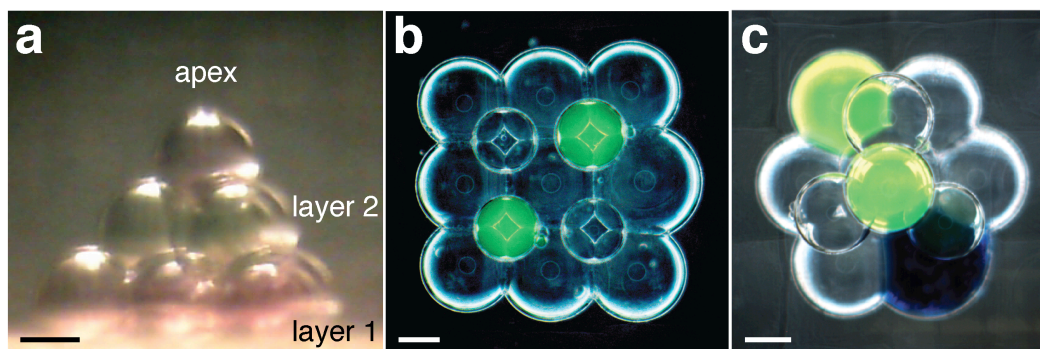


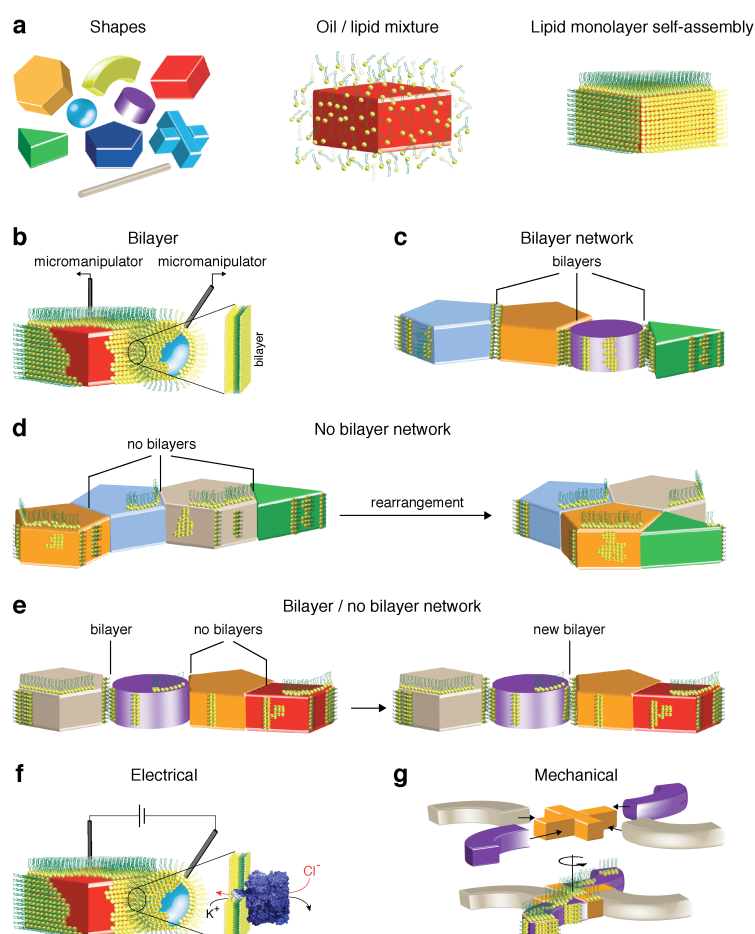
Figure 1



Packing droplets in different arrangements. (a) Aqueous droplets in a three-layered network forming a pyramid (face-centered cubic). The red (layer 1 and apex) and green droplets (layer 2) contained 5-carboxytetramethylrhodamine (5-cTAMRA) and fluorescein, respectively. (b) Droplets could be arranged in other lattice orientations (100) and (c) hexagonal close packing (111) on a patterned surface. Here, some droplets contained 10 mM pyranine (green) or 100 mM xylene cyanol (blue). Whether two droplets formed an interface bilayer could be controlled by changing the droplet size, e.g., if the droplets on a higher layer were smaller than those below, they did not touch each other but only communicated with the bottom droplets. Scale bars, 200 μm .

