

## †Electronic Supplementary Information

# Interactive Deformable Electroluminescent Devices Enabled by an Adaptable Hydrogel System with Optical/Photothermal/Mechanical Tunability†

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## **Methods:**

### **Materials:**

Polyvinyl alcohol [PVA, Kuraray, Inc., Mowiol 20-98; MW (molecular weight): 125,000; degree of hydrolysis: 98.0 to 98.8%], graphene oxide (GO, Xiwang Technology, Inc.), ammonium hydroxide (Acros Organics, Inc., 30% aqueous solution), dopamine hydrochloride (Alfa Aesar, Inc., 99%), sodium tetraborate (Alfa Aesar, Inc., anhydrous, 99.5%), polydimethylsiloxane (PDMS, Sylgard 184, Dow Corning, Inc.), zinc sulfur (Glowbug, Inc., particle size: ~ 20  $\mu\text{g}$ ) and Ecoflex<sup>®</sup> 00-30 (Smooth-On, Inc.) were used as received without further purification.

### **Synthesis of V-Slime, V-SP, V-SG, and V-SPG:**

A 10 wt % PVA aqueous solution was prepared by adding 10.00 g of PVA into 90.0 g deionized (DI) water followed by stirring vigorously at 80 °C for 2 hours. A GO aqueous suspension was prepared by adding 0.20 g of GO into 9.80 g of DI water and shaking on a mixer (Fisher analog vortex, Model: 9454FIALUS) for 2 hours. The polydopamine (PDA) nanoparticles were synthesized by the polymerization of dopamine in a solution at pH = 9. Samples of 40.0 mL ethanol and 90.0 mL DI water were mixed; subsequently, a certain amount of ammonium hydroxide was added to the mixture to adjust the pH to 9. A sample of 0.50 g dopamine hydrochloride was then added to the solution and stirred vigorously for 24 hours to ensure a complete reaction. Subsequently, the synthesized PDA was centrifuged at 2400 g force for 30 minutes and washed twice with DI water. At last, the PDA was heated at 65 °C for 24 hours. The PDA suspension was then obtained by uniformly dispersing 0.30 g PDA nanoparticles into 9.70 g DI water. A borax solution was prepared by adding 4.00 g sodium tetraborate into 96.0 g DI water under vigorous stirring for 2 hours.

Slime, SP, SG, and SPG were fabricated by mixing different amounts of PVA solution, GO suspension, PDA suspension, and DI water in a beaker under vigorous stirring for 20 min to ensure a uniform dispersion (see Table S1 for the ratios of the materials). Then, a pre-determined amount of borax solution was added to the mixture while stirring. The mixture would gel immediately, which was then sealed and left still for 24 hours to reach uniformity.

**Table S1.** Amount of different ingredients for V-Slime, V-SP, V-SG, and V-SPG (Unit: g).

		PVA solution (10 wt %)	GO suspension (2 wt %)	PDA suspension (3 wt %)	DI water	Borax solution (4 wt %)
V-Slime	1-10	10	0	0	5	1
	2-10	10	0	0	4	2
	3-10	10	0	0	3	3
	4-10	10	0	0	2	4
V-SP	1-10	10	0	1	4	1
	2-10	10	0	1	3	2
	3-10	10	0	1	2	3
	4-10	10	0	1	1	4
V-SG	1-10	10	1	0	4	1
	2-10	10	1	0	3	2
	3-10	10	1	0	2	3
	4-10	10	1	0	1	4
V-SPG	1-10	10	1	1	3	1
	2-10	10	1	1	2	2
	3-10	10	1	1	1	3
	4-10	10	1	1	0	4

**Fabrication of E-SPG:**

8.00 g of V-SPG was charged into a polystyrene petri dish and placed for 24 hours for degassing. Subsequently, the V-SPG was put into a refrigerator at -20 °C for 24 hours. The frozen V-SPG was then transferred into 30.0

mL 1.50 M trisodium citrate solution for 24 hours to salt out. Subsequently, the salted-out V-SPG was added into DI water for 24 hours until it reached equilibrium.

**Fabrication of electroluminescent (EL) dye/PDMS layer using doctor blade coating:**

4.00 g of EL dye was mixed with 2.00 g PDMS (base to curing agent = 10 to 1). A custom-made doctor blade tool with a gap height of 0.20 mm was used to spread the EL dye mixture on a pre-cleaned glass (20 cm × 40 cm) at a speed of 2.0 cm/s prior to being dried in an oven at 60 °C for 24 hours. The film was then peeled from the glass and cut into the pre-determined size.

**Fabrication of pressure responsive EL device:**

A copper tape (1.2 cm × 2.6 cm) was taped onto a glass substrate. Then a layer of EL dye/PDMS (1.6 cm × 3.0 cm) was placed atop the copper tape. Subsequently, 4.00 g of V-SPG (thickness: ~ 4.4 mm) was put onto the EL dye/PDMS layer and spread into a larger area (ca. 2.4 cm × 3.8 cm) evenly. Two wires from the power source were connected to the copper and V-SPG, respectively. One glass rod (diameter: 6 mm, round-end) was used to apply force (1.1 N) (corresponding pressure: 9.73 kPa) to the V-SPG layer for 1 second.

**Fabrication of laser engravable EL device:**

The preparations of the copper tape and EL dye/PDMS layer were the same as the above-mentioned steps. Then, 2.00 g of V-SPG 4-10 (thickness: ~ 1.7 mm) was put onto the dye layer and spread into a larger area (ca. 2.5 cm × 4.8 cm) evenly. A laser engraver (Glowforge model: BASIC) with a power of 16 W and a scanning speed of 500 mm/s was used to engrave V-SPG 4-10. Two wires from the EL power were connected to the copper and V-SPG 4-10, respectively.

**Fabrication of stretchable EL device for linear actuator:**

A cured PDMS (base to curing agent = 30 to 1, thickness: 7 mm, cured at 60 °C for 24 hours) was cut into a 2 cm × 8 cm stripe as the substrate. The PDMS stripe was attached to the two ends of the actuator (UPC: 787502202094, ECO LLC) using silicone adhesive sealant (Model 80050, Permatex). A stripe of E-SPG 1-10

(1 cm × 4 cm × 2 mm) was attached to the PDMS substrate as the bottom electrode. Then, an EL dye/PDMS layer (1.5 cm × 7.5 cm) was attached to E-SPG 1-10 and PDMS substrate. Subsequently, another larger E-SPG 1-10 (1.2 cm × 6 cm × 3.2 mm) was attached to the dye layer. Two wires from the EL power were connected to the two E-SPGs, respectively.

