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Transient deformation dynamics of particle laden droplets in electric field

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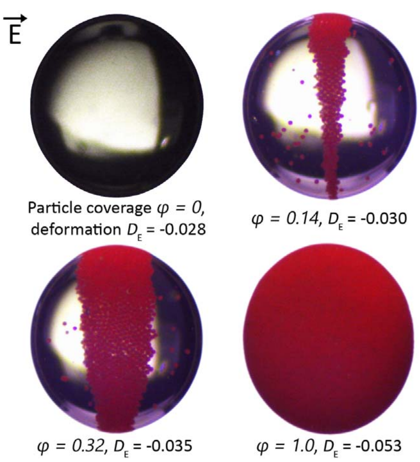
GRAPHICAL ABSTRACT



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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,

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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]:

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

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: $R = \sigma_{in}/\sigma_{ex}$, $S = \varepsilon_{ex}/\varepsilon_{in}$, $\lambda = \mu_{ex}/\mu_{in}$. Dimensional and dimensionless parameters for our system (silicone oil suspended in castor oil) are listed in Table 1 in the Materials section.

?A3B2 tlsb=-0.03w?>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.

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

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 $d_{\parallel}$ 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

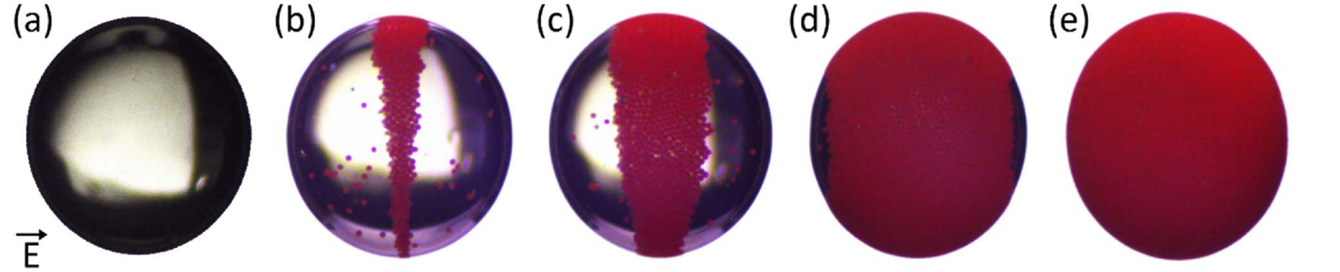

Fig. 1. Silicone oil droplets with different particle coverage and suspended in castor oil. Silicone oil droplets (diameters 2.0 mm) (a) without and (b)–(e) with red polyethylene particles subjected to a DC electric field of strength 200 V mm^{-1}. The particle coverage for the five droplets are respectively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

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 two liquids [39] due their contrast in electric conductivities and dielectric permittivity. Depending on the relative electric properties of the fluids and particles, such a droplet can be stretched to a prolate, or compressed to an oblate shape [9,38] when subjected to an applied electric field. The polarity of the induced droplet surface charge depends on the charge relaxation time $\tau_{e,in} = \varepsilon_0\varepsilon_{in}/\sigma_{in}$ of the droplet relative to that of the external fluid $\tau_{e,ex} = \varepsilon_0\varepsilon_{ex}/\sigma_{ex}$ [40,42]. Since the surrounding castor oil conducts better than the silicone oil droplet, the dipole moment of the droplets in Fig. 1 is aligned antiparallel with the electric field direction and the electric stress exerts a compressive force on the droplets, i.e. an oblate (negative) deformation occurs ($d_\perp > d_\parallel$).

When electric charges reach the droplet interface, electrohydrodynamic circulation flows are induced by tangential electric stresses. The direction of the circulation flows is given by the relative magnitudes of the electric conductivities and dielectric constants of the fluids, and the induced flows can transport surface particles to the droplet electric equator area or to the electric pole areas [39] respectively. In our present oil in oil system, the polyethylene particles are structured in ribbons at the droplet surfaces where the particle packing increases with time and also with the applied electric field strength [36]. In the deformation experiments described in the following, we limited the applied electric field to 300 V mm^{-1} to avoid effects such as electrically induced Quincke rotation [11,12,47], droplet breakup [38,48,49] or spinning particle domains [9,11,12].

