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Dielectric and ferroelectric hysteresis properties of 1–3 lead magnesium niobate–lead titanate ceramic/Portland cement composites

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Abstract

In this work, lead magnesium niobate–lead titanate (PMN–PT) ceramic was cut and filled with Portland cement (PC) to produce 1–3 connectivity PMN–PT/PC composites. Dielectric and ferroelectric hysteresis properties of these composites with PMN–PT ceramic volume content of 60% were investigated. Room temperature dielectric constant (ε_r) at 1 kHz of the PMN–PT/PC composite was found to be ≈1500. At higher frequency (20 kHz), the dielectric constant was reduced to the value of ≈1300. Ferroelectric (polarization–electric field) hysteresis loops at 10–90 Hz and varying electric field were measured. The ''instantaneous'' remnant polarization (P_{ir}) at 50 Hz and at the electric field of 7 kV/cm of the PMN–PT/PC composite was found to be ≈10 μC/cm^2. These values of 1–3 composites therefore are promising when compared to previous results of composites at similar conditions.

Keywords: B. Composites; C. Dielectric properties; C. Ferroelectric properties; PMN–PT

1. Introduction

Piezoelectric-cement based composites have been developed in order to meet the acoustic matching of concrete for structural health monitoring application [1,2]. One of the mostly used piezoelectric ceramic materials is lead zirconate titanate (PZT) as it has very high piezoelectric coefficient (d_{33}) and dielectric constant (ε_r) and most of the piezoelectric-cement based composites were produced using PZT as the piezoelectric ceramic in the composite [1,2,7–13] from the previous investigated works [1–18]. Lead magnesium niobate–lead titanate ceramic, $Pb(Mg_{1/3}Nb_{1/2})O_3$–$PbTiO_3$ (PMN–PT) like PZT can be used in the application of smart structures. PMN–PT ceramic has a high dielectric constant (ε_r), high d_{33} value and an ideal curie temperature (T_c) at ≈150 °C compare to PZT at ≈400 °C [19,20]. For cement-based composite produced with piezoelectric materials other than PZT such as PMN–PT very little is known. In this research, PMN–PT/PC composite material was produced in the form of 1–3 connectivity from lead magnesium niobate–lead titanate (PMN–PT) and Portland cement. The dielectric and ferroelectric hysteresis behavior of the composites were then investigated.

2. Experimental

Columbite technique was first used to produce PMN powder. $MgNb_2O_6$ or MN (JCPDS file no. 88-0708) powder was first produced by calcining magnesium carbonate hydroxide pentahydrate $(MgCO_3)_4Mg(OH)_2\cdot5(H_2O)$ and Nb_2O_5 at 1150 °C. MN powder was mixed with lead oxide (PbO) in ethanol and milled together in a zirconia ball mill for 24 h, and were calcined at 800 °C to produce PMN powder. Lead titanate ($PbTiO_3$) was produced in a similar manner using mixed oxide method from PbO and TiO_2. XRD of the PMN and PT were determined to ensure their purity [21] before mixing the powder together at molar ratio of 0.67PMN–0.33PT in ethanol and milled together for 30 min (Fig. 1). PMN and PT powder were then pressed into pellets before sintering at 1250 °C for 2 h using a hydraulic press to form disk samples of 15 mm diameter and 2 mm thickness. During calcining and sintering, PbO and

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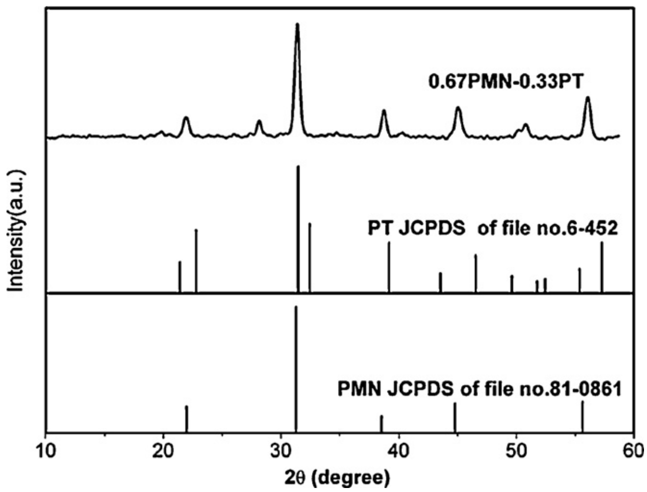

Fig. 1. X-ray diffraction patterns of PMN–PT.

