<u>Electronic Supporting Information (ESI)</u>

Mono-emulsion droplet stretching under direct current electric field

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In this section, we provide the details of the classification of flow types around an emulsion interface, simulation technique, theoretical model and the comparison between simulation and theoretical results.

1) Classification of various flow types

On the basis of the flow types around an emulsion interface under a direct current electric field, we can classify the emulsion deformation as prolate 'A', prolate 'B' and oblate that depends on permittivity ratio ($S = \varepsilon_d/\varepsilon_b$) and conductivity ratio ($R = \sigma_d/\sigma_b$), where the subscripts *d* and *b* denote the droplet liquid and bulk liquid, respectively, as shown in Fig. S1.

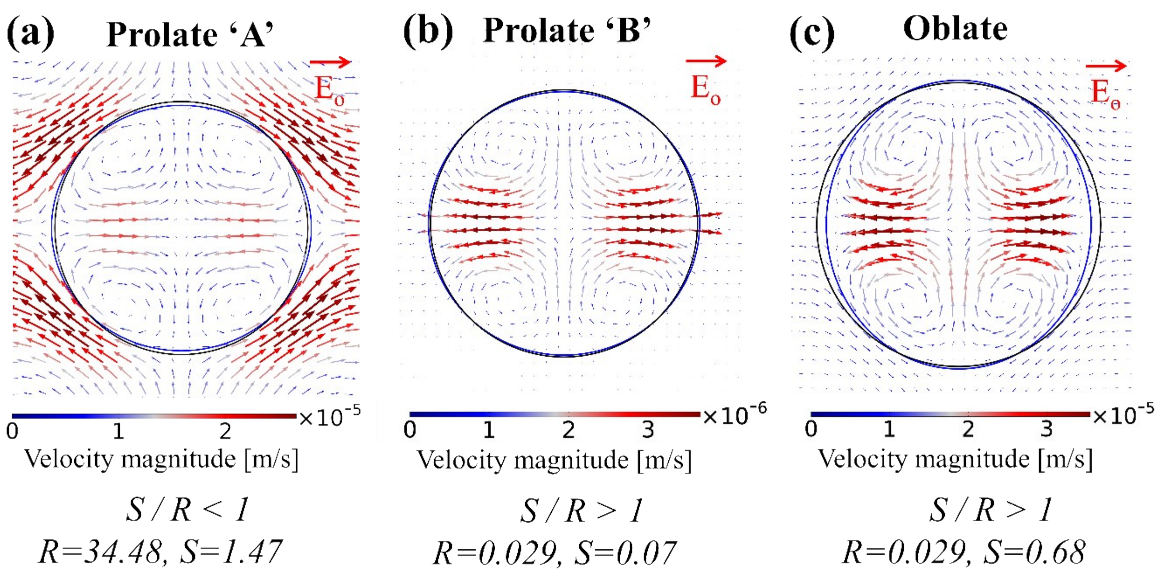

Fig. S1 Classification of various flow types obtained from numerical simulation. The black and blue lines show the initial and final droplet shapes, respectively.

It can be seen from Fig. S1 that if $S/R < 1$, the flow direction is from equator to pole and resulting shape of the droplet is always prolate. On the other hand, if $S/R > 1$, the flow direction is from the pole to equator and drop shape can be prolate or oblate.

2) Simulation Model

We solve the problem by coupling the Navier-Stokes momentum equation with the level set equation and governing equations of the electric field.

I. Problem Description

As shown in Fig. S2, a spherical droplet of radius r and diameter $a = 2r$, density ρ_d, viscosity μ_d, electrical permittivity ε_d and electrical conductivity σ_d is suspended in immiscible liquid of density ρ_b, viscosity μ_b, electrical permittivity ε_b and electrical conductivity σ_b. The interfacial tension between the liquids is represented by γ. The system is subjected to a direct current electric field, when a voltage ($\phi = \phi_\infty$) is applied to the supply electrode. To characterize the liquid properties across the interface, we define the property ratios as the following: $\Upsilon = \rho_d/\rho_b$, $\Gamma = \mu_d/\mu_b$, $S = \varepsilon_d/\varepsilon_b$ and $R = \sigma_d/\sigma_b$

