

## Short Review of 3D Printed Piezoelectric Sensors

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### Abstract

Recently, 3D printing technology has gained increased attention in the manufacturing industry because it allows the manufacturing of complex but sophisticated structures as well as moderate production speed. Owing to advantages of 3D printers, such as flexible design, customization, rapid prototyping, and ease of access, can also be advantageous to sensor developments, 3D printing demands have increased in various active device fields, including sensor manufacturing. In particular, 3D printing technology is of significant interest in tactile sensor development where piezoelectric materials are typically embedded to acquire voltage signals from external stimuli. In regard with piezoelectricity, researchers have worked with various piezoelectric materials to achieve high piezoelectric response, but the structural approach is limited because ceramics have been regarded as challenging materials for complex design owing to their limited manufacturing methods. If appropriate piezoelectric materials and approaches to design are used, sensors can be fabricated with the improved piezoelectric response and high sensitivity that cannot be found in common bulk materials. In this study, various 3D printing technologies, material combinations, and applications of various piezoelectric sensors using the 3D printing method are reviewed.

**Keywords:** Piezoelectricity, Tactile sensor, 3D printing, 3D printed sensors

### 1. INTRODUCTION

With the advent of 3D printing technology, diverse, complex, and sophisticated design products can be realized at moderate production speeds and costs compared with conventional manufacturing processes. Owing to these advantages, 3D printing technology has been widely investigated in various fields, including nanogenerators and sensors [1-4]. In particular, nanogenerators are typically powered by piezoelectricity, a triboelectric, or a hybrid method. Self-powered sensors adopt these nanogenerators as a power source or the sensing signal itself can be acquired by the above-mentioned mechanisms [5-7]. To realize sensors based on piezoelectricity with high output and efficiency, more research on the development of materials and structures is needed. Piezoelectricity is the ability of the piezoelectric materials to generate an electric charge in response

to the applied mechanical or electrical stresses. Piezoelectric ceramics, such as lead zirconate titanate (PZT), barium titanate (BTO), and potassium sodium niobate, are widely used as sensors and actuators because of their high piezoelectricity and relatively low manufacturing costs. However, these piezoelectric ceramics are considered inappropriate materials for flexible devices because of their brittleness and low ductility [8-10].

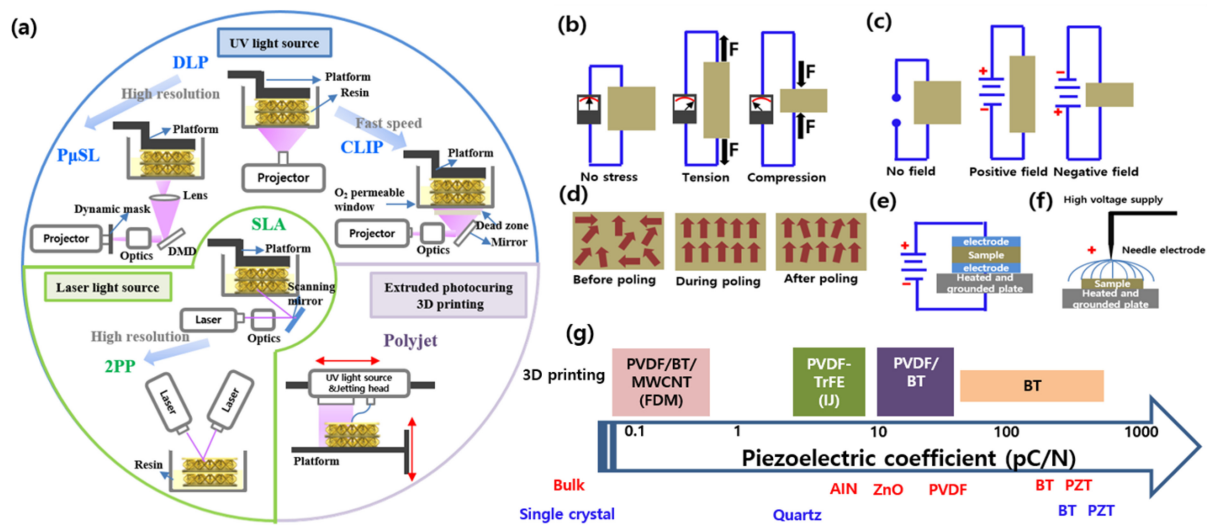
Manufacturing flexible piezoelectric objects using ceramics in conventional methods is challenging. Although 3D printers were introduced decades ago, commercially available 3D printing equipment was initially focused on polymers; therefore, the addition of ceramics to 3D printers seemed implausible. Owing to the increasing demands for complex but sophisticated objects with a small volume, 3D printing technology has been actively investigated in the last decade, and this advanced technology can now be applied to ceramics [11-14]. Consequently, 3D printing technology mixing ceramic and polymer continues to be developed owing to the demand for efficient implementation of complex structures, mass production, and various field applications. In particular, a typical vat photopolymerization (VP) stands out as a ceramic 3D printing method. The 3D printing of piezoelectric material using the VP method improves sensitivity and piezoelectric properties owing to its structural diversity; therefore, VP method-based 3D printing using different material options were suggested for various sensor applications [15-17].

