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Lili Li

The Pennsylvania State University, Xi'an Jiaotong University

Shujun Zhang

Blatek Inc., The Pennsylvania State University, Xi'an Jiaotong University, shujun@uow.edu.au

Zhuo Xu

Xi'an Jiaotong University

Fei Wen

Xi'an Jiaotong University

Xuechang Geng

Blatek, Inc.

See next page for additional authors

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1-3 piezoelectric composites for high-temperature transducer applications

Abstract

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Authors

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1–3 piezoelectric composites for high-temperature transducer applications

Lili Li^{1,2}, Shujun Zhang^{1,2,4}, Zhuo Xu¹, Fei Wen¹, Xuecang Geng³,
Hyeong Jae Lee² and Thomas R Shrout²

¹ Electronic Materials Research Laboratory, Key Laboratory of the Ministry of Education & International Center for Dielectric Research, Xi'an Jiaotong University, Xi'an 710049, People's Republic of China

² Materials Research Institute, Pennsylvania State University, University Park, Pennsylvania 16802, USA

³ Blatek Inc., State College, Pennsylvania 16801, USA

E-mail: soz1@psu.edu

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Abstract

High-temperature Pb(Zr,Ti)O₃/epoxy 1–3 composites were fabricated using the dice and fill method. The epoxy filler was modified with glass spheres in order to improve the thermal reliability of the composites at elevated temperatures. Temperature-dependent dielectric and electromechanical properties of the composites were measured after ageing at 250 °C with different dwelling times. Obvious cracks were observed and the electrodes were damaged in the composite with unmodified epoxy after 200 h, leading to the failure of the composite. In contrast, composites with > 12 vol% glass sphere loaded epoxies were found to exhibit minimal electrical property variation after ageing for 500 h, with dielectric permittivity, piezoelectric coefficient and electromechanical coupling being on the order of 940, 310 pC N⁻¹ and 57%, respectively. This is due to the improved thermal expansion behaviour of the modified filler.

(Some figures may appear in colour only in the online journal)

1. Introduction

High-temperature ultrasonic transducers are widely used in various applications, including high-temperature non-destructive testing (NDT) and well logging for oil, gas and geothermal industries [1–4]. Most of the oil wells' temperature is below 175 °C; however, it is about 200 °C for high-pressure and high-temperature wells, and further increases to 250 °C and above for geothermal wells. Thus, high-temperature ultrasonic transducers with operational temperatures ≥ 250 °C are desirable [5, 6]. Transducers using conventional PZT monolithic ceramics have been fabricated for different ultrasonic applications at elevated temperature; however, they suffer from the low sensitivity, low resolution and poor signal-to-noise ratio, associated with the low thickness coupling factors and high acoustic impedance of the ceramics [7].

1–3 piezoelectric composites offer advantages of increased thickness electromechanical coupling factors, reduced lateral vibration mode across the width of the resonator, and tailored acoustic impedance, leading to the

improved resolution and bandwidth of transducers [8–11]. In addition, 1–3 composites show improved conformability and a large reduction of thermal stress in the transducers due to the compliancy of the polymer phase [12, 13]. Previous work on high-temperature 1–3 piezoelectric composites was focused on LiNbO₃/cement and PZT/epoxy structure [14–19]. LiNbO₃/cement composites were reported to be functional above 400 °C; however, the low electromechanical coupling of LiNbO₃ single crystal limited their applications. Conventional 1–3 PZT/epoxy composites are limited in the temperature usage range of <180 °C, due to the relatively low glass transition temperature (T_g) and high thermal expansion coefficient (TEC) of the polymer fillers. Thus, internal stress induced by the different TEC of ceramics and polymers, may give rise to cracking and debonding in the composites and leads to structural failure. In order to obtain lower TEC in the epoxy, one effective approach is to introduce inorganic filler particles and/or short fibres with low TEC into the epoxy resin matrix, where fused silica (TEC = 0.5 ppm °C⁻¹) has been widely used as filler [20, 21].

In this work, high-temperature 1–3 composites fabricated using high Curie temperature (T_c) PZT ceramic (TRS203,

⁴ Author to whom any correspondence should be addressed.

