain constant d_h, d_h is about 102 pC/N for the sample B. Figs. 3–5 are dielectric constant ε', hy-

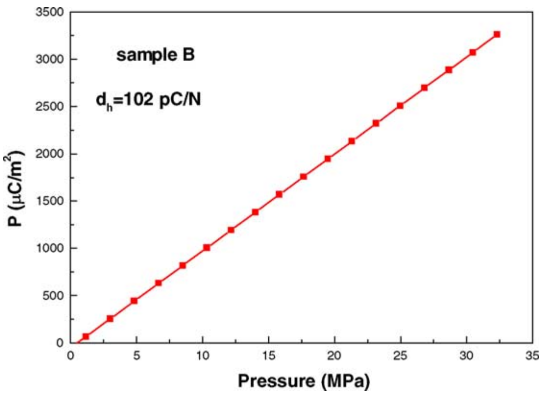

Fig. 2. P vs. pressure for sample B.

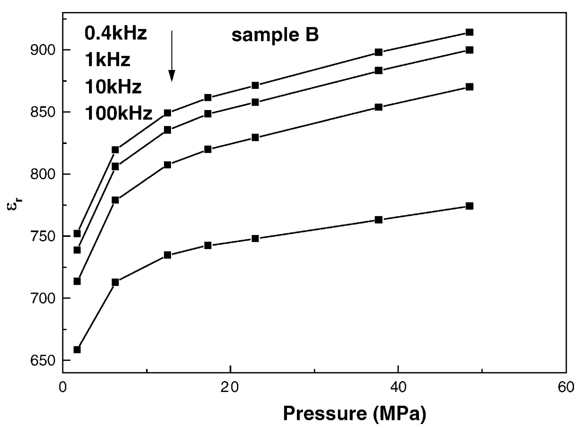

Fig. 3. ε_r vs. pressure for sample B.

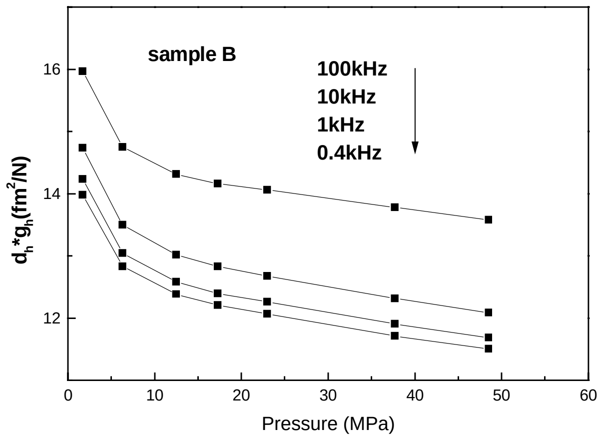

Fig. 5. $d_h \times g_h$ vs. pressure for sample B.

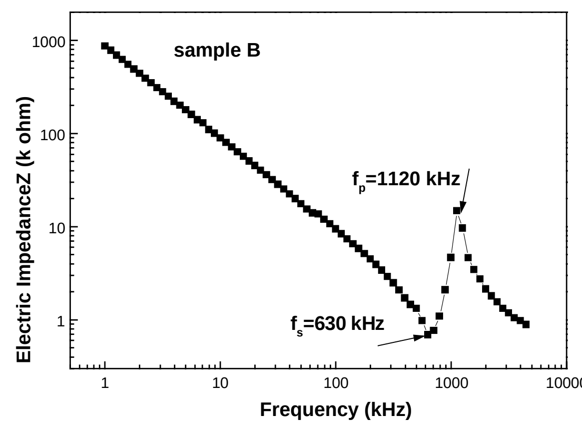

Fig. 6. Z vs. frequency for sample B.

drostatic piezoelectric voltage constant g_h, and figure of merit $d_h \times g_h$ dependence of the hydrostatic pressure at different frequency. The dielectric constant ε' increase with pressure, and reduce with frequency, respectively. The hydrostatic piezoelectric voltage constant g_h, and figure of merit $d_h \times g_h$ decrease with pressure, and increase with frequency, respectively. Fig. 6 is the electrical impedance Z dependence of frequency. The series resonant frequency f_s and parallel resonant frequency f_p of the thickness mode resonance in the composite B were determined by the curve of the electrical impedance Z versus frequency. Only a strong thickness resonance can be observed, but its third and fifth harmonics cannot be discovered. The curve of the electrical impedance Z versus frequency curve is very clean and tidy during the frequency range from 1 kHz to 10 MHz.

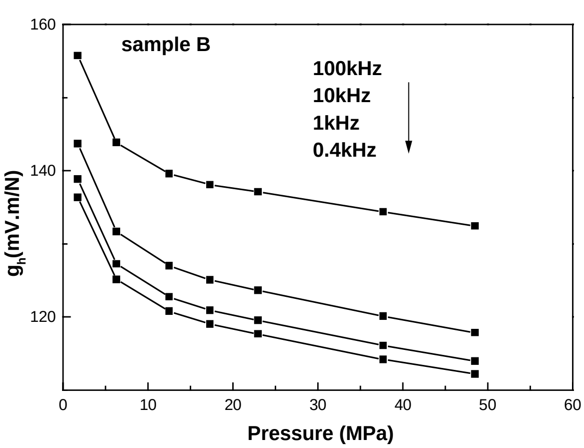

Fig. 4. g_h vs. pressure for sample B.

4. Conclusions

The single crystal/polymer 1–3 composites with different PMN-PT68/32 volume fraction have higher electromechanical coupling factor k_t, hydrostatic piezoelectric voltage constant g_h, figure of merit $d_h \times g_h$, and lower dielectric constant ε', acoustic impedance Z than that of the PMN-PT single crystal, PZT ceramics and PZT ceramics/polymer 1–3 composites. These good properties may be used for new generation underwater acoustic transducer applications.

Acknowledgements

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