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14. ABSTRACT
This study involves the exploration of 3D printing dielectric elastomer actuators (DEAs) as a fundamental basis for electrical actuation and control. Soft robotics is an emerging field in which active biological systems can be mechanically approximated by active “smart” materials, enabling a synergistic integration between sophisticated external controls and bio-inspired actuation. Soft robots are designed to be entirely deformable, closely mimic biological motion, provide insight into biomechanical systems, and enable modes of actuation not feasible in traditional “hard” robotic systems. This renders soft robots as prime candidates for additive manufacturing using an

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Report Title

Final Report: STIR: Toward Programmable Dielectric Elastomers for Actuation and Control

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This study involves the exploration of 3D printing dielectric elastomer actuators (DEAs) as a fundamental basis for electrical actuation and control. Soft robotics is an emerging field in which active biological systems can be mechanically approximated by active “smart” materials, enabling a synergistic integration between sophisticated external controls and bio-inspired actuation. Soft robots are designed to be entirely deformable, closely mimic biological motion, provide insight into biomechanical systems, and enable modes of actuation not feasible in traditional “hard” robotic systems. This renders soft robots as prime candidates for additive manufacturing using an extrusion-based 3D printer. At the fundamental materials level, a micron-scale resolution, multi-material 3D printer is capable of sandwiching polymers, hydrogels, and metallic nanoparticles with a complexity and density that cannot be accomplished with conventional two-dimensional techniques. By combining this approach with recent advances in the development of dielectric elastomer actuators, we will investigate 3D programmable dielectric elastomers for actuation and control. Specifically, here we focus on replacing the common electrode materials used for DEAs with ionic hydrogels, and 3D printing layered composites of conductive hydrogel/dielectric elastomer/conductive hydrogel to fabricate fully 3D printed soft actuators that generate bending motions due to electrical stimuli.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

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TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

Number of Presentations: 0.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

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Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

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TOTAL:

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Books

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Received	Book Chapter

TOTAL:

Patents Submitted

Patents Awarded

Awards

- Conquer Paralysis Now Challenge Grant Winner (2016)
- Moore Foundation Inventor Fellows Finalist (2016)
- Extreme Mechanics Letters (EML) Young Investigator Award (2015)

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
Ghazaleh Haghiashtiani	1.00	
Ryan Han	0.10	
FTE Equivalent:	**1.10**	
Total Number:	**2**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
Michael McAlpine	0.00	
FTE Equivalent:	**0.00**	
Total Number:	**1**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

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Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Total Number:

Names of other research staff

NAME PERCENT_SUPPORTED

FTE Equivalent:
Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

See attachment

Technology Transfer

- Ed Habtour, Prognostics and Diagnostics Team Lead, U.S. Army Research Laboratory; Frank Gardea, Postdoctoral Fellow, U.S. Army Research Laboratory (Collaboration in terms of transfer of material samples for characterization).
- Dr. Jack Norfleet, Medical Simulations and VR at the Army Research Laboratory, who funds Dr. Rob Sweet's research group at UMN and UWash. They are interested in developing highly lifelike organ models for surgical practice in the field. Our 3D printed soft actuators may be incorporated into these organ models which we are developing to generate lifelike, moving organ models for more accurate surgical practice.

Final Report - Grant # W911NF-15-1-0469
(Reporting Period: October 2015 – August 2016)

