

## Droplet-Based Interfacial Capacitive Sensing

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### 1. Mathematical Derivations for the Mechanical-to-Capacitive Sensitivity

#### Eq. S1 Mechanical Deformation

The overall deformation of the sensing chamber, which includes the deflection of the flexible membrane and the compression of the elastic separation layer, will lead to the change of the interfacial contact area, and therefore, result in variation of the capacitive measurements. More specifically, small deflections of circular membranes ( $y_m$ ) can be mathematically predicted according to the classic thin-plate theory,<sup>31</sup> while elastic deformation of the separation layer ( $y_l$ ) are well adapted to the linear strain-stress relationship. The overall mechanical deformation of the droplet ( $y$ ) is a sum of the membrane deflection and the elastic deformation of the separation layer under external force ( $F$ ).

$$y_m = \frac{3(1-\nu_m^2)R^2}{4\pi E_m T_m^3} F \quad (\text{S1a})$$

$$y_l = \frac{HF}{S_l E_l} \quad (\text{S1b})$$

where  $\nu_m$  and  $E_m$  represent Poisson ratio and Young's modulus of the sensing membrane and  $E_l$  is Young's modulus of the separation layer.  $R$  and  $T_m$  are the radius and thickness of the membrane, and  $H$  is the height of the sensing chamber.  $S_l$  shows the deformed region along the sensing chamber, an experimentally determined constant.

### Eq. S2 Deformation-Induced Capacitive Change

As the droplet shape deforms under the external mechanical load, the electrolyte-electrode contact area will vary, which is proportional to the change of the overall interfacial capacitance.

$$\Delta C_{EDL} = c_o \Delta A = c_o \left( \frac{V_d}{H-y} - \frac{V_d}{H} \right) \approx \frac{c_o V_d}{H^2} (y_m + y_l) \quad (S2)$$

Where  $c_o$  is the unit area capacitance of the EDL at the electrolyte-electrode interface and  $V_d$  indicates the volume of the electrolyte droplet.

Combining Eq. S1 and S2, the governing equation for the overall mechanical-to-capacitive sensitivity of the droplet sensors can be established as Eq.1 and the two mechanical deformation constants  $\alpha$  and  $\beta$  in Eq.1 can be derived as follow:

### Eq. 1 Mechanical-to-Capacitive Sensitivity

$$\frac{\Delta C_{EDL}}{P} = c_o \alpha \left( \frac{V_d R^2}{H^2} \right) \beta \quad (1)$$

$$\alpha = \frac{3(1\nu_m^2)}{4E_m T_m^3} \quad (a)$$

$$\beta = \frac{\pi}{S_1 E_1} \quad (b)$$

## 2. Evaluation of droplet relaxation

The transient response of the droplet was characterized by applying and removing an external load and monitoring the membrane and the droplet shape change through a high-speed camera (1,200 fps).

**Fig. S1** Snapshots of the droplet relaxation process, recovering to a unstressed state from being deformed.

