Electronic Supplementary Material (ESI) for Soft Matter.
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Electronic Supplementary Material for

Electrorotation of particle-coated droplets: from fundamentals to applications

Z. Rozynek,[a,b*] J. Banaszak,[a] A. Mikkelsen,[a] K. Khobaib,[a] and A. Magdziarz[a]

[a]Faculty of Physics, Adam Mickiewicz University, Uniwersytetu Poznańskiego 2, 61-614 Poznań, Poland
[b]PoreLab, The Njord Centre, Department of Physics, University of Oslo, Blindern, N-0316 Oslo, Norway
*Corresponding author: zbiroz@amu.edu.pl

Figure S1: At weak E-fields (up to around 250 V·mm^{-1}), the deformation magnitude deformation increased as power law of the electric field strength with an exponent slightly above 2. This is particularly apparent for larger droplets (with a diameter above 1 mm) that deform more than smaller droplets. During the droplet deformation, the surface area increases, thus more charges may accumulate on the droplet's interface contributing to the increase of the total electric stress acting on the droplet. At stronger electric field strengths (above around 400 V·mm^{-1}), the magnitude of droplet deformation is roughly proportional to E^2.

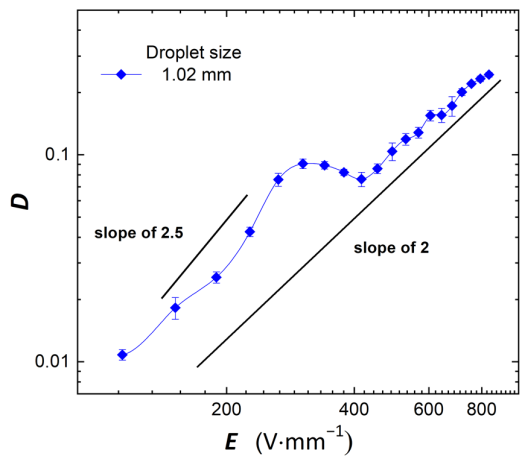


Fig. S1. A log-log plot of the deformation magnitude vs. the *E*-field strength.

Figure S2: It appears that the data for droplets smaller than 0.85 mm collapse and follow $\omega^2 \propto E^2/E_{cr}^2$ relationship. Though, the droplet rotation rate at high field strength (i.e., $E^2 \gg E_{cr}^2$) goes through a non-monotonic transition with increasing droplet size (above 0.72 mm).

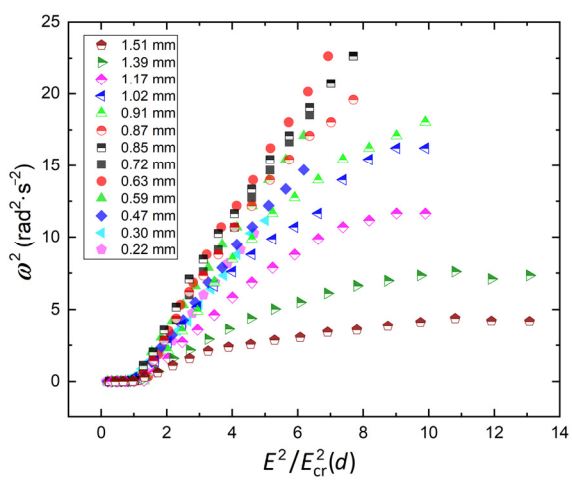
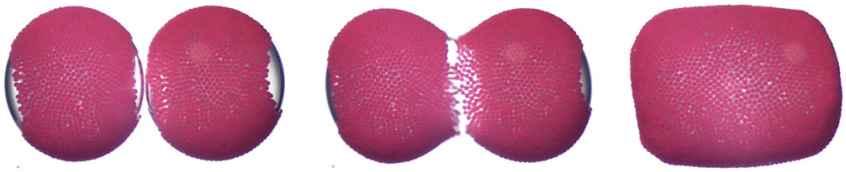


Fig. S2. Re-plotted data series presented in **Fig. 3a,b**. The argument values for each data series were normalized by the corresponding value of E_{cr} taken from panel **Fig. 3c**.