STIR: Toward Programmable Dielectric Elastomers for Actuation and Control

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Objective

This study involves the exploration of 3D printing dielectric elastomer actuators (DEAs) as a fundamental basis for electrical actuation and control. Soft robotics is an emerging field in which active biological systems can be mechanically approximated by active "smart" materials, enabling a synergistic integration between sophisticated external controls and bio-inspired actuation. Soft robots are designed to be entirely deformable, able to slip into small spaces, run for long periods of time, closely mimic biological motion, provide insight into biomechanical systems, and enable modes of actuation not feasible in traditional "hard" robotic systems. Biological analogies of soft robots – such as cephalopods – are powerful and efficient due to their inherently three-dimensional actuation mechanisms. This renders soft robots as prime candidates for additive manufacturing using an extrusion-based 3D printer. At the fundamental materials level, a micron-scale resolution, multi-material 3D printer is capable of sandwiching polymers, hydrogels, and metallic nanoparticles with a complexity and density that cannot be accomplished with conventional two-dimensional techniques. By combining this approach with recent advances in the development of dielectric elastomer actuators, we will investigate 3D programmable dielectric elastomers for actuation and control. Specifically, here we focus on replacing the common electrode materials used for DEAs with ionic hydrogels, and 3D printing layered composites of conductive hydrogel/dielectric elastomer/conductive hydrogel to fabricate fully 3D printed soft actuators that generate bending motions due to electrical stimuli.

Approach

- We begin with the selection of materials for compliant electrodes of the actuator. This involves optimizing the chemical composition of the ionic hydrogels, selected as the electrode material, both for printability and performance.
- We investigated different options for the electroactive dielectric layer and the passive layer of the actuator and selected the appropriate materials.
- Mechanical characterization of selected materials was performed with the purpose of characterizing material properties and assuring the compliancy of ionic hydrogel electrodes to the dielectric layer, and the higher elastic modulus of the passive layer.
- After selection and optimization of individual materials for printability, different surface engineering strategies were investigated for efficient printing of the hydrophilic hydrogel electrodes on the hydrophobic silicone layers, to overcome these material incompatibilities.
- Next, full devices were fabricated via comprehensive, multi-material 3D printing.
- Finally, the 3D printed actuators were subjected to high electric fields to study their actuation performance and verify the feasibility of the concept.

Relevance to Army

This work represents a first-of-its-kind study toward the freeform fabrication of tether-less biochemical-based mechanical devices and soft robots that can serve a multitude of army applications. First, soft robots can be developed to generate electrically-driven mobility with a simple structural design and control, and can be harnessed for mechanical camouflage and slipping into small spaces in the field. Second, like other dielectric elastomer transducers, the developed device can be used in sensor mode, thus enabling programmable sensory-motor systems with integrated feedback. Third, the sensing capabilities of the hybrid hydrogel/elastomer composites can be broadened to be applied for non-destructive evaluation and structural health monitoring via 3D printing of conformal and geometrically complex devices. Ultimately, by manipulating the concept in different modes of actuation and control, the soft DEAs can find applications in developing dynamic mechanical skins, surface textures, smart prosthetics, and soft robotics.

Accomplishments for Reporting Period

a) Materials Selection and Optimization

In this study, we primarily chose a unimorph configuration for the DEA to explore a bending mode of actuation. The unimorph configuration comprises an electroactive dielectric elastomer, with electrodes on both sides, attached to a stiffer passive layer. Upon application of high electric fields, the in-plane expansion of the dielectric layer will be translated to an out-of-plane bending motion due to the restriction imposed by the passive layer[1] (Figure 1). Thus, three different materials should be selected for the electrodes, dielectric elastomer, and passive layer of the DEA, respectively. In addition to the mechanical and electrical properties required in the selection of each of these materials, additional rheological and other specifications should be taken into consideration in order to enable the incorporation of these materials in the 3D printing process. For instance, a common scheme for selection of these materials is that they should exhibit shear-thinning behavior. In other words, the selected material should have lower viscosity at the imposed shear rates by the printing process in order to flow through the nozzle, but maintain their shape once they have been deposited onto the substrate. In addition, they should have a fast curing rate and preferably, require no post processing in an effort to keep the fabrication on the printer platform, thus simplifying the process. For this purpose, UV curable materials seem to be good candidates, since a UV source can easily be incorporated into a 3D printing unit. Besides these general considerations, the selection process for each of the individual materials is detailed below.