#### **Fabrication of finger bending monitoring EL device:**

A stripe of E-SPG 1-10 (0.8 cm × 2 cm × 2 mm) was attached to a nitrile glove substrate *via* a double-sided tape (3M 9474LE) as the bottom electrode. Then, an EL dye/PDMS layer (1 cm × 2.6 cm) was attached to the E-SPG. Subsequently, another larger E-SPG 1-10 (1.3 cm × 3 cm × 3.5 mm) was attached to the EL dye/PDMS layer. Two wires from the EL power were connected to the two E-SPGs, respectively.

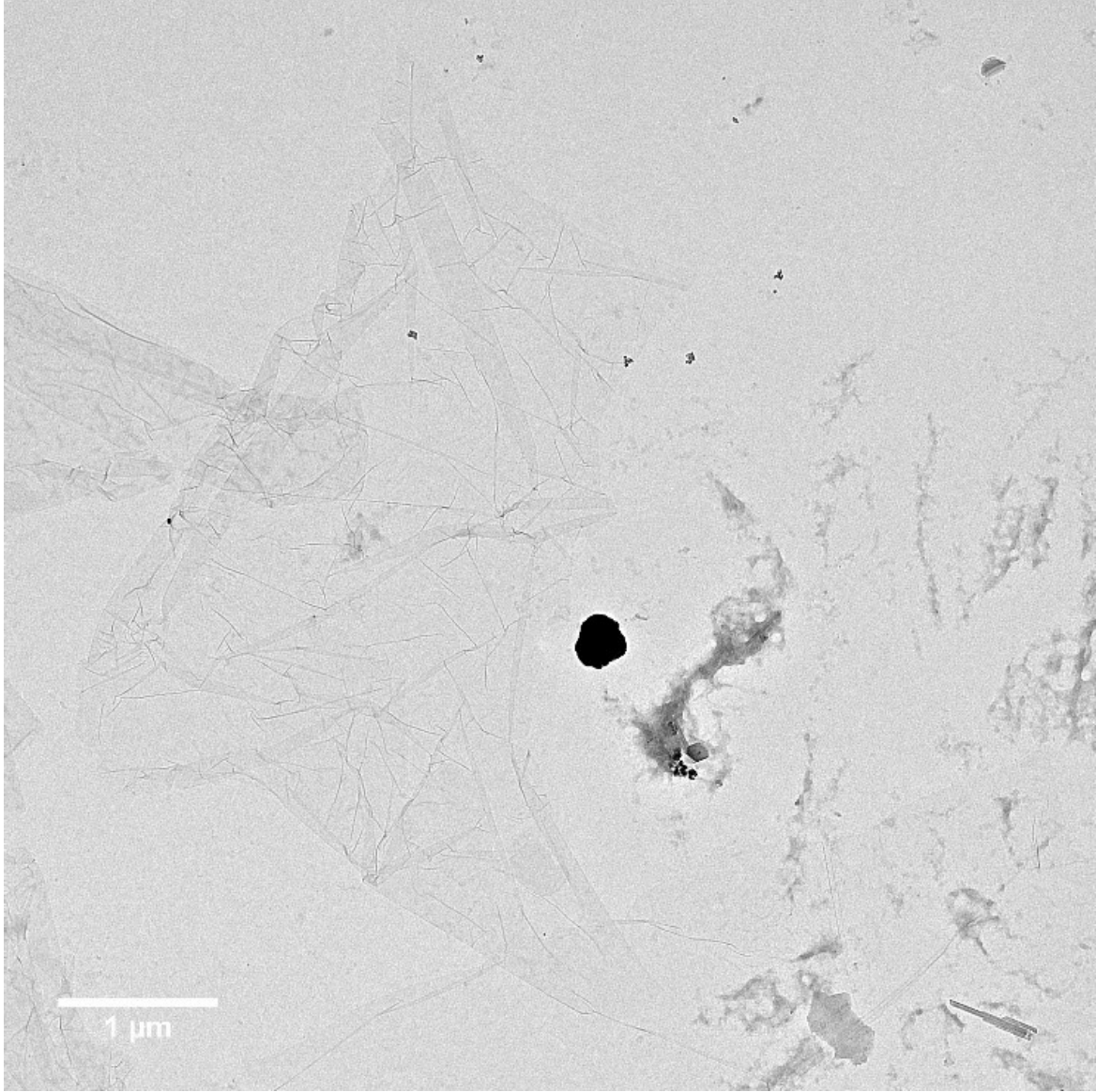
#### **Fabrication of bulging EL device:**

A VHB tape (4 cm × 4 cm) was attached to a sheet of polyethylene terephthalate (PET, 10 cm × 10 cm). Ecoflex® and PET/VHB were then cut into a circular shape with a diameter of 2.5 cm. The PET/VHB was attached to another sheet of PET by peeling the backing of the VHB. The circular Ecoflex® was stacked on the PET/VHB layer. A tube that was connected to a syringe was put in between the Ecoflex® and PET/VHB layer for inflation/deflation. A silicone adhesive sealant (Permatex 80050) was used to seal around the stack. 2.00 g of V-SPG 1-10 was evenly spread onto the bulging device as the bottom electrode. Then, an EL dye/PDMS layer (4 cm × 4 cm) was attached to the V-SPG 1-10. Subsequently, an E-SPG 1-10 (3.5 cm × 3.5 cm × 3.5 mm) was attached to the EL dye/PDMS layer. Two wires from the EL power were connected to the V-SPG 1-10 and E-SPG 1-10, respectively.

