Supplementary Information: Electrowetting without External Voltage using Paint-on Electrodes

Collin B. Eaker[a], Ishan D. Joshipura[a], Logan R. Maxwell[a], Jason Heikenfeld[b], Michael D. Dickey[a,]*

[a]Department of Chemical and Biomolecular Engineering,

North Carolina State University, Raleigh, NC 27695-7905, USA

[b]Department of Electrical Engineering and Computing Systems,

University of Cincinnati, Cincinnati, OH 45221-0030, USA

Electrochemical Impedance Spectroscopy (EIS)

The capacitance of the electrowetting system was measured using EIS and compared for both DI H_2O (Fig. S1a and S1c) and DI H_2O/SDS (Fig. S1b and S1d) solutions. EIS was performed at 0 V, 0.1, 0.2, and 0.3 vs. the open-circuit potential and repeated three times. The capacitance of the electrowetting system was determined based on model fits of the two spectra. DI H_2O and DI H_2O/SDS were determined to be 0.781 $\mu F/cm^2$ and 0.298 $\mu F/cm^2$, respectively. Theoretical electrowetting curves based on the measured capacitance for their respective solutions follow the experimental data shown in Fig. 1F. As expected, the capacitance for the DI H_2O/SDS is lower than DI H_2O. In the former solution, a lipid bilayer forms at the interface of the H_2O and EGaIn oxide. As a result, the overall thickness increases for the dielectric.