The electrodes should remain conductive at high deformations and compliant to the dielectric elastomer without adding significant stiffness to the actuator.[2] They also need to sustain a high number of stretching cycles and remain conductive after large deformations. Much of the published DEAs have used hand-painted carbon grease or carbon films[3,4] that have poor reliability and are not available in printable forms. Nanomaterials, such as carbon nanotubes[5], graphene, and silver nanoparticles and nanowires, have improved performance in comparison to conventional electrodes, but they are difficult to integrate into advanced actuator designs[6]. The invention of a highly stretchable, transparent ionic conductor based on polyacrylamide hydrogels has enabled the fabrication of DEAs that can sustain voltages above 10 kV, and hence produce significantly larger strains[7]. Recently, researches have proposed the integration of higher molecular weight polymers

into the composition of such ionic hydrogels and other materials including silicones, to modify their composition and rheological properties, thus rendering them suitable for use in the 3D printing process.[8]

By incorporating this strategy, we formulated the composition of the UV curable, polyacrylamide-based ionic hydrogel (Table 1). Lithium Chloride (LiCl) was used to induce ionic conductivity in the hydrogel composition. In addition, the hygroscopic properties of this salt improves the water retention of the hydrogels, thus preventing their drying out which would inhibit their performance as electrodes in the DEA.[9] The 3D printing of this hydrogel was optimized based on the nozzle tip diameter, extrusion pressure, and printing speed. A UV cure system was utilized during the printing process to ensure the curing of each layer prior to the deposition of subsequent layers (Figure 2a). It was observed that the 3D printed hydrogels maintain their transparency and stretchability (Figure 2b and 2c).

For the dielectric layer, we selected silicone elastomer, which is one of the materials of choice in developing DEAs due to its fast electromechanical response, low mechanical loss, and availability in a wide range of viscosities, curing rates, and mechanical properties. We tested different commercially available silicones including TC 5005 A/B-C (BJB Enterprises), Semicosil 912 (Wacker), Silopren UV Electro 225-1 (Momentive), and a modified silicone composition developed in the work discussed earlier, which involved the addition of a second UV curable, high molecular weight silicone adhesive sealant (Loctite 5039 Nuvasil) to the Semicosil 912 silicone elastomer at a 3:2 weight ratio[8]. We employed the latter as the dielectric elastomer layer of our DEA. Finally, we chose the UV curable, one-part silicone sealant Loctite 5084 Nuvasil as the passive layer due to its higher stiffness compared to the dielectric elastomer and its non-corrosiveness. The optimal conditions for printing these two materials were determined. It was also observed that exposing the printed materials to UV light only after the complete deposition of each individual layer results in a finer surface integrity for the dielectric and passive layer. Thus, this method was applied to ensure the uniformity of these layers to avoid the dielectric breakdown and failure of the device.

Furthermore, mechanical characterization of 3D printed samples of the three materials were performed to ensure that the hydrogel electrodes are compliant to the dielectric layer, and the passive layer has a higher stiffness comparing to the dielectric layer to yield out-of-plane motion. The elastic modulus of the passive layer, silicone dielectric elastomer, and ionic hydrogel electrodes were estimated as 420 kPa, 84 kPa, and 8.95 kPa, respectively (Figure 3).

b) Overcoming Material Incompatibilities and 3D Printing the DEA

A major challenge in 3D printing the DEA device is the deposition of the hydrogel electrodes on the silicone dielectric layer due to the highly hydrophobic nature of silicone vs. the hydrophilic properties of the hydrogel. This leads to the weak interactions between the droplet and the surface that causes surface dewetting. Dewetting causes significant loss of resolution as adjacent droplets coalesce into bigger droplets. There exist several promising surface engineering strategies to minimize these material incompatibilities. For instance, plasma treatment is a common method used specifically in microfluidics to decrease the hydrophobicity of silicones. However, the treated surfaces are not stable and often suffer from hydrophobic recovery.[10] In addition, this process

necessitates cleanroom facilities and the removal of printed structures from the printer platform, which is not favorable to our purpose of devising fully 3D printed structures. Other methods of modifying silicone surfaces involve chemical processing such as the use of surfactants.

