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Electrical and elastic properties of 1–3 PZT/epoxy piezoelectric composites

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Abstract This paper focused on the electrical and elastic properties of 1–3 PZT/epoxy composites fabricated by a dice-fill method. When PZT volume fractions are 10–35 vol.%, the piezoelectric constants d_{33} of the PZT/epoxy composites are about 170–270 pC/N and permittivity ε_r measured at 1 kHz is 80–350. The composites also possess lower acoustic impedance Z of 4–9 Mrayl, higher thickness electromechanical coupling coefficient k_t of 65% and better flexibility C_{33} of 13–33 GPa. Three mode resonant characteristics were analyzed based on the sizes and microstructural periodicity of composites, which had a great effect on k_t. Finally, these electrical and elastic properties of the composites were compared with the prediction based on a well-known homogenization model.

Keywords 1–3 piezoelectric composite · Fabrication process · Piezoelectric property · Dielectric constant

1 Introduction

Piezoelectric materials that transfer electrical energy to mechanical energy were widely used for electromechanical transducers. As the most promising candidate for high-frequency underwater hydrophone and biomedical imaging, 1–3 piezoelectric composites show many advantages over bulk piezoelectric ceramics: lower acoustic impedance (Z), higher coupling coefficient (k_t), lower mechanical quality factor (Q_m)and dielectric loss (tgδ) and better design flexibility [1–4]. Methods developed for processing fine-scale 1–3 piezoelectric composites include: dice-fill, lost mold, RIE(reactive ion etching)-LIGA, fiber processing and so on [3–6]. Among these methods, the simply and cheap processing of dice-fill has made it the standard commercial method in forming 1–3 piezoelectric composites for high-frequency application.

This paper focused on the piezoelectric and electromechanical properties of 1–3 PZT/epoxy composites with 10–35 vol.% PZT contents, and the resonant characteristic and elastic properties were also analyzed here. In addition, the experimental results were compared with the prediction based on the well-known homogenization model proposed by Smith [6, 7].

2 Experimental procedure

Commercially available PZT (LQ-PZT, Zr/Ti atomic ratio= 0.516:0.484; average particle size: 0.97 μm; Sakai Chemicals Co, Ltd., Japan) powder and epoxy resin (resin/hardener weight ratio=100:36, Bushler, USA) were used as the active ceramic and passive polymer, respectively. The relative densities of the bulk PZT samples sintered at 1200 °C for 2 h were all over 96%. The 1–3 piezoelectric composites were fabricated by a dice-fill process: first, PZT plates were periodically cut in two perpendicular directions by an automatic dicing saw (two different diamond saws were used here with the saws thickness of 350 and 150 μm, respectively). After the kerfs were filled by epoxy, the samples were degassed and cured at room temperature for more than 24 h. Then both sides of the samples were lapped until the rods appeared and silver glue was used as the surface electrode dried at low temperature (<150°C). Finally,

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the composites were polarized along the rod direction at 100°C for 15 min with the poling electric field of 2 kV/mm. Table 1 gives the dimensions of PZT rods and the periodic space and Fig. 1 shows the PZT rods after cutting.

PZT volume content can be calculated from the density of 1–3 composites. The dielectric properties and resonant characteristics were analyzed using a HP4194A impedance analyzer. The piezoelectric constant d_{33} was measured at 110 Hz by using a quaisistatic ZJ-A3 d_{33} meter (average value from 30 dots). Composites PE2 series were polished to different thickness to determine the effect of the thickness on the electrical properties.

3 Results and discussion

The theoretical model was used to calculate the elastic, piezoelectric and dielectric properties. The physical parameters of epoxy and monolithic PZT from references [8, 9] were used as the input data for calculation.

Figure 2 shows the variations of d_{33}, ε and g_{33} with PZT volume content. When PZT volume fractions were 10–35 vol.%, the piezoelectric constants d_{33} of the PZT/epoxy composites were about 170–270 pC/N and permittivity ε_r measured at 1 kHz was 80–350, both increase with increasing PZT volume content. However, the experimental values were lower than those calculated from the theoretical model. It was mainly due to the unhomogeneous structure of the composites and the discrepancy of physical parameters between real PZT bulks and those from references. Piezoelectric voltage constant g_{33} means the sensitivity of a receiving voltage, which was calculated from: $g_{33}=d_{33}/\varepsilon$. As shown in Fig. 2, the experimental values of g_{33} correspond well with the theoretical model, and the composites possessed the highest value of 177 (10^{-3} m^2/V) with PZT 10 vol.%. In addition, the theoretical curve showed that g_{33} of the composites would always be higher than that of pure PZT when PZT fraction is higher than 7 vol.%.