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**Fig. 1.** (a) Schematic of vat photopolymerization 3D printing methods. Schematics of the piezoelectricity principle for a (b) direct and (c) inverse piezoelectric effect. Schematics of the (d) poling process, (e) contact poling, and (f) corona poling. (g) Comparison of 3D printed materials piezoelectric properties and conventional piezoelectric materials.

For 3D printed sensors, various parameters, such as force, displacement, pressure, and strain, must be considered [15-19]. Advantages, such as higher productivity and precision, can result from 3D printing of strain, pressure, tactile, wearable, and hybrid sensors [20]. Therefore, we reviewed past studies of 3D printing technologies, particularly those on piezoelectric materials and sensors using VP. Finally, the future of piezoelectric-based ceramic 3D printing for sensor studies are discussed.

## 2. 3D PRINTING VIA PHOTOPOLYMERIZATION

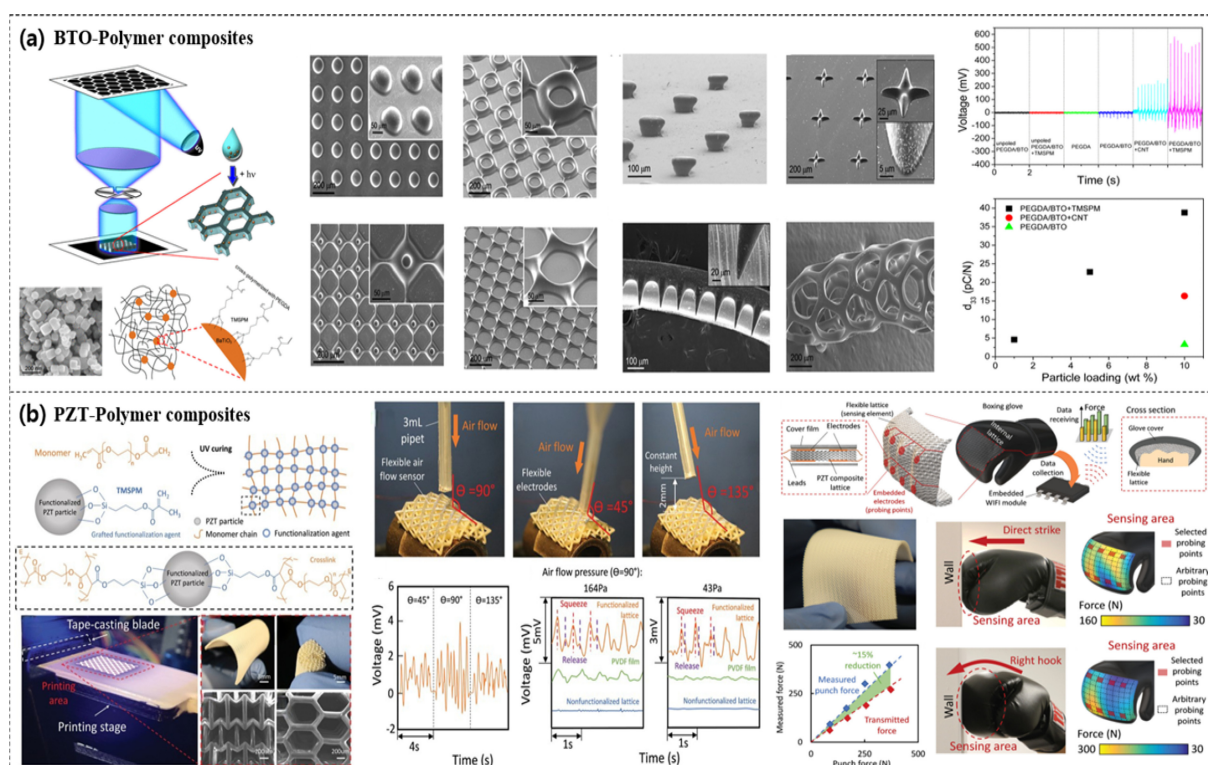
Note that 3D printing is also called additive manufacturing (AM) because it transforms 3D models into objects by stacking materials, such as polymers, ceramics, and metals, layerwise. The VP method is a 3D printing technology that forms a shape by placing a liquid photopolymer resin in a container and selectively irradiating light to cure and laminate it [21-23]. Fig. 1 (a) shows various photopolymerization-based 3D printing technologies [24].