We observed that after the application of diluted solutions of surfactant (e.g., Triton X-100) to silicone and evaporation of the solvent, the wetting of uncured hydrogel inks was improved. However, this method did not significantly enhance the adhesion of these materials, as the printed hydrogel layers separated from the underlying silicone layer after curing. We assume that the presence of UV curing could potentially affect the adhesion and bonding of material layers. Thus, we employed a different surface engineering strategy that takes advantage of the UV element present in the fabrication process. This method involves the application of benzophenone (BP) photoinitiator, which has been used to activate elastomer surfaces in UV assisted polymer grafting and is shown to improve the interfacial bonding of elastomer/hydrogel hybrids.[11] Upon treatment of the elastomer surface with BP and exposure to UV light, the BP molecule generates radical sites that can abstract a hydrogen atom from the elastomer. This then facilitates the reaction of the hydrogel monomer, acrylamide in this case, with the generated radicals, thus leading to graft polymerization and chemical bonding of the hydrogel to elastomer.[11-14] For this purpose, we applied aliquots of 10 wt.% Benzophenone solution in acetone onto the 3D printed silicone structures. After the absorption of BP, the surface of silicone was dried using nitrogen gas and the subsequent hydrogel layers were printed while the whole structure was exposed to UV light. This method resulted in depositing hydrogel films with uniform surfaces on the silicone layer with an improved interfacial bonding (Figure 4a).

Finally, we designed and 3D printed the full device (Figure 4b). We performed an iterative process of device fabrication and testing to find a starting point for the number of layers of each material (i.e., the thickness of each layer) that would lead to actuation. Specifically, for the dielectric layer, there is a tradeoff between the layer thickness and the resulting actuation rate, since the lower dielectric thickness yields a higher actuation, but increases the chance of dielectric breakdown. Our recent 3D printed actuator has dimensions of ~30 × 10 × 1.15 mm with a dielectric layer thickness of approximately 360 μm and one layer of hydrogel deposited as each electrode.

c) Preliminary Actuation

To investigate the actuation performance of the 3D printed DEA, one end of the device was anchored to a glass slide while the other end was left unconstrained (Figure 5a). After applying high voltage to the DEA, the out-of-plane motion of the device was observed. Quantitative data on the actuator tip displacement was obtained by recording the actuation and analyzing the recorded video file using Tracker software which provides a video analysis and modeling tool. As can be seen in Figure 5b and as expected, the DEA exhibits higher displacement with increasing applied voltage. Currently, a ~2 mm displacement at 4 kV applied voltage has been accomplished for the developed devices (Figure 5b and 5c). At 4.1 kV of applied voltage, electrical arcing occurred which constrained the maximum voltage applied to the device, and eventually, led to dielectric breakdown and device failure. The observed actuation shows promise of incorporating hydrogels as electrodes and 3D printing of soft actuators. Current efforts are focused on improving the performance of the devices to achieve higher actuation rates, thus broadening its range of applications in various fields including, but not limited to soft robotics.

Collaborations and Technology Transfer

- Ed Habtour, Prognostics and Diagnostics Team Lead, U.S. Army Research Laboratory; Frank Gardea, Postdoctoral Fellow, U.S. Army Research Laboratory (Collaboration in terms of transfer of material samples for characterization).
- Dr. Jack Norfleet, Medical Simulations and VR at the Army Research Laboratory, who funds Dr. Rob Sweet's research group at UMN and UWash. They are interested in developing highly lifelike organ models for surgical practice in the field. Our 3D printed soft actuators may be incorporated into these organ models which we are developing to generate lifelike, moving organ models for more accurate surgical practice.

Resulting Journal Publications During Reporting Period

- In progress (coming soon)

Graduate Students Involved During Reporting Period

- Ghazaleh Haghiashtiani, PhD student in mechanical engineering, currently working on this project.
- Ryan Han, master's student in mechanical engineering, occasionally assisted Ghazaleh in the 3D printing process and design for this project.