Round 1–3 PZT/epoxy piezoelectric composites mainly consist of three resonant modes: planar-mode, thickness-mode and lateral-mode resonance. Here the resonant frequencies of thickness-mode and planar-mode are determined by the typical size of samples, yet lateral-mode resonance is related to the microstructural periodicity of samples. Figure 3 shows the variations of electrical impedance with decreasing thickness of the samples, where the resonant peaks can be catalogued into the following modes: First, the planar-mode resonance usually appeared at a lower frequency (f_p). The larger diameters of samples PE2 put the planar-mode to a lower frequency so it was beyond this frequency band (100 kHz–10 MHz). Second,

Table 1 Feature parameters of the fabricated PZT/epoxy composites.

Samples	PZT vol.%	PZT rods (μm)	Distance between rods (μm)
PE1	10.5	250	450
PE2	17.8	350	450
PE3	20.2	400	450
PE4	32.9	250	220

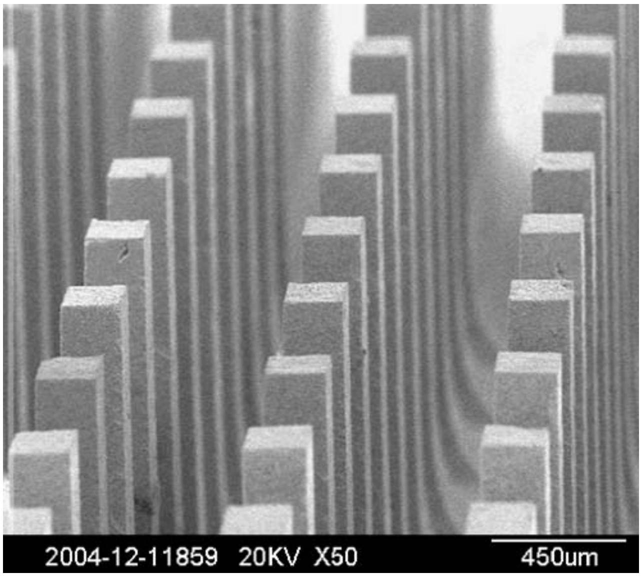

Fig. 1 PZT rods after cutting

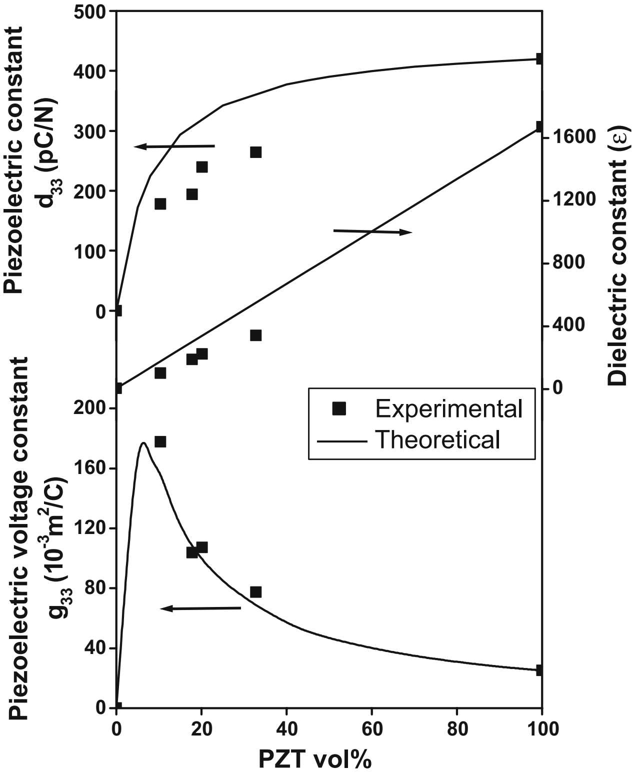

Fig. 2 Variations of piezoelectric constant d_{33}, dielectric constant ε and piezoelectric voltage constant g_{33} with PZT volume fraction