PMN–PT powders were also used as excess powders to cover the sample in order to prevent lead volatilization.

These PMN–PT ceramic were then cut and filled with normal Portland cement (The American Society for Testing and Materials Type I cement) to produce 1–3 connectivity PMN–PT/PC composite using 60% PMN–PT by volume. A schematic diagram of PMN–PT/PC composites with 1–3 connectivity is illustrated in Fig. 2. Thereafter, the composites were placed for curing at 60 °C and 98% relative humidity for 5 days before measurements. An impedance meter (Hewlett Packard 4194A) was used to obtain the capacitance and the dissipation factor (tan δ) of the composites at room temperature and at frequency of 1 kHz. The relative dielectric constant (ε_r) was then calculated from the following equation:

$$\varepsilon_r = \frac{Ct}{\varepsilon_0 A}$$

where C is the sample capacitance, t is the thickness, ε_0 is the permittivity of free space constant (8.854×10^{-12} F m^{-1}), and A is the electrode area. The room temperature ferroelectric hysteresis (P–E) loops were characterized using a computer controlled modified Sawyer–Tower circuit. The electric field was applied to a sample by a high-voltage ac amplifier with the input sinusoidal signal with a frequency of 10–90 Hz from a signal generator.

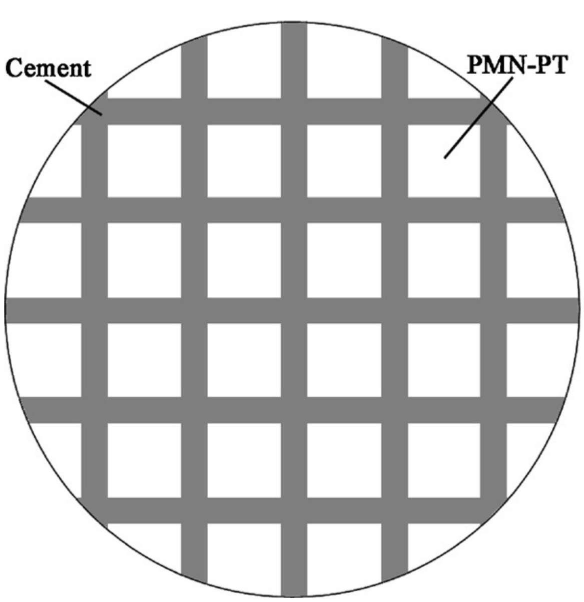

Fig. 2. Schematic diagram of 1–3 PMN–PT/PC composites.

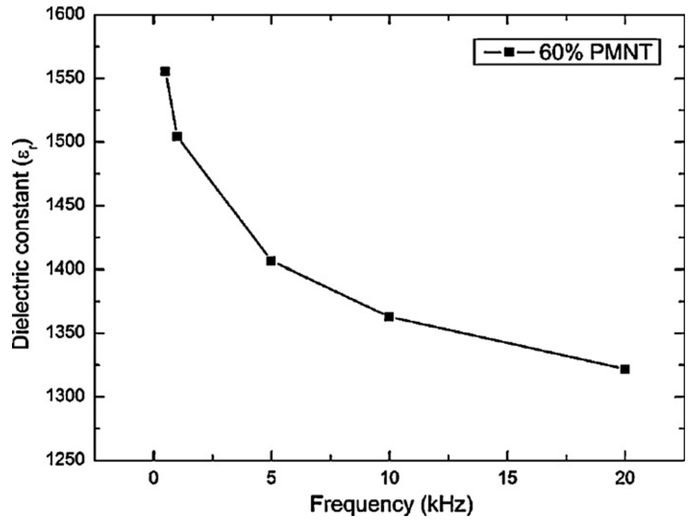

Fig. 3. Dielectric constants of PMN–PT/PC composites as plotted against frequency.

