

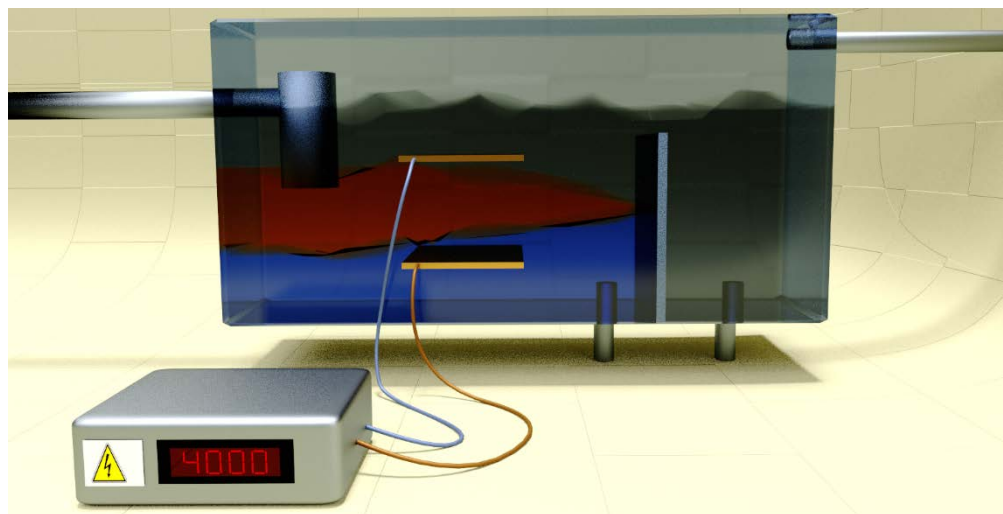
Report

Electrocoalescence III

Status and summary report from activities

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ABSTRACT

The report briefly sums up the status from SINTEF Energy's research activities in the area of electrocoalescence, and gives more detailed overview of the different activities in Electrocoalescence III project.

Experiments on surface characterisation of water drops in crude has been performed in a bespoke test rig. Water drops were stressed by electric fields, and the response of the surface recorded.

The drop behaviour was modelled by level-set methods and molecular modelling with good results.

A technique for producing drop on demand by electrostatic stressing of a water meniscus is developed. One can produce water drops of desired sizes in the range of tens to hundreds of micrometres.

A benchtop model coalescer for testing coalescence efficiency for realistic crude oil processes, and optimizing process parameters is designed and tested with good results.

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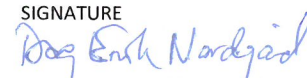
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1 BACKGROUND

1.1 Short history

Separation of water from crude oil is an important process in exploitation and refining of crude oil. Water drops will precipitate with time as they are heavier than oil. This is the mechanism used in so-called gravity separators. If water drops in the emulsion are small, sedimentation speed is slow and the residence time in the tanks becomes long. In some cases, stable emulsions are formed, and gravity is not sufficient to separate the water from the oil. It was early discovered that by applying electrical fields one may speed up separation [1]. This effect is achieved by electrostatic forces pulling drops together and making them merge into larger drops; facilitating electrocoalescence (EC).

While hydrodynamic forces are short-range forces, electrostatic forces act over longer distances. One has experienced that it is not straightforward to optimize electrocoalescence; crude oil qualities vary and valid models of electrocoalescence have not been available.

Historically electrocoalescence was achieved by applying large electrodes into gravity separators energized either with AC or DC. Lately so-called compact coalescers have arrived at the market, allowing turbulent flows and usually applying field over shorter distances using insulated electrodes [2]. Separators are used onshore and offshore in a late stage of crude oil production, and onshore for removing saline water in desalinizers before refining processing. Size reduction, energy efficiency and separation efficiency are important challenges in design and operation of electrocoalescers.

Some larger reviews of electrocoalescence have been published [3, 4, 5, 6]. We will here briefly sum up the experiences, experimental techniques, models and questions that have been established in a series of projects run at SINTEF Energy on this topic before concentrating on the specific results achieved in the latest project. The first project, EC I, started with studies of droplets and drop pairs in model oils and effects of electric fields on multidrop systems. Then in the next project, EC II, surface chemistry and studies of drops and drop pairs in crude oils followed. In the last project, EC III, we have mainly been targeting crude oil systems and the drop surface behaviour as surface properties is seen as a main obstacle for coalescence.

The teams working on this has comprised electrical engineers, physicists, mechanical engineers and chemists who together have targeted this multidiscipline problem.

1.2 Compact coalescers

Compact coalescers are often used in combination with larger sedimentation tanks, cyclones etc. for the last polishing of the crude oil targeting water cuts below 0.5 %. The flow can often be turbulent. With high shear, water drops are frequently brought close and in contact which aids coalescence. On the other hand, larger drops may be ripped apart. Small dimension will also often result in water bridging between electrodes forming a short circuit. The electrodes are therefore normally covered with dielectric insulation.

In our studies we have mainly targeted low water-cut emulsions (i.e. ~ 2 % water cut and below) studying drops in the 50-500 μm diameter range. Typically drops will be then several drop diameters apart.

1.3 How the surface of the water drops affect the emulsion stability

The properties of water in oil emulsion are determined by the water, the oil and the properties of the interface between the water drops and the oil. Substances from the oil and water may adsorb to the interface and properties may vary widely. The surface properties have a strong effect on coalescence. For coalescence to occur one first have to drain the oil film between the water drops. Next the stabilizing surface film has to break before the water drops merge.

Coalescence of emulsion drops covered by protective stabilising surface films occurs at two stages, namely in the film thinning to a critical thickness at which point the film becomes unstable, and in the final film rupture. The rate of film thinning is dependent on the rate at which the interface moves the film. Hence, this process is affected by properties of the entire colloidal system, i.e. viscosity and density of the two phases, concentration and type of chemical components (surfactants) at the interface, forces acting between the interfaces, interfacial tension and its gradient, interfacial shear, dilatational viscosity and elasticities and the size of the interacting droplets. The final film rupture may occur due to flow instabilities, temperature fluctuations, electric fields or Marangoni effects. Hence, demulsification of water-in-crude oil emulsions is commonly achieved by means of thermal, electrical, mechanical and/or chemical methods.

In the start (EC I) the studies were focusing towards model oils allowing depiction of the drops, with some studies of drops and drop pairs covered with asphaltenes. In the next phase (EC II) one aimed at more realistic conditions and worked with real crudes developing NIR camera techniques for studying singular drops and drops falling onto water surfaces. Finally in the last project (EC III) one focused on more detailed studies of surface behaviour under transient electric fields and also on development and testing of a model coalescer for measuring coalescence efficiency of real systems.

1.4 Physics of coalescers

When two water drops in an emulsion is brought in contact they will after some time coalesce; the water phase in the two drops gets in contact. When the surface is clean this occurs instantaneously, while for drops covered with a layer of amphiphilic substances this may take time. When drops are suspended in oil the drop impacts will depend on gravitational and flow forces. *Flow forces are near field forces* where the pressure from liquid flow acts upon the drops. In flows with shear and turbulence this results in more frequent drop collisions. When drops are pushed together, the oil between them must be drained away before they can get in contact. In large separators with stagnant emulsions, drop impacts are rare; mainly resulting from larger drops with higher sedimentation speed overhauling smaller slower drops.

Electrostatic forces on the other hand acts over longer distances and may help to bring drops together. There are two different forces acting, one is the so-called electrophoretic force, acting on drops having a net charge. These are either attractive or repulsive forces depending on charge polarities and direction of the electric field. Drops will acquire charge from induction when hitting metallic conductive surfaces. Unless a drop has a charge resulting from specific chemical properties, the charges on a drop will leak away due to losses to the surrounding crude oil. The charge loss depends on the conductivity and time constant of the surrounding crude oil. Coalescence due to electrophoresis will mainly be a process taking place at or close to surfaces. The other force is the so-called dielectrophoretic force. These forces act in divergent electric fields on drops that can be polarized, and will attract drops towards increasing fields. A drop in a uniform field will create a divergent field around itself and will therefore attract other drops. Coalescence induced by dielectrophoretic forces will therefore be a bulk process.

A water drop in an electric field will be polarized, stretch and may become unstable from forces acting on the charges at the poles (Figure 1-1). The stretching will depend on the field and on the surface tension of the interface. The dynamics of the deformation depend on the surface stiffness and other properties. Therefore electrostatic stretching may be used to investigate surface properties, as an alternative to pendant drop investigations. This technique is established in EC I and EC II and is further developed in EC III.

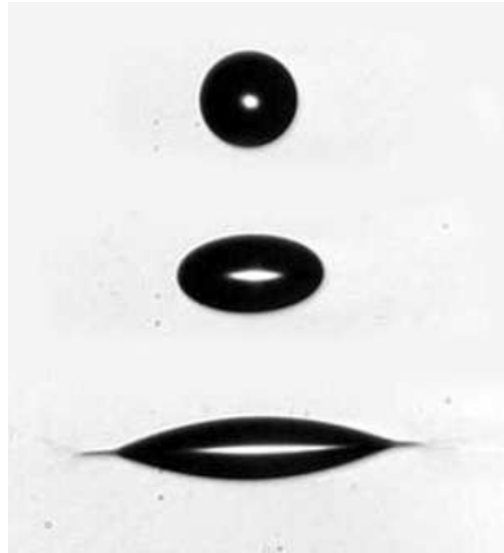


Figure 1-1: Water drops stretched in an electric field until destabilisation.

When two drops get close to each other they will each deform the field around them and attractive forces may result in coalescence as seen in Figure 1-2. These forces have a limited range as shown in Figure 1-3. Here the forces are scaled by the distance between the drops (s) relative to the radius of the smaller drop (R_2). We see that when the smaller drop is more than one diameter away then the attractive forces falls quickly off. So the dielectrophoretic force on drop pairs is a proximity effect.

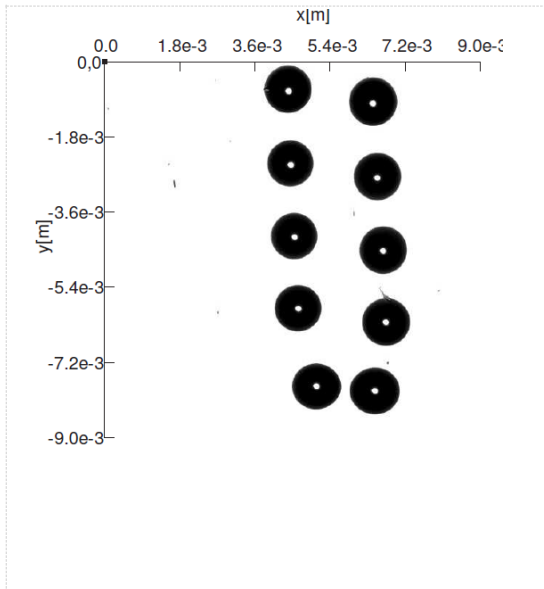


Figure 1-2: Falling drops attracted by horizontal electric field.

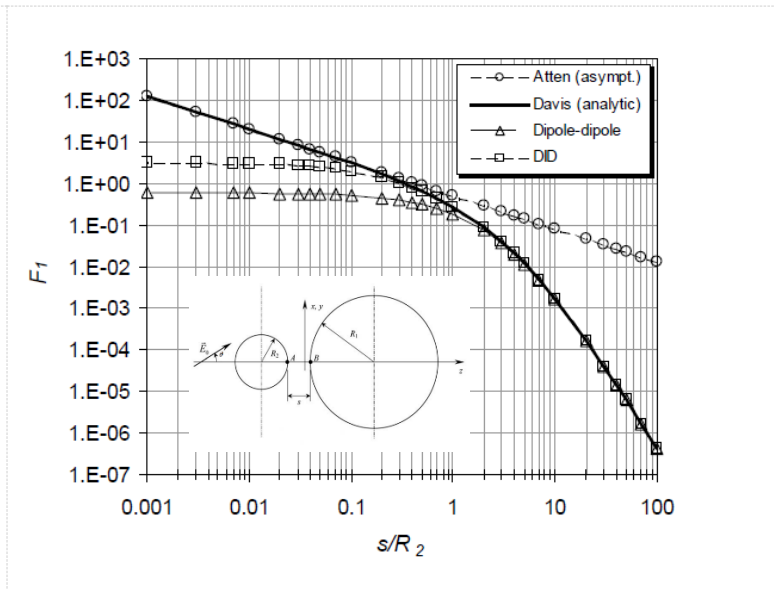


Figure 1-3: Forces between two water drops of different size at distance s . Calculations from different sources.