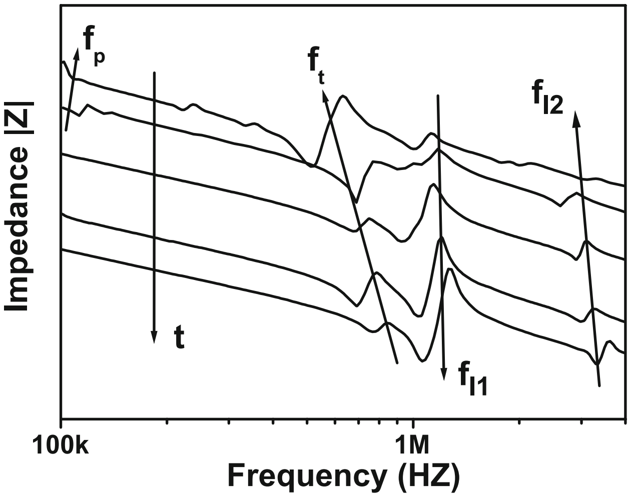

Fig. 3 Variations of impedance with frequency for PE2 series

the thickness-mode resonance, which is the typical and useful mode for such electromechanical transducers, would happen at a higher frequency (f_t) than planar-mode. In this mode PZT rods vibrate in the fundamental length longitudinal 33 mode and the unclamped piezoelectric coupling coefficient k_{33} is applicable for this mode. Since the composites are treated as a monolithic transducer material, the resonance of the composite corresponding to the 33 mode of the PZT rods will be referred to as thickness-mode resonance. As seen from Fig. 3, the thickness-mode resonance was obvious when the thickness was much larger than the periodicity of a sample (see the top curve). However, as the thickness decreased, f_t transferred to a higher frequency, and when f_t and f_{l1} became too closer, the thickness-mode would easily couple with the lateral-mode (see the others except the top line). Third, lateral-mode resonance is undesired for 1–3 composites. If the thickness-mode resonance is strongly mixed with the lateral modes, and consequently the transducer using such a composite will work in a very inefficient way. This situation must be avoided by maintaining a fine periodicity in the distribution of the phases as was pointed out in reference [10].

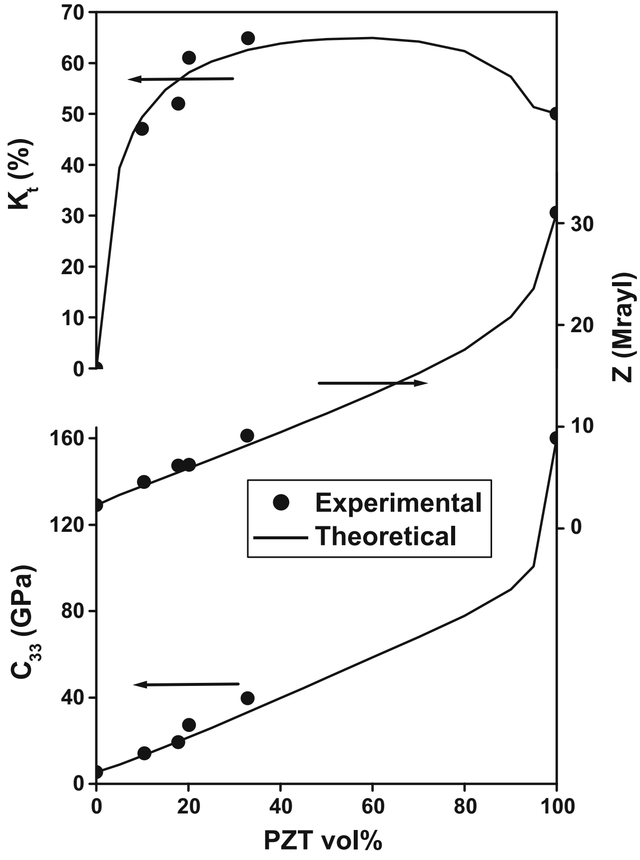

Fig. 4 Variations of thickness electromechanical coupling coefficient k_t, acoustic impedance Z and elastic stiffness coefficient C_{33} with PZT vol.%: experimental values (*dots*) compared with theoretical calculations (*line*)