In Fig. 2 we display the transient deformation of the droplets in Fig. 1. The transient droplet deformation of a droplet is governed by three characteristic times: the charge relaxation time τ_e, which is the time scale for the interface to acquire its steady-state surface charge density distribution, the capillary time $\tau_c = \mu a/\gamma$, which is the time scale for droplet relaxation due to capillary forces, and the flow time scale $\tau_f = \mu/(\varepsilon_0\varepsilon_{ex}E^2)$, which is the time scale for transporting charges by hydrodynamic convection.

In the inset in Fig. 2a, shortly after the electric field is turned on, we observe a transient prolate to oblate anomaly, i.e. the measured droplet deformations evolve from positive (stretched out) to negative (compressed). This appears to be most pronounced for particle covered droplets that are not fully covered. A similar anomaly has been reported by computational studies of droplets in [50] and characterized by the Saville number Sa_{ex}, which is defined as the as the ratio $\tau_{e,ex}/\tau_{c,ex}$, and is a number characterizing the charge transport towards the droplet interface [42,50]. For the 2 mm sized pure silicone oil droplet used in our experiments, Sa_{ex} is about 4.5, indicating that it takes more time to accumulate charges at the interface than to transport charges away from the interface. Thus, at early times, the droplet behaves as a perfect dielectric, and the droplet deforms along the electric field direction.

For a given electric field, the drop deformation D_E increases with particle coverage φ (see Fig. 2a). A silicone oil droplet fully covered by polyethylene particles (violet diamonds) has a steady-state deformation D_E twice as large compared to a particle-free silicone oil droplet (red squares). This result could potentially be due to: (i) reduced resistance to deformation related to reduced surface tension, (ii) increased elastic stress from the particle shell, or (iii) an increase in electrical stress. Since the contributions from (i) and (ii) can be neglected for Pickering shells that are not jammed [35], we are left with (iii) for the present situations. In numerical models [11,35,45], charge convection has been shown to weaken the steady-state oblate deformation by distorting the surface charge density profile and weaken tangential stress. The electric Reynolds number Re_E represents the ratio of the charge relaxation time scale τ_e to the flow time scale τ_f. A small electric Reynolds number (< 1) indicates that it takes more time to transport charges by convection than the time it takes to build up charges at the droplet interface. Since $Re_E \approx 1$ for the silicone oil in castor oil system, charge relaxation by convection influences the steady-state droplet deformation. Without electrohydrodynamic flow, no charge convection is present in and around the fully covered droplets, thus explaining the larger electric stress and droplet deformation. In addition, the insulating polyethylene particles at the droplet surface decrease the effective conductivity of the droplet which then leads to a larger charge accumulation at the droplet interface [35].

Esmaeeli and Sharifi developed a simple model for the transient droplet deformation given by [51]: $D(t) = D_E[1 - \exp(-t/\tau_d)]$, where D_E is the steady-state deformation and τ_d is the time scale that governs the deformation dynamics: $\tau_d = \frac{\mu_{ex} a}{\gamma}\frac{(2\lambda+3)(19\lambda+16)}{40(\lambda+1)}$. This model strictly applies to pure droplets not covered by particles.

In Fig. 2b, $\ln(1 - D(t)/D_E)$ is plotted versus time (after the application of an electric field of strength 200 V mm^{-1}) for the same droplets in Fig. 1 and Fig. 2a. The plots are linearly fitted to calculate the slope (which is the inverse of τ_d). We note the observed exponential relaxation also for particle laden drops. For a more complete model, charge convection, finite charge relaxation and transient fluid inertia has to be included. Here we can only refer to computational and numerical work[44,45,50], since to the best of our knowledge, there are currently no particle-interface models available for the time dependent deformation of particle covered droplets. However, some capsule models have been used to describe the steady-state deformation of particle laden droplets, for instance elastic models where the particle layer has shear elasticity [11,52,53] or a fluid shell description where the particle layer can be considered fluid during deformation [35].

3.2. Effect of surface particles on droplet deformation time

The time for charges to accumulate at interfaces of layered or inhomogeneous dielectrics is given by the Maxwell-Wagner charge relaxation time, which is independent of the applied electric field [54]: $\tau_{MW} = \frac{\varepsilon_{in} + 2\varepsilon_{ex}}{\sigma_{in} + 2\sigma_{ex}}$, τ_{MW} is approximately 0.9 s for a silicone oil droplet in castor oil system. Because the silicone oil and the polyethylene particles have much smaller electric conductivities than the castor oil, and their dielectric constants are similar, the Maxwell-Wagner charge relaxation time of the particle laden droplets will be approximately the same as the particle-free droplets in castor oil. For the silicone oil droplet (red squares) in Fig. 2a, the deformation time t_D, defined as the time for droplets to reach the arbitrarily set threshold of 95% of their steady-state deformation D_E, is measured to be $t_D \approx 1.9$ s.