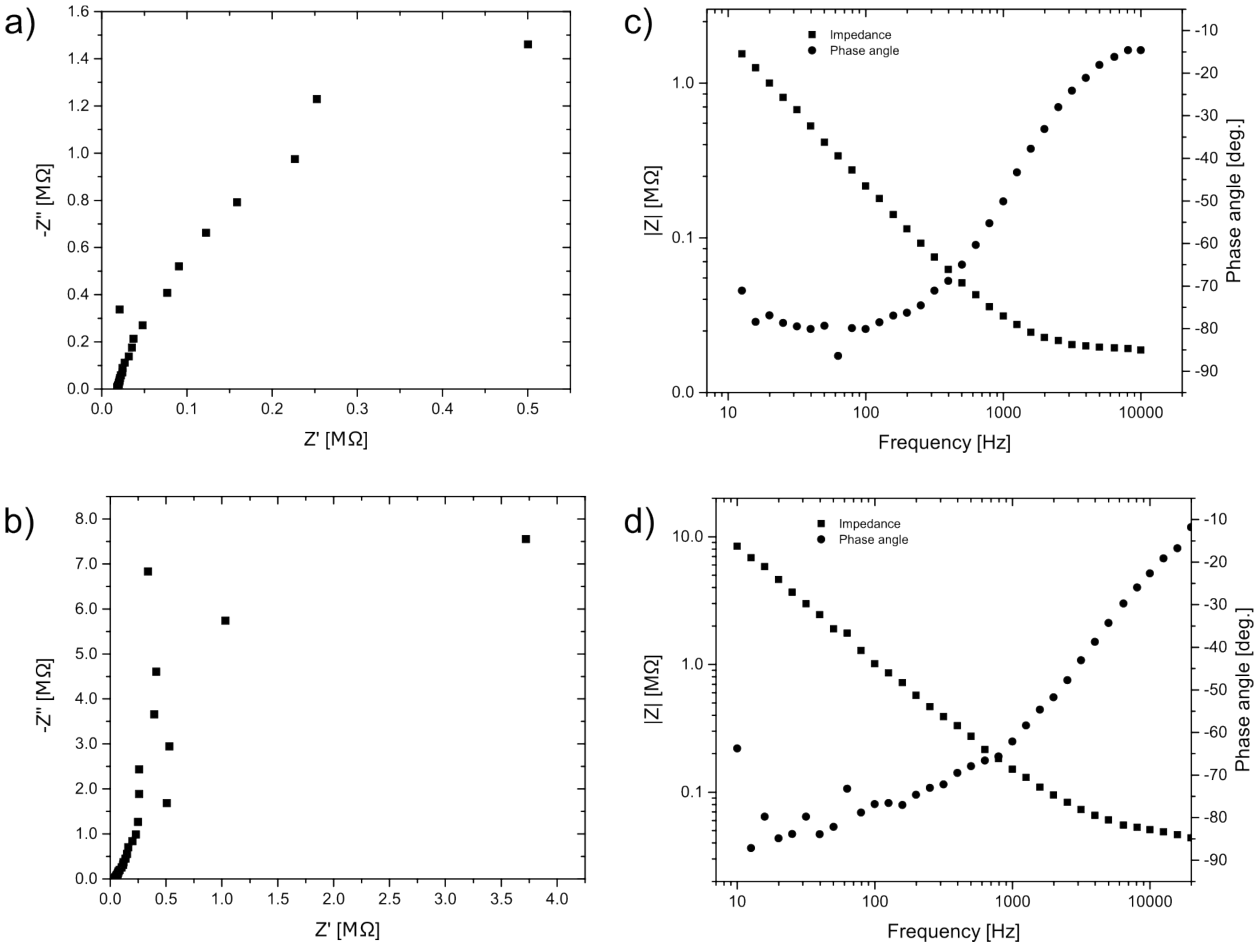

Fig. S1. Electrochemical Impedance Spectroscopy (EIS): EIS was performed on Bio-Logic SP-200 potentiostat from 100 kHz to 10 Hz using a two-electrode system. Figure S1 shows representative Bode (left) and Nyquist (right) plots at 0 V (vs. open-circuit potential). Nyquist plots for DI H_2O (Fig. S1a) and DI H_2O/SDS (Fig. S1b) compare the real (Z') and imaginary (-Z") components of the system impedance. Bode plots for DI H_2O (Fig. S1c) and DI H_2O/SDS (Fig. S1d) compare the magnitude of impedance (|Z|) and phase angle of the system.

Dielectric Thickness Calculations

Previous experiments show that passivating oxide layer thickness is approximately 1-3 nm. We also sought to determine the thickness of the adsorbed surfactant layer (to find the total dielectric thickness). We use a simple model of capacitance (Equation S1):

$$\frac{1}{C} = \frac{1}{C_{ox}} + \frac{1}{C_{LB}} \qquad \text{(S1)}$$

where C is the capacitance measured from EIS between the pure DI H_2O drop and the liquid metal (0.781 μF/cm^2), C_{ox} is the capacitance of the gallium oxide layer, and C_{LB} is the capacitance created by the lipid bilayer in dodecane oil. Putting these values in terms of the dielectric permittivity and thicknesses yields Equation S2:

$$\frac{1}{C} = \frac{d_{ox}}{\varepsilon_{ox}} + \frac{d_{LB}}{\varepsilon_{LB}} \qquad \text{(S2)}$$

here, the d_{ox} and d_{LB} represent the thickness of the oxide layer and bilayer, respectively, and ε_{ox} and ε_{LB} represent the dielectric permittivity (with dielectric constant values of 10 and 2.1 for the oxide and bilayer, respectively). Solving the equation for d_{LB} yields a bilayer thickness of approximately two nm. The calculated thickness for the lipid bilayer is in agreement with similar electrowetting systems that utilize lipid bilayers on metal oxides.[17]

Hysteresis Effects

Previous electrowetting systems utilizing low voltage have shown significant hysteresis (i.e. the electrowetting curves depend strongly on previous states of the system). We demonstrate that by utilizing AC potentials, we can mitigate or eliminate this hysteresis. Fig. S2 shows the data (from which Fig. 2a is derived), in which forward and reverse sweeps of potential are utilized. These data indicate that the direction of the sweep is not a significant factor in the contact angle change, with a maximum contact angle difference of 4-5 degrees between the forward and reverse sweeps.

