Enhancing dropwise condensation through active transport of drops using electrowetting







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ABSTRACT

Dropwise condensation is used for heat transfer applications, including industrial coolers and water harvesting. Most research aimed on increasing heat transfer is focussed on passive approaches like surface engineering. More recently the active approach of electrowetting has been used to enhance heat transfer. Motivated by these studies, we use electrowetting on functionalized substrates to actively transport condensate drops off the surface.

In this research we explain the workings of active transport and investigate its effect on the heat transfer. By experimentally measuring the condensation rate as well as looking at indirect signs of improved heat transfer on the breath figure, we show that active transport does improve heat transfer.

The electrode design used in this study for active transport is not optimized. We introduce a force balance model that we use to predict the critical radius, and thus efficiency of thousands of different electrode designs for active transport. This model can be used to further enhance active transport.

In this research we present a new experimental setup designed and built to experimentally determine the condensation rate. This modular setup we use to determine the condensation rates for different types of functionalized substrates. We show that, contrary to what was published before, zigzag interdigitated electrodes do not enhance but actually decrease the condensation rate due to pinning at the electrical traps. Interdigitated electrodes show a slight increase in condensation rate, but not as much as with active transport.

We investigate several methods to determine the condensation rate, both experimentally based water collection methods and image analysis based techniques. We show that even though most techniques show comparable condensation rates, only one method proved to be accurate enough to distinguish differences between different types of substrates.

Even though dropwise condensation shows superior heat transfer over filmwise condensation, it is barely used in (industrial) applications due to surface degradation. We study surface degradation during dropwise condensation with electrowetting and use Liquid Infused surfaces in an attempt to create durable surfaces as a first step towards applications of dropwise condensation with electrowetting.

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Before you lies the master thesis of Harmen Hoek, showing the work that has been done over the course of a twelve month period.

This thesis is the result of 667 hours of condensation, spread out over 76 individual experiments, on 28 different substrates, during which 329,956 images of the condensation pattern was recorded, storing 6.04TB of data.

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ACRONYMS

- DWC dropwise condensation
- FWC filmwise condensation
- EW electrowetting
- IDEs interdigitated electrodes
- ZZEs zigzag patterned interdigitated electrodes
- SLIPS Slippery Liquid Infused Porous Surfaces
- LIs Liquid Infused surface(s)
- PCF Physics of Complex Fluids group
- CAH contact angle hysteresis
- NCG non-condensable gases
- LV Labview
- TL Thorlabs
- PDMS polydimethylsiloxane
- ITO indium tin oxide
- RH relative humidity
- FOV field of view

1 INTRODUCTION

Condensation is the change of physical state of matter from the gas phase into the liquid phase. The word condensation comes from the Latin word *condensare*, meaning 'press close together'. With condensation of water, water molecules cluster together to form a liquid. Condensation can be split up in homogeneous and heteregeneous condensation. Homogeneous condensation occurs spontaneously and randomly inside a uniform substance in the abscence of nucleation sites, whereas heterogeneous condensation, which is much more common, occurs on impurities such as dust particles or microscopic roughness. Heterogeneous condensation can be further differentiated in bulk condensation, where the vapor condenses in a gas phase like fog, and surface condensation is generally differentiated in two types: filmwise condensation (FWC) and dropwise condensation (DWC). Both types of condendensation are shown in figure 1.1.

FWC occurs when the condensed liquid wets the surface. Condensation takes place onto the liquid film as it grows thicker and slides down under the influence of gravity. DWC may occur when vapor condenses on a surface not fully wetted by the condensate. The vapor condenses and forms droplets that grow in size by direct condensation of more vapor, or by coalescence of neighboring droplets until gravity pulls down the droplet. While FWC occurs on hydrophilic surfaces, DWC occurs on hydrophobic surfaces.

DWC was first reported by Schmidt *et al* in 1930 [1] where they showed that the heat transfer coefficient for DWC is much higher than for filmwise condensation. Later measurements showed heat transfer coefficients for DWC 10-30 times higher than FWC. [2–4] Reason for this increase heat transfer with DWC is the absence of the liquid film that acts as a thermal resistance decreasing the heat transfer from the vapor to the surface. For this reason DWC is the preferred mode of condensation in heat transfer applications. The process of DWC and the improved heat transfer is explained in more detail in chapter 2.

But long before the first reports of DWC in science, it has been omnipresent in nature as dew formation [5] where humid air condenses and transforms into liquid water. DWC is used to harvest water out of the air in arrid climats by animals like the Desert Beetle where an array of hydrophilic bums surrounded by a hydrophobic background on the Beetle's back nucleates water vapor and forms drops. [6, 7] The Litoria caerulea, a green tree frog in tropical northern Australia uses condensation in the dry season to get water. The frog is cooled down overnight, and during the warm dry day they get into warm humid tree hollows where water condensation occurs on their body. [8] Also plants use DWC to harvest water, such as the Namib desert plant. [9]

2 | INTRODUCTION



Figure 1.1: Filmwise condensation (a) versus dropwise condensation (b).

Applications of DWC

In (industrial) applications condensation is commonly used to extract heat from vapor. Most electricity produced globally is generated by a steam cycle. [10] The efficiency of the steam cycle depends on the amount of energy that is transferred from the energy source to water that vaporizes to power a turbine. Here condensation plays an important role to cool the vapor down below the saturation temperature so it can be reused in the cycle.

Also in smaller systems where heat exchangers play an important role, like refrigeration and air conditioning systems, condensation plays an important role. [11] Smaller more efficient condensation systems could significantly increase energy efficiency and reduce size. [12]

Another application of DWC is water-harvesting systems. [13, 14] Water shortage is a major problem of our modern society that is expected to get worse due to climate change and increased human population. The United Nations estimated that more than 5 billion people could suffer water shortages by 2050. [15] Collection of water out of foggy wind or thermal desalination systems to turn seawater into fresh water are suggested as remedies. [16]

These are just a few examples where condensation plays an important role. Condensation also has many useful applications in agricultural [17], industrial and domestic applications, such as sanitation and sterilization [18], food processing, chemical processing [19] or anti-icing [20].

Passive approaches to improve condensation rate

The performance of these applications of DWC all depend on the amount of condensation; the more water condenses, the higher the heat transfer. The main bottleneck to condensation performance is the removal of condensate from the condensing surface. Condensate that stays on the surface acts as a thermal barrier for new condensation, as will be explained in section 2.1.2.

Over the years a lot of effort has been put into increasing condensate removal from

the surface.

Hybrid surfaces of high wetting contrast have been used that allow for seamless integration of FWC and DWC to enhance condensate drainage of the surface. [21, 22] On these surfaces drops grow on the hydrophobic patches until they touch the hydrophilic patches resulting in rapid transfer of the condensate to these patches allowing for new condensation to occur on the hydrophobic patches. This method of patterned wettability has been reported to improve condensate collection 19% [23] to 63% [22] compared to non-patterned hydrophobic surfaces.

Boreyko *et al* [24] and Miljkovic *et al* [25, 26] used superhydrophobic nanostructured surfaces to enhance drop mobility. When drops coalesce on these surface, the resulting drop can jump from the surface due to the release of excess surface energy. Miljkovic *et al* reported a 30% increase in the heat transfer coefficient using these surfaces. However, these improvements were only achieved at low supersaturations, at high supersaturations the flooding of the nanostructured surface led to a 40% decrease in heat transfer.

Park *et al* [6] combined macroscopic bumb topography used by the Namib desert beetle to maximize vapor diffusion flux with apex geometry of cactus spines and slippery pitcher-plant-inspired nanocoating to create a 3D slippery asymmetric bump surface. Their surface allows enhanced drop condensation rates on the top of the bumps and fast removal away from these bumps. They observed sixfold-higher exponent of growth rate, faster onset, higher steady-state turnover rate, and a greater volume of water collected.

Other improvements in condensation rates have been made by reduction in contact angle hysteresis with the use of liquid impregnated textured surfaces [27, 28], with superhydrophobic grooved surfaces [29] and Microdrop Self-Propelling Al Surfaces [30], to just name a few.

Active approaches to improve condensation rate

Historically, the majority of research done on improved DWC have relied on passive approaches like surface engineering mentioned above. Active techniques require external forces, e.g. electric fields or vibrations.

Recently Baratian *et al* [31] and Dey *et al* [32] made efforts to improve the heat transfer in DWC using an active technique called electrowetting (EW). EW is the increase of wettability using an electric field. Using functionalized electrode designs they reduced contact angle hysteresis of condensate drops allowing for shedding at smaller radii. At the same time they reported enhanced drop coalesce due to drop alignment to the electrostatic minima resulting in faster drop growth and lower surfaces coverages. Using heat transfer measurements without non-condensable gases they reported an increase in heat transfer of 50–60% [31] and \sim 400% [32] using a different electrode design and with volume estimations using image analysis. Similar functionalized surfaces with EW were used for this study. More about EW and

its enhancing effect on condensation in section 2.3.

Another DWC enhancing technique involving electric fields is from Miljkovic *et al* [33]. Succeeding their research mentioned above, they used nanotextured jumpingdrop surfaces in combination with an external electric field that limits drop return to the condensing surface, further enhancing the condensation rate. Using this electricfield-enhanced condensation technique they reported a ~50% increase over their conventional jumping-drop condensation.

Current challenges and limitations

Even with the amount of research done on enhancing DWC, DWC has has been hardly realized in industrial applications. [34]

Promoting DWC on a surface requires a hydrophobic surface, which usually are fragile. Despite years of effort, coating degradation is still a major problem leading to loss of surface hydrophobicity and consequently surface flooding. Industrial tests have been performed by implementing dropwise condensers in evaporators of power stations and sea water evaporators, but no significant increase in condensation rate has been observed in these long term experiments due to surface degradation. [35] Increasing stability of the coatings often leads to increased thickness resulting in added thermal resistance. Research by Holden *et al* [36] even showed that a coating thickness of $60\mu m$ was enough to fully counteract the benefits of DWC compared to FWC, even though the surface showed stable DWC > 22,000 hours. The perfect combination of a hydrophobic, thin and durable surface has yet stayed out.

A recent study by Preston *et al* [37] focussed on this problem by trying a coating of graphene. Graphene displays hydrophobic behavior thus promoting DWC and resists degradation. Heat transfer measurements showed an improvement by a factor of 4 compared to FWC, while showing superior robustness. Experiments over 2 weeks still showed DWC, while fluorocarbon coatings under similar conditions completely degraded under 12 hours.

Other efforts for more stable surfaces with DWC have led to Liquid Infused Surfaces (LIs). With LIs an oil layer lubricates a porous surface forming a superhydrophobic surface on which the condensate drop 'floats'. This results in extremely low contact angle hysteresis and special properties such as self-healing. LIs are used in this research and are discussed in detail in section 2.5.

1.1 RESEARCH GOALS

In this field of research we can identify 2 challenges. The first one being to create a surface with a heat transfer coefficient as high as possible, and secondly increasing the stability of this layer to allow for practical applications.

This thesis aims to continue the work done by Baratian *et al* [31] and Dey *et al* [32] on improving heat transfer rates with DWC using EW. More specifically we aim to use functionalized substrates to actively transport drops down the surface to increase the volumetric condensation rate. We aim to combine EW with surfaces that can withstand long term use without degrading.

To achieve these goals we set the following subgoals:

- Improvement of the experimental setup. The current setup used for condensation experiments is cumbersome to use; coolant leaks are common and substrates are difficult and time consuming to replace. In addition, condensate collection, a direct measure for average heat transfer rates, is not possible. The goal is to improve the ease of use of the setup and allow for water collection;
- Quantitative measurement with different substrates. Repeat experiments with similar functionalized substrates used by Baratian *et al* [31] and Dey *et al* [32] to determine the condensation rate and verify their conclusions;
- Design and test functionalized electrode designs for active transport of condensate, i.e. actively move drops off the substrate using EW.
- Develop and test the stability of substrate coatings that can be used in combination with EW and show long term stability.

1.2 THESIS OUTLINE

A theoretical background of (dropwise) condensation and EW is given in the Theoretical Aspects in **chapter 2**. **Chapter 3**, the Experimental Aspects gives a full overview of the redesigned experimental setup, experimental procedures and data processing. The Results are presented and discussed in **chapter 4**. Here several topics will be discussed, including a comparison of techniques to determine the condensation rate, active transport, comparison of several functionalized substrates, a model to improve active transport electrode designs and finally the results of a detailed study into surface degradation. **Chapter 5** contains the Conclusions of all the results. And the last chapter, **chapter 6** elaborates on future research in this field of topic with the Outlook.

2 | THEORETICAL ASPECTS

2.1 CONDENSATION THEORY

Condensation involves the change of phase from vapor to liquid and is initiated by a temperature difference between the vapor and cooled surface, called subcooling. Condensation is inextricably linked to mass and heat transfer. Vapor from the bulk moves towards the cooled liquid-vapor interface and is converted into liquid. During the process of condensation heat is transferred. For vapor to condense the latent heat must be extracted from the interface by either conduction or convection. This heat transfer is the reason behind many applications of condensation.

Dropwise condensation (DWC) is preferred over filmwise condensation (FWC) by its enhanced heat transfer coefficient, as pointed out in the introduction. With DWC, drops that condensed on the surface from the vapor grow by three processes: 1) direct condensation of vapor molecules onto drop surface, 2) surface diffusion of small embryos and 3) by coalescence with neighboring drop. The first two growth processes are common in the early stages of DWC, the last one kicks in when drops are big enough they touch. This multifaceted growth makes DWC a complicated cyclic process that can be studied by looking at the breath figure: the moisture pattern, as will be discussed in section 2.1.1.

The total heat transfer with DWC is complicated too. Large drops have a larger surface area / mass ratio compared to small drops. This makes large drops grow slower compared to their smaller companions. Larger drops do also have a larger thermal resistance isolating the surface from new condensation. Therefore smaller drops are preferred in DWC as will be further elaborated in section 2.1.2.

2.1.1 Phases of dropwise condensation

DWC is a cyclic process of 3 distinct phases that can be identified from the breath figure that is the moisture pattern on the surface. These phases are 1) nucleation and growth of drops without coalescence, 2) coalescence driven growth; 3) shedding of the drop. A snapshot of each phase is shown in figure 2.2.

Each phase has its own characteristics including a characteristic radius that defines the transition between phases, these are shown in figure 2.1.

We consider a vertically mounted surface cooled on one side at temperature T_c , and with a constant vapor flow on the other side at temperature T_v in the bulk.



Figure 2.1: Condensation phases without electrowetting characterized by the drop radius.



Figure 2.2: Breath figures of the condensation process without electrowetting. At t=60s after introducing vapor in the chamber phase I: nucleation is visible. At t=303s the drop growth is governed by phase II: coalescence. At t=492s phase III: shedding is visible simultaneously with renucleation as the condensation cycle repeats.

2.1.1.1 Phase 1: nucleation

In DWC, when a clean surface is exposed to (water) vapor, the first phase is nucleation, shown in figure 2.2a. Nucleation is the phase transition of a water nucleus from vapor to liquid. It occurs when the vapor temperature is brought below the saturation temperature (or above the saturation pressure).

Why does nucleation happen? The first vapor nucleation occurs by thermal fluctuations of the smallest nucleus that is thermodynamically stable, that is the nucleus that requires the least amount of energy to change phases and does not evaporate after condensing.

The energy involved to nucleate a drop in homogeneous nucleation, that is without a surface, is the free energy of a drop plus the energy required to form the contact area of the liquid-vapor: [38]

$$W_{hom} = -\Delta e \ V + S\gamma = -\frac{4\pi R^3}{3}\Delta e + 4\pi R^2\gamma$$
(2.1)

where R is the drop radius, Δe the change in free energy, V the drop volume, S the drop surface area and γ the surface tension. The energy is thus lowered by phase transition from vapor to liquid, but prevented by the formation of the liquid-vapor interface. An energy barrier has to be overcome.

The smallest possible nucleus size that can overcome this energy barrier, thus condenses on the surface and does not evaporate again, follows directly from equation 2.1 by finding radius corresponding to energy barrier dW/dR = 0:

$$\frac{dW_{hom}}{dR} = -4\pi R^2 \Delta e + 8\pi R\gamma = 0$$

$$\Rightarrow R_{n,hom} = \frac{2\gamma}{\Delta e}$$
(2.2)

To account for heterogeneous nucleation, a geometric function $f(\theta)$ is introduced, to consider the energy reduction due to the presence of the substrate:

$$W_{het} = W_{hom} f(\theta) \tag{2.3}$$

For the spherical cap model of bubble nucleation, which is the most common nucleation model, the geometric factor is $f(\theta) = 1/4(2 + \cos \theta)(1 - \cos \theta)2$, [38, 39] or simplified as $f(\theta) \approx \sin \theta$ [40].

Using this expression and rewriting Δe using the Clapeyron equation, we get a final expression for $R_{n,het}$: [4]

$$R_{n,het} = \frac{2\gamma T_{sat}}{h_{fg}\rho_l \Delta T} f(\theta)$$
(2.4)

Where T_{sat} is the saturation temperature of the vapor, $\Delta T = T_{sat} - T_w$ the subcooling, h_{fg} the latent heat of condensation and ρ_l the drop density. $f(\theta)$ shows a direct contact angle dependency, which is, larger contact angle drops require more energy to be formed.

The liquid drop is thus only stable for a radius bigger than R_n , and thus nucleation will occur above this radius. For condensation of saturated water vapor around $20^{\circ}C$, $R_n \sim 40$ Å.

By the heterogeneity of the surface, nucleation occurs heterogeneously on the surface resulting in a nonuniform distribution of the drops.

After nucleation, drops keep growing partially by direct absorption of vapor molecules, but mainly via surface diffusion of small embryos around the drop. Vapor molecules thus do not form new drops once all nucleations sites are occupied, but diffuse in bigger drops. This results in a constant number of drops on the surface, and constant growth of these drops. This also means that in this early condensation phase, drops are widely separated: $R_c >> R$, that is the distance between the drops (the coalescence radius) is much larger than the radii of the drops. [41]

2.1.1.2 Phase 2: coalescence

Drops keep growing and the characteristic length scales converge: $R \approx R_c$, that is, the drop radii are similar to the distance between the drops. The coalescence phase kicks in, see the breath figure in figure 2.2b. When condensate drops touch, they coalesce into a single new drop with a volume equal to the sum of the volume of the initial drops. This new drop is (approximately) located at the center of mass of the initial drops. Coalescence is favorable in terms of energy, as it lowers the total surface energy. The coalescence radius at which coalescence starts depends on the nucleation site density of the surface, with a typical value of $R_c \sim 2.6 \mu m$ [40].

Coalescence events happen in a fraction of a second, and often start a chain reaction of coalescence events. An example of such a chain reaction of coalescence events is shown in figure 2.3. The prominent drops before the coalescence event are contoured with green, after the event the final drop is contoured with red.

During the coalescence phase the drop size distribution is quite narrow. Coalescence causes drops to be further apart, making it more likely for smaller drops to coalescence. While direct absorption and surface diffusion of vapor into drops still results in drop growth, the dominant growth mechanism is coalescence.

Coalescence has the noticeable effect of creating clean space on the surface, as the total surface area the drops occupy decrease with a coalescence event. On these clean areas a second generation of drops will nucleate and grow, resulting in a bimodal distribution. [42]



Figure 2.3: An example of a chain reaction of coalescence events. At t=o a large number of drops is present close together, the biggest drops encircled with green. At t=3s a single coalescence event caused a chain reaction coalescing all the drops into one, encircled with red.

2.1.1.3 Phase 3: shedding

Drops keep growing by direct absorption, surface diffusion and primarily by coalescence until the weight exceeds the pinning forces acting on the drop and the drop slides down the surface, a phenomena called drop shedding, visible in figure 2.2c. The radius at which drop shedding occurs is called the critical shedding radius R_{crit} .

The critical shedding radius is found by solving the force balance of the drop. The drop weight can be expressed as

$$F_g = \rho g \frac{2}{3} \pi R^3 \tag{2.5}$$

where $2/3\pi R^3$ is the drop volume approximated as a hemisphere, i.e. a drop with contact angle $\theta = 90^\circ$, ρ is the drop liquid density and g the gravitational acceleration.

The pinning force acting on the drop contact line, ignoring any roughness/ geometry factors [43–45], is [46]

$$F_p = 2R\gamma\Delta\cos\theta \tag{2.6}$$

where *R* is the radius of the drop-substrate contact area, γ is the surface tension of the drop, and $\Delta \cos \theta = \cos \theta_R - \cos \theta_A$ the contact angle hysteresis (CAH) defined as the difference between the receding θ_R and advancing θ_A contact angles.

The force balance of a drop at R_{crit} is than [47]

$$F_{p}(R_{crit}) = F_{g}(R_{crit})$$

$$\rho \frac{2}{3} \pi R_{crit}^{3} = 2R_{crit} \gamma \Delta \cos \theta$$

$$\Rightarrow R_{crit} = \sqrt{\frac{3}{\pi}} \lambda_{c} [\Delta \cos \theta]^{1/2}$$
(2.7)

Where $\lambda_c = \frac{\gamma}{\rho g}$ the capillary length.

Shedding drops grow in size sweeping the surface from other drops below making the drop grown in size and leaving behind a clean surface for new drop nucleation and growth.

The different condensation phases are well defined and easily recognized when looking at the breath figures. But only at the start of the condensation process, when the surface is still clean, only one phase is present at once. Due to the heterogeneity of the surface drops are not evenly spaced and the coalescence phase kicks in at different times on the surface and re-nucleation occurs only at specific locations. Consequently, new generations of drops are created at different times everywhere on the surface. This behavior makes condensation a local cyclic process of the phases mentioned above. At every moment in time one of these phases can be distinguished locally on the surface.

2.1.2 Heat transfer

Applications of DWC depend on the efficiency of the system, i.e. the amount of heat transfer between bulk vapor and surface. A higher heat transfer means more condensation per unit area.

The total heat transfer Q between bulk vapor and bulk coolant is a sum of the convective heat transfer through the vapor and coolant and heat conduction through the (glass) substrate separating vapor and coolant and the condensate, considering FWC, i.e. the whole substrate is covered by a condensate layer. See figure 2.4. Assuming laminar flow of the coolant and vapor, the total heat transfer is given by: [48]

$$Q = \frac{T_v - T_c}{R} \tag{2.8}$$



Figure 2.4: A schematic showing the contributions to the total heat transfer. The coolant at temperature T_c increases by convective heat transfer towards the glass substrate. Inside the substrate and condensate water film conductive heat transfer linearly decreases the temperature. On the vapor side convective heat transfer results in a final temperature increase to T_v .

where T_v is the vapor bulk temperature, T_c the coolant bulk temperature and R the thermal resistance between the vapor bulk temperature and coolant temperature. The total thermal resistance is a sum of the individual resistances of the system, including the capillary resistance of the drop curvature, interfacial resistance of the liquid-vapor interface. All these are discussed in detail by Khandekar and Muralidhar [4]. As an approximation we include only the major contributions of the thermal resistance, as:

$$R = \frac{1}{A} \left(\frac{1}{h_v} + \frac{L_g}{k_g} + \frac{1}{h_c} + \frac{L_w}{k_w} \right)$$
(2.9)

where h_v and h_c are the heat transfer coefficients of the vapor and coolant, respectively, k_g and L_g the thermal conductivity and thickness of the substrate, respectively, and k_w and L_w the thermal conductivity of the condensate on the substrate, respectively.

For DWC equation 2.9 does not hold as the total surface area is not covered by a liquid film and the thickness of the water depends on the drop size. In general one can consider a reduction in thermal resistance, due to an increase in bare surface area, i.e. loss of thermal resistance term L_w/k_w .

From equation 2.8 follows directly that the total heat transfer is linear proportional to the subcooling $Q \propto \Delta T = T_v - T_c$.

Determining the heat transfer rate can be done by experimental heat flux measurements or measuring the average volumetric condensation rate $\langle v \rangle$. Direct heat transfer measurements measure the temperature difference between two points of a material with known thermal resistance and calculate the heat transfer using equation 2.8.

Volumetric condensation rates measure the mass Δm of the shedded condensate

over a period Δt , from which follows the average heat transfer rate $\langle Q \rangle$ using the latent heat of vaporization h_{fg} : [49]

$$\langle v \rangle = \frac{\Delta m}{\Delta t} = \frac{\langle Q \rangle}{h_{fg}}$$

$$\Rightarrow \langle Q \rangle = \langle v \rangle \cdot h_{fg}$$
(2.10)

This latter method of average volumetric condensation rates is used in this research.

Besides experimental heat transfer measurements, breath figures can also reveal signs of increased heat transfer. These signs including surface coverage, shedding radius, shedding frequency and drop coalescence will be discussed below.

2.1.2.1 Decreased surface coverage

=

Equation 2.9 shows an important dependency of the thickness of the condensate layer L_w on the thermal resistance. Condensate on the surface increases the thermal resistance between vapor and coolant, decreasing the heat transfer. This isolating or blanketing effect [40] is the reason heat transfer of DWC exceeds that of FWC in similar conditions. The permanent water film in FWC acts as a permanent thermal resistance, while for DWC this thermal resistance is only present locally.

To increase the heat transfer in DWC it is therefore the objective to reduce the amount of condensate on the surface, that is, reducing the surface coverage of the breath figure, where the surface coverage is the ratio: surface area covered by drops/ substrate area. Already in the earliest works on DWC formation of 'cleaned' or low-surface-coverage regions was considered to have important implications for heat transfer. [50] Lower surface coverage means more bare surface is available for nucleation of new drops and fewer drops that isolate the surface.

2.1.2.2 Decreased shedding radius

Another indication of increased heat transfer is a decrease in R_{crit} . The heat flux through small drops is larger than through bigger drops. [35, 51, 52], This is beacause they are thinner and thus having a smaller thermal resistance (equation 2.9). In fact, the majority of the heat transfer occurs through the small drops. [52]

In addition to that, a smaller R_{crit} also means drops shed down earlier than larger drops, assuming the same drop growth rate. These drops will thus clear the surface sooner to make place for nucleation of new drops, with higher heat transfers.

It has even been shown that under certain circumstances drop growth stops completely for large drops due to evaporation at high vapor flow rates. Such large drops will only shed after a coalescence event with a neighbouring drop.

In literature this indication of increased heat transfer has been discussed well. Khandekar *et al* found an increased heat transfer of over 200% by decreasing the critical shedding radius from 4mm to 1mm. [53] Following simulations they report a correlation between the heat transfer and shedding radius as:

$$\frac{Q}{A} = 0.19 R_{crit}^{-1.2} \tag{2.11}$$

Here, Q/A is the heat transfer per unit area $[MW/m^2]$ and R_{crit} in [mm]. Similar increases in heat transfer were found by Tower and Westwater [54], Rose *et al* [35] and with numerical simulations. [40, 52]

A smaller critical shedding radius thus results in an increased heat transfer. One way of decreasing the critical shedding radius is by decreasing the contact angle hysteresis using electrowetting, which will be discussed in 2.2.

2.1.2.3 Increased shedding frequency

A decrase in R_{crit} also means an increase in shedding frequency, that is the number of drops that shed per unit time, assuming the drop growth remains the same. Shedding drops grow in size sweeping the surface from other drops and leaving behind a clean surface. This makes shedding extremely efficient in increasing the total condensation rate and in the earliest works on DWC, sweeping initiated by shedding was found to have a significant effect on the heat transfer [55]. Sweeping removes drops from the surface below the critical shedding radius, reducing the surface coverage and increasing overall heat transfer.

2.1.2.4 Increased drop coalescence

Another sign of increased heat transfer is the increase in drop coalescence. Drop coalescence strongly accelerates drop growth. [5] The average drop radius obeys a power law of the form $\langle R \rangle \sim t^{\mu}$, where μ is a growth exponent. For single drop growth, e.g. in the earliest phase of condensation, there is no coalescence and $\mu = 1/3$. With coalescence this factor scales to $\mu = 1$. [5]

In addition to accelerated drop growth does coalescence have the consequence of leaving free space on the surface for new nucleation and growth. Since two drops occupy more surface area compared to a single drop with the volume of the two drops combined.

This striking behavior makes coalescence extremely efficient in increasing the heat transfer. Increasing drop coalescence can also be achieved by electrowetting, where electrostatic drop alignment and drop oscillations results in enhanced coalescence (section 2.3).

2.2 ELECTROWETTING

Electrowetting (EW) is the increase of wettability between a liquid and insulated electrode substrate that is caused by an electric field between the two. This increased wettability is observed experimentally as the increase in drop contact angle. The electric field is applied by a potential difference between two electrodes. Historically this is done with an electrode below and insulated surface and other electrode in the conductive drop. Newer methods include an interdigitated electrode surface below a dielectric layer in the substrate, see figures 3.5 and 3.6. By applying a potential difference between the two electrodes an electric field is generated between the two electrodes. A dielectric layer between drop and electrodes prevents electrolysis.

Electrowetting can be subdivided in DC electrowetting and AC electrowetting. DC electrowetting involves applying a DC voltage between the electrode and liquid. When applying a voltage between electrode and liquid, the electrode will accumulate a surface charge, which consequently causes the accumulation of counter-ions near the interface. These ions lower the interfacial energy and induces an increase in contact angle. [56]

With DC electrowetting both the advancing and receding contact angles increase by the same amount as a result of the electric field, hence resulting in no change in the hysteresis of the drop. [56]

With AC electrowetting an AC voltage is applied between electrode and liquid. Due to the alternation in voltage, the advancing contact angle experiences an increase at maximum voltage amplitude, while the receding contact angle remains unchanged at minimum voltage amplitude [57]:

$$\cos\theta_a^{EW} = \cos\theta_a^0 + 2\eta_{rms} \tag{2.12a}$$

$$\cos\theta_r^{EW} = \cos\theta_r^0 + 0 \tag{2.12b}$$

where θ_a^{ew} , θ_r^{ew} , θ_a^0 and θ_r^0 are the advancing and receding contact angles due to EW and advancing and receding contact angles without EW, respectively, and η is the electrowetting number defined as [56]:

$$\eta_{rms} = \frac{\epsilon_0 \epsilon U_{rms}^2}{2h\sigma} \tag{2.13}$$

with ϵ_0 and ϵ the vacuum and relative permittivity of the insulator, respectively, U_{rms} the root-mean-square of the externally applied potential difference, h the insulator thickness and σ the surface tension of the drop.