3. Results and discussion

The dielectric constant (ε_r) is shown against the frequency in Fig. 3 where the effect of frequency on the dielectric constant can be seen. The dielectric constant can be seen to decrease with increasing frequency where ε_r is ≈1500 at 1 kHz and ε_r is ≈1300 at 20 kHz. Moreover, when compared to the previous published results of 0–3 PMN–PT/PC composites [2], where ε_r is ≈500–700 for 50–70% PMN–PT, the dielectric constant of these 1–3 PMN–PT/PC composites can be seen to be much greater (almost doubled that of 0–3 composites). The dielectric

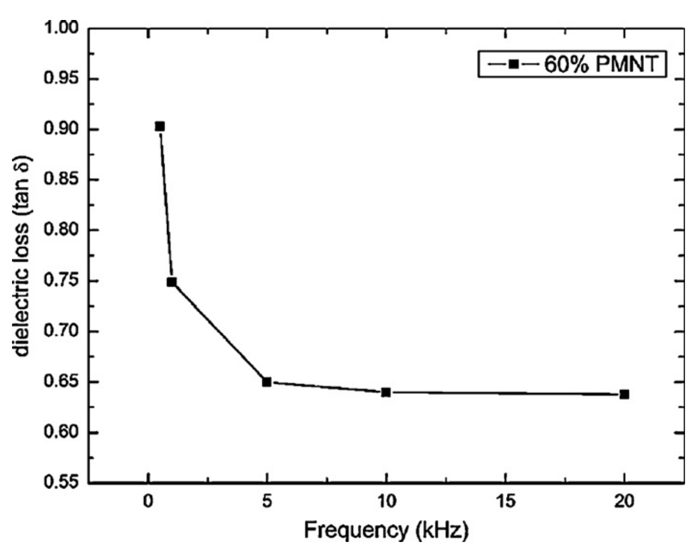

Fig. 4. Dielectric loss of PMN–PT/PC composites as plotted against frequency.

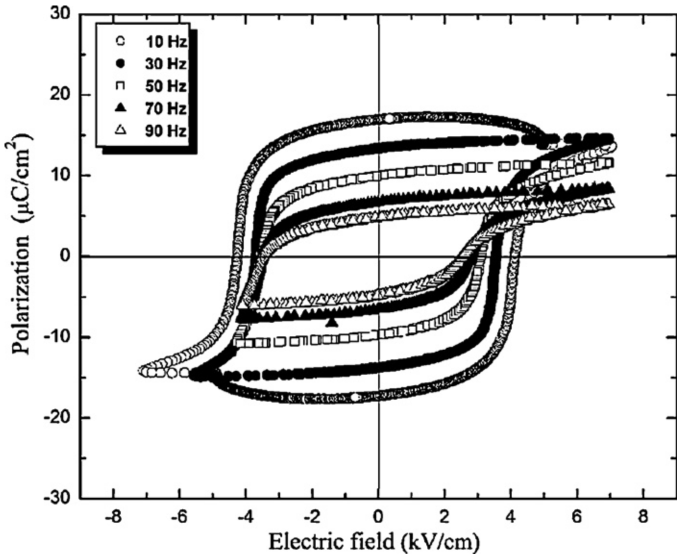

Fig. 5. Ferroelectric polarization–electric field (*P*–*E*) hysteresis loops of PMN–PT/PC composites at 10–90 Hz.

value therefore becomes closer to that of the pure PMN–PT ceramic where the measured ε_r is ≈2000 at 1 kHz. In addition, the dielectric loss of the composites (Fig. 4) was found to decrease with increasing frequency where the dielectric loss values reduced from 0.9 to 0.65 when the frequency increased from 0.1 to 20 kHz.

Hysteresis loops were plotted at 7 kV/cm and with varying frequency from 10 to 90 Hz (Fig. 5). For comparison, we define the *y*-axis intercept at a given applied field as the ''*instantaneous*'' remnant polarization (P_{ir}), while the *x*-axis intercept as the ''*instantaneous*'' coercive field (E_{ic}). Ferroelectric polarization–electric field (*P*–*E*) hysteresis loops at 10–90 Hz and varying electric field were observed. Although sharp end characteristic of normal hysteresis loop is seen for these composites, lossy appearance indicated by the increase in the total loop area can also be seen as a consequent of the cement matrix presenting in the 1–3 composites. Therefore, it should be noted here that fully saturated *P*–*E* loops cannot be achieved for these composites as a result of their highly lossy characteristics due to the presence of the weak conducting ions in the cement matrix [5]. Nonetheless, polarization–electric field (*P*–*E*) hysteresis loops measurements were still possible. For the ''instantaneous'' remnant polarization (P_{ir}) measured at 50 Hz and at the electric field of 7 kV/cm of the PMN–PT/PC composites was found to be ≈10 μC/cm^2. The effect of the frequency on P_{ir} and E_{ic} of PMN–PT/PC composites can be seen in Fig. 6(a) and (b), respectively. Quite clearly, it can be seen that P_{ir} decreases from 17 to 5 μC/cm^2 as frequency increases from 10 to 90 Hz. The E_{ic} also decreases from 4.2 to 2.5 kV/cm with increasing frequency (10–90 Hz). However, at a particular frequency and electric field used, these values of 1–3 composites are found to be significantly higher than previously published results of 0–3 composites. It is understandable that 1–3 composites essentially contain the piezoelectric ceramic fully aligned in one direction (1 connectivity prism) which allows the ceramic to possess greater dielectric