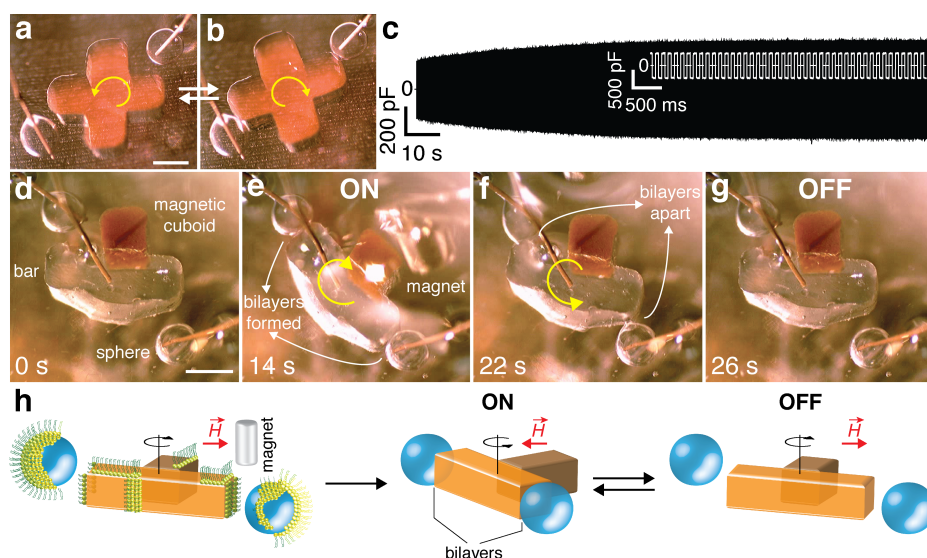
Specific communication paths defined by α HL pores in a 3D droplet network. (a) A 9-droplet square array containing 1 M KCl, 10 mM Tris.HCl, pH 7.0, formed the bottom layer. The four droplets at the vertices (1', 2', 3', 4') contained α HL heptamer ($\sim 83 \mu\text{g mL}^{-1}$). Four droplets (1, 2, 3, 4) containing α HL heptamer ($\sim 83 \mu\text{g mL}^{-1}$) were placed on the bottom layer to form a second layer, and finally a droplet containing pyranine (10 mM in 1 M KCl, 10 mM Tris.HCl, pH 7.0) was placed on top of the assembly without the help of a magnet. (b) After 4 d, a color change was observed in all the droplets of the second layer. (c) After an additional ~ 12 h, the four vertex droplets of the base layer became green, turning darker after another 12 h (d). (e – h) The vertex droplets could be removed one-by-one without compromising the structural integrity of the assembly. All the droplets contained DPhPC (1 mg mL^{-1}), and were assembled in a 1:1 (v/v) mixture of hexadecane / silicone oil AR20 containing DPhPC (2 mg mL^{-1}). Scale bars (a, e), $200 \mu\text{m}$ (b – d, same scale as a; f – h same scale as e).

