

# Supplementary Information:

## Voltage-Driven Ion Flux Promotes Emulsification at the Water|Oil Interface:

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Figure S1: Glass cell Images of a two-phase system over the course of 5 min. A 3 mL aqueous top layer containing 0.1 M  $[\text{NBu}_4][\text{Cl}]$  was pipetted over 3 mL of DCE containing 0.1 M  $[\text{NBu}_4][\text{PF}_6]$ . No emulsification was observed after 5 minutes of the two phases being in the presence of one another.

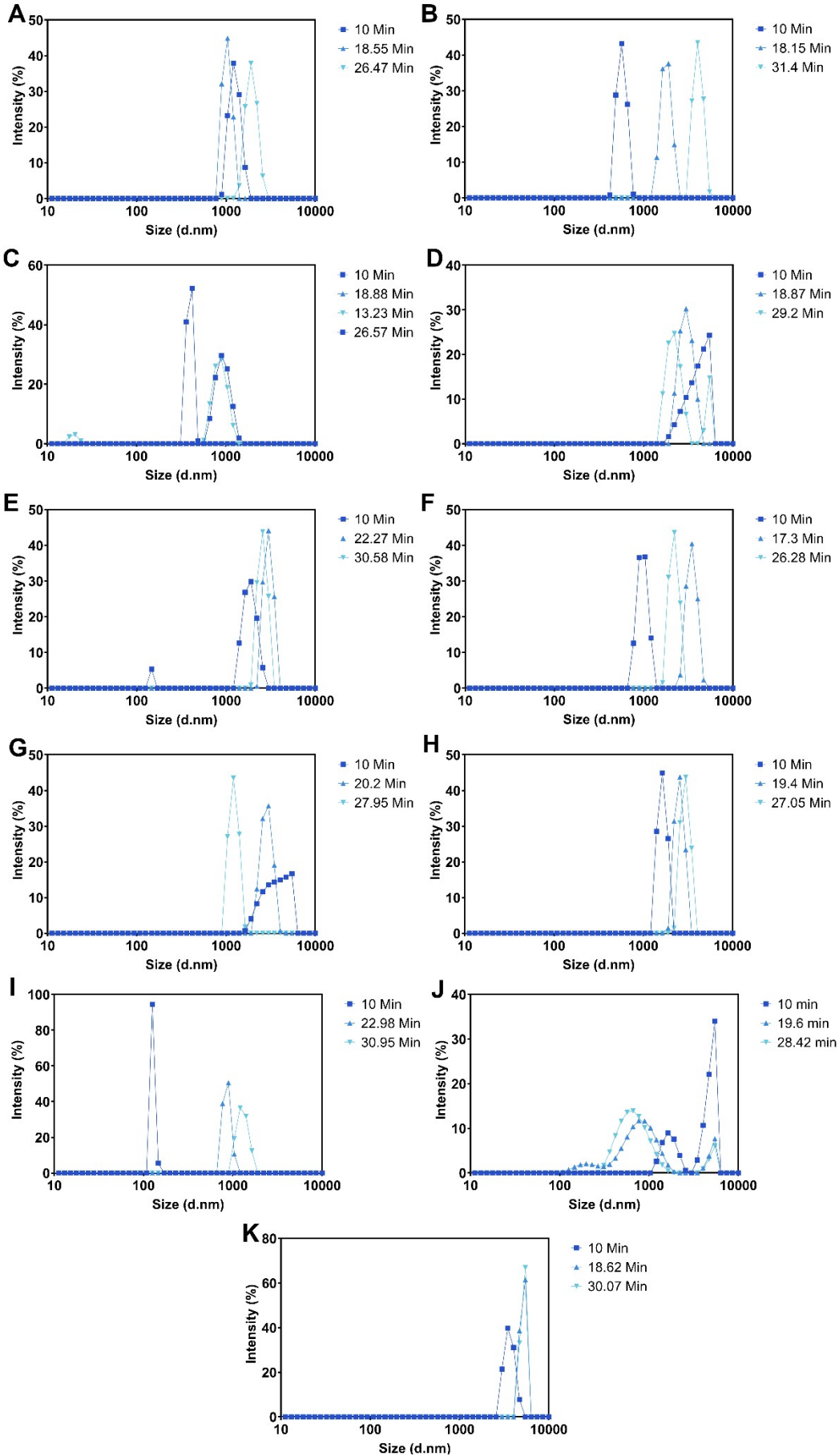
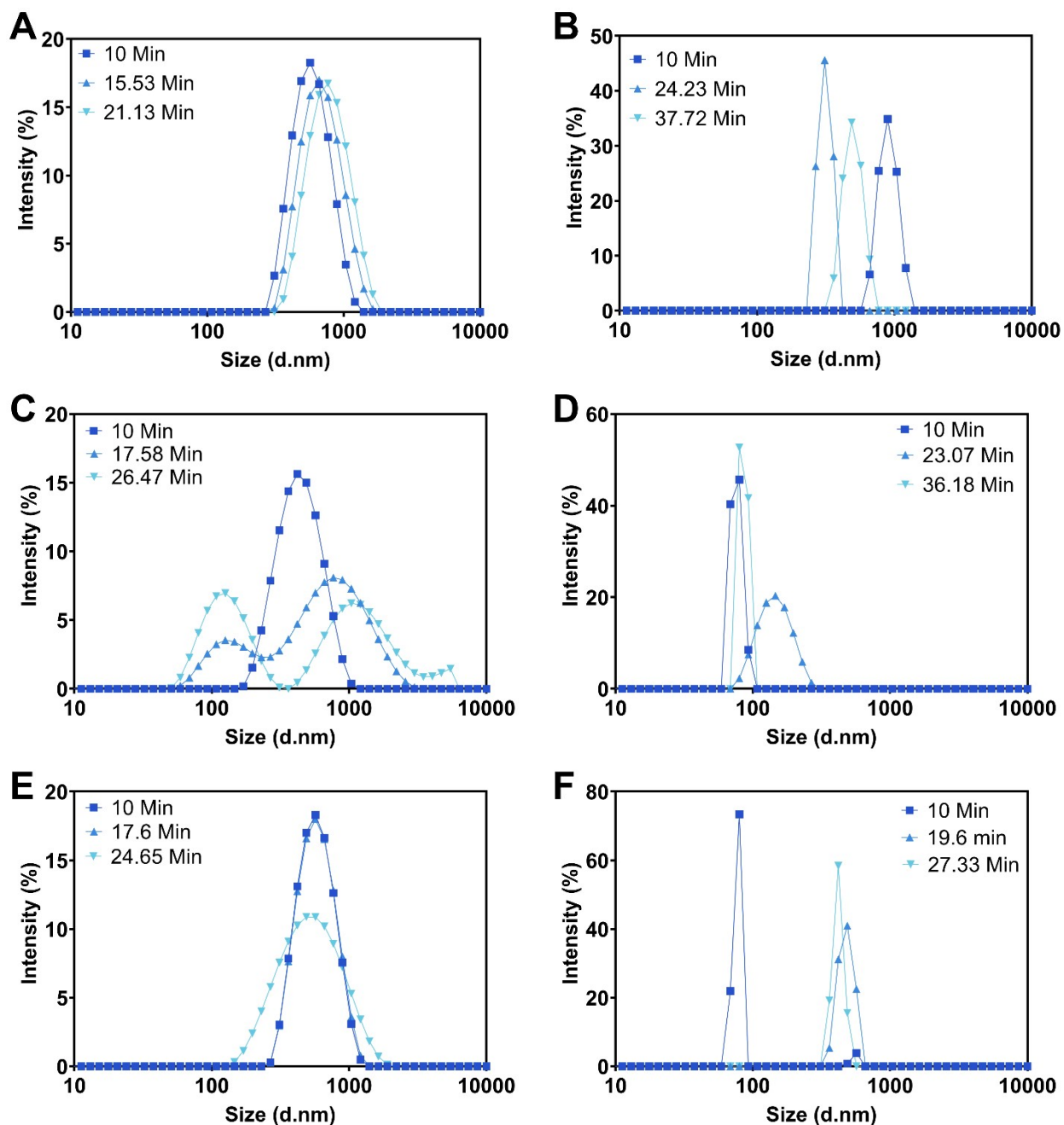