Awards, Honors and Appointments

- Conquer Paralysis Now Challenge Grant Winner (2016)
- Moore Foundation Inventor Fellows Finalist (2016)
- Extreme Mechanics Letters (EML) Young Investigator Award (2015)

Additional Information

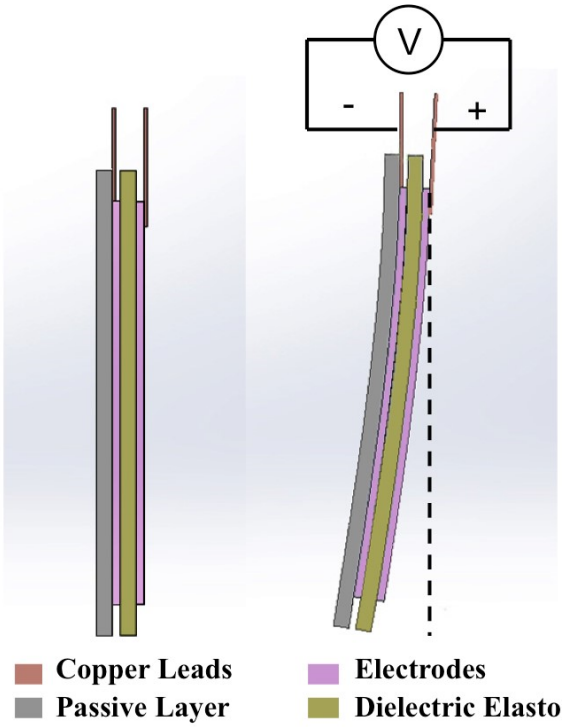

Figure 1. Schematic of the layered structure for a dielectric elastomer unimorph actuator.

Role	Component	wt. %
Monomer	Acrylamide (AAm)	7.90%
Crosslinking Agent	MBAA	0.13%
Photoinitiator	Irgacure 1173	0.08%
Ion Induction	Lithium Chloride (LiCl)	21.48%
Thickening Agent, Rheology modifier	Polyacrylamide (PAM, Mol. Wt.=5,000,000)	3.16%
Solvent, Rheology Modifier	Ethylene Glycol	37.60%
Solvent	Ultra-pure H_2O	29.64%

Table 1. Chemical composition of the ionic hydrogel.