This unequal addition of the contact angle, in contrast to DC electrowetting, results in an increase in hysteresis:

$$\Delta \cos \theta^{EW} = \cos \left(\theta_r^{EW} - \theta_a^{EW} \right)$$

= $\Delta \cos \theta^0 - 2\eta^{rms}$ (2.14)

As a result, drops have an increased mobility on the surface due to electrowetting. The pinning force acting on the drop is thus altered by electrowetting. Rewriting equation 2.6 with the addition of electrowetting gives:[32]

$$F_p = 2R\gamma\Delta\cos\theta \\\approx 2R\gamma\left(\Delta\cos\theta_0 - \alpha\beta U_{rms}^2\right)$$
(2.15)

where the contact angle hysteresis $\Delta \cos \theta$ is approximated as $\Delta \cos \theta_0 \alpha \beta U_{rms}^2$. $\Delta \cos \theta_0$ is the value of $\Delta \cos \theta$ for $U_{rms} = 0$ V, β the ratio of the effective dielectric capacitance per unit area (estimated to be roughly $\beta \sim 8.5 \times 10^{-6} F/N/m$ [32]) and α a coefficient characterizing the efficiency of the contact line depinning ($\alpha \sim 1$). [58]

The pinning force thus decreases with electrowetting voltage. It must be noted that this decrease does not go on with decreasing voltage, but stabilizes at moderate voltages. [58, 59]

2.3 ELECTROWETTING AND CONDENSATION

Electrowetting can be used to actively control the mobility of drops in a breath figure evolution during DWC. This was first done by Baratian *et al* in 2018 where they showed the effects of electrowetting on the breath figures using /aclDEs (see section 3.2.1) on the breath figures. [31]. EW induces two main effects when used with DWC. First the decrease in contact angle hysteresis results in a decrease in pinning force that consequently results in a decreased critical shedding radius. Secondly, due to drop alignment to the electrostatic minima of the electric field and oscillations induced by the alternating voltage, coalescence is enhanced. The latter depends on the electrode design and attenuation pattern. Depending on the design and attenuation pattern additional effects might appear like electrical forces pulling drops down.

2.3.1 Interdigitated electrodes (IDE)

The simplest design used for condensate manipulation using electrowetting by Baratian *et al* are interdigitated electrodes (IDEs). To show the effects of electrowetting with IDEs on DWC, snap shots of the breath figure during the condensation process are shown in figure 2.6. The updated condensation phase diagram is shown in figure 2.5.

With EW, the initial phases of DWC are similar to condensation without EW. Drops nucleate at radius R_n and start coalescing when drops touch at radius R_c , as described in section 2.1.1.1 (figures 2.6a and 2.6b). The effect of EW starts getting visible when the drops grow in size. The drops experience an electrical force by the electrical field induced by EW. The electrostatic energy landscape is not uniform on the surface, but shows drop size dependent minima on the surface that follow directly from solving the Poisson equation for an alternating potential, done in Supplemental Material S9 of Baratian et al [60]. The electrostatic minima are located at the edges of the electrodes for small drops and move towards the center of the gap for larger drops. For drops of size $\mathcal{O}(w_g)$ and bigger, with $w_g = 2b$ the width of a gap, the minimum is located at the center of the gap. As the drops grow in size they migrate to these size-dependent energy minima and start aligning, see figures 2.6c and 2.6d. While the electrostatic minimum moves towards the center of the gap smoothly, the drops show a sudden alignment at the center of the gap when the drop size exceeds the critical radius of gap alignment $R_g \sim 0.3 w_g$, see figure 2.6d. At $R_g \sim 0.3 w_g$ drops migrate to the center from both sides of the gap touch starting a coalescence cascade. This cascade results in the sudden horizontal alignment of the drops at the center of the gaps. This drop alignment to the electrostatic minimum is one of the reasons drop growth and thus heat transfer is enhanced.

The aligned drops keep growing by direct condensation and coalescence. As a matter of fact, the growth rate for aligned drops is larger, as smaller drops move towards these bigger ones. Drops grow until they reach the critical shedding radius $R_{crit,IDE}$ when the weight overcomes the hysteresis and the drop sheds down, see figure 2.6f. But $R_{crit,IDE} \neq R_{crit}$.

The decreased hysteresis due to EW (equation 2.14) results in a decreased pinning

force. The force balance on the drop from equation 2.7 can be rewritten using the modified pinning force with EW from equation as:

$$F_{g} = F_{p}$$

$$\rho g \frac{2}{3} \pi R_{crit,IDE}^{3} = 2R_{crit,IDE} \gamma (\Delta \cos \theta_{0} - \alpha \beta U_{rms}^{2})$$

$$\Rightarrow R_{crit,IDE} = \left[\frac{3}{\pi} \gamma_{c}^{2} (\Delta \cos \theta_{0} - \alpha \beta U_{rms}^{2})\right]^{1/2}$$
(2.16)

Hence, for $U_{rms} > 0$ we see that $R_{crit,IDE} < R_{crit}$ from equation 2.7. Drops thus shed at smaller radii with EW. In section 2.1.2 was shown that a smaller shedding radius and decreased surface coverage are a direct indication of increased heat transfer. Therefore, condensation with EW does increase the heat transfer.

Preliminary heat transfer measurements result from Baratian *et al* showed an increase of the heat transfer of 50-60%. [31] They also showed a reduction in surface coverage of 10-12%. This reduction in surface coverage is (partially) caused by the enhanced coalescence with EW, a benefit of condensation with EW on IDEs that is not included in $R_{crit,IDE}$.

Coalescence might even be further enhanced by reducing the ac frequency to drive drops at their resonance frequency. This results in individual drop resonances that shake the drops clearing an area around the drop from smaller drops. Furthermore lower frequencies have shown to reduce the CAH more for smaller drops. [61] Unpublished research by Baratian *et al* [62] shows increased heat transfer with lower ac frequencies as a result of contact line depinning due to the increased oscillation range of the drop.



Figure 2.5: Condensation phases with EW on IDEs characterized by the drop radius.



Figure 2.6: Breath figures of DWC under EW with IDEs. At t=o the surface is clean; the green overlay shows the location of the IDEs. At t=84s nucleation happens randomly on the surface. At t=129s coalescence is visible and drops start aligning at the edge of the gaps between electrodes. At t=213s Coalescence continue and larger drops are aligned at center of the gap. At t=357s drops are mainly aligned at the center of the gap, the location of the electrostatic minimum. At t=573s first shedding occurs.

2.3.2 Zigzag patterned electrodes (ZZE)

Where IDEs do enhance coalescence and reduce the critical shedding radius by contact angle hysteresis reduction, the electric forces acting on the drop only align the drops horizontally. A variation of this electrode design that does induce vertical forces on the drop is the zigzag patterned interdigitated electrodes (ZZEs) design, first reported by Dey *et al* in 2018. [32] See section 3.2.1 for a description of the design.

The condensation phases with ZZEs, shown in figure 2.10, in the initial growth period are similar to those with IDEs design (2.5): drops nucleate, grow by coalescence and align horizontally at radius R_g . But the snapshots of breath figures in figure 2.9 show additional behavior, i.e. vertical alignment of the drops that can be explained by calculation the electrical forces acting on a drop.

To calculate the net electric force acting on the drop caused by the zigzag design, we assume a drop on a substrate with ZZE touching two adjacent electrodes, schematically shown in figure 2.8a.

The normal electrical force acting on the drop, per unit contact line of the drop, can be written as: [32, 63]

$$\bar{F}_e = F_e \hat{n} \approx \beta \gamma U_{rms}^2 \hat{n} \tag{2.17}$$

where \hat{n} is the unit vector normal to the contact line and β the ratio of the effective dielectric capacitance per unit area (estimated to be roughly $\beta \sim 8.5 \times 10^{-6} F/N/m$ [64]).

The electrical force acts on the drop in the direction normal to the drop contact line, and since the drop overlaps two electrodes, the drop is pulled on from 2 sides. The horizontal components of \overline{F}_e will result in horizontal alignment of the drops, similar to IDE electrodes. With the drop centered, the total energy of the system is minimized. The vertical components on the other hand will add up and result in a net force pulling down on the drop. This electrostatic behavior on a drop is widely used for drop manipulation in microchannels. Due to the vertical asymmetric shape of the electrode the drop overlap with the electrodes is asymmetric: the lower half of the drop has a bigger overlap with the electrodes than the top half. Figure 2.7 shows electrostatic energy landscape of a drop with on two coplanar electrodes separated by a gap from De Ruiter et al from 2014. [65]. The top graph shows the horizontal alignment behavior of a drop. The minimal electrostatic energy is achieved with the drop being centered on the electrodes: the horizontal alignment of drops. The bottom graph shows the vertical alignment behavior of a drop. When a drop 'touches' both electrodes, an electrical circuit containing two parallel plate capacitors in series formed. The asymmetric overlap results in a net electrical force pulling the drop onto the electrodes to minimize the electrostatic energy.



Figure 2.7: Drop trapping in electrostatic potential wells. The electrostatic energy (black solid line) and the resulting electrostatic force (red dashed line) are shown for two coplanar electrodes seperated by a narrow gap. Figure adapted from De Ruiter *et al.* [65].

The vertical component of the force with ZZEs can be estimated by evaluation of the normal vector as an integral, like:

$$F_{e\downarrow} \approx 2\beta\gamma U_{rms}^2 \int \hat{n} \cdot \hat{v} \, dl$$

$$\approx 2\beta\gamma U_{rms}^2 \int \cos \alpha \, dl$$

$$\approx 2\beta\gamma U_{rms}^2 \Delta l \qquad (2.18)$$

where the integration is over the contact line that overlaps the underlying active electrodes. dl is the drop contact line element, with \hat{n} the unit vector that is perpendicular to this contact line, and \hat{v} the unit vector in the vertical direction. α is the angle between \hat{n} and \hat{v} , and thus is $dl \cos \alpha$ the horizontal projection of the line element dl. The total length of the horizontal projection of dl is than the integration of this as $\int \cos \alpha \, dl$. Due to the symmetrical shape of the drop, the vertical components of the electrical force cancel out where they both overlap the electrode. The contact line that overlaps the electrode that is not canceled out due to the asymmetrical shape of the electrodes results in a net vertical force pointing down. The horizontal projection of this piece of contact line can be approximated as $\int \cos \alpha \, dl \approx \Delta l$. Δl is thus the horizontal distance between the intersections of the electrode and drop, see figure 2.8a. The factor 2 is introduced due to the double overlap of the drop with the electrodes left and right.

From equation 2.18 we see that a drop touching two ZZEs experiences a electrical force pulling it down. The force directly depends on the opening angle of the electrode. Sharper electrodes will result in a larger electrical force and drop transport down at smaller radii. A drop will slide down when the weight assisted by the electrical force overcomes the pinning force on the drop at the characteristic radius R_t . But due to the repetitiveness of the electrode design, drops will only be transported down one electrode period. At the converged gap between two adjacent electrodes, the top part of the drop has a larger electrode overlap compared to the bottom part, resulting in an electrostatic force working against the drop weight, as shown in figure 2.8b.

Drops will accumulate at the smallest gap resulting in vertical alignment of the condensate drops, as shown in the breath figure in figures 2.9d and 2.9e. Drops are only transported down once they touch both electrodes (neglecting any dissipation forces), resulting in transport starting close to the smallest gap, moving up where the distance between the electrodes increases, best visible in figure 2.9d.

Drops will be trapped until the drop weight overcomes the trapping and pinning force at the critical shedding radius $R_{crit,ZZ}$:

$$F_{g} = F_{p} + F_{t}$$

$$\rho \frac{2}{3} \pi g R_{crit,ZZ}^{3} = 2R_{crit,ZZ} \gamma \left(\Delta \cos \theta_{0} - \alpha \beta U_{rms}^{2}\right) + 2\beta \gamma U_{rms}^{2} \Delta l$$

$$\Rightarrow R_{crit,ZZ} = \left[\frac{3\gamma}{\pi \rho g} \left(\Delta \cos \theta_{0} - \alpha \beta U_{rms}^{2}\right)\right]^{1/2} + \left[\frac{2\gamma}{3\pi \rho g} \beta U_{rms}^{2} \Delta_{l}\right]^{1/3} \quad (2.19)$$

$$= R_{crit,IDE} + \left[\frac{3\gamma}{2\pi \rho g} \beta U_{rms}^{2} \Delta_{l}\right]^{1/3}$$

Where $F_t = -F_{e\downarrow}$ is the electrical trapping force.

The addition of the zigzag electrodes thus results in an increased shedding radius compared to the IDEs: $R_{crit,zz} > R_{crit,IDE}$. Where the electrostatic force assists the drops down toward the smallest gap, at radii smaller than with IDEs, the same force works against the drops at the smallest gap, resulting in an increased net shedding radius.

Dey *et al* showed that even though the shedding radius increases, the increased coalescence still results in a net heat transfer increase. They came to this conclusion by looking at indirect evidence of increased heat transfer using image analysis; no actual heat transfer or water collection experiments were performed. [32]

Comparing the condensation phase diagrams without electrowetting (figure 2.1) with the ones from IDEs (figure 2.5) and ZZEs (2.10) we notice an interesting behavior. Each extra electrode dimension that is added to the electrode design results in an extra phase in the condensation process. Without electrowetting there are 3 distinct phases, adding 1D IDEs results in 4 distinct phases and 2D ZZ electrodes in 5 distinct phases. Higher complexities in electrodes translates in higher complexities in DWC.

As will be presented in the results of section 4.3.1, active transport adds another dimension of electrodes and thus another phase in the condensation process.



Figure 2.8: Situation sketch of a drop on ZZEs. In (a) the drop experiences a electrical force pulling the drop down towards the apex of the gap. In (b) the drop is trapped at the gap apex by the electrical force.



Figure 2.9: Breath figures of DWC under EW with ZZEs. At t=o the surface is clean; the green overlay shows the location of the ZZEs. At t=44s drops start aligning horizontally at the apex where $R \approx R_g$. At t=63s coalescence due to gap alignment causes a rapid increase in drop size close to the apex. Drop are transported to the smallest gap. At t=117s drops further up the electrodes, where the gap, thus R_g is larger, also moved down. At t=217 the breath figure shows 2D alignment of drops stuck at the electrical traps. At t=379s shedding is initiated.



Figure 2.10: Condensation phases with EW on ZZEs characterized by the drop radius.

2.4 DROP IN MOTION

So far we assumed that a condensate drop is static, that is, it is not in motion. When a drop reaches its critical shedding radius it starts sliding (or rolling [66]) down and experiences a dynamic friction force F_d . The friction force plays an important role in resisting the downward motion of the drop that results (assuming the drop does not gain mass) in a constant sliding velocity.

The origin and size of the friction force depends on the type of surface and has been widely studied on both liquid and solid surfaces. Smith *et al* studied the dynamic of droplet shedding for liquid-infused surfaces (see section 2.5) in 2013, where they argued the total dissipation is the sum of the viscous dissipation within the drop, in the oil film beneath the drop and in the wetting ridge near the three-phase contact line. [66] A lot of research has been focussed on drop sliding and contact line friction of solid surfaces. [67] 't Mannetje *et al* studied the sliding velocity for drops on solid Teflon surfaces. They found a critical sliding velocity by solving

the force balance of the driving forces and resistive dissipative forces, where the dissipative forces are decomposed in a bulk contribution due to the Poiseuille-like flow profile and into an edge contribution due to the local contact flow patterns close to the contact line. [59]

Calculating the dynamic friction force is irrelevant for the electrode designs mentioned above; once a drop moves down, it keeps moving down.¹ However, as it turns out, with the double layered ZZEs this dynamic friction force becomes relevant, as will be explained in sections 4.3 and 4.5 where the workings and model of active transport are introduced.

The total dynamic friction force for the latter solid surface is determined by solving the force balance on the drop.

The total dynamic friction can be estimated as the sum of the bulk contribution F_{bulk} and edge contribution F_{cl} :

$$F_d = F_{bulk} + F_{cl} = \frac{4A^2\mu v\rho}{V} + 8R\xi v \approx vR(6\pi\mu\rho + 8\xi)$$
(2.20)

where $A \approx \pi R^2$ is the drop area, V the drop volume, v is the drop sliding velocity, μ the drop viscosity and $\xi = 71 \pm 10 \ mPa \ s \ [68, 69]$ the contact line friction coefficient.

The dynamic friction force is a linear function of the drop sliding velocity, which in turn depends on the net force acting on the drop.

2.5 LIQUID INFUSED SURFACES

Surface degradation is one of the main challenges with DWC. The rough hydrophobic surfaces degrade much faster due to the influence of condensing water vapor than smooth surface used for FWC, as elucidated in the introduction. An innovative technique for solving this problem is with the use of Liquid-Infused surfaces, used in this research.

Slippery Liquid Infused Porous Surfaces (SLIPS) or in short Liquid Infused surface(s) (LIs) are a type of substrates that achieve non-wetting properties using a lubrication layer. A porous substrate contains pockets of lubrication liquid rather than air to create a superhydrophobic surface on which the drop 'floats', as shown in 2.11. The hydrophilic lubrication liquid, usually an oil, remains in the textured substrate due to lower surface energies with the cavities being wet. [70] Inspiration for LIs came from the Nepenthes pitcher plant, in which microstructures

trap a thin layer of water, resulting in a slippery surface in contact with legs of insects. [71]

¹ There are situations where these forces become relevant for sliding drops. High friction forces result in slow sliding speeds. These slow moving drops do contribute significantly in the thermal resistance (the blanketing effect) and are thus included in some models including from Yamali *et al.* [40]

LIs have extremely low contact angle hysteresis of < 2.5 degrees [72] and other special properties such as self-healing, where the physical damage to the surface is restored within 0.1-1s by lubricant flow toward the damaged area by surface-energy-drive capillary action refilling the voids. [73] Applications of LIs include anti-corrosion at wind turbines and oil rigs [74], ship hull friction reduction [71], anti-icing for power lines [75] and aircraft wings [76] and medical applications [71], naming just a few.

More recently applications of LI surfaces in combination with EW systems has been studied by Hao *et al* [77]. They found that such a surface provides completely reversible EW responses for voltages over 500V and negligible contact line pinning.

Creating stable LI surfaces requires several design criteria. The main criteria established by Wong *et al* [72] are: 1) lubrication and liquid must be immiscible, 2) the lubrication oil must wet and spread on the solid surface; 3) the solid substrate must have higher affinity for the lubricating liquid over the probing liquid.

Depending on the surface energies of the liquid, lubricant and porous solid, several thermodynamic states are possible. The drop can either be cloaked by the lubricant, or not, and the lubricant can either encapsulate, impregnate or not wet the porous solid at all. All these states are explained by Smith *et al* [66]. The thermodynamic state the system is in helps us understand the behavior of the substrate, especially when studying surface degradation.

In this research we used Silicone oil as lubricant on a substrate coated with Polydimethylsiloxane (PDMS). See section 3.2.2 for the experimental details. Silicone oil is a hydrocarbon, thus immiscible with water and known to spread on PDMS [66] making it a good lubricant.

The check whether the drop is encapsulated by the oil or not, we calculate the oil-water spreading coefficient $S_{ow(a)}$. At room temperature $\gamma_{wa} = 72.2 \text{ mN/m}$, $\gamma_{ow} = 45.1 \text{ mN/m}$ [78] and $\gamma_{oa} = 21.2 \text{ mN/m}$ [79], we find:

$$S_{ow(a))} = \gamma_{wa} - \gamma_{ow} - \gamma_{oa}$$

= 6.6 mN/m (2.21)

So the drop is encapsulated by the Silicone oil, as shown in figure 2.11. During condensation experiments, the condensed drop is cooled below room temperature, resulting in an increase in the surface tension of water and an even larger spreading coefficient.



Figure 2.11: Situation sketch of a drop on a LIs.
3 EXPERIMENTAL ASPECTS

The homemade experimental setup for condensation experiments is schematically shown in figure 3.1 with the corresponding flow diagram in figure 3.2.

The full documentation of the experimental setup with extended experimental protocols, coating procedures, chamber assembly, 3D models, lists with parts and more can be found in appendix A.

Deionized water (Millipore Synergy UV, 18.2 $M\Omega$ cm) is heated on a hot plate (RCT Basic, IKA labortechnik). Ambient air is blown through the water using an aquarium pump (0886-air-550R-plus, Sera) and the flowrate is monitored by a flowmeter (AWM5101VN flowmeter, Honeywell). The water on the hot plate reaches an equilibrium temperature after 30-45 minutes. During this heat up time the water vapor is bypassed away from the condensation chamber using the bypass valve. The chamber is kept dry with a steady dry Nitrogen flow. During an experiment the humidified air is guided into the condensation chamber through two inlets at the bottom of the chamber. Vapor that condenses in the tubing collects at the lowest point in the tubing and is let out via the condensate-drain valve to the drain.

The humidified air passes in front of a vertically mounted cooled sample where the vapor partially condenses. The remaining vapor exits the chamber through a fine grid of holes at the top of the chamber. Temperatures inside the chamber are measured by multiple thermistors (TCS651m AmsTECHNOLOGIES and Thorlabs TSP-TH) using a DAQ card, a homemade voltage divider with $10K\Omega$ resistors, a complimentary LabView script and the Thorlabs TSP01 Application. Thermistors are located at the inlet of the chamber, close to the sample in the middle of the chamber, the outlet of the chamber, in the coolant behind the sample, in the heated water on the hot plate and the ambient air. These thermistors give the amount of subcooling in the chamber and ensure reproducibility of experiments. A humidity sensor (Thorlabs TSP01) is placed either outside the chamber or inside the chamber to measure the relative humidity. The transparent sample is cooled using a commercial cooler (Haake-F3-K, Thermo Fisher Scientific) by passing cooled water trough a coolant cell behind the sample.

For experiments with water collection the condensate on the sample slides down off the sample into a funnel that is connected to a collection tube or to a hydrophobically coated (Rustoleum Neverwet multi-surface coating) plexiglas funnel that direct the water to a collection tube on the balance (Nimbus Analytical Balance NBL 254i). The balance is readout using the ADAM DU software in realtime. To improve the lifetime of the coating in the funnel, the funnel is purged with a minimal flow of instrumental air to keep the temperatures down.

Images of the condensation process are recorded using a 4K camera (Point Grey,

FL3-U3) fitted with a 20X zoom lens (Z125D-CH12, EHD) for magnification. The sample is back illuminated using an LED light source (MB-BL305-RGB-24-Z, Metabright). Light passes through a window in the coolant cell, through the transparent coolant fluid, transparent sample, condensation chamber and heated window before being captured by the camera system. The heated window prevents condensation on the view port for the camera and is made of an indium tin oxide (ITO) coated sheet of glass through which a current is passed using a current source (Delta Elektronika Power Supply E 030-1).

High ambient humidity in the lab causes condensation on the coolant cell window when the dew point is reached. This decreases the light through-put to the camera. To counteract this, the humidity close to the coolant cell window is brought down with a steady flow of dry instrumental air.

For condensation experiments with EW, an alternating voltage is applied to transparent IDEs on the sample using one or two function generators (Agilent 33220A) and voltage amplifier(s) (Trek PZD700A).



Figure 3.1: Schematic of the experimental setup.



Figure 3.2: Flow diagram of the experimental setup.

3.1 REDESIGNED EXPERIMENTAL SETUP

The experimental setup used for obtaining the results in this thesis was custom designed and built for the purpose of this research.

3.1.1 New experimental setup

Earlier condensation experiments at the Physics of Complex Fluids group (PCF) were performed using a homemade condensation setup used for several publications [31, 32]. The condensation chamber was made out of plexiglas sheets that were glued and bolted together. The substrate was glued in place to seal if from the coolant water behind the substrate and EW connection wires were glued directly on the sample. Leaks between the coolant cell and chamber were common, changing the sample was complicated and time consuming and condensate collection was not possible. All together reason for designing and building a new condensation setup.

For easier, more extensive and efficient condensation experiments, several design criteria for a new condensation setup were resolved. These included easier switching of samples, condensate collection, more reliable EW connections that are faster to make, ease to expand or change the setup with future experiments in mind and high reproducibility of specific experimental conditions with the aids to measure it.

The new condensation setup was digitally designed in 3D in Google SketchUp and for the most part 3D printed with the thermoplastic acrylonitrile styrene acrylate (ASA). A picture of the setup is shown in figure 3.3. ASA was chosen for its good mechanical properties, UV stability and high chemical resistance.

The biggest part of the setup, i.e. the back wall of the condensation chamber with coolant cell, is CNC cut out of impermeable high-density polyethylene (HDPE). Several 3D printed versions with ASA were tested, but not one of these was perfectly water sealed as the pressurized coolant cells started leaking. CNC cutting proved to be the best solution. The setup is designed to be fully modular, consisting of various parts that are bolted together, see figures 3.4a and 3.4b for an assembled and disassembled render. This makes redesign and replacement of specific parts possible, such as sample holder for different types of samples.

The substrate is sealed from the coolant cell with a silicone ring and pressed in place with a sample holder that is bolted into the back wall of the chamber. This creates a perfect seal and makes replacing a sample a matter of minutes. EW connections are made through the sample holder using spring-loaded contacts. The connections are made when the sample holder is pressed against the sample and since they no longer exposed to the water vapor, they don't influence the experiments.

When volumetric condensation measurements are required, a newly designed sample holder with funnel can be used. This funnel connects directly to standardized laboratory centrifuge tubes for static water collection experiments or to the hydrophobically coated funnel that directs the water to a collection box on the balance for dynamic water collection experiments. See section 3.1.2 for an extended description about water collection.

The front of the condensation chamber slides out vertically after removing the top lid of the chamber and the new PCB connection for the heated window, for easy and fast access to the substrate.

The redesigned flow diagram, shown in figure 3.2, includes a by-pass valve for preexperiment stabilization of the water vapor, a condensate drain valve for draining the condensed vapor in the tubing and valves for Nitrogen and instrumental air purging. The whole setup uses standardized pneumatic tubing with quick release fittings that allows for easy expansion of the setup.

For illumination of the breath figure for image recording a new holder was designed, also shown in figures 3.4a and 3.4b. For image recording purposes the illumination source hold a circular aperture to get circular light transmission through the drops, see section 3.4.1.

3.1.2 Water collection

To approximate the average heat flux during a condensation experiment, the volumetric condensation rate $\langle v \rangle$ was obtained by collecting the condensate that rolls of the substrate. Water collection was done with two different approaches, discontinuous water collection and continuous water collection.

Discontinuous water collection collects the condensate that slides of the surface in a funnel that directs it in standardized laboratory centrifuge tubes. Water is collected over the full duration of an experiment, after which the water is weighted to determine the condensation rate (calculation in section 3.4.2.1). The setup in



Figure 3.3: A part of the experimental experimental setup. On the left the circular back illumination is visible, in the middle the condensation chamber with the heated window on the right, on the right the camera lens is visible.

discontinuous water collection is shown in the figures 3.4a and 3.4b.

Continuous water collection funnels the condensate to a hydrophobically coated funnel that directs the condensate to a collection box on a balance. The \sim 15 cm long funnel is coated with Rust-Oleum NeverWet Multi-Surface hydrophobic coated that is applied with at least 3 base layers. The balance is connected to a computer to read out the mass of the condensate in real time. Afterwards the (local) average condensation rate is determined by linearly fitting of the data, see section 3.4.2.2.

There are advantages of continuous water collection over discontinuous water collection. These include

- the possibility to change experimental conditions, such as EW voltage, during an experiment; therefore eliminating the extra step of restarting the experiment and pre-heating the condensation chamber. Also, since the experimental conditions between two experiments are never exactly the same, this also gives the option to get reference measurements between the actual measurements;
- 2. more accurate condensation rates. Discontinuous experiments always includes the lower condensation rate during the heat-up phase of the chamber at the

start of an experiment, therefore underestimating the actual condensation rate;

3. the possibility to exclude experimental errors that are caused by the incidental condensate from places other than the substrate from leaking into the collection tube.

The accuracy in determining the condensation rate with both these methods and two other image analysis methods is tested during this research. See sections 3.4.2 and 4.2.

If chosen for no water collection experiments, but solely image recording of the breath figure, the sample holder with built in funnel can be swapped out for a (non protruding) sample holder without funnel. Similarly the bottom of the chamber can be swapped out for a solid bottom to seal the hole when no tube is present.

3.1.3 Chamber stability and reproducibility

We tested the performance of the new setup by characterizing the results in two groups: stability and reproducibility. With stability we look at the experimental conditions within one experiments, how stable are these conditions. With reproducibility we check the experimental conditions with the same settings between multiple experiments.

Chamber stability

We performed an 83-hour condensation experiment without EW to check the stability of the chamber. These 83 hours is an extreme case as usual experiments last only 2 hours. We thus expect the fluctuations mentioned here to be a worst-casescenario.

Table 3.1 lists average sensor values and standard deviations from this experiment. The abbreviations Labview (LV) and Thorlabs (TL) stand for Labview and Thorlabs, respectively, referring to the brand of thermistors, electronics and readout software used. In the condensation chamber sensors are located at 3 locations: at the bottom, i.e. at the vapor inlets, in the middle close to the sample (~1 cm from the sample for the Labview sensor, ~2 cm for the Thorlabs sensor) and at the vapor outlet at the top of the chamber. The coolant cell behind the sample has its own thermistor and also the flow are, relative humidity and ambient temperature are measured continuously. The average subcooling during an experiment is defined as $T_{middle,LV} - T_{coolant cell,TL}$.

We note barely any fluctuations in the coolant cell, while the condensation chamber temperatures fluctuate with about 1.5°C. These fluctuations come forth from the delicate equilibrium the setup depends on. Fluctuations in for example ambient temperature or flow rate cause a new equilibrium in the water boiler resulting in different chamber temperatures. Also, during an 83-hour experiment the water boiler has to be refilled twice. Even though this is done very slowly, the equilibrium water temperature is different afterwards. The flow rate depends on the water level in the boiler. A full boiler results in flow rate of 3.5 SLPM, an almost empty boiler in 3.95 SLPM.

This data does not show the initial chamber fluctuations during heat-up when the vapor is just introduced into the dry and cold chamber. After the introduction of vapor into the chamber it takes about 11 minutes to reach temperatures within 5% of the overall-averages. It takes 30-45 minutes, depending on the vapor temperature, to reach full equilibrium.

The chamber relative humidity is about 5-10% before vapor is introduced into the chamber. At equilibrium the relative humidity is 100%.

	Sensors	Value
$T_{middle,LV}$	1	$33.7 \pm 1.5^{\circ}C$
$T_{middle,TL}$	1	$34.8\pm0.8^\circ C$
$T_{\mathbf{top,LV}}$	2	$33.4 \pm 1.6^{\circ}C$
$T_{bottom,LV}$	2	$35.8 \pm 1.8^{\circ}C$
$T_{\rm coolant\ cell,TL}$	1	$10.44\pm0.02^\circ C$
Flow rate	1	3.5 – 3.95 SLPM
RH lab	1	$52.3 \pm 4.1\%^{*}$
$T_{ambient,TL}$	1	$22.6\pm0.2^\circ C^*$

 Table 3.1: Average sensor values during an 83-hour experiment. * = these values vary significantly between experiments.