Stereolithography (SLA) is one of the most widely used VP technologies, which uses an ultraviolet (UV) laser beam to selectively cure a polymer resin. Because SLA cures resin in units of a laser spot, the resolution of the product is relatively good, but low process speed is a trade-off. Two-photon polymerization uses a femtosecond ( $10^{-15}$  second) pulsed laser to selectively cure photopolymers near the focal point of lasers; thus, it has 250 times higher resolution (200 nm). In contrast, digital light processing

(DLP) projects a light beam instead of a laser to cure resin, layerwise. As the resin is cured in a unit area, the DLP is a fast 3D printing method. Owing to the relatively poor resolution of DLP compared with SLA, projection micro-stereolithography (P $\mu$ SL) is suggested as a promising approach with a high resolution (up to 0.6  $\mu$ m) and fast production capability. Continuous liquid interface production is known as a rapid printing method because it does not require intermittent stationary steps for resin curing. A PolyJet 3D printer is similar to a typical inkjet printer, but a liquid photopolymer is cured onto the build tray instead of jetting ink droplets onto paper. PolyJet is a precise and fast method capable of 3D printing a multi-material and multi-color with a small batch just by adding multiple print heads. To date, various 3D printing technologies based on VP have been introduced. Details of VP 3D printing methods are described in reference [24].

## 3. 3D PRINTABLE PIEZOELECTRIC MATERIALS

The aforementioned 3D printing methods are feasible candidates to realize ceramic objects. In particular, if piezoelectric materials are used, active devices, such as sensors and actuators, can be created by a 3D printer. Piezoelectric materials can transform strain caused by applied mechanical (direct piezoelectric effect) or electrical stresses (inverse piezoelectric effect) into electrical energy, as shown in Figs. 1 (b) and (c) [25].



**Fig. 2.** (a) Functionalized BTO-polymer composites, 3D printed piezoelectric microstructures and their piezoelectric voltage. Reprinted with permission from Ref. [27]. Copyright (2014) American Chemical Society. (b) Functionalized PZT-polymer composites and piezoelectric properties for a pressure sensor. Reprinted with permission from Ref. [2]. Copyright (2019) John Wiley & Sons

As shown in Fig. 1 (d), the piezoelectric activity of these piezoelectric materials can be obtained by the poling process, wherein a strong electric field is applied across piezoelectric materials such that randomly oriented dipole domains are aligned. The poling is divided into contact and noncontact (corona) processes. In contact poling, the sample is sandwiched between bias and grounded plate, and electrodes on the sample are in contact (see Fig. 1 (e)). On the other hand, in corona poling, the material is placed on a grounded surface and a strong electrical field is applied to the material through a needle, which is not in physical contact with the material (see Fig. 1 (f)) [26].