$T_c \sim 380^\circ\text{C}$) and glass spheres modified epoxy (to reduce the TEC of the epoxy) were studied. Temperature-dependent properties of the composites were investigated; the temperature stability and reliability (high-temperature ageing) were evaluated.

2. Experimental procedures

PZT composites were fabricated using the conventional dice and fill method [22]. The piezoelectric ceramics used in this study were commercially available high T_c PZT (TRS203, TRS Technologies Inc.). The temperature-dependent properties of TRS203 were compared with conventional PZT5A (TRS200, TRS Technologies Inc.). For the passive phase, Duralco 4703 polymers (Cotronics Corp.) with various volume ratios of glass spheres ($\sim 0\%$, 4%, 12% and 20%) were investigated. The kerf and pillar widths were controlled to be 0.27 mm and 0.81 mm, respectively, with PZT volume fraction being on the order of 56%. The glass sphere modified epoxies were then backfilled into the kerfs and cured at 120°C for 4 h and then aged at 230°C for 16 h. The final thickness of the composites was about 3.4 mm, giving fundamental thickness resonance frequency of ≤ 500 kHz.

The glass transition temperatures T_g of pure and glass sphere modified epoxies were determined using dynamic mechanical thermal analysis (DMTA) (defined as the maximum value of mechanical loss), using a dynamic mechanical analyser (DMA Q800, TA Instruments), on samples with dimensions of $25\text{ mm} \times 10\text{ mm} \times 4\text{ mm}$. Thermal expansion measurements were carried out in the temperature range $30\text{--}275^\circ\text{C}$ using a dilatometer (DIL 402 PC, NETZSCH), on samples with dimensions of $25\text{ mm} \times 5\text{ mm} \times 5\text{ mm}$. The TEC, α , was calculated according to

$$\alpha_T = \frac{1}{L_0} \times \frac{\Delta L}{\Delta T}, \quad (1)$$

where L_0 is the specimen length at room temperature [23]. The densities of the pure and modified epoxies were measured by the Archimedes method. The longitudinal velocities of the epoxies were determined by the ultrasonic pulse-echo method using a 15 MHz longitudinal wave transducer (Ultra Laboratory Inc.). The acoustic impedance Z of the epoxies was then determined according to

$$Z = \rho V_L, \quad (2)$$

where ρ is the density and V_L is the longitudinal wave velocity. The dielectric permittivity and loss as a function of temperature were determined from the capacitance and loss measured by an HP4284A LCR meter connected to a computer-controlled furnace, while the electromechanical coupling factor was determined by the resonance and anti-resonance frequencies measured using an HP4194 impedance phase-gain analyser, according to the IEEE Standard [24].

3. Results and discussion

3.1. Properties of pure and modified epoxies

The properties of pure and modified epoxies were investigated. Figure 1 shows the temperature-dependent TEC for epoxies

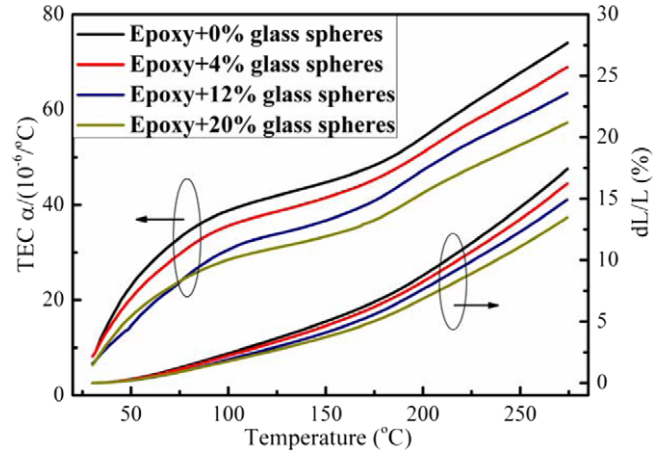


Figure 1. TEC as a function of temperature for pure and glass loaded epoxies. (Note that the left arrow illustrates TEC and the right arrow illustrates dL/L .)

