Drop pinch-off from hydrophobic heat exchanger plates

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Drop pinch-off from hydrophobic heat exchanger plates

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Chapter 1: Introduction

1.1 Goals
The goal of this study is to increase the knowledge on the causes and consequences of drop pinch-off, as occurring in hydrophobic polymer heat exchangers. To this end, three aspects have been investigated.

Re-entrainment, which is caused by drop pinch-off, has been studied in a small scale heat exchanger to quantify the effect of drop pinch-off on the overall heat and mass transfer rate.

Experiments are performed in single plate mock-ups to find by which mechanism drops break off a heat exchanger plate. Various quantities, like the edge shape, plate orientation vapour velocity and surface tension have been varied to allow the construction of a semi-empirical correlation to predict the drop size after pinch-off.

Finally, several experiments have been performed at well-defined conditions which allow validation of numerical simulations of drop pinch-off in forced convective flow.

1.2 Topics
The industrial problem: re-entrainment from heat exchanger plates
Polymer heat exchangers are suitable for recovering heat from corrosive or fouling gas streams. Such gas streams are produced for example by ceramic furnaces or during biomass incineration. Water vapour, which is present in the gas mixture, condenses dropwise on hydrophobic plates. Many corrosive components, like SO₂, SO₃, HF, HCl and HNO₃, dissolve readily in the condensate. Collection of the condensate allows easy extraction of these components from the gas stream, resulting in lower gas cleaning costs. This study investigates what the effect of re-entrainment of condensate into the gas stream is on both the overall heat transfer and gas cleaning efficiency in a compact polymer heat exchanger, and how the negative effect can be minimised.

Part of the re-entrained drop are swept out of the heat exchanger and impact on the walls of downstream piping or equipment. Continued impaction of re-entrained drops during years of operation can damage the downstream piping or equipment. To avoid this, no re-entrainment should occur at the normal heat exchanger operating conditions. In this report, re-entrainment is measured at various operational conditions, showing under which conditions downstream equipment might be damaged.

Re-entrainment has been studied in a heat exchanger, (§2.3.1), made from polyvinylidene-fluoride (PVDF). The advantage of a PVDF heat exchanger over more commonly used metal heat exchangers is that the material is resistant to almost any corroding or otherwise aggressive substance or mixture, and anti-
fouling characteristics of the teflon-like substance PVDF are very good. Furthermore, water condenses dropwise on PVDF, whereas water condenses filmwise on most other materials. The heat transfer rates during dropwise condensation are higher than the heat transfer rate during filmwise condensation, making dropwise condensation a desirable phenomenon.

The disadvantage of PVDF compared to more commonly used metals is that the heat transfer rate of PVDF is much lower. Coating a metal wall with a hydrophobic polymer combines the advantage of dropwise condensation with the high heat transfer rates of metals. Unfortunately, polymer coated metals degrade during prolonged operation, while the PVDF heat exchanger has proven to be fouling-resistant and uphold dropwise condensation during years of experimenting. Thus, at industrial conditions where corrosion or fouling prohibits the use of regular or coated metal heat exchangers, a compact PVDF heat exchanger can be a good alternative (although less efficient, it works at least).

The large internal plate area and the presence of many small drops in the heat exchanger result in a large area for air-liquid interface. This facilitates cleaning of the gas by adsorbing water-soluble pollutants, like hydrofluoric, hydrochloric and sulphuric acid into the liquid. In this thesis it will be shown that when all condensate is removed without re-entrainment, this results in high cleaning rates of a gas stream without additional investment cost.

However, when the air mixture velocity in the PVDF heat exchanger is too high, condensate drops break off the plates and are re-entrained in the gas stream. Because no study on this phenomena has been found in literature, re-entrainment during dropwise condensation has been studied. Methods to measure adsorption of water-soluble components and re-entrainment are presented in Chapter 2. Small scale experiments show the effect of vapour velocity and condensate mass flux on re-entrainment and show the effect of re-entrainment on heat and mass transfer at a typical industrial condition.

**Insight in re-entrainment: study of the pinch-off of a single drop from a vertical plate**

Although these experiments quantify re-entrainment in a compact condenser, they yield little insight in the mechanism that causes re-entrainment or by which methods re-entrainment could be reduced. These topics have therefore been investigated in single-plate mock-ups of a hydrophobic heat exchanger plate. In Chapter 3, experiments on drop pinch-off from a horizontal hydrophobic heat exchanger plate at ambient conditions are presented. Drop shapes during pinch-off are used to calculate various forces acting on drops, pinching off of plates with various edge shapes. Furthermore, drop sizes after pinch-off have been determined.