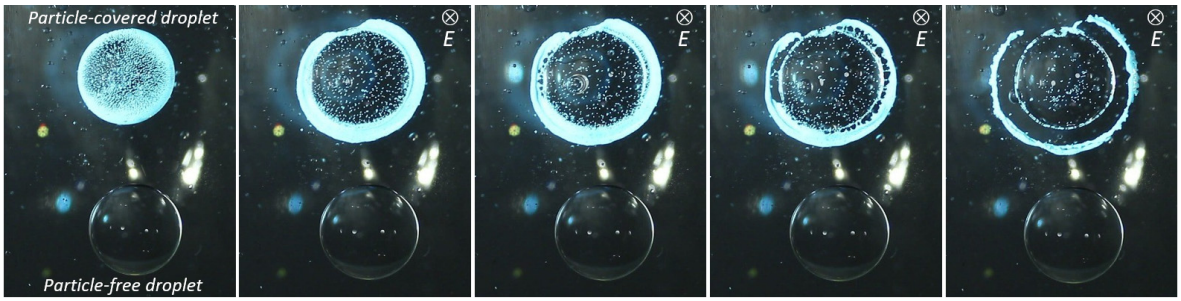
Figure S3: Two silicone oil droplets partly covered by 50-µm PE particles (left panel) coalesced to form an arrested Pickering droplet (right panel). The coalesced droplet had a jammed-particle layer that kept the droplet non-spherical even after the *E*-field was turned off.



t = 0 s *t* = 0.5 s *t* = 20 s

Fig. S3. (left) Two silicone oil droplets just before electrocoalescence and (right) the resulting coalesced droplet. The diameter of each original droplet was ∼1.5 mm.

Figure S4: Two silicone oil (50 cSt) droplets (particle-free and particle-covered) formed in castor oil. Application of a strong *E*-field of around 850 V·mm^{-1} resulted in irreversible detachment of particles from the droplet's surface. The droplets were viewed through an optically transparent ITO electrode along the *E*-field direction.



t = 0 s *t* = 1 s *t* = 1.2 s *t* = 1.5 s *t* = 2 s

Fig. S4. Detachment of surface microparticles from a silicone oil droplet (diameter of ~ 1.7 mm). Application of a strong *E*-field of around 850 V·mm^{-1} caused the particles to crumple and eventually irreversibly detach from the surface of the droplet. The droplet is imaged parallel to the direction of the *E*-field through transparent electrodes. See also the corresponding **Movie S7**.

Movie S1. Four PS particle-covered droplets (diameter 0.22–1.02 mm) at different *E*-field strengths (125–835 V·mm^{-1}).

Movie S2. Electrorotation of droplets (diameter of ~ 1.39 mm) with rigid and deformable shells composed of PE particles. The rigid shell was made by joining particles through heating using a microwave oven. During heating of a PE particle-covered droplet, hairy polymeric structures were formed sticking out of the shell. The droplet with a hairy shell rotated significantly slower than the droplet with a smooth shell owing to the increase of the magnitude of drag coefficient.

Movie S3. A silicone oil droplet (diameter of ~ 0.7 mm) covered by non-ohmic clay mineral particles (Li-fluorohectorite). Subjected to an *E*-field of ~ 270 V·mm^{-1} the droplet slowly rotated with at a rate of around 0.2 rad·s^{-1}. The rotation rate was increased by increasing the *E*-field strength up to ~ 300 V·mm^{-1}. However, at *E* ~ 330 V·mm^{-1} the clay particle-covered droplet stopped rotating. The dipole moment on the droplet covered by the clay film flipped at E ~ 330 V·mm^{-1}, and thus, the *E*-field acted on a droplet by stretching it along the E-field direction.

Movie S4. Electrorotation used for changing shape of a droplet and porosity of its particle shell. A droplet covered with 50-µm PE particles electrorotate in *E*-field of 350 and 600 V·mm^{-1}. The movie was sped up 4 times.

Movie S5. A droplet with diameter ∼2 mm covered with 50-µm PE particles (red and cyan) forming a Janus shell subjected to *E* = 700 V·mm^{-1}. The movie was sped up 4 times and then 10 times.

Movie S6. Breaking the droplet agglomerates down to individual droplets. Optical microscope images of silicone oil emulsions droplets covered by 10-µm PS particles. Castor oil was used as a continuous medium. An *E*-field of 200 V·mm^{-1} was applied to promote electrocoalescence of droplets, whereas a stronger *E*-field of 500 V·mm^{-1} was applied to induce electrorotation of droplets and droplet clusters to help the clustered droplets to separate. The electric field was applied in horizontal direction, and the movie was sped up two times.

Movie S7. Detachment of surface microparticles from a silicone oil droplet (diameter of ~ 1.7 mm). Application of a strong field of ~ 850 V·mm^{-1} caused the particle to crumple and eventually irreversibly detach from the surface of the droplet. The droplet is imaged parallel to the direction of the *E*-field through transparent electrodes.