## Behaviour of water droplets falling in oil under the influence of an electric field.

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#### **Abstract**

The use of electric field is a promising technique for separating stabile water-oil emulsions. Charges induced on the water droplets will cause adjacent droplets to align with the field and attract each other. The present work outlines the efforts to model the forces that influence the kinematics of droplets exposed to electric field when falling in oil. Mathematical models for these forces are briefly presented with respect to the implementation in a multi-droplet Lagrangian framework. The droplet motion is mainly due to buoyancy, drag, film-drainage, and dipole-dipole forces. Attention is paid to internal circulations, non-ideal dipoles, and the effects of surface tension gradients. Experiments are performed to observe the behavior of two falling water droplets exposed to an electric field perpendicular to the direction of their motion up to the droplets coalescence. The droplet motion is recorded with a high-speed CMOS camera. The optical observations are compared with the results from numerical simulations where the governing equations for the droplet motion are solved by the RK45 Fehlberg method with step-size control and low tolerances. Results, using different models, are compared and discussed in details. Observation concerning the contribution of the different forces acting on the water droplets kinemtics is presented.

## Introduction

The oil extracted from offshore reservoirs will normally contain a large and, during the reservoir lifetime, increasing percentage of water in the oil. When the water-oil mixture is passed through the pressure relief valve an emulsion with a high percentage of small water drops is formed. Before the oil is pumped on-shore or into tankers, it is desirable to extract the water from this emulsion. Today the separation tanks are mainly built or operated as gravity separators with low flow rates and long residence times; lasting from minutes to tens of minutes. The residence time mainly depends on the sedimentation velocity of the smallest drops (e.g.  $d < 100 \mu m$ ). Electrostatic fields are to some extent used to help smaller drops to coalesce in to larger drops that sediment quicker as presented by Eow et al. [1]. The combination of an electric field and a moderate turbulent flow is a promising and compact technique for separating stabile water-oil emulsions, see Atten [2]. Charges induced on the water droplets will cause adjacent droplets to align with the field, attract each other and eventually coalesce. The sedimentation velocity increases proportionally to the square of the diameter, and therefore one wishes to get the smallest water droplets to coalesce into larger droplets. The present work outlines the forces that influence the kinematics of falling spherical droplets exposed to an electric field. Mathematical models for these forces are presented and discussed with respect to the implementation in a multi-droplet Lagrangian framework. The spherical droplet motion is mainly due to buoyancy, drag, film-drainage, and dipole-dipole forces. General and physically meaningful models for these forces are needed in order to establish simulation models for the behavior of water-oil emulsion.

In the present work, models for the forces that are believed to dictate the motion of droplets falling in oil under the influence of an electric field are reviewed and discussed in details. A modeling framework that properly account for the effect of the different forces on the droplet motion is used as suggested by Chiesa et al. [3]. Experiments are performed to observe the behavior of two falling droplets released simultaneously into oil. The electrical field applied perpendicularly to the direction of the motion induces charges on the water droplets. This causes adjacent droplets to align with the field and attract each other. A comparison between observations and predictions is presented in order to assess the performance of the modeling framework used in this work.

## Theoretical background

The trajectory of a spherical droplet i is calculated by integrating Newton's second law. The law equates the droplet inertia with the forces acting on it, and reads:

$$\frac{dx_i}{dt} = v_i \tag{1}$$

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$$m_i \frac{dv_i}{dt} = F_{\text{fluid}} + F_{\text{ext}} + F_{\text{d-d}},$$
(2)

where  $m_i$ ,  $x_i$ , and  $v_i$  are the mass, position, and velocity of the droplet.  $F_{\rm fluid}$  represents the vector of forces acting from the fluid on the droplet,  $F_{\text{ext}}$  is the external force vector, and  $F_{\text{d-d}}$ represents the inter-droplet force vector. Droplet tracking with droplet-droplet interaction has a high computational cost. It is therefore important to keep the computational work necessary to calculate the particle forces as low as possible since the forces have to be calculated for each particle. Finally models should be easily implementable in a numerical code.