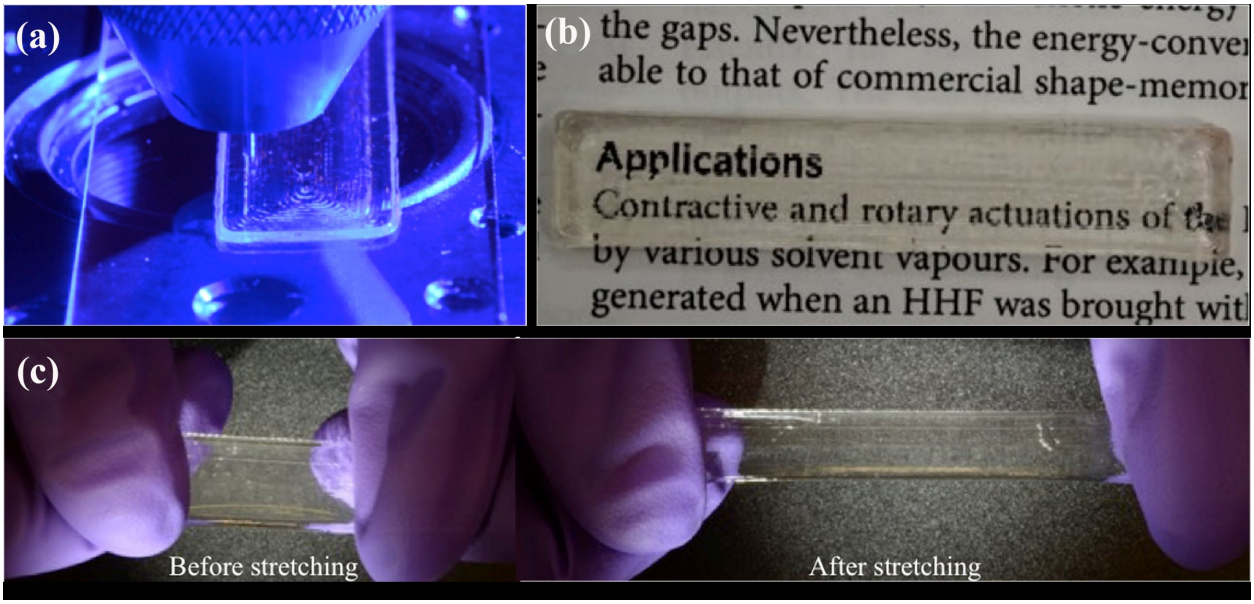

Figure 2. 3D printed ionic hydrogel. (a) Layer by layer printing of the hydrogel ink under UV light exposure. (b) Transparency of a 3D printed hydrogel sample. (c) Stretchability of a 3D printed hydrogel sample.

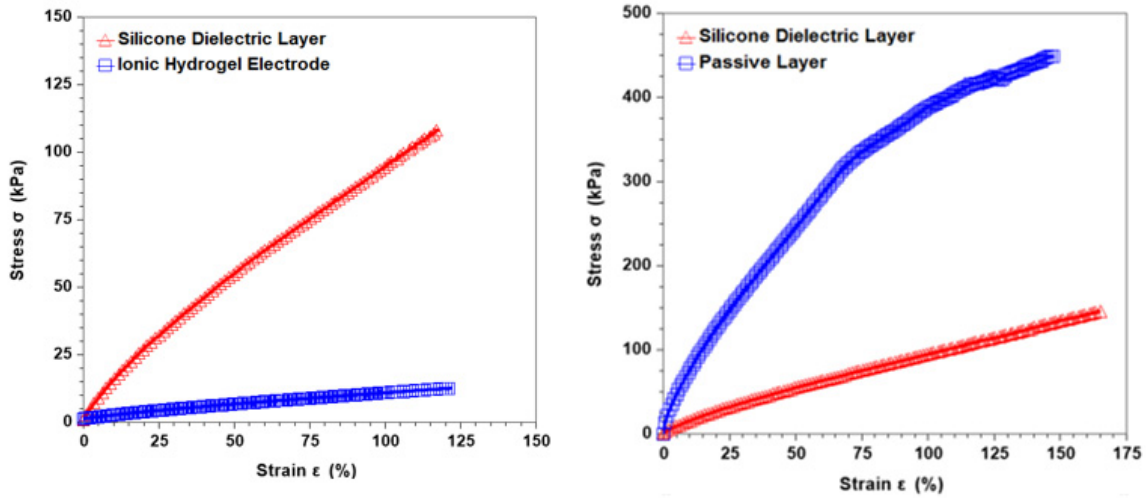

Figure 3. Comparison of stress-strain plots of hydrogel and passive layer with silicone dielectric.