Fig. 3 shows the deformation time plotted versus the particle coverage for droplets with diameters ranging from 1.0–2.5 mm and subjected to a DC electric field of strength 200 V mm^{-1}. The deformation time increases when

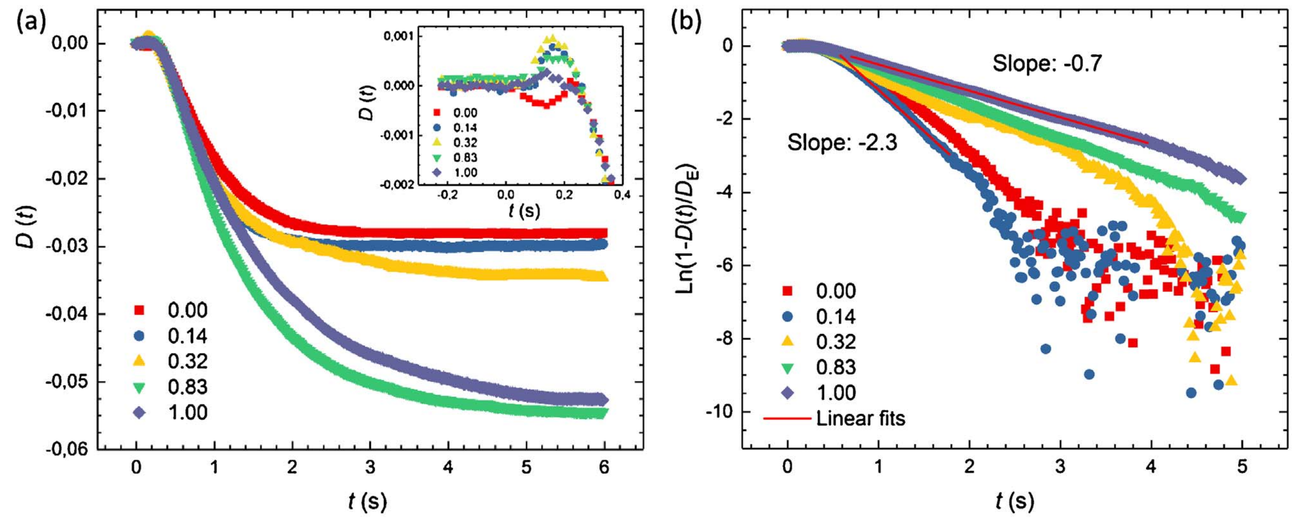

Fig. 2. Transient droplet deformation. (a) Transient deformation of 2 mm sized silicone oil droplets without and with surface particles when subjected to a 200 V mm^{-1} electric field (see Materials and Methods section for details on the measurements). The particle coverage φ ranges between 0 and 1. (b) ln(1-$D(t)/D_E$) plotted versus time.

more polyethylene particles are added to the droplet surface, which can be attributed to changes in the electric properties (conductivity and dielectric constant) of the droplet interface by the added particles. In a recent work, we concluded that the electric conductivity of a polyethylene Pickering droplet (fully particle covered droplet) is approximately 30% of a silicone oil droplet when using particles that are 50 μm (the same size as we use in the present work) [35]. Viscous dissipation due to the presence of the polyethylene particle layer might also contribute to the increased deformation time [55]. Similar to the experimental work reported in ref. [56] dealing with particle-free silicone oil droplets, we also find that the deformation time of particle covered droplets decreases with the applied electric field strength.

3.3. Deformation versus electric capillary number

The electric capillary number Ca_E is the ratio of the electric stress $\varepsilon_{ex}E_0^2$ to the capillary stress γ/a. For Taylor's model to be valid, Ca_E must be sufficiently small for the deformation to be linear in Ca_E. If the electric stress becomes larger than a critical capillary stress (which is working to restore the deformed droplet to a spherical shape), the surface tension can no longer balance the electric stress and the droplet breaks up into smaller droplets [48]. Fig. 4 displays the steady-state deformation of different sized silicone oil droplets with particle coverage ranging from 0 (no surface particles) to 1 (fully covered droplets) plotted versus the electric capillary number. As expected, the droplet deformation for all the droplets is linear with Ca_E (i.e. proportional to the droplet radius and electric field squared) for small deformations when surface tension is strong enough to overcome deformations due to electric stresses. For larger Ca_E values, the droplet deformations start to deviate from Taylor's theory. It is also observed that particle-free droplets deform less than particle covered droplets.