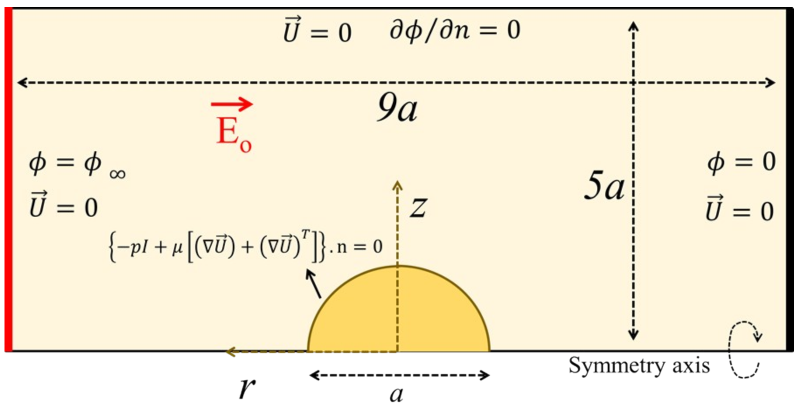

Fig. S2 Axisymmetric cylindrical domain used in the numerical simulation. Boundary conditions for fluid flow and electric field are specified

To model the problem, we considered a two-dimensional axisymmetric cylindrical domain. To avoid the effects of domain truncation on the droplet deformation, the radius and height of the numerical domain were fixed to be nine and five times greater than the diameter of the droplet a , respectively. We applied the no slip boundary condition ($\vec{U} = 0$) to all the walls. The bottom of the domain is the axis of symmetry. The left side of the domain is the supply electrode ($\phi = \phi_\infty$) while the side right of the domain is grounded ($\phi = 0$). The top of the domain was electrically insulated ($\partial\phi/\partial n$). At the droplet interface, we specified the initial fluid interface$\left[\{-pI + \mu[(\nabla\vec{U}) + (\nabla\vec{U})^T]\}.n = 0\right]$.

II. Governing Equations

A. Level set method

The interface between the two liquids is evolved by a fixed contour of a level set function $\xi = 0.5$ in the level set equation expressed as

$$\frac{\partial\xi}{\partial t} + \nabla\cdot(\vec{U}\xi) = \chi\nabla\cdot\left(\Psi\nabla\xi - \xi(1-\xi)\frac{\nabla\xi}{|\nabla\xi|}\right) \tag{S1}$$

Here, $\vec{U}$ is the velocity field inside the domain and ψ is the parameter that controls the thickness of the interface, χ is the stability parameter known as the re-initialization parameter and $\xi(1-\xi)\frac{\nabla\xi}{|\nabla\xi|}$ is known as the artificial flux. The term in the Laplacian in equation (S1) can be considered of as a diffusion term that is trying to enlarge with the interface width. This is countered by the flux divergence. The thickness of the interface ψ is maintained by the equilibrium of these two terms

For this two phase fluid system, the density ρ, viscosity μ, conductivity σ and permittivity ε are represented by the function of level set variable ξ expressed as

$$\rho = \xi(\rho_d - \rho_b) + \rho_b \tag{S2a}$$

$$\mu = \xi(\mu_d - \mu_b) + \mu_b \tag{S2b}$$

$$\sigma = \xi(\sigma_d - \sigma_b) + \sigma_b \tag{S2c}$$

$$\varepsilon = \xi(\varepsilon_d - \varepsilon_b) + \varepsilon_b \tag{S2d}$$

The value of the level set variable ξ is "1" for the outside liquid represented by subscript "b" and changes to "0" for the droplet liquid represented by subscript "d".

B. Fluid motion equations

The Navier-Stokes equation is coupled with the Level set equation in conjunction with the applied electric field and electric stresses to study the dynamics of the mono-droplet under different electric field strengths. The flow is assumed incompressible and the flow regime is laminar. The incompressible Navier-Stokes equation is expressed as

$$\rho\left(\frac{\partial\vec{U}}{\partial t} + (\vec{U}\cdot\nabla)\vec{U}\right) = \nabla\{-pI + \mu[(\nabla\vec{U}) + (\nabla\vec{U})^T]\} + \vec{F}_{st} + \vec{F}_e \tag{S3a}$$

$$\nabla\cdot\vec{U} = 0 \tag{S3b}$$

Here, F_{st} is the surface tension force whereas F_e is the volumetric force due to Maxwell stresses.

