## Supporting Information

## **Stretchable 3D Lattice Conductors**

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### 1. Experimental procedure

Fabrication of lattice conductors: We use a projection microstereolithography system to fabricate water-soluble hollow scaffolds. The printing process has been reported elsewhere and outlined in Fig. S1<sup>[1,2]</sup>. Once dried with air for 2 min (Fig. 1e), the hollow scaffold is filled with tin-catalyzed silicone elastomers (mold max NV14, base:crosslinker=100:7 by weight, Smooth-on). The filled elastomers are cured for 12 h at 25 °C (Fig. 1f). After that, the structure is immerged in 1 mol/L NaOH solution for 4 h. The elastomer structures are ready after washing in DI water for 2 min and air-drying for 2 min (Fig. 1g). Then, the elastomer lattices are dipped in a mixture of dopamine (2 mg/mL, Sigma-Aldrich) and 10 mM Tris-HCl (pH 8.5, bioWorld) for overnight. After the dipping, the elastomer lattice is rinsed with water and dried with nitrogen for 2 min. We then make a hydrogel pre-solution by mixing 2.34g acrylamide (Sigma-Aldrich), 0.006g N,N-methylenebisacrylamide (Sigma-Aldrich), various grams ammonium persulfate (APS, Sigma-Aldrich), 22.5 ml DI water as a solvent, 7.5 ml Glycerol (Sigma-Aldrich) and 6 g NaCl. Once adding 0.08 ml N,N,N',N'-tetramethylethylenediamine (TEMED, Sigma-Aldrich)<sup>[3]</sup>, we record the time. At various time points, the elastomer structures are dipped into the hydrogel pre-solution for five seconds. The coated structures are baked in an oven at 60° for 30 min, followed by UV irradiation (8W, 254 nm) for 20 min. The manufacturing throughput is estimated as 4.47 mm<sup>3</sup>/min. The thicknesses of the hydrogel coatings are measured using an optical microscope (Nikon ECLIPSE LV100ND) to image the cross sections of hydrogel-elastomer laminates (Fig. S3).

*Mechanical characterization*: The elastomer lattices and lattice conductors are cyclically stretched by an Instron tester with rate 0.0167 mm/s (model 5942, Instron). Finite element calculations are carried out in a commercial finite-element software, ABAQUS 6.10.1(**Fig. 2e**). The elastomer beams are taken to obey eight-chain model with parameters  $\mu$ =30 kPa and  $n_1$ =2.9 (see **Eq. 2**)<sup>[4]</sup>. The models are discretized by C3D8R elements and the result accuracy is ascertained through mesh refinement studies. The loadings in the simulations are displacement-controlled.

*Electrical characterization*: The resistances of the lattice conductors are measured using four-point-probe method sourced by a 60 Hz AC voltage 5-28V<sup>[3, 5]</sup>. The LEDs are connected in series with the hydrogel-elastomer structures powered by the 60 Hz AC voltage (20V).

*Expanding balloon*: Similar to the fabrication of elastomer architectures, an elastomer balloon with diameter ~1.5 cm and wall-thickness 500  $\mu$ m is fabricated based on a hollow water-dissolvable scaffold (**Fig. S8**). Once fabricated, the elastomer balloon is encapsulated within the third-order lattice conductor and activated by a controlled air pressure. The balloon diameter variation is measured from the image sequences. The resistances on two ends of the lattice conductors are measured using the four-point-probe method.

*Wastewater monitoring*: A lattice conductor with  $\sim$ 30µm hydrogel coating is first dipped in DI water for more than 2 hours to ensure equilibrium swelling of the hydrogel, and then dipped in the CuSO<sub>4</sub> solution for 5 min. Then the lattice conductor is taken out of the solution, and the residue water on the surface is quickly absorbed and removed by a tissue paper. The lattice conductor is subsequently mounted on a four-point-probe setup to measure the resistance. The resistance measurement only takes ~15s. Repeated measurements are conducted when the concentration of the CuSO<sub>4</sub> solution is increased step-by-step by adding concentrated CuSO<sub>4</sub> solutions.

# 2. Theoretical analysis of stress-strain behavior of the lattice conductors

Considering the geometrical deformation in **Eqs. 1-2**, we can formulate the free energy of the *i*th order lattice (**Fig. 2a**) as

$$W_i(\lambda_i) \approx v_i W_1(\lambda_{i,1}) \tag{S1}$$

where *i*=2 and 3,  $\lambda_i$  is the overall uniaxial stretch,  $\lambda_{i,1}$  is the stretch of the smallest beam,  $v_2 \approx 3\sqrt{3}\pi d^2 / (4l^2)$  and  $v_3 \approx 27\pi^2 d^4 / (16l^4)$  are the volume fraction of the second and third order structure, *d* is the beam diameter and *l* is the beam length. The elastomer can be modeled using eight-chain model with the free energy density expressed as<sup>[4]</sup>

$$W_1(\lambda_1) = \mu n_1 \left( \frac{\beta_1}{\tanh \beta_1} + \ln \frac{\beta_1}{\sinh \beta_1} \right)$$
(S2)

where  $\mu$  is the shear modulus of the hydrogel-elastomer hybrid,  $n_1$  is the average Kuhn number of the polymer chains,  $\beta_1 = \Gamma^{-1} \left( \sqrt{(\lambda_1^2 + 2\lambda_1^{-1})/(3n_1)} \right)$ ,  $\lambda_1$  is the uniaxial stretch,  $\Gamma^{-1}()$  is the inverse Langevin function, and the Langevin function is written as  $\Gamma(x) = \coth x - 1/x$ .