In the following sessions, models for the forces that are believed to dictate the motion of droplets falling in oil under the influence of an electric field are reviewed and discussed in details. The modeling framework proposed by Chiesa et al. [3] is used in the present work. The electric force between the particle and the droplet is modeled with either the analytical expression obtained by Davis [4], the DID model by Siu et al. [5] or the point dipole model by Klingeberg et al. [6]. When describing the motion of a falling water droplet the effect of internal circulation induced in the droplet has to be taken into account. Internal circulation reduces the viscous part of the drag force and therefore the drag coefficient needs to be corrected in order to account for this reduction as outlined by Happel and Brenner [7]. Furthermore, the surface tension varies over the droplet surface by the effect of surfactant on the interface and by elongation of the droplet, caused by the electric field. This leads to interfacial stresses that inhibit the creation of internal circulation. LeVan [8] suggests how to take into account the effect of surface tension gradient in our numerical framework. The model proposed by Vinogradova [9] takes into account the slip between the liquid film and the approaching spheres.

## Modeling the fluid-droplet and body forces

Fluid droplet forces are transfered from the fluid to the droplets through friction and pressure difference. These forces are expressed exactly by the following surface integral:

$$\frac{1}{V_{\rm d}}F_{\rm fluid} = \frac{1}{V_{\rm d}} \int_{\mathcal{A}_{\rm d}} \left( -p_s n_{\rm d} + \tau_d \cdot n_d \right) \, \mathrm{d}A \tag{3}$$

where  $V_d$  is the volume of the droplet.  $p_s$  is the pressure at the droplet surface,  $n_d$  represents the unit outward normal vector and  $\tau_d$  is the shear stress tensor at the droplet surface. The pressure and the friction on the interface are unknown and Eq. (3) has to be modeled. In the Lagrangian framework, the models for the surface integral attempt to provide particular physical meanings.

The 'steady-state' drag force acts on a droplet in a uniform pressure field when there is no acceleration of the relative velocity between the droplet and the conveying fluid. The force reads:

$$F_{\rm d} = \frac{1}{2} \rho_{\rm c} C_{\rm d} A |u - v| (u - v), \tag{4}$$

Surfactants on the interface and elongation of the droplet, caused by the electric field, give a variation in the surface tension. The surface tension gradient leads to interfacial stresses that inhibit the creation of internal circulation. The surface tension gradient is included in the formula by LeVan [8] for the drag coefficient:

$$C_d = \frac{24}{Re_d} \frac{3\lambda + 2 + 2\kappa(\mu_c r_d)^{-1} + 2/3\gamma_1(\mu_c|u-v|)^{-1}}{3\lambda + 3 + 2\kappa(\mu_c r_d)^{-1}}, \quad (5)$$

where also the surface dilational viscosity  $\kappa$  is taken into account. However, in the present work surface dilational viscosity is neglected,  $\kappa=0$ . In Eq. (5) it is assumed that the interfacial tension varies as follows  $\gamma=\gamma_0+\gamma_1 cos\psi$  where  $\psi$  is measured from the front stagnation point.

The *virtual mass force*  $F_{\rm vm}$  is an unsteady force that describes the acceleration of fluid when a particle and the fluid have a relative acceleration. It reads:

$$F_{\rm vm} = \frac{\rho_{\rm c} V_{\rm d}}{2} \left( \frac{Du}{Dt} - \frac{dv}{dt} \right) \tag{6}$$

We assume that the droplets have no net charge, hence the electric field as a far field force can be neglected. On the other hand, the electric field gives rise to dipole-dipole interactions between the droplets, which are modeled as inter-droplet forces. Then the gravity is the only *external force* and the buoyancy force is given by:

$$F_{b} = (\rho_{d} - \rho_{c}) g V_{d} e_{g}$$

$$\tag{7}$$

where g and  $e_{\rm g}$  are the modulus and the direction of the gravity.

In the present work, the effects of the pressure gradient, the Basset history force and the lift forces have been neglected. The pressure difference over a small droplet is negligible due to the size of the droplets. The contribution from the gravity is handled separately. Lift forces are due to droplet rotation and shear forces, and can therefore be neglected when a rigid sphere or droplet is falling in a stagnant fluid. Due to the small size of the spheres and the high viscosity of the oil, the particle time scale is very small. Thereby follows that the Stokes number is small and the Basset history force can therefore be neglected [10].