Voltage shape and frequency will influence the efficiency of compact coalescers. Effects of forces will vary with voltage type. For electrophoretic forces the direction of the forces varies with the polarity of the applied voltage, while dielectrophoretic forces on water drops will always be attractive, both for AC and DC. The voltage shape also matters. For many effects it is the integral of the forces that matter. An AC voltage with a high RMS value (i.e. square wave AC with an RMS value equal to the peak voltage) has a higher efficiency than a voltage with a low RMS value as e.g. a sine voltage with an RMS value of 0.7 times the peak value. From this point of view, DC voltages seem preferential.

In a compact coalescer there will usually be an insulating layer with a low conductivity covering the electrodes to avoid short circuit from water bridging. The DC case is simple, only conductivities matter and the crude oil with a high conductivity will almost short circuit the electric field: The voltage drop occur across the electrode covering, resulting in a low field in the emulsion to drive electrocoalescence (red curve) as shown in Figure 1-4 [7]. However, if one uses AC at higher frequencies the potential distribution will be capacitive and governed by permittivity of electrode covering and crude oil. As permittivity of crude and electrode covering will be quite similar the potential distribution will now be even (Blue line). The limiting frequency is given by the so-called relaxation time constant of the crude $\tau = \epsilon_0 \epsilon_r / \gamma$, where $\epsilon_0 \epsilon_r$ is the permittivity and γ is the conductivity of the liquid. The applied frequency, f , has to be above a certain limit given by this time constant to get any voltage across the emulsion: $f > \gamma / 2 \epsilon_0 \epsilon_r$. One has to be aware that the conductivity varies a lot with temperature as shown in Figure 1-5, where the conductivity for a number of crude oils is plotted vs. the inverse absolute temperature. The figure shows how crude oil conductivities can vary between room temperature and 160 °C. To get an efficient coalescence process the frequency has to be adjusted according to the conductivity of the oil at the processing temperature. Knowledge on oil conductivity is important to evaluate coalescer efficiency.

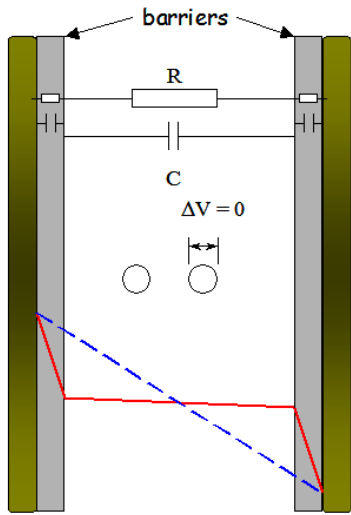


Figure 1-4: Electric field in coalescer with and without surface covering.

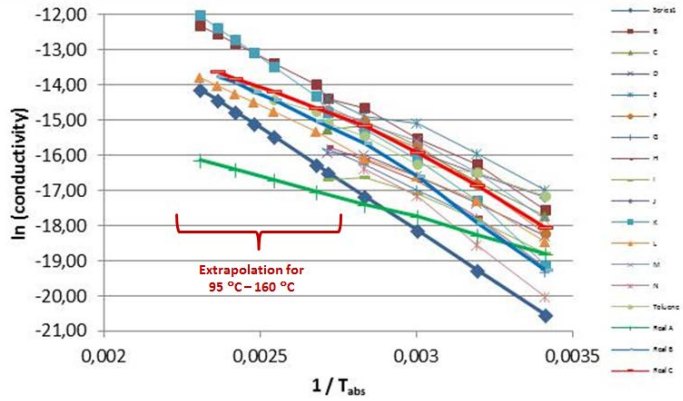


Figure 1-5: Conductivity vs inverse absolute temperature.

In order for the electric field to activate drop merging, drops need to be close. With a low water cut drop distances will often exceed the drop diameters. Assuming monodisperse drops the ratio between interdrop distance and drop diameter will only depend on water cut, not on drop diameter. Figure 1-6 shows that for 5 % water cut or below the drop distances will easily exceed two drop diameters. In 1993 Atten showed that in order to explain electrocoalescence there had to be a shear movement in an emulsion to get drops close enough for the drops to “see” each other [8].

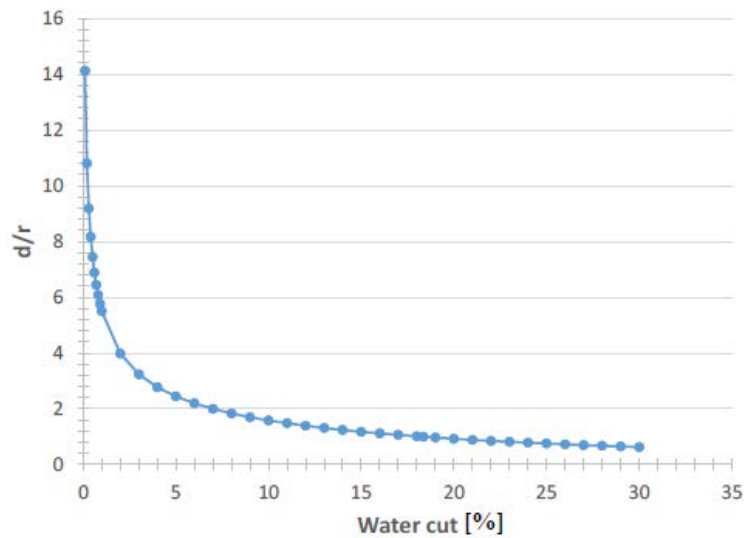


Figure 1-6: Reduction of drop distance d/r vs water cut for mono-disperse water drops.

In a conventional separator, one often talks about coalescence time, being the time needed for two meeting drops to merge. First, time is needed to drain the oil between the drops, then when drops rest in contact it will often take time for the drop surfaces to break and the drops to coalesce. This time is often stochastic and will vary with the surface properties of the drops. When two drops meet in a shear flow (see Figure 1-7) they will stay close (within attraction distance, e.g.) for a certain time called coincidence time. If the coincidence time is longer than the coalescence time drop merger will occur. Electrostatic attraction will increase coincidence time, improve draining and also reduce coalescence time by introduce new mechanisms and improve “adhesion”.

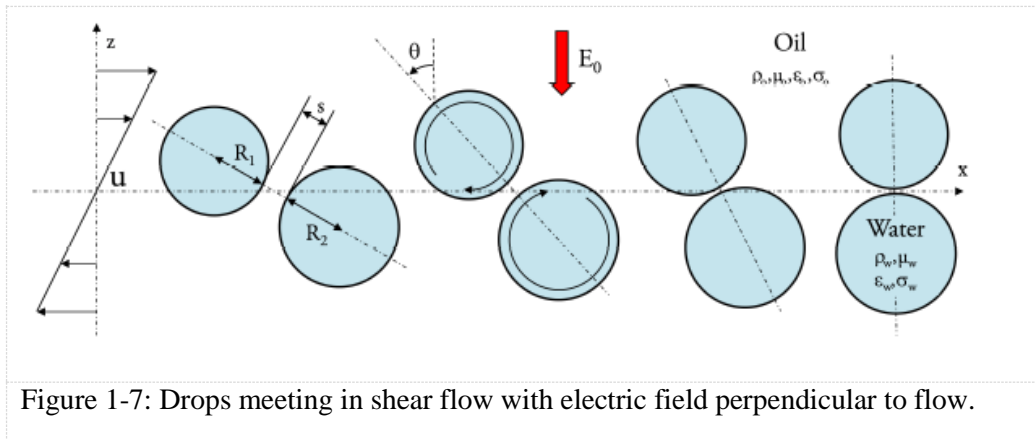


Figure 1-7: Drops meeting in shear flow with electric field perpendicular to flow.

In an early phase (EC I) much work was done to analyse drop movement in electric fields under turbulent conditions to analyse coincidence frequency [9]. If such a model could be combined with a functional coincidence criterion – either causal or stochastic - electrocoalescence could be modelled. Alas, the coalescence criterion was still evasive, and much work was done to reveal this.

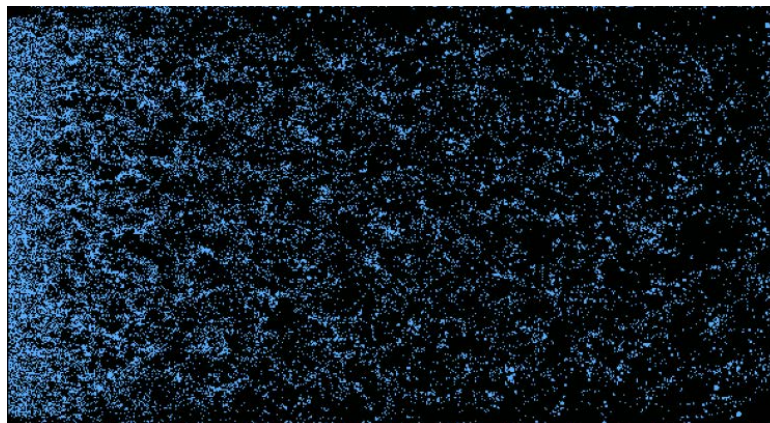


Figure 1-8: Modelling drops in turbulent flow [10]. Coalescence modelled with about 30 000 droplets is simulated simultaneously in a 3 cm× 6 cm two-dimensional channel with electric field.

When two water drops approach in an electric field opposite charges will concentrate at the areas where they are closest and the drops will be attracted to each other, see Figure 1-9 . It was discovered in EC I that the forces could be so strong that the surface deformed into a fast Taylor cone that penetrated into the neighbour drop. This was the normal coincidence for water drops in neat model oils. However, for crude oils and when adding surface covering substances to the oil the behaviour could be more complex [11]. Drops could meet,

exchange charge and be pushed apart, the cone formed could initiate a liquid jet push the approaching drops apart, or they could just be kept together. The hunt for a simple coalescence criterion still remains unsolved. However, it seems likely that the answer has to consider the surface properties of the drops.

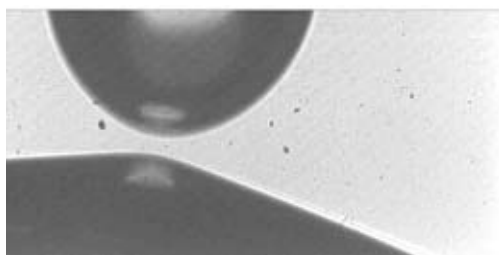


Figure 1-9: Examples of drop merger by Taylor cone instability.

1.5 Activities in electrocoalescence projects

1.5.1 EC I project

The objective in this project was to build fundamental knowledge and models for the physical processes active in electrocoalescence that could serve as basis for designs of a new generation of compact water separators for use in offshore oil production, hereunder:

- Experimental observations of the coalescence process.
- Establish fluid dynamical and electrostatic models of the coalescence process.
- Find possibilities and restrictions for the application of electrocoalescers in respect to varying qualities of crude oil.

The project has had three main activities:

- Experimental investigations of singular and drop pairs in electrostatic fields. Naphthenic model oils and model oils stabilized with asphaltenes were used. High speed video recordings of free falling drops and drop pairs in electric fields were studied [12].
- Building electro-hydrodynamic models that could calculate multiparticle movement under influence of liquid flow and electrostatic forces [13].
- A large experimental investigation with observations of particle movement in turbulent flow with applied electric fields was done using particle image velocimetry [14].

