## Supplementary Information

# Reversible, voltage-activated formation of biomimetic membranes between triblock copolymer-coated aqueous droplets in good solvents

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#### 1 CSI Formation: Adjacent Droplets and Droplet-on-Hydrogel Methods

Copolymer stabilized interfaces are formed using two approaches, the first involving two adjacent aqueous droplets (Figure S1a) and the second involving one aqueous droplet and a planar hydrogel (Figure S1b). The droplet-hydrogel method<sup>1</sup> is specifically applied to obtain specific capacitance  $(C_m)$ measurements. Due to the low monolayer tension achieved in certain oil mixtures by rapid assembly of copolymer monolayers at the droplet-oil interfaces, droplets sag causing them to flatten significantly when resting on the bottom of the PDMS substrate. With the extremely low tension, the bottom-up view provided by the inverted microscope (i.e., see Figure 1b and Figure 2 in the manuscript) is thus unable to provide an accurate estimate of the bilayer ellipticity and area for  $C_m$  calculations. The droplet-onhydrogel approach<sup>2</sup> allows for direct visualization of the bilayer area (Figure 4a in the manuscript) to overcome the challenge of estimating ellipticity in the case of extremely low monolayer tension. As described in the manuscript, CSI area is manipulated by varying the magnitude of the applied dc voltage. The resulting planar CSI interfaces are viewed from below through a 4x objective lens on an Olympus IX51 inverted microscope, and images are acquired using a OI Click CCD camera controlled by QCapture software. CSI images are post-processed using the ImageJ to calculate the area of the interfaces at every voltage level. In parallel, electrical measurements are obtained via two Ag/AgCl electrodes: one inserted into the droplet and one inserted into the conductive hydrogel. For CSIs formed between two adjacent droplets, two Ag/AgCl electrodes are used with one inserted into each droplet.



Figure S1: (a) CSI formed side-by-side. (b) CSI formed by droplet-on-hydrogel technique.

## 2 Electoporation of CSIs



Figure S2: Representative current measurements at dc voltages near the rupture potential for CSIs formed in: (a) hexadecane, (b) decane, (c) 1:3 AR20-hexadecane, and (d) 1:1 AR20:hexadecane. The transient increases in current are attributed to electroporation of the membrane, which leads to rupture.

### 3 References

- 1. J. R. Thompson, A. J. Heron, Y. Santoso and M. I. Wallace, *Nano letters*, 2007, 7, 3875-3878.
- 2. L. C. M. Gross, A. J. Heron, S. C. Baca and M. I. Wallace, *Langmuir*, 2011, 27, 14335-14342.