# Modeling droplet-droplet forces

The inter-droplet forces are the film thinning forces, due to the drainage of the fluid between the droplets, and the electric forces due to polarization of the conductive water droplets. The *film-thinning force* is caused by drainage of the liquid film between two approaching droplets. The derivation of the formulas usually requires that the gap between the particles is small  $h \ll a$  and that the flow is within Stokes regime  $Re_dh \ll a$ .  $a = (r_1r_2)/(r_1+r_2)$  is the reduced radius. When the particles are very close, a slip will occur and avoid a zero impact velocity. The formula of Vinogradova [9] for the fil-thinning force includes a slip distance b and reads:

$$F_{\rm f} = -\frac{6\pi\mu_{\rm c}a^2(v_r \cdot e_{\rm r})}{h} \left\{ \frac{2h}{6b} \left[ \left( 1 + \frac{h}{6b} \right) \ln\left( 1 + \frac{6b}{h} \right) - 1 \right] \right\} e_r. \tag{8}$$

The electric forces are due to polarization of the conductive water droplets. Consider an uncharged spherical droplet placed in an insulating medium. The droplet is furthermore subjected to an electric field  $E_0$ . The field outside a dielectric sphere of permittivity  $\varepsilon_d$  corresponds exactly to the electric field of a dipole located at the sphere centre. The value of this dipole moment p depends on the sphere size, permittivity and the strength of the electric field. Due to the polarization of the droplet, the poles will have charges of same magnitude but opposite polarity, preserving zero net charge. In a homogeneous field, the net force on the droplet is zero. Subjected to an inhomogeneous field the droplet will experience a stronger field at one pole than at the other, resulting in a net force acting on the droplet in the direction of the field gradient, a phenomenon called dielectrophoresis. The resulting force is given by  $F = (p \cdot \nabla)E$ . If the permittivity of the drop  $\varepsilon_d$  is higher than the permittivity of the surrounding medium  $\varepsilon_{oil}$ , the drop will move towards the high field region. An inhomogeneous electric field may for instance be set up by nearby point charge or another dielectric droplet. In the latter case the electrostatic force attracts the two droplets, given that  $\varepsilon_d > \varepsilon_{oil}$ .

## Point dipole model

For large droplet distances  $|d|/r_d \gg 1$  we can approximate the electrostatic interaction between two droplets as the force between two dipoles located at the sphere centres. This is frequently referred to as the *point-dipole* approximation. The forces in radial direction  $F_{\rm r}$  and tangential direction  $F_{\rm t}$  read [6]:

$$F_{\rm r} = \frac{12\pi\beta^2 \varepsilon_{\rm oil} |E_0|^2 r_2^3 r_1^3}{|d|^4} \left( 3k_1 \cos^2 \theta - 1 \right) \tag{9}$$

$$F_{\rm t} = -\frac{12\pi\beta^2 \varepsilon_{\rm oil} |E_0|^2 r_2^3 r_1^3}{|d|^4} k_2 \sin(2\theta), \tag{10}$$

where  $\theta$  is the angle between the direction of the electrical field  $E_0$  and the relative droplet position vector d.  $\beta$  is defined as:  $\beta = \frac{\mathcal{E}_d - \mathcal{E}_{\mathrm{oil}}}{\mathcal{E}_d + 2\mathcal{E}_{\mathrm{oil}}}.$   $k_1 = 1$  and  $k_2 = 1$  in the point dipole approximation. The point-dipole model is not valid when the droplets are approaching each other. In the literature there are different approaches to find the dipole-dipole forces beyond the point dipole approximation for multiple particles of arbitrary size and position.

# The multiple image method

A promising method, the *multiple image method*, was presented by Yu et al. [11]. The two first terms in the multiple image method gives the dipole induced dipole model (DID) [5], which is simple and numerical efficient. Siu et al. [5] show that the DID model is in good agreement with experimental values for  $|d|/r_1 > 0.1$  for equally sized conductive particles. The DID model reads as Eq. (9) and (10), where the coefficients  $K_1 \neq 1$  and  $K_2 \neq 1$ , see Siu et al. [5] for further details. In the limit  $|d| \rightarrow \infty$  the coefficients  $K_1$  and  $K_2$  approach unity and we recover the point dipole model given by Eq. (9) and (10).

## The analytical solution

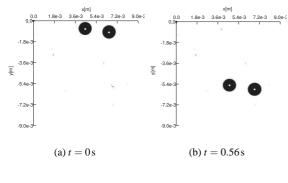
Davis [4] found an analytical solution to Laplace's equation for two conducting spheres of arbitrary size, discplacement and net charge, using bi-spherical coordinates. The exact solution for uncharged spheres is given by:

$$F_r = 4\pi\epsilon_{\text{oil}}|E_0|^2 r_2^2 \left(F_1 \cos^2 \theta + F_2 \sin^2 \theta\right)$$
 (11)

$$F_t = 4\pi \varepsilon_{\text{oil}} |E_0|^2 r_2^2 F_3 \sin(2\theta),$$
 (12)

where the parameters  $F_1$ ,  $F_2$ , and  $F_3$  are complicated series depending on the ratios  $|d|/r_2$  and  $r_1/r_2$ . Unfortunately, the computational cost required for calculating  $F_1-F_3$  is high in a multi-droplet situation. However, the exact solution is excellent for benchmarking other models in cases with two particles/droplets. For large drop separations  $|d|/r_1\gg 1$  the force components  $F_1-F_3$  approach the values of the point-dipole Eq. (9) and (10). For small separations  $|d|/r_1<1$ ,  $F_2$  and  $F_3$  takes constant values while  $F_1$  diverges, see Atten [2].