C. Leaky dielectric model

When both liquids are loosely conducting , R ($=\sigma_d/\sigma_b$)~ $O(10)$, subsequently, under the assumption of a quasi-electrostatic field, the electric field can be separated from the magnetic field and the conservation of free charge can be written as

$$\frac{\partial q_f}{\partial t} + \nabla \cdot \vec{J}_i = 0 \tag{S4}$$

Here, it is assumed that both the droplet and the outside liquids follow the Ohmic conduction law, and the current density $\vec{J}_i$ is related to the electric field $\vec{E}$ by $\vec{J}_i = \sigma \vec{E}$ where σ is the electrical conductivity as defined by equation S2c. By Gauss law, we can obtain the following equation

$$\nabla \cdot \left(\varepsilon_o \varepsilon \frac{\partial \vec{E}}{\partial t} + \sigma \vec{E} \right) = 0 \tag{S5}$$

The electric field $\vec{E}$ is defined in terms of the electric potential ϕ as

$$\vec{E} = \nabla \phi \tag{S6}$$

The electric force F_e can be calculated by taking the divergence of the Maxwell stress tensor σ_M

$$F_e = \nabla \cdot \sigma_M \tag{S7}$$

$$\sigma_M = \varepsilon_o \varepsilon (\vec{E} \cdot \vec{E} - E^2 I/2) \tag{S8}$$

For the two dimensional axisymmetric domain the stress tensor can be expanded as

$$\sigma_M = \varepsilon_o \varepsilon \begin{bmatrix} \{E_r^2 - (E_r^2 + E_z^2)/2\} & 0 & E_r E_z \\ 0 & (E_r^2 + E_z^2)/2 & 0 \\ E_r E_z & 0 & \{E_z^2 - (E_r^2 + E_z^2)/2\} \end{bmatrix} \tag{S9}$$

III. Benchmarking the simulation model

To benchmark the simulation model, we calculated the deformation at varying electric capillary number which is the ratio of the electric stress to the capillary stress for a castor droplet in silicone oil, and compared the results from Taylor's theory and the experimental results. Here, Taylor's deformation parameter is denoted by D, which is calculated by $D = \frac{(\alpha - \beta)}{(\alpha + \beta)}$, where α and β are the droplet semi-axis length parallel and perpendicular to the direction of the electric field, respectively.

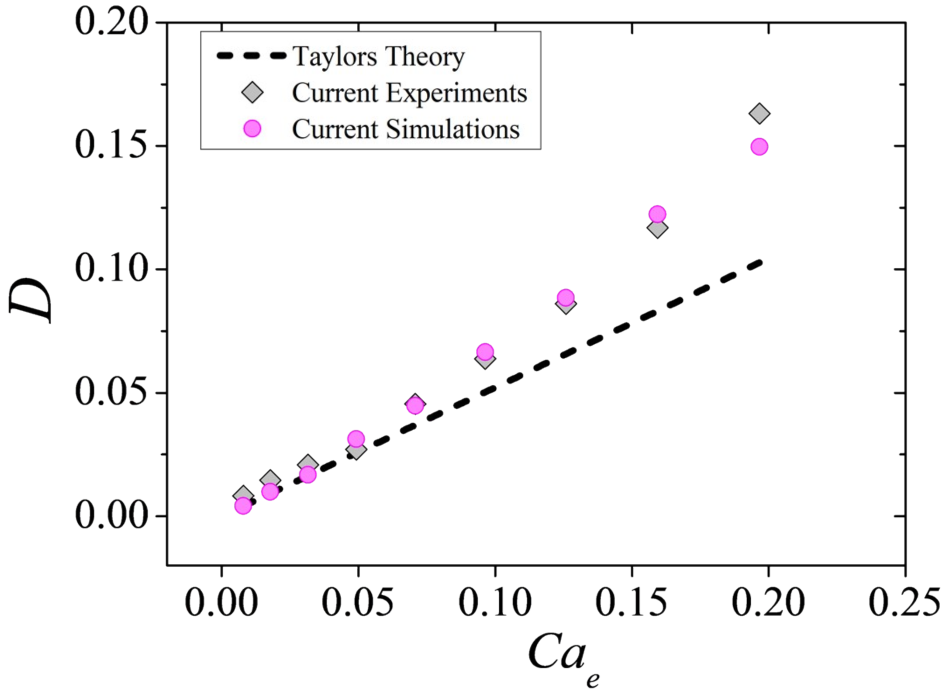

Fig. S3 Comparison between simulation and experimental results obtained for loosely conducting castor droplet in silicone oil under direct current electric field. Dashed line represents the first order asymptotic theory of Taylor[1, 2]