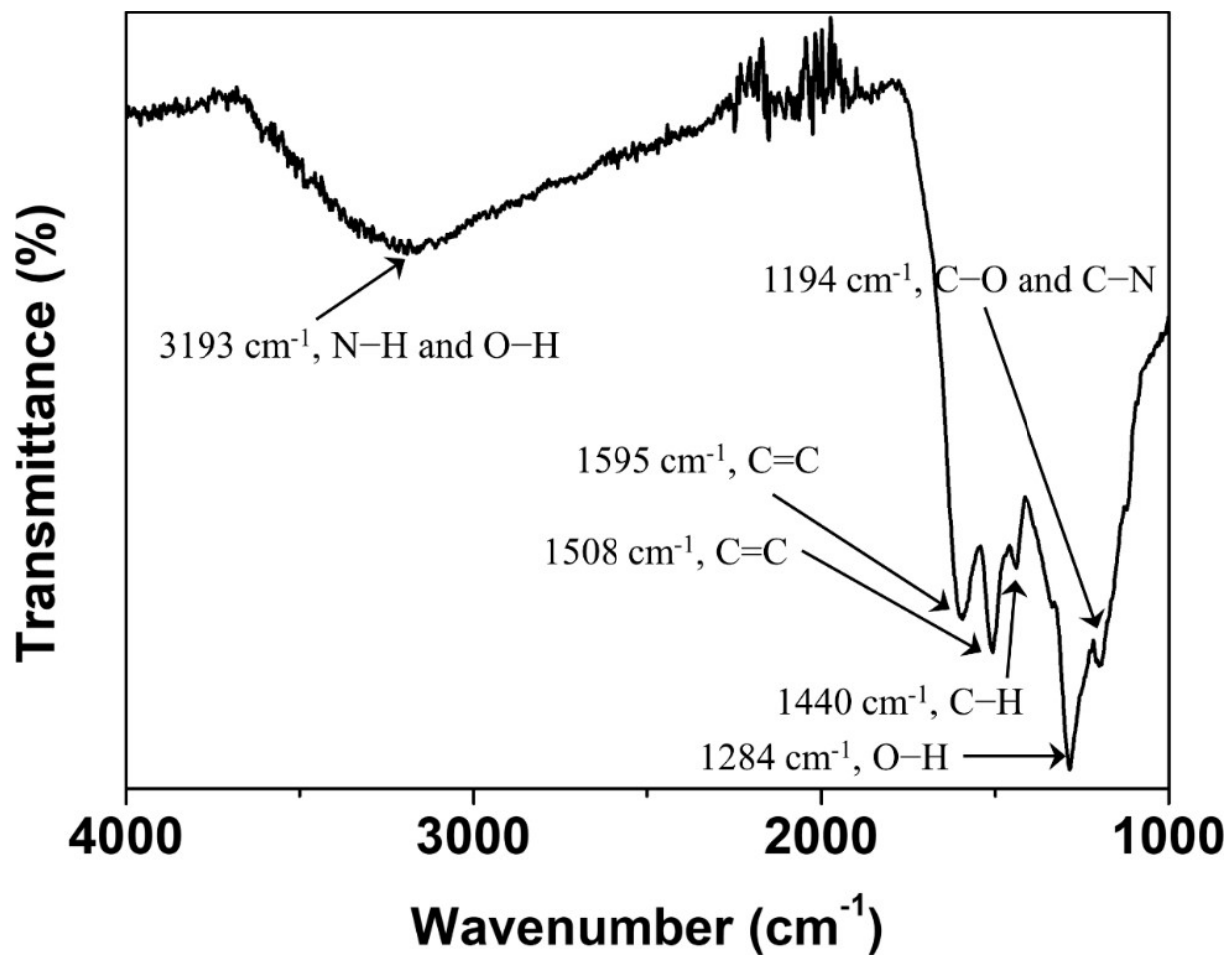
#### **Characterization:**

The transmittance of SPG was recorded on a PerkinElmer ultraviolet/visible/near-infrared Lambda 900 spectrophotometer. The surface images of the samples were taken on an AmScope ME 520TA optical microscope. The electroluminescent spectra of the samples were recorded on a Cary Eclipse Fluorescence Spectrometer (Agilent, Inc.) with a slit of 20 μm. The mechanical tests were performed on an Instron 5869

universal tester. The rheological tests were performed on a TA Instruments AR-G2 rheometer. The impedance change was recorded on a Hioki LCR meter (voltage: 1 V; Model: IM 3533-01). The dynamic light scattering was recorded on an ALV compact goniometer system with multi-detectors (CGS-3MD, Germany). The XRD patterns were recorded on a Bruker D2 diffractometer using a graphite monochromator with Cu K $\alpha$  radiation. All the digital photos and videos were captured with a OnePlus 7 Pro or an iPhone 12 Pro Max.

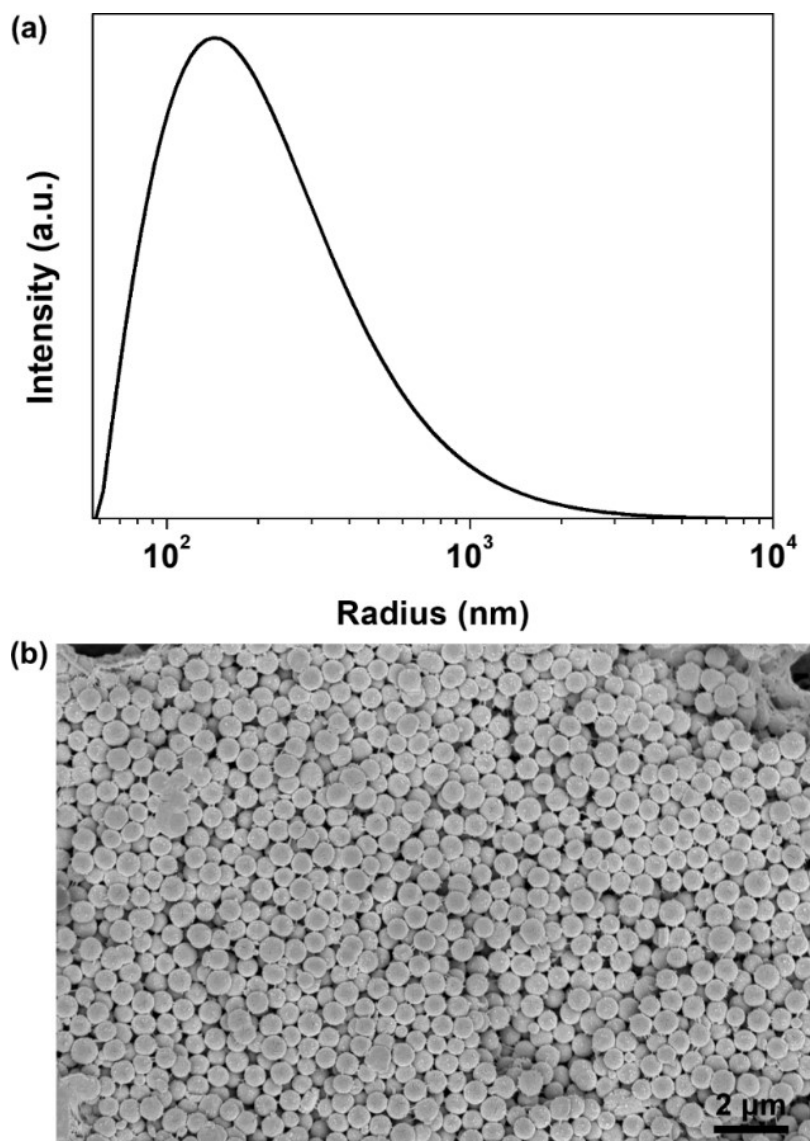


**Figure S1.** TEM image of the GO nanosheets.



**Figure S2. FTIR spectrum of PDA.** Here, the band at 3193 cm<sup>-1</sup> is assigned to the stretching vibration of N-H, and O-H. The characteristic peaks at 1595 and 1508 cm<sup>-1</sup> are assigned to C=C bonds. The small peaks at 1440 and 1284 cm<sup>-1</sup> are assigned to the deformation vibration of C-H and O-H. Also, the peak at 1194 cm<sup>-1</sup> is assigned to the stretching vibration of C-O and C-N.<sup>1,2</sup> These results confirm that PDA was synthesized successfully.