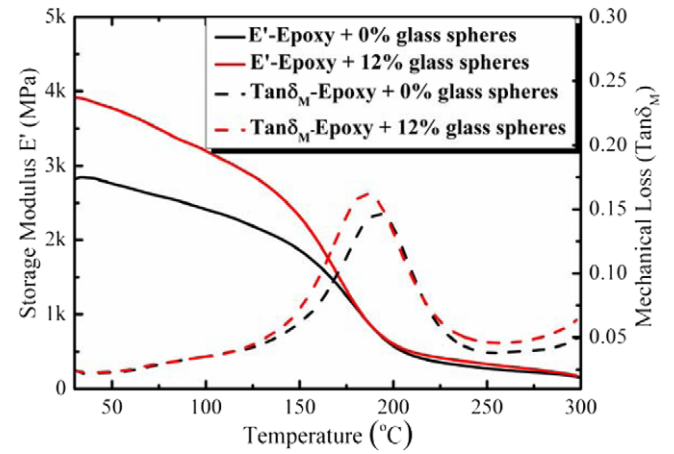


Figure 2. Temperature-dependent storage modulus and $\tan \delta_M$ for pure and 12% glass modified epoxy.

modified with different volume ratios of glass spheres. It was found that TEC increased as a function of temperature for all the studied epoxies, but decreased with increasing glass sphere volume ratio, with values being on the order of $68\text{ ppm }^\circ\text{C}^{-1}$ and $59\text{ ppm }^\circ\text{C}^{-1}$ at 250°C for pure and 12% glass loaded epoxies, respectively.

Figure 2 shows the temperature-dependent storage elastic moduli for the epoxy matrix with 0% and 12% glass spheres. It can be observed that the storage modulus E' for the epoxies decreased as temperature increasing and exhibited a sudden drop at about 190°C , indicative of the softening of the epoxies. It should be noted that the incorporation of glass spheres increases the room temperature storage (elastic) modulus of the epoxy matrix, being 2800 MPa for the pure epoxy and increased to 4000 MPa for 12% glass sphere modified epoxy, without sacrificing the glass transition temperature T_g (peak point of damping- $\tan \delta$). The temperature-dependent dielectric properties for the epoxies are given in figure 3. It was found that the dielectric permittivity and dielectric loss for all the epoxies increased at about 200°C . Of particular interest is that the dielectric permittivity increased and the dielectric

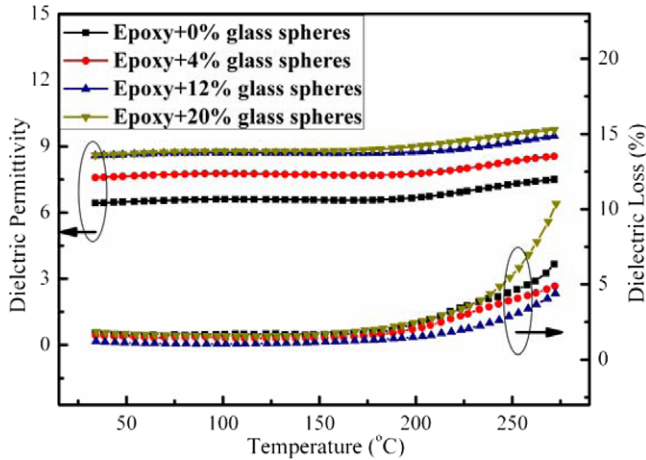


Figure 3. Temperature-dependent dielectric permittivity and loss measured at 10 kHz for pure and glass loaded epoxies. (Note that the left arrow illustrate dielectric permittivity and the right arrow illustrate dielectric loss.)

Table 1. Material parameters for pure and glass sphere loaded epoxies.

Parameter	Epoxy (0%) ^a	Epoxy (4%)	Epoxy (12%)	Epoxy (20%)
ρ (g cm ⁻³)	1.85	1.93	2.17	2.03
Z (Mrayl)	4.8	5.1	6.4	5.5
T_g (°C) ^b	192	195	187	204
TEC at 250 °C (ppm °C ⁻¹) ^c	68	63	59	52
Dielectric loss at 250 °C	5%	4%	3%	6%
Dielectric permittivity at 250 °C	7	8	9	9

^a Epoxy (x%) means epoxy with x vol% glass spheres.

^b T_g is the glass transition temperature.

