3D-printing of a piezocomposite material with high filler content for transducer applications

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Abstract—**This work describes the process of synthesizing and printing a novel (0-3) piezocomposite material using solution mixing technique and digital light processing technique. This piezoelectric composite material was developed using a photopolymer, grey resin and lead magnesium niobate and lead titanate (PMN-PT) with particles size 5 µm. The 3D-printing of this thick film was achieved using the piezocomposite material with high filler content upto 60% w/w of PMN-PT. Results showed a satisfactory print resolution, a thick film with high flexibility and exceptionally high d_{33} coefficient of 49 pm/V during single point scan and an average of 67 pm/V during full surface scan measured using the laser vibrometer technique.**

Keywords—Piezocomposite; Photopolymer; PMN-PT; 3D-Printing; Composite; Digital light processing

I. INTRODUCTION

Fabrication of piezoelectric transducers via 3D- printing offers the possibility of manufacturing low-cost, miniature, highly detailed and highly sensitive devices. Recently, the use of piezocomposite materials in the manufacture of sensors and transducers has been growing [1], in areas such as energy harvesting [2], medical prosthetics, sonar applications [3], damping mechanical vibrations [4] and with recent work describing the process of 3D printing a fully working piezocomposite-based microphone [5].

3D-printing of piezocomposite materials creates the possibility of combining the most useful properties of each constituent material such as the piezoelectric sensitivity of the ceramic and the flexibility of the polymer [6] to achieve the desired design goal when designing transducer applications. The ability to 3D-print piezocomposite materials via additive manufacturing processes depends on combining piezoelectric ceramic nanoparticles and an ultraviolet curable (UV) polymer to form a piezocomposite mixture with the (0-3) connectivity pattern. A piezoelectric composite exhibits the (0-3) pattern where the piezoelectric ceramic particles are dispersed within a polymer matrix [7].

Previously, (0-3) piezocomposite materials consisting of barium titanate ($BaTIO_3$), and Poly (ethylene glycol) diacrylate (PEGDA) [5] was the most commonly used for additive manufacturing due to the high natural piezoelectric response of $BaTIO_3$ and photoactive property of PEGDA. However, such (0-3) piezocomposite material exhibits low piezoelectric response, with d_{33} coefficients two orders of magnitude lesser than the piezoelectric ceramic [5]. Obtaining higher d_{33} coefficients by increasing the concentration of the ceramic nanoparticles trades off the ease of processability of the piezocomposite during printing. A high particle concentration often results in a composite with high viscosity and also significantly increases attentuation of light through the material due to scattering from the particles which inhibits the ability to form intricate geometries and considerably reducing functionality [8].

In this work, we describe the fabrication, 3D-printing and characterization of novel 3D printable piezocomposite material consisting of a photopolymer, grey resin loaded with high concentration of lead magnesium niobate and lead titanate (PMN-PT) using a bottom-up digital light processing process to print a thick film. This procedure of 3D printing the novel piezocomposite has the possibility of being used to manufacture flexible thin/thick films of varying shapes with high piezoelectric properties.

II. MATERIALS AND METHODS

The materials used in this experiment comprise a commercial photopolymer grey resin (formlabs, Somerville, MA), lead magnesium niobate and lead titanate, size 5 µm (American Elements, LOS ANGELES, USA).

A. Material Synthesis and 3D-Printing

The piezocomposite materials were first synthesized by the process of solution mixing before 3D printing the sample. 40% w/w grey resin was mixed with 60% w/w PMN-PT and put in a THINKY AER-250 planetary mixer (Intertronics, Oxfordshire, England) and mixed for 6 min at 1500 rpm and de-foamed for 4 min at 1200 rpm to obtain a homogenous solution and to get rid of all air bubbles within the composite mixture.

To print the thick film, CAD models and slices were prepared using the manufacturer's in-house software (Asiga Composer). The piezocomposite mixture was then placed in the built tray of the 3D printer as shown in Fig. 1. The 3D printed thick film was fabricated using a commercially available 3D printer (Asiga Pico2 HD). This printer uses a 385 nm UV LED, and operates using the bottom-up digital light processing technique. The light source is positioned underneath the build tray containing the piezocomposite, which rests on a tray with a transparent teflon base. The build process begins with the approximate positioning of the build tray 100 µm from the teflon base and over-exposure of the resin to create a "burn-in" layer which ensures that the built

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part sticks to the build block and further provides an anchor for each subsequent build layer. The build progresses by raising the platform by the desired build layer thickness, repopulating the tray underneath the platform with the resin. The next layer is then cured starting from the build tray base, with the gelation point progressing upwards towards the previously printed layer. For the turgid suspension described here, where particle size and spacing between particles are far greater than the wavelength, the wavelength would not be expected to influence the cure depth.

Finally, silver paint was applied as the top electrode and bottom electrode before the polarization process. The printed sample was poled by placing the sample in silicon oil heated to a constant temperature of 120 °C under a constant electric field of 7 kV applied across the surface of the sample for 3 h.