**Figure S3.** (a) DLS diagram and (b) SEM image of the synthesized PDA nanoparticles.

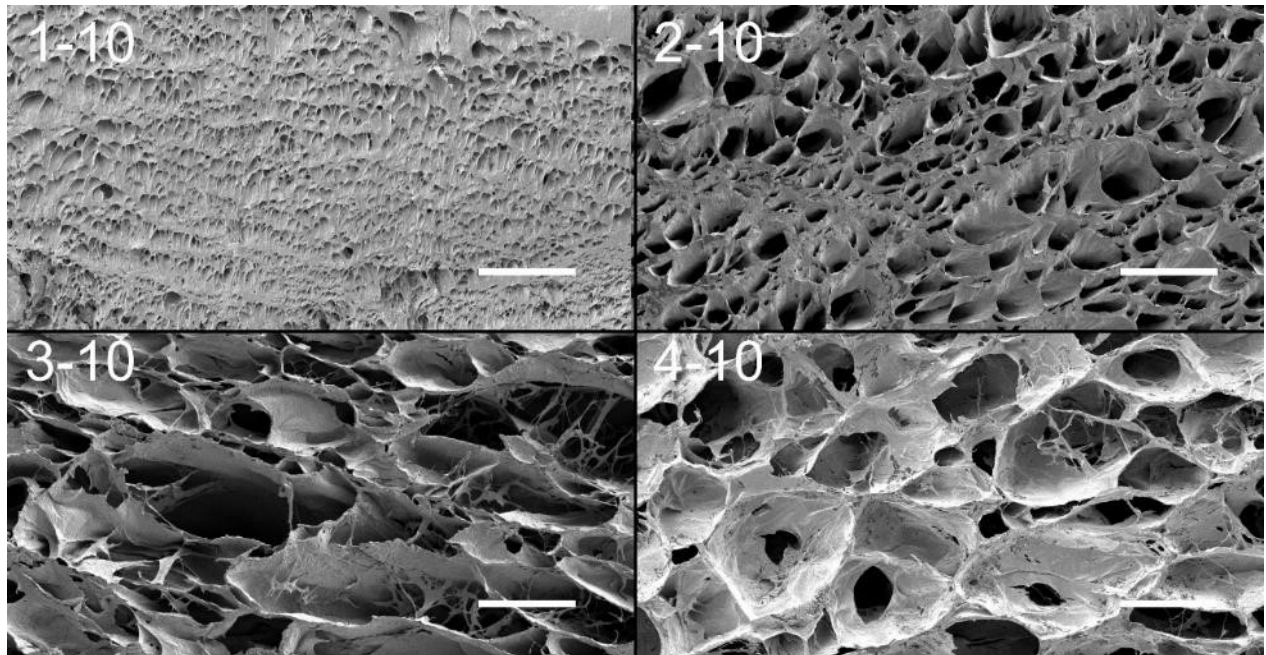
The number average radius and weight average radius of PDA nanoparticles were calculated to be 286 and 703 nm, respectively. The polydispersity was calculated to be 2.46. The number average radius and PDI were calculated using the following equations:

$$\text{Number fraction } N_x = \frac{I_x}{\int I_x dx}, \text{ where } I_x \text{ is the relative intensity;}$$

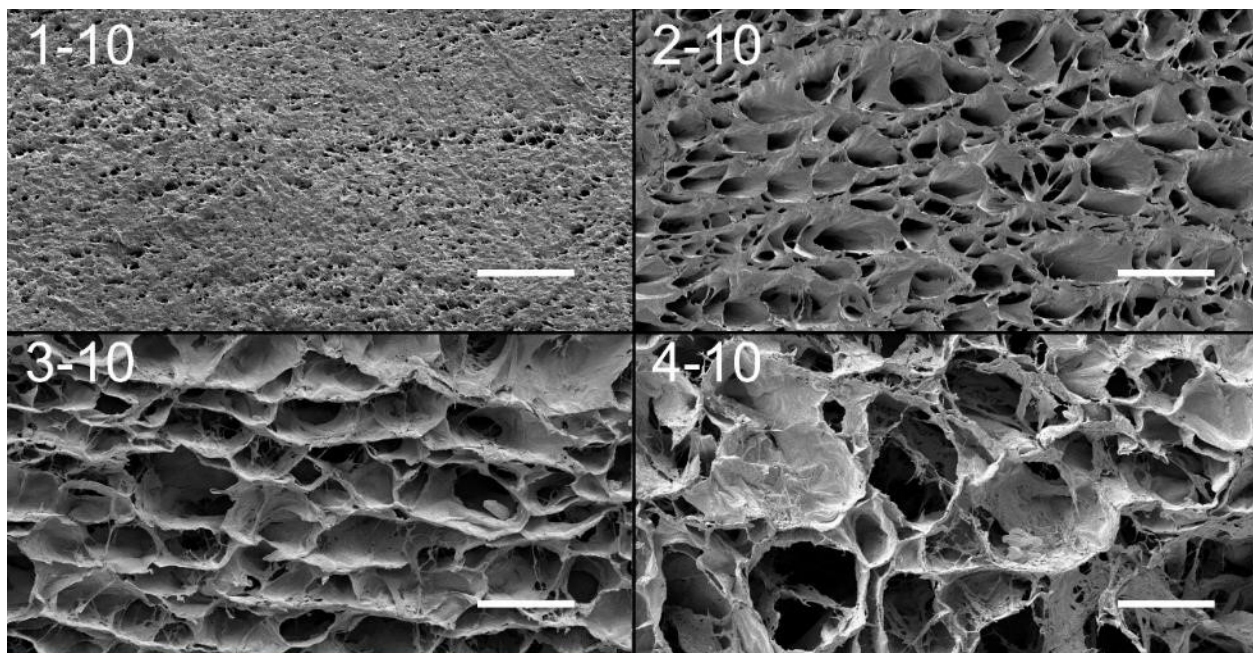
$$\text{Number average radius } R_n = \frac{\sum R_x N_x}{\sum N_x}, \text{ where } R_x \text{ is the radius when the relative intensity is } I_x;$$