^c TEC is the thermal expansion coefficient.

loss decreased, as glass spheres volume ratio increased from 0% to 12%.

Table 1 summarizes the properties of the pure and modified epoxies. The density and the acoustic impedance of the epoxies were found to increase with glass sphere volume ratio increased from 0% to 12%, above which, the acoustic impedance decreased, due to the fact that the viscosity of the epoxy was too high with 20% glass spheres, leading to the air bubbles in the epoxy [25, 26]. Cracking and debonding may happen in the composites with excessive bubbles when the temperature increased due to the internal stress induced by different TEC of epoxy and air bubbles. This may lead to structural failure of the 1–3 composites and affect the thermal stabilities of the composites. This problem would be much more severe when the air bubbles are at the interface of ceramic rod/epoxy. It should be noted that the glass transition temperatures were similar, being on the order of 192–204 °C, regardless of the glass sphere loading.

3.2. Properties of monolithic PZT ceramics

Prior to the fabrication of high-temperature 1–3 composites, the properties of high T_c monolithic PZT ceramics (TRS203) were measured and compared with PZT5A (TRS200).

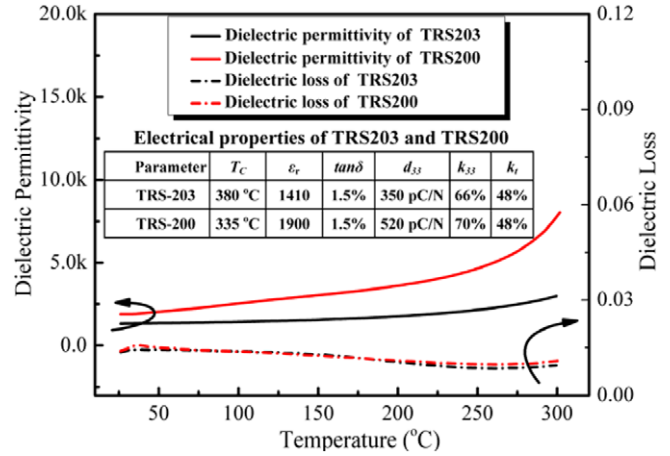


Figure 4. Dielectric permittivity and loss as a function of temperature for TRS203 and TRS200. (Note that the left arrow illustrates dielectric permittivity and right arrow illustrates dielectric loss.)

Figure 4 shows the temperature dependence of dielectric permittivity and loss of TRS203 and TRS200 (measured at 1 kHz), the room temperature properties were given in the small inset table. The dielectric losses of both ceramics were found to be <2% (25 °C–300 °C). The temperature coefficient of dielectric permittivity of TRS203 was found to be 0.46% °C⁻¹ in the temperature range 25 °C–300 °C, being much lower than that of TRS200 (1.2% °C⁻¹), demonstrating improved temperature stability. It was found that TRS200 exhibited higher d_{33} and k_{33} ($d_{33} = 520$ pC N⁻¹, $k_{33} = 70\%$) compared with TRS203 ($d_{33} = 350$ pC N⁻¹, $k_{33} = 66\%$); however, the T_c of TRS203 (380 °C) was much higher than that of TRS200 (335 °C). Figure 5 shows the temperature dependence of the electromechanical coupling factor k_{33} for high T_c PZT (TRS203) and compared with TRS200. Of particular significance is that the coupling factor k_{33} of TRS203 maintain the same value from room temperature to 300 °C, indicating that TRS203 exhibited high thermal stability with broadened temperature usage range up to 300 °C.

3.3. Thermal stability and reliability of 1–3 composites

In this section, the room temperature properties, thermal stability and reliability of 1–3 composites were evaluated. The room temperature properties of 1–3 composites are summarized in table 2. The measured piezoelectric coefficients d_{33} were found to be on the order of 310 pC N⁻¹, slightly lower than that of monolithic PZT ceramic (~350 pC N⁻¹), while the calculated electromechanical coupling factors k_t were found to be 57%, higher than the thickness coupling of monolithic ceramics (~48%). The acoustic impedance and mechanical quality factors Q_m of the composites were found to be on the order of 20–22 Mrayl and <50, respectively, lower than those values of PZT ceramic (30 Mrayl and ~80), which will benefit the high sensitivity and broad bandwidth transducer applications.