Figure 3



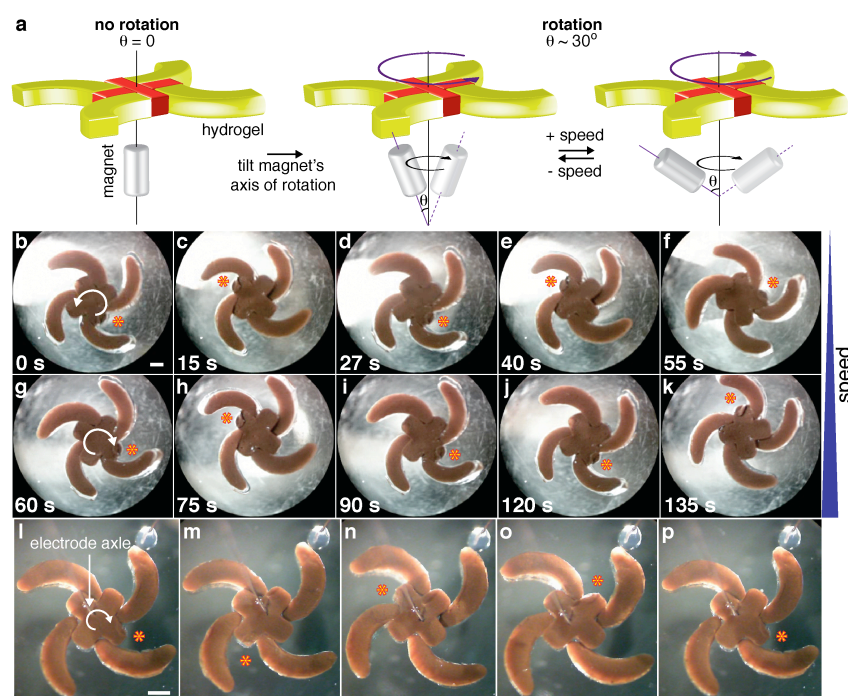
Lipid-coated hydrogel networks. (a) Hydrogel objects with various shapes immersed in a lipid / oil solution become coated with a lipid monolayer. (b) When two lipid-coated shapes are brought together with a micromanipulator, a bilayer can be formed at the interface. (c) At high lipid concentrations, a stable bilayer network is formed. (d) When the shapes are pressed against each other, the bilayers rupture at the hydrogel interfaces, and a network coated with a single external lipid monolayer is obtained. The positions of the hydrogel shapes in both bilayer and no-bilayer networks can be rearranged to change the network topology. (e) Networks can be formed with and without bilayers between specific hydrogel objects. In such a network, a new bilayer can be created between two shapes (which did not originally have a bilayer) by pulling them apart and forming the bilayer. (f) Bilayer networks can be used to form functional electrical and mechanical devices. For example, α HL pores can be used to carry an ionic current between two hydrogel shapes separated by an interface bilayer. (g) A hydrogel rotor is an example of a mechanical device.

Figure 4



Soft-matter mechanical devices. (a, b) An agarose cross (colored with 5-cTAMRA) was used as a switch to connect and disconnect two agarose spheres (diameter 1 mm) on Ag/AgCl electrodes, one sphere was connected to the ground and the other to the active end of a head stage. An electrical circuit was formed when the two poles of the cross formed bilayers with the spheres (1 mg mL⁻¹ DPhPC in hexadecane). In this case, the rotation of the hydrogel switch was performed manually using a steel needle. (c) Representative trace showing the capacitance increase during bilayer formation. (d – g) We automated the 'ON-OFF' function of the hydrogel switch by using a magnetic field. The switch was made by connecting a hydrogel cube loaded with paramagnetic beads and an empty hydrogel bar. Having the magnetic beads in only one hydrogel piece increased the spatial resolution with which the switch could be controlled. The switch was immersed in a lipid / oil mixture (1 mg mL⁻¹ DPhPC in 1:1 v/v hexadecane / silicone oil AR20). A wire was inserted in the empty hydrogel bar to act as an axle. A cylindrical Nd magnet (height 5 mm, radius 2.5 mm) was used to rotate the switch clockwise so that the poles of the hydrogel bar formed bilayers ('ON' state) with the hydrogel spheres. A counter-clockwise rotation of the switch separated the leaflets disconnecting the circuit ('OFF' state). Yellow arcs with arrowheads denote the clockwise and counter-clockwise rotation of the switch. The time in seconds is shown. The 'ON-OFF' cycle was repeated several times. (h) Representation of reversible bilayer formation using the switch. Scale bars (a and d), 1mm.

Figure 5



Reversible rotation of a hydrogel rotor. A hydrogel cross and four crescents were used to construct a rotor. (a) The rotor, loaded with paramagnetic beads, rotated under an external magnetic field. The rotation depended on the magnet's axis of rotation: when $\theta = 0$, the assembly did not rotate; when $\theta \sim 30^\circ$, the assembly rotated. The direction of rotation of the hydrogel assembly changed with the magnet's angular velocity. At lower speeds the assembly rotated counter to the rotation of the magnet, but at higher speeds the assembly switched its direction of rotation to match that of the magnet. (b – k) Movie snapshots of the rotor's counter clockwise and clockwise rotation at slow and fast speeds of the magnet, respectively. The time in seconds is shown. (l – p) Four crescents and a cross (both shapes contained magnetic beads) were assembled manually to form a rotor, which was rotated manually around an Ag/AgCl axle, which also acted as an electrode. Both mechanical assemblies were coated with a lipid monolayer in a lipid / hexadecane mixture (1 mg mL^{-1} DPhPC). The colored star denotes a defect in the cross shape used as a reference point to follow the rotation. Scale bars (b and l), 1 mm.