Supporting Information

Biomimetic hydrophilic islands for integrating elastomers and hydrogels of regulable curved profiles

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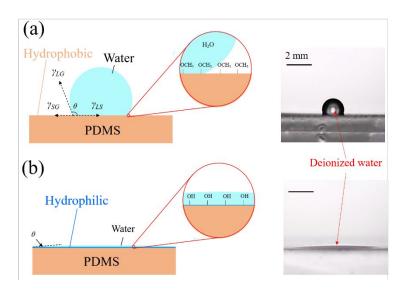


Figure S1. A water droplet on PDMS of different surface hydrophilicities. (a) A water droplet beads up on hydrophobic surface of PDMS. (b) A water droplet wets thoroughly on hydrophilic surface of PDMS. The volumes of water droplets are 2 μ L. Scale bars: 2 mm.

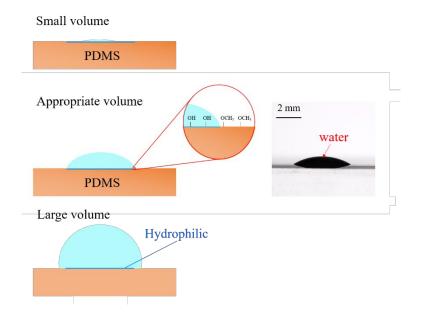


Figure S2. A water droplet on the hydrophilic island on the hydrophobic surface of PDMS. When the volume is small, the water droplet wets the hydrophilic island same as that on a uniformly hydrophilic surface. When the volume is intermediate, the water droplet has constant contact area with PDMS and its profile changes as volume increases. When the volume is sufficiently large, the water droplet expends across hydrophilic/hydrophobic intersection, recovering to that on a uniform hydrophobic surface.

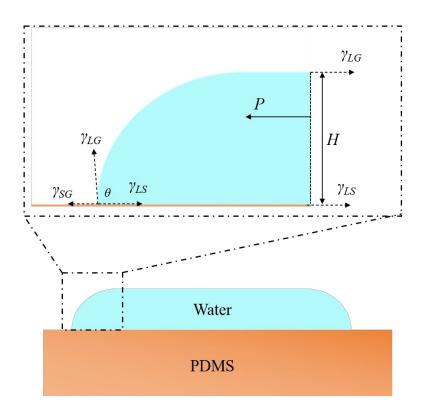


Figure S3. Mechanics analysis of a water droplet on a PDMS elastomer. When the volume of water droplet is sufficiently large, the radius of the droplet exceeds the capillary length of water and the gravitational effects prevail. The water droplet is flattened by gravity and takes a pancake shape at equilibrium with thickness of *H* as illustrated. Take a portion of the liquid for analysis, force balance in the horizontal direction requires that $P + \gamma_{SG} = \gamma_{LG} + \gamma_{LS}$, where *P* is the hydrostatic pressure, integrated over the entire thickness of the droplet, which equates to $\frac{1}{2}\rho gH^2$. Recall Young's equation which describes the equilibrium of forces at the triple point,

$$\gamma_{SG} = \gamma_{LG} \cos \theta + \gamma_{LS}$$
. Therefore, we obtain $H = \sqrt{\frac{2\gamma_{LG} (1 - \cos \theta)}{\rho g}}$.

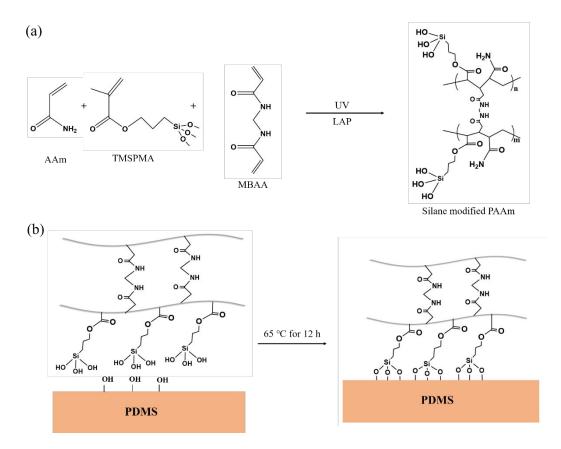


Figure S4. (a) Chemistry of modifying PAAm hydrogel with silanes. (b) Chemistry of silane condensation to form covalent interlinks between PAAm hydrogel and PDMS elastomer.

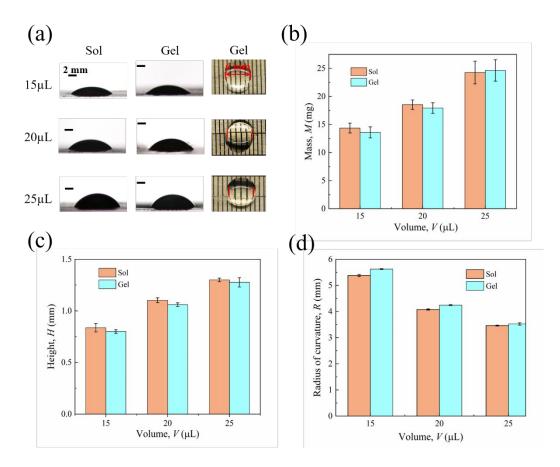


Figure S5. Sol-gel transition of gelatin hydrogel with high degree of fidelity of profile. (a) Side view and top view compare. Compare of (b) mass, (c) height, and (d) radius of curvature before and after sol-gel transition.