The thermal stability and reliability of PZT (0% & 12%) composites were evaluated by ageing the composites at 250 °C up to 200 h and 500 h, respectively. The dielectric behaviour

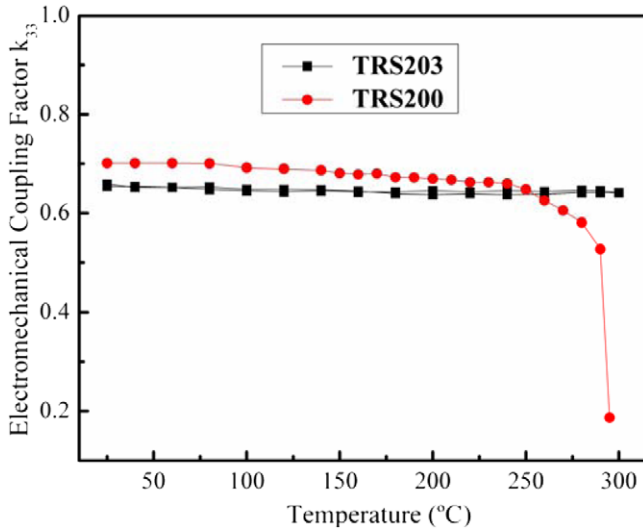


Figure 5. Electromechanical coupling factor k_{33} as a function of temperature for TRS203 and TRS200.

Table 2. Electrical properties of high-temperature 1–3 composites.

Parameter	PZT (0%) ^a	PZT (4%)	PZT (12%)	PZT (20%)
Dielectric permittivity	900	900	940	950
Dielectric loss	1.0%	1.0%	1.0%	1.0%
d_{33} (pC N ⁻¹)	310	310	310	310
k_t (k_{33})	57%	57%	57%	57%
Z (Mrayl)	20	20	21	22
Q_m^b at RT	45	45	50	50
Q_m at 250 °C	30	30	25	25

^a PZT ($x\%$) means epoxy with x vol% glass spheres.

^b Q_m is the mechanical quality factor.

as a function of temperature and ageing time for PZT (0%) and PZT (12%) composites are given in figures 6(a) and (b), respectively, with the electromechanical coupling factor shown in the small insets. The dielectric permittivity for PZT (0%) was found to be 900 at RT, increasing to 1520 at 250 °C, while the corresponding dielectric loss was found to be 1.5% at RT, increased to 5% at 250 °C. The electromechanical coupling factor was found to be 57% at RT, slightly increasing to 60% at 250 °C, with minimal variation. Similar dielectric and electromechanical behaviours were also observed for PZT (12%), as shown in figure 6(b).

However, obvious cracks were observed in the unmodified epoxy after ageing for 200 h, as shown in figure 7, leading to the damage of electrodes on PZT (0%) composite. The piezoelectric coefficient d_{33} and electromechanical coupling k_t were found to be 170 pC N⁻¹ and 43%, respectively, indicating the failure of PZT (0%) composites. In order to delineate the degradation mechanism, the surface of the PZT (0%) composites was polished and re-electroded. It was found that both dielectric and electromechanical properties followed the same trend as the virgin sample, indicating that the property degradation of PZT (0%) was attributed to the structural failure instead of the depolarization of the PZT ceramic, which may be caused by the internal stress induced by the different TECs of PZT ($\alpha \sim 2$ ppm °C⁻¹ at 250 °C) and the pure epoxy ($\alpha = 68$ ppm °C⁻¹ at 250 °C) [27].