Chamber repoducibility

For the reproducibility of the experimental conditions in the chamber we collected the data of 60 experiments (>150 hours of data). The averages and standard deviations are listed in table 3.2. Only condensation chamber temperatures are listed here, as these fluctuate most. To change the experimental subcooling, we change the vapor temperature by setting a different hot plate temperature. Hot plate temperatures of 150, 190 and 200°C were the most common ones.

We observe temperature fluctuations of 2.8-3.8°C between experiments, showing that duplicating exact experimental conditions is not possible. These errors arise from both the inaccurate setting of the hot plate temperature as well as the ambient fluctuations mentioned previously. These fluctuations are not directly translated into our results, as we normalize our data using the subcooling per experiment, see section 4.1.

$T_{\text{hot plate}} [^{\circ}C]$	150	190	200
$T_{\mathbf{bottom}} [^{\circ}C]$	36.9 ± 3.7	37.7 ± 3.3	42.3 ± 3.2
$T_{\mathbf{middle}} [^{\circ}C]$	34.3 ± 3.4	36.1 ± 2.8	38.9 ± 3.8
$T_{\mathbf{top}} [^{\circ}C]$	33.9 ± 3.5	35.7 ± 2.8	37.3 ± 3.2

Table 3.2: Average chamber temperatures of 60 experiments (>150 hours of data).

3.1.4 Recommendations for further improvement

The current experimental setup is based off the first iteration of the experimental setup that was used at Physics of Complex Fluids group (PCF). Many of the deficiencies and impracticalities of this setup were solved by the new experimental setup. However, after almost 700 hours of condensation experiments, several shortcomings have come to light. Some recommendations for further improvement to the setup inude: Humidity control. Currently only experiments are 100% relative humidity can be carried out with this experimental setup. Humidity control could be achieved by a commercial system or my a home-made feedback loop dat mixed dry air with humid air via two electronic valves. With such a system an accurate and actively controlled equilibrium can be achieved in a shorter time and the effect of humidity on condensation rates can be investigated; Pre-equilibration of the condensation chamber before starting an experiment. The current experimental procedure introduces a flow in the chamber at the start of an experiment, followed by a significant equilibration period of the chamber reaching equilibrium temperature and humidity. This equilibration time is impractical for experiment while still deteriorative for the substrate. A shutter based system in which the substrate is isolated from the vapor could be a possible solution; A fully closed system system to allow experiments with other fluids than water. Currently the non-condensed water vapor exits the condensation chamber through holes in the top lid, while condensed vapor on other places than the substrates leak out of the bottom of the chamber. A closed loop-system would allow to reuse these fluids and vapor. The full experimental documentation in appendix A lists several more recommendations for improvement.



(a)



Figure 3.4: 3D renderings of the main parts of the experimental setup.

3.2 SUBSTRATES

The substrates consist of a 70x70mm glass substrate covered with a thin transparent patterned electrode pattern of indium tin oxide (ITO), which is covered with a dielectric layer and hydrophobic top coating, which will be specified below. The ITO electrodes are obtained via photolithography and etching in a solution of 18% HCl, creating gaps between the electrodes. In the condensation chamber roughly $1500mm^2$ (30% of total substrate area) of the sample was exposed directly to the water vapor, due to the clamping system holding the sample in place.

3.2.1 Electrode designs

Three different types of ITO designs are used to control the behavior of the condensate on the surface: IDEs, single layered ZZEs and double layered ZZEs.

Figure 3.5 shows a schematic of a substrate with IDEs where the electrodes are enlarged for clarity. The electrodes consist of two parallel horizontal electrodes on the top and bottom of the substrate with vertical IDEs. The ITO electrodes are covered by a dielectric layer and a top coating, as shown in the horizontal cross section in figure 3.6. By applying a voltage over the two phases, a local electric field is generated. The pitch of the electrodes is defined as $pitch = w_e + w_g$ where w_e is the width of the electrode and w_g the width of the gap between two electrodes. Two designs of IDEs are used with a pitch of $50\mu m$ and $200\mu m$ and equal width-gap ratios. During condensation experiments the sample is mounted vertically with gravity pointing along the IDEs.

Figure 3.7 shows a schematic overview of ZZEs, with again the electrodes enlarged for clarity. The IDEs here are triangularly shaped introducing an extra dimension in the vertical direction defined as length *d*. The horizontal cross section is equal to the IDEs shown in figure 3.6. The gap width 2b is defined as the shortest distance between the electrodes, the electrode width 2a + 2b as the widest point of the electrodes. Two designs of ZZEs are used with a lengths $d = 1500\mu m$ and $d = 3000\mu m$, $2b = 50\mu m$ and $2a + 2b = 250\mu m$. The substrate is mounted with the triangular electrodes pointing opposite to the gravitational force. The gap between the electrodes has the same dimensions as the electrode.

The third electrode design is an extension of the ZZEs design where two pairs of ZZEs are stacked with a vertical shift of half a period, as shown in figure 3.8. The second layer of IDEs were aligned using an optical microscope, which was found to be 99% accurate. The two pairs of electrodes are separated by a dielectric layer creating four isolated electrodes, as shown in figure 3.9. Actuation of the electrodes is done using two voltage sources with amplifiers which can be controlled to pulse out of phase of each other using a Labview program.



Figure 3.5: Schematic top view of IDEs.



Figure 3.6: Schematic side section-cut view of IDEs and ZZEs.



Figure 3.7: Schematic top view of ZZEs.



Figure 3.8: Schematic top view of double layered ZZEs for active transport.



Figure 3.9: Schematic side section-cut view of double layered ZZEs for active transport.

3.2.2 Substrate coatings

Two types of coatings were used to isolate the electrodes and creating a hydrophobic top layer.

The substrates with ITO IDEs and single layered ZZEs were fabricated as Lls. See section 2.5 for more details about Lls. The ITO electrodes were obtained via photolithography.

For the top coating Polydimethylsiloxane (polydimethylsiloxane (PDMS), Sylgard-184) was mixed in a 10:1 base:agent mass ratio and degassed in a vacuum chamber for 20 minutes. The substrate with ITO electrodes was spin coated at 2000 rpm for 5 minutes to obtain a 9.3μ m thick insulating layer that separates the electrodes from the drops and to act as porous medium for oil-infusion. This is thinner than the expected value of 13μ m.[80] After spin coating the substrate was baked for approximately 3 hours at 60°C. The substrate was then soaked in Silicone oil (FC-70, Sigma Aldrich) for hours to infuse the porous PDMS. Right before a condensation experiment the substrate was taken out of the oil and the excess oil was blown off with Nitrogen.

The substrates with double layered ZZEs were fabricated with Teflon coatings instead LIs, due to the poor adhesion of PDMS to the dielectrics layers needed for the double electrode design.

The electrode design was fabricated in a two time repeating process of photolithography followed by spin coating a layer of SU-8 (BRAND) to act as an insulating layer between the electrodes and the drops.

The substrate was then spin coated with Teflon AF 1600 (DuPont) at 500rpm for 45 seconds to create a layer of approximately $2\mu m$ thick. The substrate was heated on a hot plate for a couple of seconds to evaporate the solvent and baked at 195°C for approximately 2 hours.

3.2.3 Surface characterization

Several substrates were characterized to determine the contact angle hysteresis (CAH) as a function of applied voltage. This was done for LI and Teflon coated substrates before and after 40+ hours of condensation experiments with EW to study the effect of surface degradation.

The EW curves are shown in figure 3.10. $\Delta \cos \theta$ and η are defined in equations 2.14 and 2.13, respectively.

We approximated the observed behavior as a linear decreases of hysteresis with $\eta \sim U^2$, followed by a plateau. This behavior follows the EW equation (equation 2.14) up to the plateau, i.e. the saturation regime caused by local breakdown at the contact line.[81]

Before condensation the CAH for LIs decreases rapidly with voltage to basically zero. The oil lubrication layer makes a drop 'float' on the surface reducing the pinning forces. This behavior is also visible in the image taken during the measurements, shown in figure 3.11. Figure 3.11a shows the water droplet deforming the oil lubrication layer causing the oil to creep up and form a meniscus, similar to figure 2.11. For comparison does figure 3.11b show a water drop on a Teflon coated surface.

In contrast to the LI CAH, the Teflon surface does show a finite CAH, even at large voltages. After condensation, both Teflon and LIs degraded. At lower EW numbers ($\eta < 0.2$) the CAH for Teflon and LI are similar. However, Teflon does show larger errors indicating more local pinning.

Comparing the CAH in the saturation regimes, we observe that the CAH for LI is always lower than Teflon. The increase in CAH from before to after EW for LI is a factor of 9, while this increase for Teflon is only a factor of 2.

The surface degradation of both coating with and without EW during condensation experiments is studied in detail in this research. For the results, see section 4.6.



Figure 3.10: Contact angle hysteresis $\Delta \cos(\theta)$ as a function of the EW number η due to AC EW.



Figure 3.11: Images from Optical Contact Angle measuring device (OCA) showing (a) a drop on a IDE LIs before condensation with EW at 350V and (b) a drop on a IDE Teflon coated surface before condensation with EW at 150V.

3.3 EXPERIMENTAL PROCEDURES

Before each experiment water on the hot plate was heated for at least 90 minutes, while being purged with ambient air to reach an equilibrium. The vapor was redirected away from the chamber to the drain valve. 20 minutes before the start of the experiment the water cooler was turned on with flow through the coolant cell. The condensation chamber was kept dry with a steady flow of Nitrogen through the chamber.

With the camera an image of the sample without drops was recorded to be used as background image for the image analysis. Any surface damage or permanent bubbles in the coating is removed from the analysis using this image. An image at high shutter speed was recorded that revealed the electrodes on the surface. This image was used to determine the conversion factor from pixels to micrometers.

For discontinuous water collection experiments a collection tube was connected to the sample holder. At the start of the experiment the Nitrogen flow was turned off, the by-pass valve was turned to allow flow in the chamber and the image recording was started. After 120 minutes the valve was closed, the image recording stopped and the collected water in the collection tube was measured. Since the humidity and temperature in the chamber at the start of the experiment are low, the chamber reaches an equilibrium about 30 minutes into the experiment.

For continuous water collection experiments the hydrophobically coated funnel was connected and directed into a collection container on the balance. At the start of the experiment the Nitrogen flow was turned off, the by-pass valve was turned to allow flow in the chamber and the image recording was started. The chamber was then allowed to equilibrate for at least 45 minutes. Then for variable periods of at least 45 minutes the water condensate mass was recorded for different EW voltages or actuation pulses.

All experiments are performed at 100% relative humidity (RH) in the bulk vapor (values are lower closer to the condenser surface) and in the presence of non-condensable gases (NCG).

With volumetric condensation measurements each sample was not used more than 16 hours.

3.3.1 Sample actuation

When using EW, sample actuation, e.g. AC waveforms and frequency, is one of the primary variables. The biggest part of this study used Lls (see section 2.5 and 3.2.2). With Lls the EW voltage was 300V up to 450V, with an AC frequency of 1kHz. Teflon coated surfaces (see section 3.2.2)) were used for active transport (section 4.3), due to the complex layered electrode design. These double layered ZZEs were actuated with different voltages due to different dielectric layer thickness. The top layer was actuated at 125V at 1kHz, the bottom layer at 150V at 1kHz. The layers were attenuated out of phase for 2 seconds each using a custom build Labview program. Teflon surfaces were also used to compare surface degradation (section 4.6) and for calculations in the model (section 4.5). The AC EW voltage here was 150V for Teflon and the frequency 1Khz.

For pulsed EW, a 2 second burst at 150V 1kHz was given every 15 seconds.

3.4 DATA PROCESSING

3.4.1 Image analysis

The high contrast color images recorded of the condensation process are loaded onto a computer for analysis using a home-made program in Mathworks MATLAB. The goal of the image analysis is to extract the exact location and size of every drop in the image and from there estimate the total volume on the surface and condensation rate.

This procedure is complicated by the fact that each drop shows circular holes in the image due to the transmitted light of the circular light source as well as the complication of overlapping drops in the image due to limited image resolution, poor focus or the physical problem large contact angles where small drops can exist partially underneath large drops.

The key for reliable image analysis is in the quality of the recorded images. The recorded images need to be focused carefully, the light source has to be circular and the contrast between drops and the background must be high and even (no vignetting).

A shortened version image analysis steps is shown in figure 3.12. First the original image (figure 3.12-(a)) is converted to a grayscale image (figure 3.12-(b)). Then a background image, that is an image with no drops on the surface recorded before a

condensation experiment, is subtracted from it and the contrast is increased (figure 3.12-(c)). Otsu's method is used to automatically determine the binary threshold to convert the image to a binary image. The Otsu's method determines the binary threshold in a manner such that the combined spread (variance) of black and white pixels is minimal. Using the obtained threshold, each image is then converted to a binary image, see figure 3.12-(d). In order to remove the holes inside the drops the separate background components (regions of connecting black pixels) are detected. Each connected component with a threshold eccentricity and radius are filled (inset figure 3.12-(e)-(i)). Filling based on the eccentricity and radius prevents filling connected drops. Aforementioned, some drops are slightly connected in the binary image (inset figure 3.12-(e)-(ii)). Separating these drops starts by taking the distance transform of the image where each foreground (white) pixel is transformed into the closest distance to a background (black) pixel. The resulting image (inset figure 3.12-(e)-(iii)) shows a gradient from white to black where the darker the color, the further that pixel is inside a drop. The tiny local minima are removed and the watershed transform is applied to the distance transform and translated to the binary image. The watershed transform turns local minima in the foreground pixels (the shortest connection between two connection drops) into background pixels, thus splitting the connected drops (inset figures 3.12-(e)-(iv) and 3.12-(e)-(v)).

The non-circular holes in the drops are filled and the connected components of the drops are determined. Each component now represents one drops. For drops not touching the border of the image (non-boundary drops) is assumed that they are circular. Assuming a spherical projection of the drops on the surface the radius and center of each drop is calculated. The volume of each drop is calculated using a spherical cap with a contact angle determined from separate contact angle measurements.

Drops touching the border of the image (boundary drops) undergo an extra analysis step. Each of the components contours are circle fitted from which the radius and center location is determined. The volume inside the frame is determined based on the radius, the part of the drop inside the image and the assumption that the drop is a hemisphere.

The end result of the image analysis is shown in figure 3.12-(f) where each drop is numbered uniquely.

Now that the location and center of each drop is known, data can be extracted from the breath figures. This data includes the radius of shedding drops, surface coverage and total volume in the breath figure. The total volume assumes a constant drop contact angle of 115° to calculate the volume of the spherical caps inside the frame. For drops touching the edge of the frame, the volume inside the frame is estimated assuming hemispherical drops minus (half) spherical caps.

Proper drop detection is mainly a matter of proper image recording. High contrast images with uniform lighting and circular light transmission through the drops are crucial.



Figure 3.12: The basic image analysis procedure for determining the location and size of every drop. The original image (a) is converted to grayscale (b). The background image is subtracted and the contrast is increased (c). The image is converted to a binary image (d), holes inside drops are filled and connecting drops are separated (e). Holes in drops are filled, the separate components are detected and the boundary drop detection is applied (f).

3.4.2 Condensation rates

The average volumetric condensation rate ($\langle v \rangle$) is the mass of vapor that condenses on the surface per unit time. The condensation rate is determined in 5 different methods, of which 4 give an exact condensation rate, shown in the diagram of figure 3.13. 2 of these methods are water collection techniques (weighing): the discontinuous water collection method (disc-method) where water was collected for 2 hours straight, and the continuous water collection method with intra-averages (cont-method), where water was directly guided to a balance to get a direct measurement. The other 2 techniques are image analysis based. The automated image analysis method (auto-method) determines the total shedded volume using a simple algorithm, this is either done fully automatically or partially manually. The second image analysis technique was the calculation method (calc-method) where the average drop critical shedding radius and shedding frequency was used to determine $\langle v \rangle$. This latter method was used by Dey et al [32] to show an increased condensation rate for ZZEs.

The 5th technique, cont-method with inter-averages measures the condensation rate using continuous water collection and calculates the change compared to a direct (no EW) reference directly before or after the measurement.

All techniques will be discussed in detail below. The accuracy and (dis)advantages of each technique are discussed in section 4.2.



Figure 3.13: Branch diagram showing the 4 tested methods to determine the average condensation rate $\langle v \rangle$.

3.4.2.1 disc-method (discontinuous water collection)

Discontinuous water collection collects condensate over a period Δt on a surface with area A. The condensate is collected outside the condensation chamber and its mass M is determined post-experiment. $\langle v \rangle$ is than determined by averaging the condensation rates over N experiments and by normalization with the area, time and subcooling, the latter one being possible by the linear relationship between the condensation rate and subcooling (equations 2.8 and 2.10). This gives an average volumetric condensation rate of

$$\langle v \rangle_{disc} = \frac{1}{N} \sum_{n}^{N} \frac{V_n}{A_n \Delta T_n \Delta t_n} = \frac{1}{N \Delta t A} \sum_{n}^{N} \frac{V_n}{\Delta T_n} \quad \left[m^3 s^{-1} K^{-1} m^{-2} \right]$$
(3.1)

Here we restrict the average to experiments with the same duration Δt and condensation substrate area A.

The condensation rate of each experiment V_n is normalized using the average subcooling of the entire experiment.

3.4.2.2 cont-method (continuous water collection with intra-averages)

With continuous water collection, condensate from the surface is directed via a funnel to a collection box on a balance where the mass is recorded realtime. This allows for switching experimental conditions during experiments, e.g. EW voltage, and directly see the effect of it. The period Δt over which water is collected varies between 30-60 minutes. In Δt about 35-50 drops are collected on the balance, which, due to pinning inside the funnel, is less than the number of shedding events. The recorded change in mass is linearly fitted to calculate an average condensation rate. For N of these periods spread out over multiple experiments, the average condensation rate is:

$$\langle v \rangle_{cont} = \frac{1}{AN} \sum_{n}^{N} \frac{V_{n,fit}}{\Delta t_n \Delta T_n}$$
(3.2)

The condensation rate of each period $V_{n,fit}$ is normalized using the average subcooling ΔT_n of that period.

This method of averaging the exact condensation rates over multiple periods and experiments is referred to as intra-averages. A variation of the cont-method with inter-averages is explained in section 3.4.2.5.

3.4.2.3 auto-method (image analysis with automated shedding algorithm)

The automated condensation rate is determined using the recorded images of the breath figure during condensation. The recorded images are processed in MATLAB to determine the total volume in the field of view for each frame using a constant contact angle of 115° (section 3.4.1), this is differing from the calc-method ahead where the drop is assumed to be hemispherical for calculative simplicity. The total shedded volume is automatically estimated by the addition of negative volume changes between 1250 individual frames (1/3 fps) between 50-62.5 minutes. After 50 minutes the condensation chamber is assumed to be at equilibrium. Experiments have shown that fluctuations between analysis windows of 1250 frames beyond 30 minutes are less than 8%. If, due to the image analysis errors, the automated algorithm proves to be too inaccurate, e.g. there are large spikes in the volume due to watershed transform failure, the shedded volume is determined semi-manually by adding up the volumes of shedding drop manually.

The final average condensation rate is averaged and normalized similar to the disc-method, resulting in:

$$\langle v \rangle_{auto} = \frac{1}{NA_{FOV}\Delta t} \sum_{n}^{N} \frac{V_n}{\Delta T_n} \quad \left[m^3 s^{-1} K^{-1} m^{-2} \right]$$
(3.3)

Where A_{FOV} is the area of the field of view of the camera and $\Delta t = 12.5$ minutes.

3.4.2.4 calc-method (image analysis with average calculation)

The calc-method calculates the condensation rate based on the average shedding radius $\langle R_{sh} \rangle$ and frequency $\langle f_{sh} \rangle$ from the image analysis. For calculative simplicity the drop is assumed to be hemispherical. For consistency the average shedding radius and frequency are determined from the same 1250 frames as the auto-method (between 50–62.5 minutes). In these 1250 frames, the number of shedding drops is on average 36.

A drop is considered a shedding drop if a) it moves down (slowly) and b) it does not pin elsewhere on the sample, thus moves out of frame continuously.

Estimating the drop as a hemisphere, the average condensation rate for N independent experiments is:

$$\langle v \rangle_{calc} = \frac{2\pi}{3NA_{FOV}\Delta t} \sum_{n}^{N} \frac{\langle R_{sh} \rangle_{n}^{3} \langle f_{sh} \rangle_{n}}{\Delta T_{n}} \quad \left[m^{3} s^{-1} K^{-1} m^{-2} \right]$$
(3.4)

Where $\Delta t = 12.5$ minutes.

This method is used by Dey et al [32] to estimate the condensation rate for different types of samples.

3.4.2.5 cont-method (continuous water collection with inter-averages)

The experimental side of continuous water collection with inter-averages is identical to the cont-method described in 3.4.2.2. The inter-averages refers to the processing of the data. Instead of averaging the exact condensation rates of all periods, inter-averages uses a direct reference, i.e. no-EW, period right before or after the measurement. The condensation rate $\langle v \rangle^{EW}$ is divided by the condensation rate of the reference $\langle v \rangle^{ref}$ to determine a percentile change in condensation rate. The average percentile change $\langle \langle v \rangle^{ew} / \langle v \rangle^{ref} \rangle$ is obtained by averaging multiple (*N*) of these percentile changes:

$$\left\langle \frac{\langle v \rangle_{cont}^{ew}}{\langle v \rangle_{cont}^{ref}} \right\rangle = \frac{1}{N} \sum_{n}^{N} \frac{\langle v \rangle_{cont,n}^{ew}}{\langle v \rangle_{cont,n}^{ref}}$$
(3.5)

If a reference is taken before and after the measurement, the one after is used, if no reference is available before or after, the dataset is not used.

An example inter-averages is shown in figure 4.8 from the results section where the linear fitting and averaging is shown.

The main advantage and reason for development of this technique is the nonsusceptibility to changes in experimental conditions between experiments, and within experiments. Experimental conditions like subcooling or flow rate tend to fluctuate a lot between experiments, and even within long experiments. Calculating exact averages results in large errors, while using a direct reference minimizes these errors. The biggest disadvantage is that no exact condensation rates can be calculated.

3.4.3 Fitting of critical shedding radius

The temporal evolution of the critical shedding radius is measured in section 4.6 for determining the amount of surface degradation.

We assume surface degradation is an ephemeral process; surfaces degrade and reach a constant degraded state after some time. This is visible in the surface critical radii plot in figure 4.17a of the results section. This allows us to fit two lines to the data: an initial fit to reach the constant value, and a constant line for the equilibrium data.

First the equilibrium data is fitted with a horizontal line to the equilibrium data, i.e. the data that remains relatively constant compared to the data at the start of an experiment. Assuming the critical radius converges to the equilibrium value, we fitted an exponential function to all the data with the boundary condition that it converges to the equilibrium value ($R_{crit,equilibrium}$). This function has the form of

$$R = R_{\text{crit,equilibrium}} \cdot \exp\left[b \cdot \exp\left(c \cdot t\right)\right]$$
(3.6)

Where *b* and *c* are fitting parameters. Using the fitting toolbox in Matlab we find the best fit.

4 RESULTS AND DISCUSSION

In this chapter we present the results. **Section 4.1** starts off by comparing the average heat transfer of multiple types of substrates as a function of subcooling. **Section 4.2** compares different methods that were used to determine the condensation rate. In **section 4.3** the workings of active transport condensation are explained and the efficiency is discussed. **Section 4.4** compares the condensation rate of a wide range of types of samples acquired using the continuous water collection technique. **Section 4.5** presents a model for active transport that can be used to optimize future active transport electrode designs. Finally, in **section 4.6** the surface degradation during long term condensation experiments is quantified.

4.1 SUBCOOLING DEPENDENCY

We measured the average condensation rate with discontinuous water collection (disc-method, see section 3.4.2.1) experiments of 2 hours. We did this for 4 different types of samples: no EW and EW with IDE of 200µm pitch, IDE of 50µm pitch and ZZEs with a length of $3000 \mu m$. For each experiment the average subcooling was determined and the average heat flux followed directly from the condensation rate using equation 2.10 with $h_{fg} = 2.26 \ kJ \cdot kg^{-1}$. [82] The data is shown in figure 4.1 with errorbar corresponding to the average deviations of $\Delta \langle v \rangle \approx 125 \ mm^3 \cdot s \cdot m^{-2} \Rightarrow \Delta \langle q'' \rangle \approx 2.6 \ W \cdot m^{-2}$ from experimental data. We observed a linear increase in average heat flux as a function of the subcooling for small subcoolings, as fitted with the dashed line. This trend follows the predictions from equation 2.8 and is also observed in literature. [35, 83] However, for subcoolings above $\sim 35^\circ \mathrm{C}$ this trend seems to break down. This nonlinear behavior is thought to be a result of the sweeping behavior. At higher subcoolings drops grow faster and consequently is sweeping more common. As a result the average critical shedding radius further down the surface, where sweeping is more common, reduced and thus the average heat transfer, as explained in section 2.1.2.2. The importance of shedding on the heat transfer is well known as vertical elongated surfaces show higher average heat transfers than horizontal elongated surfaces, as Yamali et al [40] showed using a heat transfer model. Only when sweeping is rare linear behavior is observerd. Similar nonlinear behavior has been shown in literature, as summarized by Khandekar *et al.* [4]

We observed similar average heat flux for all types of samples using this discontinuous water collection technique. We do note that the error the heat transfer is large and thus causes an inaccuracy in this data. With our current experimental setup exact reproducibility and stability of the experimental conditions are not feasible, as explained in section 3.1.3. The reason we do not measure any improvements exceeding the error concludes that neither of the electrode designs have major improvements on the heat transfer.

The improvements we might be looking for are too small to measure it using the disc-method, makes us seek for a better method to measure the average heat transfer, done in the next section.

The linear behavior of the subcooling on average heat flux (and condensation rate) allows us to normalize our condensation rates by division with the subcooling, which is done from here on.



Figure 4.1: The average heat flux as a function of condensation subcooling for different types of (functionalized) LI substrates. Data collected using discontinuous water collection (disc-method). Data with subcooling <35K is fitted linearly (black line).

4.2 COMPARISON OF TECHNIQUES TO DETERMINE THE CONDENSATION RATE

To study the different alternatives in determining the average volumetric condensation rate ($\langle v \rangle$), $\langle v \rangle$ was determined using 4 different methods. These methods were discontinuous water collection (disc-), continuous water collection (cont-), image analysis shedding algorithm (auto-) and average calculation from image analysis (calc-), described in detail in section 3.4.2.

Each of the 4 methods was used to determine $\langle v \rangle$ for 4 different types of substrates: no EW (0V), IDEs with 50-50 μm gap-electrode widths, IDE 200-200 and ZZEs with $d = 3000 \mu m$ long electrodes. The total number of combination acquired during these experiments is thus 16.

The resulting $\langle v \rangle$ are shown in the boxplot of figure 4.2 where $\langle v \rangle$ is normalized with the average subcooling. The subscripts following the type of substrates on the x-axis indicate the total number of datasets used for that datapoint. Each dataset being a 2 hour experiment. While the disc- and cont-method could not be measured simultaneously, the auto- and calc-method could be used simultaneously with either of those methods.

We will go into detail for each method, validating the accuracy and limitations of each method and differences between the types of substrates.



Figure 4.2: The average volumetric shedding rate per unit area per degree of subcooling for 4 different configurations (oV, IDE 200-200, IDE 50-50 and ZZE d=3000) on LIs, calculated with the 4 tested approaches: discontinuous water collection (**disc**-method), continuous water collection with intraaverages (**cont**-method), automated volume addition from image analysis (**auto**-method) and calculated based on averages from image analysis (**calc**method).

The subscripted numbers indicate the total number of datasets used for that datapoint. Red marks indicate the median, the bottom and top edges of the box indicate the 25th and 75th percentiles, respectively, the whiskers extend to the most extreme data points not considered outliers and the outliers are marked with red dots. For the disc-method only data with subcooling below 35°C is included. For image analysis data only correctly detected data is included (flawless datasets). Methods auto- and calc- use data between 50 and 62.5 minutes from the start of the condensation experiment (equilibrium data).

4.2.1 disc-method (discontinuous water collection)

We observe similar condensation rates of $\sim 10 \ mm^3 s^{-1} K^{-1} m^{-2}$ for all types of substrates when collecting water using the disc-method. We assume these condensation rates underestimate the actual (equilibrium) condensation rates, as the 2-hour measuring time includes the 30-45 minute heat-up time of the condensation

chamber (section 3.1.2).

There is no increased condensation rate visible for any of the substrate types, in accordance with section 4.1.

The disc-method being the most straight-forward technique, is not accurate enough to distinguish small differences in the condensation rates between the different types of substrates. This is due to limitations of the experimental setup, where fluctuations in temperature and flow-rate exceed the (possible) small differences in condensation rates between the different types of substrates.

4.2.2 cont-method (continuous water collection with intra-averages)

Using the cont-method we observe overall higher condensation rates compared to the disc-method. This increase comes from the lack of the heat-up phase of the condensation chamber.

Comparing the different types of substrates and considering the medians, we do see an increased condensation rate for IDE substrates, and decreased rate for ZZEs, both compared to the 0V reference. This could indicate that EW with IDE substrates increase the condensation rate, while EW with ZZEs decreases the condensation rate. But the statistics are poor for the IDE-type substrates where only few datasets are used, and the error bars are large for the reference and ZZEs, making hard conclusions impossible.

The cont-method is in general the most accurate method of the 4 in the section to determine the condensation, although figure 4.2 does not show that directly. With the cont-method data is normalized using the temporal evolution of the chamber temperature, instead of the overall average. In this way, fluctuations in temperature are accounted for more accurately. Also, the heat-up phase of the condensation chamber is not included in the final condensation rate. Compared to the image analysis methods below, this method is straight-forward and unquestionable.

This method formed the basis for an even more accurate method of continuous water collection with inter-averages, which reduces errors by excluding fluctuations in experimental conditions between and within long experiments. This method, described in section 3.4.2.5, which only determines percentile differences from the reference, is used in section 4.3 for active transport and in section 4.4 for several other types of substrates.

4.2.3 auto-method (image analysis with automated shedding algorithm)

The auto-method is in contrast to the disc- and cont-methods, not an weighing, but image analysis method. Based on the breath figures recorded of a part of the substrate the condensation rate is estimated using an algorithm.

The resulting condensation rates are similar to the disc- and cont-methods.

