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Electronic Supplementary Information [ESI]

Breakups of encapsulated surfactant-laden aqueous droplet under DC electric field

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In the ESI, we provide details regarding the calculation of the electric and capillary stresses acting on the core droplet. Here, we also provide measured interfacial tension (γ_{12}) values corresponding to non-dimensional surfactant concentration (C^*).

We consider a spherical core droplet of radius r_c that is encapsulated by an outer shell of radius r_s $(r_c = \beta r_s)$ and suspended in immiscible ambient liquid under a direct-current (DC) electric field. The stresses acting on point P ($\theta = 0$) at the core interface are analyzed.

Calculation of electric stress

The DC electric field is applied to the double-emulsion droplet. Owing to the discontinuity of the electrical properties, electric stresses are generated at the interfaces, which are given by the Maxwell stress tensor.

$$\sigma_M = \varepsilon(\vec{E}\vec{E} - E^2 I/2) \tag{S1}$$

The increase in the stresses at the core interface can be conveniently represented by normal and tangential stress components:

$$\left\|(\sigma_M \cdot \vec{n}) \cdot \vec{n}\right\| = \left\|\varepsilon(\vec{E} \cdot \vec{n})^2 - \varepsilon(\vec{E} \cdot \vec{t})^2\right\| \tag{S2}$$

$$\left\|(\sigma_M \cdot \vec{n}) \cdot \vec{t}\right\| = \left\|\varepsilon(\vec{E} \cdot \vec{t})^2\right\|. \tag{S3}$$

Here, $\|A\|$ indicates an increase in A, and $\vec{n}$ and $\vec{t}$ represent unit vectors normal and tangential to the interface, where $\vec{n} \equiv r$ and $\vec{t} \equiv \theta$.

$$\left\|(\sigma_M \cdot \vec{n}) \cdot \vec{n}\right\|_{12} = \left(\varepsilon_2 {E_{n2}}^2 - \varepsilon_1 {E_{n1}}^2\right)/2 - \left(\varepsilon_2 {E_{t2}}^2 - \varepsilon_1 {E_{t1}}^2\right) \tag{S4}$$

$$\left\|(\sigma_M \cdot \vec{n}) \cdot \vec{t}\right\|_{12} = \left(\varepsilon_2 E_{t2} E_{n2} - \varepsilon_1 E_{t1} E_{n1}\right) \tag{S5}$$

Here, $E_n = \partial\phi/\partial n$ and $E_t = \partial\phi/\partial t$. According to the continuity of the normal component of the electric current density and the continuity of the tangential component of the electric field at the interface, $E_{n1} = E_{n2}/R_{12}$ and $E_{t1} = E_{t2}$, respectively. Then, the above equations can be simplified as follows:

$$\|(\sigma_M \cdot \vec{n}) \cdot \vec{n}\|_{12} = \varepsilon_2(1 - S_{12}/R_{12}){E_{n2}}^2/2 - (1 - S_{12}){E_{t2}}^2 \qquad \text{(S6)}$$

$$\|(\sigma_M \cdot \vec{n}) \cdot \vec{t}\|_{12} = \varepsilon_2(1 - S_{12}/R_{12})E_{t2}E_{n2}. \qquad \text{(S7)}$$

The electric-field components are determined by the gradient of the electric potential (ϕ) in the spherical coordinate system:

$$\vec{E}_i = -\left(\hat{r}\frac{\partial}{\partial r} + \hat{\theta}\frac{\partial}{r\partial\theta}\right)\phi_i, \qquad \text{(S8)}$$

where i = 1, 2, and 3 for the core, shell, and ambient liquids, respectively.