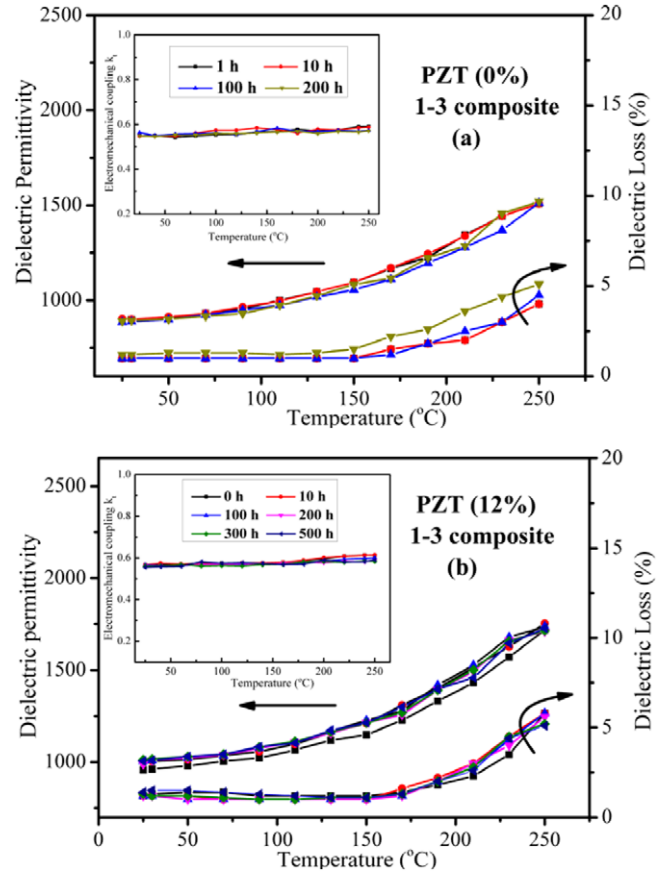


Figure 6. Temperature-dependent electrical properties for 1–3 composites ageing at 250 °C for various times (a) PZT (0%) 1–3 composite and (b) PZT (12%) 1–3 composite. (Note that the left arrows illustrate dielectric permittivity and the left arrows illustrate dielectric loss.)

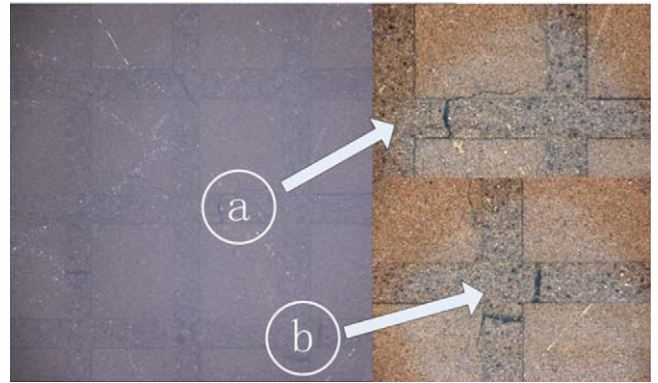


Figure 7. Photos of PZT (0%) composite after ageing for 200 h.

In contrast to PZT (0%) composite, PZT (12%) composite exhibited improved thermal reliability when aged at 250 °C. As shown in figure 6(b), no obvious variation in the dielectric permittivity, dielectric loss and electromechanical coupling were observed with ageing times up to 500 h. The dielectric permittivity of PZT (12%) composite was found to be 1010 after ageing for 500 h, slightly higher than the value before ageing ($\epsilon_r = 950$), while the dielectric loss, piezoelectric coefficient d_{33} and electromechanical coupling k_t were found to maintain the same value of 1.5%, 300 pC N⁻¹ and 56%,