We conclude that the auto-method is reasonably accurate for determining the condensation rate, but not as accurate as the experimental disc- and cont-methods, and also not accurate enough to determine the differences between different types of substrates.

The calculated condensation rate with the auto-method does depend on many parameters, which is not the case for the previous 2 methods. These parameters include the estimated contact angle for calculation of the volume and the image recording field of view (FOV) size ($\sim 10 \times 7.5mm$) and location. The location of the FOV is important considering that at the bottom of the substrate shedding is more common and thus here the condensation rate is higher than at the top of the substrate.

Another limitation is the current algorithm used to estimate the shedded volume where the negative change in volume between 2 consecutive frames is considered the shedded volume. This algorithm works well enough when volume changes are large, i.e. the shedding radius is large. For small shedding radii shedding events are more common, and consequently the volume change between frames is smaller. Individual drop growth and drops entering at the top of the FOV (partially) counteract the negative volume change resulting in a decrease in calculated shedded volume. This has shown to underestimate the condensation rate for active transport (section 4.3) for example, where the critical shedding radius is decreased by 50%.

The calculated increase in condensation rate for the ZZEs, shown in figure 4.2, is incorrect as a result of local pinning due to the electrode design. With ZZEs drops entering the FOV from the top are sometimes pinned inside the FOV. This results in a sudden increase in volume inside the FOV, and consequently an extra contribution to the shedded volume once the drop leaves the FOV many later. For substrates without this local pinning drops entering the FOV leave the FOV in a split second, not resulting in the entering drop to be counted as shedded volume. Thus the auto-method is unsuitable for more complex electrode designs such as ZZEs.

One last advantage of the auto-method is the limited amount of data needed for a good average. Generally 2-10 minutes of recorded images is enough, depending on the shedding frequency.

4.2.4 calc-method (image analysis with average calculation)

The calc-methods uses the average critical shedding radius and average shedding frequency from the image analysis to estimate the condensation rate. This method was also used by Dey *et al* [32] to show an increased condensation rate for ZZEs compared to no EW.

The calculated condensation rates using the calc-method are a factor 2–5 smaller compared to all the other methods. The condensation rates have similar values for OV and IDE types of substrates, but several times larger for ZZEs.

The calc-method is one of the simplest techniques to determine the condensation

rate, as it only requires an average shedding radius and frequency. All of which can be determined relatively easily from the recorded images, not requiring time- and resource intense image analysis like the auto-method. As it turns out, this method is oversimplified and not accurate to estimate the condensation rate. Condensation rates are extremely underestimated since the contribution of sweeping is not included at all, in contrast to the disc-, cont- and (to some extent) the auto-method. Even though drops shed at an average critical radius, many drops do not grow this big because they are removed by a shedding event. As discussed before, this decrease in the average maximum radius significantly increases the condensation rate. This method would work well for a condensation surface facing downward, where sweeping is not present.

The calc-method also overestimated the condensation rate for more complex types of electrode substrates like the ZZ configuration, as was the case with the automethod too. How to define the critical shedding radius and frequency with substrates with ZZEs is questionable due to local pinning of drops at electrical traps due to the electrode design. Drops move to the electric trap at variable critical radii (visible in figure 2.9), but shed down at a radius several factors larger. Picking a single value for the shedding radius and frequency is not possible. And in addition to that does the camera only captures a small part of the substrate, which makes it hard to distinguish a shedding drop from a drop that moves to or between electrical traps.

We conclude that the calculated increase in condensation rate using the calcmethod of condensation with ZZEs is incorrect and that in general the calc-method is inaccurate to estimate the condensation rate. The reported increased condensation rate for ZZE compared to no EW in Dey *et al* [32] is thus questionable.

4.2.5 Conclusions

In summary, the disc-, cont- and auto-method give similar condensation rates and can all be used to calculate the condensation rate. The calc-method was shown to be inaccurate and should not be used.

Between the disc-, cont- and auto-method, the weighing techniques are more accurate by nature, as these methods are a direct method to determine the condensation rate. The auto-method requires several assumptions and estimates and is influenced by the way the images are recorded. The auto-method also shows inaccurate values for more complex electrode substrate, such as ZZEs, due to its multi-phased shedding behavior.

Between the discussed methods, the most accurate one is the cont-method with intra-averages. This method excludes the heat-up phase of the chamber and normalizes the condensation rate with the temporal evolution of the subcooling. The disc-method includes the heat-up phase and uses the overall average temperature.

	Accuracy	Ease of use	Usable for complex substrates	Exact condensation rate
disc-method	+	+-	++	У
cont-method (intra-averages)	++	-	++	у
auto-method	-	+	-	у
calc-method		++		у
cont-method (inter-averages) (section 3.4.2.5)	+++		++	n

Table 4.1: Comparison of the 5 investigated methods to determine the condensationrate. Scale from best to worst: ++ / + / +- / - / --. y=yes, n=no.

Table 4.1 shows a summary of all techniques with pros and cons. It also includes the cont-method with inter-averages, used in section 4.3 and 4.4.

None of the methods however is accurate enough to show differences between substrates. This is mainly a limitation of the experimental setup. Experimental conditions, such as subcooling and flow-rate between and even during experiments are not constant. The errors caused by these fluctuations exceed the actual differences in condensation rates between the different substrates.

It can be concluded from these results is that none of the substrates increase the condensation rate by a factor of 2 or more. This motivated us to find a more accurate collection method (cont-method with inter-averages, as discussed before in section 4.2.2) and an even more efficient substrate type, which turned out to be active transport.

4.3 ACTIVE TRANSPORT

This section gives an overview of this new technique. First the workings of active transport will be explained in section 4.3.1. Here the different phases of the active transport condensation process are described using the breath figures. Section 4.3.2 is focussed on the efficiency of active transport: does active transport improve heat transfer?

All active transport substrates were in contrast to the other substrates Teflon coated (see section 3.2.2).

4.3.1 Workings of active transport

DWC is a repetitive process of different phases, as explained in section 2.1.1. At any moment in time different phases can be present simultaneously on the surface. These phases change with EW and become more complicated with sophisticated electrode designs. Without EW, the 3 phases can be identified: nucleation, coalescence and shedding.

Introducing EW in its simplest form using IDEs result in an extra phase of horizontal gap alignment (figure 2.5) that enhanced coalescence. Introducing another dimension in the electrodes is achieved using ZZEs, and an extra phase of vertical alignment emerges (figure 2.10) at the electrical traps.

Finally we introduce a third dimension by stacking two layers of ZZEs in a double layered ZZEs configuration. The condensation process is shown in the image series in figure 4.3. The overlay in the breath figures show the location of the top layer of electrodes in grey, and the bottom layer in red. At any time only one layer of electrodes is active, switching back and forth every 2 seconds. The drop of interest is encircled with a red dashed line. Condensation on a clean surface shows at first similar behavior to single ZZEs condensation and thus similar condensation phases can be identified. We identify the phases nucleation (figure 4.3a-4.3b), coalescence (figure 4.3b-4.3c), horizontal gap alignment (figure 4.3b-4.3d) and vertical alignment when the drops move to the smallest gap (figure 4.3d). All similar to single layered ZZEs.

The difference between single and double layered ZZEs comes from the active part of the actuation for the latter one. With a drop trapped at an electric trap, the layer causing the trapping is turned off and simultaneously the other layer of electrodes is turned on. For this active layer the drop is halfway down and hence the drop is not at an electric trap anymore but experiencing a downward electric force pulling it away from the location where it was trapped before. But since drops move at finite speeds, it might not be able to clear the passive electric trap before it is turned on again and be pulled up back on the trap.

The electrical force (equation 2.18) and weight (equation 2.5) pull down on the moving drop, while the pinning force (equation 2.3.1) and dynamic friction force (equation 2.20) counteract this movement. The sliding velocity that follows from this force balance is $v \sim R^2$. Full derivation of the force balance is done when introducing the model for active transport in section 4.5.1. Thus if a drop is big enough, it gains enough speed to clear the electric trap.

But if a drop is too small, it will not gain enough speed to clear the electric trap in the time it is off, and when the actuation switches, the drop is pulled back up and centered again on the electric trap. This 'sliding down – pulled back up'-behavior is referred to as drop gap-oscillation (figure 4.3e-4.3g) and can occur for 30 seconds or longer.



Figure 4.3: The different stages in DWC with active transport. The overlay shows the top electrodes (gray) and bottom electrodes (red). (a) Clean surface with nucleation. (b) Coalescence of droplets with alignment on bottom electrode edge. (c)-(d) Coalescence and transport of droplet to smallest gap between electrodes (pinning points). (e)-(f) Drop gap-oscillations. (g)-(j) Active transport off the electrode surface.

An example of drop gap-oscillation is shown in the image series in figure 4.4. Each image shows an overlay with the active electrodes in red and the passive electrodes in black, note that these thus change from image to image. At t = 0 a drop moves in from the top being pulled down by the active ZZEs. At t = 9s the electrodes switched, turning off the electrical trap that would have trapped the drop is at t = 0s and t = 3s. However, the drop does not gain enough speed to clear the gap due to the dynamic friction forces and is pulled back up when the sample actuation switches between top and bottom electrodes, visible at t = 15s. This process repeats several times (t = 15 - 33s). Every oscillation the drop gains more mass allowing it to move down further than before, as indicated with the yellow arrows and distances. With the last oscillation at t = 33s the drop is large enough, consequently gains enough speed and clears the gap.

Once a drop clears one electric trap, it is able to clear all electric traps below resulting in active transport of the drop from electric trap to electric trap in a conveyer-like behavior (figure 4.3h-4.3j).

Drop gap-oscillations start as soon as the drop is at the electric trap, at radius $R_o = R_t$. The oscillation amplitude increases with each oscillation up to a maximum of $R_{crit,active}$; when the drop is actively shedded down.



Figure 4.4: Image series of the drop gap-oscillation during active transport. The red and grey overlay show the location of the electrodes in the substrate. Red indicates the active electrode, gray the passive electrode. The yellow arrow indicates the distance it needs to travel to clear the electrical trap. The drop moves to the electrical trap at t=0, is trapped and oscillates between t=9s and t=33s and shedded down at t=36s.

The drop gap-oscillation around the electric trap and active transport off the surface are unique to the double ZZE design. Compared to single layered ZZEs, drops do not only move to the electric traps, but also start oscillating at these traps. This introduces a new phase of (drop) gap-oscillations, shown in the updated phase diagram of figure 4.5.

The main difference between double and single layered ZZEs is the net direction of the electric force. Where the electric force with single layer ZZEs is creating permanent electric traps on the surface where the electric force is always pointing upward, with double layered ZZEs these electric traps are only temporarily. When the actuation switches between the two electrode layers, the electric traps change location on the surface, allowing drops to be pulled down by the active electrode layer away from the traps. In this way the critical shedding radius is decreased compared to no EW.



Figure 4.5: Condensation phases with EW on double layered ZZEs (active transport) characterized by the drop radius.

4.3.2 Efficiency of active transport

To quantify the efficiency of active transport condensation we look at several clues that indicate an increased heat transfer such as surface coverage and critical shedding radius, described in section 2.1.2, as well as direct measures of the volumetric condensation rate $\langle v \rangle$.

Figure 4.6 shows breath figures of the condensation process with no EW (4.6a), EW with single layered ZZEs ($d = 3000\mu m$) (4.6b) and active transport using double layered ZZEs (4.6c). There is a clear difference visible between the breath figures. First of all, the surface coverage is different. Both the reference and the ZZEs show a higher surface coverage compared to active transport.

Figure 4.7b shows the average surface coverage for active transport, reference, pulsed EW (on ZZEs $d = 3000 \mu m$ electrodes) and EW with ZZEs. For each configuration the surface coverages is an average from over 25 hours of condensation. We observe a reduction in the surface coverage for active transport attenuated surfaces of -22% from 52.1% without EW to 30.9% with active transport. This reduction is a clear indication of increased heat transfer as explained in section 2.1.2.1. Fewer drops on the surface means a decrease in the total thermal resistance and more surface area available for nucleation of new drops. Baratian *et al* [31] reported a decreased surface coverage for condensation with IDE electrodes of -10-12% and a corresponding heat transfer increase of +50-60%.

The small decrease in surface coverage for ZZEs and pulsed EW can be attributed to the enhanced drop coalescence in the early condensation stages.



Figure 4.6: Breath figures of DWC without EW (a), EW with single layered ZZEs (b) and EW with double layered ZZEs (active transport) (c). All on Teflon coated substrates.

Another clear indication of increased heat transfer is the decrease in critical shedding radius for active transport. The critical shedding radius is shown in figure 4.7a for the 4 configurations, each datapoint an average from over 50 shedding events. For active transport we see a decrease of -51% from an average of 542 μm for the reference to 275 μm for active transport condensation. At the same time we measured an increased shedding frequency of +700% from 1.4/min to 9.8/min on average. With active transport it took on average 103 seconds for a drop to grow from nucleation size to the critical radii, while for the no-EW configuration it took 450 seconds on average.

This increased shedding frequency with the decrease in critical shedding radius is a clear sign of increased heat transfer, as described in sections 2.1.2.2 and 2.1.2.3.



Figure 4.7: Critical shedding radius (a) and surface coverage (b) for 4 types of (functionalized) substrates.

To confirm these indirect measures of increased heat transfer, we performed water collection experiments with continuous water collection with inter-averages (section 3.4.2.5) to calculate the improvement in condensation rate. The condensation

rate is a direct measure of heat transfer (equation 2.10).

Figure 4.8 shows the continuous water collection results of an active transport experiment where the mass normalized by the subcooling is shown versus time. After preheating the condensation chamber (Time=0), condensate was collected for roughly \sim 35 minutes without EW before active transport was initiated. After \sim 30 minutes one of ZZE layers was turned off, creating a single layered ZZE configuration. Again after \sim 30 minutes EW was turned off completely for a final \sim 35-minute period as a reference. After linear fitting and comparison with the direct references the improvements in heat transfer for the active transport and ZZ configurations were determined as explained in section 4.8. The fitted lines are shown in orange, with the dash-dot lines indicating the references and the average condensation rates for each period is indicated with the mg/min/K number.

The percentile increase in condensation rate for active transport was found to be +11.5% using this method. Active transport thus increases the heat transfer. On the other hand, we found a decrease in heat transfer of -20.1% using single layered ZZEs. This will be discussed in the next section.

Due to experimental issues only one dataset with active transport was used to determine this percentage increase. Extra experiments have to be performed to support this exact number, as will be discussed in the outlook.

Experimentals fluctuations are clearly visible in this dataset when comparing the condensation rates of the references an hour apart. These fluctuations make it hard to compare almost identical condensation rates, as discussed in the stability performance of the experimental setup in section 3.1.3. Using the cont-method with inter-averages and normalization with the subcooling, we manage to reduce this error. The percentages of improvement calculated in a similar matter for more types of substrates are presented in the next section.



Figure 4.8: Continuous water collection on Teflon coated substrates with interaverages. The mass *M* normalized with the subcooling (ΔT) is shown versus time. 35 minutes of reference (no EW) condensation is followed by a 30-minute period with active transport, 30-minute period with single layered ZZEs and finally 35-minute reference period. All periods are individually linearly fitted (orange line). Reference periods (dot-dash lines) are used to calculate the percentage improvement.

4.4 CONTINUOUS WATER COLLECTION WITH INTER-AVERAGES

Different techniques to determine the condensation rate were investigated in section 4.2. None of the techniques to determine the condensation rate exactly were accurate enough to show any differences in condensation rates between different substrates with the current experimental setup. The main reason for this limitation are the fluctuations in experimental conditions between experiments and within long experiments that introduce large errors. Therefore we developed a new technique where instead of determining the actual condensation rate we look at changes in the condensation rate within an experiment, where we use the no-EW case as reference. See section 3.4.2.5 for full description of this technique.

We performed condensation experiments where we switched between reference and EW every 30-45 minutes without stopping the experiment. These experiments were performed for 6 different types of substrates: ZZEs with lengths $d = 1500 \mu m$ and $d = 3000 \mu m$ with continuous EW, ZZEs with length $d = 3000 \mu m$ with pulsed EW (short pulse every 2 seconds), IDEs with 50-50 μm gap-electrode widths, IDEs 200-200 and double layered ZZEs with active transport actuation.

By having a direct no EW reference before or after the period with EW we were able to eliminate experimental fluctuations between experiments and partially within experiments. Each experiment gives multiple condensation rate improvements
$(\langle v \rangle / \langle v \rangle_{\rm no \; EW})$ which were averaged to get the average improvement of that substrate type.

Figure 4.9 shows the condensation rate improvements for all types of substrates. Note that the number of data points per configuration is limited, and thus actual values might differ slightly.



Figure 4.9: The average percentage improvement of 6 types of functionalized substrates compared to the reference (no EW). Calculated using continuous water collection with inter-averages.

Both ZZ configurations show a decreased condensation rate of -12% and -6.6% for ZZEs with lengths $d = 1500\mu m$ and $d = 3000\mu m$, respectively, compared to the reference. The design of the ZZEs creates electrical pinning points on the surface where drop radii exceed the shedding radius without EW. This increase in critical radius results in a higher thermal resistance and consequently decreased heat transfer. The electrode gradient for $d = 1500\mu m$ electrodes is sharper compared to d = 3000 electrodes. This sharper gradient results in bigger electrical pinning forces, bigger shedding radii and thus lower heat transfer. This conclusion is contradictory to the conclusions of Dey et al [32], where an increased condensation rate of $\sim +400\%$ was calculated. But, concluded earlier in section 4.2, the method used to estimate the condensation rate by Dey et al is inaccurate and does output an incorrect increase in the condensation rate due to the complex shedding behavior of ZZEs.

When instead of continuous EW we pulse $d = 3000 \mu m$ ZZEs, we observe a slight improvement of the condensation rate of +1.7%. The electrical traps are temporarily turned off allowing drops to shed down by gravity at smaller radii, while still benefiting from the increased coalescence of the ZZEs design. The electrodes were turned on for a short pulse every 2 seconds, which was an arbitrary pattern. One might expect further improvement by optimizing this pulse pattern. IDEs show an increased condensation of +3.8% and +6.2% for $200 - 200\mu m$ electrode-gap and 50-50 electrodes, respectively. The enhanced coalescence with IDEs in combination with decreased hysteresis improves the condensation rate. Our calculated improvements are not as substantial as the reported values by Baratian *et al* [31, 62], where using heat transfer measurements improvements of +50 - 60% were observed. We assign this difference to the absence of non-condensable gases (NCG) in the experiments of Baratian *et al* which are present in our experiments. It has been found that the presence of NCG can result in a significant reduction in heat transfer. [84, 85] Minkowyez and Sparrow [86] even found that as little as a half percent of air in steam can reduce the heat transfer by -50%.

Small amounts of NCG in the vapor can cause a large buildup of NCG at condensing interface. As a consequence the partial pressure of the vapor at the interface is reduced, lowering the temperature at which the vapor condenses. [87] But the reason the efficiency of IDEs decreases in the presence of NCG could lie in the reduced efficiency of the system that drives the improved heat transfer of IDEs in the first place, i.e. the reduced surface coverage. Preston *et al* [37] reported that without NCG the heat transfer with DWC is +500 - 700% higher than FWC, while with NCG this improvement is reduced to only +30 - 40% improvement. It thus seems when there is more bare surface available for condensation, i.e. the lower the surface coverage, the more the heat transfer is reduced by the presence of NCG. And since IDEs reduce the surface coverage by enhanced coalescence and reduced shedding radii, the reduction in heat transfer because of NCG is bigger for IDEs.

Using active transport with double layered ZZEs we do see an improved condensation rate of +11.5%, as mentioned before in section 4.3.2. This improvement over the other configurations is a result of the unique electrode design and actuation pattern that combines the increased coalescence behavior of ZZEs with the addition of active transport away from the electrical traps. This conveyor-like transport of condensate drops has not been reported in literature before. The system can be further optimized with better electrode design, which is done using a force balance model in the next section.

4.5 NUMERICAL MODEL FOR ACTIVE TRANSPORT

The increased condensation rate of +11.5% for active transport condensation is achieved using essentially an arbitrary electrode design and actuation pattern. Many parameters can be tuned to improve the efficiency of active transport, including electrode width, height and spacing, as well as actuation time and EW voltage. To optimize these parameters we developed a simple numerical force balance model that allowed us to check thousands of combinations of parameters and see the effect on the critical shedding radius and thus the efficiency of the system.

This model is conceptually simple in the sense that it uses a simple force balance to predict the critical shedding radius, but becomes mathematically complicated due to the complex geometry of the zigzag electrode design. This section presents the model starting with the development of the model in sections 4.5.1 to 4.5.3. In section 4.5.4 we use this model to find the *generic op-timization equation*, a single equation that gives the most optimal combination of electrode dimensions.

The model is written in MATLAB. The full code can be found in appendix B.

4.5.1 Force balance

Reducing the critical shedding radius R_{crit} will result in increased heat transfer (section 2.1.2.2). For simple systems without EW or with EW using IDEs drops shed at R_{crit} anywhere on the surface once the pinning forces are overcome by gravity. For ZZEs drops always accumulate at the electric traps, i.e. at the smallest distance between electrodes, before shedding down, regardless of their initial location. $R_{crit,ZZ}$ is thus always achieved at these specific locations. With active transport the situation is similar as drops are transported to the electric

with active transport the situation is similar as drops are transported to the electric traps. But now the electric traps are interchanged periodically for electric forces pulling the drop down using the second layer of electrodes that is vertically offset by half a period. As explained in the workings of active transport in section 4.3.1, a drop has to clear the passive electric trap before it is made active again to actively shed down the surface. By solving the force balance of a drop at one of these passive electric trap locations we are able to estimate the smallest drop radius R_{crit}^{1} that is able to do this.

Figure 4.10 shows the situation sketch used for the model. A droplet of size R is located halfway down the active (green colored) ZZEs. The area between two alternating electrodes is referred to as the gap and has the same dimensions as the electrodes. The repeating ZZEs have a height of d, and a distance of 2b between them at the smallest point, known as the gap. The horizontal protruding part of each electrode has width a, making the total width of each electrode 2b + 2a.

We assume 4 forces are acting on the drop: weight F_g (equation 2.5), pinning force due to contact angle hysteresis F_p (equation 2.3.1), electrical force F_e (equation 2.18) and the dynamic friction F_d (equation 2.20) when the drop is sliding. These forces are indicated with red arrows in figure 4.10.

The dynamic friction force F_d is a linear function of the drop sliding velocity. For a drop to clear the electric trap is needs to have have a velocity large enough to cover a distance R_{crit} in Δt , the velocity that follows from this called the critical velocity v_{crit} :

$$v_{crit} = \frac{R_{crit}}{\Delta t} \tag{4.1}$$

We thus set this velocity as a boundary condition. For velocities smaller than v_{crit} , the dissipation force will be smaller, but the drop will never clear the electric trap

¹ For notational convenience we write $R_{crit,active}$ as R_{crit} in this chapter from here on.



Figure 4.10: Situation sketch with electrode parameters and forces acting op the drop.

and active transport will not be initiated.

The dynamic friction force that follows from the critical shedding velocity is the maximum dynamic friction force the drop will experience, and is only reached at the moment of shedding when $R = R_{crit}$.

Rewriting equation 2.20 using the critical velocity from equation 4.1 gives the expression for the maximum dynamic friction force:

$$F_d^{max} = F_d(v = v_{crit}) = \frac{R_{crit}^2}{\Delta t} (6\pi\mu\rho + 8\xi)$$
(4.2)

The force balance acting on the drop in the middle of an electrode at the moment of active transport initiation ($R = R_{crit}$) in vertical direction, can thus be written as

$$F_{e\downarrow} + F_g + F_p = -F_d^{max} \quad \text{for } R = R_{crit}$$
(4.3)

where $F_{e\downarrow}$, F_g , F_p and F_d^{max} are defined in equations 2.18, 2.5, 2.6 and 4.2, respectively. Here the condition $R = R_{crit}$ is a direct result from the dynamic friction force that is evaluated for the maximum velocity the drop reaches in time Δt , which is sufficient to determine the critical shedding radius.

Evaluation of Δl

Solving the force balance of equation 4.3 requires an exact expression of $F_{e\downarrow}$ and thus evaluation of Δl , the horizontal distance between the intersects of the electrode edge with the drop. Δl , is calculated analytically in the model by solving the intersection of a circle and line.

The electrode edges are defined as two linear lines mirrored around x = 0. Since we assume the drop to be horizontally centered on the electrodes, the problem is

symmetric and we thus solve the problem for one electrode edge.

The electrode edge is defined as a line with slope -d/a. The electrodes are spaced by 2b at y = -d/2 and 2b + 2a at y = d/2. We define the angle between x = -band the inside of the electrode as $\alpha = \tan^{-1}(d/a)$. This results in an intersection of the line with x = 0 at $y = -d/2 - b/\tan(\alpha) = -d/2 - (bd)/a$. Thus the line describing the edge of the electrode is

$$y_{line} = -\frac{d}{a}x - \frac{d}{2} - \frac{b}{a}, \quad -(a+b) \le x \le -b, \\ a, b, d > 0$$
(4.4)

The drop is defined as a circle centered on (x, y) = (0, 0):

$$x^{2} + y_{circle}^{2} = R^{2} \Rightarrow y_{circle} = \pm \sqrt{R^{2} - x^{2}}, \quad x^{2} < R^{2}$$
 (4.5)

Solving the equality $y_{line} = y_{circle}$ for x using equations 4.4 and 4.5, we find

$$x_{1,2} = -\frac{(ad^2 + 2bd^2 \pm a(4R^2a^2 + 4R^2d^2 - a^2d^2 - 4abd^2 - 4b^2d^2)^{1/2}}{2(a^2 + d^2)}, \quad R > \frac{a}{2} + b$$
(4.6)

as the horizontal intersect points between the electrode edge and drop. If the drop is too small to overlap the electrodes, Δl is zero. As a boundary condition we thus set $\Delta l = 0$ for $R \leq \frac{a}{2} + b$, giving the final expression for Δl :

$$\Delta l = |x_1 - x_2|$$

$$= \begin{cases} \left| -4R2a^2 - 4R^2d^2 + a^2d^2 + 4abd^2 + 4b^2d^2 \right|^{1/2} \times \left| \frac{a}{a^2 + d^2} \right| & \text{if } R > \frac{a}{2} + b \\ 0 & \text{if } R \le \frac{a}{2} + b \end{cases}$$

$$, \ a^2 + d^2 \neq 0.$$
(4.7)

4.5.2 Model assumptions

For model simplicity, we implemented several assumptions. These assumptions include the assumptions made in the definitions of the forces, and the assumptions below.

Assumption 1: single electrode gap

The model assumes a drop only overlaps a single gap between two infinitely wide electrodes. In reality once a drop exceeds the width of the overlapping electrodes, i.e. when it touches 2 more gaps, a second electrical force is induced that pulls the drop down, as visible in figure 4.4. Setting a model boundary condition for the maximum drop size not to exceed the width of one set of electrodes (R < 3(a/2+b)) would result in very limited model results, and is therefore not implemented. In some situations the model will thus underestimate the critical drop radius.

For the vertical direction the drop is not allowed to exceed the electrode size, since

this would result in strong electrical trapping forces. Thus, R < d/2. As it also turns out, in the most efficient double layered ZZEs designs, the height of the electrode exceeds the width of the electrode by a factor of 5. It is thus less likely for the model to be limited by this boundary condition.

Assumption 1:

$$R_{crit} = \begin{cases} NaN & \text{if } R < d/2 \\ NaN & \text{if } R < 3(a/2+b) \\ R & \text{if otherwise} \end{cases}$$
(4.8)

Assumption 2: centered drop and static forces

The drop is assumed to be centered between two electrodes at all times, and thus the forces acting on the drop are constant at all times. As a consequence drop acceleration is zero and the drop sliding velocity is constant.

When a drop moves down, the electrical force is assumed to be constant and equal to the value at the center the gap: $F_{e\downarrow}(t) = \text{constant}$. In reality as the drop slides down the electrical force decreases as the horizontal projection of the overlap Δl decreases.

Assumption 2:

$$\sum F(t) = ext{constant}$$
 (4.9)

Assumption 3: constant pinning force

Both electrodes are assumed to be in the same plane, and attenuated with the same potential Urms. In reality the two layers of electrodes are separated by a dielectric layer, and to compensate for this, the deeper electrodes are attenuated with a higher potential.

Assumption 3:

$$U_{rms}(t) = \text{constant} \land \Delta \cos \theta_0(t) = \text{constant}$$

 $\Rightarrow f_p(t) = \text{constant}$
(4.10)

4.5.3 Model validation

Validation of the model was done by checking the implementation of the individual forces and by studying the model behavior on changing one parameter once at a time. Validation as such is a first step towards full validation of the model, which would require experimental verification. All the model parameters and constants are shown in table 4.2. The values are referred to as the standard values and match the experimental values as close as possible. Unless otherwise specified the results shown used these values.

4.5.3.1 Force balance

Using the separate forces from equations 2.5, 2.6, 2.18, and 2.20, the model dependency on the drop radius was studied.

Constan	t Value	Dimensi	ioMeaning
a	100×10^{-6}	т	Horizontal projection of electrode edge.
			See figure 4.10.
b	$25 imes 10^{-6}$	т	Half the minimum width of the gap between
			electrodes. See figure 4.10.
d	$1500 imes 10^{-6}$	т	Vertical projection of electrode edge.
			See figure 4.10.
U	150	V	EW voltage.
t	2	S	on/off time of the top and bottom electrode.
$\Delta \cos \theta_0$	0.206	1	Difference between the cosines of the receding and advancing contact angles for $U_{rms} = 0V$.
θ	115	deg	Advancing contact angle of drop on surface.
α	1	1	A constant coefficient characterizing the effi- ciency of the ac-EW induced CAH reduction mechanism.
β	$8.5 imes 10^{-6}$	$F/(N \cdot m)$	Ratio if the effective dielectric capacitance per unit area and γ . [32]
ho	997	kg/m^3	Density of water.
8	-9.81	m/s^2	Gravitational acceleration.
γ	72.8×10^{-3}	N/m	Surface tension of water-air interface.
μ	1×10^{-3}	$Pa \cdot s$	Viscosity of water drop.
ξ	$71 imes 10^{-3}$	$Pa \cdot s$	Contact line friction coefficient. [68, 69]

 Table 4.2: Default model parameters.

Figure 4.11a shows the individual forces acting on the drop as a function of the drop radius.

The pinning force, shown in red, increases linearly with the radius. This linear behavior is what we expect based on equation 2.6: $F_p \sim R$. The gravitational force, shown in blue, for small drop radii is almost negligible. For larger radii it increases fast, confirming the $-R^3$ behavior of the drop volume and thus $F_g \sim -R^3$.

