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Fabrication of high-aspect-ratio micro piezoelectric array by powder injection molding

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ABSTRACT

A powder injection molding process to fabricate a high-aspect-ratio piezoelectric microarray was developed. The reverse shaped sacrificial mold insert was developed by X-ray micromachining, and then insert-type injection molding was conducted to fabricate the piezoelectric microarray structure. For the micro-powder injection molding, rheological properties of the powder binder mixture were evaluated and analyzed. The measured flow behavior exponent was 0.44 and the flow activation energy was 83 kJ/mol^{-1}. Based on the analyzed rheological property, injection molding conditions were optimized. The rheological property and the injection molding conditions were crucial to ensure complete filling. Using the optimized conditions, two high-aspect-ratio piezoelectric microarrays were fabricated: (i) 20-μm patterns with 1:5 aspect ratio and (ii) 40-μm patterns with 1:10 aspect ratio.

1. Introduction

Piezoelectric ceramic are important functional materials because of their unique electrical property of energy transduction. The ferroelectric and piezoelectric nature of piezoelectric ceramics has led to numerous transducer applications such as actuators, sensors, capacitors, resonators, and high-power transducers [1–5]. However, a single-phase piezoelectric ceramic is not suitable for underwater acoustics and medical imaging applications because of its high acoustic impedance. To match the acoustic impedance, piezoelectric/polymer composite that consists of active piezoelectric ceramic and passive polymer matrix has been developed [6,7]. The piezoelectric/polymer composite, especially the 1-3 connectivity piezoelectric/polymer composite has numerous advantages such as high electromechanical property, low acoustic impedance, and suppression of the parasitic lateral mode [8].

Two methods to fabricate 1-3 piezoelectric/polymer composite have been developed. First method is a top-down approach that carves out the bulk piezoelectric ceramic by using direct mechanical machining, e.g., diamond saw [9], ultrasound [10], laser ablation [11], and deep reactive ion etching [12]. Among these, the dice-and-fill method that uses a dicing saw to carve out the bulk piezoelectric ceramic is the most common method [13]. However, the dice-and-fill method has several drawbacks. The sizes of the rod and gap are limited by the thickness of the saw (normally $>$ 50-μm). The shape of rod is restricted to be square. Thermal and mechanical damage by dicing causes degradation of piezoelectric property. Finally, production is slow and expensive.

To overcome these drawbacks the bottom-up approach which uses the piezoelectric ceramic powder to fabricate the green body has been developed. Casting [14,15], micro-pressing [16], embossing [17,18], injection molding [19–21], and fused deposition method [22] have been investigated to fabricate 1-3 piezoelectric/polymer composite. Among the various near-net-shape manufacturing methods, powder injection molding (PIM) is one of the most promising candidates because of its high production rate and shape complexity [23,24]. Although some previous research has used PIM with piezoelectric ceramic materials [19], the size of the pattern was no less than hundreds of micrometers because of demolding difficulties. Even though there were few research reports about the fabrication of micro-scale feature [25], the taper angle which is not proper for the piezoelectric performances was required to demold the injection molded structure. Alternatively, the lost mold method which uses the reverse shaped sacrificial polymeric mold has been used to replicate high-aspect-ratio ceramic micro-structure without taking the taper angle. Piezoelectric ceramic structures with 50-μm patterns and 1:6 aspect ratios have been produced [26]; this is the current minimum size of implementable patterns of high-aspect-ratio piezoelectric micro-structures produced using this method.

For the injection molding of high-aspect-ratio micro-structure,

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the rheological property of the powder binder mixture is critical. The implementable minimum pattern size can be significantly decreased by using an optimized powder binder mixture. Despite its importance, little research has evaluated or analyzed the rheological property of piezoelectric powder binder mixture for PIM [27]. Another major problem is the technical difficulty of making a precise mold insert. Due to the resolution limit, the conventional mechanical machining technique cannot easily fabricate pattern sizes <50-μm. To fabricate the precision mold insert with the same level of accuracy as the size of the target structure, a new technique is required. An X-ray micromachining method known as LIGA (from the German Lithographi, Galvanoformung, Abformung) process can be used to fabricate micro-scale mold inserts. Synchrotron X-ray micromachining can precisely fabricate high-aspect-ratio micro-structure with excellent sidewall roughness [28–30].

In this research, we demonstrated a PIM process to fabricate high-aspect-ratio piezoelectric microarray. We used X-ray micromachining to fabricate a precise reverse shaped mold insert with micrometer-scale accuracy. The rheological property of powder binder mixture was evaluated and analyzed to find the optimal injection molding conditions to fill the micro-sized cavity completely. Two high-aspect-ratio structures were fabricated: (i) 20-μm patterns with 1:5 aspect ratios, (ii) 40-μm patterns with 1:10 aspect ratios. Experimental methods for the fabrication of micro pattern and the analysis of rheological property for the piezoelectric powder binder mixture were introduced.