Additional experiments on drop pinch-off using ethane, water and ethanol at various pressures and vapour velocities are presented in Chapter 4. These measurements reveal the effect of liquid and vapour density, surface tension and
vapour velocity on the size of drops after pinch off. Results are compared to measured drop sizes after pinch-off from a vertical plate, showing the relevance of plate orientation.

**Pinch-off at near-critical conditions**

Numerical simulations of systems of changing topology, of which drop pinch-off is an example, have been performed using a diffuse interface model. An important difference with previously performed numerical simulations on drop pinch-off is that here, vapour shear forces, gravity nor surface tension can be ignored during pinch-off. Experiments using ethane at near-critical conditions have been performed to allow validation of these numerical calculations.

Performing experiments at near-critical conditions has the following advantage. Because the interface at near-critical conditions is much thicker than molecular dimensions, it is possible to perform numerical simulations with limited or no scaling of the interfacial thickness.

Up to this point, it is still disputed how various dimensionless numbers should be adapted when the interfacial thickness is scaled. Because these are the first experiments on drop pinch-off without density matching, performed in forced convective flow, validations of numerical experiments and the effect of interface scaling at non-zero Bond, Weber and Reynolds number at a wider range of conditions is possible.

1.3 Structure of this thesis

Tree subjects are treated in this report: re-entrainment in a small scale heat exchanger, experiments to determine the pinch-off mechanism and experiments an near-critical conditions to validate numerical simulations of pinch-off.

In the next chapter (Chapter 2), re-entrainment in a small scale heat exchanger is treated. Background on heat and mass transfer is presented in section 2.2. The test rig is described in section 2.3. Measured heat transfer, mass transfer and re-entrainment rates are presented in section 2.4. These are used to calculate the influence of re-entrainment on overall heat transfer rates (section 2.5) and gas cleaning efficiency (section 2.6) in a heat exchanger array.

Pinch off of water drops from a horizontal hydrophobic plate is studied in chapter 3. Various forces acting on a drop are calculated in section 3.2. A single plate mock up is described in section 3.3, experimental results are presented in section 3.4.

In Chapter 4, drop pinch-off at elevated pressure is studied. The test rig is described in section 4.2, while experimental results are presented in section 4.3. Most experiments are performed using ethane at near-critical conditions. These are used to validate a numerical simulation of a drop that pinches off, section 4.4 Some experiments, presented in section 4.3, have been performed using water and ethanol to study drop pinch-off at a wider range experimental conditions. The conclusions and recommendations are presented in chapter 5.
Chapter 2: The influence of re-entrainment on heat transfer and adsorption in a hydrophobic heat exchanger

2.1 Introduction
Measurements in a small scale heat exchanger have been performed to investigate whether a compact PVDF heat exchanger can be used to clean polluted gas streams and what the influence of re-entrainment is on the heat transfer rate and the adsorption rate.

To this end, three processes are investigated. First of all, re-entrainment rates are measured at various experimental conditions (§2.4.1). Secondly, heat transfer rates are measured at various vapour and coolant inlet conditions (§2.4.2). The adsorption of water-soluble pollutants is measured using ammonia. Adsorption rates are presented in §2.4.3. An adaptation to the heat exchanger to increase mass transfer is implemented and tested.

In section 2.5 measured re-entrainment rates are combined with experimentally determined heat transfer rates to calculate the effect of re-entrainment on the total amount of transferred heat in a heat exchanger cascade. In section 2.6, measured re-entrainment rates and adsorption rates are combined to calculate the effect of re-entrainment on the gas cleaning performance of a heat exchanger cascade.

Before the experimental results are presented, theory on heat and mass transfer during dropwise condensation is presented in section 2.2. Heat transfer from gas mixture to coolant is treated paragraph 2.2.1. A method to calculate the amount of ammonia in the condensate assuming no mass transfer limitation is presented in paragraph 2.2.2. This method requires the calculation of the temperature of drops on the heat exchanger plate, which is constructed in paragraph 2.2.3.