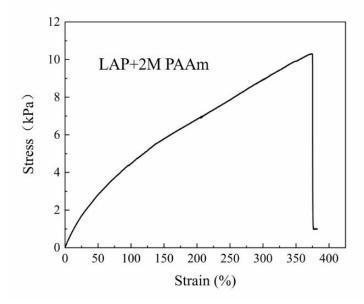


Figure S6. Stress-strain curve of the PAAm hydrogel used for ionotronic luminescent device. The samples have 10 mm in length (along the direction of stretch), 90 mm in width, and 2 mm in thickness. Samples are loaded on a testing machine (Instron 5966) with a 100 N load cell and pulled at a velocity of 60 mm/min. Three parallel samples were tested.

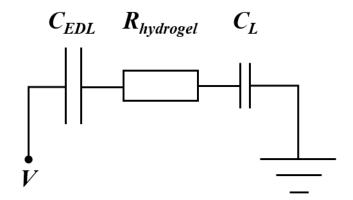


Figure S7. Equivalent circuit of ionotronic luminescent device. Neglecting the resistance of metallic wires, the equivalent circuit of the ionotronic luminescent device composes of three elements in series: a capacitor due to the electric double layer between ITO and PAAm hydrogel, a resistor due to the resistivity of PAAm hydrogel, and a capacitor due to the layer of ZnS:Cu particles dispersed PDMS elastomer. The capacitance per unit area of the electric double layer is on the order of 10⁻² F m⁻², resistivity of PAAm hydrogel containing 10 mol L⁻¹ lithium chloride is on the order of $10^{-2} \Omega m$, and thickness of luminescent layer is on the order of 10^{-4} m. The area of hydrogel is on the order of 10⁻⁵ m², permittivity of luminescent layer is on the order of 10⁻¹¹ F m⁻¹ (provided the relative dielectric constants of PDMS and ZnS be 2.5 and 8.5 respectively). The applied voltage has a frequency of 1 kHz. With the above parameters, we estimate the equivalent impedances of the electric double layer, PAAm hydrogel, and luminescent layer to be $|Z_{EDL}| = |1/j\omega C_{EDL}| \sim 10^2 \Omega$, $|R_{hydrogel}| = \rho l/s \sim 1 \Omega$, and $|Z_L| = |1/j\omega C_L| \sim 10^2 \Omega$. $10^9 \Omega$. In a series circuit, the voltage drops across a load is proportional to the impedance of the load. Because $|Z_L|$ is much larger than $|Z_{EDL}|$ and $|R_{hydrogel}|$, the applied voltage is mainly dropped across the luminescent layer. In particular, for an applied voltage of 2.5 kV, the voltage drops across the electric double layer is much below 1 V so that the device can work stably without electrolyzing the PAAm hydrogel. Indeed, we did not observe any noticeable bubbles during the experiments.



Figure S8. More hydrogel patterns on elastomer, including heart, star, square, circle and triangle shapes, as well as a "SUSTech" pattern and a "2020" pattern.

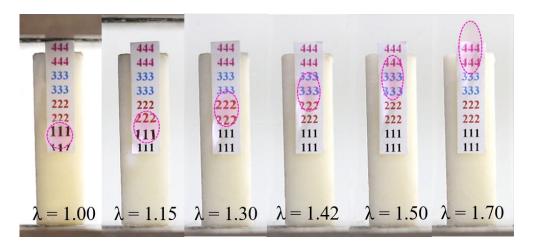


Figure S9. Soft tunable lens accommodates to the deformation of PDMS and changes its focal length accordingly.

Movie S1. 90° peeling test of hydrogel–elastomer hybrid. A polyacrylamide hydrogel/PDMS elastomer bilayer is fixed on the stage of a 90° peeling apparatus. A flexible but inextensible PET film is glued on the top surface of polyacrylamide hydrogel and fastened by the clamps of a loading machine (100 N load cell, Instron 5966). The machine pulls the PET film at a speed of 60 mm min⁻¹. The movie is played at $3 \times$ speed.

Movie S2. Selective light up of ionotronic luminescent device. Polyacrylamide hydrogels of various volumes (i.e. heights) are arranged into a pattern of "3" and cured on a luminescent layer, which is spin-coated on an ITO glass. The bottom of the ITO glass is adhered to a sheet of transparent glass using a double-side tape (very high bond, VHB). The transparent glass is supported by a home-made frame such that a digital camera can be used to capture videos from the bottom. Another piece of ITO is adhered to the crosshead of a mechanical testing machine using VHB. A sinusoidal voltage of amplitude 2.5 kV and frequency 1 kHz is applied on two ITO glasses. As the top ITO glass moves down with the crosshead, it contacts the hydrogels of highest heights to trigger the light up of pattern "1", and then pattern "7" and "3" sequentially.

Movie S3. Soft tunable lens. A gelatin hydrogel lens is fabricated on the 6-mm hydrophilic island of a PDMS elastomer, which is fastened onto a tensile machine at two ends. A 3D printed wedge with a slant edge is placed in front of the lens and a camera is used to capture the image of the Arabic numbers on the slant edge. The lens initially focuses on "111". As the PDMS elastomer is lengthened at a constant speed of 20 mm min⁻¹, the gelatin hydrogel lens deforms and changes its focus length, showing the Arabic numbers of "222", "333" and "444" with declining magnification factor. The feasibility of soft tunable lens is attributed to the strong, stretchable and transparent adhesion between gelatin hydrogel and PDMS elastomer. The movie is played at 5× speed.