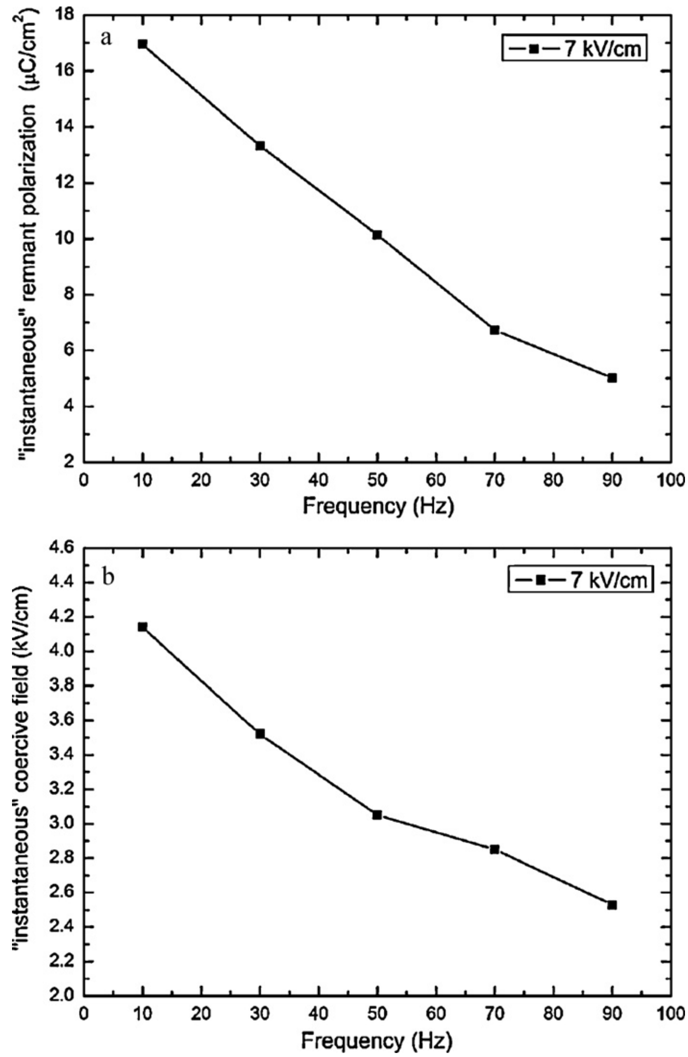

Fig. 6. The effect of the frequency on (a) P_{ir} and (b) E_{ic} of PMN–PT/PC composites.

properties and ferroelectric behavior giving less loss that would otherwise occur in the 0–3 composites. In 0–3 composites, the piezoelectric ceramic existed as random particles surrounded by cement matrix with high loss caused by the conducting ions [5,18,22]. In addition, less contact surface areas between the two materials would improve the properties of 1–3 composites when compared to 0–3 composites having the same volume content of ceramic [11].

4. Conclusions

The dielectric results of 1–3 PMN–PT/PC composites were found to be relatively high (ε_r = 1500 at 1 kHz) and is close to the pure PMN–PT ceramic (ε_r = 2000). Ferroelectric polarization–electric field (*P*–*E*) hysteresis loops of these composites show lossy appearance indicated by the increase in the total loop area as a consequent cement matrix presenting in the 1–3 composites. However, sharp end characteristics can still be observed. It was found that P_{ir} decreases from 17 to 5 μC/cm^2 as frequency increases from 10 to 90 Hz. E_{ic} value, on the other hand, decreases from 4.2 to 2.5 kV/cm. At 50 Hz and at the electric field of 7 kV/cm, measured P_{ir} of the PMN–PT/PC composite was found to be ≈10 μC/cm^2. At a similar frequency and electric field used, these values of 1–3 composite are found to be significantly higher than previously published results of 0–3 composites.

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