The project gave clear evidence on which mechanisms that govern the electrocoalescence, and on importance of voltage shape and frequency. Also computer models that can be used for studies of coalescer designs were established. Users came aware of possibilities and restrictions for achieving.

1.5.2 EC II Project

The overall objective was to establish know-how and criteria for design and operation of electrocoalescers for realistic water-in-crude-oil emulsions to enable efficient use of the electrocoalescer technology for different well conditions:

- Verify new drop-drop coalescence model for real oils (e.g. heavy crudes, asphaltenes, resins, solid particles etc).
- Establish methods for quantifying electrocoalescence efficiency criteria for crude oils.
- Build model oil/water emulsion test apparatus/rig.

- Reduce use of emulsion breaking chemicals and improve quality of produced water.
- Contribute to increased production in reservoirs with high water content by efficient oil water separation.
- Enable possibilities for subsea and downhole electrocoalescers.

In the electrocoalescence II project one focused on studying the formation of electrostatically induced surface instabilities of drops that would bridge the gap between drops and result in drop merger. This was studied in model oils and also in real crude oils, where a near infrared imaging technique has been developed to also study more realistic cases. Analytical and numerical models were developed to facilitate development of code that will be able to simulate behaviour in a full multi-drop emulsion in compact coalescers. The building blocks were established, but still some work remains to link models with experiments [15].

Coalescence in flowing emulsions has been studied in small scale cell allowing optical observations of drops and also using a rheometer. The results show the importance of having a shear in the flow and of choosing a correct frequency to improve separation efficiency. It is shown how oil conductivity - which is an important parameter for optimizing the frequency applied voltage of a compact coalescer - will vary with crude quality and temperature [15].

More detailed we can mention:

- Drop instabilities –responsible for the coalescence process has been studied in a model system and modelled.
- The drainage time between drops -and the time widow for coalescence between adjacent drops – has been studied and modelled.
- The dynamics of the instabilities has been studied and analytical expressions derived.
- A large setup for studying details in drop coalescence for drops meeting in a shear with a background electric field has been built. This will facilitate test of coalescence models for later inclusion in multi-drop computer models.
- Drop surface rheology has been studied in model systems using both by drops stretched stepped electric field and by a pendant drop rheometer. Influence of surface elasticity and bulk oil viscosity on the Taylor theory for surface instability has been and studied and analytical expressions derived.
- It is found that one in a real crude oil system can consider drops as charge neutral and that the forces acting upon the drops from the applied electric field, is due to electrostatic polarisation of the drops.
- A technique for studying drops in real crude oils by applying NIR-cameras has been developed.
- Studying drop coalescence in a real crude oil shows that model oil systems are not directly relevant for what occurs in a real crude oil.
- A test cell allowing observation drop behaviour in a flowing emulsion has been designed and built. The cell is suited for verification of drop coalescence criteria established in experiments in stagnant oil.
- Coalescence efficiency and dependence on voltage waveform level and shape and shear rate has been studied in a customized rheometer.
- Crude oil conductivity – important for choosing the frequency of the voltage in a compact electrocoalescers –has been measured for several crude oils. It is shown how the conductivity depends on asphaltene content and temperature. The influence of asphaltene aggregate was also studied by dielectric spectroscopy.
- Numerical models for surface covered drops in electric field has been developed allowing for a theoretical modelling of the coalescence behaviour of water drops in a crude oil with a background electric field.

1.5.3 EC III project

The main objective of the EC III project was:

- To establish basic knowledge on the electrocoalescence process to support design and operation of electrocoalescers for emulsions in heavy crude oils.

Sub-objectives were in the start – after a reduced support - adjusted to:

- Experimental verification and mathematical modelling of water drop pair behaviour in shear fluid flow and electric fields.
- Experimental investigation and mathematical modelling of mechanisms facilitating and hindering drop coalescence of water drops with surface-active components in crude oils.
- Characterization of water drop surface rheology using mechanical and electrostatic excitation.
- Investigation of coalescence efficiency for model oil and crude oil emulsions in model coalescer.

The project was organized in three activities, described more in detail in chapters 2 to 5:

- Drop stability in model oil and crude oil.
- Movement of drop pairs in shear flow.
- Testing of coalescence efficiency of crude oil emulsion samples.

1.6 Modelling

During the course of the electrocoalescence projects, a versatile modelling toolbox has been developed.

- **Particle-tracking CFD.** In this approach [9], the water drops in oil are modelled as particles, which is a fair assumption for small drops. A particle-tracking module was developed and added to the in-house CFD code SPIDER. Forces due to the electric field were included, and there was a four-way coupling between the particles and fluid. The action of turbulence on the particles was also accounted for, see Figure 1-8. The particle module is quite flexible, and could be included in other CFD codes, if desired.
- **Detailed simulation of drop dynamics.** A modelling approach based on the level-set method has been developed, see e.g. [16]. This method enables the detailed simulation of drop dynamics, e.g., the oscillation due to a step-change in the electric field. Topological changes, such as break-up and coalescence, can also be handled, although it is challenging to describe the relevant forces. Our model simultaneously accounts for the effect of an electric field, and of surfactants present at the interface. This combination is unique. The model has been further developed in EC III, to include the effect of asphaltenes. This is described in Section 3.
- **A multiscale method to simulate asphaltene-covered droplets.** This newly developed method employs simulations both on the molecular level and on the drop level. It is described in Section 3.

1.7 Experimental techniques and resources established in the projects

1.7.1 Optical studies

An experimental environment for studying drops and drop pairs in electric fields is established, shown in Figure 1-10. Temperature could be varied up to 80 °C. The set-up allowed for high-frame rate video recordings of drop behaviour using a normal fast camera. Voltages up to 20 kV, with varying shape and frequency is available. Lately a vertical moving stage is introduced. With this one can elevate the test cell as a drop is falling so it is kept in view at the optical axis for longer times. Resolution of the images using high focal depth optics is down to 2.5 $\mu\text{m}/\text{pixel}$, and highest full resolution frame rate is 1730 fps. This set-up

allows observations similar to what can be done with a pendant drop instrument, except that one can study fixed volume drops. Using voltage steps it is extremely well suited for studying fast excitation of drop surfaces, important for drop stability and emulsion behaviour.

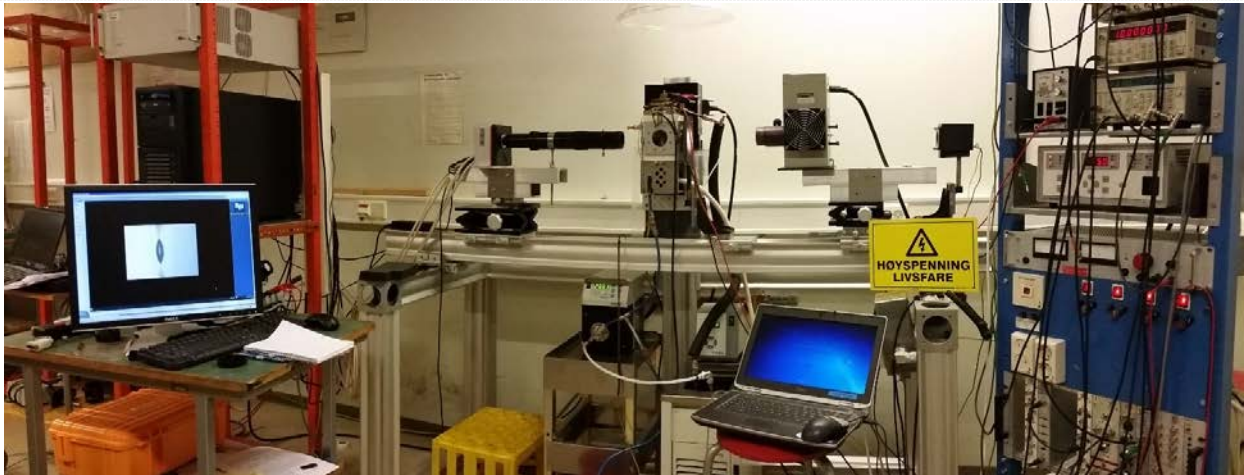


Figure 1-10: Optical bench for studying electrocoalescence. The temperature controlled test cell in the middle is mounted on a lifting stage.

1.7.2 NIR camera

To facilitate observations of water drop behaviour inside crude oil a near infrared (NIR) camera was purchased (Cheetah 640 from Xenics). Crude oils will normally be transparent for near infrared light. One exception is oils with high particle content where light is scattered not absorbed. The camera had a frame rate of 1700 fps and a resolution of 640 x 512 pixels. The camera allows for studies of drop dynamics and surface properties similar to studies of sizzling oil drops water, except that now one can study water in oil which is a non-inverted system. The camera fits well with the optical set up described above.

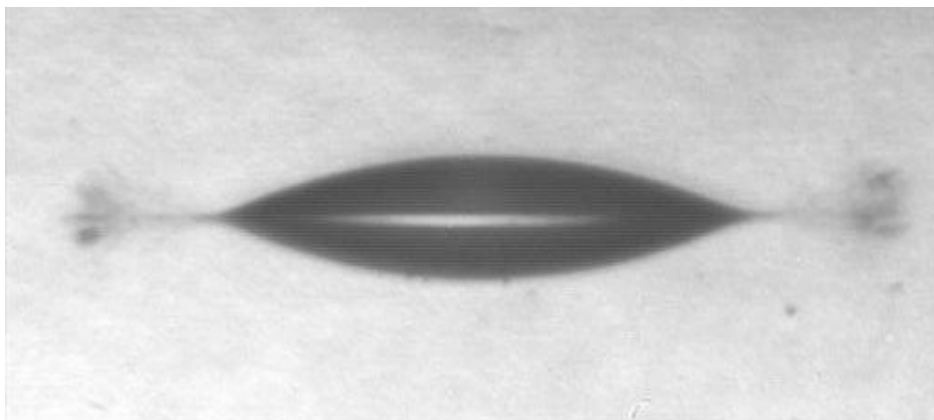
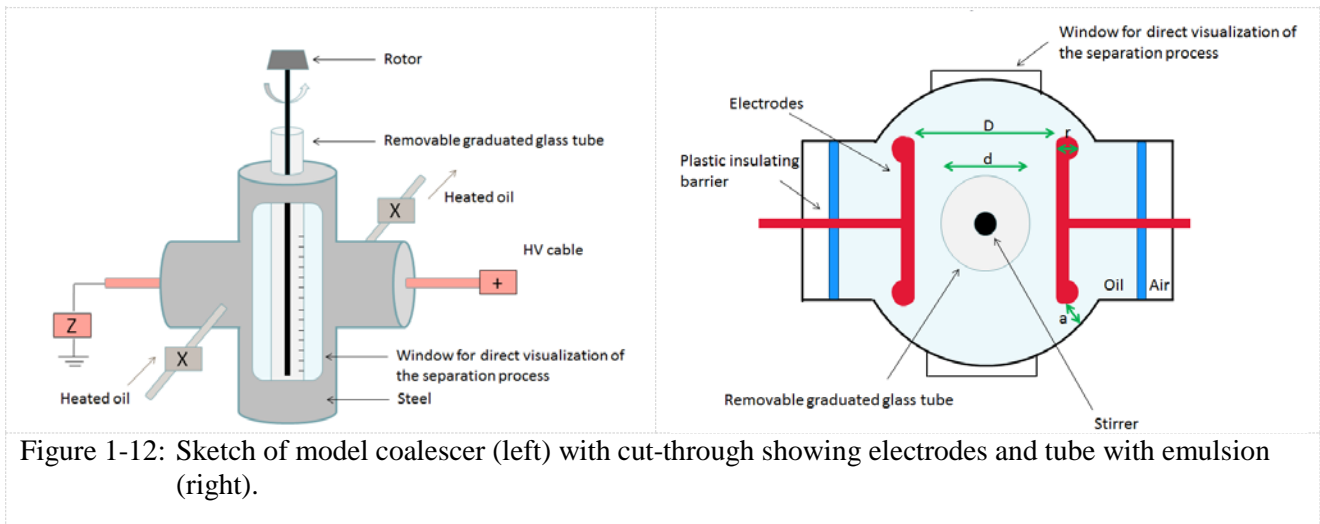


Figure 1-11: NIR Camera pictures of water drop breakup in crude oil.

