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Transient deformation dynamics of particle laden droplets in electric field

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ABSTRACT

We study the transient deformation dynamics after application of uniform DC electric fields to particle laden droplets with different polyethylene particle coverage. Presence of interfacial particles result in reduced electrohydrodynamic circulation flows and charge convection, which in turn result in slower transient droplet deformation compared to pure uncovered droplets, and as well to increased steady-state droplet deformation. A prolate-oblate deformation transition is observed immediately after the application of the electric field. This anomaly is not visible or greatly reduced for droplets fully covered by particles.

1. Introduction

The dynamics of soft materials subjected to electric fields have lately attracted much attention in a variety of areas such as the dynamics of pendant and sessile droplets [1,2], electrorheological response in emulsions [3–5], vesicle manipulation [6–8] and colloidal particle manipulation at droplet interfaces [9–13]. Deformation of droplets plays key roles in many industrial applications and/or natural processes such as microfluidic systems (chemical reactors), combustion systems, electrohydrodynamic ink-jet printers, emulsification, mixing processes, biological cell systems or enhanced oil recovery [14–17].

It is energetically favourable for particles to bind at droplet interfaces, thus confining particle movement to within the interface. This has proven essential for a variety of studies and applications, including material development [10,18,19], emulsions stabilization [20–22], two-dimensional particle systems [23–25] and particle structuring [9,13,26]. The deformation of particle-free and particle laden droplets has previously been investigated by mechanical compression [27,28], microfluidics focusing devices [29], in hydrodynamic shear flows [30–33] and by magnetic or electric fields [9–12,34–38].

To understand the electric response of such complex systems, it is important to quantify the dynamics and time scales of simpler systems, for instance single droplets, for which the surface particle coverage φ is systematically increased from zero to full particle coverage. Here we study the transient deformation of weakly conductive droplets of silicone oil with different particle coverages. The electrohydrodynamic deformation of weakly conducting droplets without particles is described by Taylor's model [39], which has subsequently been developed by Melcher [40] and others [41–43]. The model is based on the assumptions that the two fluids have finite electric conductivities which yields a charge build-up at the droplet interface creating an interfacial electrical shear stress. In addition to a normal stress component balanced by the droplet surface tension, the electric stress has a tangential component that sets up viscous electrohydrodynamic flows.

Assuming that the deformations are small, and that the time required for the interface to acquire its steady-state surface charge density distribution, is much shorter than the convective time, the Taylor-model deformation is proportional to the applied electric field squared, and the droplet deformation is given by the electric properties of the fluids [39]:

$$D_E = \frac{d_{\parallel} - d_{\perp}}{d_{\parallel} + d_{\perp}} = \frac{9a\varepsilon_0\varepsilon_{\text{ex}}E_0^2}{16\gamma S(2+R)^2}\left[S(R^2+1) - 2 + 3(RS-1)\frac{2\lambda+3}{5\lambda+5}\right],$$

where $d_{\parallel}$ and $d_{\perp}$ respectively are the droplet axis parallel and perpendicular with the electric field direction, ε_0 the vacuum permittivity, ε_{ex} the relative dielectric constant of the surrounding exterior fluid, a the droplet radius, γ the interfacial surface tension between the droplet and exterior fluid, while the dimensionless numbers R, S and λ are the conductivity, dielectric constant and viscosity ratios, respectively: $R = \sigma_{\text{in}}/\sigma_{\text{ex}}$, $S = \varepsilon_{\text{ex}}/\varepsilon_{\text{in}}$, $\lambda = \mu_{\text{ex}}/\mu_{\text{in}}$. Dimensional and dimensionless parameters for our system (silicone oil suspended in castor oil) are listed in Table 1 in the Materials section.

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Table 1
Set of parameters for a silicone oil droplet suspended in castor oil. The top section of the table lists dimensional parameters for the droplet and the bath medium, while the bottom section lists dimensionless groups. The electric field is set to 200 V mm^{-1}.

Phase	ε_r	σ (S m^{-1})	μ (Pa s)	ρ (kg m^{-3})	a (mm)	γ (mN m^{-1})
Droplet (silicone oil)	2.8	5.6×10^{-12}	0.05	959	1.0	4.5
Medium (castor oil)	4.7	5.6×10^{-11}	0.75	960		
	Ca_E	Re_E	Sa_{ex}	S	λ	R
	0.4	1.3	4.5	1.7	15	0.1

Since Taylor's pioneering work on electrohydrodynamics [39], which is considering small deformations, several theoretical and computational investigations have been worked out to predict the transient deformation observed in experiments. Due to the challenges in coupling the interfacial charge distribution to the induced fluid flow, the transient charge relaxation and the charge convection driven by the interfacial flow are most often neglected in models. Taylor's model predicts both prolate and oblate steady-state deformations, but does not include charge convection and transient deformation effects. These effects are important to predict the right droplet shape evolution and to improve the accuracy of predictions [44,45].

For particle covered droplets, the electric properties of the surface particles are of importance, as they may suppress DC electric field induced electrohydrodynamic flows and stretch the droplet if the electrical conductivity of the particles is sufficiently high [9]. Suppression of electrohydrodynamic flows is also expected and observed in this case if the particle surface coverage is high (φ approximately or above 90%) [11,35]. For sufficiently low particle concentration, the particles form thin electric-equatorial ribbons at the droplet surface in applied DC electric field, however in this case there may still be circulation flows in the particle-free electric-polar areas. For fully covered droplets, a capsule type model has to be used, for example an elastic model [11] or a fluid shell description [35].