2.2 Theory on heat and mass transfer during dropwise condensation in the PVDF heat exchanger

2.2.1 Heat transfer during dropwise condensation.
The primary use of the PVDF heat exchanger is to recover heat from waste gas streams, e.g. those leaving industrial furnaces or incinerators. Adaptations to the heat exchanger that increase the overall heat transfer rate are therefore highly desirable. In this paragraph, the way in which the heat exchanger works is presented. After this, some adaptations improving the heat transfer rate are proposed and tested. A schematic impression of the PVDF heat exchanger is given in fig. 2.2.1. A detailed description is given in paragraph 3.1.
2.2 Theory on heat and mass transfer

As shown in fig. 2.2.1, the heat exchanger is operated in cross flow. Cold coolant enters the heat exchanger at the top and flows down through small channels, heating up. Hot gas mixture enters the heat exchanger from the left side and flows horizontally, cooling down. As a result, temperature difference between gas mixture and coolant, separated by the walls of the heat exchanger plate is largest in the top-left corner where hot gases and cold coolant flow into the heat exchanger. The temperature difference is lowest in the lower-right corner, where both gas mixture and coolant leave the heat exchanger. A schematic of the temperature profiles perpendicular to the heat exchanger plate are presented in fig. 2.2.2.

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Fig. 2.2.1 Schematic of gas mixture flow, coolant flow and temperatures in PVDF heat exchanger. Light colour denotes high temperature of the fluid in front of the plate, a dark colour denotes low temperature.

Fig. 2.2.2 Schematic of temperature profiles in PVDF heat exchanger. Top: centre of vapour channel. Bottom: centre of liquid channel. The insert to the right indicates the position on the heat exchanger plates.
The three pictures, from left to right, show schematics of the temperature profile at the gas intake, the temperature profile after a short distance in gas flow downstream direction, and the temperature profile after condensation has set in. At the gas inflow, temperature of the gas at the plate surface equals the gas mixture inlet temperature, which results in a high temperature difference over the plate separating gas and coolant. Conductive heat transfer, for example through the plate, is described by the well-known phenomenological equation 2.2.1:

\[ Q = -\lambda \nabla T \]  \hspace{1cm} 2.2.1

where \( Q \) is the heat transfer rate per unit area and \( \nabla T \) is the temperature gradient. \( \lambda \) is the heat transfer coefficient. Typical values for \( \lambda \) at ambient conditions (1 bar, 20ºC) are 0.024 W/mK for air, 0.17 W/mK for PVDF and 0.6 W/mK for water [16,19,23,29].

As the gas flows through the heat exchanger, temperature in the gas near the wall decreases, until the water vapour partial pressure equals the saturation pressure at the wall. Water vapour in the gas stream condenses, forming small drops on the heat exchanger plate. Condensation of water vapour releases the heat of condensation, increasing the heat flux through the plate. Drops on the plate grow due to condensation and coalescence[1,6,15,17,37]. At a certain size, gravitational and vapour shear forces remove the drop from the plate.

All heat that is transferred from gas mixture to coolant must be conducted through the plate. To increase the heat transfer rate from gas to coolant, the temperature difference over the plate or the heat transfer coefficient should be increased, or the plate thickness has to be decreased. The heat transfer coefficient of PVDF cannot be changed, and decreasing the plate thickness limits the strength of the plate. To increase the local heat transfer rate from gas mixture to coolant, the temperature difference over the plate will be increased. Three methods to achieve this will be tested:

1) The first method is to increase the coolant flow rate. The average rise in coolant temperature decreases as the coolant flow rate increases. This will increase the temperature difference between cold coolant and hot gas mixture.

2) The second method is to increase the gas mixture velocity. As the gas mixture velocity increases, more gas is fed to the heat exchanger. Consequently, The decrease in gas temperature decreases, increasing the average temperature over the plate. Furthermore, increasing the gas velocity might change the vapour flow from laminar to turbulent, resulting in an increased mass and heat transfer rate.

3) The third method that will be tested is the use of hydrophobic PAL inserts on the gas side. The inserts serve two goals. First of all, the inserts act as static mixers, increasing convective heat transfer and vapour transfer in the gas mixture. Furthermore, the hydrophobic inserts are expected to affect drainage of condensate. This influences the average condensate drop size, thereby changing heat transfer from gas to coolant.
2.2 Theory on heat and mass transfer

2.2.2 Mass transfer of water-soluble substances from gas mixture to condensate

In the ceramics industry, hydrofluoric acid has to be removed from the gas leaving ceramics ovens. Because of their corrosive nature, environmental impact and experimental hazards, adsorption experiments using these substances are not desirable in the partially aluminium test rig at the university of Technology in Eindhoven. To overcome this problem, a non-condensable water-soluble substance is chosen to investigate adsorption in condensate during condensation in the pilot plant test rig.