1.7.3 Model Coalescer

A bench top model coalescer is built for checking coalescence efficiency of emulsions with or without de-emulsifiers. It is inspired from a rheometer design but with very significant improvements of electrode systems, avoiding bare electrodes. Thus coalescence processes taking place will be representative for a bulk process as in a real compact coalescer. The coalescer consists of a rotating bob inside a glass tube where the emulsion is placed. Shear in the resulting Pouseille flow may therefore be varied. The glass tube with the emulsion is placed in between two electrodes in an oil bath. The background electric field is close to uniform, and the oil bath is temperature regulated. The voltage across the electrode gap is controlled by a high voltage amplifier, allowing variation of voltage shape and frequency. The set-up is electrically screened for safe operation.



1.7.4 EHD generation of water droplets (range 20-200 microns) in oil

The injection of water droplets or droplet pairs in oil is obtained by EHD technique, with an obtainable diameter range of 20-200 microns. The method is based on the method developed in ECII, which consists in applying a multistage voltage pulse on an electrode facing a water meniscus hanging at the tip of a grounded metallic needle of about 1 mm outer diameter (Figure 1-13). A multistage pulse of about 1 to 2 ms duration and thousands of volts amplitude is generated by coupling a waveform generator Agilent 33500B and a high voltage amplifier Trek 20/20C. The droplet injection can be observed by high speed video (10000 fps). Although the detachment of the droplets from the meniscus is always obtained after the complete shutdown of the high voltage, the drops are observed being electrically charged. Improvements of the multistage pulse allow reducing that droplet charge to less than 0.01 pC. Droplet pairs can be injected by applying two successive pulses, with a sufficient delay to avoid the disintegration of the first droplet.

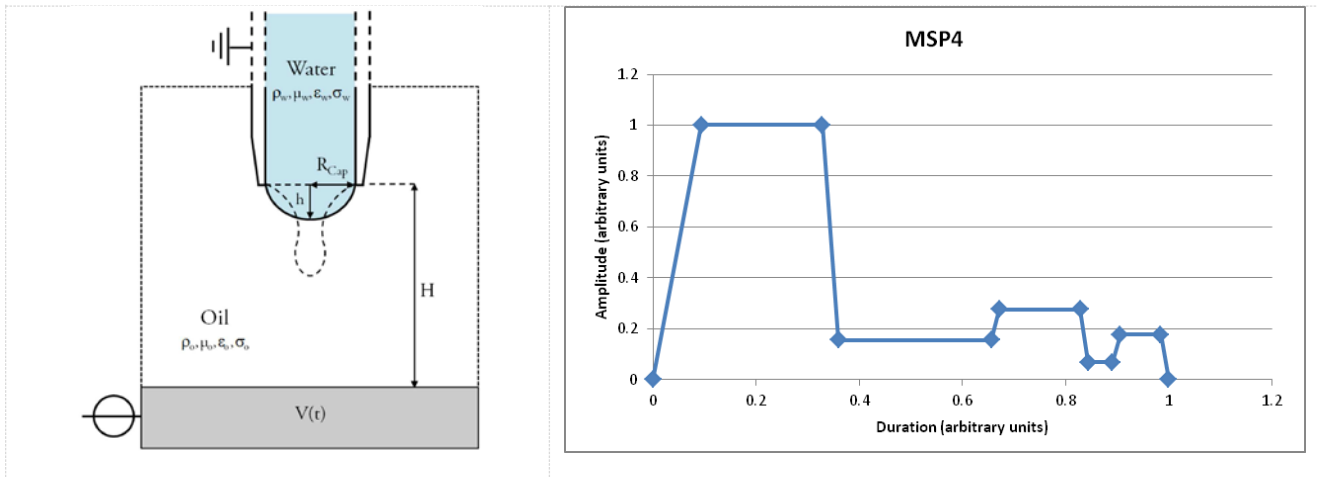


Figure 1-13: EHD injection principle and example of multistage pulse (variable maximum voltage V and duration Δt).

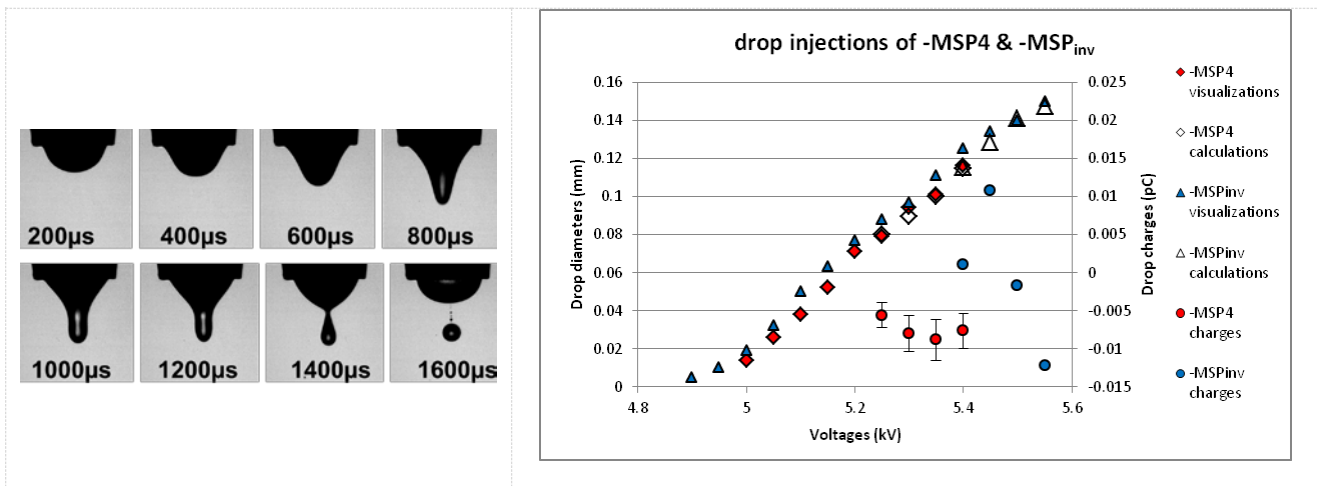


Figure 1-14: Injection of droplet and range of droplet diameter and charge vs maximum applied voltage.

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2 EXPERIMENTAL INVESTIGATIONS OF SURFACE BEHAVIOUR

2.1 Introduction

The purpose of the experimental work has been to measure the dynamic behaviour of single water droplets and coalescence of drop pairs in different oils when the drops are subjected to an electric field. The water drop behaviour is expected to be a function of different system parameters such as interfacial tension, elasticity and viscosity/temperature, together with applied voltage amplitude, frequency and waveform.

In earlier studies, single water droplets were suspended on a solid surface to achieve very long ageing and observation times. However, it was observed that even the most hydrophobic surfaces would influence drop stability by electrostatic interaction with the resting water droplet, sometimes giving strange results. Attempts were also made to suspend droplets in a density gradient column, but these experiments gave very unpredictable interfacial tension values. Hence, it was decided to design a new test set-up for observations of free falling droplets. The first year of this project was mainly used to build, program and test the experimental set-up. This is further described in Section 1.7.

MSc student Torstein Penne has performed a large part of the experiments with single droplets during the last year of the project, supervised by Svein Magne Hellesø and Gunnar Berg. This work has been done in close cooperation with the modelling part of the project, leading to two joint papers (see Section 3 for more details). The main results are presented in the Master's thesis by Torstein Penne [2.1].

Drop-drop coalescence was studied for a droplet falling by gravity towards a stationary, larger water drop at the bottom of the test cell. This part of the project was mainly completion of experimental work from the preceding project Electrocoalescence II, leading to a journal paper on drop coalescence in crude oil [2.2].

The main outcome of the sub-project can be summarized as follows:

- Build, program and test an experimental set-up for observations of free falling droplets.
- 1 MSc (2015).
- 1 journal article in a reputable international journal.

2.2 Experimental approach

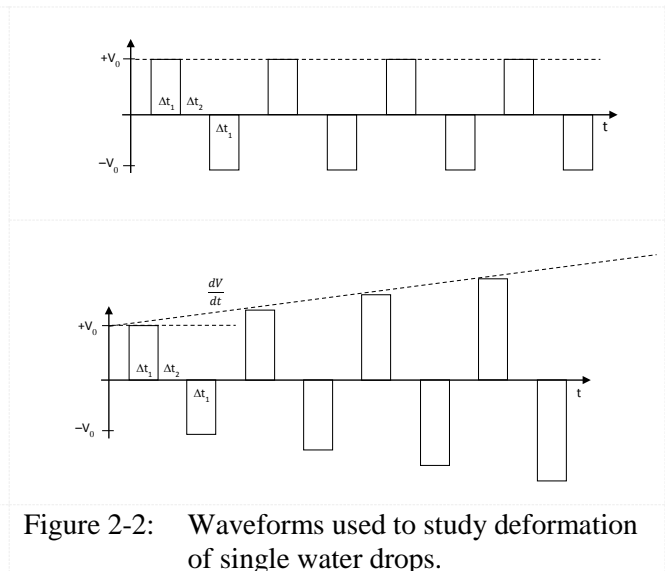
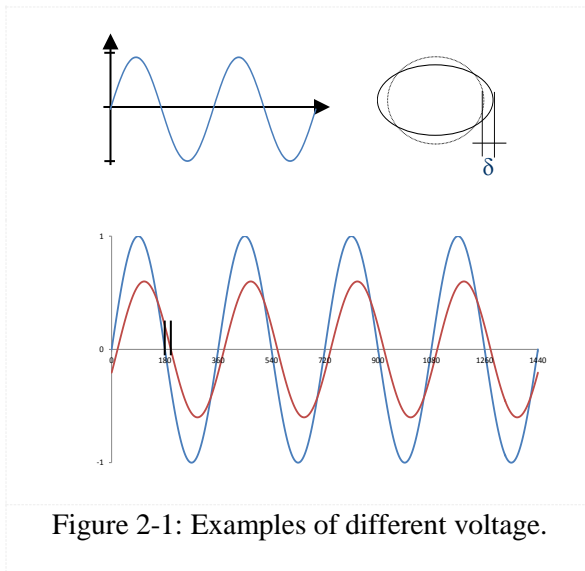
The dynamic behaviour of single water droplets was studied in different liquids:

- Model oil Marcol 52. This transparent oil was added 0.005 wt% surfactant Span 80 for better control of the interfacial tension.
- Crude oils A, B, C, D and E. Crudes B and E had very low viscosity and were very difficult to study due to the high velocity of the falling water droplets.
- Crude D added 1000 ppm demulsifier EB-8228.

The water used in the experiments was MilliQ-filtered water added 3.35-3.36 wt% salt (NaCl), with an estimated relative permittivity of $\epsilon = 82$ at room temperature.

In all experiments a vertical electric field configuration was used, giving 2D rotational symmetry which is easier to use in numerical modelling. By applying different voltage shapes it is possible to study different properties of free falling water drops.

- With no voltage applied, terminal velocity and drag coefficient can be measured for varying degrees of surface covering. It is assumed that an electric field will modify the drag coefficient of drops with polar surfactants.
- Using a sine wave voltage the water drop surface will oscillate with twice the excitation frequency. As the drop surface dynamics are dominated by viscous effects, higher oscillation modes are strongly damped. The experiments will determine the viscous damping of the drop. In this case the maximum drop elongation a_{\max} and oscillation amplitude $\delta = a_{\max} - a_{\min}$ is measured as a function of voltage frequency.
- Using a pulsed voltage waveform the drop surface will be strongly deformed for a short time duration (impulse response). Surface oscillations can be studied during excitation and relaxation of the drop. Repeated voltage pulses enable several observations/experiments using the same water drop, ensuring that the surface ageing time and drop diameter is constant. The experiments will determine drop stretching, oscillation resonance frequencies and damping caused by interfacial tension and elasticity (depending on drop ageing time), bulk oil viscosity (depending on temperature), and applied voltage amplitude. As the drop will be subjected to consecutive voltage pulses there is a possibility that the interfacial layer can be altered by the surface stretching. This should be observed as a change in e.g., drop axis ratio $(a/b)_{\max}$, characterizing the interface stability. Two different variations of this experiment are possible. Using an increasing or decreasing voltage amplitude in the pulse train will then give several data points on a drop elongation curve (a/b as a function of reduced electric field E^*), strongly reducing the required number of experiments.
- A sine wave voltage is used to find the phase shift between the applied electric field and drop deformation. To reduce the influence of bulk viscosity the voltage frequency should be kept as low as possible. In this work a frequency of 5 Hz was used, giving a drop oscillation frequency of 10 Hz. It is then possible to measure several deformations for each drop.