The corona poling method has been used for both polymer composites with a large area and thin films to minimize the chance of dielectric breakdown of the samples. The properties of the polarized samples can be confirmed by monitoring piezoelectric coefficients. Fig. 1 (g) shows the piezoelectric coefficients obtained via 3D printing (top) and conventional techniques (bottom) [25]. Compared with conventional bulk materials on the lower side of the arrow in Fig. 1 (g), 3D printed materials have relatively low piezoelectric properties owing to mixed composition materials. In addition, the 3D printing process requires extra optimization in every step, such as the preparation

of material, conditioning exposure, and post-processing.

One of the most important factors in 3D printed piezoelectric materials is selecting an appropriate piezoelectric powder with high piezoelectric response, which can be stably dispersed in the liquid photocurable resin. For this reason, to achieve high piezoelectricity, a wide range of materials have been reviewed by functionalizing the surface of piezoelectric particles and optimizing the content as well as using various materials, such as polymer, ceramic powder-polymer, nanoparticle-polymer composites, and multi-materials [2, 27-30].

### 3.1 Polymer

In addition to piezoelectric ceramics, polyvinylidene fluoride (PVDF) is a promising candidate for a piezoelectric component in the 3D printer because  $\beta$  phase PVDF is flexible and has moderate piezoelectric properties [31]. For example, in a previous study when PVDF powder was dissolved in a solvent and subsequently combined with liquid photopolymer, PVDF/PR solvent with a ratio of 1:10 (PVDF:DMF) and 2 wt% PVDF/PR had the highest piezoelectric properties with  $\pm 0.053$  nA and 0.014 pC/N [32]. Although PVDF-included 3D printed composites can be realized,

**Table 1.** Summary of reported sensors using VP 3D printing

Material	Method	Performance	Application	Refs.
TMSPM/ PZT/Resin	P $\mu$ SL	Piezoelectric charge constant ( $d_{33} \approx 42 \text{ pC N}^{-1}$ )	Piezoelectric flexible air flow sensor, Self-sensing boxing glove	[2]
TMSPM/BNNT/Resin	P $\mu$ SL	Sensitivity: 24 mV kPa $^{-1}$	Piezoelectric strain sensor	[30]
PZT/ MWCNT electrode	MM-P $\mu$ SL	Capacitance change ratio ( $\Delta C/C_0$ ): -0.04-0.06	Tactile sensor	[31]
PPTMGA-40/ionic hydrogel	DLP	The tensile strength: 15.7 MPa Elongation at break: 414.3%	Piezoresistive strain sensor Electric switch, Wearable device	[33]
PUA/PEDOT:PSS	DLP	Sensitivity: $1.666 \times 10^{-4} \text{ S N}^{-1} \text{ m}^{-1}$	Piezoresistive pressure sensor	[16]
HDDA/PDMS/carbon grease	DLP	Tensile strength: 17.4 kPa	Piezoresistive strain sensor, Self-healable actuator	[34]
MWCNT/Resin	DLP	Elongation: 100 %	Capacitive pressure sensor	[35]
PAAm-PEGDA/MgCl $_2$ /VHB tape	DLP	Sensitivity: 0.84 kPa $^{-1}$	Capacitive pressure, Strain sensor, Wearable devices	[36]

their inadequate piezoelectric properties warrant future ceramic composite research.

### 3.2 Piezoelectric ceramic-polymer composites

Owing to excellent mechanical flexibility and piezoelectric response, research on piezoelectric ceramic-polymer composites using PZT and BTO has been conducted [2, 27, 30]. To enhance the mechanical-to-electrical conversion efficiency, ceramic powders are being functionalized.

Fig. 2 (a) shows the BTO nanoparticles chemically modified with acrylate surface groups, which form covalent linkages with the polymer matrix under light exposure [27]. The modified 10 wt% BTO composites using a DLP 3D printer exhibited piezoelectric coefficients ( $d_{33}$ ) of approximately 40 pC/N, which is over 10 times larger than those of the composites with unmodified BTO nanoparticles. Fig. 2 (b) shows functionalized PZT nanoparticles in the UV-sensitive monomer matrix using P $\mu$ SL [2]. The voltage output of functionalized PZT powder loading over 74 wt% exhibited the highest  $d_{33}$  (from 27 pC/N to 110 pC/N), as compared to unfunctionalized PZT nanoparticles. These results show that appropriate functionalization of ceramic powders is critical to achieve high piezoelectric properties.