The electric potential (ϕ_i) for each phase is obtained by solving the governing Laplace equation $(\nabla\phi_i = 0)$ of the charge conservation with the following boundary conditions: (1) $\phi_1(r_c,\theta) = \phi_2(r_c,\theta)$; (2) $\phi_2(r_s,\theta) = \phi_3(r_s,\theta)$; (3) $\sigma_1\partial\phi_1/\partial r(r_c,\theta) = \sigma_2\partial\phi_2/\partial r(r_c,\theta)$; (4) $\sigma_2\partial\phi_2/\partial r(r_s,\theta) = \sigma_3\partial\phi_3/\partial r(r_s,\theta)$; (5) $\phi_1(0,\theta)$ is bounded; (6) $\phi_3(r,\theta) = E_o r cos\theta, where\ r\rightarrow\infty$.

$$\phi_1 = 9\xi r E_o cos\theta, \qquad \text{(S9a)}$$

$$\phi_2 = 3\xi(R_{12} + 2)\left[(r/r_s) - \varsigma(r_s/r)^2\right]r_s E_o cos\theta \qquad \text{(S9b)}$$

Here,

$$\xi = 1/[(R_{12} + 2)(R_{23} + 2) + 2\beta^3(R_{12} - 1)(R_{23} - 1)],\ \zeta = \beta^3(R_{12} - 1)/(R_{12} + 2). \qquad \text{(S10)}$$

Substituting Eq. (S9) into Eq. (S8) and substituting the resulting equation into Eqs. (S6) and (S7) yields the electric stress components at point P:

$$F_E = \|(\sigma_M \cdot \vec{n}) \cdot \vec{n}\|_{12} = 81\xi^2 S_{23}({R_{12}}^2 - S_{12})\varepsilon_3 {E_o}^2/2 \quad \text{(S11)}$$

$$\|(\sigma_M \cdot \vec{n}) \cdot \vec{t}\|_{12} = 0. \quad \text{(S12)}$$

Calculation of capillary stress

Initially, when there is no electric field, the core droplet retains a spherical shape, and the capillary stress is given by the Young–Laplace equation:

$$F_i = 2\gamma_{12}/r_c. \quad \text{(S13)}$$

When the electric field is applied, the core droplet deforms, and the capillary stress becomes

$$F_f = 2\gamma_{12}(1/r_{c1} - 1/r_{c2}). \quad \text{(S14)}$$

where r_{c1} and r_{c2} represent the principal radii of curvature of the core droplet. The equation of the deformed surface of the droplet is given as

$$\bar{R} = r\left\{1 + \frac{2}{3}D(3\cos^2\theta - 1)\right\},\quad D = \frac{(\alpha - \bar{\alpha})}{(\alpha + \bar{\alpha})}. \quad \text{(S15a,b)}$$

Here, α and $\bar{\alpha}$ represent the semi-major and semi-minor axes of the deformed core droplet, respectively.

The principal radii of curvature of the deformed core droplet (ellipsoid) are given as

$$r_{c1} = \frac{\beta^2}{\alpha} \text{ and } r_{c2} = \frac{\alpha^2}{\beta}.$$

Using Eqs. (S17a) and (S17b), for $D \ll 1$, the following can be derived:

$$\left(\frac{1}{r_{c1}} + \frac{1}{r_{c2}}\right) = \frac{8}{\alpha} - \frac{6r_c}{\alpha^2}. \quad \text{(S16a)}$$

Thus, the change in the capillary stress can be expressed as

$$F_f - F_i = 2\gamma_{12}/r_c - \gamma_{12}\left(8/\alpha - 6r_c/\alpha^2\right). \quad \text{(S17)}$$

Measured interfacial tension values

We measured the interfacial tension between aqueous core-silicone oil (γ_{12}) at different surfactant concentration ($C_{Tween\ 80}$) represented in terms of non-dimensional surfactant concentration (C*). The results for few specific cases are provided in Table S1.

Table S1. Measured interfacial tension (γ_{12}) corresponding to non-dimensional surfactant concentration (C*)

Sr. No	**Non-dimensional Surfactant Concentration (C*)**	**Interfacial Tension (γ_{12}) [N/m^3]**
1	0	0.0353
2	0.05	0.0271
3	0.15	0.0189
4	1	0.0133