2.3 Summary and conclusions

The experiments using square wave voltages show that surface properties of a water drop may change very fast, meaning that the effect of previous deformations cannot be neglected. Depending on the oil, previous deformations can weaken or strengthen the drop. This is seen in the results respectively as an increase or decrease in stable and maximum aspect ratio of the water drop between two consecutive deformations. The effect is larger for crude oils with higher concentration of surfactants, and is easily seen in Figure 2-3. The effects of increasing vs decreasing square waves are not as apparent, and the temperature of the oil seems to have a larger effect on the effective surface tension than the signal sequence.

Droplet oscillation frequency is the parameter that shows the least change due to previous deformations. The fact that oscillation frequency values are so similar could mean excitation by square waves is an effective way of determining the actual resonance frequency of an excited droplet.

The contractive forces from surface tension come from the drops resistance against increases in surface area, and step excitation of a water drop will give (over)damped surface oscillations. All damping coefficients have shown an increase with temperature. This is an indication that treating an electrically excited water drop as a simple damped harmonic oscillator is oversimplifying the problem. For a harmonic oscillator the results would imply that all the oils tested have increased viscosity at increased temperatures, which is not the case. Looking not only at the values of damping coefficients, but also at the qualitative behaviour of the droplets at different temperatures it is clear that the viscosities are lower at higher temperatures. This is observable as e.g., an increase in settling speed. At 50 °C the amplitude ratio of consecutive oscillations is not constant, as would be expected by underdamped oscillations, but largest between the two first oscillations. This implies that the oscillations do not act as an underdamped oscillator.

The theory of harmonic oscillators implies that the phase shift of drop deformation will decrease with viscosity. Oils C, D and D with demulsifier confirm this, and with more experiments Marcol is expected to do the same. It seems that not only the oil viscosity, but also the adsorption of impurities on the drop surface has an effect on the phase shift. The phase shift of deformation is related to energy loss in the droplets. These results imply that if drop heating by energy loss in deformation is an issue, the effect of surface active components in the oil must also be considered.

The fact that previous deformations affect the deformation and stability of the droplets could explain why electrocoalescers might be ineffective. Ideally the electrocoalescer should take into consideration not only the chemical properties of the oil/water mixture, but also for how long the two phases have been mixed. Properly controlling the amount of time before the oil/water mixture is sent into the electrocoalescer, and the residence time in the electric field, might be crucial to effective separation.

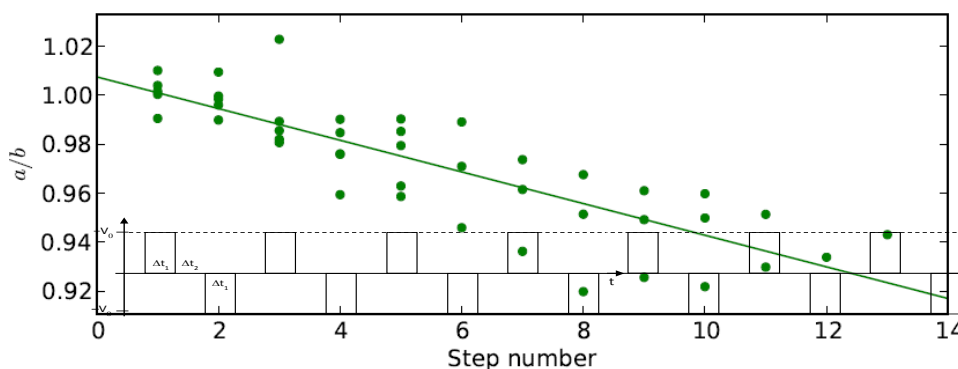


Figure 2-3: Example of observed reduction in drop elongation (semi-axis ratio a/b) in crude A during 14 consecutive constant voltage steps. The pulse width and pulse distance is 50 ms.

Drop-drop coalescence experiments were performed in Grane crude oil. In this study a bipolar square voltage (BSV) was used to get stable electrostatic forces. Using a BSV waveform gives a phase independent electrostatic force on non-charged drops, which is equivalent to applying a DC field, but with the advantage of a capacitive voltage distribution in the electrode gap. The high voltage was applied to the lower electrode after the smaller drop was released, to ensure that the falling drop was uncharged. Experiments based on the use of a NIR video camera provides new and unique results concerning electrocoalescence in crude oil which should be of interest for the application of compact electrocoalescers. The electric field induces forces that affect both the water drop motion in crude oil and the coalescence process. Up to a threshold field, the surface charge density redistributed just after bridging of the two drops and charge exchange slows down the coalescence process. Above this threshold field, the coalescence was not total and two different transitions were found: From full coalescence to partial coalescence at 60 °C (see Figure 2-4) and to non-coalescence at 40 °C. Each of these two scenarios have been observed in the case of model oils without resins and asphaltenes, but the fluids properties determining either the partial coalescence or the bouncing behaviour are not yet well characterized. The peculiarity of the studied crude oil is that there was a scenario change by changing the working temperature, which affects many properties of the system.

For model oils, the transition to partial coalescence or bouncing is not really characterized in terms of applied field, interfacial tension, properties of the oil-like permittivity and viscosity, water conductivity, presence of surface-active compounds, etc. The generation of a thread connecting the two drops and its initial shape appear to play a crucial role: a small length to radius ratio of the water bridge should lead to partial coalescence, whereas a large one should promote electrical bouncing. In case of a water drop falling in a crude oil, the problem is further more intricate because of the presence of various surface-active compounds that modify the static interfacial tension and might influence the dynamic evolution of the interface subjected to a shear stress. The presence of asphaltenes and resins is of primary importance because these compounds can build an interfacial film promoting viscoelastic behaviour of the drops. The present study suggests the existence of such a film at 60 °C, but its absence or its negligible influence at 40 °C. Therefore, the understanding of the phenomenon of electrocoalescence in crude oils requires first a better knowledge of the interfacial film whose rheological properties are far from being fully clarified.

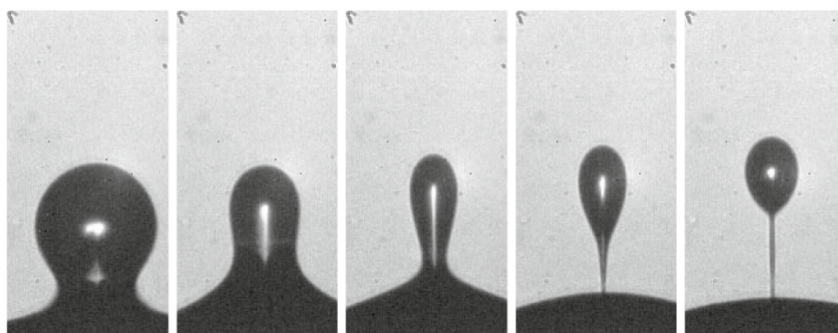


Figure 2-4: Partial coalescence in Grane crude oil at 60 °C.

2.4 Deliverables

Publications

- 2.1. M.Sc. thesis Torstein Penne: ["Electrical Excitation of Water Drops in Oil"](#), NTNU, Trondheim 2015.
- 2.2. S.M.Hellesø, P.Atten, G.Berg, L.E.Lundgaard: ["Experimental study of electrocoalescence of water drops in crude oil using near-infrared camera"](#), *Exp. Fluids*, vol. 56, 2015.

3 MODELLING OF INTERFACIAL BEHAVIOUR

3.1 Introduction

One of the main hypotheses behind the present project is that in order to understand the electrocoalescence phenomenon, one needs to understand droplet dynamics in an electric field. In the previous electrocoalescence project (ECII), it was shown that surface-active agents (surfactants), which induce a varying interfacial tension, can significantly affect droplet dynamics. In practice, other components, such as asphaltenes, will also be present at the interface. The goal of the present sub-project has been to describe their influence on the forces acting at the drop interface, and hence on the droplet dynamics. The present sub-project has cooperated closely with the experimental activity (Section 2), and this has led to two papers [3.4, 3.5].

Due to the limited available resources, the bulk of the research has been carried out in the form of a PhD project. The candidate, Åsmund Ervik, was hired in January 2013 and the thesis was submitted in March 2016. (The slight delay with respect to the three-year duration is due to parental leave.) Ervik has been supervised by Bernhard Müller (NTNU) and Svend Tollak Munkejord (SINTEF). As part of the doctoral work, Ervik spent a week at G2ELab (Section 4) in Grenoble, and six months (October 2014 - March 2015) at Imperial College, London, collaborating with Professor Erich A. Müller and others in the Molecular Systems Engineering group.

One MSc candidate, Morten Olsen Lysgaard, was supervised as part of this activity during autumn 2014 and spring 2015. The supervisors were Åsmund Ervik, Bernhard Müller and Helge Holden (NTNU) and Svend Tollak Munkejord (SINTEF).

The main outcome of the sub-project can be summarized as follows:

- 1 PhD (to be defended in June 2016).
- 1 MSc (2015).
- 1 journal article in a reputable international journal and 5 more expected.
- 2 conference articles.
- Further development of the level-set code from ECII, in particular with a model to simulate elastic interfaces.
- A parallel version of the level-set code (with a reduced model subset at the moment).
- A framework to ease the set-up of relevant molecular-dynamics simulations (open source).
- A web application for obtaining molecular models for a large number of chemicals (open source).

In the following, the main results will be briefly summarized. The deliverables, which consist of Åsmund Ervik's PhD thesis and 8 associated papers, as well as Morten Olsen Lysgaard's MSc thesis, are listed in Section 3.7. These deliverables contain detailed descriptions of the work, much this detail is omitted here for the sake of clarity and brevity.

3.2 Our approach

An important motivation behind the present work is the hypothesis that to understand emulsion behaviour, understanding single-drop and drop-pair behaviour is essential. Thus we have focused the attention to the dynamics of a single drop, and the associated physical modelling. In particular, modelling and experiments should be carefully set up to study the same phenomena, as described in [3.3, 3.5].

In ECII, our level-set code was equipped to handle the influence of electric fields and insoluble surfactants, a combination which was quite unique. However, it could not handle one important effect which is observed for crude oils, namely, the tendency for the interface to be rigid. It is believed that large molecules present in the crude oil, called asphaltenes, are responsible for this behaviour. As part of the work in ECIII we have constructed a multi-scale approach that can handle this effect. It was demonstrated that this method is able to capture the surface rigidity, see Figure 3-1.