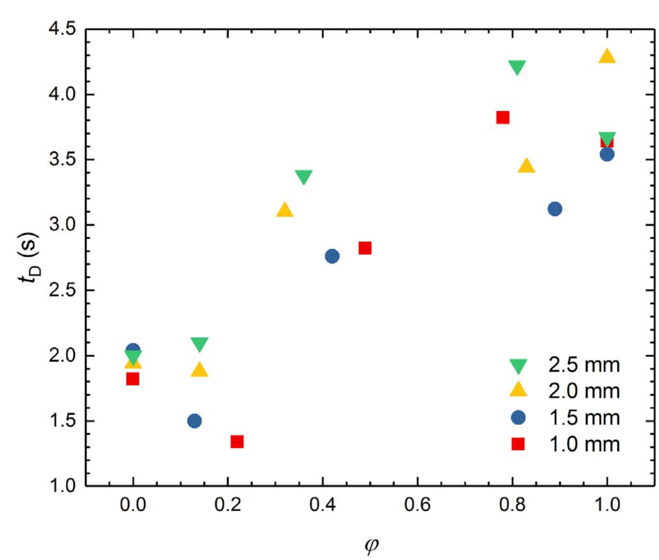

Fig. 3. (a) Deformation time (time for droplets to reach the arbitrarily set threshold of 95% of their steady-state deformation D_E) plotted versus the particle coverage φ (particle covered surface area/droplet surface area) for droplets with diameters ranging from 1.0–2.5 mm and subjected to a DC electric field of strength 200 V mm^{-1}.

4. Conclusions

We show that electrically insulating polyethylene particles at the interface of silicone oil droplets results in a larger deformation compared to particle-free droplets. We conclude that this can be caused by reduced charge convection and reduced electric conductivity of the interface when the polyethylene surface coverage is high. We also find that the deformation time increases with increasing particle coverage. This may not only be caused by diminished electrohydrodynamic flows and interfacial charge convection, but can possibly also be due to particle capillary interactions. Immediately after the electric field is turned on, we observe a transient prolate to oblate anomaly, possibly caused by a delay in the interfacial charging.

Author contributions

A.M., Z.R., P.D. and J.O.F. planned all experiments. A.M. and Z.R. performed the experiments at NTNU, Norway. A.M., Z.R. and K.K. performed the experiments at AMU, Poland. A.M. wrote the manuscript. A.M., Z.R., P.D. and J.O.F. contributed in all discussions towards the finalization of the manuscript.

Competing financial interests

The authors declare no competing financial interests.

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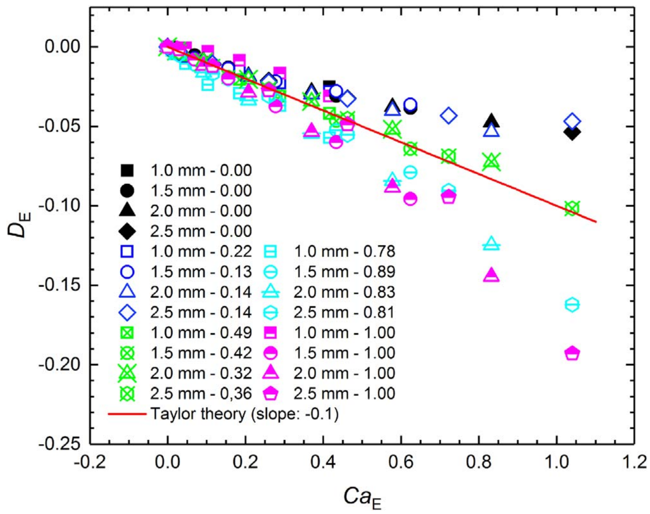

Fig. 4. Steady-state deformation of particle covered silicone oil droplets (diameters 1.0–2.5 mm) plotted versus the electric capillary number $\mathrm{Ca_E} = \varepsilon_{ex} E_0^2 \gamma / a$. The particle coverage φ ranges between 0 for no surface particles, and 1 for droplets fully covered by a porous particle layer. The strength of the applied electric field was incrementally increased from 50 to 300 V mm^{-1} (in steps of 50 V mm^{-1}).

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