



## Surface-condensed piezoelectric fibers and composites



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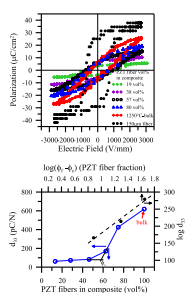
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### HIGHLIGHTS

- Surface-condensed PZT fibers exhibit more favorable manipulation strength.
- The  $k_p$ ,  $f_r$ ,  $P_r$  and  $P_s$  of PZT fibers largely increased while the aspect ratio  $>2.8$ .
- The  $d_{33}$  increased with volume fraction of PZT fibers after a threshold 58 vol%.
- Boric acid-PVA surface-condensed a capsule-like web to protect PZT fibers.

### GRAPHICAL ABSTRACT



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### ABSTRACT

An extrusion process that combined the surface condensation reaction was used to fabricate PZT fibers to exhibit a smooth surface and a circular morphology, with favorable manipulation strength. The sintered fibers showed more tetragonality. The electromechanical coupling coefficient and the resonant frequency of the PZT fibers had increased, while the aspect ratio of length to diameter ( $R$ ) was larger than 2.8. Single PZT fibers with  $R > 2.8$  showed larger remanent and saturation polarization values than the 1-3 composite and the PZT bulk. The dielectric constant and saturation polarization were in proportion to the volume filling percentage of the PZT fibers in the 1-3 composite. The piezoelectric coefficient increased with the fraction of the PZT fibers after a threshold volume fraction of 58 vol% was reached. As a proposed mechanism, the boric acid-PVA condensation reaction was initiated at the extruded fiber surface to form a capsule-like web to protect the extruded PZT fiber surface, increase the fiber strength, maintain the circular morphology, and facilitate fiber manipulation.

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## 1. Introduction

Lead zirconate titanate (PZT) is an excellent ferroelectric and piezoelectric material near the morphotropic phase boundary (MPB) at a Zr/Ti mole ratio of 52/48 [1–3]. The ferroelectric and piezoelectric properties are dependent on the microstructure, crystallization of the material, and the geometric arrangement as

well as composition of the composite [4–7]. However, stiff ceramic piezoelectrics have drawbacks, including high densities, limited strains, brittleness, and lack of conformability.

PZT fibers are useful in composite structures for several actuating and sensing applications, such as vibration control, energy harvesting, high-strain actuators, and high-frequency ultrasonics for medical imaging. The fine, strong ceramic fibers combine in a polymer matrix with limited stiffness and toughness to form the PZT–polymer composites, which show promising electromechanical energy conversions. In the 1-3 type composite, the active PZT fibers are arranged in 1 dimension (1D) and the passive polymer

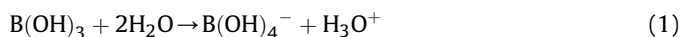
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filler in 3 dimensions (3D). The polymer acts as a protector to transfer stress, and modifies the dielectric properties and mechanical energy conversion efficiencies of the composite [6]. This kind of fiber composite also offers increased conformability to the more irregular and curved surfaces, which is critical for large-scale coverage.

The PZT–polymer composites have been prepared using various methods. Cochran et al. [8] researched injection molding and laser machining methods for preparing ultrafine scale 1-3 connectivity, piezoelectric ceramic–polymer composites. Their methods were associated with high tooling costs and dimensional changes, and experienced cross-sectional tapering and material re-deposition. Other methods, including thermal and chemical removal of the sacrificial polymer molds [9], controlled placement of sintered piezoelectric fibers [10], and the lost silicone mold technique [11], have limited commercial use because they impose constraints on volume fractions, minimum dimensions, material quality, cost, time, and repeatability of manufacture. Although the viscous suspension spinning process (VSSP) also produces fine-scale fibers, a microscopic investigation revealed that the VSSP produces non-homogeneous fibers [12].

In the 1-3 structures, a standard, aligned, 1-directional property plays a major role because of the 1D polarization. To maximize actuation and to achieve practical electromechanical energy conversion efficiency, the PZT fibers used in the 1-3 composites offer greater strength and conformability potential for large-area coverage and directional actuation [13,14]. The prepared fibers are required to possess specific characteristics. Smooth-fiber surfaces, uniform circular cross sections, low porosity, and small grain sizes are required for strength reinforcement, electromechanical energy conversion, and volume packing in composites [15].

Polyvinyl alcohol [PVA,  $(\text{CH}_2\text{CHOH})_n$ ] has a film-forming ability and an excellent binding property. It also exhibits a surfactant property because it contains hydrophilic and hydrophobic groups in its molecular structure [16]. Boric acid ( $\text{H}_3\text{BO}_3$ ) dissolved in a water solution generates a borate ion  $[\text{B}(\text{OH})_4^-]$  because the hydroxyl ion combines with water, as shown in (1):



PVA is sensitive to the boric acid solution. The  $\text{B}(\text{OH})_4^-$  plays the role of crosslinker because it joins 4 hydroxyl ( $\text{OH}^-$ ) ions to the hydroxyl group in the PVA, which results in a condensation reaction [17]. The PVA- $\text{B}(\text{OH})_4^-$  then establishes a network to enfold any object in a capsule-like structure, protecting it from chemicals and humidity, further enhancing the mechanical strength.