3) Theoretical Model

The radial and tangential component of electric stress acting on the droplet under AC electric field with frequency ω can be given as [3]

$$\sigma_{Mr} = \frac{9}{4}\varepsilon_o\varepsilon_2{E_o}^2\left[\frac{(R^2 - 2qR^2 + 1) + a^2\omega^2(q-1)^2}{(2R+1)^2 + a^2\omega^2(q+2)^2}\right]\cos^2\theta + \frac{9}{4}\varepsilon_o\varepsilon_2{E_o}^2\left[\frac{(q-1)(R^2 + a^2\omega^2)}{(2R+1)^2 + a^2\omega^2(q+2)^2}\right] \quad \text{(S10)}$$

$$\sigma_{M\theta} = \frac{9}{4}\varepsilon_o\varepsilon_2{E_o}^2\left[\frac{R(Rq-1)}{(2R+1)^2 + a^2\omega^2(q+2)^2}\right]\sin\theta\cos\theta \quad \text{(S11)}$$

Where, R = resistivity ratio = 1/conductivity ratio = 1/R. The above equations can be simplified in our case by putting $\omega = 0$

$$\sigma_{Mr} = \frac{9\varepsilon_o\varepsilon_b E_o^2}{4(2/R+1)^2}\left[\left(1/R^2 - \frac{2S}{R^2} + 1\right)cos^2\theta + 1/R^2(S-1)\right] \quad \text{(S12)}$$

$$\sigma_{M\theta} \sim \frac{\varepsilon_o\varepsilon_b E_o^2}{(2/R+1)^2}\left[1/R(\frac{S}{R} - 1)\right]sin\theta cos\theta. \quad \text{(S13)}$$

The radial hydrodynamic stress components in the two liquids (droplet and ambient) are given as [1]

$$\sigma_{Hrd} = \frac{A}{r}5\mu_d(1 - 3cos^2\theta), \quad \sigma_{Hrb} = \frac{A}{r}3\mu_b(1 - 3cos^2\theta) \quad \text{(S14a,b)}$$

The radial stress jump can be given as

$$\sigma_{Hrd} - \sigma_{Hrb} = \frac{A}{r}5\mu_d(1 - 3cos^2\theta) + \frac{A}{r}3\mu_b(1 - 3cos^2\theta) \quad \text{(S15)}$$

When the droplet is un-deformed, the capillary stress across the interface is given by the Young-Laplace equation as

$$P_{undeformed} = P_{in} - P_{out} = \frac{2\gamma}{r} \quad \text{(S16a)}$$

Here, r is the radius of un-deformed droplet. As, the droplet stretches, its capillary stress given by the Young-Laplace equation becomes

$$P_{deformed} = P_{in} - P_{out} = \gamma\left(\frac{1}{r_1} + \frac{1}{r_2}\right) \tag{S16b}$$

where, r_1 and r_2 are the principal radii of curvature of the deformed droplet. The capillary stress difference between the un-deformed and deformed configuration can be expressed as

$$\Delta P_\gamma = \frac{2\gamma}{r} - \gamma\left(\frac{1}{r_1} + \frac{1}{r_2}\right) \tag{S16c}$$

The equation of the deformed surface of the droplet is given as [3]

$$\bar{R} = r\left\{1 + \frac{2}{3}D\left(3\cos^2\theta - 1\right)\right\},\quad D = \frac{(\alpha - \beta)}{(\alpha + \beta)} \tag{S17a,b}$$

Here, α and β are the semi- major and semi- minor axis of the deformed droplet.

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

$r_1 = \frac{\beta^2}{\alpha}$ and $r_2 = \frac{\alpha^2}{\beta}$

Using equation (S17a,b), $\alpha = r + \frac{4}{3}rD$ and $\beta = r - \frac{2}{3}rD$ and for $D \ll 1$, it can be derived that

$$\left(\frac{1}{r_1} + \frac{1}{r_2}\right) = \frac{8}{\alpha} - \frac{6r}{\alpha^2} \tag{S18a}$$

It can also be derived that

$$\left(\frac{1}{r_1} + \frac{1}{r_2}\right) = \frac{40}{6r - \beta} - \frac{150r}{(6r - \beta)^2} \tag{S18b}$$