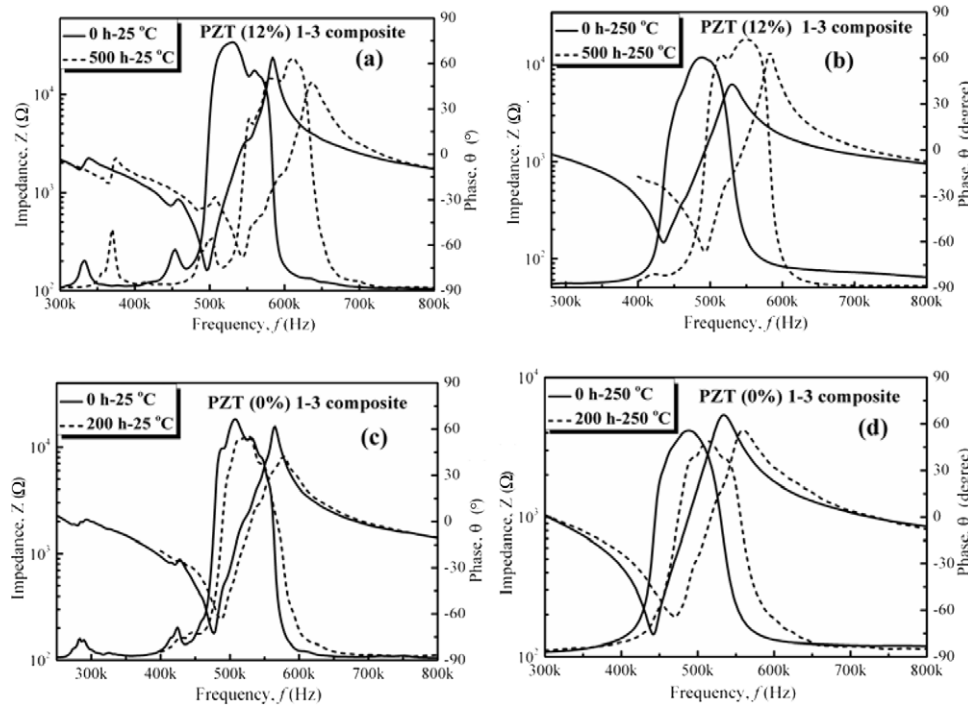


Figure 8. Measured electrical impedance and phase before and after ageing for 1–3 composites: (a) PZT (12%) composite at RT, (b) PZT (12%) composite at 250 °C, (c) PZT (0%) composite at RT and (d) PZT (0%) composite at 250 °C. (Note that after ageing for 200 h, the electrode of PZT (0%) composites was damaged, which was removed and re-electroded. The impedance and phase spectrum of PZT (0%) composite after 200 h were measured after re-electrode, as given in (d).)

respectively. Of particular significance is that no structural failure was observed with high-temperature ageing. This improved thermal reliability was attributed to the low TEC (at 250 °C) of epoxy (12%) ($\alpha = 59 \text{ ppm } ^\circ\text{C}^{-1}$) compared with the pure epoxy ($\alpha = 68 \text{ ppm } ^\circ\text{C}^{-1}$).

Figure 8 shows the electrical impedance and phase angle of PZT (0% and 12%) composites before and after ageing. As shown in figures 8(a) and (b), relatively high phase angle (70° at 250 °C) was observed after ageing for 500 h, indicative of a low damping (low loss) of PZT (12%) composite, demonstrating minimal property degradation. However, for PZT (0%) composite, low phase angle (47° at 250 °C) was observed, as shown in figures 8(c) and (d). The resonance and anti-resonance frequencies of both 1–3 composites were found to shift to higher frequencies after ageing, revealing the increasing of the effective velocities induced by the hardening of the polymer after ageing. It was observed from figure 8 that for both composites, the thickness coupling factors increased (from 57% to 60%) but the mechanical quality factor Q_m decreased (from 45 to 30 for PZT–0% and 50 to 25 for PZT–12% composites), as the temperature increased to 250 °C. This is due to the fact that as the temperature increased, the polymer became softer and the thickness mode became pure due to the less clamping from the soft polymer, which resulted in the enhancement of k_t . On the other hand, as the temperature increased, the electrical impedance at resonance frequency increased, while the magnitude of the electrical impedance at anti-resonance frequency decreased, indicated the increased damping and mechanical loss. And the increased mechanical loss leads to decrease in mechanical quality factor Q_m .

4. Conclusion

In conclusion, the thermal stabilities and reliability of high-temperature 1–3 piezoelectric composites were studied. Temperature-dependent dielectric and electromechanical properties of the composites were measured after ageing at 250 °C with various dwelling times. The reliability of the composites was improved using glass sphere modified epoxy fillers, which has lower thermal expansion coefficient. Obvious cracks were observed and the electrodes were damaged in PZT (0%) composite after ageing for 200 h, leading to the structural failure. In contrast, neither electrical properties variation nor structural failure was observed for PZT (12%) composite after ageing at 250 °C up to 500 h, due to the decreased thermal expansion of the modified filler, making it a potential candidate for NDE type transducer applications at elevated temperature.

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