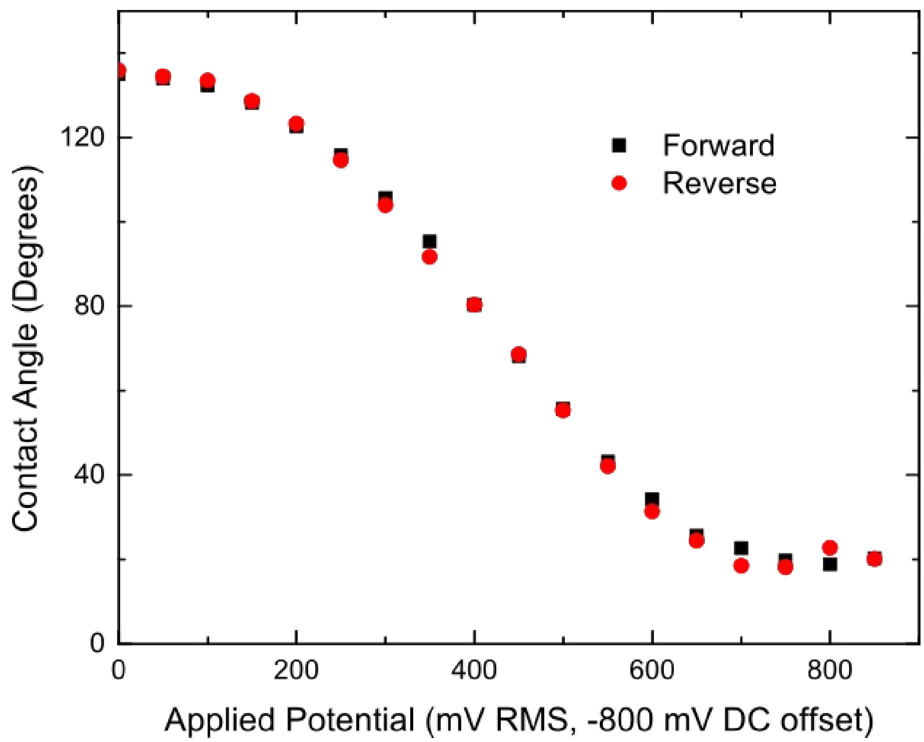

Fig. S2: Hysteresis data for AC electrowetting curves. Electrowetting data for DI H_2O/1 wt% SDS on a smooth film of EGaIn. Each data point represents the average of two runs, with a 100 Hz sine waveform applied for 1 minute. A -800 mV DC offset was used to account for the potential of zero charge. Forward and reverse runs were taken with the same drop, in the same position on the substrate.

Short Circuit: Fig. 4b indicates that shorting the top and bottom electrodes would create a Faradaic current for the platinum, copper, and stainless steel electrodes (but not for gallium or EGaIn). We tested this assertion by physically closing the circuit for 30 seconds and measuring the resulting current. As shown in Supplementary Fig. S3, while platinum, copper, and stainless steel all showed a decaying current (with wetting behavior), the gallium and EGaIn showed neither.

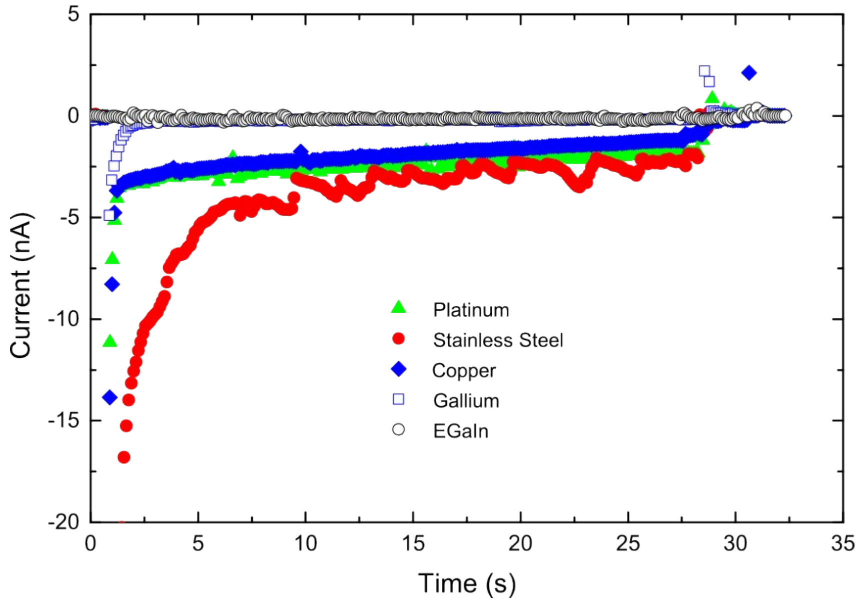

Fig. S3: Short-circuit current data for each top electrode. Current for a drop of DI H_2O on a smooth film of EGaIn with the circuit closed for approximately 30 seconds.