The maximum dynamic friction force is shown as a purple dotted line, as this force is only acting on the drop when it is in motion at the critical velocity v_{crit} . This velocity, and thus the dissipation force plotted is only acting on the drop at a radius equal to the critical radius. The dissipation force increases quadratically as we expect from equation 2.20: $F_d \sim R^2$. The yellow line shows the dissipation force when the drop is static: $F_d = 0$.

The behavior of the electrical force, shown in yellow, due to the asymmetrical shape of the electrodes is more complicated. At small drop radii, the drop is not touching the electrodes, thus $F_e = 0$ for R < a/2 + b. When the drop increases in size and touches the electrodes, the total overlap determines the size of the force. This overlap is characterized by Δl (equation 4.7). Δl scales as $-\sqrt{R^2 + C_1R}$, with $C_1[m]$

a constant arising from the electrode dimensions a, b and d, which for small R is approximately to $-\sqrt{C_1R}$ (series expansion around R = 0). Thus, for the electric force we find $f_e \sim \sqrt{R}$ for small R. This square-root behavior is visible in figure 4.11a as the sudden decrease when the drop touches the electrodes.

Figure 4.11b shows the force balance from equation 4.3. The sum of the electrical, gravitational and pinning force is shown in blue. The maximum dynamic friction force is shown as a red dotted line and is only valid for a drop of radius R_{crit} . Solving the force balance for R gives R_{crit} , shown with a red cross. The radius at which the drop is trapped and start oscillating (R_o) is found by solving the force balance for $f_e = 0$. R_o is shown as a yellow cross. Between R_o and R_{crit} the drop starts moving down but is unable to achieve the critical velocity v_{crit} in Δt and is thus pulled back up once the actuation switches.

The sum of the electrical, gravitational and pinning force in blue shows linear behavior for R < a/2 + b. This is when the drop is not touching the electrodes $(f_e = 0)$ and the drop size is small thus the weight is negligible $(f_g \approx 0)$. The dominant force here is thus the pinning force, which scales linearly with R. As soon as the drop touches the electrodes, the electric force increases fast as the overlap of the drop with the electrodes increases. If the drop is at its critical velocity, the quadratic growing dissipation force also contributes. Thus the dominant force on the static drop that overlaps the electrodes is the electric force $(f_e \sim \sqrt{R})$, and thus the sum of the forces decreases with a root. For large R the weight of the drop is the dominant force $(f_g \sim -R^3)$, and thus the sum of the forces scales as the power of 3 for large R.

We conclude that the implementation of the forces is correct, as the forces scale as expected.



Figure 4.11: Forces acting on a drop versus drop radius calculated using the model for a fixed lateral position of the drop. (a) Shows the individual forces, (b) shows the force balance from equation 4.3.

4.5.3.2 Model behavior on specific parameters

The model is written in the first place to optimize the electrode design for more efficient active transport condensation, which is achieved by reducing the critical shedding radius. Investigating the dependency of the critical radius on these parameters helps us understand the model and serves as a first step towards active transport optimization.

Figure 4.12 shows the change in critical shedding radius as a function of each of the 8 model parameters from table 4.2. One parameter is varied at once, the others are kep at their default value. For each parameter we calculated the critical radius by running the model.

The dependence on the d (vertical electrode length) is shown in figure 4.12c. With all other parameters kept constant we observe an increase in the critical radius with increasing d. For small d the electrode gradient (angle between gravity direction and electrodes) is large, and thus the electric force is large. With increas-

ing length, this angle decreases, and for $R \to \infty$ the electrodes run parallel and the electric force is zero, as visible with the line approaching the $F_e = 0$ -case.

The dependence of R_{crit} as a function of the minimal gap width (2*b*) between the electrodes is shown in figure 4.12b. For b = 0, the critical shedding radius is smallest, for large *b*, it approaches the $f_e = 0$ -case, where the drop sheds down by its weight before it even touches the electrodes. Changing *b* does not change the gradient of the electrodes, and thus the smaller *b*, the better. The practical reason *b* is finite is to connect the electrodes with each other.

The dependence on the electrode width (~a) in figure 4.12a is more complicated. For $a \rightarrow 0$ the electrodes run parallel and $f_e = 0$. With increasing *a* the gradient increases and thus the critical radius decreases rapidly. But there is a minimum! As *a* increases, the distance between the electrodes also increases, and thus the drop size at which the electric force starts acting on the drop increases as well (R > a/2 + b). This delicate balance between large electric force and an early electric force results in an optimal value for *a*. This optimum *a* does of course depend on the other electrode parameters *d* and *b*, and thus evaluation of all three of these parameters is needed to find the best, or a good combination of these parameters. This multi-parameter optimization is done in section 4.5.4.

The critical radius as a function of EW voltage is visualized in figure 4.12e. For U = 0, the electric force is always zero. For large R both the $f_e = 0$ -case and $f_e > 0$ -case decreases. The $f_e = 0$ case decreases since the pinning force depends on U_{rms}^2 (equation 2.6). At a certain voltage f_p turns negative according to equation 2.6. This is physically not possible, and we have to keep in mind that equation 2.6 is an approximation that hold for well for small voltages, but breaks down at large voltages (see section 3.2.3). [68]

For the same reason mentioned above, we get non-physical results for small $\Delta \cos \theta_0$. For large $\Delta \cos \theta_0$ the pinning force increases linearly, as predicted by equation 2.6.

For the dependence of the critical radius on t in figure 4.12f, we see a rapid decrease with actuation time. For short actuation times the drop has very limited time to clear the gap and thus needs a high velocity. This high velocity is only obtained with large drop sizes, thus a large R_{crit} . However, longer actuation times will reduce the shedding frequency, something the model does not include.

Figure 4.12g shows the dependence on the the drop viscosity. Since $F_d \sim \mu$ we see linear dependence. Small viscosity fluids (such as methanol) have a smaller dissipation force compared to high viscosity fluids (like viscous oils).

Finally, in figure 4.12h shows the dependence on the contact line friction coefficient ξ . The used value from literature from 't Mannetje *et al* [68] of $\xi \approx 71Pa \ s$ has a fitting error of $\pm 10Pa \ s$. The full error range is plotted, and only a (linear) deviation of $\Delta R_{crit} < 1\mu m$ is observed. The exact value of ξ is thus irrelevant in this model.

In conclusion the model shows the physical behavior we expect. For large EW voltages U and/or small contact angle hysteresis $\Delta \cos \theta_0$ the approximate form of the pinning force introduced in equation 2.6 does not hold anymore. However, this is only a first step into full validation of the model. Proper validation will require experimental verification, what has not been done for this thesis.



Figure 4.12: The dependence of the critical shedding radius R_{crit} on 8 model parameters (see table 4.2).

4.5.4 Electrode optimization

In section 4.5.3 we found that for most active transport parameters going in one direction will result in a smaller critical shedding radius and thus increased heat transfer. For example by decreasing the hysteresis ($\Delta \cos \theta_0$) or increasing the EW voltage (*U*). Many of these parameters are hard to control in reality. The maximum EW voltage is limited by the breakdown of the dielectric layer and the hysteresis depends on the coatings being used.

The major optimization parameters are the electrode dimensions d, a and b. These parameters are designable and show a more complex relationship where smaller dimensions do not always result in smaller shedding radii, as was shown by changing a in figure 4.12a. Smaller feature sizes are also not always possible by manufacturing limitations. Finding a good ratio between these parameters is thus an important step towards more efficient active transport systems.

In order to find a more optimal set of electrode dimensions that will decrease the critical shedding radius, we ran the model for model for 500,000 combinations of these parameters. For *b* we choose 5, 10, 25 and 50 μm . Each value of *b* was ran for 500 values of *d* between 10-3000 μm (~ $6\mu m$ resolution) and each set of *b* – *d* for 250 values of *a* between 10-1500 μm (~ $6\mu m$ resolution). In each iteration the critical shedding radius was calculated by solving the force balance numerically.

The results are shown in the interpolated contour plot in figure 4.13 for the two extreme values of b: $b=5 \ \mu m$ and $b=25 \ \mu m$, ignore the red line for now. The red cross indicates the current electrode design used.

Due to the single-electrode boundary conditions for only about 60% of the combinations a solution was found.

We observe in figure 4.13 that smaller *a* does not always result in a smaller R_{crit} . This observation agrees with the result from figure 4.12a where the minimum R_{crit} was found for an *a* larger than the smallest *a*. Decreasing *d* however, always results in a smaller shedding radius, but not always at the same rate. For larger values of *a*, the rate of decrease in R_{crit} is smaller. From a manufacturing standpoint this is an important result. Even though smaller electrodes will result in smaller R_{crit} , the percentile decrease in R_{crit} is much smaller than the percentile decrease of the feature size. Larger electrodes are easier to manufacture and electrically more reliable, and with a relatively small increases in R_{crit} the efficiency of the system increases only slightly. There is a large margin around the optimal value for which the design will work almost as well.

Finally we observe that for smaller values of b, R_{crit} is also smaller for the same values of a and d. This result corresponds with the observations in figure 4.12b, where smaller b will always reduce R_{crit} . For larger values a and d, b becomes negligible and results are very similar. b does not change the gradient of the electrodes, and is introduced for the practical reason of connecting electrodes in-line. b has to be chosen as small as possible.



Figure 4.13: Model bruteforcing results of 500,000 possible combinations of *a*, *b* and d for two (a) $b = 5\mu m$ and (b) $b = 25\mu m$. The red line shows the estimation of the generic optimization equation. The red cross shows the current electrode design ($b = 25\mu m$, $a = 100\mu m$, $d = 1500\mu m$).

(1)

Generic optimization equation 4.5.4.1

Using the brute-force results, we approximated a single equation that gives the best d for a given a and b, an equation referred to as the generic optimization equation. This equation can be used for designing more efficient double layered ZZEs, where the input is limited by the smallest possible feature size.

Deriving the generic optimization equation from the brute-force results was done by finding a where R_{crit} is minimal for each d and b. The resulting data points were fitted with the simplest polynomial possible. A second-order polynomial in this case of the form:

$$a = p_1(b) \cdot d^2 + p_2(b) \cdot d + p_3(b)$$
(4.11)

where p_1 , p_2 and p_3 are the fitting coefficients. The raw data and fits are shown in figure 4.14.

Figure 4.14: Fitting of the R_{crit} -minima from the model for 4 values of b using equation 4.11.

Each *b* gives a unique set of fitting coefficients p_1 , p_2 and p_3 . Each fitting parameter only depends on b, and we thus fitted a polynomial to all the coefficient to find the dependence on b. The most, but proper fits were obtained for linear fits for p_1 and p_2 and a second order polynomial fit to p_3 :

$$p_1(b) = p_{1a} \cdot b + p_{1b} \tag{4.12a}$$

$$p_2(b) = p_{2a} \cdot b + p_{2b} \tag{4.12b}$$

$$p_3(b) = p_{3a} \cdot b^2 + p_{3b} \cdot b + p_{3c} \tag{4.12c}$$

Here $p_{1a,1b}$, $p_{2a,2b}$, $p_{3a,3b,3c}$ are fitting coefficients. The fits are shown in figure 4.15.

Combining equations 4.11 and 4.12, we find a generic optimization equation for *a* as function of *d* and *b*:

$$a = (p_{1a} \cdot b + p_{1b}) \cdot d^2 + (p_{2a} \cdot b + p_{2b}) \cdot d + (p_{3a} \cdot b^2 + p_{3b} \cdot b + p_{3c}) \quad (4.13)$$

Figure 4.15: Fitting of the fitting parameters p_1 , p_2 and p_3 from equation 4.11 using equation 4.12.

Using the fitting parameters from the brute-force model, simplifying and rounding the fitting parameters up to 3 significant numbers, we find the generic optimization equation:

$$a = 0.14b + d(655b + 0.058) - 1.0 \times 10^3 b^2 - d^2(1.7 \times 10^5 b - 8.4) + 1.5 \times 10^{-6}$$
(4.14)

where the units of *a*,*b* and *d* are in meters.

For a given *b* and *d* this equation gives an *a* that corresponds to the smallest R_{crit} for that *b* and *d*. To validate this equation, it is plotted with the contour plot in figure 4.13 for $b = 5\mu m$ and $b = 25\mu m$. It indeed follows the minimal values.

Solving equation 4.14 for d, gives the final expression of the generic optimization equation:

This generic optimization equation can be used to improve future electrode design for active transport by providing a matching a for a given value of b and d. The limitation of electrode design is the smallest possible feature size. The smaller the feature size, the smaller the critical shedding radius, and thus the more efficient the condensation process. By selecting a minimum feature size and setting this value as a and b, one can find corresponding electrode height d using equation 4.15 to have the smallest critical shedding radius possible.

As an example, with the electrode dimensions used for this research: $d = 1500\mu m$, $a = 100\mu m$ and $b = 25\mu m$ (table 4.2), we find $R_{crit} \approx 135\mu m$ using the model. This value is lower than the experimental value of $275\mu m$, reported in section 4.3.2, but as noted before, several experimental parameters such as the hysteresis are not well defined and small fluctuations result in large fluctuations in R_{crit} .

Using the model, we estimate that by setting $a = 125\mu m$ and leaving the other parameters unchanged, R_{crit} reduces to $115\mu m$, a decrease of -15%. Picking a smaller

feature size of $d = 500 \mu m$, leaving $b = 25 \mu m$, we find an optimal $a = 42 \mu m$ and a corresponding $R_{crit} \approx 55 \mu m$, a decrease of -60% from the current value.

The smallest possible feature size for a reliable electrode design is about $10\mu m$. Setting this as a criterion for the gap and electrode width, i.e. $2b = 10\mu m$ and $a = 10\mu m$, we find minimal the best corresponding electrode length of $d = 117\mu m$ that reduces the critical radius to $R_{crit} \approx 15\mu m$, a reduction of almost -90% compared to the current design. Hence, it should be noted that a smaller R_{crit} also results in longer shedding times, thus less frequent sweeping of the surface below and an increased blanketing effect. Effects like these are not included in the model; these and several other suggested improvements are mentioned in the outlook.

4.6 SURFACE DEGRADATION

We studied the effects of condensation and EW on the degradation of the samples in order to understand the stability of our surfaces and reliability of our data. This was done by long condensation experiments over 40 hours with PDMS LIs and Teflon surfaces with and without EW. During these experiments condensate was collected and its mass was measured continuously. The camera took pictures of the breath figures continuously from which the critical shedding radii were determined. Due to the temperature and flow rate fluctuations in the condensation chamber within and between experiments, the measured volumetric condensation rate was not accurate enough to compare the small changes in condensation rate between the experiments. These experimental setup limitations are explained in section 3.1.3.

The amount surface degradation was therefore determined by looking at the critical shedding radius. The critical shedding radius is a direct measure of the pinning forces acting on the drops. The bigger the critical radius, the larger the pinning forces and the more degraded the surface. Keeping the shedding radius low results in increased heat transfer (section 2.1.2).

Figure 4.16 shows breath figures of the condensation process for the 4 configurations: LIs without EW (LI no EW), Teflon without EW (Teflon no EW), LIs with EW (LI EW) and Teflon with EW (Teflon EW), for 5 moments in time. The temporal evolution of the critical radius is shown in figure 4.17b for the first two hours of condensation, and in figure 4.17a for 40 hours of condensation. The temporal evolutions shows the raw data points from the image analysis as dots. The raw data is fitted by a horizontal line for the equilibrium data (solid lines) and exponential increase at the start (dotted lines). Hereby we assume surface degradation is an ephemeral process; surface degrade and reach a constant degraded state after some time. The calculation for the fitting is elucidated in section 3.4.3. Surface characterization was discussed in section 3.2.3 using the EW curves.

10 minutes into condensation, the LIs show a smaller critical shedding radius compared to the Teflon samples. The oil in the LIs creates a low friction surface where drops slide down early with little hysteresis, as is also visible in the EW

Figure 4.16: Breath figure series of 4 types of (functionalized) substrates during 2400 minutes of condensation.

curve. The sliding speed is considerably lower than on Teflon as the viscous dissipation is higher. The Teflon surfaces show an important difference between EW and no EW. With EW the hysteresis is decreased resulting in a smaller shedding radius. This difference is not evidently visible for the LIs. The LIs do by nature have a much smaller hysteresis and therefore the added benefit of EW is small.

120 minutes into condensation, we observed an increase of critical radius for LIs from $100\mu m$ to $400\mu m$. This increase is caused by the decrease in hysteresis due to removal of (access) oil from the surface. The low surface energy silicone oil encapsulates the high energy surface energy water drops, as explained in section 2.5 Each shedding condensate drop rolling of the surface takes with it a finite amount of oil, increasing the hysteresis of the surface and consequently increasing the shedding radius. For LIs there is very little difference between EW and no EW in the first 120 minutes and no surface pinning points are visible due to the lubricating or self-healing behavior of the oil on the surface.

The critical radius of the Teflon surfaces does show different behavior for EW and no EW. The decrease in hysteresis with EW results in a lower critical radius compared to no EW. The steady state critical radius for Teflon surfaces with EW is reached within the first hour, while without EW this steady state is only reached after 20 hours of condensation. This difference in equilibration time suggests that with EW the surface degrades much faster compared to no EW. We did notice that Teflon surfaces are not as homogeneous as LIs. Small scratches on the Teflon surfaces resulted in pinning points on the surface. These pinning points cause drop trapping and consequently increased the shedding radii by a factor of almost 2 at these pinning points. This non-uniformity explains the large deviation in data points in figure 4.17a.

1200 minutes into the condensation we observed a steady state for all the samples. After this point the critical shedding radii did not change.

The LIs showed an increase of the shedding radius within the first 20 hours before reaching the equilibrium value. We assume that at this point all Silicone oil is removed from the surface and condensation is happening on the bare PDMS surface. The removal of the oil is assumed to be increased due to the oil encapsulation on the drop (calculation in section 2.5); as every shedding drop takes down a finite amount of oil.

For LIs with EW we observed a large increase in critical radius by a factor of 10 from $100\mu m$ at the start, to $1200\mu m$ after 1200 minutes of condensation. While for LIs without EW a smaller increase by a factor of 4.5 from $100\mu m$ to $450\mu m$ was observed. This difference suggests that surface degradation significantly increases with EW. A possible explanation for this effect is that with EW not only the oil is removed from the surface, but penetration of the water into the porous dielectric PDMS coating breaks down the coating resulting in dielectric breakdown. Figure 4.19 shows an optical microscope of the LI surface after 40 hours of DWC with EW, where damage is clearly visible on the electrodes. The continuous current through the water in the PDMS layer might result in the creation of bubbles by electrolysis. [57] These bubbles might start a spread of damage through the dielectric, resulting in the patterned damage visible in figure 4.19. The damage to the PDMS coating breaks down the rough structure, increasing wetting of the surface and consequently resulting in a much larger R_{crit} . Without EW the PDMS layer remains intact, and thus still shows hydrophobic behavior.

We conclude that LIs, in the current form, are more suitable for short term experiments as the EW and non-EW situations show similar amounts degradation, making comparison feasible. Since for most of this study experiments did not exceed 2 hours, LIs were the most appropriate option. LIs showed the smallest critical radii and were totally uniform by the self-healing properties, whereas Teflon surfaces showed local pinning points at small scratches or damaged coatings.

LIs also proved to work well with EW, where the decrease in hysteresis caused by EW was even negligible at the start of the experiments due to the extremely low hysteresis of the surfaces by nature. For this reason EW is assumed to show even greater improvements with smaller feature sizes where earlier coalescence increases the early drop growth rate or surfaces with larger hysteresis.

For long term experiments, Teflon surfaces still proved to be superior over LIs due to oil depletion with LIs. With EW the depletion of the oil might even result in dielectric breakdown and breakdown of the porous PDMS resulting in increased wetting.

Figure 4.17: Critical shedding radius (R_{crit}) evolution during 43 hours (a) and first 2 hours (b) of condensation for 4 types of (functionalized) substrates.

Figure 4.18: A breath figure of condensation with IDE EW on Teflon showing a drop at R_{crit} without pinning (encircled green) and a drop with $R > R_{crit}$ that is pinned due to surface damage (encircled orange).

Figure 4.19: Optical microscope image of the LI surface after 40 hours of condensation with EW.

5 CONCLUSION

5.1 ACTIVE TRANSPORT

In this thesis we presented a new technique of active transport to improve the average heat transfer during DWC using EW. Active transport uses 2 isolated stacked ZZEs that are vertically offset by half a period. Using out-of-phase actuation we managed to actively transport condensate down the surface in a conveyor-like behavior between electrical traps. We identified the modified condensation phases that play a role in active transport, including drop gap-oscillations where drops oscillate around an electrical trap if their masses are too small to overcome the dynamic friction forces.

We quantified the improvements of active transport on the heat transfer using the breath figures and by volumetric condensation measurements. We observed a decrease in surface coverage of -22% from 52.1% without EW to 30.9%. The critical shedding radius decreased by -51% while the shedding frequency increased by +700% from an average cycle-time of 450 seconds without EW to 103 seconds with active transport. All these are indications of increased heat transfer. Furthermore, we measured an increased condensation rate of +11.5% using the method of continuous water collection with direct references (inter-averages). This new method for determining the changes in the condensation rate eliminates many experimental fluctuations. Due to experimental issues only one dataset was used to determine the volumetric condensation rate improvement, thus exact improvement values might differ. We conclude that active transport is feasibile and does improve the heat transfer compared to other electrode design and no EW.

5.2 ACTIVE TRANSPORT MODEL

Many parameters influence the efficiency of active transport, including electrode width, height and spacing, as well as actuation time and EW voltage. To study the effects of these parameters and further optimize the electrode design for future research, we developed a numerical force balance model to estimate the critical shedding radius R_{crit} . Using the model we showed that there is an ideal combination of electrode dimensions for a set minimum feature size, i.e. smaller is not always better. By brute forcing thousands of possible combinations of electrode dimensions we estimated the *generic optimization equation*: a single equation to find the best electrode dimensions. Using this equation we for the smallest possible feature size for a reliable electrode design, i.e. $10\mu m$, we find a decrease in R_{crit} of almost -90% from $135\mu m$ to $15\mu m$, indicating there is room for improvement making active transport even more efficient. However, it should be noted that a smaller R_{crit} also increases shedding time and decreases shedding frequency decreasing

the overall heat transfer of the substrate. We conclude the model physically behaves as expected, but requires validation with experiments.

5.3 SUBCOOLING DEPENDENCY

We experimentally studied the dependence of the subcooling on the heat transfer using discontinuous water collection, where water was collection over a period of 2 hours to estimate the average condensation rate. We observed a linear increase in heat transfer for small subcoolings ($\Delta T < 35K$), analogous to theory. For large subcoolings we note an increase in heat transfer exceeding the linear trend that we assign to shedding. For large subcooling shedding is more common causing drops further down the surface to be removed from the surface before reaching R_{crit} , thus effectively reducing the average R_{crit} .

5.4 EXPERIMENTAL SETUP AND COMPARISON OF CONDEN-SATEION RATE METHODS

In our effort to accurately determine the condensation rate for different substrates, we developed a new experimental setup. This setup was designed using 3D modeling in-house and 3D printed or CNC-cut. This new setup improves on many fronts to the old experimental setup. Several improvements include easier switching of samples, modularity for future expansion and water collection.

The average condensation rate can be determined in different ways.

We compared 2 image analysis methods and 2 experimental methods to determine the condensation rate for 4 different types of substrates. One of these methods, the calc-method, is used in literature to estimate the condensation rate using the average radius and shedding frequency for ZZEs. We found that all methods give comparable condensation rates, except for the calc-method that highly underestimates the condensation rates, and indicates incorrect increases for condensation with ZZEs. We conclude that this method is inaccurate and should not be used. We conclude the most accurate analysis method to be the continuous water collection method, where many experimental fluctuations are excluded. However, none of these methods proved to be accurate enough to show differences between substrates, that are expected to show different condensation rates.

5.5 CONTINUOUS WATER COLLECTION

The method we developed that proved accurate enough to compare substrates was continuous water collection with direct references (inter-averages). Using this method we showed that ZZEs do not improve the condensation rate, as reported by Dey *et al*, but actually decrease the condensation rate by -6.6% up to -12%, depending on the design. Even though coalescence is increased with ZZEs, the electric traps increase R_{rcit} and consequently increase the thermal resistance.

When pulsing the same ZZEs, we noted a small improvement of +1.7% by temporarily disabling the electric traps.

EW with IDEs showed improvements of +3.3% up to +6.2% depending on the electrode design. The enhancement in coalescence and decreased hysteresis result in faster drop growth and decrease of R_{crit} . These percentages are smaller than previously reported values in literature. We believe this difference is due to the presence of NCG in our system where that is not the case for Baratian *et al.* Especially for low surface coverage situations, like with IDEs, this results in a significant reduction in heat transfer.

5.6 SURFACE DEGRADATION

Lastly we experimentally studied surface degradation of Teflon and Lls with and without EW by looking at the change in R_{crit} during 40 hour experiments. In all cases we found some form of surface degradation but on different timescales. In the first two hours the degradation of Lls is similar regardless of the use of EW or not, making Lls very suitable for short term experiments. The Teflon surfaces on the other hand showed mutually different behavior. Where degradation with EW happens within the first hour, without EW it takes up to 20 hours to fully degrade. Simultaneously R_{crit} is suppressed for EW due to decreased hysteresis. After 40 hours of condensation the Teflon surfaces show similar degradation, even though both degraded at different rates. The Lls degraded more than Telfon due to depletion of the lubrication oil. Each drop is encapsulated by oil removing a finite amount with every shedding event. After 40 hours this resulted in condensation on the bare PDMS. With EW the depletion of the oil might even result in dielectric breakdown and breakdown of the porous PDMS resulting in increased wetting and consequently a R_{crit} twice that without EW.

Teflon surfaces did show heterogeneous pinning points where drops got stuck and the R_{crit} increased by several factors, resulting in large errors in the average R_{crit} . On the other hand, LIs degraded homogeneously due to the self-healing behavior of the lubrication oil.

We conclude that LIs do decrease contact angle hysteresis with dropwise condensation, but are only stable for hours, after which Teflon surface are still more reliable.

6 OUTLOOK

Over the past years a lot of effort has gone into active enhancement of DWC using EW by Baratian, Dey *et al* [31, 32, 62]. It has shown to be a successful technique to increase heat transfer. With this knowledge our research set out to improve on the design using double layered ZZEs. We have shown that using this design we are able to actively transport drops off the surface well below the critical solely weight-driven shedding radius, thus enhancing heat transfer.

The enhanced heat transfer can be beneficial for applications like water-harvesting or heat transfer in electronic heat management or power plants.

However, extensive water collection experiments are required to statistically substantiate our findings, as some of the data presented is statistically poor at the moment. This is the case for the active transport as well as the other continuous water collection experiments.

We believe that the double layered ZZEs design can be further optimized to increase the efficiency even more. The model that was presented in this thesis is a good starting point for that.

The model itself can be improved on several fronts. The single-electrode assumption should be extended to multiple electrodes and the model should be verified and calibrated with actual experiments to, among other things, increase the accuracy in the hysteresis. The current model is time independent; as a result longer actuation times will decrease the critical shedding radius. While in reality longer actuation time also decrease shedding frequency and velocity and accordingly reduce the benefits of sweeping and increase thermal resistance.

The home-built experimental setup presented in this thesis has shown to be a major improvement over the previous iteration. The setup is easier in use, and experimental conditions are better defined and measured. The current main bottleneck of the setup are the large fluctuations of vapor temperature that arise from uncontrollable factors like ambient lab conditions. Commercially available vapor generators that actively control temperature and humidity using feedback loops are a good option for this.

The lack of industrial applications of DWC is mainly due to fragile hydrophobic surfaces needed for DWC. Our efforts towards increased heat transfer using EW would be purposeless if this problem remains unsolved. An advantage of EW to control shedding behavior is the lack of surface modification that many passive techniques require, as the electrodes are hidden in the surface. This allowed us to use LIs as top coating with the main benefits being extremely low CAH and self-repairing. However, during long term experiments the lubrication oil depleted com-

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pletely from the surface, resulting in the complete loss of hydrophobicity. Further development of the LIs is needed to prevent oil depletion. This could be achieved by selecting oils that do not encapsulate drops and evaporate little, or using an oil reservoir that continuously self-replenishes the lubricant. [72] We were also unable to combine LIs with active transport due to the complex electrode design. More surface engineering is needed to overcome this problem.

We see this hybrid solution of EW with LIs as a promising combination to enhance heat transfer for long periods.

The research done on EW with DWC has increased our understanding of the physical changes. However, to our knowledge, no research has studied the effect of EW on the nucleation of drops. Even though no significant effects are visible on the millimeter-scale, the effects at the micro- and nanoscale remain unknown. These effects can be studied using a Quartz crystal microbalance or using an optical microscope and high speed recording.

The author genuinely hopes this research will continue within the PCF group or elsewhere, eagerly to see further developments of the unique combination of DWC and EW.

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A DOCUMENTATION EXPERIMENTAL SETUP

Documentation of the second-iteration Experimental Setup for Condensation Experiments

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This document is the full documentation of the experimental setup for dropwise condensation experiments.

Section 1 gives a general introduction to the experimental setup with the workings of the setup, design choices and explanation of the 3D model and samples.

Section 2 contains a detailed experimental protocol for setting up and experiment, running an experiment and analyzing the data produced during an experiment. This section also contains detailed lists with recommended settings and workings of software needed to acquire data.

Section 3 gives a detailed step-by-step explanation to assemble the complete experimental setup. This section can be skipped if the setup is already installed and working.

Section 4 gives a list of all the 3D parts, tubing and other parts with ordercodes and more. This section is helpful when parts are missing, broken or spare parts are needed.

Section 5 gives an overview of all the important files this documentation comes with.

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1 General introduction

The home-made experimental setup is schematically shown in figure 2 with the flow diagram in figure 1. A picture of the tubing at the experimental setup is shown in figure 3. All the valves are shown in figure 4.

De-ionized water is heated on a hot plate (RCT Basic, IKA labortechnik). Ambient air is blown through the water using an aquarium pump (0886-air-550R-plus, Sera) and the flowrate is monitored by a flowmeter (AWM5101VN flowmeter, Honeywell). The water reaches an equilibrium temperature after 30-45 minutes.

During an experiment the humidified air is guided into the condensation chamber through two inlets at the bottom of the chamber.