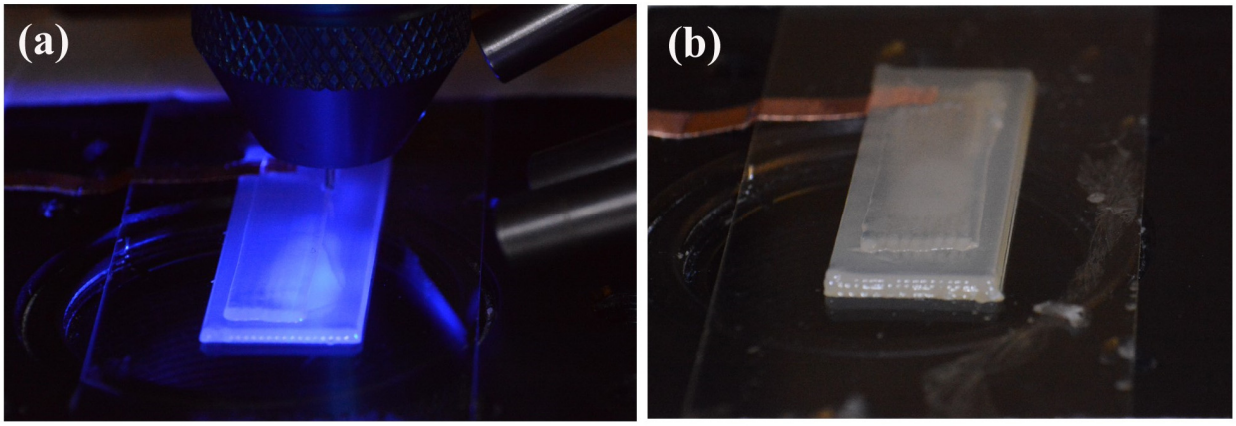

Figure 4. Multi-material 3D printing of the dielectric elastomer actuator. (a) Uniform deposition and bonding of the hydrogel on the silicone-based passive layer treated with Benzophenone. (b) Final 3D printed dielectric elastomer actuator structure.

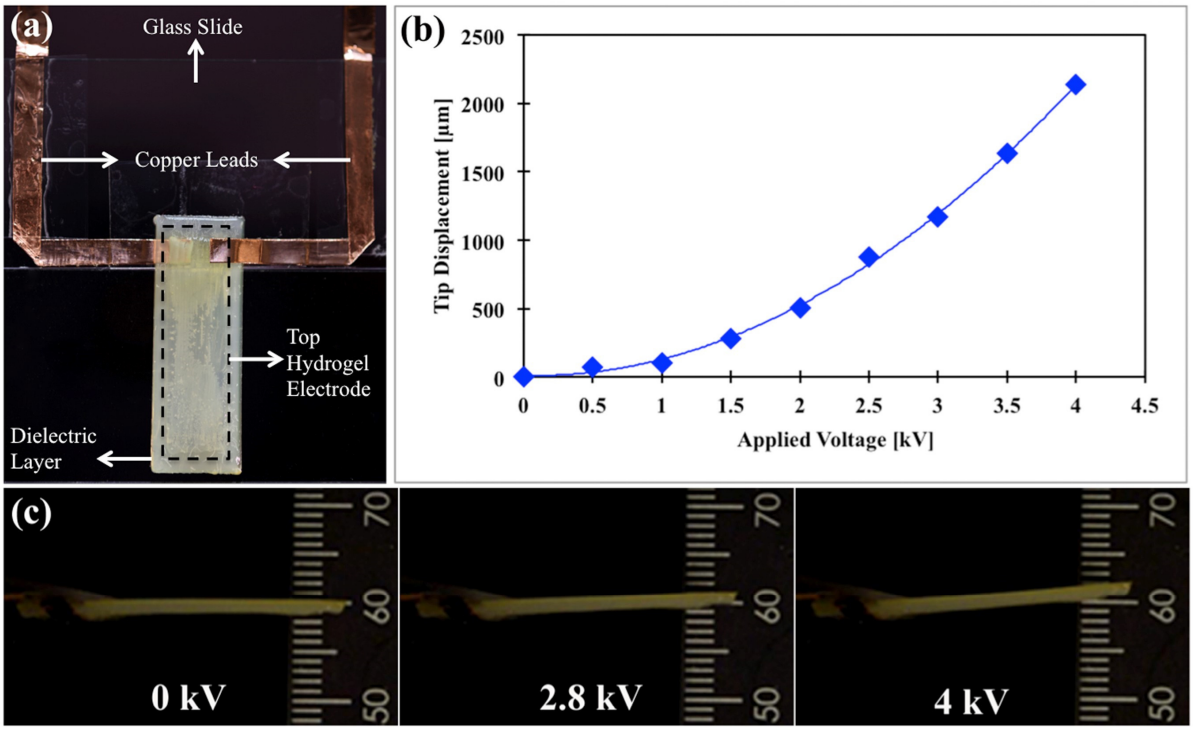

Figure 5. Actuation performance of the 3D printed device. (a) The test device anchored to a glass slide at one end. (b) The displacement of actuator's tip under the application of high electric fields. (c) The tip of the actuator exhibited a ~1 mm and 2 mm displacement at 2.8 kV and 4 kV.

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