The corresponding uniaxial nominal stress can be expressed as

$$s_i(\lambda_i) = \frac{\partial W_i(\lambda_i)}{\partial \lambda_i}$$
(S3)

where *i*=2 and 3. The theoretical predictions from **Eq. S3** can consistently match the stress-stretch behaviors of the lattice conductors at various orders using parameters  $\mu \approx 30$  kPa,  $n_1 \approx 2.9$ ,  $\varsigma \approx 0.08$  and  $\alpha \approx -0.33$  (**Fig. 2b**). It is noted that the compressibility  $\alpha$  can be validated by experiments shown in **Fig. S6**. To further validate the theoretical model, we vary the beam aspect ratios d/l of the third order structures and

test their stress-strain behaviors which are also consistent with the theoretical predictions (Fig. S5).

#### 3. Supplementary figures



**Figure S1**. Schematic to show the 3D-printing process of the water-dissolvable scaffolds<sup>[2]</sup>. First, a 3D hollow computer-aided design (CAD) model is sliced into a series of images with a prescribed spacing along the vertical direction. These 2D slice images, illuminated with UV/blue light from a light emitting diode, are sequentially projected onto a resin bath, where the photoresin is capped in a prescribed height by a printing glass stage. The exposed resin is solidified, forming a layer structure bonded onto the printing stage. To eliminate the adhesion between the solidified resin and bath, an oxygen permeable membrane (Teflon fluoropolymer, CSHyde, USA) is attached on the bottom, inducing a thin layer (~5-20µm) of the oxygen-rich dead zone to quench the photopolymerization. As the printing stage is lifted off, the fresh resin can flow back. By lowering down the stage by a prescribed height and illuminating the resin with another slice image, the second layer can be printed and bonded onto the first layer. By repeating these processes, we can print a hollow scaffold with nearly arbitrary 3D architectures.



**Figure S2.** Dopamine molecules are oxidized in an acid solution and then polymerized overnight<sup>[6]</sup>.



**Figure S3.** (a) Optical microscope images to show hydrogel layers with thickness  $11.1 \mu m$  and  $53.6 \mu m$ , respectively. (b) The thicknesses of hydrogel layers for various ammonium persulfate (APS, photoinitiator) concentrations and time after adding N,N,N',N'-tetramethylethylenediamine (TEMED, crosslinking accelerator). We dipcoated hydrogel layers on the elastomer structures at different time points after adding the crosslinking accelerator TEMED into the hydrogel pre-solution.



**Figure S4**. Experimentally measured and theoretically fitted (a) nominal stresses and (b) resistance of a hydrogel bar sample in functions of applied uniaxial strains. The theory for the stress-strain behavior follows **Eq. S2** with parameter shear modulus  $\mu \approx 0.4$  kPa and  $n_1 \approx 4$ .



**Figure S5**. Experimentally measured and theoretically predicted stress-stretch behaviors of the third-order lattice with various beam aspect ratios.



**Figure S6.** The lateral stretch in a function of the uniaxial stretch of the third-order lattice.



**Figure S7**. (a-c) SEM images of PDA coatings on a mold max elastomer substrate at (a) unstretched, (b) stretched and (c) relaxed states. (d) Schematics to show the PDA coating on an elastomer structures in various states. At the stretched state, the PDA coating is fractured into islands; however, due to high adhesion between the PDA and the elastomer, the PDA islands are reassembled into a PDA coating after relaxation. (e) Similarly, we expect a PDA interfacial layer sandwiched between a hydrogel layer and elastomer substrate should follow a similar behavior during the substrate stretching and relaxing. The non-debonding between the PDA and elastomer substrate stretching and relaxation. Since the maximum stretch of the elastomer is 3.5 (**Fig. 2b**) and the maximum stretch of the hydrogel is more than 5 (**Fig. S4**), the hydrogel will remain intact before the elastomer failure.



**Figure S8**. (a) The fabrication process of an elastomer balloon similar to the fabrication process of the elastomer structure shown in **Fig. 1e-i**. (b) Experimental setup for expanding the elastomer balloon.

## 4. Supplementary movies

**Movie S1**: Cyclical stretching of the third-order elastomer structure. The movie speed is 10 times of the real experiment. The stretch goes from 0 to 7, returns to 0 and then increases until beam fracture.

**Movie S2**: An elastomer heart-mimic encapsulated within a lattice conductor is expanding cyclically using air pressure (three cycles).

## Reference

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