Thus, the equation (S16c) takes the form as

$$\Delta P_{\gamma} = 2\gamma\left[\frac{1}{r} - \left(\frac{4}{\alpha} - \frac{3r}{\alpha^2}\right)\right] \tag{S19}$$

4) Comparison between simulation and theoretical model results

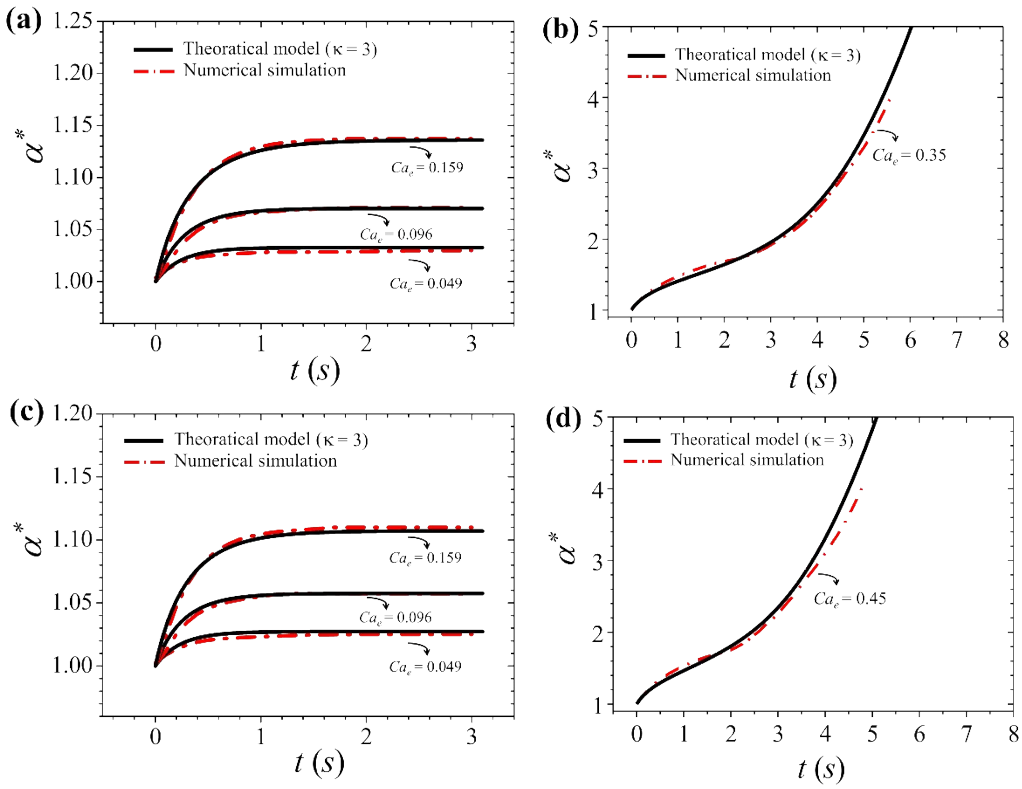

Fig. S4 Comparison between simulation and theoretical model (equation (12)) results. (a-b) $(\Gamma, S, R) = (1, 1, 20)$, (c-d) $(\Gamma, S, R) = (1, 0.1, 10)$, where Γ, S and R are the viscosity, conductivity and permittivity ratios between droplet and bulk liquids.

We consider two case studies of droplet stretching under direct current electric field adopted from a reference [4] for a comparison between simulation and theoretical model (equation (12)) results. The physical parameters for the two case studies are $(\Gamma, S, R) = (1, 1, 20)$ and $(\Gamma, S, R) = (1, 0.1, 10)$, respectively, where $\Gamma, S\ and\ R$ are the viscosity ratio, permittivity ratio and conductivity ratio between droplet and bulk liquid. The results for equilibrium (Fig. S4a and Fig. S4c) and non-equilibrium (Fig. S4b and Fig. S4d) degree of deformation for both the cases are as shown in Fig. S4

References

(1) G. Taylor, *Proceedings of the Royal Society of London. Series A. Mathematical and Physical Sciences*, 1966, 291, 159-166.

(2) J. Melcher and G. Taylor, *Annual review of fluid mechanics*, 1969, 1, 111-146.

(3) S. Torza, R. Cox and S. Mason, *Phil. Trans. R. Soc. Lond. A*, 1971, 269, 295-319.

(4) R. Sengupta, L. M. Walker and A. S. Khair, *Journal of Fluid Mechanics*, 2017, **833**, 29-53.