Long-term Closed Circuit: We performed long-term experiments to demonstrate that the wetting without an external potential could be sustained for a long period of time (hours, rather than minutes or seconds). We utilized DI H_2O in the experiments, because the H_2O/SDS solution separates too quickly from the top electrode when short-circuited. This experiment is noteworthy for several reasons: first, it shows that the wetting can be maintained for at least one hour, reversibly; although the droplet initially pins on the surface when the voltage is removed, the initial contact angle can be restored by moving the droplet back and forth on the substrate. By closing the circuit again, in the same location, the droplet can once again decrease its contact angle. Additionally, this shows what appears to be oxide growth on the surface, indicating that the short-circuiting is causing dielectric breakdown.

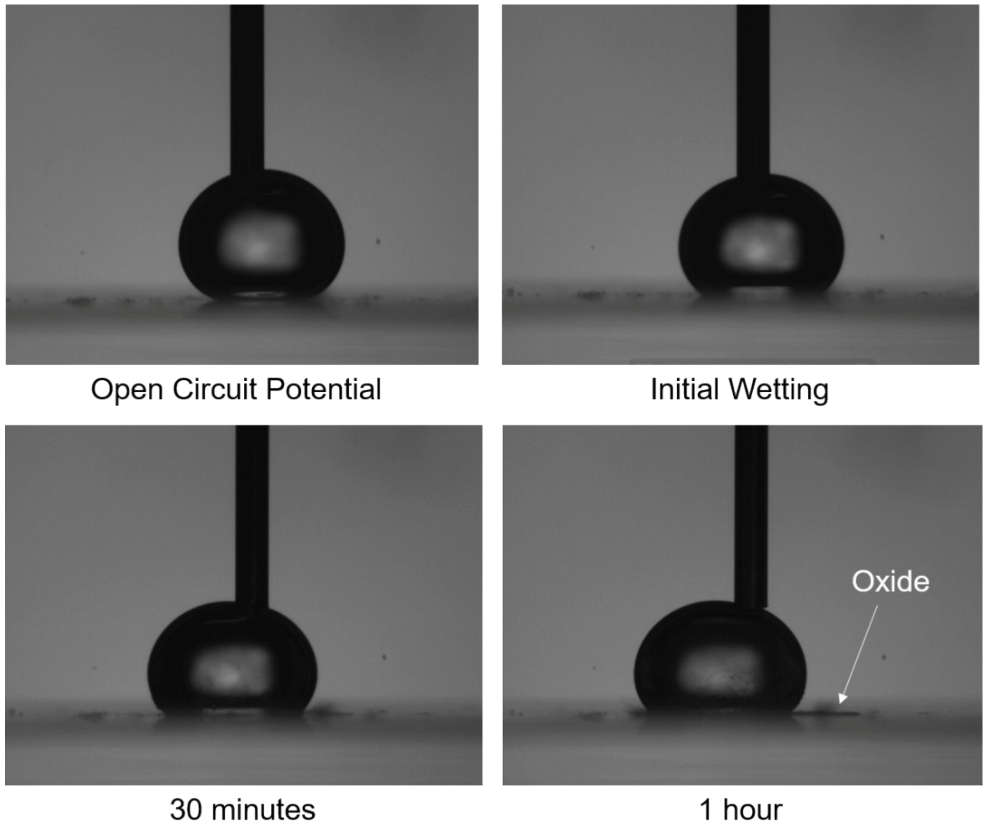

Fig. S4: Long-term wetting. Droplet of DI H_2O on a smooth film of EGaIn. The images show the progress of the drop at open circuit potential, the initial wetting after the circuit is closed (after approximately 2 seconds), after 30 minutes, and after 1 hour.

AC Electrowetting (Supplementary Movie S1)

Droplet of DI H_2O/1 wt% SDS is dispensed from a 310 μm stainless steel syringe needle in a solution of dodecane/0.2 wt% sorbitan trioleate on a liquid metal substrate. An AC potential is applied (750 mV, 100 Hz sine waveform) with a -800 mV DC offset. Droplet dramatically changes contact angle against the liquid metal. When the potential is removed, the contact angle returns to its original state.

Electrowetting without external potential (Supplementary Movie S2)

Droplet of DI H_2O/1 wt% SDS is dispensed from a 310 µm stainless steel syringe needle in a solution of dodecane/0.2 wt% sorbitan trioleate on a liquid metal substrate. The substrate is connected electrically to the top electrode, causing the droplet to immediately decrease its contact angle and form satellite droplets on the surface. Disconnecting the electrodes causes the droplet to bead back up.