Here we investigate how surface particles influence transient droplet deformation by both weakening the charge convection and by strengthening the charge relaxation. We quantify the effects of surface particles on droplet deformation times, steady-state deformation and prolate-oblate anomalies for a range of droplet sizes, coverages and applied electric field strengths.

2. Material and methods

The current experiments were performed in an optical square acrylic cuvette (10 × 10 × 45 mm), with two copper plates constituting electrodes. The distance between the electrodes was 7.8 mm. Castor oil (Sigma-Aldrich 83912, density 0.961 g cm^{-3} at 25 °C, electrical conductivity ~60 pS m^{-1}, relative permittivity 4.7 at 25 °C, and viscosity 0.75 Pa s) was poured in the cuvette, and silicone oil (VWR Chemicals, Rhodorsil® 6678.1000, density 0.96 g cm^{-3}, electrical conductivity ~5-6 pS m^{-1}, relative permittivity 2.8 at 25 °C, and viscosity 0.05 Pa s) droplets without and with red polyethylene particles (REDPMS-0.98 45–53 µm, relative permittivity 2.1 and density ~0.98 g cc^{-1} purchased from Cospheric LLC) were placed inside the castor oil using a micropipette. The polyethylene particles were dispersed in the silicone oil, and the concentration was numerically characterized by weight percent. To avoid aggregation, the samples were placed in an ultrasonic bath for 5 min and mechanically shaken. There is a small density difference between the fluids, as well as between the fluids and the particles, however, the droplet sedimentation velocity was sufficiently small enough to enable us to neglect sedimentation effects. DC electric fields with strength between 0 and 300 V mm^{-1} were applied by connecting the electrodes to a high voltage amplifier (5HVA24-BP1Ultravolt®, Advanced Energy®), controlled by a voltage signal generator and monitored by an oscilloscope.

The droplet dynamics was studied and recorded through an IDS camera (UI-3590CP-C-HQ R2, IDS Imaging Development Systems GmbH) with magnifying lenses (Thorlabs, High-Magnification Zoom Lens Systems). Movies and images with a resolution of 1028 × 768 (XGA) and 50 fps framerate with uEye Cockpit software, were recorded.

The transient droplet deformations were estimated by analyzing 50 fps. The recorded movies were converted to frames by using a JPG converter software. In each frame, the edge of the droplets was recognized and fitted with an ellipse using ImageJ software. The axes of the droplets parallel and perpendicular to the electric field direction were measured from the fitting procedure. The droplet deformation is defined here as $D = (d_{\parallel} - d_{\perp})/(d_{\parallel} + d_{\perp})$, where $d_{||}$ and $d_{\perp}$ are the drop axes parallel and perpendicular to the electric field direction, respectively. We calculated the deformation for each frame and normalized it by the deformation before the electric field was turned on, by subtracting the average deformation of the first hundred frames. This latter procedure was necessary because the ImageJ software cannot distinguish the edge of the droplet from the edge of the surface particles and therefore measured some of the particle droplets to be slightly deformed (up to $D = 0.01$) even before the electric field was applied.

We define here the particle coverage of the droplets as $\varphi = S/A$, where A is the surface area of the droplet and $S = 2\pi ah$ [46] is the surface area of the particle ribbon film (a spherical segment defined by cutting a sphere with a pair of parallel planes), a the radius of the droplet and h the width of the ribbon. φ is defined as 1 when the droplet is fully covered by particles, and 0 when the droplet is particle-free.

3. Results and discussions

3.1. Transient deformation of particle laden droplets

We study the deformation of polyethylene laden silicone oil droplets that are suspended in castor oil and subjected to DC electric fields. Fig. 1 presents images of five silicone oil droplets (diameters 2.0 mm) with various polyethylene particle coverages, where the ratio between the particle coverage ranges between 0 and 1, all subjected to a DC electric field of strength 200 V mm^{-1}. When uniform DC electric fields are applied in this way, charges accumulate at the interface between the

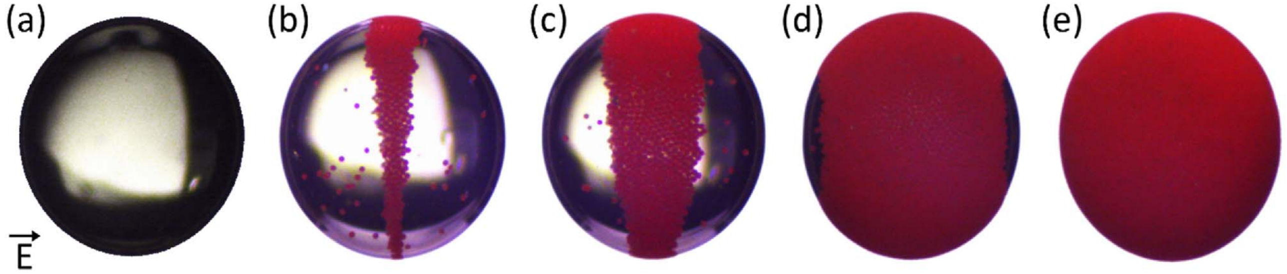

Fig. 1. Silicone oil droplets with different particle coverage and suspended in castor oil. Silicone oil droplets (diameters 2.0 mm) (a) without and (b)–(e) with red polyethylene particles subjected to a DC electric field of strength 200 V mm^{-1}. The particle coverage for the five droplets are respectively (a) 0, (b) 0.14, (c) 0.32, (d) 0.83 and (e) 1.