$$\text{Weight average radius } R_w = \frac{\sum R_x^2 N_x}{\sum R_x N_x};$$

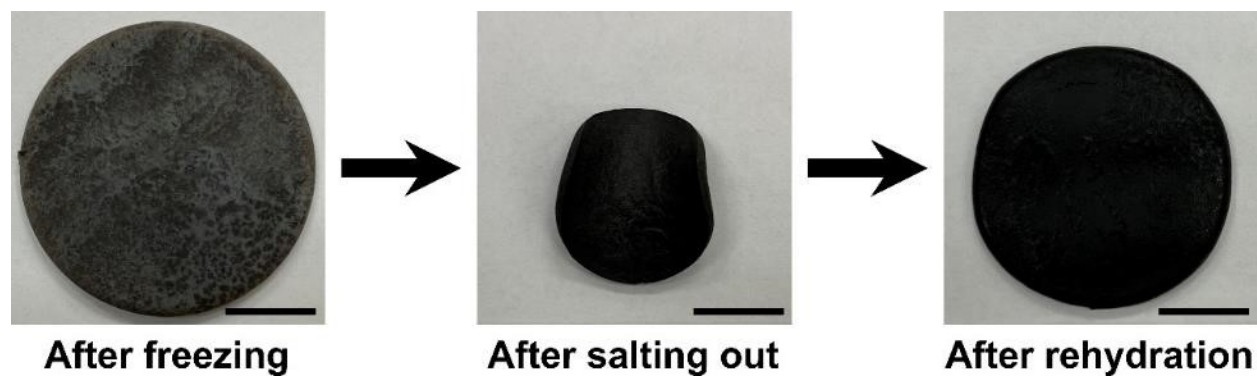
Polydispersity  $PDI = \frac{R_w}{R_n}$



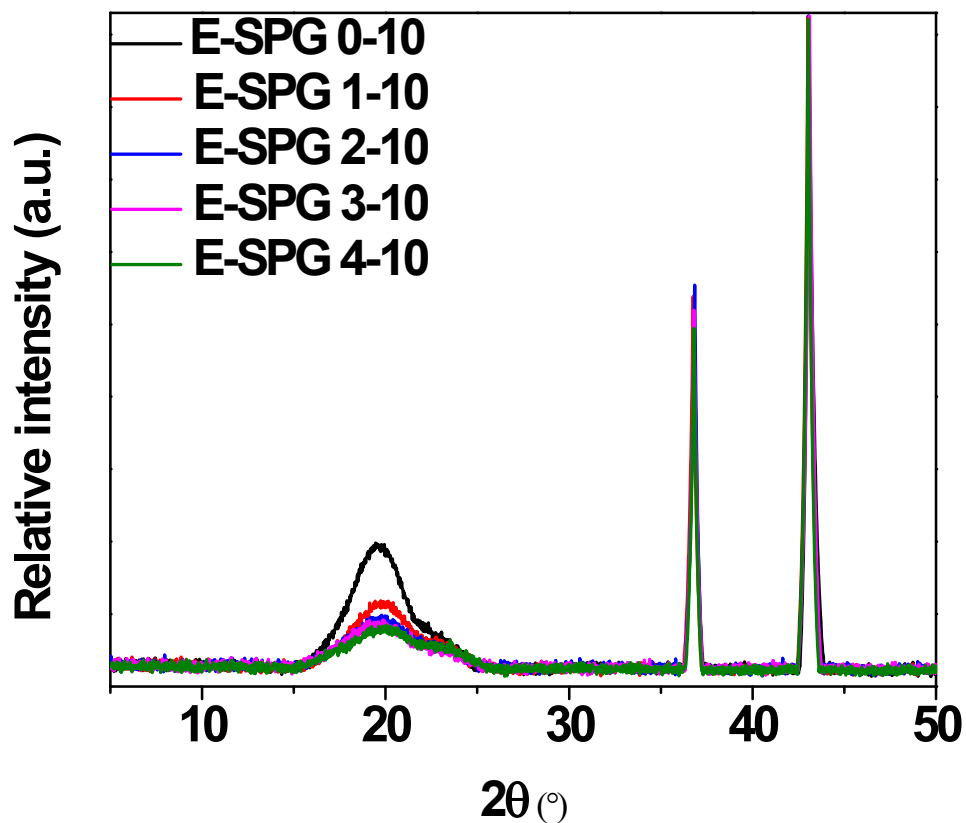
**Figure S4.** SEM images of E-SPG with different borax to PVA ratios (scale bars = 25 μm).