Figure S2: Dynamic light scattering measurements of aqueous droplets formed in an organic phase, when a 5 mA current is applied on a two-phase system over the course of 5 min using a Pt wire ( $r = 1$  mm). Data shown corresponds with cases shown in Table 1. A) The anode was placed in an aqueous solution containing 0.1 M  $[\text{Nbu}_4][\text{Cl}]$ , and cathode was placed in an organic (DCE) solution containing 0.1 M  $[\text{Nbu}_4][\text{PF}_6]$ , B) The Cathode was placed in an aqueous solution containing 0.1 M  $[\text{Nbu}_4][\text{Cl}]$ , and anode was placed in an organic (DCE) solution containing 0.1 M  $[\text{Nbu}_4][\text{PF}_6]$ , C) The Cathode was placed in an aqueous solution containing no salt, and anode was placed in an organic (DCE) solution containing 0.1 M  $[\text{Nbu}_4][\text{PF}_6]$ , D) The anode was placed in an aqueous solution containing no salt, and cathode was placed in an organic (DCE) solution containing 0.1 M  $[\text{Nbu}_4][\text{PF}_6]$ , E) The Cathode was placed in an aqueous solution containing 1M NaOH, and anode was placed in an organic (DCE) solution containing .01 M  $[\text{Nbu}_4][\text{PF}_6]$ , F) The anode was placed in an aqueous solution containing 1M NaOH, and cathode was placed in an organic (DCE) solution containing 0.1 M  $[\text{Nbu}_4][\text{PF}_6]$ , G) The anode was placed in an aqueous solution containing 1M  $\text{H}_2\text{SO}_4$ , and cathode was placed in an organic (DCE) solution containing 0.1 M  $[\text{Nbu}_4][\text{PF}_6]$ , H) The Cathode was placed in an aqueous solution containing 1M  $\text{H}_2\text{SO}_4$ , and anode was placed in an organic (DCE) solution containing 0.1 M  $[\text{Nbu}_4][\text{PF}_6]$ , I) The Cathode was placed in an aqueous solution containing no salt, and anode was placed in an organic (DCE) solution containing 0.1 M  $[\text{Nbu}_4][\text{TPB}]$ , J) The Cathode was placed in an aqueous solution containing no salt, and anode was placed in an organic (DCE) solution containing 0.1 M  $[\text{Nbu}_4][\text{Br}]$ , K) The Cathode was placed in an aqueous solution containing no salt, and anode was placed in an organic (DCE) solution containing 0.1 M  $[\text{NH}_4][\text{ClO}_4]$ . DLS measurements were taken 3 times, unless specified, for 60 runs at 1.64s/run, with a 120s equilibration time, and a equilibration temperature of 25 °C, as per instrument suggestion.



**Figure S3:** Dynamic light scattering measurements of aqueous droplets formed in various organic phases, when a 5 mA current is applied on a two-phase system over the course of 5 min using a Pt wire ( $r = 1$  mm). Data shown corresponds to two-phase electro emulsification with variations on the organic solvent used. Here the cathode was placed in an aqueous solution containing no salt, and the anode was placed in an organic solution containing 0.1 M  $[\text{Nbu}_4][\text{PF}_6]$ . DLS measurements for the water and oil phases can be seen for 1,2-Dichloroethane (A, B respectively), Chloroform (C, D respectively) and methylene chloride (E, F respectively). DLS measurements were taken 3 times, unless specified, for 60 runs at 1.64s/run, with a 120s equilibration time, and a equilibration temperature of 25 °C, as per instrument suggestion.