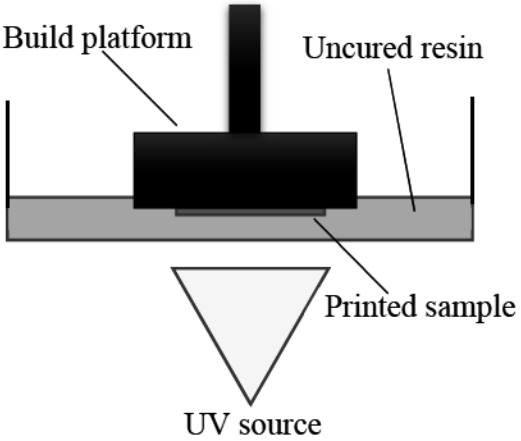

Fig. 1. Shows the process of 3D- printing the novel piezocomposite material using the bottom-up digital light processing.

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

A. 3D- Printing

The 3D-printed thick film sample consisting of 60% w/w PMN-PT and 40% w/w grey resin is shown in Fig. 2. The printed sample was very flexible, smooth and detailed. The thick film had a size 2 cm x 2 cm and a thickness of 0.5 cm. The cure depth for the piezocomposite with 60% w/w loading PMN-PT was estimated to be 13.39 µm, restricting build step sizes to less than 50 µm

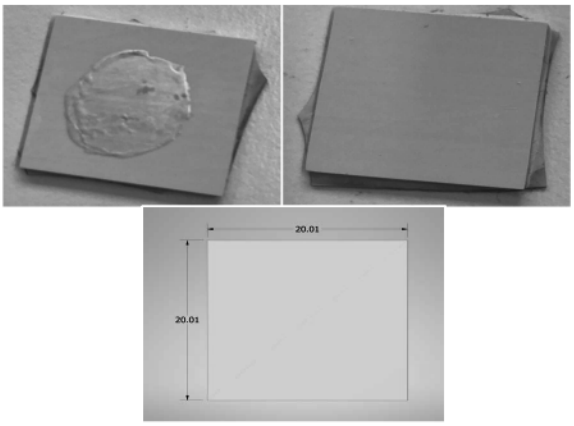

Fig. 2(a). Shows two 3D printed thick films. (b). Shows the CAD file used to print the thick films. The printed samples were flexible, detailed, smooth and fully cured.

B. Piezoelectric properties

The piezoelectric coefficient d_{33} of the 3D printed thick film sample was determined using a laser doppler vibrometer (Polytec MSA100-3D, Waldbronn, Germany). A sinusoidal AC voltage of $10V_{p-p}$ was applied across the axis of charging and the resulting displacement measured. Single point scans and full surface scans were obtained and all measurements were averaged over 20 cycles, using complex averaging. A full surface scan was performed with a total of 2000 scan points to obtain the average d_{33} shown in table 1. The thick film responded at a resonant frequency of 15.201 kHz; however, all the scans were obtained below resonance at 15 kHz. The d_{33} coefficient measured by the laser vibrometer was also compared with that obtained using a quasi–static piezoelectric d_{33}/d_{31} meter (Institute of Acoustics, Chinese Academy of Sciences, Beijing, China). The results obtained are compared in table 1. The measurement obtained by d_{33} meter was low compared to the LDV result, this could be as a result of the bending effect of the thick film sample [9].

Table 1. Comparison of d_{33} coefficient obtained from the LDV and d_{33} meter.

Sample	LDV (pm/V) Single point Scan	LDV (pm/V) Full Surface Scan (Average)	d_{33} meter (pC/N)
3D-printed thick film	49	67	2.1

IV. CONCLUSIONS

The development and 3D printing of a novel (0-3) piezocomposite material consisting of grey resin mixed with Lead Magnesium Niobate Lead (PMN-PT) is presented in this work. The piezocomposite material was created to exhibit a (0-3) structure before the printing process. The thick film was successfully printed using the piezocomposite with PMN-PT loading of 60% w/w. The thick film was flexible, smooth and fully cured. The piezoelectric property of the printed thick film was determined by the 3D LDV experiment. The printed film responded across all frequencies ranging from 1 kHz to 20 kHz during the frequency sweep with its resonant frequency at 15.201 kHz and an average d_{33} of 67 pm/V taken below resonance. Although, different d_{33} values were obtained during single point scans, this seemed to be due to an agglomeration of PMN-PT nanoparticles across different regions within the bulk of the piezocomposite.

The thick and flexible structure of this piezocomposite coupled with the piezoelectric properties and suitable print resolution offers promise as a functional material, which can be used in applications in medical transducer, composite design, and wearable and implantable electronics. Further work is needed to investigate the microstructure, mechanical properties of these piezocomposite and to further enhance their d_{33} value and to 3D print functional applications.

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