**Figure S5.** SEM images of E-Slime with different borax to PVA ratios (scale bars = 25  $\mu\text{m}$ ).

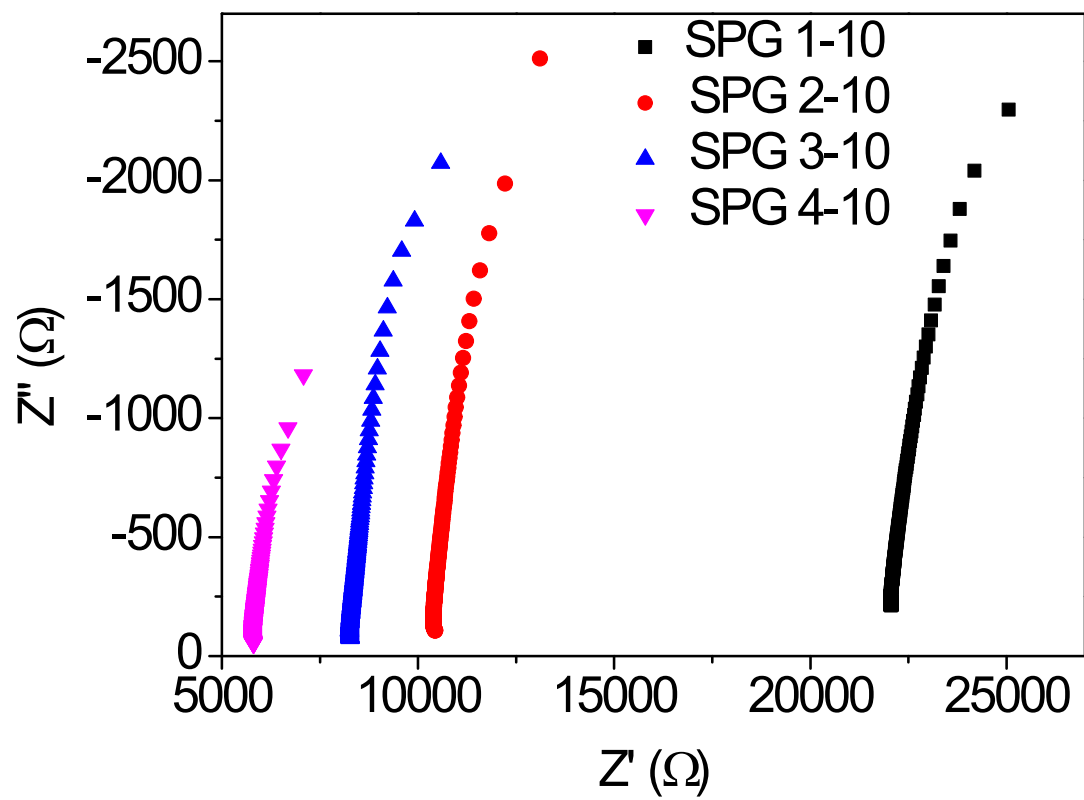


**Figure S6.** Digital photos of the frozen, salted-out, and rehydrated SPG 1-10 (scale bars = 2 cm).

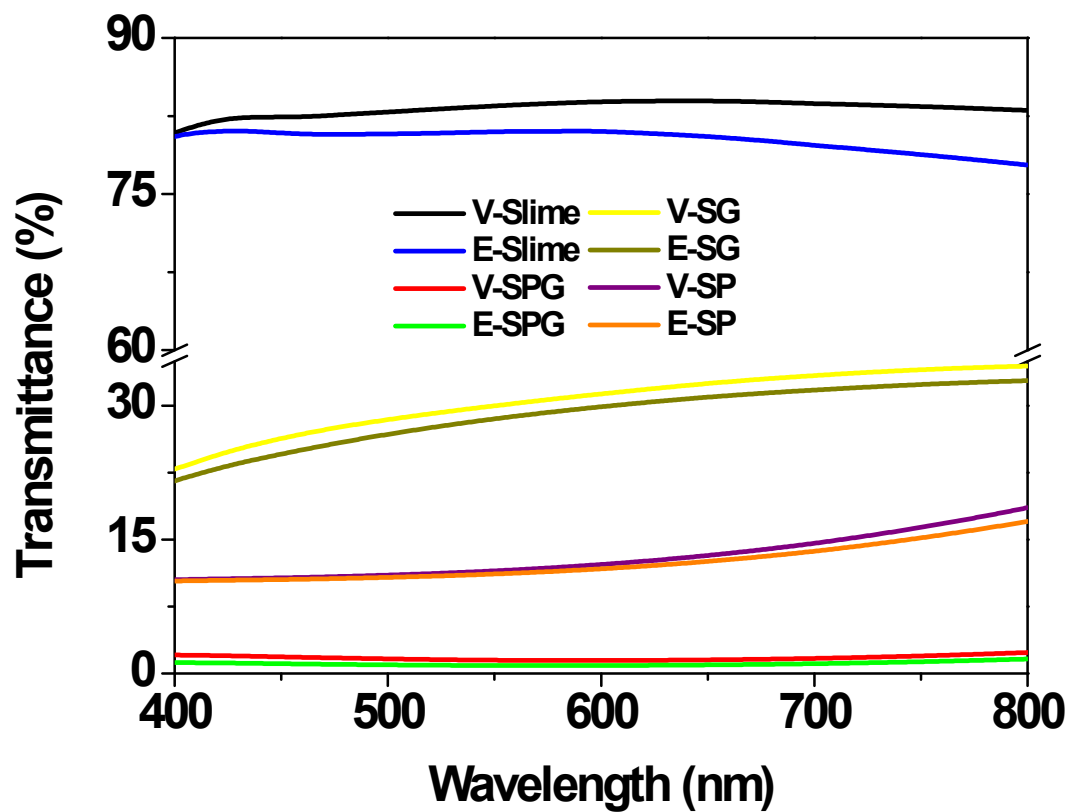


**Figure S7.** XRD patterns of E-SPG with different borax to PVA ratios.

All the samples were dried under 50 °C for 24 hours before XRD characterization. These patterns were recorded on an aluminum holder. The peaks at around 20° correspond to the (101) crystal plane of PVA.<sup>3,4</sup> The characteristic peaks at 36.8° and 43.0° are assigned to the aluminum holder. The intensities of all the patterns were normalized to these two peaks, which were chosen as the internal standards for comparing the crystallinity of different SPGs. The positions of the peaks didn't shift with the borax to PVA ratio, indicating that the increasing borax to PVA ratio does not change the structure of the PVA crystal phase. The intensity of the peaks decreased with an increasing borax to PVA ratio, exhibiting a lower crystallinity due to the constrained movement of the PVA chains.<sup>5,6</sup>