Thickness-mode electromechanical coupling coefficient k_t, acoustic impedance Z and elastic stiffness constant C_{33} are evaluated through the following equations:

$$k_t^2 = \frac{\pi f_r}{2f_a}\,\mathrm{tg}\,\frac{\pi(f_a - f_r)}{2f_a} \tag{1}$$

$$Z = \rho_{\mathrm{com}} \cdot v_{\mathrm{com}},\; v_{\mathrm{com}} = f_r \cdot 2t \tag{2}$$

$$C_{33}^D = 4\rho_{\mathrm{com}} t^2 f_p^2 = 4\rho_{\mathrm{com}} N_t^2 \tag{3}$$

Where $f_r = f_s = f_{\min}$, $f_a = f_p = f_{\max}$; R1 is the real impedance at the resonant frequency $f_{\min}$; C^T is the static capacitance of

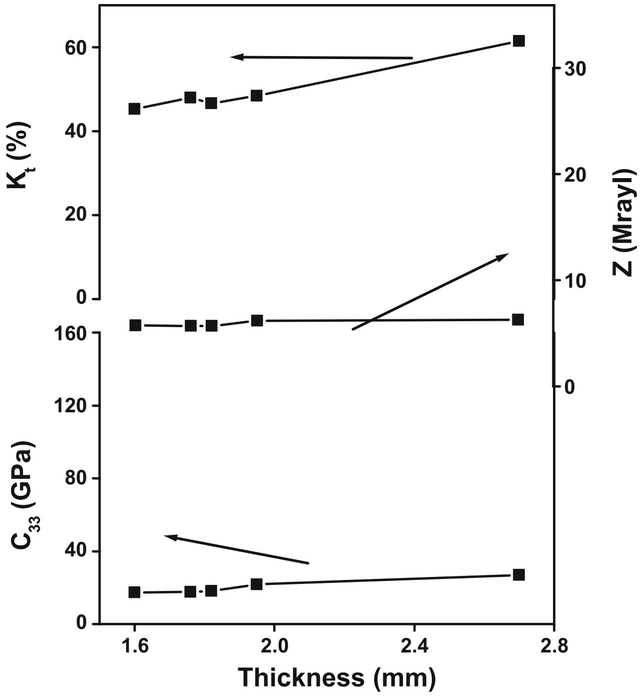

Fig. 5 Variations of thickness electromechanical coupling coefficient k_t, acoustic impedance Z and elastic stiffness coefficient C_{33} with thickness for PE2 series

1–3 composite at 1 kHz; ρ_{com}, v_{com} represent for the density and acoustic velocity of 1–3 composite; and t is the thickness. These properties k_t, Z and C_{33} as functions of PZT volume content and thickness are listed in Figs. 4 and 5, respectively, compared with their theoretical values.

Figure 4 shows that thickness electromechanical coupling coefficient k_t increased greatly when PZT volume fractions were lower than 20%. And sample PE4 with PZT 32.9 vol.% possesses the highest k_t of 65%, very close to that of single phase PZT (k_{33}=70%). The higher k_t values than pure PZT (50%) were mainly due to the 33 mode of PZT rods. The acoustic impedance Z and the elastic stiffness coefficient C_{33} both increased almost linearly with PZT vol.%, which is a good agreement between the model and experiment supporting the explanation of the great dependence of these properties on PZT volume fraction. In all, the piezoelectric composites possessed much lower Z (3–10 Mrayl) and better flexibility (13–33 GPa) than those of pure PZT.

Figure 5 illustrates the electromechanical coupling coefficient k_t increased with increasing thickness, being different from the case for the effective k_t values [2]. This was mainly caused by the coupling effect of the thickness-mode and the lateral-mode, as seen from Fig. 3. The acoustic impedance Z tended to be stable with decreasing thickness for the same composition of samples, for it is an intrinsic property of material. However, the elastic stiffness coefficient C_{33} ascended lightly with the thickness for the same composition. This slight increase was mainly attributed to a more homogeneous structure with a higher thickness.

4 Conclusion

In this work, 1–3 PZT/epoxy piezoelectric composites were fabricated by a dice-fill method. Quaisistatic electrical properties and dynamic properties drawn from resonant characteristics were systematically investigated. The results show that the mode-coupling was intensive as the thickness decreased. The obtained 1–3 piezoelectric composites possess better properties than pure PZT, such as: higher piezoelectric voltage constant g_{33} ($78.5–177\times10^{-3}$ m^2 V) and higher thickness electromechanical coupling coefficient k_t (48–65%), lower acoustic impedance Z (3–10 Mrayl) and better design flexibility (13–33 GPa).

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