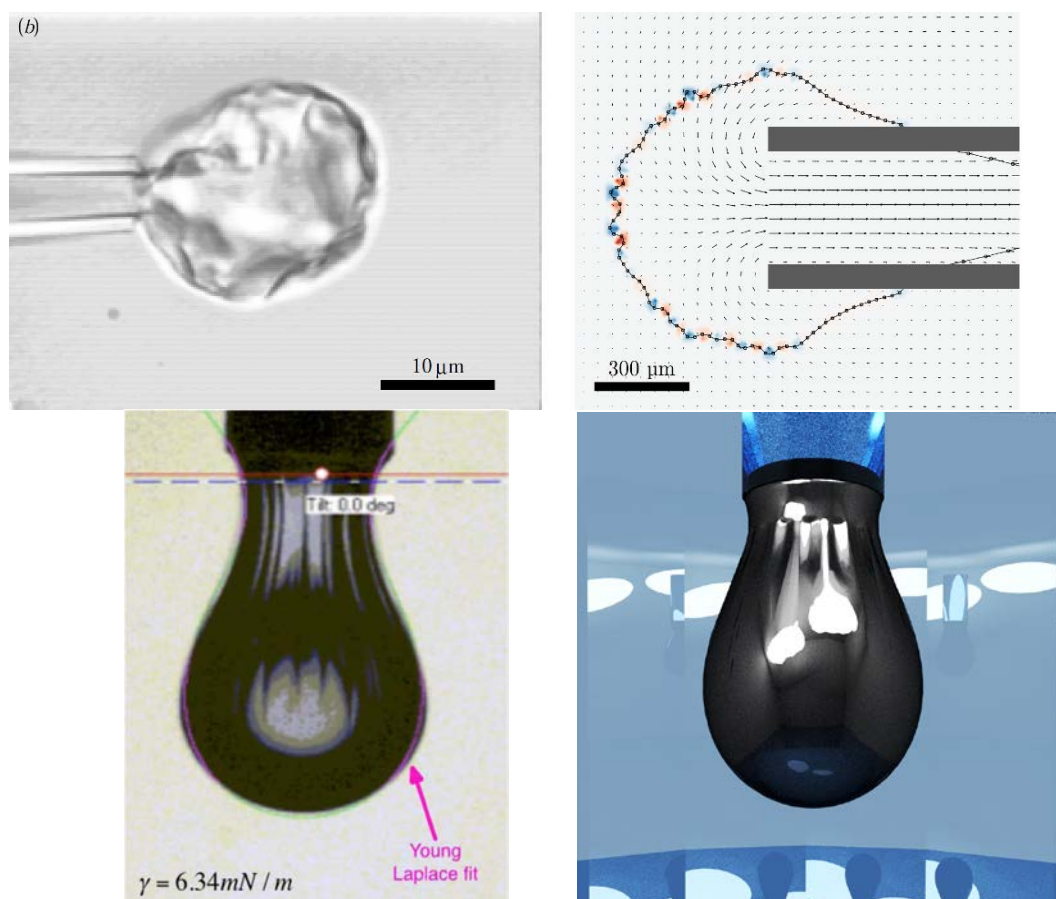


Figure 3-1: Pulling water out of an asphaltene-covered drop leads to crumpling. Top row: very small droplet where gravity is unimportant, symmetric crumpling regime. Bottom row: larger droplet where gravity and interfacial properties together lead to rotationally symmetric crumpling regime. Right: Our simulations. Left: Experimental observations, by Yeung et al.¹ (top) and by Pauchard et al.² (bottom).

3.3 Studies of water drops in model oil

In order to augment our physical understanding and aid model development, experiments should be performed with well-defined systems. In the case of electrocoalescence, this means that the study of model oils with known properties is particularly relevant. Standard modelling approaches normally assume the fluids to be pure. In the laboratory, however, it is essentially impossible to obtain perfectly pure organic fluids: the only reported falling drop experiments in the literature which obtain pure fluid behaviour are

¹ A. Yeung et al., On the interfacial properties of micrometre-sized water droplets in crude oil, *Proc. R. Soc. A* (1999) **455** 3709-3723

² V. Pauchard et al., Asphaltene-Laden Interfaces Form Soft Glassy Layers in Contraction Experiments: A Mechanism for Coalescence Blocking, *Langmuir* 2014 **30** (43), 12795-12803

conducted with metallic liquids such as mercury, molten lead or molten beryllium trioxide. The inability to obtain pure fluid behaviour in ordinary liquids was the subject of much research in the first half of the 20th century; see e.g. the book by Leal³. The currently accepted hypothesis is the stagnant cap model which explains how surfactants cause small drops to fall slower than predicted by pure fluid theory. However, in publication [3.6], together with Dr. Erik Bjørklund from project partner Wärtsilä, we demonstrate that the stagnant cap model is not internally consistent, and propose a new model which we argue is superior.

In the experimental and simulation work in this project, the challenge of unknown surfactants present in even high-purity oil was overcome by “overwhelming” the surfactants present in the oil by adding Span 80 surfactant in known quantities, whose effect could be modelled by our level-set code, see Figure 3-2. This figure shows the stretching of a water drop falling in Marcol oil with Span 80. Two things are worth noting here. First, the simulation not accounting for surfactant gives larger deformation than in experiments, but a steady-state deformation that agrees very well with the Taylor model for equilibrium deformation (which assumes pure fluids). Second, when surfactant is accounted for in the level-set code, the calculated deformation agrees with the experiment to within the experimental uncertainty.

Figure 3-2 (right) illustrates the effect of surfactant in a different way, by comparing four different simulation results. The four quadrants show the same 0.5 mm diameter water droplet subjected to a 400 V/mm electric field, but with different amounts Span 80. The black vectors illustrate the force at the interface. For the case of no surfactant, the force is perpendicular to the interface. As the surfactant concentration is increased, the interfacial force gets a tangential component from the Marangoni force. This force works against the motion of the interface, damping the deformation. However, the addition of surfactant also decreases the interfacial tension, which gives a larger deformation. This two-fold effect of adding surfactant is explained in more detailed in the paper.

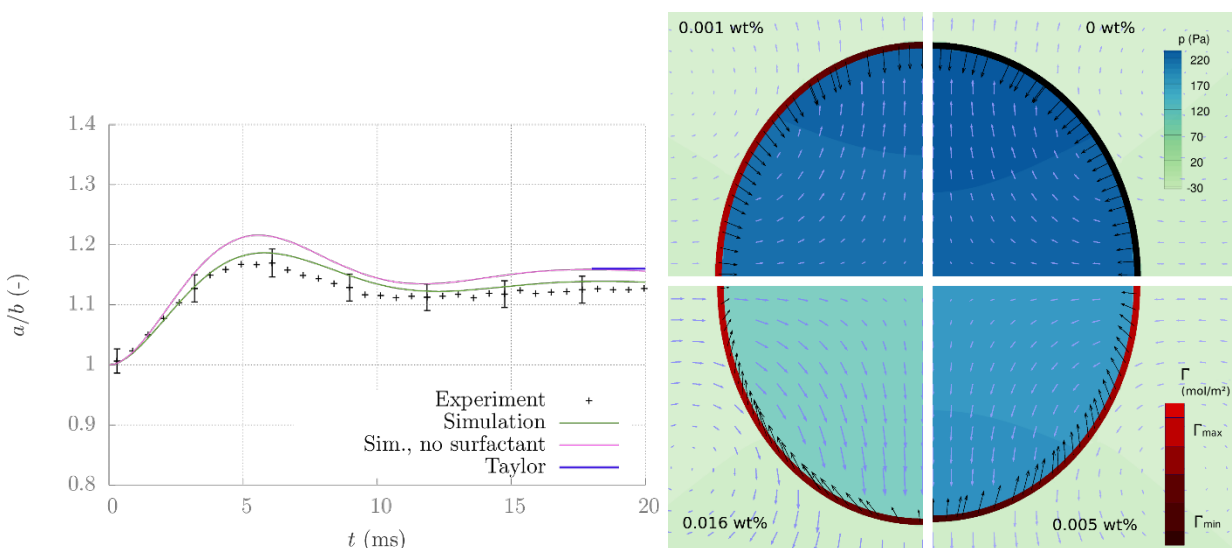


Figure 3-2: Left: Relation between semi-major and semi-minor axis. Simulation and experimental results for a 958 μm water drop falling through Marcol with 0.015 wt% Span 80 subjected to a 400.0 V/mm field [3.3]. Right: The effect of surfactant concentration on normal and tangential interfacial stress for a 0.5 mm diameter drop subjected to a 700 V/mm electric field. Comparison of the pressure field (blue/green), the vector $\gamma\kappa n - 100 \nabla(\gamma)$ (black vectors), the flow field (sky-blue vectors) and the surfactant concentration at the interface (red to black) [3.5].

³ L. G. Leal. *Advanced transport phenomena: fluid mechanics and convective transport processes*. Cambridge University Press, 2007.

Finally we have observed in our experiments a significant hysteresis effect when repeatedly stretching drops at high surfactant concentrations. But these effects are only seen for the case where the applied deformations are first small and then increased, not in the opposite case when the deformations are first large and then decreased. We have investigated whether this effect can be explained just by the hydrodynamics and surfactant transport, and have found this not to be the case. One may speculate that the small initial deformations cause a phase change in the adsorbed surfactant layer, causing it to become more pliable. Furthermore, the effect of interfacial elasticity may be important also for model-oil systems.

3.4 Studies of water drops in real crude oil

The main objective of the collaboration with Imperial College was to construct a multiscale model for simulating the interface between water and real crude oils. This multiscale model consists of a nanoscale and a macroscale component, which are linked by parameters computed at the nanoscale being used as input on the macroscale. The macroscale model consists of the existing level-set code extended with an immersed boundary method for simulating the elastic interface. This method was implemented in the MSc work of Morten Olsen Lysgaard. The nanoscale model consists of molecular dynamics simulations performed using state-of-the-art models for the crude oil components, in particular the asphaltenes. These models have been (and are being) developed at Imperial College. Results from the multiscale model show great promise, and have been the subject of an oral presentation at the Petrophase conference in June 2015 (a prestigious and selective international conference), and at the Tekna separation conference in September 2015. The model is described in the MSc thesis of Morten Olsen Lysgaard [3.4] and in the submitted publication [3.9], from which Figure 3-1 is taken. The model can be summarised using Figure 3-3:

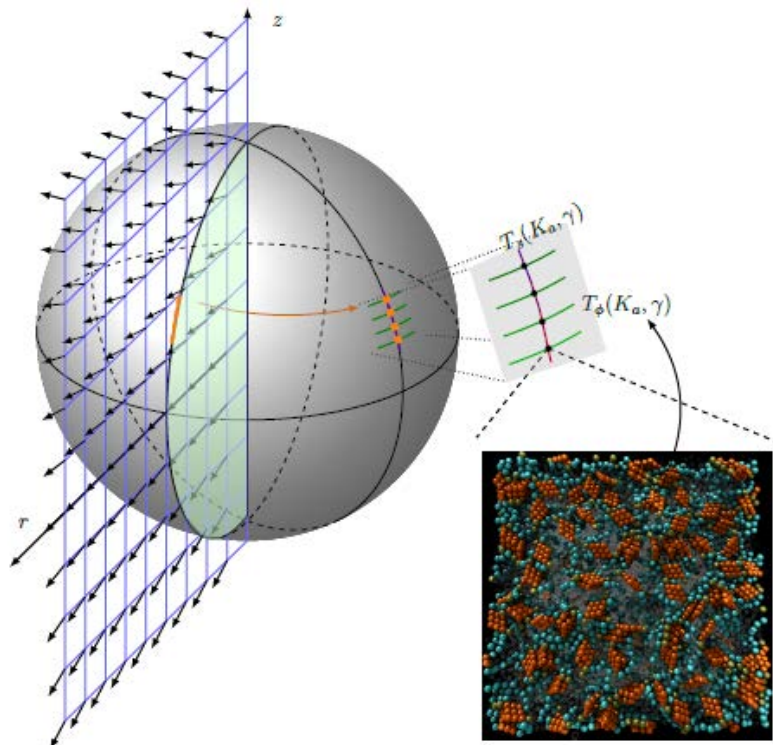


Figure 3-3: The asphaltene-covered drop is shown in grey. The grid with vectors illustrates the macroscale simulation domain, where the interface is tracked with the level-set (green hemicircle) and immersed-boundary (orange points) methods. The immersed-boundary method takes into account the tensions T_s and T_ϕ illustrated with green and red lines in the gray zoom-in area. The tensions are calculated using the interfacial elasticity and tension, K_a and γ , which are calculated from the nanoscale method (lower right corner).