**Figure S8.** Impedance plot for SPGs (measuring voltage: 1.0 V).



**Figure S9.** Transmittance of V/E-Slime 1-10, SG 1-10, SP 1-10, and SPG 1-10 with a thickness of 4 mm.

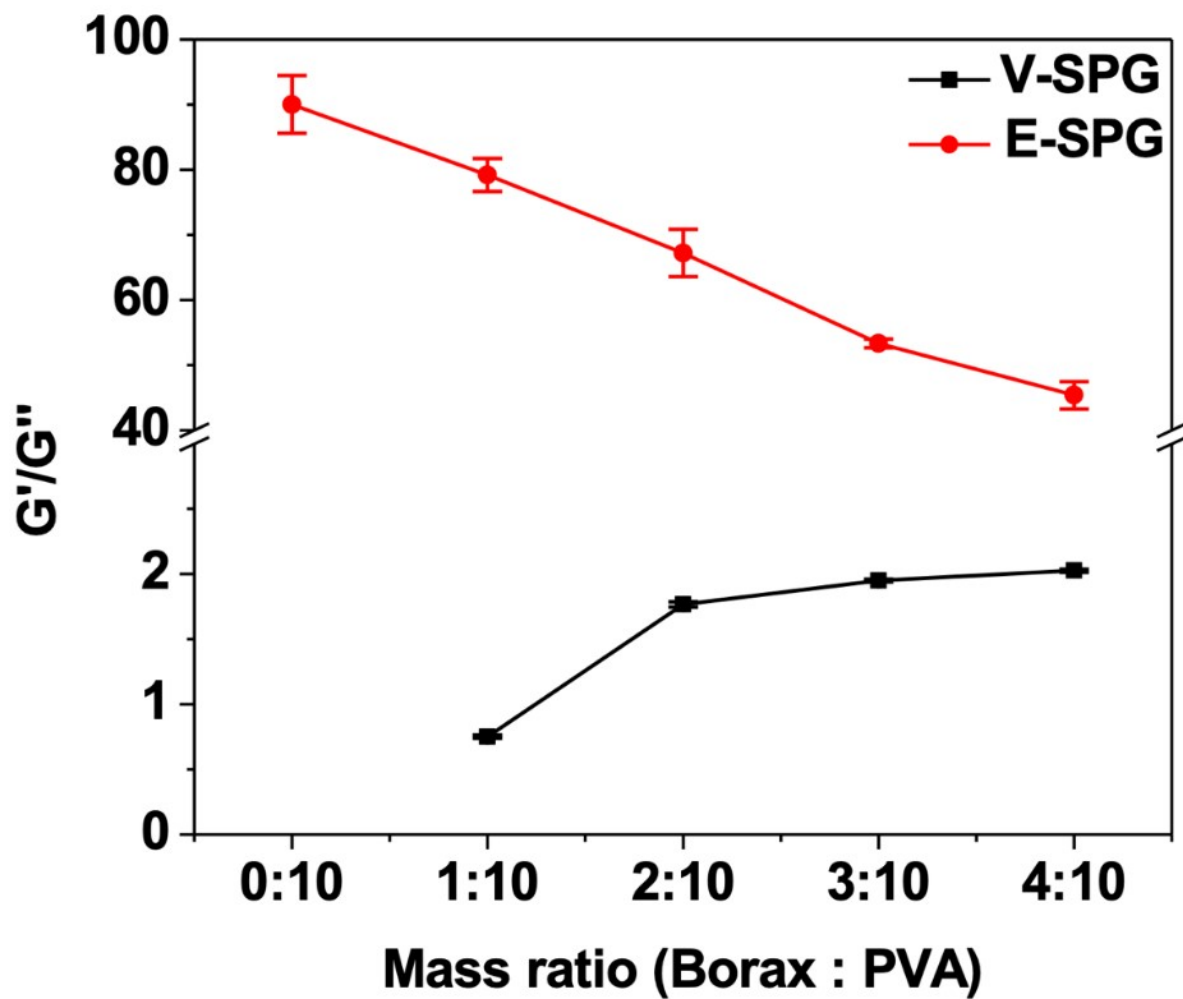
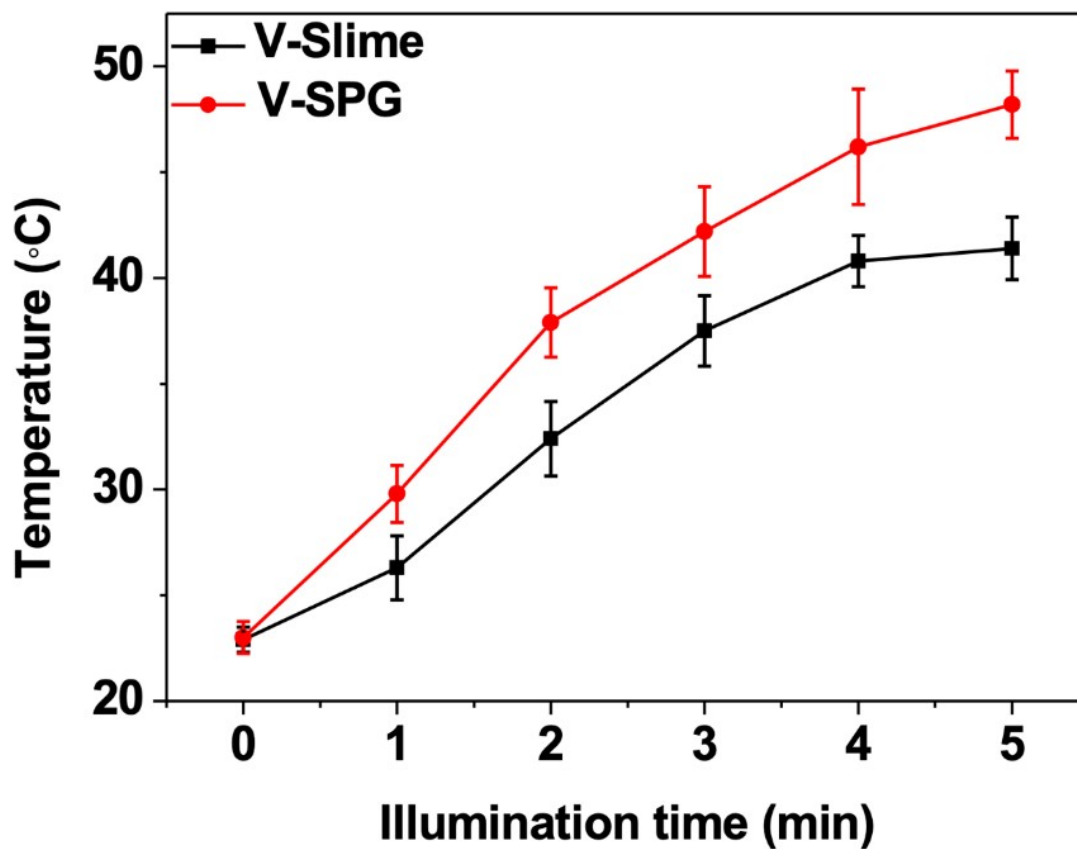


Figure S10. Storage modulus/loss modulus ratio of V- and E- SPG.





**Figure S11.** Photothermal performance of V-slime and V-SPG 4-10.

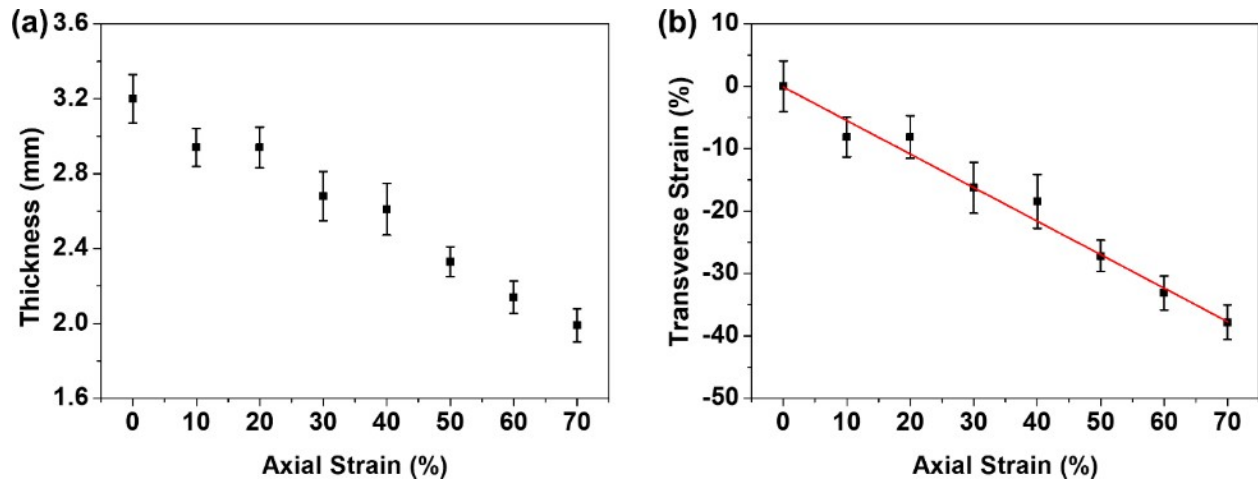


Figure S12. (a) Thickness of E-SPG under varying axial strains; (b) Relationship between transverse and axial strains, accompanied by a linear fit to determine the Poisson's ratio.