### 3.3 Nanoparticle-polymer composites

To overcome the high stiffness of inorganic piezoelectric ceramics, piezoelectric nanocomposites using nanofillers have been suggested. For example, a functionalized boron nitride nanotubes (BNNTs) polymer composite was produced by the 3D

printer, and this 0.2 wt% BNNTs polymer composite exhibits a sensitivity of 120 mV/(kPa·wt%) over a broad press region (1–400 kPa) [29]. This is attributed to the effective stress transfer between BNNTs and polymers. Note that a small fraction of piezoelectric composites in a resin is capable of functioning piezoelectricity.

### 3.4 Multi-materials

Recently, the 3D printing of multi-materials with heterogeneous properties has gained attention because it allows manufacturing products with various properties that respond to external stimuli, such as temperature, humidity, or structures, with rigid and flexible elements.

One notable research on multi-materials 3D print was reported by Professor Xiaoyu Zheng's research group at UCLA [30]. The multiple functional materials were volumetrically printed in a single step, thereby resulting in a complex 3D structure to integrate sensor use. These active multi-materials include polymer, ceramic, semiconducting, and magnetic materials. This approach is critical for enabling multifunctional sensors bypassing the need for multiple sensor integrations.

## 4. SENSORS USING 3D STRUCTURE

The VP-based 3D printing technology has been applied to develop sensors and they are summarized in Table 1. The main applications of 3D printed sensors are to monitor strain, pressure, or tactile sensing.

#### 4.1 Strain Sensor

A strain sensor array on fingers using 0.2 wt% functionalized BNNTs polymer composite has been reported to improve interface stress transfer and piezoelectric output [29]. A structure was designed to boost the piezoelectric response with a maximum sensitivity of 24 mV/kPa, which is 10 times higher than that of a flat BNNTs-based sample. Moreover, printed piezoelectric materials were used for the strain sensor array on the fingers by transferring the postures of the hand to electric signals using 14 piezoelectric strain sensors. When the hand gripped a ball, different output signals from bent fingers were recorded because it represents the locations where strain sensors deformed. These displayed results argue that the 3D printed conformal devices can measure force distribution on the contact area and different motions of the robotic hand.

#### 4.2 Pressure Sensor

Fig. 2 (b) displays piezoelectric sensors based on functionalized PZT powder polymer composite with structural flexibility as well as high sensitivity and detection. The functionalized 74 wt% PZT powder composites approach the upper bound of the piezoelectric constant  $d_{33}$  [2]. These microarchitectures are inserted into a boxing glove to demonstrate pressure sensing and spatial mapping capabilities.

#### 4.3 Tactile sensor

A tactile sensor using 3D structured multi-materials is demonstrated in Ref. [30]. The composite materials with different damping coefficients detect elastic wave propagation within the architectures using volumetrically deposited electrodes to demonstrate internal stress sensing. The results demonstrate that 3D printing can volumetrically deposit multiple functional materials into complex architectures based on localized electrostatic attraction and is capable of a 3D-printed sensor with tactile, impact, and shape-sensing features.

### 5. CONCLUSION

Thus far, piezoelectric materials for 3D printed sensors have been studied using various approaches, primarily by mixing the piezoelectric powder with polymer. Owing to their 3D structure, various 3D printed sensors can sense piezoelectric response better than conventional bulk piezoelectric sensors.

In particular, selections of ceramic powders with high piezoelectricity, incorporations between functionalized inorganic and organic materials, and an optimized concentration of ceramic powder are feasible approaches to fabricate sensors with an improved piezoelectric response and high sensitivity. For example, multi-material 3D printing with diverse colors or materials can be realized by combining different properties of materials. Consequently, these printed objects can be adopted as sensors for tactile, impact and shape-sensing features via integrated manufacturing processes. However, improvement of piezoelectric properties of 3D printed objects is necessary to effectively utilize 3D printing technology for sensors. If both materials and structural design studies are carefully investigated in parallel, 3D printing technology is expected to achieve improved piezoelectric properties and flexibility, which has not been achieved by conventional methods; therefore, it can further be adopted not only by sensors but also by actuators in the near future.

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