Using this method, the effect of mechanical (as opposed to Gibbsian) interfacial elasticity on the flow inside and around droplets can be taken into account. As shown in Figure 3-1, simulations using this approach show very close agreement with experimental results in the literature. In future work, this approach can be extended to studying the effect of other crude-oil components, effects of temperature and pressure, etc.

3.5 Molecular modelling tools

As part of the collaboration with Imperial College London, two open-source molecular simulation tools were developed and used in the multiscale method. The first of these tools is called Bottled SAFT, a web application providing molecular simulation parameters for over 6000 fluids. Bottled SAFT is freely available at www.bottledsaft.org and is described in more detail in paper [3.8]. Bottled SAFT is linked to the other tool, raaSAFT, which is a molecular simulation framework that uses the parameters to enable molecular simulations in a straight-forward fashion. Together, these tools provide an unprecedented ease-of-use of molecular simulations. The use of GPU acceleration through the underlying HOOMD-blue⁴ open-source simulation code (from U. Michigan) means raaSAFT can be used to study very large systems over very long time scales. Paper [3.7] describes raaSAFT in detail, and a benchmark polymer simulation with 1.2 million atoms is used to demonstrate the capabilities of the tool; this simulation would take more than a year to complete using conventional approaches.

3.6 Conclusions and suggestions for further work

3.6.1 Conclusions

The use of CFD modelling to analyse phenomena such as electrocoalescence enables a quantitative discussion of each modelled physical effect. The description of the dynamics of droplets subjected to an electric field is one important stepstone to understanding the electrocoalescence phenomenon. Good quantitative agreement has been obtained between simulations and experiments for model-oil systems with added surfactant. The most important remaining hurdle is the description of interfacial properties for real crude oil-water systems. We have obtained important initial results for this system by applying the multi-scale approach developed in this project. This approach shows great promise, but should be tested further and quantitatively validated against experiments.

3.6.2 Suggestions for further work

- Explore further the elastic-interface model for electrocoalescence. One particular effect which has not been incorporated in the present model, due to time constraints, is the bending rigidity of the interface. Including this would be straight-forward. Other avenues of investigation include the effects of different asphaltene molecular structures, the effects of other crude oil components, the effects of temperature and pressure, etc.
- Experimental investigations using the pendant drop tensiometer, in order to provide further validation of the model, will also be of interest in future work.
- Complete the parallelization of our level set/immersed boundary model, in order to enable the simulation of multiple drops in a flow field.
- Continue the line of research with closely coupled experiments and modelling, e.g., using a model system with controlled additions of asphaltenes.
- Modelling the presence and movement of charges at the interface, as well as the effect of the electric field on the interfacial configuration of asphaltenes, will also be interesting topics.

⁴ J. A. Anderson et al., General purpose molecular dynamics simulations fully implemented on graphics processing units, *Journal of Computational Physics*, **227** (10) 2008, 5342-5359.

3.7 Deliverables

The PhD thesis [3.10] by Åsmund Ervik was submitted in March 2016, and was successfully defended on 24 June 2016. The associated papers [3.1–3.3] have been published. The associated papers [3.5-3.9] are under consideration for publication or have recently been accepted for publication. The papers and the thesis can be read using the provided links.

The MSc thesis by Morten Olsen Lysgaard [3.4] was submitted in June 2015 and received an A grade.

Publications:

- 3.1. Å. Ervik, K.Y. Lervåg, S. T. Munkejord: "[A robust method for calculating interface curvature and normal vectors using an extracted local level set](#)", *Journal of Computational Physics* **257**, Part A, 259–277, 2014. & [External link](#).
- 3.2. Å. Ervik, S. T. Munkejord, B. Müller: "[Extending a serial 3D two-phase CFD code to parallel execution over MPI by using the PETSc library for domain decomposition.](#)" In: *10th International Conference on Computational Fluid Dynamics in the Oil & Gas, Metallurgical and Process Industries, CFD2014*. SINTEF, Trondheim, Norway, 17–19 June 2014. & [External Link](#).
- 3.3. Å. Ervik, S. M. Hellesø, S. T. Munkejord, B. Müller: "[Experimental and computational studies of water drops falling through model oil with surfactant and subjected to an electric field](#)". In: *18th International Conference on Dielectric Liquids, ICDL 2014*, IEEE, Bled, Slovenia, 30 June–3 July 2014. & [External link](#).
- 3.4. M. O. Lysgaard: "[Development of an immersed boundary method for simulating contaminated fluid interfaces in two-phase flow](#)". Master's thesis, Dept. of Mathematical Sciences, Norwegian University of Science and Technology (NTNU), June 2015. & [External Link](#).
- 3.5. Å. Ervik, T. Penne, S. M. Hellesø, S. T. Munkejord, B. Müller: "[Influence of surfactants on the electrohydrodynamic stretching of water drops in oil.](#)" *Submitted to Physical Review Fluids*, 2016.
- 3.6. Å. Ervik, E. Bjørklund: "[The admissible surfactant distributions and velocities for small falling drops](#)" *Submitted to Journal of Fluid Mechanics*, 2016.
- 3.7. Å. Ervik, G. Jiménez-Serratos, C. Herdes, E. A. Müller: "[raaSAFT: a framework enabling coarse-grained molecular dynamics simulations based on the SAFT-g Mie force field](#)". *Accepted for publication in Computer Physics Communications*, 2016. & [External Link](#).
- 3.8. Å. Ervik, A. Mejía, E. A. Müller: "[Bottled SAFT: a web app providing SAFT- \$\gamma\$ Mie force field parameters for thousands of molecular fluids](#)". *Accepted for publication in Journal of Chemical Information and Modelling*, 2016. & [External Link](#).
- 3.9. Å. Ervik, M. O. Lysgaard, C. Herdes, E. A. Müller, S. T. Munkejord, B. Müller: "[A multiscale method for simulating fluid interfaces covered with large molecules such as asphaltenes.](#)" *Accepted for publication in Journal of Computational Physics*, 2016.
- 3.10. Å. Ervik, "[Multiscale modelling using molecular dynamics and interface-capturing methods for two-phase flow simulation of droplets covered with surfactants or asphaltenes, and applications to electrocoalescence](#)". PhD thesis, Dept. of Energy and Process Engineering, NTNU. Successfully defended 24 June 2016.

4 STUDIES IN GRENOBLE: EXPERIMENTS ON DROPLET PAIR INTERACTION.

4.1 Introduction

Among the conclusions of ECII project it was stated that experimental data on the electrocoalescence of small droplets will be useful to improve the understanding of the dynamics of water-oil interfaces and to validate numerical models. The goal of the researches proposed in Grenoble concerned initially experiments in shear flow, but as a first step falling small droplets in stagnant fluid and under electric field aligned with the droplet pair axis was addressed. That simpler configuration already showed the difficulty on generating and controlling very small droplet pairs (in the range 20-200 microns diameter) and was found easier to compare with models, thanks to the axial symmetry.

The experimental research has been carried out as the PhD project of You Xia, supervised by Jean-Luc Reboud (Grenoble Electrical Eng. Lab. – Univ. Grenoble Alpes). Comparisons with the results of numerical models performed using commercial software Comsol Multiphysics™ helped characterizing the experimental conditions and the behaviour of droplet pairs under electric field.

The main results will be briefly summarized hereafter. The deliverables are You Xia's PhD thesis (submission planed in March 2016) and associated papers: 2 conferences during the PHD and 2 papers in preparation.

4.2 Development of experimental set-up

First main challenge is the generation of very small droplet pair (range 20-200 microns) aligned with electric field. Droplet-on-Demand generation by EHD method was improved: its main interest is the control, in the required range, of the diameter of droplets injected from a single metallic needle, which is obtained by varying the applied high voltage (the desired range cannot be obtained with other methods). Nevertheless, it was observed that the droplets were a little electrically charged. Improvements of the method were proposed to control the droplet charge and characterise from both electrical and hydrodynamic point of view the initial state of the droplet pair. Special attention was paid to the visualisations: the droplets are very small and thus their falling velocity is also very small (typically 0.1 mm/s, in Marcol™ model oil): accurate observation by video involves strong zooming and multiple angle of view (4 CCD cameras + high speed camera). Moreover the influence of convection flows and remaining electric space charge in oil may be strong.

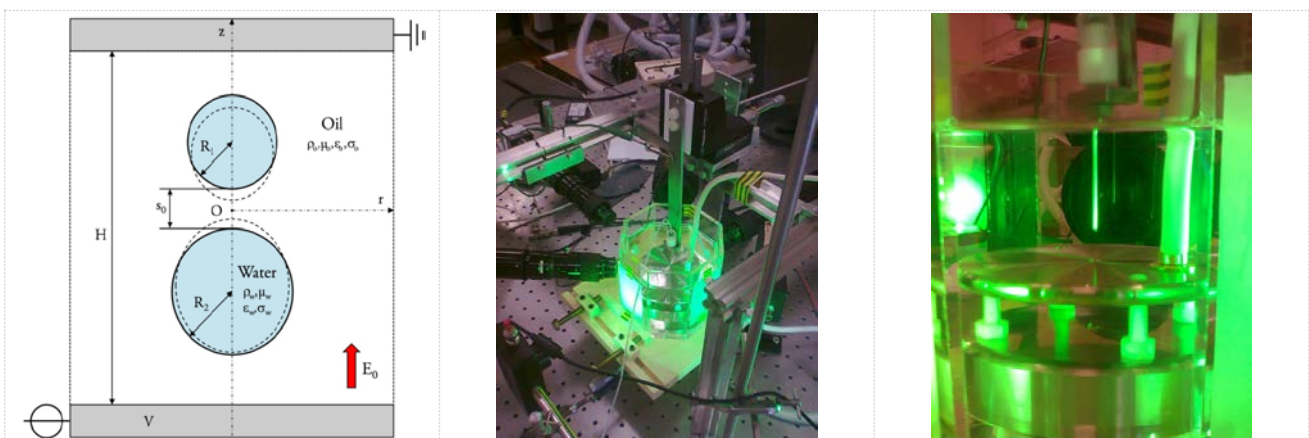


Figure 4-1: Left picture: scheme of droplet pair configuration in electric field; central picture: octagonal plexiglas cell, video cameras and optics; right picture: zoom on metallic needle and HV electrode).

4.3 Validation of drop diameter and charge from falling velocity and coalescence conditions

Electrical and hydrodynamic characterisation of the droplet pair and its coalescence are mainly deduced from the analysis of falling velocities, with and without applied DC electric field. As illustrated on Figure 21, a complete data set of droplet position and velocity is deduced from video. Cross analyses of different angle of view allow verifying that axis of symmetry of the drop pair, electric field and gravity are well aligned and a few high speed videos (8000 fps) of the coalescence process were obtained.

Modelling the different terms of hydrodynamic and electrostatic interactions between droplets allows deducing from the recorded velocities their respective mass and electric charge. When coalescence occurs, a record of the resulting single droplet velocity, with and without applied voltage, allows controlling the mass and charge conservations and validating the method (Figure 21).

A first data set was constituted of about 40 different cases, with varying droplets pair (with a limited diameter range to remain with falling velocities between 0.1 and 0.3 mm/s) and varying applied DC or AC voltage. Analyses of the results and experimental uncertainties allow performing some recommendations for future work. First comparisons with numerical simulations using Comsol Multiphysics™ software are promising for the stage preceding the merging of the droplet interfaces. Further comparisons with the models developed in Trondheim should be the next step.