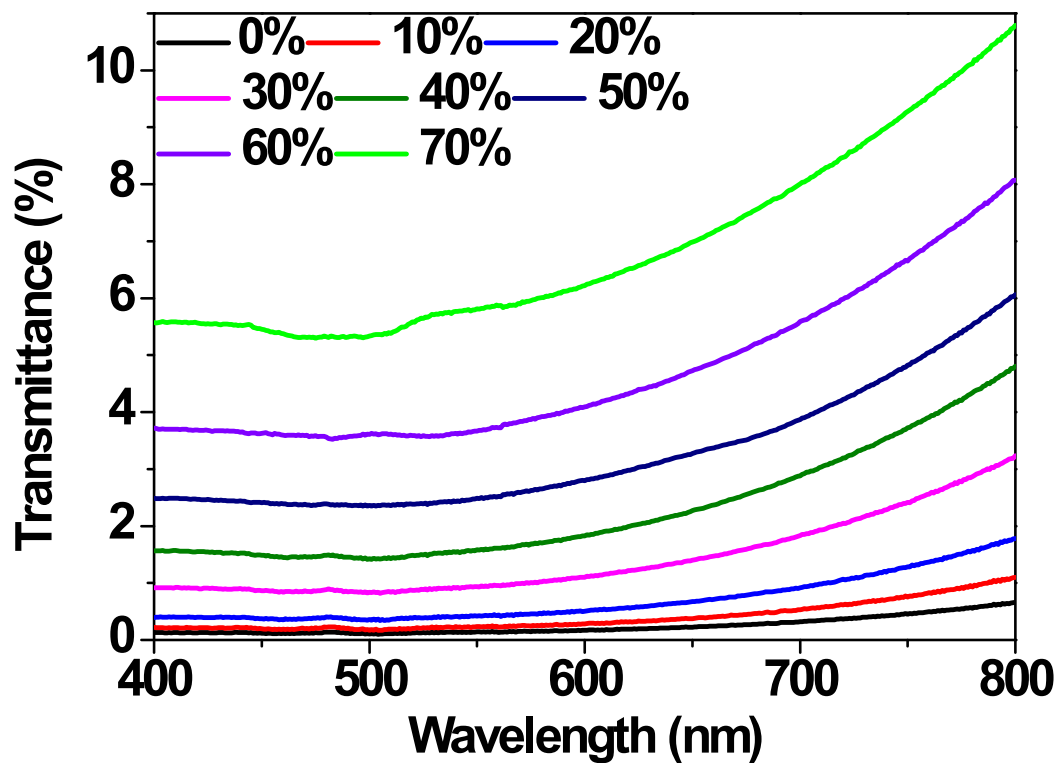
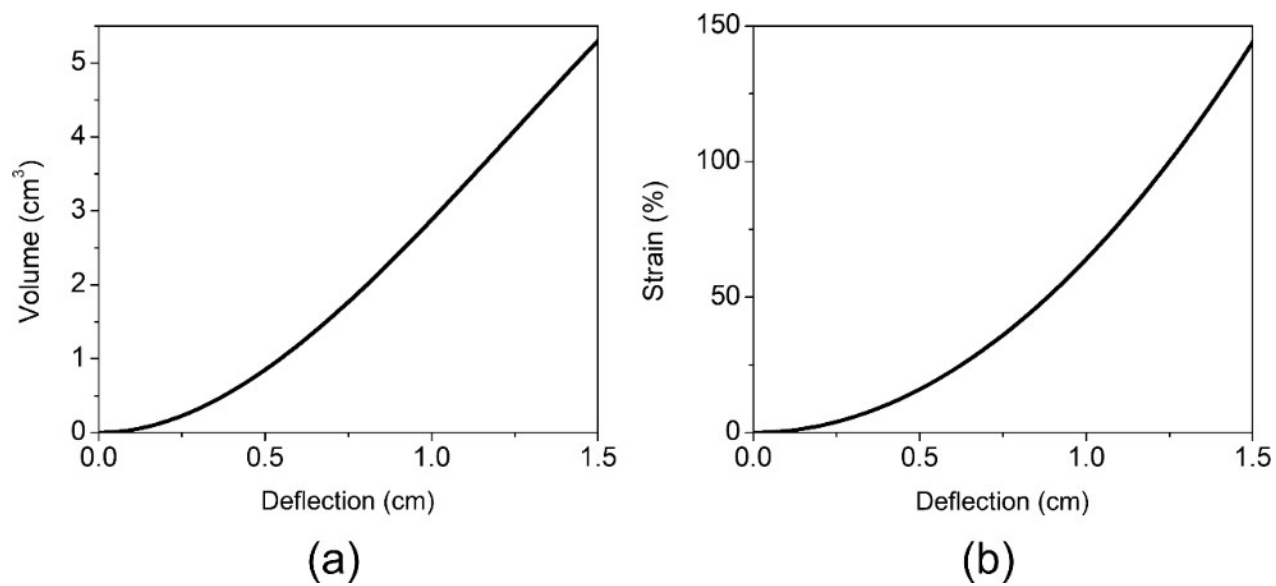


Figure S13. Transmittance of E-SPG 1-10 (thickness: 2.0 mm) under different strains.



**Figure S14.** Change of volume and strain in the bulging EL device as a function of deflection.

**Movie caption:**

Movie S1: V-SPG based soft EL device for pressure responsive application.

Movie S2: V-SPG based soft EL device applied for printing board application.

Movie S3: E-SPG based stretchable EL device applied for stretchable strain sensing.

Movie S4: E-SPG based stretchable EL device applied for stretchable and compressible strain sensing.

Movie S5: E-SPG based stretchable EL device applied for finger bending monitoring.

Movie S6:  $2 \times 2$  bulging EL device array for dynamic display.

## References:

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