2. Experimental procedures

2.1. Materials for powder injection molding process

Synthesized $Pb(Mg, Nb)O_3$–$Pb(Zr, Ti)O_3$ [PMN-PZT] powder was used in this study. Table 1 summarizes characteristics of piezoelectric ceramic powder. The particle-size distribution was measured using a particle-size distribution analyzer (Horiba LA-950V2). The apparent and tap density were measured using a tap density volumeter (Bettersize BT-300). The pycnometer density, used as a reference value for the theoretical density, was measured using an automatic helium pycnometer (Micrometrics AccuPyc 1330). Fig. 1 shows the morphology of the powder observed by the scanning electron microscopy (SEM Philips XL30S FEG). The agglomeration of irregularly-shaped fine particles was observed.

A mixture of paraffin wax (PW), polypropylene (PP), polyethylene (PE) and stearic acid (SA) was used as a binder system. PP and PE were designed for the primary binder which strengthens the primary products and PW was designed for the secondary binder, which enhances the rheological property of the feedstock. SA was used as the surfactant. Characteristics of binders are summarized in Table 2.

2.2. Mold inserts fabrication via X-ray micromachining process

X-ray micromachining was used to fabricate sacrificial mold inserts. An X-ray mask was prepared using UV lithography. The dimensions of the patterns were 25–150 μm. A 20-μm Cr/100-μm Au seed layer was deposited on a 200-μm-thick polyimide sheet. A UV photoresist (SU-8 3010, MicroChem Corp.) was spin-coated on the seed layer. A rod-array-shaped structure with 20-μm-thick photoresist was fabricated using UV photolithography, then a gold layer to block the X-ray was electroplated up to 17 μm thick. Using the obtained X-ray gold mask, the positive photoresist, poly(methyl methacrylate) (PMMA; Good fellows Corp.) was selectively exposed to synchrotron X-rays. X-ray energy was adjusted to 4 kJ/cm^3 which is known as an optimal dose to ensure the clear removal of exposed PMMA. After X-ray exposure, the exposed PMMA mold inserts were developed using a GG developer (a chemical mixture of 60 vol% 2-(2-butoxyethoxy) ethanol, 20 vol% tetrahydro-1, 4-oxazine, 5 vol% 2-aminoethanol, and 15 vol% deionized water) for 48 h. After each step, the uniformity of fabricated pattern was analyzed using an optical microscope.

Table 1
Characteristics of PMN-PZT ceramic powder.

Particle size (μm)			Density (10^3 kg/m^3)		
D_{10}	D_{50}	D_{90}	Apparent	Tap	Pycnometer
0.31	0.52	1.01	1.62 (20%)	2.54 (32%)	7.98

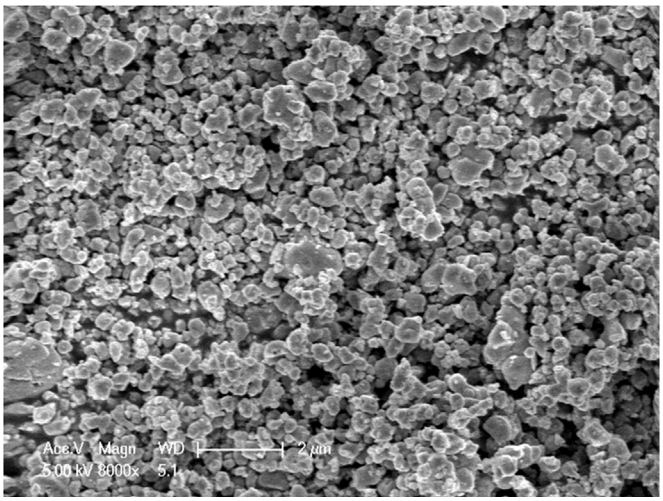

Fig. 1. Morphology of PMN-PZT powder.

2.3. Micro structure fabrication via powder injection molding process

The entire PIM process including mixing, injection molding, debinding and sintering were conducted to fabricate the high-aspect-ratio micro piezoelectric arrays. Powders were mixed using a twin-screw extruder mixer with at mixing temperature of 160 °C and solid loading of 45 vol%. The appropriate solids loading were determined based on a previous torque rheometer experiment [31]. The rheological property of powder binder mixture was evaluated using a strain-controlled plate type rheometer (MCR101 Anton Paar). As changing the strain rates and temperatures, shear viscosity was measured. Injection molding process was carried out using a molding machine (TR 30EH, Sodick Plustech). The micromachined mold inserts were attached to the mold base, then PMN-PZT feedstock was injected. Molding conditions were optimized while observing the incomplete-filling defects using an optical microscope. Optimized injection conditions were determined as injection pressure of 60 MPa, injection temperature of 160 °C and mold temperature of 65 °C. To remove the PMMA mold inserts, the green body was immersed in acetone at 35 °C for 4 h. After removing the mold inserts, two step debinding process were conducted for the binder removal. Solvent debinding was carried out in a bath of N-hexane at 45 °C for 12 h and the thermal debinding was conducted in argon (Ar) atmosphere as setting the holding temperature at 250 and 400 °C [31]. Sintering process was conducted in a close crucible at 1250 °C for 2 h in air atmosphere [32]. Dimensions of fabricated high-aspect-ratio micro-arrays were analyzed using SEM.