The humidified air passes in front of a vertically mounted cooled sample where the vapor partially condenses. The remaining vapor exits the chamber through a fine grid of holes at the top of the chamber. Temperatures inside the chamber are measured by multiple thermistors (TCS651m AmsTECHNOLO-GIES and Thorlabs TSP-TH) using a DAQ card, a home-made voltage divider with 10K Ohm resistors a complimantary LabView script and the Thorlabs TSP01 Application. Thermistors are located at the inlet of the chamber, close to the sample, the outlet of the chamber, in the coolant behind the sample, in the heated water on the hot plate and the ambient air. These thermistors give the amount of subcooling in the chamber and ensure reproducibility of experiments. A humidity sensor (Thorlabs TSP01) is placed either outside the chamber or inside the chamber to measure the relative humidity.

The transparent sample is cooled using a commercial cooler (Haake-F3-K, Thermo Fisher Scientific) by passing cooled water trough a coolant cell behind the sample. The condensation on the front of the sample slides down off the sample into a funnel that is connected to a water collection tube or to a home-made hydrophobically coated funnel that direct the water to a collection tube on the balance (Nimbus Analytical Balance NBL 254i). The balance is readout using the ADAM DU software in realtime.

Images of the condensation process are recording using a 4K camera (Point Grey, FL3-U3) fitted with a 20X zoom lens (Z125D-CH12, EHD) for magnification. The sample is back illuminated using a LED light source (MB-BL305-RGB-24-Z, Metabright). Light passes through a window in the coolant cell, through the transparant coolant fluid, transparant sample, condensation chamber and heated window before being captured by the camera system. The heated window prevents condensation on the view port for the camera and is made of an ITO coated sheet of glass through which a current is passed using a current source (Delta Elektronika Power Supply E 030-1).

For condensation experiments with electrowetting, an alternating voltage is applied to transparent interdigitated electrodes on the sample using a function generator (Agilent 33220A) and voltage amplifier (Trek PZD700A).

The flow diagram in figure 1 shows several valves. Using the by-pass valve it is possible to by-pass the vapor flow from the heater to a drainage beaker. This allows pre-equilibration of the water on the hot plate without flow through the condensation chamber.

Since all the tubing is not isolated, vapor condenses inside the tubing on its way to the chamber and reduces the flow rate. The condensate-drain valve is located at the lowest point in the tubing and allows for drainage of the condensate during the experiments.

For drying the condensation chamber before or after an experiment, a Nitrogen inlet is located in the tubing system.

To prevent condensation on the outside of the coolant cell at high humiditiy in the lab, instrumental air can be purged over the outside of the coolant cell. The same instrumental air valve allows airflow through the hydrophobically coated funnel used for water collection experiments to the balance to prevent damage to the coating.



Figure 1: Flow diagram of the experimental setup.



Figure 2: Schematic of the experimental setup.



Figure 3: Top view of the tubing at the setup.



Figure 4: (a) Nitrogen valve for drying out the chamber. (b) By-pass valve for directing vapor to the chamber (current position) or to the drain. (c) Drain valve for draining condensate from the tubes.

1.1 Motivation of design choices

This second iteration of the condensation setup improves on many of the complicated and time consuming tasks the old setup required. The main design choices are made with the following preferences in mind: 1) easy sample swapping, 2) fast electrowetting connections, 3) easy to expand or change the

setup with future experiments, 4) water collection and 5) reproducible chamber conditions and with accurate condition measurements.

These preferences are accomplished by 1) having the sample being held in place with a sample holder and bolts, 2) using spring-loaded POGO connections, 3) having a modular design which allows for redesigning, printing and swapping out specific parts, 4) having sample holders with funnels that connect to tubes or a balance and 5) equipping the setup with multiple temperature sensors, a flow meter and a humidity sensor to accurately measure the conditions in the chamber and a by-pass in the system to reach good vapor equilibrium.

1.2 Recommendations for further improvement

Some recommendations for further improvements to the setup include:

- Humidity control. The current setup creates vapor of 100% humidity only. This could be achieved by a home-made feedback loop of 2 electronic valves with dry air and humid air that are mixed to achieve the desired humidity, or a commercially available system.
- Pre-equilibration of the condensation chamber before experiments. Currently when an experiment is started, water vapor is led into the chamber after which it takes up to 40 minutes to reach an equilibrium temperature in the chamber. This could be done by closing off the sample with a shutter that is opened once the chamber reaches equilibrium.
- Heated pipes from the water boiler to the chamber to prevent heat loss and condensation on the chamber.
- Hydro dynamically designed coolant cell to for laminar flow and better heat transfer.
- Fully closed system to allow experiments other fluids than water.
- No non-condensable gasses.

1.3 3D model

The experimental setup is designed in Google SketchUp and SOLIDWORKS 2018. It is designed to be fully modular, as shown in figure 5, consisting of multiple separately 3D printed and CNC cut parts that are bolted together. This makes redesign and replacement of specific parts possible, such as sample holders for different types of samples.

The latest 3D model file as of writing this manual is setup2018_v16_newsampleholders.skp. Since Google SketchUp models are 3D meshes, it cannot be used for CNC cutting, which requires 3D solids. So the main condensation chamber is also available in SOLIDWORKS 2018 with filename Main chamber.SLDPRT.

A SOLIDWORKS 2018 Assembly is also available for high quality renders with filename SetupAssembly_Aug122019.SLDA Parts for printing can either be created by exporting individual layers to .stl files, or by using these already exported files. See section 5 for an overview of all the parts and its filenames.

Suggested is that all 3D printed parts are printed at 100-micron resolution with the thermoplastic acrylonitrile styrene acrylate (ASA). ASA is known for good mechanical properties, UV stability and high chemical resistance. It is commonly used for outdoor applications.

1.4 Main chamber

Figure 6 shows the technical drawing of the main (CNC cut) condensation chamber with important measurements. All holes for bolts have a tolerance. The holes in the top and bottom of the chamber fit two pneumatic G1/4" fittings with BSPP thread (thread is not in model and needs to be done manually).

The chamber has many extra holes for extension modules to be installed. When not in use, these holes can be filled up using rubbers stoppers. See section 4.4 for suggested stoppers.

A list of suggested bolts and nuts can be found in section 4.3.

1.5 Sample holder and samples

Samples are held in place using a sample holder. Sample holder are 3D printed frames that presses the sample on a silicone ring with bolts that go through the main chamber.

The experimental setup only support samples of 70x70mm. Samples need to be transparent to allow back illumination and image recording. It is therefor recommended to use ITO electrodes when using electrowetting.

To use samples with electrowetting, the sample holders have spring-loaded contacts at specific locations. Section 4.4 lists the recommended spring-loaded contacts with ordercode. Figure 7 shows the locations of these contacts. The 70x70mm sample is shown in grey, the outlined shape in the middle is the open area in the sample holder and the white circles inside the grey area are the spring-loaded contact holes. There is also a mirrored sampleholder available with electrowetting connects on the left side of the sample.

When performing pulsed electrowetting experiments, or double attenuation-electrowetting experiments, connect the function generator(s) via USB to a computer and run the Labview program Agilent33120A_sinepulsed_v5 (designed by Daniel Wijnperle).

1.6 Images of the setup

Figure 8 shows the hydrophobically coated funnel for dynamic water collection experiments. The funnel connects directly to the sample holder and directs the water to a container on the balance. Note that the top of the funnel is sealed off to prevent water leaking out of the condensation chamber entering the funnel. The funnel is purged with instrumental air to prevent coating damage due to heat and humidity. Figure 9 shows the front and back side of the main condensation chamber with coolant cell and more. Figure 10 shows the water boiler and a close-up of the sample, sample holder, electrowetting connections and more during an experiment.

1.7 Stability and Reproducibility

We tested the performance of the new setup by characterizing the results in two groups: stability and reproducibility. With stability we look at the experimental conditions within one experiments, how stable are these conditions. With reproducibility we check the experimental conditions with the same settings between multiple experiments.

	Sensors	Value
$T_{\mathbf{middle},\mathbf{LV}}$	1	$33.7 \pm 1.5^{\circ}C$
$T_{\mathbf{middle},\mathbf{TL}}$	1	$34.8\pm0.8^\circ C$
$T_{top,LV}$	2	$33.4\pm1.6^\circ C$
$T_{\rm bottom,LV}$	2	$35.8\pm1.8^\circ C$
$T_{\text{coolant cell,TL}}$	1	$10.44\pm0.02^\circ C$
Flow rate	1	3.5 - 3.95 SLPM
RH lab	1	$52.3 \pm 4.1\%^*$
$T_{\text{ambient}, \text{TL}}$	1	$22.6\pm0.2^\circ C^*$

Table 1: Chamber stability

Chamber stability

We performed an 83-hour condensation experiment without EW to check the stability of the chamber. These 83 hours is an extreme case as usual experiments last only 2 hours. We thus expect the fluctuations mentioned here to be the worst-case-scenario.

Table ?? lists average sensor values and standard deviations from this experiment. The abbreviations LV and TL stand for Labview and Thorlabs, respectively, referring to the brand of thermistors, electronics and read-out software used. In the condensation chamber sensors are located at 3 locations: at the bottom, i.e. at the vapor inlets, in the middle close to the sample (~1 cm from the sample for the Labview sensor, ~2 cm for the Thorlabs sensor) and at the vapor outlet at the top of the chamber. The coolant cell behind the sample has its own thermistor and also the flow are, relative humidity and ambient temperature are measured continuously. The average subcooling during an experiment is defined as $T_{\rm middle,LV} - T_{\rm coolant \ cell,TL}$.

We note barely any fluctuations in the coolant cell, while the condensation chamber temperatures fluctuate with about 1.5K. These fluctuations come forth from the delicate equilibrium the setup depends on. Fluctuations in for example ambient temperature or flow rate cause a new equilibrium resulting in different chamber temperatures. Also, during an 83-hour experiment the water boiler has to be refilled twice. Even though this is done very slowly, the equilibrium water temperature is different afterwards. The flow rate depends on the water level in the boiler. A full boiler results in flow rate of 3.5 SLPM, an almost empty boiler in 3.95 SLPM.

This data does not show the initial chamber fluctuations during heat-up when the vapor is just introduced into the dry and cold chamber. After the introduction of vapor into the chamber it takes about 11 minutes to reach temperatures within 5% of the overall-averages. It takes 30-45 minutes, depending on the vapor temperature, to reach full equilibrium.

The chamber relative humidity is about 5-10% before vapor is introduced into the chamber. At equilibrium the relative humidity is 100%.

Chamber reproducibility

For the reproducibility of the experimental conditions in the chamber we collected the data for 60 experiments (>150 hours of data). The averages and standard deviations are listed in table ??. Only condensation chamber temperatures are listed here, as these fluctuate most. To change the experimental subcooling, we change the vapor temperature by setting a different hot plate temperature. Hot plate

$T_{hot plate}$	150	190	200
$T_{\mathbf{bottom}}$	36.9 ± 3.7	37.7 ± 3.3	42.3 ± 3.2
$T_{\mathbf{middle}}$	34.3 ± 3.4	36.1 ± 2.8	38.9 ± 3.8
T_{top}	33.9 ± 3.5	35.7 ± 2.8	37.3 ± 3.2

Table 2: Chamber reproducibility

temperatures of 150, 190 and 200 degrees were the most common ones. We observe temperature fluctuations of 2.8-3.8K between experiments, showing that duplicating exact experimental conditions is not possible. These errors arise from both the inaccurate setting of the hot plate temperature as well as the ambient fluctuations mentioned previously.



 $\label{eq:second} Figure \ 5: \ Modularity \ of \ the \ experimental \ setup, \ designed \ in \ \texttt{Google SketchUp} \ and \ \texttt{SOLIDWORKS 2018}.$









Figure 8: The hydrophobically coated funnel that direct droplets to a balance for (dynamic) water collection experiments.



Figure 9: The main condensation chamber. (a) Front view showing the heated window connections, tube for (static) water collection experiments, the camera lens, light source and more. (b) Back view showing the bolts and wingnuts that hold the sampleholder with sample in place, the coolant cell, tubing and more.



Figure 10: (a) The heated plate (boiler) showing are being purged through the water. (b) An inexperiment picture taken of the sample and (old design) sample holder showing the spring-loaded POGO electrowetting connections, thermistors, sample, condensate collection funnel and more.

2 Experimental Protocol

This protocol describes step-by-step all the procedures for doing condensation experiments. From sample preparation to data analysis.

A useful logbook template for these experiments is available in the enclosed files. See section 5.

2.1 Sample preparation

Samples for condensation experiments are either coated with Teflon, or liquid-infused on PDMS coating. This protocol only describes the coating procedure for liquid-infused samples. The setup only supports $70 \times 70mm$ samples.

2.1.1 Sample coating

- 1. Prepare the PDMS mixture for spincoating. PDMS part A and part B (Sylgard 184, Dow Corning) mixed in a 10:1 (weight:weight) ratio and steered during 2 minutes.
- 2. Place PDMS in vacuum desiccator for degassing (20 minutes).
- 3. Spincoat sample with PDMS for 5 minutes at 2000 rpm for a coating thickness of $10\mu m$ or 1 minute at 4000 rpm for a coating thickness of $20\mu m$. [1] Ensure a dust free sample before spincoating.
- 4. Bake the sample for 2 hours at $60^{\circ}C$.
- 5. Soak the sample with Silicone oil for at least 2 hours.
- 6. Blow off the access oil before using for an experiment. Wipe the back side of the sample clean with a a tissue to prevent oil from contaminating the coolant water.
- 7. Scratch the coating off the electrode connections for the spring-loaded electrowetting connections.

2.1.2 Removing old sample

- 1. Open the condensation chamber by removing the top lid and open the front by disconnecting the PCB connector and sliding the plexiglas front upward.
- 2. Drain the coolant fluid from the coolant cell by disconnecting the outlet hose in the top of the chamber.
- 3. Remove the sample holder by disconnecting the electrowetting connections to the sample holder, bending the thermistors away and removing the bolts that hold the sample holder in place.
- 4. Pull the sample out using the notch on the bottom of the sample holder.

2.1.3 New sample mounting

At this point the chamber should be open and no sample is mounted.

1. Place the silicone ring in the main chamber. Make sure the ring lays flat to prevent cracking of the sample when pressing the frame. Tip: spray water on the silicone ring or chamber to help keep the ring in place for the next steps. See figure 11.



Figure 11

- 2. Put the sample on top of the silicone ring and place the sample holder on top. Secure the sample holder with at least 8 bolts (ok, 4 works fine usually when you're lazy) and tighten evenly by going around multiple times. Don't over-tighten the bolts, only little force is needed for a good seal. See figure 11.
- 3. Reconnect the coolant outlet hose on the top of the chamber and test the seal by turning the cooler on. It is common for air bubbles to appear at the start that are in the circuit.
- 4. Connect the electrowetting connections to the sample holder and bend the thermistors back into place.
- 5. Close the condensation chamber by sliding the plexiglas front in from the top and closing the top of the chamber with the lid. Connect the PCB connector to the front window for the heated window. See figures 12 and 13.
- 6. Purge the chamber with dry nitrogen to prevent condensation on the sample.

2.2 Experiment preparation

It takes approximately 90 minutes for the water on the hot plate and 20 minutes for the water cooler to reach equilibrium temperatures. See section 2.6 for additional settings.

- 1. Refill boiler flask with milli-Q water half way up to the logo.
 - 15



- 2. Make sure the chamber valve (figure 4 or 1) is set to the bypass (handle pointing left).
- 3. Turn on the hot plate with stir and turn on the air pump to purge the water.
- 4. Turn on the water cooler.
- 5. Turn on the following sensors / electronics:
 - (a) Airflow sensor power adapter;
 - (b) Red multimeter adapter;
 - (c) Light source;
 - (d) Cooler thermometer;
 - (e) Heated ITO window;
 - (f) Function generator + amplifier if EW experiments are performed.



Figure 13

- (g) Open the instrumental air valve slightly to purge air over the plexiglas window in the coolant cell to prevent condensation and to cool the coated funnel when performing dynamic water collection experiments.
- 6. Start the Labview measurements program (temperature_control_TE_v3_harmen-9.vi) and the Thorlabs TSP01 Application. Note that Labview automatically saves the data, but that for Thorlabs you have to save the data manually afterwards.
- 7. Create the folder structure on the pc for saving the data:
 - (a) In the directory c\Data\Condensation Data create a folder with the date like YYYY-mm-dd.
 - (b) In that folder create a folder for each recording like YYYY-mm-dd_Rec001_0V.
 - (c) Inside that folder create a folder Raw Data.
 - (d) Inside that folder create three folders for the background, electrodemap and experimental images: YYYY-mm-dd_Rec001_0V_bg, YYYY-mm-dd_Rec001_0V_em, YYYY-mm-dd_Rec001_0V_data.
- 8. Start up the camera software Point Grey FlyCap 2.
 - (a) Open the settings window. In Camera Settings uncheck the checkboxes for Auto and set the following settings:
 - i. Brightness: 0,000%;
 - ii. Exposure: 1,415%;
 - iii. Sharpness: 1024;
 - iv. Hue: 0,000deg;
 - v. Saturation: 100,000%;
 - vi. Gamma: 1,000;
 - vii. Shutter: variable, see below;
 - viii. Gain: 0,000dB;
 - ix. FrameRate: 1;
 - x. W.B.(Red): 520;
 - xi. W.B.(Blue): 655;



xii. Power: Checked.

- (b) In the same settings window, in Custom Video Mode, set the video resolution to $4K\ 4:3$ resolution (mode 10).^2
- (c) Open the recording settings. set the following settings:
 - i. Recording mode: Streaming;
 - ii. Capture: variable, see below;
 - iii. Image Format: PNG;
 - iv. Compression level: 1;
 - v. Save as interleaved: Unchecked;
 - vi. Save as raw: Unchecked;
 - The recording mode will be set later.
- 9. If performing a dynamic experiment (real-time weighing using using the balance), the balance (ADAM 254i) should be setup.
 - (a) Connect the balance to the computer using USB.
 - (b) To set the balance correctly, press Setup and navigate through the menu until SERIAL appears, than press Setup again. Set the serial interface paramters as:
 - i. ENABLE: ON;
 - ii. BAUD: 9600;
 - iii. PARITY: NONE;
 - iv. STABLE: OFF;
 - v. CONTIN: ON;
 - vi. PERIOD: 1;
 - vii. FORMAT: STANDARD;
 - (c) On the computer, open the software ADAM DU, open a new data window and press Start New Data Collection Session.
 - (d) Select from the Data menu Enable Custum Data Collection Panel to save the datetime stamp as well.
 - (e) From the dropdown menu for Select Balance/Scale select PGW/PW/NBL/EBL, select the right COM port, set the Baud/Speed to 9600 and Mode to n,8,1. The actual recording is started right before the experiment.
 - (f) Select Continuous Data Logging from the Data menu to continuously write the data to a file.

2.3 Experiments

With the coolant water and heated water at equilibrium temperatures, the experiment can be started. It is important to first record a background image, that is an image with no droplets on the sample, for the image analysis afterwards and an image of the electrodemap to use as a length conversion factor.

1. Align and focus the camera:

 $^{^{2}}$ July 2019: I noticed black vertical lines on the left side of the frame due to some malfunctioning of the camera. If this is the case, set the resolution to 3968x3000 and shift the field of view 32 pixels to the right. Keeping the black bar results in the border droplet detection failing when analyzing the images with MATLAB afterwards.

- (a) Spray some droplets on the substrate.
- (b) Align the camera to the top of the substrate as much as possible. Make sure the camera is perpendicular to the sample (so all of the droplets are in focus).
- (c) Focus the camera on the droplets using the camera stage translations. Note that due to the spherical shape of the droplets and narrow aperture of the lens (large depth of field), there is a large range where the camera is approximately in focus. This translates to about 9 rotations of know of the perpendicular direction of the translation stage.
- (d) Make sure the light source is circular (done using the 3D printed circular aperture, see section 3.9) and the transmissions through the droplets in the center.
- (e) Blow the droplets carefully of the surface using Nitrogen. Make sure that the funnel is dried out as well.
- (f) Set the Shutter under Camera Settings in the settings window to 45ms to see the electrodes. Make sure the electrodes are vertical (small deviations are corrected for in the image analysis), rotate the camera slightly to fix this.
- (g) From this point on, do not bump into the table or move the camera.
- (h) Turn on the heated window.
- (i) If performing a static experiment (no real-time weighing of the condensate), connect the collection tube to the sample holder. See figure 14.





- (j) If performing a dynamic experiment (real-time weighing using using the balance), connect the collection tube to the balance. Make sure the pipe leading into the collection box on the balance does do not touch the collection box on the balance.
- 2. Record electrodemap and background images:
 - (a) Set the shutter speed to 45ms for the electrodemap images (you should see the electrodes). Select the _em folder in the recording window. Use for the filename the folder name. Record 5-10 images.
 - (b) Set the shutter speed to 90-100ms for the background images. Select the _bg folder in the recording window. Use for the filename the folder name. Record 5-10 images.



- 3. Start of the experiment:
 - (a) Select the _data folder in the recording window, set the Recording mode to record 1 out of every 3 images for 0 images (infinite recording at 1/3fps, standard). Check section 2.6 for more recommended recording settings.
 - (b) Select the correct electrowetting frequency and voltage and check the voltage using the voltmeter (with the sample not connected). When performing pulsed electrowetting experiments, or double attenuation-electrowetting experiments, connect the function generator(s) via USB to a computer and run the Labview program Agilent33120A_sinepulsed_v5 (designed by Daniel Wijnperle).
 - (c) If performing a dynamic experiment (real-time weighing using using the balance), the recording has to be started. Press Confirm Comms. Settings and Start Collecting Data. The session should now display the balance reading multiple times per second.
 - (d) Close the Nitrogen flow purge in the chamber.
 - (e) Ensure the drain valve (figure 4 or 1) is closed.
 - (f) Turn the chamber valve from the bypass position to the chamber position (handle pointing right). At the same start recording images and write down the time, this is needed for synchronizing the images with the temperature data and the actual flow in the chamber.
- 4. During an experiment it can be useful to drain condensate in the inlet tubing by opening the drain valve (figure 4 or 1) for a few seconds every half an hour or so. This prevents water for spitting up to the sample or heated window.
- 5. End of the experiment:
 - (a) Stop recording images, turn the chamber valve to the bypass position (handle pointing left) and write down the time.
 - (b) Turn off electrowetting and the heated window.
 - (c) Open the drain value and open the top of the chamber.
 - (d) Slowly open the Nitrogen purge through the chamber to dry the sample.
 - (e) Stop the Labview measurements program and write down the exact time of stopping. This is needed since Labview does not save this internally! Go to the saved folder (named data on the Desktop) and open the file in with Excel Importer. Save the data as .xlsx like YYYY-mm-dd_Rec001.
 - $(f) Stop the logging in Thorlabs. Manually save the data by pressing the save icon like {\tt YYYY-mm-dd_Rec001}.$
 - (g) Stop the data collection in ADAM DU. Open the file in Notepad and save with UTF8 encoding (otherwise MATLAB is unable to open the file).
- 6. Repeat steps 2 to 5 for the other experiments.

Important notes for the experiments:

- 1. Image quality is the most important aspect good data. Even though the image analysis work relatively well with poor images, the best results are obtained with good images! If the droplets are not in focus, stop the experiment and start over.
- 2. For safety: do NOT leave EW and/or the heated window on when working in the chamber or close to it.

2.4 After experiments

- 1. At the end of the experiments turn off the sensors / electronics:
 - (a) Airflow sensor power adapter;
 - (b) Red multimeter adapter;
 - (c) Light source;
 - (d) Cooler thermometer;
 - (e) Heated ITO window;
 - (f) Function generator + amplifier if EW experiments were performed.
- 2. Put on the lid on the condensation chamber and slightly turn on the Nitrogen flow (a small overpressure is enough) if the sample is reused later .
- 3. Scan the logbook to PDF and add each logbook entry to the ${\tt Raw}$ Data folder.

2.5 Data processing

Each experiment produces 5 sources of data: images of the condensation process captured by Point Grey, temperatures and flow rate data from Labview, temperatures and humidity from Thorlabs, condensate mass from the ADAM balance and the labnotes with the timestamps of different processes during each experiment.

The goal of the data analysis is 1) to process the data into readable MATLAB-format, and 2) to link the different data sources with one another.

Figure 15 shows the data processing flow diagram and the different scripts used to process the data. All scripts are briefly explained below.

MathWorks MATLAB is required to analyze the data. All scripts are tested in version R2018b.



Figure 15: Data processing flow diagram. Data sources are depicted with a dotted line, analysis steps with a solid line. Note the dependency between the different data sources and analysis.

2.5.1 Image processing

Workings of the image analysis

The high contrast color images recorded of the condensation process are loaded onto a computer for analysis using a home-made program in Mathworks MATLAB. The goal of the image analysis is to extract the exact location and size of every droplet in the image. This procedure is complicated by the fact that each droplet shows circular holes in the image due to the transmitted light of the circular light source as well as the complication of overlapping droplets in the image due limited image resolution, poor focus or the physical problem large contact angles where small droplets can exist partially underneath large droplets.

The key for reliable image analysis is in the quality of the recorded images. The recorded images need to be focused carefully, the light source has to be circular and the contrast between droplets and the background must be high and even (no vignetting).

A shortened version image analysis steps is shown in 16. First the original image (figure 16-(a)) is converted to a grey scale image (figure 16-(b)). Then a background image, that is an image with no droplets on the surface recorded before an condensation experiment, is subtracted from it and the contrast is increased (figure 16-(c)). Otsu's method is used to automatically determine the binary treshold to convert the image to a binary image. The Otsu's method determines the binary threshold in a manner such that the combined spread (variance) of black and white pixels is minimal. Using the obtained threshold, each image is then converted to a binary image, see figure 16-(d). In order to remove the holes inside the droplets the separate background components (regions of connecting black pixels) are detected. Each connected component with a threshold eccentricity and radius are filled (inset figure 16-(e)-(i)). Filling based on the eccentricity and radius prevents filling connected droplets.

Aforementioned, some droplets are slightly connected in the binary image (inset figure 16-(e)-(ii)). Separating these droplets starts by taking the distance transform of the image where each foreground (white) pixel is transformed into the closest distance to a background (black) pixel. The resulting image (inset figure 16-(e)-(iii)) shows a gradient from white to black where the darker the color, the further that pixel is inside a droplet. The tiny local minima are removed and the watershed transform is applied to the distance transform and translated to the binary image. The watershed transform turns local minima in the foreground pixels (the shortest connection between two connection droplets) into background pixels, thus splitting the connected droplets (inset figures 16-(e)-(iv) and 16-(e)-(v)).

The non-circular holes in the droplets are filled and the connected components of the droplets are determined. Each component now represents one droplets. For droplets not touching the border of the image (non-boundary droplets) is assumed that they are circular. Assuming the a spherical projection of the droplets on the surface the radii and center of each droplet is calculated. The volume of each droplets is calculated using a spherical cap with a contact angle determined from separate contact angle measurements.

Droplets touching the border of the image (boundary droplets) undergo an extra analysis step. Each of the components contours are circle fitted from which the radius and center location is determined. The volume inside the frame is determined based on the radius, the part of the droplet inside the image and the assumption that the droplet is a hemisphere.

The end result of the image analysis is shown in figure 16-(f) where each droplet is numbered uniquely.



Figure 16: The basic image analysis procedure for determining the location and size of every droplet. The original image (a) is converted to grey scale (b). The background image is subtracted and the contrast is increased (c). The image is converted to a binary image (d), holes inside droplets are filled and connecting droplets are separated (e). Holes in droplets are filled, the separate components are detected and the boundary droplet detection is applied (f).

Using the image analysis

MATLAB-script: DropDectionMethod2_29.m. The high resolution images to calculate the radii and location of every droplet in the frame. The exact workings of the script are beyond the scope of this manual.

The script requires the following external scripts in the same folder to work: border_detection_3.m, centroid_location_function_2.m, centroidanglecorrection.m, circfit.m, DropDetectionFunction_9.m, DropLocationEvolution.m, Electrodemap_LocationPlot_2.m, fill_drops.m, imrotate2.m, merge_images.m, plotting_results_function.m, SheddedVolumeFunction_1.m and UnitCellSplitting.m.

To analyze the images. Fill in the Settings-section in the script. Most likely only the electrowidth, gapwidth, contact angle CA_1 and total recording time t have to be changed.

Running the script requires to select a background image to ignore permanent dark spots on the surface and an electromap image as conversion factor from pixels to meters. All steps are explained when running the script.

The data is stored in the temporary **results** folder where the MATLAB script is located. The data contains the temporal evolution of the radii, the location of all droplets, surface coverage, total volume on the surface and more.

Preview movies of the raw images can be created using the script create_preview_movies.m.

Further analysis can be done using the following additional scripts:

• CriticalRadius_Shed_Time_v2.m: allows skipping through the images and manually selecting droplets that are shedding in order determine the critical shedding radius.



- DropTracking_Coalescence_Version1_9.m: allows skipping through images and manually selecting coalescence events by selecting parent (droplet(s) after a coalescence event) drops and its children (droplets before a coalescence event) in order to determine the center of mass evolution during an experiment.
- DropTacking_Coalescence_Visualization_Version1_1.m: used in addition to DropTracking_Coalescence_Version1_9.m in for visualization of the coalescence events.
- RadialDistributionT_8.m: plots the radial distribution (radius vs number of drops) for a specific normalized location (0-4) and time range.
- RadialDistributionX_2.m: plots the radial distribution for a specific time and normalized location range.
- AWA_rhat_vs_location_1.m: calculates the area weighted average and r-hat for specific x-locations for specific unit cells (used for publication[2].
- DropTrackingVersion2_7.m: allows skipping through images to follow a specific droplet. Skipping through large number is possible to semi-automatically follow a droplet. Growth of a single droplet can be plotted and visualized.

Further details of these scripts can be found in the scripts themselves.

Important note on image analysis for large datasets. The current code is unable to handle large datasets (> 50,000 files). This is due to the terrible way MATLAB allocates variables. The code will be able to run and analyze the frames, but fails to write the temporary parfor data to final variables. Even when analyzing a smaller set, pre-allocation causes the same problem. Variables like Radiil grow above 1.5GB for example. Rewriting big chunks of the code is necessary to fix this. For now manually select a range of images to analyze.

2.5.2 Temperature data processing

Temperature data includes the data from Labview (.xlsx) and Thorlabs (.txt). Use the script Experiment_Temperature_Plots_May2019 to analyze the data. Note that the exact times of starting an experiment, stopping an experiment and stopping the recording of Labview are mandatory to synchronize all the data.

2.5.3 Balance data processing

The weight data from the balance (.csv) should be analyzed after the temperature data, as it requires the exact experiment subcooling during an experiment. Note that the .csv-file should be formatted as UTF-8 in order for MATLAB to read it. By default the ADAM DU software does not export it in this format. The data can be easily converted using Notepad.