## **Results and Discussions**



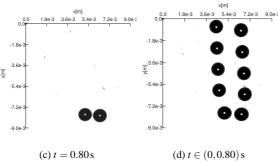


Figure 1: Experimental observation of the kinematic of two water droplets falling in a stagnant oil under the influence of an electric field.

Experiments are designed for visual observation of water droplets in oil under the influence of electric field stress, see [3]. Two sub-millimeters sized water droplets are released within an upper ground electrode from a glass capillary coated with gold to avoid static charge transfer from glass to the particle. The two water droplets fall freely in Nytro 10X oil and after  $t=0.56\,\mathrm{s}$  an electric 50Hz field of magnitude 280V/mm is applied. The electric field is horizontally applied thus perpendicular to the direction of the droplets motion. A sinusoidal voltage with frequency 50Hz is used. Droplet interactions and coalescence are recorded with the Phantom V4 high speed CMOS camera.

Numerically, the governing equations (1) and (2) are solved with a Runge-Kutta Fehlberg 4-5 solver with step-size control.

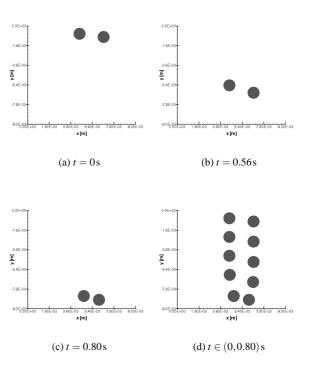


Figure 2: Visual prediction of the kinematic of two water droplets falling in a stagnant oil under the influence of an electric field. The models of Davis, LeVan, Vinogradova are employed.

Accurate simulations are ensured by using a relative tolerance of  $10^{-5}$  and an absolute tolerance of  $10^{-25}$ . The modeling framework outlined in [3] is used in the present work to predict the kinematics of two water droplets simultaneously released in oil. The radius of the smallest droplet placed on the left in the experiments is  $r_1 = 533 \,\mu\text{m}$  and the radius of the biggest one is  $r_2 = 553 \,\mu\text{m}$ . Uncertainty in measured droplet diameter is less than  $10\mu m$ . The position of the droplets is recorded with a high speed CMOS camera and it is digitally extracted from the sequential frames [3]. Fig. 1 shows a series of frames at different times. At time t = 0.0s, see Fig. 1 a) the droplets are released in the oil. The droplets fall down because of gravity and at time t = 0.56s an electric field perpendicular to the direction of the droplets motion is applied. Fig. 1 b) shows the droplets position at the instant when the field is applied. Fig. 1 c) shows the droplets position at time  $t = 0.80 \,\mathrm{s}$ , when the droplets are just about to coalesce. The electrical field induces charges on the water droplets and this causes adjacent droplets to align with the field and attract each other. Fig. 1 d) shows a sequence of frames taken at a time interval  $\Delta t = 0.20$ s where the effect of the electric field on the droplet kinematics is observable. The biggest droplet is observed to fall at a higher speed than the smallest one and therefore the angle between the center of the droplets and the normal to the droplets motion increases. This trend is reversed when a field normal to the droplet motion is applied and the droplets tend to align with the field. Fig. 2 shows a visual prediction of the kinematics of the two falling droplets when the Davis model is employed together with the models of LeVan, and Vinogradova as previously explained. The observed y-coordinates of the two droplets are here compared to the predictions obtained when using different models for the electric forces. Fig. 3 shows a comparison between the predicted and observed y-coordinate of the two droplets as a function of time. The Davis, the DID and the point dipole models are used to-

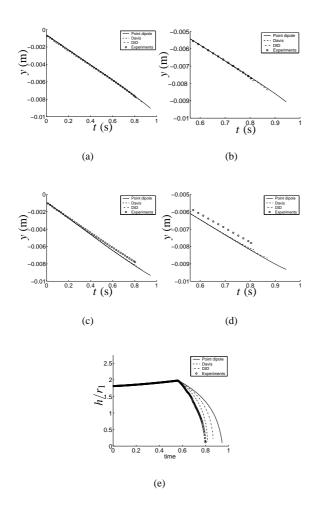


Figure 3: Observed and predicted vertical coordinate versus time of droplet 1 a), b) and of droplet 2 c),d). Different models for the electrical forces are adopted together with the models of LeVan, and Vinogradova.