The use of condensation of PVA and boric acid has successfully prepared PZT fibers in our previous study [18]. The boric acid condensed with PVA in PZT paste that resulted in the paste viscosity increase or green fiber hardening during the extrusion process. The stability of fabrication is needed to improve to promote the piezoelectric and polarization properties. In this work, we further modified the extrusion method that combined a surface condensation process to prepare and manipulate the flexible PZT fibers that had sufficient green body strength to maintain circular fiber morphology. The extruded fibers without boric acid composition were pulled through a boric acid solution bath to produce a condensation reaction. The various as-extruded PZT fibers were then sintered and formed into 1-3 PZT-epoxy composites. The crystallinity, microstructures, dielectric, piezoelectric and ferroelectric properties of the PZT fibers and 1-3 composites were studied. A surface condensation mechanism was also proposed.

## 2. Experimental procedures

A minimal amount of 85 wt% PZT (PZT/(PZT + PVA solution)) powders (BETACERA, Taiwan) and PVA (10 wt% aqueous solution) were mixed and agitated mechanically. The PZT pastes were formed by milling the as-prepared (PZT + PVA) mixtures in a 3-roller mill over multiple cycles. The PZT fibers were fabricated by extruding the PZT pastes using syringe pushing, controlled by a robot dispenser. The extruded PZT fibers were pulled through a 1–5 M boric acid solution and collected using a mechanical scrollbar, as shown in Fig. 1. The various concentrations of the boric acid solution and PVA molecules on the extruded PZT fibers generated crosslinking, condensation, and dehydration reactions around the fiber surface during the pulling process. The as-extruded and surface-condensed PZT fibers were then dried at an ambient temperature to achieve green body strength. The diameters of the prepared green PZT fibers ranged from 250 to 1800  $\mu\text{m}$  in a circular morphology. The final sintered diameters were approximately 150–1080  $\mu\text{m}$ .

After debinding at 600 °C for 2 h and sintering at 1250 °C in a PbO protection atmosphere for 2 h, the PZT fibers were manually aligned in parallel in a mold. A polymer was then syringed into the mold to form the 1-3 composite. The polymer was composed of epoxy (EPON 828) mixed with a hardening agent (DETA). The obtained composite was deaerated using a centrifugation process. The deaerated composite was subsequently cured at 90 °C for 1 h. The composites were then removed from the mold and were cut perpendicular to the longitudinal direction of the PZT fibers. Silver paste was used as electrodes on the cut ends of the 1-3 composite.

When the piezoelectric materials were first prepared, the crystallized grains and domains were randomly oriented. A poling process was required to establish a dominant domain alignment, and thus, maximize the energy conversion efficiency. The polarization of the PZT bulks, fibers, and 1-3 composites was performed under an electric field intensity 2 kV  $\text{mm}^{-1}$  in a silicone oil bath at 80 °C for 15 min. The poling direction was in the longitudinal direction of the fibers.

The crystallinity and microstructures of the PZT bulks, fibers, and composites were observed at room temperature using X-ray diffraction equipment (D2 PHASERTM) and the FESEM (JSM – 6335F NT). Bulk density was measured using Archimedes' method. The P–E curves, dielectric constant, and piezoelectric coefficient were measured and evaluated. The dielectric constant, resonant frequency and electromechanical coupling coefficient of the fibers, bulk, and composites were measured and calculated using an impedance analyzer (Agilent 4294A) at room temperature. The piezoelectric coefficient was measured using a Berlincourt  $d_{33}$  piezo meter (Channel Products Inc. USA). The P–E curves were measured using a Sawyer-Tower circuit.

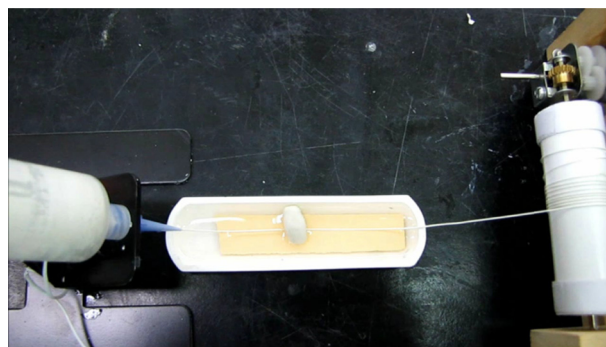


Fig. 1. The photograph of the extruded PZT fiber pulled through a boric acid solution bath and the condensation reaction occurred during the process.

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