



Highly flexible piezoelectric 0–3 PZT–PDMS composites with high filler content



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ABSTRACT

Flexible piezoelectric composites offer alternative and/or additional solutions to sensor, actuator and transducer applications. Here high density, highly flexible piezoelectric composites with 0–3 connectivity using filler volume fractions up to 50 vol.% are realized by solution casting of dispersions of ferroelectric $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$ (PZT) in poly-dimethylsiloxane (PDMS). Excellent piezoelectric properties (permittivity ϵ_r up to ~ 40 , piezoelectric charge constant d_{33} up to 25 pC/N, piezoelectric voltage coefficient g_{33} up to 75 mV m/N), electrical properties (conductivity σ about 1×10^{-6} S/m at 1000 Hz) and mechanical properties (storage modulus E' up to 10 MPa, loss modulus E'' less than 0.5 MPa, limited creep and stress relaxation) have been realized. The high flexibility combined with excellent properties of these composites opens new ways to 'soft touch' applications in a variety of transducer and sensor applications.

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1. Introduction

Piezoelectric composites with 0–3 connectivity are of paramount importance for sensors, actuators and transducers in a variety of fields. In particular, flexible soft composite materials relate to transducer and sensor applications for the generation and detection of underwater acoustic signals (sonar), medical diagnostic systems (e.g. micropumps in micro-/nano-fluidic devices) and tactile sensors (energy scavenger for aerospace/automotive/domestic devices or touch-base switches for the consumer market). For each of the above mentioned applications, a different balance in properties is required. In many cases the most important requirements are that the composite films should be flexible and soft in the appropriate thickness range and still possess as good as possible piezoelectric properties [1–3].

Due to the increasing demands on structural performance for these electronic devices, there has been extensive research carried out in the design and fabrication of polymer–matrix composites (PMCs) in recent years [4–7]. Composite materials allow for optimization of electrical, magnetic and mechanical effects and this resulted in the emergence of many advanced functional materials. These PMCs offer several advantages over other types of materials: their ability to be tailored with properties by varying the volume fraction of the ceramic inclusions and their ease of production including the option to realize easily different sizes and shapes [8–10].

The most widely used ceramic for such composite materials is (ferroelectric) lead zirconate titanate $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$ or PZT because of its excellent electromechanical properties [11,12]. The properties of PMCs will depend not only on the nature of the phases but also the connectivity of the ceramic particles and the matrix. For the matrix of 0–3 composites various polymers are used.

In a previous study, we reported on the processing and characterization of 0–3 lead zirconate titanate/liquid crystalline thermoset/polyamide (PZT/LCT/PA) composites [13]. Hot-pressing was utilized for the fabrication and the effect of the volume fraction of PZT on the piezoelectric and dielectric properties was studied. The experimental data of permittivity and piezoelectric constants were compared with several theoretical models (Jayasundere, Yamada and Lichtenecker) for these 0–3 composites. In order to assess the correlation of the experimental data with the theoretical models, the experimental data obtained from PZT/PA composites were also included. The matrix used leads to relatively stiff composite materials.

An interesting matrix material choice is poly-(dimethylsiloxane) (PDMS), a silicon-based elastomer with repeating unit of $\text{SiO}(\text{CH}_3)_2$. Due to its very low glass transition temperature T_g , PDMS exhibits rubbery behavior at room temperature. Properties such as elastic behavior, resistance to high temperatures, resistance to radiation and chemical attack make PDMS suitable for a wide range of applications in electrical and optical devices [14,28].

In the present work, 0–3 PZT–PDMS composites were fabricated with volume fractions up to 50 vol.% PZT ceramic particles by solution casting. The electrical, dielectrical and mechanical properties

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were investigated as a function of ceramic volume fraction and frequency. These PZT–PDMS composites offer the advantage of high flexibility in comparison with other 0–3 composites, even with 50 vol.% PZT. These composites possess the ability to attain various sizes and shapes, each with high flexibility (Fig. 1a) due to the exceptional elastic behavior of PDMS, combined with good functional properties. Only a few papers report on flexible composites with relatively high ceramic volume fractions. Sakamoto et al. [15,16] studied the electro-active properties of composites of PZT up to about 30 vol.% in a poly-urethane matrix. Ba–Sr–titanate–zirconate up to 40 vol.% in an epoxy matrix was studied by Yang et al. [17] and these authors conclude that the Yamada model is inadequate because of the unknown shape factor. However, due to the nature of these matrices, the elastic moduli (not reported) are probably much higher than for PDMS composites.

Liou and Chiou [18] described the incorporation of Ba–Sr–titanate up to about 60 vol.% in PDMS rubber and studied in particular the dielectric tunability. However, their materials contained a bulk porosity of 4–7% and a surface porosity of 18–24%. These authors also observed a relatively high shape factor of 14.0 for the Yamada model and attributed that to clustering of the filler and the surface effect. No study was made of the flexibility. Sharma et al. [19] used PDMS composites with up to 32 vol.% PZT for active damping. No comparison of the experimental permittivity and elasticity results was made with theoretical expressions. Romasanta et al. [20] incorporated up to 8 vol.% Ca–Cu–titanate in PDMS and studied their electro-mechanical response. Also these authors observed a relatively high shape factor of 12.5 for the Yamada model, stated that their results are in agreement with others and conclude that the Yamada model is applicable for these composites. As far as the authors are aware of, no other papers studied the mechanical properties of flexible 0–3 composites in some detail.