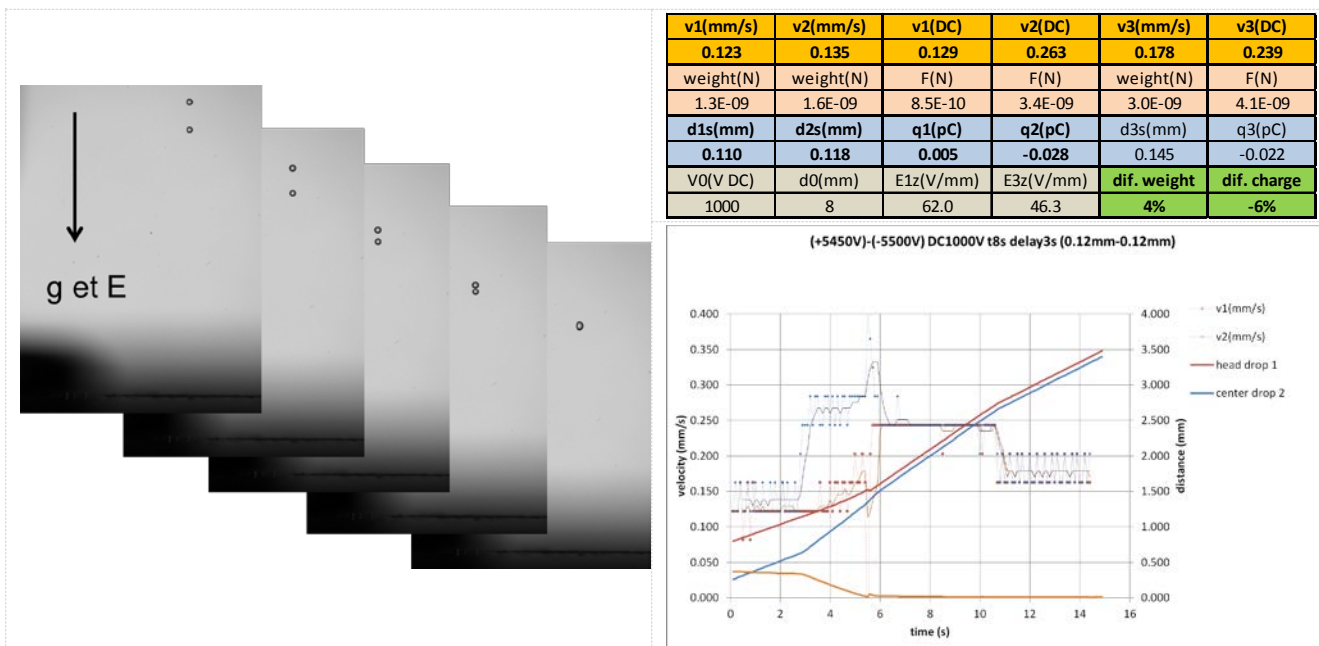


Figure 4-2: Example of successive states of falling droplet pair and electrocoalescence under DC field. Figure on the right shows, with respect to time, the position and velocity of the droplets, before and after coalescence. From applied voltage (1 kV DC, from $t = 3\text{s}$ to 11s) and spacing (8 mm) between the needle and the electrode, the electric field varies a little with vertical position around the average value of 54 V/mm. Conservation of mass and charge are verified in that case respectively within 4% and 6%.

4.4 Deliverables

Experimental resources

Preliminary planning, the PhD thesis [4.5] by You Xia would be submitted in March 2016, and defended in May 2016. The associated papers [4.1-4.2] have been published. The associated paper [4.3] is under preparation.

Publications

- 4.1. Y. XIA, J.-L. REBOUD, P. ATTEN, 2014, "[Influence of viscosity and interface tension on water droplet injection in oil](#)", 18th. Int. Conf. on Diel. Liq. (ICDL) IEEE, June30-July3, Bled, Slovenia
- 4.2. Y. XIA, JL.REBOUD, P. ATTEN, 2014, "[EHD Injection of Droplet Pairs of Micrometric Size in Dielectric Liquids](#)", 9ème conférence de la Société Française d'Electrostatique, 27-29 août 2014, Toulouse, France,
- 4.3. Y. XIA, JL.REBOUD, P. ATTEN. "[Experiments on droplet pair interaction in electric field](#)". *SFE conference, Poitiers, 2016.*
- 4.4. Y.XIA. "[Experiments on EHD Injection, Interaction and Electrocoalescence of Water Droplet Pairs in Oil](#)". PhD thesis. Univ. Grenoble Alpes. 2016.

5 MODEL COALESCER

A new bench size model coalescer was built with an internal Couette flow in the emulsion, with insulated electrodes and a large active volume to provide a uniform electric field in the bulk oil. Several parameters were studied to determine their influence on the electrostatic dehydration process of water-in-crude-oil emulsions for two different crude oils. Temperature and shear rate were found to be the adjustable process parameters to increase separation efficiency.

5.1 Why a model coalescer?

During the project Electrocoalescence II, a commercial electro-rheometer was used to evaluate electro-coalescence efficiency on emulsions. Though this device was potentially a powerful tool for investigating the behaviour of water-in-crude oil emulsions under electric fields, it had some characteristics which limited its suitability in such kind of studies. There are concerns that using viscosimeters may not be representative for the bulk process taking part in a compact coalescer. In the coalescer the electric field is fairly homogeneous and electrodes are insulated, while in the viscosimeter the electric field will be in-homogeneously concentrated at sharp edges in the bob, and maybe more severe the electrodes are metallic and the electrode gap very small resulting in drop charging. One is therefore afraid that the coalescence will be a surface process with charged water drops, not representative for the realistic bulk process with charge neutral drops

5.2 Building an in-house model coalescer

During the first part of the project we focused our efforts on the designing and building of a new bench size model coalescer with an internal couette flow in the emulsion, and with insulated electrodes and a large active volume to provide a uniform electric field in the bulk oil. This all allows for independent adjustment of all parameters of interests. Therefore during the project, several parameters have been studied to determine their influence on the electrostatic dehydration process of water-in-crude-oil emulsions.

Figure 5-1 shows a schematic illustration of our in-house model coalescer. The chamber is filled with a heating electrically insulating liquid which circulates through a Julabo MC heating bath to provide a controlled temperature during the experiments. The emulsion is kept in a glass cylinder with a acrylic rotor, providing shear to the emulsion. The electric field is applied by two uncovered plane electrodes. High voltage is fed to the electrodes from an amplifier, supplied by a function generator. Voltage amplitude and waveform is recorded by an oscilloscope. The emulsion separation is recorded by a camera, taking pictures on regular intervals.

This model coalescer presents many advantages over the electro-rheometer used during ECII. The electrodes are completely insulated; meaning that the droplets are not subjected to any metallic surfaces hence no possibility of charged droplets. Moreover, the dehydration is observed visually via a window in the test cell.

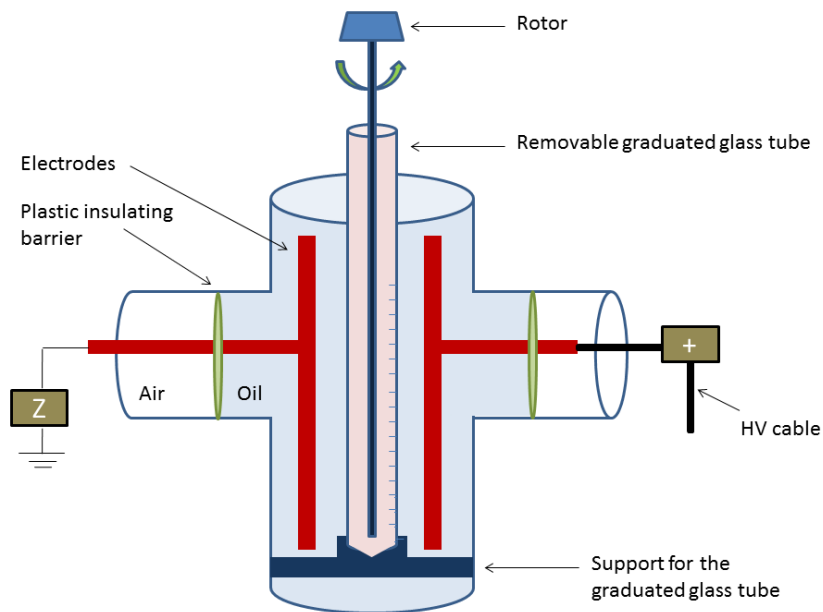


Figure 5-1: Schematic of the model coalescer.

The main outcome of the sub-project can be summarized as follows:

- Design and construction a new bench size model coalescer.
- 1 journal article in a reputable international journal.

5.3 Dehydration efficiency of AC fields on water in crude oil emulsions

Destabilization of water in crude oil (Heidrun) emulsions was investigated from two different batches provided by Statoil. Several parameters were studied: temperature, shear rate, water cut of the emulsion and also frequency and waveform of the electric field. The results are thoroughly discussed in the journal article [5.1]

The results obtained for the dehydration efficiency of the two different batches of Heidrun are collected in the Table 5-1.

Table 5-1: Dehydration efficiency of two batches of emulsions with Heidrun oil and water depending on process parameters.

	Heidrun 1		Heidrun 2	
	Parameters	Separation efficiency	Parameters	Separation efficiency
Frequency	500 Hz	76 %	500 Hz	60 %
	1000 Hz	76 %	700 Hz	55 %
	5000 Hz	76 %	1000 Hz	60 %
Shear rate	0 rpm	66 %	0 rpm	10 %
	60 rpm	78 %	60 rpm	55 %
	120 rpm	76 %	120 rpm	62 %
	240 rpm	30 %	240 rpm	65 %
Water content	5 %	56 %	5 %	45 %
	10 %	68 %	10 %	62 %
	20 %	76 %	20 %	69 %
Temperature	22 °C	76 %	22 °C	58 %
	40 °C	84 %	40 °C	65 %
	60 °C	90 %	60 °C	75 %
Wave form	Sinusoidal	66 %	-	-
	Square	76%		
Droplet size	-	-	10-40 µm	75 %
			10-30 µm	66 %
			10-20 µm	64 %
			5-10 µm	58 %
			3-7 µm	42 %

- Influence of frequency: For both crude oils tested, there is no significant relation between frequency and dehydration efficiency.
- Influence of shear rate: the rotor speed has a large influence on separation rate and efficiency. For Heidrun 1, at high rotor speed (240 rpm), the dehydration efficiency collapses. It becomes even less efficient than when there is no shear applied. After 60 minutes only 30% of the water present in the emulsion can be retrieved (65% when there is no shear). For an emulsion with a typical viscosity value of 25 cP at 22 °C, we find that the flow becomes unstable when the rotor speed exceeds 28 s-1 or about 180 rpm. For Heidrun 2, with a viscosity of 40 cP at the same temperature, the flow becomes unstable when the rotor speed exceeds 260 rpm, explaining why we don't observe the same phenomenon at this shear rate.

- Influence of water content and droplet sizes: the best separation yield is obtained for higher water cuts. This was expected since a higher water cut means a shorter distance between droplets increasing the probability of meeting, hence coalescence. The same reasoning can be made about droplet size. The probability for droplets to meet is higher when those droplets are bigger.
- Influence of temperature: dehydration efficiency increases with increasing temperature. Sedimentation speed is proportional to $1/\eta$; an increase in temperature will cause a drop of the viscosity of the emulsion hence a higher sedimentation speed.
- Influence of wave form: Theory predicts the square waveform to be 1.41 times more effective than the sine waveform. This trend was almost consistent with the results obtained here, that showed a ratio between separations obtained with square waves to separations obtained with sine waves to be between 1.1 and 1.2. However for the same RMS value, the sinusoidal wave ($U_{RMS} = U_{max}/\sqrt{2}$) form is more efficient than the square wave form ($U_{RMS} = U_{max}$). Consequently, for the same energy input, the sinusoidal wave form is more efficient than the square one.

5.4 Deliverables

Publications

- 5.1. C.Lesaint, G.Berg, L.Lundgaard and M.-H.G.Ese: "[A Novel Bench Size Model Coalescer: Dehydration Efficiency of AC Fields on Water-in-crude-oil Emulsions](#)" Paper accepted for publication in IEEE Trans. Diel. And El.Ins, 2016.
- 5.2. C. Lesaint, M-H. G. Ese, M. B. Augdal: "[Dehydration efficiency of AC electrical fields on water-in-crude oil emulsions](#)" Summer student report, 2013.
- 5.3. H. Vassmyr: "[Dehydration Efficiency of AC Fields on Water-in-Crude-Oil Emulsions](#)" Summer student report, 2014.



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