gether with the models of LeVan, and Vinogradova to predict the time of coalescence and droplets kinematic of the droplets. The results obtained by the Davis model predict well the time of coalescence and the overall kinematic of the two droplets. The simulation obtained by the other two models overestimated the collision time. This result is coherent to the results presented by Chiesa et al. [3]. The point dipole model underestimates the effect of the electric forces on the kinematic of the droplets. In the numerical prediction, the biggest droplet is falling at a higher speed than the smallest one as observed in the experiment. The predicted fall of the smallest droplet up to time t = 0.56 s agrees well with the experimental observations see Fig. 3a) and b). On the other hand the predicted fall of the biggest droplet is slightly overestimated, see Fig. 3c) and d). This is most probably due to the uncertainty related to the measured droplets size. There is not any reason to believe that the drag model needs to be reviewed in order to account for the vicinity of the other droplet. The drag force acting on the droplets is effected by the present of a second droplet in its vicinity when the droplets lye in the boundary layer of the neighboring one. In the present case, the boundary layer is very small due to the high viscosity of the oil in which the droplets are moving. The predicted and observed normalized distance  $h/r_1$  between the droplets centers as a function of time is plotted in Fig. 3e). The time to collision predicted by means of the Davis equations well agrees with the coalescence time observed in the experiments.

#### Observations

The motion of two fluid droplets simultaneously released in oil is predicted by means of the modeling framework proposed by Chiesa et al. [3]. The effect of internal circulation induced in the droplets is taken into account together with the variation of the surface tension of the droplets due to the electric field. The electric field is applied normally to the motion of the droplets after 0.56s of free fall. The droplets tend to align with the field before they eventually coalesce with each other. This behavior is well predicted by the model proposed by LeVan Eq. (5) for the drag force, by Vinogradova Eq. (8) for the film-thinning force and the Davis' analytical expression. The use of different models to take into account the effect of electrical fields is also assessed. The 'time of coalescence' predicted by means of the Davis equations well agrees with the collision time observed in the experiments.

#### References

- J. S. Eow, M. Ghadiri, A. O. Sharif, and T. J. Williams. Electrostatic enhancement of coalescence of water droplets in oil: a review of current understanding. *Chem. Eng. J.*, 84:173–192, 2001.
- [2] P. Atten. Electrocoalescence of water droplets in an insulating liquid. *J. Electrostatics*, 30:259–270, 1993.
- [3] M. Chiesa, J. A. Melheim, A. Pedersen, S. Ingebrigtsen, and G. Berg. Forces acting on water droplets falling in oil under the influence of an electric field: numerical prediction versus experimental observations. Submitted to European Journal of Mechanics, 2004.
- [4] M. H. Davis. Two charged spherical conductors in a uniform electric field: forces and field strength. Rand. Corp. Memorandum RM-3860-PR, January 1964.
- [5] Y. L. Siu, T. K. Wan Jones, and K. W. Yu. Interparticle force in polydisperse electrorheological fluid: Beyond the dipole approximation. *Comput. Phys. Commun.*, 142:446–452, 2001.
- [6] D. J. Klingenberg, F. van Swol, and C. F. Zukoski. The small shear rate response of electrorheological suspensions. II. extensions betond the point-dipole limit. *J. Chem. Phys.*, 94(9):6170–6178, 1991.
- [7] J. Happel and H. Brenner. Low Reynolds number hydrodynamics. Martinus Nijhoff Publishers, The Hague, The Netherlands, 1983.
- [8] D. M. LeVan. Motion of droples with a Newtonian interface. *J. Colloid Interface Sci.*, 83(1):11–17, 1981.
- [9] O. I. Vinogradova. Drainage of a thin liquid film confined between hydrophobic surfaces. *Langmuir*, 11:2213–2220, 1995.
- [10] E. E. Michaelides. Hydrodynamic force and heat/mass transfer from particles, bubbles, and drops - the Freeman scholar lecture. J. Fluids Eng. - Trans. ASME, 125:209– 238, March 2003.
- [11] K. W. Yu and T. K. Wan Jones. Interparticle forces in polydisperse electrorheological fluids. *Comput. Phys. Commun.*, 129:177–184, 2000.