2. Experimental

2.1. Materials

The PZT powder used in this research is a commercial half-product PZT5A4 (Morgan Electro Ceramics, Ruabon, UK), a soft PZT with 1 mol% Nb added as dopant. Before use it was thermally treated, as reported in [13] where further details can be found. The average size of the filler is about 1.0 μm and the details of the filler size also can be found in [13]. The polymer used is linear vinyl-terminated poly(dimethylsiloxane) (PDMS17 with $M_w = 17,200$ g/mol; ABCR GmbH & Co) cross-linked with the four-functional siloxane, tetrakis(dimethylsiloxane) (ABCR GmbH & Co). The hydrosilylation reactions were catalyzed by cis-dichlorobis (diethylsulphide) platinum (II) catalyst (Strem Chemicals, Inc) previously

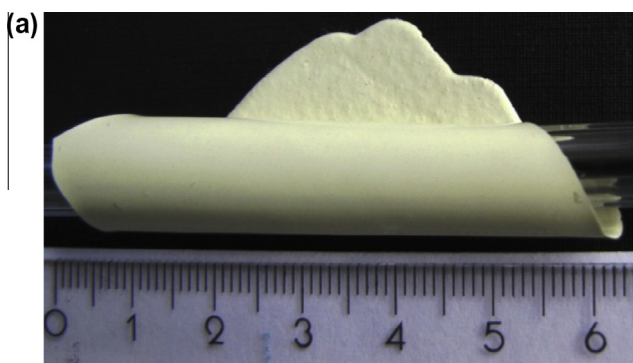


Fig. 1a. Photograph showing the flexibility of a 40 vol.% PZT–PDMS solution casted composite film by folding over a pen (thickness ~ 280 μm).

dissolved in a toluene solution. Esteves et al. [14] reported on the details of the hydrosilylation addition reaction to obtain cross-linked (tri-dimensional network) PDMS composites by reacting functional end groups on the PDMS chains with a multifunctional cross-linker in the presence of a catalyst. We used the same hydrosilylation addition reaction to realize the PZT–PDMS composites.

2.2. Fabrication of composites

In brief, the materials PZT (10–50 vol.%) and PDMS (90–50 vol.%) were mixed in a speed mixer (DAC 150 FVZ) at 3000 rpm for 2 min and the appropriate amount of cross-linker is added and mixed again for 1 min and finally the catalyst is added and mixed for 1 min. This mixture is directly solution casted on a polycarbonate sheet and subsequently dried and cured under vacuum initially at 60 $^{\circ}\text{C}$ for 20 h and at 110 $^{\circ}\text{C}$ for 5 h. Attempts were made to realize composites with 60% PZT but the viscosity of the dispersion appeared too high to be able to cast the material properly. Composites with specific dimensions of 14 mm in diameter and 200–275 μm thickness were cut from the composite films. Circular gold electrodes with a thickness of 300 nm and an area of $7.85 \times 10^{-5} \text{ m}^2$ were sputtered on both sides of the composites using an Edwards sputter coater (model S150B). The poling of the electroded sample is performed by applying an electric field of 12 kV/mm (Heinziger 10 kV power supply) at 120 $^{\circ}\text{C}$ for 30 min in a silicone oil bath to ensure uniform heating. The electric field was kept on during cooling to room temperature.

2.3. Measurements

The relative density of the composites were measured by the displacement of the solvent (2 wt.% sodium dodecyl sulphate (Merck) in ultra-pure water) using a dynamic contact angle measuring instrument/tensiometer (Dataphysics DCAT 21). Impedance data of the composites were collected by an impedance analyzer (EG&G Princeton Applied Research, Model 1025) coupled with a potentiostat (Potentiostat/Galvanostat, Model 283) at room temperature at a frequency range of 10 mHz–5 MHz. The piezoelectric charge constant was measured with a d_{33} meter (Piezotest, PM300) at a fixed frequency of 110 Hz. The d_{33} and ϵ_r values obtained at 110 Hz were used to calculate the piezoelectric voltage coefficient g_{33} according to:

$$g_{33} = d_{33}/\epsilon_0\epsilon_r \quad (1)$$

where d_{33} is the piezoelectric charge constant in pC/N, ϵ_0 is the permittivity of free space (8.85×10^{-12} F/m) and ϵ_r is the relative permittivity of the composite. The microstructure of the composites was examined by SEM (FEI, Quanta 3D FEG). The mechanical properties of the composite films were tested in a tensile mode on DMA (TA Instruments Q800 series) at room temperature. The static elastic modulus E of the composites was measured by performing stress–strain tests with a strain rate of $0.5\% \text{ min}^{-1}$. Creep measurements were conducted at 0.1 MPa, a stress level within the linear viscoelastic region while stress relaxation measurements were conducted at a constant strain of 0.55%. Both were monitored for 140 min. The dynamic behavior of pure PDMS and 40 vol.% PZT–PDMS composites were studied using a temperature ramp and multi-frequency mode. The specimens were heated at a constant rate of 3 $^{\circ}\text{C}/\text{min}$. While heating, the specimens were deformed at constant amplitude over a single frequency. The mechanical properties were also measured over a range of discrete frequencies. The frequencies 1, 5 and 10 Hz were used in this study. The dynamic tests were conducted at constant force amplitude of 5 μm with a preload force of 0.01 N. The temperature range used was from -120 $^{\circ}\text{C}$ to $+120$ $^{\circ}\text{C}$ at a frequency of 1 Hz.

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