Use the script Continuous_water_collection_July_2019.m to analyze the data. Exact times of starting an experiment and stopping an experiment are mandatory. Additionally the times between which electrowetting was on can be used to linearly fit the data separately for different stages in the experiment.

2.6 More settings

2.6.1 Recording settings

Shooting in 1/3 frames per second is the most common settings. Higher speed recordings are limited by the writing speed of the images to the drives / USB connection speed. Using memory buffering it is

Resolution	Buffered	Streamed	
4k 4096x2160	May 762f 27a 21fpg	1fps continuous	
PNG images	Max 7051, 578, 211ps		
4k 4000x3000	Max 574f, 38s,	0.0fpg.continuous	
PNG images	15.1fps	0.91ps continuous	
4K 4096x2160	Max 930f, 44s,	4.76fpg continuous	
H.264 movie	21.14fps	4.701ps continuous	
4K 4000x3000	May 670f 45a 15fpg	2 57fpg continuous	
H.264 movie	Max 0701, 458, 151ps	3.57 lps continuous	
3K 3072x1620	Max 1453f, 54s,	1.72fpg_continous	
PNG	27fps	1.721ps continous	

Table 3: Experimentally determined maximum frame rates for several recording settings.

possible to record at higher fps for short durations. Infinite0 recording is only possible for streaming. Table 3 lists some experimentally determined frames rates for different settings. All values are determined with a USB3.0 connection and a resolution of 4000x3000.

Note that recording in H.264 movie is possible, but not recommended for the image analysis.

2.6.2 Subcooling temperatures

With the current setup it is hard to reach a specific subcooling temperature. Each vapor temperature is reached by an equilibrium of the hot plate heating the water and the cold ambient air being pumped through the water.

Figure 17 shows the linear regression of the set hotplate temperature and the corresponding equilibrium temperatures in the bottom and the middle of the chamber. The temperature in the middle of the chamber is used to calculate the subcooling as:

$$\Delta T = T_{middle} - T_{cooler} \tag{1}$$



Figure 17: Experimentally determined equilibrium chamber temperatures for several set hot plate temperatures.

3 Experimental setup assembly

This section contains a step-by-step guide to assemble the experimental setup, mainly the condensation chamber. This section by itself is not sufficient to assemble the full setup. Other sections in this document can be used for that.

3.1 Hole threading

Thread the holes in the top and bottom of the main chamber (Main Chamber.SLDPRT) and the two holes in the chamber bottom (bottomfiller_v12.stl) with G1/4" BSPP thread.

3.2 Coolant window

Insert the plexiglas coolant window (56.0mm x60.5mm) and glue into place using 3M Scotch-Weld DP8005 Liquid Acrylic Adhesive glue. See figure 18. Note that other glues like superglue, NOA or standard epoxy are not sufficient to create a pressure resistant seal.



Figure 18

3.3 Chamber frame

Attach the chamber frame (frame_v12.stl) using 4 (or 8) M3 bolts and nuts. See figure 19.

3.4 Chamber bottom

Put the chamber bottom (bottomfiller_v12.stl) in place by sliding it in place from the inside of the chamber frame and secure it using two M3 bolts with M3 wingnuts. See figure 20.



Figure 19





3.5 Mounting to optical posts assembly

The setup can be mounted to the 1/2-inch optical posts assembly using 4 to 8 M6 bolts on the 90-degree angled mounts (e.g. Thorlabs RA90). See figure 21.

3.6 Chamber side walls

Two pieces of 173.5mm x 36.0mm of 6mm thick plexiglas can be slid into the chamber frame to close the sides of the chamber. See figure 22.



Figure 21

3.7 Chamber front assembly

Mount the handle (handle_v12.stl) to a plexiglas sheet of 174mm x 122mm of 6mm thick using 2 M3 bolts and nuts. Mount the PCB connection (pcbmount_v9.stl) using two M3 bolts and nuts. The heated ITO window can be held in place using 4 small bolts with spacers and nuts and wired to the the PCB connector (not shown in images). See figure 23.

3.8 Tubing assembly

Tubing for coolant and vapor flow is connected with 4 G1/4" pneumatic fittings. See figure 24. The flow diagram can found in figure 1.

3.9 Light source assembly

For the back illumination light source a custom made frame can be used. The light source itself is slid into the main frame (lightsource_mainframe_v9.stl) and held in place using the circular aperture (lightsource_aperture_v9.stl using 3 M3 bolts and nuts.

The controller is mounted onto the baseplate (lightsource_controllermount_v9.stl) and held in place using two M3 bolts and nuts.

The whole assembly can be mounted to the 1/2-inch optical posts assembly using 4 M6 bolts on the 90-degree angled mounts (e.g. Thorlabs RA90).

See figure 25.



Figure 22

Alternatively the controller can be mounted to a 1/2-inch optical post using 2 M6 bolts and 90-degree angled mounts. See figure 26.

3.10 Collection box

Since the chamber is not watertight, water will leak out during longer condensation experiments. The collection box is designed to collect this water and easily empty. Glue 2 or 4 20mm x 10mm x 2mm magnets (Neodymium) in the collection box arms (collectionbox_arms_v10.stl) and 2 to 4 in the water collection box itself (collectionbox_v10.stl) using standard epoxy or super glue. Make sure the polarities of each set are inverted so the collection box attaches to the arms. Mount the arms to the bottom of the main chamber using 4 M6 bolts and nuts.

See figure 27.

3.11 Sample holder

The sample holders for water collection (sampleholder_funnel_mirrored_v15.stl, sampleholder_funnel_v12.stl and sampleholder_funnel_v15.stl) have holes for spring-loaded contacts that make the electrowetting





Figure 24

connections. Put 6 spring-loaded contacts in the holes and glue into place using epoxy.



Figure 25



Figure 26



Figure 27

4 List of parts

This section contains all the necessary parts for the setup. These include the 3D printed, CNC cut and tubing parts.

4.1 3D parts

All 3D parts are listed in the figures 28, 29 and 30. The captions correspond to the filenames. Suggested is that all 3D printed parts, except for the main chamber, are printed at 100-micron resolution with the thermoplastic acrylonitrile styrene acrylate (ASA). ASA is known for good mechanical properties, UV stability and high chemical resistance. It is commonly used for outdoor applications. The main chamber part needs to be fully watertight since the coolant water flows through here under pressure. Many tests have shown that 3D printing is not sufficient for a good seal. Therefor it is suggested to CNC cut the main chamber out of HDPE, plexiglas or Delrin.

4.2 Tubing

The experimental setup uses pneumatic tubing and fittings of two sizes. The tubing for Nitrogen and instrumental air flow uses tubing with an outer diameter of 6mm, while the vapor and coolant water flows through 8mm tubing. Note that officially the tubing is only made for pneumatic purposes, but it works for water flow as well.

The parts needed for tubing are listed in table 4.

4.3 Bolts and nuts

The minimal requirement of bolts and nuts for the setup is shown in table 5. Note that the optical posts assembly parts to support the setup are not included!

4.4 Other parts

The electrowetting connections are made using spring-loaded contacts in the sample holders. Possible contacts are: Contact, Connector, SMT, Spring Loaded Pin, Point, 2 A, 7.3 mm from Harwin, partnumber: P70-1030045R, Farnell ordercode: 2664913.

Holes in the chamber that are not in use can be filled using rubbers stoppers. Suggested are tapered silicone/rubber stoppers with a diameter of 3 to 3.3mm. Search Ali-express for: 'T type solid High Temp Tapered end ensures a snug fit silicone rubber stopper seal plug tubes 3MM 3.3mm 3 3.0 3.1 1/8" MM hole' (if still available).

For proper sealing of the sample, use generic silicone or something similar.

To glue the plexiglas coolant-window into the condensation chamber use texttt3M Scotch-Weld DP8005 Liquid Acrylic Adhesive glue.

4.5 Electronics

The electronic devices used are listed in table 6.

Part name	Usage	Partnumber Legris	Ordercod Farnell	e Amount needed
Pneumatic Fitting, Female, El- bow, 8 mm, 13.5 mm, G1/4	Tube fitting for vapor and coolant inlet and outlet	3199 08 13	4253085	4
Pneumatic Fitting, Male, BSP, Straight Adaptor, 8 mm, 8 mm, G1/4	Tube fitting for cooler.	3101 08 13	4251787	2
Pneumatic Fitting, Equal, El- bow, 8 mm, 13.5 mm	Optional elbow for easier tube management.	3102 08 00	4251908	3 (op- tional)
Pneumatic Fitting, D8, Y Adap- tor, 8 mm, 13.5 mm	Y-adapter for merging vaport flow with Nitrogen and splitting vapor/Nitrogen for double cham- ber entrance.	3140 08 00	4252690	2
Pneumatic Fitting, Equal, Tee, 8 mm, 13.5 mm	Tee for drain channel.	3104 08 00	4251969	1
Pneumatic Fitting, Unequal Union, Straight Adaptor, 8 mm, 13.5 mm	Unequal join for Nitrogen inlet.	3106 06 08	4252044	2
Pneumatic Fitting, D6, Y Adap- tor, 6 mm, 10.5 mm	Y-adapter for splitting instru- mental air to coolant window drying and dynamic collection funnel.	3140 06 00	4252676	1
Pneumatic Tubing, Ester, PUR (Polyurethane), Blue, 6 mm, 4 mm, 9 bar, 25 m	Tubing for Nitrogen and instru- mental air.	1025U06 04	4251520	1
Pneumatic Tubing, Semi Rigid, Nylon (Polyamide), Green, 8 mm, 6 mm, 25 bar, 25 m	Tubing for coolant and vapor. Tubing is officially only pneu- matic, but works fine for fluids.	1025U08 02	1454053	1
N/A	3-way valve with 8mm pneu- matic tube connections for main vapor control: bypass or cham- ber.	N/A	N/A	1
N/A	2-way valve with 8mm pneu- matic tube connections for drain- ing condensate.	N/A	N/A	1
N/A	2-way valve with 6mm pneu- matic tube connections for Nitro- gen and instrumental air control.	N/A	N/A	2

Table 4


 $(a)\ {\tt Main\ chamber.SLDPRT}$

Figure 28







(a) pcbmount_v9.stl



(c) sampleholder_funnel_mirrored_v15.stl







(f) tubeconverter_v9.stl

sampleholder_funnel_v12.stl

Figure 30: Parts.

Location of bolts	Standard length	Diameter (M) [mm]	Amount	Type of
Location of boits	[mm]		needed	nut
Sample holder	35	4	8-12	WN
Chamber frame	15	3	4	Ν
Chamber bottom	20	3	2	WN/N
Light source	15	3	3	WN
Light source controller	15	3	4	N
PCB mount	10	3	2	Ν
Chamber front handle	15	3	2	N
Lightsource to Thorlabs RA90	10	6 (1/4"-20 Thread)	4-6	none
Chamber to Thorlabs RA90	15	6 (1/4"-20 Thread)	4-8	none
Collection box	20	6 (1/4"-20 Thread)	4	N

Table 5: Minimal required RVS bolts and nuts for the setup (excluding the supporting optical posts assembly). N=nut, WN=wing nut.

Device

Brand

Hot plate Aquarium pump Flow meter Thermistors Thermistors Humidity sensor Water cooler CameraZoom lens Light source Function generator Amplifier Current source Balance

RCT Basic, IKA labortechnik 0886-air-550R-plus, Sera AWM5101VN flowmeter, Honeywell TCS651m AmsTECHNOLOGIES Thorlabs TSP-TH Thorlabs TSP01 Haake-F3-K, Thermo Fisher Scientific Point Grey, FL3-U3 Z125D-CH12, EHD $\rm MB\text{-}BL305\text{-}RGB\text{-}24\text{-}Z,\ Metabright}$ Agilent 33220A Trek PZD700A Delta Elektronika Power Supply E 030-1 Nimbus Analytical Balance NBL 254i

Table 6: Electronic devices used.

5 List of files

This documentation comes with a folder with important files, listed below. The actual files can also be found on https://drive.google.com/drive/folders/1kbDgM8KvgmLR2vMjDW3PR6JbP4sZbNCF?usp=sharing (for as long as this is available).

```
+---Labview
       Untitled 2 (SubVI).vi
        temperature_control_TE_v3_harmen-9.vi
       Agilent33120A_sinepulsed_v5.vi
+---Matlab
I.
   create_preview_movies.m
L
    Т
    +---Image analysis
1
           DropDetectionMethod2_29_beta.m
L
   UnitCellSplitting.m
    Т
       1
           border_detection_3.m
    Т
       1
    Т
       1
           centroid_location_function_2.m
           centroidanglecorrection.m
    T
       T
       circfit.m
       Ι
           DropDetectionFunction_9.m
           DropLocationEvolution.m
    Т
       Electrodemap_LocationPlot_2.m
    Ι
           fill_drops.m
    T
    Т
       imrotate2.m
           merge_images.m
       Т
           plotting_results_function.m
    Τ
           SheddedVolumeFunction_1.m
    T
    Т
        Т
       \---Extended image analysis
    T
               DropTrackingVersion2_7.m
               RadialDistributionT_8.m
    RadialDistributionX_2.m
    1
               AWA_rhat_vs_location_1.m
    Т
               \tt CriticalRadius\_Shed\_Time\_v2.m
    T
               DropTacking_Coalescence_Visualization_Version1_1.m
               DropTracking_Coalescence_Version1_9.m
    Т
    Т
      --Temperature analysis
    +-
T.
   1
           Experiment_Temperature_Plots_May2019.m
1
    Т
    \---Balance analysis
T
           Continuous_water_collection_July_2019.m
+-
   -3Dmodel
setup2018_v16_newsampleholders.skp
   +---SolidWorks Assembly
```

SetupAssembly_Aug122019.SLDASM
 +SolidWorks main chamber
Main chamber.SLDPRT
setup2018 v14 solidworks.PDF
setup2018 v14 solidworks.SLDDRW
setup2018 v14 solidworks drawing v2 PDF
setup2018 v14 solidworks drawing v2.ipg
/ \Parts for printing
sampleholder_funnel_v15.stl
sampleholder_funnel_mirrored_v15.stl
chambertop_v12.stl
handle_v12.stl
frame_v12.stl
bottomfiller_v12.stl
sampleholder_flat_v12.stl
collectionbox_arms_v10.stl
collectionbox_v10.stl
lightsource_mainframe_v9.stl
lightsource_aperture_v9.stl
lightsource_controllermount_v9.stl
tubeconverter_v9.stl
pcbmount_v9.stl
ARCA_sampleholder_part1_v1.stl
ARCA_sampleholder_part2_v1.stl
ARCA_sampleholder_part3_v1.stl
sampleholder_funnel_v12.stl
\png
ARCA_sampleholder_part2_v1.png
ARCA_sampleholder_part1_v1.png
ARCA_sampleholder_part3_v1.png
bottomfiller_v12.png
chambertop_v12.png
<pre>collectionbox_arms_v10.png</pre>
<pre>collectionbox_v10.png</pre>
frame_v12.png
handle_v12.png
lightsource_aperture_v9.png
<pre>lightsource_controllermount_v9.png</pre>
lightsource_mainframe_v9.png
pcbmount_v9.png
sampleholder_flat_v12.png
<pre>sampleholder_funnel_mirrored_v15.png</pre>
sampleholder_funnel_v12.png
<pre>sampleholder_funnel_v15.png</pre>
tubeconverter_v9.png

41

\---Other
 Logbook_template_Apr162019.docx
 Logbook_template_Aug022019.docx

References

- G. Casquillas, F. Bertholle, E. Terriac, and M. Le Berre, "PDMS thickness vs spin-coating speed," *Elveflow*, vol. 1, pp. 2–3, 2005.
- [2] D. Baratian, R. Dey, H. Hoek, D. Van Den Ende, and F. Mugele, "Breath Figures under Electrowetting: Electrically Controlled Evolution of Drop Condensation Patterns," *Physical Review Letters*, vol. 120, no. 21, p. 214502, 2018.

B | CODE FOR MODEL

B.1 MODEL matlab-CODE MODEL VERSION 7

```
1 %% forcebalance_activetransport_version7.m
```

```
% Assumptions:
```

- % force balance droplet halfway on zigzag electrode.
- % 29-10-2019, version 4, 19:40 Numerical solve gives 2 solutions for R, looking at graphs the smallest one seems to be the right one all the time. Possible cause is that
- % the R<electrode (Fe=0) is also included as a solution for large R. Without EW this is the solution! Seems to hold for variable d, where the upper limit is constant: no
- % other force other than Fe depends on d!
- % 30-10-2019, version 5, 17:15 Version 5 solves a miscalculation in calculation of alpha (tan-->atan), includes the spherical cap volume for the gravitational force,
- 11 % simplifies delta_l in earlier stage.
 - % 30-10-2019, version 6, 22:56 Version 6 has rewritten the dependency calculation. It is generalized (only one loop is needed for all variables) and made faster. Boundary
 - % conditions are checked more carefully now to prevent errors.
 - % 11-11-2019, version 6, 11:47 Version 7 replaces the net-force (free fall) with a dissipation force (bulk + CL) based on [Mannetje2011].
- 16 % If model only gives one solution (does not include fe=0), than that is because the condition [..&..] is not solved. condition [..|..] is solved though! cannot be done automatically with vpasolve.

%% Initialization

```
clear all, clc
```

```
21 try, clf(1), clf(2), clf(3), clf(4), end
```

```
%%
```

```
syms x y R d a b rho g Dcosthetazero beta U t CA gamma alpha2 mu xi %CA in
degrees, rest in SI units, Dcosthetazero unity
```

%% Settings

```
26
```

```
analyze_graphs = 1; %solve force balance for R with all parameters
below. Display results visually in graphs.
R_range = [1,300].*1e-6; %range of R to plot the force plots
for
R_value_um = 115; %for fplot of circ and line
```

```
31
                           0; %this mode on/off
   analyze_var =
                           xi; %chose from vars: d a b rho g Dcosthetazero
       var_name =
           beta U t CA gamma alpha2
       var_range =
                           [61:.1:71]*1e-3;
           %Dcosthetazero: [.1:.002:.35 .355:.005:.8]; %216val
                           [0:100:1400 1410:10:2300 2310:100:3500
           %d:
36
               3750:250:12000]*1e-6; %151val
           %a:
                           [0:3:75 75:110 113:3:210 220:15:810]*1e-6; %135val
                           [0:5:600]*1e-6; %121val
           %h:
           %U:
                           0:165; %166val
                           [.4:.01:2 2.1:.1:10]; %241val
           %t:
           %xi:
                           [61:.1:71]*1e-3; %100val
41
                           [0.000297 0.00056 0.0019 0.004 0.001 0.00212
           %mu:
               .0562];Hexane %Methanol IPA blood water milk olive_oil %7val:
                increase vpasolve_R_range
                           linspace(0.000297,.0562,200) %hexane to olive_oil
           %mu:
                200val: increase vpasolve_R_range
   analyze_bruteforce =
                           0; %bruteforce large range of a,b,d values to find
        an optimum
       brute_values =
                           250; %number of values to try per var %15 for 4
46
           parameters takes 83min (depends on how many actual solutions there
            actually are)
       brute_range_d =
                           linspace(10e-6,3000e-6,2*brute_values);
       brute_range_a =
                           linspace(10e-6,1500e-6,brute_values);
                           [5 10 25 50]*1e-6; %linspace(0,400e-6,brute_values
       brute_range_b =
           ):
       brute_parpoolsize = 8; %number of parpool workers for bruteforce. If
           one is already open, this function does not work.
51
   % model settings
   vpasolve_R_range =
                           [0 2000e-6]; %search range for critical radius
   boundary_conditions =
                           [R>0, R<d/2]; %extra boundary conditions besides</pre>
       delta_l_condition. In current mode only variables [a,b,d] are allowed
       to be in BC
56 %% Estimated worktime bruteforcing
   if analyze_bruteforce
       %optionspersec = 10.14; % poolsize of 16. 11835s for 120000 options
           (44.1% solutions) => 10.14 options/s (1-11-2019)
       optionspersec = 4.1099; % poolsize of 8. 19198s for 120000 options
           (52.8% solutions) (13-11-2019)
       brute_options = length(brute_range_d)*length(brute_range_a)*length(
           brute_range_b);
       disp(['Total bruteforce options are: ' num2str(brute_options) '. This
61
           wil take an estimated ' num2str(round(brute_options/optionspersec
           /3600,1)) ' hours, based on 13-11-2019 data for a poolsize of 8.'
           1)
       disp(['Average resolutions: d = ' num2str(mean(diff(brute_range_d)*1e6
           )) 'um, a = ' num2str(mean(diff(brute_range_a)*1e6)) 'um, b = '
           num2str(mean(diff(brute_range_b)*1e6)) 'um.'])
```

```
%% Constants
66
   % Situation sketch
   % |..a..|b b|..a..|
   % ___
   %\
         1.
           1 .
71 %
          / \
   % \
                    1
   % \ | R | /
                        d
         \setminus   /  / /
   %
         \setminus | | | /
   %
76 %
          | | | | /
   % electrode dimensions, see sketch above
   a_value =
                           100e-6; %[m] half of maximum electrode width minus
        gap width
                           25e-6; %[m] half of the minimal gap width
   b_value =
s_1 d_value =
                           1500e-6; %[m] electrode height
   % electrode attenuation
   U_value =
                           150; %[V] For active transport top layer 125V,
       bottom layer, 150V;
   t_value =
                           2; %[s] Attenuation time of each electrode layer
86
   % surface parameters
   Dcosthetazero_value =
                          .206; %.225 sept measurements, assume new Teflon
       surface, old=.2
   CA_value =
                           115; %[deg] Contact angle of droplet on surface.
       CA should be >90deg.
   alpha2_value =
                           1; %constant, might me little less than 1. See
       Dey2018SUP
91 beta_value =
                           8.5e-6; %[F/(N m)], see Dey2018SUP, approximation.
         epsilon_value = 2.1*8.8541878128*1e-12; %Par C
   %
         dithickness = 2286*10^(-9); %m, thickness (2019029) ;%thickness
   %
       dielectric, assume top layer
         beta_value = epsilon_value/(8*dithickness*gamma_value);
   %
% physical constants
   rho_value =
                           997; %[kg/m^3] Water density
                           -9.81; %[m/s<sup>2</sup>] Gravitational acceleration
   q_value =
   gamma_value =
                           72.8e-3; %[N/m] surface tension water-air
   mu_value =
                           le-3; %[Pa s] Viscosity of water
                           71e-3; %+-10 [Pa s] Contact line friction
101 xi_value =
       coefficient [Mannetje2011]
   %% Determine 1: approximately the horizontal projection of dl
   % Define two equations and intersect the two. Difference in x is l.
106 eq_circle = x^2 + (y+100e-6)^2 == R^2;
   alpha = atan(a/d);
```

end

```
s = b/tan(alpha);
   eq_line = y = -d/a * x - (d/2 + s);
111
   assume(R>0)
   eq_circle = simplify(eq_circle);
   warning off
   y_circle = solve(eq_circle,y,'Real',true); %x^2 <= R^2</pre>
116 warning on
   y_line = solve(eq_line,y,'Real',true);
   delta_l_condition = R>a/2+b;
   delta_l_condition_false = R<=a/2+b;</pre>
121 R_delta_l_condition = a/2+b;
   x_intersect = solve(y_circle == y_line,x);
   y_intersect = subs(y_line,x,x_intersect);
126 delta_l = abs(x_intersect(1)-x_intersect(2)); %exact delta l in [R,a,d,s]
   delta_l = simplify(delta_l);
   %CONDITION: if R>a/2+b --> delta_l=0
   boundary_conditions = [boundary_conditions, delta_l_condition];
131
   x_intersect_R = double(subs(x_intersect,[R d a b],[R_value_um d_value_um
       a_value_um b_value_um]));
       y_intersect_R = double(subs(y_intersect,[R d a b],[R_value_um
            d_value_um a_value_um b_value_um]));
       abs(x_intersect_R(1) - x_intersect_R(2)),1)
136 % Forces
   % Weight
   V = 0.5*(4/3)*pi*R^3; %[m^3] volume of hemisphere
   Vcap = V+(1/3)*pi*R^3*(2-3*sind(CA)+sind(CA)^3); %[m^3] volume of
       spherical cap with CA>90degreesx = -R/2;
141 F_g = Vcap*rho*g;
   % Pinning force
   F_p = 2*R*gamma*(Dcosthetazero-alpha2*beta*U^2);
146 % Electric force
   %factor 2 for both overlaps
   %CONDITION: if R>a/2+b --> delta_l=0
   %F_e = 2*beta*gamma*U^2*-delta_l;
   F_e = piecewise(delta_l_condition, 2*beta*gamma*U^2*-delta_l,0); %0
       otherwise
151
   % Dissipation forces of drop sliding down [Mannetje2011]
   deltay = R; %[m] Distance droplet has to travel in time t
   % A = pi * R^{2};
156 % h = Vcap/(rho*A);
```

```
v_crit = deltay/t; %We assume here that a is ZERO: droplet does not grow,
       thus all forces are constant when drop starts sliding: no net
       acceleration, thus constant velocity!
   % F_bulk = 4*mu*A*velocity/h;
   % F_cl = 8*R*xi*velocity;
   % F_d = F_bulk+F_cl;
161 F_d = v_crit*R*(6*pi*mu*rho+8*xi); %F_d is here the MAXIMUM dissipation we
        get: the dissipation at critical (escape) velocity!
   % % Net force to clear the gap
   % deltay = -R/2; %[m] Distance droplet has to travel in time t
166 % F_n = Vcap*rho*(2*deltay)/(t^2);
   % Balance: NOT ABLE TO SOLVE
   force_balance = simplify(F_g+F_p+F_e+F_d == 0);
171
   %% analyze_var
                           Numerically solve force balance for R, var_name
   if analyze_var
       var_all = [Dcosthetazero,alpha2,d,U,t,rho,g,gamma,beta,CA,a,b,mu,xi];
           %all variables in the problem
       var_all_subs = [Dcosthetazero_value,alpha2_value,d_value,U_value,
176
           t_value,rho_value,g_value,gamma_value,beta_value,CA_value,a_value,
           b_value,mu_value,xi_value];
       var_constants = var_all(var_all ~= var_name); %exclude the variable
           from list
       var_constants_subs = var_all_subs(var_all ~= var_name);
       data = struct();
       data.var_range = var_range;
181
       data.var_name = var_name; %the actual variable here
       data.R_crit = [];
       %save all constants as well
       data.constants = var_constants:
       data.constants_subs = var_constants_subs;
186
       if ~ismember(var_name,[a,b,d]) %substitute boundary conditions and
           check validity of them outside forloop: saves 0.1s per iteration
           boundary_conditions_subs = subs(boundary_conditions,[a,b,d],[
               a_value,b_value,d_value]);
           warning('off')
           if isempty(solve([boundary_conditions_subs],R))
191
               disp('Boundary conditions not valid. Check boundary_conditions
                    and delta_l_condition_subs'), return %check validity of
                   boundary conditions
           end
           warning('on')
           disp(['Using the boundary conditions: ' [strjoin(arrayfun(@char, [
               boundary_conditions_subs], 'uniform', 0),', ')] '.'])
           assume([boundary_conditions_subs]) %set boundary conditions for R
196
       end
```

	<pre>force_balance_subs2 = simplify(subs(force_balance,var_constants, var_constants_subs)); %substitute all constants. force_balance_subs2(R,var_name) is left</pre>
201	<pre>for i=1:size(var_range,2) var_range_i = var_range(i); disp(['Solving force balance for R for ' char(var_name) ' = ' num2str(var_range_i) '']) R_crit_i = []; %iteration R_crit</pre>
206	<pre>%set boundary conditions aka assumptions if ismember(var_name,[a,b,d]) %substitute boundary condition and check validity of them inside for loop if boundary conditions change each iteration if isequal(var_name,d), d_value = var_range_i; end if isequal(var_name,a), a_value = var_range_i; end if isequal(var_name,b), b_value = var_range_i; end if isequal(var_name,b), b_value = var_range_i; end</pre>
211	<pre>boundary_conditions_subs = subs(boundary_conditions,[a,b,d],[a_value,b_value,d_value]); if isempty(solve([boundary_conditions_subs],R)) %check validity of boundary conditions disp(['Boundary conditions not valid for current value of</pre>
216	<pre>disp(['Using the boundary conditions: ' [strjoin(arrayfun(@char, [boundary_conditions_subs], 'uniform', 0),', ')] '. ']) assume([boundary_conditions_subs]) %set boundary conditions for R end</pre>
	<pre>force_balance_subs3 = simplify(subs(force_balance_subs2,[var_name],[var_range_i])); %substitute, simplify and split equations in children (multiple solutions). force_balance_subs3(R) is left</pre>
221	<pre>if isequal(char(force_balance_subs3),'FALSE') %simplify returns FALSE: no solutions for boundary conditions disp(['No solution for ' char(var_name) ' = ' num2str(var_range_i) ' due to boundary conditions.']) data.R_crit{i} = NaN; continue end</pre>
<u>~</u> 20	<pre>for j = 1:size(force_balance_subs3,2) %solve for all subequations if strfind(char(force_balance_subs3(j)),' ') %equation has parameters in it, children required. If always children, it splits everywhere (at every > + - *,etc) force_balance_subs4 = children(force_balance_subs3(j)); else</pre>
231	<pre>force_balance_subs4 = force_balance_subs3(j); end</pre>

```
for k = 1:size(force_balance_subs4,2)
                    R_crit_k = vpasolve([force_balance_subs4(k)],R,
                        vpasolve_R_range); %note: if force_balance_subs3(j)
                        has conditions in it (&), than it is NOT solved
                    if ~isempty(R_crit_k) && R_crit_k>0 %if there are
236
                        solutions found
                        R_crit_i(end+1) = double(R_crit_k);
                        disp(['A solution for ' char(var_name) ' = ' num2str(
                            var_range_i) ' is R = ' num2str(round(double(
                            R_crit_k*1e6),1)) 'um.'])
                    end
                end
            end
241
            if isempty(R_crit_i)
                R_crit_i = NaN;
                disp(['No solution for ' char(var_name) ' = ' num2str(
                    var_range_i) '.'])
            end
246
            data.R_crit{i} = R_crit_i; %save to permanent data structure
            disp(newline)
       end
       clear i j R_crit_i R_crit_j
       assume(R,'clear') %unset boundary conditions
251
       % Plotting
       figure(1), clf(1), hold on
       for i=1:size(data.R_crit,2)
            plot(data.var_range(i),data.R_crit{i},'.','Markersize',25,'Color'
256
                ,[0.85 0.325 0.098])
       end
       xlabel(char(var_name),'interpreter','latex')
       ylabel('Critical radius [$\mu$ m]','interpreter','latex')
   end
261
   %% analyze_bruteforce Brute force for all parameters
   % The code used for bruteforcing is based on the normal numerical solving.
        The difference is the iteration over all possibilities of 3 values.
       The i-loop is replaced
   % by a new m-loop of the bruteforce table. Subsitution of constants is
       done before iteration. Boundary condition setting is straighforward
       and done ever iteration.
   % The internal loops j and k are the same as above. The disp-messages have
        been updated.
266
   if analyze_bruteforce
       brute_table = num2cell(transpose(combvec(brute_range_d,brute_range_a,
           brute_range_b)));
       var_all = [Dcosthetazero,alpha2,d,U,t,rho,g,gamma,beta,CA,a,b,mu,xi];
           %all variables in the problem
```

150 | CODE FOR MODEL

```
var_all_subs = [Dcosthetazero_value,alpha2_value,d_value,U_value,
271
           t_value,rho_value,g_value,gamma_value,beta_value,CA_value,a_value,
           b_value,mu_value,xi_value];
       var_constants = var_all(~ismember(var_all,[a,b,d])); %exclude the
           variable from list
       var_constants_subs = var_all_subs(~ismember(var_all,[a,b,d]));
       force_balance_subs2 = simplify(subs(force_balance,var_constants,
           var_constants_subs)); %substitute all constants.
           force_balance_subs2(R,a,b,d) is left
276
       brute_rcrit_temp = {};
       d_values = cell2mat(brute_table(:,1));
       a_values = cell2mat(brute_table(:,2));
       b_values = cell2mat(brute_table(:,3));
       tic
281
       p = gcp('nocreate'); % If no pool, do not create new one.
       if isempty(p)
           parpool(brute_parpoolsize)
       end
286
       parfor m=1:brute_options
           d_value = d_values(m);
           a_value = a_values(m);
           b_value = b_values(m);
291
           R_crit_m = []; %in case we have multiple solutions
           assume(R,'clear') %unset boundary conditions
           continuecode = 1;
296
           disp([num2str(m) ' d = ' num2str(d_value) ' | a = ' num2str(
               a_value) ' | d = ' num2str(b_value) '
                                                       Started ...'])
           %set boundary conditions aka assumptions
           boundary_conditions_subs = subs(boundary_conditions,[a,b,d],[
               a_value,b_value,d_value]);
           if isempty(solve([boundary_conditions_subs],R)) %check validity of
301
                boundary conditions
                disp([num2str(m) ' d = ' num2str(d_value) ' | a = ' num2str(
                   a_value) ' | d = ' num2str(b_value) ' R = NaN
                                                                           BCs
                    not valid.'])
               %brute_table{m,4}=NaN;
                continuecode = 0;
               R_crit_m = NaN;
               %brute_rcrit_temp{end+1} = {m,NaN}; continue
306
           end
           if continuecode
                disp([num2str(m) ' d = ' num2str(d_value) ' | a = ' num2str(
                   a_value) ' | d = ' num2str(b_value) ' BCs set to: ' [
                   strjoin(arrayfun(@char, [boundary_conditions_subs], '
                   uniform', 0),', ')] '.'])
```

```
assume([boundary_conditions_subs]) %set boundary conditions
311
                   for R
               force_balance_subs3 = simplify(subs(force_balance_subs2,[a,b,d
                   ],[a_value,b_value,d_value])); %substitute, simplify and
                   split equations in children (multiple solutions).
                   force_balance_subs3(R) is left
               if isequal(char(force_balance_subs3),'FALSE') %simplify
                   returns FALSE: no solutions for boundary conditions
                   disp([num2str(m) ' d = ' num2str(d_value) ' | a = '
316
                       num2str(a_value) ' | d = ' num2str(b_value) '
                                                                        R =
                                BCs not valid after simplify.'])
                       NaN
                   %brute_table{m,4}=NaN;
                   continuecode = 0;
                   R_crit_m = NaN;
                   %brute_rcrit_temp{end+1} = {m,NaN}; continue
               end
321
           end
           if continuecode
               for j = 1:size(force_balance_subs3,2) %solve for all
                   subequations
                   if strfind(char(force_balance_subs3(j)),'|') %equation has
326
                        parameters in it, children required. If always
                       children, it splits everywhere (at every > + - *,etc)
                        . . .
                        force_balance_subs4 = children(force_balance_subs3(j))
                            ;
                   else
                        force_balance_subs4 = force_balance_subs3(j);
                   end
331
                   for k = 1:size(force_balance_subs4,2)
                       R_crit_k = vpasolve([force_balance_subs4(k)], R,
                            vpasolve_R_range); %note: if force_balance_subs3(j
                            ) has conditions in it (&), than it is NOT solved
                       if ~isempty(R_crit_k) && R_crit_k>0 %if there are
                            solutions found
                            R_crit_m(end+1) = double(R_crit_k);
                            disp([num2str(m) ' d = ' num2str(d_value) ' | a
336
                                = ' num2str(a_value) ' | d = ' num2str(b_value
                                ) ' R = ' num2str(round(double(R_crit_k*1
                                e6),1)) 'um
                                                 SOLUTION FOUND.'])
                       end
                   end
               end
               if isempty(R_crit_m)
341
                   R_crit_m = NaN;
                   disp([num2str(m) ' d = ' num2str(d_value) ' | a = '
                       num2str(a_value) ' | d = ' num2str(b_value) ' R =
                       NaN
                                No soltutions.'])
```

```
end
           end
346
           brute_table{m,4} = R_crit_m; %save to permanent data structure
           %brute_rcrit_temp{end+1} = {m,R_crit_m};
       end
       assume(R,'clear') %unset boundary conditions
351
       toc
   end
   %% analyze_graphs
                            Numerically solve force balance for R
356 %This block is a modified version of the "Numerically solve force balance
       for R,var_name", but with no var_name. Only constants used.
   if analyze_graphs
       var_all = [Dcosthetazero,alpha2,d,U,t,rho,g,gamma,beta,CA,a,b,mu,xi];
           %all variables in the problem
       var_all_subs = [Dcosthetazero_value,alpha2_value,d_value,U_value,
           t_value, rho_value, g_value, gamma_value, beta_value, CA_value, a_value,
           b_value,mu_value,xi_value];
       var_constants = var_all; %exclude the variable from list
361
       var_constants_subs = var_all_subs;
       data = struct();
       data.var_range = NaN;
       data.var_name = NaN; %the actual variable here
366
       data.R_crit = [];
       %save all constants as well
       data.constants = var_constants;
       data.constants_subs = var_constants_subs;
371
       %set boundary conditions aka assumptions
       boundary_conditions_subs = subs(boundary_conditions,[a,b,d],[a_value,
           b_value,d_value]);
       warning('off')
       if isempty(solve([boundary_conditions_subs],R))
           disp('Boundary conditions not valid. Check boundary_conditions and
376
                delta_l_condition_subs'), return %check validity of boundary
               conditions
       end
       warning('on')
       disp(['Using the boundary conditions: ' [strjoin(arrayfun(@char, [
           boundary_conditions_subs], 'uniform', 0),', ')] '.'])
       assume([boundary_conditions_subs]) %set boundary conditions for R
381
       %force_balance_subs3 = simplify(subs(force_balance,var_constants,
           var_constants_subs)); %substitute all constants.
           force_balance_subs2(R,var_name) is left
       force_balance_subs3 = subs(force_balance,var_constants,
           var_constants_subs); %substitute all constants.
           force_balance_subs2(R,var_name) is left
```

```
disp(['Solving force balance for R ...'])
       R_crit_i = []; %iteration R_crit
386
       if isequal(char(force_balance_subs3),'FALSE') %simplify returns FALSE:
            no solutions for boundary conditions
            disp(['No solution due to boundary conditions.'])
            data.R_crit = NaN; return
       end
391
        for j = 1:size(force_balance_subs3,2) %solve for all subequations
            if strfind(char(force_balance_subs3(j)),'|') %equation has
                parameters in it, children required. If always children, it
                splits everywhere (at every > + - *,etc) ...
                force_balance_subs4 = children(force_balance_subs3(j));
            else
396
                force_balance_subs4 = force_balance_subs3(j);
            end
            for k = 1:size(force_balance_subs4,2)
                R_crit_k = vpasolve([force_balance_subs4(k)],R,
401
                    vpasolve_R_range); %note: if force_balance_subs3(j) has
                    conditions in it (&), than it is NOT solved
                if ~isempty(R_crit_k) && R_crit_k>0 %if there are solutions
                    found
                    R_crit_i(end+1) = double(R_crit_k);
                    disp(['A solution is R = ' num2str(round(double(R_crit_k*1
                        e6),1)) 'um.'])
                end
            end
406
       end
       if isempty(R_crit_i)
            R_crit_i = NaN;
            disp(['No solution.'])
411
       end
       data.R_crit = R_crit_i; %save to permanent data structure
       disp(newline)
       clear i j R_crit_i
416
       assume(R,'clear') %unset boundary conditions
       %% Substitute values for seperate forces and delta_l: needed for
            plotting
       % Seperate forces: as function of R
421
       F_g_subs = subs(F_g, \ldots)
            [d,a,b,rho,g,Dcosthetazero,alpha2,beta,U,t,CA,gamma], ...
            [d_value,a_value,b_value,rho_value,g_value,Dcosthetazero_value,
                alpha2_value,beta_value,U_value,t_value,CA_value,gamma_value])
                :
       F_g_subs = simplify(F_g_subs);
       F_p_subs = subs(F_p, \ldots)
426
            [d,a,b,rho,g,Dcosthetazero,alpha2,beta,U,t,CA,gamma], ...
```

	[d_value,a_value,b_value,rho_value,g_value,Dcosthetazero_value, alpha2_value,beta_value,U_value,t_value,CA_value,gamma_value])
431	<pre>, F_p_subs = simplify(F_p_subs); F_e_subs = subs(F_e, [d,a,b,rho,g,Dcosthetazero,alpha2,beta,U,t,CA,gamma], [d_value,a_value,b_value,rho_value,g_value,Dcosthetazero_value,</pre>
	; F_e_subs = simplify(F_e_subs); F_d_subs = subs(F_d, [d,a,b,rho,g,Dcosthetazero,alpha2,beta,U,t,CA,gamma,mu,xi],
436	<pre>[d_value,a_value,b_value,rho_value,g_value,Dcosthetazero_value,</pre>
441	<pre>% Evaluate delta_l delta_l_subs = simplify(subs(delta_l,[a,d,b],[a_value,d_value,b_value]));</pre>
	<pre>delta_l_condition_subs = simplify(subs(delta_l_condition,[a,d,b],[</pre>
	<pre>R_delta_l_condition_subs = simplify(subs(R_delta_l_condition,[a,d,b],[a_value,d_value,b_value]));</pre>
	<pre>delta_l_condition_false_subs = simplify(subs(delta_l_condition_false,[</pre>
446	%% Plot forces
	<pre>figure(1) clf(1) bald.en</pre>
451	<pre>notd on fplot(F_g_subs,R_range,'-','Linewidth',3) fplot(E_n_subs_R_range_'-','Linewidth',3)</pre>
	<pre>fplot(F_e_subs,R_range, '-', 'Linewidth',3) fplot(F_d_subs,R_range, ':', 'Linewidth',3) legend('\$F_g\$', '\$F_p\$', '\$F_e\$', '\$F_d^{max}\$', 'Location', 'southwest')</pre>
456	<pre>xlabel('Droplet radius [m]') ylabel('Force on droplet [N]') grid on</pre>
	<pre>xlim([R_range(1) R_range(2)])</pre>
461	% figure(2) clf(2) hold on
	<pre>R_range_boundary = double(R_delta_l_condition_subs);</pre>
400	<pre>f3 = fplot(0,R_range,'-','Linewidth',3,'Color',[0.9290 0.6940 0.1250]) ;</pre>
	<pre>f2 = fplot((-F_d_subs),R_range,':','Linewidth',3,'Color',[0.8500 0.3250 0.0980]);</pre>

```
assume(delta_l_condition_false_subs)
        f1 = fplot(simplify(F_e_subs)+F_g_subs+F_p_subs,[R_range(1)
471
           R_range_boundary], '-', 'Linewidth',3, 'Color', [0 0.4470 0.7410]);
        assume(R,'clear'), assume(delta_l_condition_subs)
        fplot(simplify(F_e_subs)+F_g_subs+F_p_subs,[R_range_boundary R_range
            (2)],'-','Linewidth',3,'Color',[0 0.4470 0.7410]);
       F_crit = simplify(subs(simplify(F_e_subs)+F_g_subs+F_p_subs,R,R_crit_k
476
            ));
        f4 = plot(R_crit_k,F_crit,'x','MarkerSize',20,'Color',[0.8500 0.3250
           0.0980]);
       R_crito = vpasolve(simplify(simplify(F_e_subs)+F_g_subs+F_p_subs) ==
           0,R,vpasolve_R_range);
        f5 = plot(R_crito,0,'x','MarkerSize',20,'Color',[0.9290 0.6940
            0.1250]);
481
       xlabel('Droplet radius [m]')
       ylabel('Force on droplet [N]')
        legend([f1 f2 f3 f4 f5],{'$F_g+F_p+F_e$','$F_d^{max}$','$F_d=0$',['$R_
            {crit}=$ ' num2str(round(double(R_crit_k)*1e6,1)) '$\mu$m'],['$R_o
            =$ ' num2str(round(double(R_crito)*1e6,1)) '$\mu$m']},'location','
            southwest'
        grid on
        assume(R,'clear')
486
       xlim([R_range(1) R_range(2)])
        %
        figure(3)
        clf(3)
491
       hold on
        fplot((delta_l_subs), R_range, '-', 'Linewidth', 3)
       xlabel('Droplet radius [m]')
       ylabel('$\Delta_l$ [m]')
        grid on
496
       xlim([R_range(1) R_range(2)])
       %% Fplot: show intersect circle and electrode
       d_value_um = 1500; %[m] electrode height
501
        a_value_um = 100; %[m] half of maximum electrode width minus gap width
        b_value_um = 25; %[m] half of the minimal gap width
       y_circle_R = subs(y_circle,R,R_value_um)
       y_line_R = subs(y_line,[R d a b],[R_value_um d_value_um a_value_um
            b_value_um]);
       y_line_R_flip = subs(y_line,[R d a b x],[-R_value_um d_value_um
506
            a_value_um b_value_um -x]);
       x_intersect_R = double(subs(x_intersect,[R d a b],[R_value_um
           d_value_um a_value_um b_value_um]));
       y_intersect_R = double(subs(y_intersect,[R d a b],[R_value_um
            d_value_um a_value_um b_value_um]));
```

```
figure(4)
       clf(4)
511
       hold on
       plotrange = [-800 800];
       fplot(y_line_R,[-a_value_um-b_value_um -b_value_um],'LineWidth',3,'
           Color',[0.85 0.325 0.098])
        fplot(y_line_R_flip,[b_value_um a_value_um+b_value_um],'LineWidth',3,'
           Color',[0.85 0.325 0.098])
       fplot(y_circle_R, 'LineWidth', 3, 'Color', [0 0.447 0.741])
516
       ints = plot(x_intersect_R,y_intersect_R,'x','MarkerSize',20,'Color'
            ,[.466 .674 .188]);
       xlabel('x [$\mu$m]')
       ylabel('y [$\mu$m]')
       tickz = 100;
       xticks(plotrange(1):tickz:plotrange(2))
521
       yticks(plotrange(1):tickz:plotrange(2))
       xlim([plotrange])
       ylim([plotrange])
       grid on
526
        format shortg %disp only significant numbers
       legend(ints,['$\Delta_l$ = ' num2str(round(abs(x_intersect_R(1)-
           x_intersect_R(2)),1)) ' $\mu$m'])
   end
```

B.2 MODEL matlab-code processing Brute-force DATA VERSION 4

```
1 %% Bruteforce_processing_version3.m
   % Created on: 14-11-2019: merged and improved version of codes
       plotting_bruteforce.m and bruteforce_equation.m
   % This script does the following:
   % 1. It finds the minimum Rcrit with corresponding a value for each value
      of d for each value of b.
6 % 2. Plots this as d vs a.
   % 3. Fits a second order polynomial to each line.
   % 4. Find the relationship between those equation by fitting (and plotting
      ) each coefficient with either a linear
           or 2nd order polynomial. So far it seemed best for linear for p(1)
        and p(2) and 2nd order for p(3).
   % 5. Plots this equation with a rounded (approximate) version for several
      values of b.
11 % 6. Calculate meshgrid of bruteforce data and plot contour plot for one
      value of b
   % 7. Plots equation for value of b shown in contour plot.
  % Changelog
   % 14-11-2019: script created
16 % 15-11-2019; version 2: swapped d and a. Generic optimization equation is
        now d(a) instead of a(d).
   % 15-11-2019; version 3: too much complications with swapping in version2.
        Now just solving d(a) for a.
   % 18-11-2019; version 4: auto saving data and plotting contour plots for
      all b
   clc
21
   %% Settings
   % Brutetable filename
   filename_mac =
                              '/Volumes/GoogleDrive/My Drive/PCF Master
      Thesis/Model Active Transport/version7/brutetable_20191118_500000val.
      mat';
26 filename_windows =
                               'G:\My Drive\PCF Master Thesis\Model Active
       Transport\version7\brutetable_20191118_500000val.mat';
   % Equation fitting
   analyze_equation =
                              1; %calculate the general equation and plot
      this data?
       coefficient_plots =
                             1; %show the seperate 3 plots of fitting of
           coefficient plots to determine b dependency? Just for visual
          verficication.
31
   plot_equation =
                             1; %plots the equation for the values of b
      below
       equation_plot_b = [5 10 25 50 100]*1e-6;
```

```
% Contourplot
<sub>36</sub> contour_mode =
                               1; %pre-sets for contour plot, see options
       below
                               20; %if mode 3, define number of steps
       contour_steps =
                           % 1: standard z-spacing lines with text
                           % 2: no z-spacing lines
                           % 3: increased z-spacing lines with text
                               0; %show colorscale as log instead of linear
41 contour_logscale =
   %% Load, manipulate and check data
   Save_DateTime = datestr(datetime('now'), 'yyyymmdd_HHMMss');
46 Save_folder = ['PROC_' Save_DateTime];
   try
       load(filename_mac)
       [Files_folder,~,~] = fileparts(filename_mac);
51 catch
       load(filename_windows)
       [Files_folder,~,~] = fileparts(filename_windows);
   end
56 if exist([Files_folder '\' Save_folder],'dir') ~= 7
       mkdir([Files_folder '\' Save_folder])
   end
   % Split brute_table in seperate arrays
61 % Only the minimum value of Rcrit is used. If more solution were found,
       these are not used.
   d_all = cell2mat(brute_table(:,1));
   a_all = cell2mat(brute_table(:,2));
   b_all = cell2mat(brute_table(:,3));
   for i=1:size(brute_table,1), brute_table{i,5}=min(brute_table{i,4}); end
66 rcrit_min = cell2mat(brute_table(:,5));
   b_uniques = unique(b_all);
   %% 1-5: analyze_equation
71 if analyze_equation
       %% 1. Find minimum R and corresponding a for all d values for all b
           values
       \alpha % approach: select 1 value for b --> qo over all d values for that b (x
           -axis) --> find the minimum rcrit for that d
       % and the corresponding a value (y-axis)
76
       if ~exist('r_b_min') %this process is long, if this var already exists
           , this process is likely already done before, than skip
           r_b_min_all = {}; %minimum rcrit for all d values for each value
               of b
           a_b_min_all = {}; %minimum a for all d values for each value of b
```

```
for i=1:length(b_uniques)
81
                b_value = b_uniques(i);
                disp(['Finding mimima for all d-values for b= ' num2str(
                    b_value) ' um ...'])
                d_b = d_all(b_all == b_value); %subset of d where b=b_value:
                    all d values for the current b value.
                a_b = a_all(b_all == b_value);
                r_b = rcrit_min(b_all == b_value);
86
                r_b_min = []; %all rcrit for b=b_value
                a_b_min = []; %corresponding a for all minima
                for j=1:length(d_b) %iterate over all d values (x-axis)
91
                    d_b_value = d_b(j); %subset of d_b where d=d_value
                    a_b_d = a_b(d_b == d_b_value); %subset of a_b where d=
                        d_value
                    r_b_d = r_b(d_b == d_b_value);
                    [r_b_d_min, idx] = min(r_b_d);
96
                    a_b_d_min = a_b_d(idx);
                    r_b_min(j) = r_b_d_min;
                    a_b_min(j) = a_b_d_min;
                end
101
                r_b_min_all{i} = r_b_min;
                a_b_min_all{i} = a_b_min;
            end
       else
            disp(['NOTE: script thinks it already found minima before, as
106
               variable r_b_min already exists, this part is thus skipped!'])
       end %exist('r_b_min')
       %% 2+3. Plot mimima lines d,a and fit 2nd order polynomials
       f_fit = figure('Position', [0 0 1100 700],'Visible','on');
       hold on
111
       legendarray = {};
       col = [0 0.4470 0.7410; 0.8500 0.3250 0.0980; 0.9290 0.6940 0.1250;
           0.4940 0.1840 0.5560]; %parula
       p1_all = [];
       p2_all = [];
116
       p3_all = [];
       for i=1:length(b_uniques)
            plot(d_b*1e6,a_b_min_all{i}*1e6,'.','Markersize',15,'Color',col(i
                ,:))
            legendarray{end+1} = ['b = ' num2str(b_uniques(i)*1e6) ' $\mu m$'
121
               ];
            p = polyfit(d_b,transpose(a_b_min_all{i}),2);
            x = linspace(min(d_b),max(d_b),200); %x for polyfit plot
            f = polyval(p,x);
            plot(x*1e6,f*1e6,'Color',col(i,:))
```

```
%legendarray{end+1} = [' $a = ' num2str(p(1)*1e6) 'd^3+' num2str(p
126
                (2)*1e6) 'd^2+' num2str(p(3)*1e6) 'd+' num2str(p(4)*1e6) '$'
                1;
            legendarray{end+1} = [' $a = ' num2str(p(1)*1e6) 'd^2+' num2str(p
                (2)*1e6) 'd+' num2str(p(3)*1e6) '$' ];
            p1_all(i) = p(1);
           p2_all(i) = p(2);
            p3_all(i) = p(3);
       end
131
       legend(legendarray,'location','northwest')
       xlabel('d [$\mu m$]')
       ylabel('a [$\mu m$]')
       title('d(a) values with lowest $R_{crit}$, p2 fitting')
136
       %Save fit plot
       saveas(f_fit,[Files_folder '\' Save_folder '\fitdata.png'])
       saveas(f_fit,[Files_folder '\' Save_folder '\fitdata.fig'])
141
       %% 4. Find relation between fits as function of b
       warning off %polyfit annoys with warnings
       %p1 pre-factor: linear works well
146
       p1 = polyfit(b_uniques,transpose(p1_all),1);
       if coefficient_plots
            p1_x = linspace(min(b_uniques),max(b_uniques),200);
           p1_f = polyval(p1,p1_x);
           f_p1 = figure;
151
           hold on
           plot(b_uniques,p1_all,'.','Markersize',40)
           plot(p1_x,p1_f)
           xlabel('b [um]')
           ylabel('$p_1$')
156
           %title('Linear fit to p(1)')
           %Save fittings plot
           saveas(f_p1,[Files_folder '\' Save_folder '\p1fit.png'])
            saveas(f_p1,[Files_folder '\' Save_folder '\p1fit.fig'])
       end %coefficient_plots
161
       %p2 pre-factor: linear works well
       p2 = polyfit(b_uniques,transpose(p2_all),1);
       if coefficient_plots
            p2_x = linspace(min(b_uniques), max(b_uniques), 200);
166
           p2_f = polyval(p2, p2_x);
           f_p2 = figure;
           hold on
           plot(b_uniques,p2_all,'.','Markersize',40)
           plot(p2_x,p2_f)
171
           xlabel('b [um]')
           ylabel('$p_2$')
           %title('Linear fit to p(2)')
           %Save fittings plot
```

```
saveas(f_p2,[Files_folder '\' Save_folder '\p2fit.png'])
176
            saveas(f_p2,[Files_folder '\' Save_folder '\p2fit.fig'])
       end %coefficient_plots
       %p3 pre-factor: second order works well
       p3 = polyfit(b_uniques,transpose(p3_all),2);
181
       if coefficient_plots
           p3_x = linspace(min(b_uniques), max(b_uniques), 200);
            p3_f = polyval(p3, p3_x);
            f_p3 = figure;
            hold on
186
            plot(b_uniques,p3_all,'.','Markersize',40)
            plot(p3_x,p3_f)
            xlabel('b [um]')
            ylabel('$p_3$')
            %title('Second order fit to p(3)')
191
           %Save fittings plot
            saveas(f_p3,[Files_folder '\' Save_folder '\p3fit.png'])
            saveas(f_p3,[Files_folder '\' Save_folder '\p3fit.fig'])
       end %coefficient_plots
196
       warning on
       % Come up with final equation
       eq_1 = [num2str(p1(1)) '*b + ' num2str(p1(2))];
       eq_2 = [num2str(p2(1)) '*b + ' num2str(p2(2))];
201
       eq_3 = [num2str(p3(1)) '*b^2 + ' num2str(p3(2)) '*b + ' num2str(p3(3))
           ];
       equation = ['a = (' eq_1 ')*d^2 + (' eq_2 ')*d + (' eq_3 ')'];
       disp(['Full equation found as: ' equation])
       % Convert equation to syms
206
       syms a d b
       eq = str2sym(equation);
       a = rhs(eq);
       assume([b>0, b<100e6])
       assume([d>0, d<3000e6])
211
       assume([a>0, a<3000e6])
       a = simplify(a);
       a_approx = vpa(a,2); %approximates are more than good enough!
       disp(['Approximate form of equation: a = ' char(a_approx)])
216
       %Rewrite as d(a)
       assume(a,'clear')
       aa = a; %we want 'a' in eq, but it's an eq now. so move to aa, and
           redefine a as a sym
       clear a
       syms a
221
       eq2 = a == aa;
       dd = solve(eq2,d);
       dd = simplify(dd);
```

```
dd = dd(1); %NOTE: unable to get one solution with assumptions. Tests
           have shown that solution 1>0 for all cases OR has the smallest
            positive value.
       d_approx = vpa(dd, 2);
226
       disp('Rewritten a(d) to d(a).')
       disp(['Approximate form of equation: d = ' char(d_approx)])
       %% 5. Plot approximate equation for several values of b
231
       if plot_equation
            f_eq = figure('Position', [0 0 900 500],'Visible','on');
           hold on
            for b_value = equation_plot_b
                a_approx_subs = subs(a_approx,b,b_value);
236
                pl1 = fplot(a_approx_subs,[0 3000e-6],'-','Linewidth',3,'Color
                    ',[0.8500 0.3250 0.0980]);
                a_subs = subs(aa,b,b_value);
                pl2 = fplot(a_subs,[0 3000e-6],':','Linewidth',3,'Color',[0
                    0.4470 \ 0.7410]);
            end
            legend([pl1 pl2],{'a approx','a full'},'location','northwest')
241
            xlabel('d [$\mu m$]')
            ylabel('a [$\mu m$]')
            title(['Eq for a, b=' strjoin(arrayfun(@(x) num2str(x),
                equation_plot_b*1e6, 'UniformOutput', false), ', ') 'um'])
           %Save equation plot
246
            saveas(f_eq,[Files_folder '\' Save_folder '\equation_plot.png'])
            saveas(f_eq,[Files_folder '\' Save_folder '\equation_plot.fig'])
       end %plot_equation
    end %analyze_equation
251
    %% 6. create meshgrid of data and plot contour plot of one value of b
   for contour_b = transpose(b_uniques)
       clear x y
256
       f_contour = figure('Position', [0 0 1300 900],'Visible','on');
       d_values_b = d_all(b_all == contour_b);
       a_values_b = a_all(b_all == contour_b);
        rcrit_min_b = rcrit_min(b_all == contour_b);
261
       x=d_values_b*1e6;
       y=a_values_b*1e6;
       z=rcrit_min_b*1e6;
       xv = linspace(min(x), max(x), length(unique(x)));
266
       yv = linspace(min(y), max(y), length(unique(y)));
        [X,Y] = meshgrid(xv, yv);
       Z = griddata(x,y,z,X,Y);
       if contour_mode == 1
271
            contourf(X, Y, Z,'--','ShowText','on')
```

```
elseif contour_mode == 2
            contourf(X, Y, Z,50,'LineColor','none')
       elseif contour_mode == 3
            [c,h] = contourf(X, Y, Z,contour_steps, '--', 'ShowText', 'on');
276
            h.LevelList=round(h.LevelList); %rounds levels to 3rd decimal
               place
            clabel(c,h)
       else %contour_mode
            disp('ERROR
                           Your selected contour_mode is invalid, try 1,2 or
               3')
            return
281
       end %contour_mode
       grid on
       shading interp
       clrbar = colorbar();
286
       xlabel('d [$\mu$m]')
       ylabel('a [$\mu$m]')
       xlim([0 max(d_all)*1e6])
       ylim([0 max(a_all)*1e6])
291
       ylabel(clrbar, 'R_{crit} [\mum]')
       legend(['b = ' num2str(contour_b*1e6) ' $\mu$m'],'Location','northwest
            1)
       if contour_logscale
296
            set(gca,'colorscale','log')
       end %contour_logscale
       %Save without fit
       saveas(f_contour,[Files_folder '\' Save_folder '\contour_b' num2str(
301
            contour_b*1e6) '.png'])
       saveas(f_contour,[Files_folder '\' Save_folder '\contour_b' num2str(
            contour_b*le6) '.fig'])
       % 7. Plot data with contour plot
       if analyze_equation
           hold on
306
            a_approx_subs = subs(a_approx,b,contour_b);
            xd = linspace(0,3000e-6,200);
            ya = subs(a_approx_subs,d,xd);
            plot(xd*1e6,ya*1e6,':','Linewidth',5,'Color','red')
            xlim([0 3000]), ylim([0 1500])
311
            legend({['b = ' num2str(contour_b*1e6) ' $\mu$m'],['Fit: $a = '
               char(a_approx) '$']},'Location','northoutside')
       end %analyze_equation
       %Save with fit
       saveas(f_contour,[Files_folder '\' Save_folder '\contour_fit_b'
316
           num2str(contour_b*1e6) '.png'])
       saveas(f_contour,[Files_folder '\' Save_folder '\contour_fit_b'
            num2str(contour_b*1e6) '.fig'])
```

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end %for contour_b = b_uniques