Experiments have been performed to determine the influence of solubility and mass transfer limitations on adsorption of a non-condensable water-soluble component from the gas stream into the condensate in the PVDF heat exchanger.

Ammonia (NH₃) is chosen as the absorption test substance for the following reasons:

- Comparable physical properties:
- Not condensable
- High solubility
- Chemically stable
- Not corrosive to the test rig
- Ratio of the partial pressure in vapour to the concentration in the condensate is almost independent of the partial pressure.
- Limited environmental and personal risk
- Accurate determination of concentration possible
- Easily available, low cost

Calculation of the equilibrium concentration of ammonia

The solubility of ammonia is calculated to compare the amount of dissolved ammonia in the condensate with the equilibrium concentration. This will show up to which extend mass transport limits the gas cleaning ability of the heat exchanger. The amount of ammonia that can dissolve in water is given by the Henry equation, 2.2.2

\[ P_{\text{NH}_3,\text{eq}} = H \cdot c_{\text{eq}} \]

where \( P_{\text{NH}_3,\text{eq}} \) is the equilibrium ammonia partial pressure, \( H \) is the Henry constant and \( c_{\text{eq}} \) is the equilibrium ammonia concentration in water (kg/kg). When the actual ammonia pressure is higher than the equilibrium pressure, ammonia dissolves in the liquid. When the partial pressure of ammonia is lower than the equilibrium pressure, ammonia is released by the solution.

The Henry constant is independent of the partial pressure of ammonia and total pressure, although it depends on temperature. A graphical presentation of the Henry constant of ammonia at temperatures between 20 and 95ºC according to Perry[29] is given in fig. 2.2.3.
The total amount of ammonia in a solution is higher than the amount given by the Henry equation, because part of the ammonia reacts with water, forming ammonium and hydroxide:

\[ \text{NH}_3 + \text{H}_2\text{O} \rightarrow \text{NH}_4^+ + \text{OH}^- \]  \hspace{1cm} 2.2.3

To calculate the total amount of ammonia that can dissolve in the condensate, the amount of dissociated NH\textsubscript{3} needs to be calculated. The molar ammonia concentration, \([\text{NH}_3]\), is the number of moles of NH\textsubscript{3} per litre water. The equilibrium NH\textsubscript{4}\textsuperscript{+}-concentration depends on ammonia concentration \([\text{NH}_3]\) and the acidity of the solution\([\text{H}_3\text{O}^+]\):

\[ K_z = \frac{[\text{H}_3\text{O}^+][\text{NH}_3]}{[\text{NH}_4^+]} \]  \hspace{1cm} 2.2.4

\[ K_d = [\text{OH}^-][\text{H}_3\text{O}^+] \]  \hspace{1cm} 2.2.5

where the square brackets refer to the concentration in mole/l. 

\(K_z\) is the acidity constant of ammonia in water and \(K_d\) the dissociation constant of water. Both are independent of the composition of the solution, while temperature variations have a negligible effect. Their values are:

\(K_z = 5.75 \times 10^{-10}\) mole/l

\(K_d = 1.0 \times 10^{-14}\) mole²/l²

The acidity of a solution is commonly presented via the pH, which is defined by:

\[ \text{pH} = -\log[\text{H}_3\text{O}^+] \]  \hspace{1cm} 2.2.6

The total ammonia concentration \([\text{NH}_3]_{\text{tot}}\) is the sum of the concentrations of ammonia and ammonium:

\[ [\text{NH}_3]_{\text{tot}} = [\text{NH}_3] + [\text{NH}_4^+] \]  \hspace{1cm} 2.2.7
The conversion $\xi$ is defined by:

$$\xi = \frac{[\text{NH}_4^+]}{[\text{NH}_3]_{\text{tot}}}$$  \hspace{1cm} 2.2.8

Because we know that the total amount of positive and negative ions in a solution are identical (2.9), we can calculate $\xi$ from measured values of the total amount of ammonia.

$$[\text{OH}^-] = [\text{H}_3\text{O}^+] + [\text{NH}_4^+]$$  \hspace{1cm} 2.2.9

The calculation has been performed in the following way, see table 2.2.1.

**Table 2.2.1 Seven steps to calculate the conversion of ammonia in $[\text{NH}_4^+]$ in a aqueous solution without other acids or bases.**

<table>
<thead>
<tr>
<th>Step</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>pH is chosen</td>
</tr>
<tr>
<td>2</td>
<td>$[\text{H}_3\text{O}^+]$ is calculated using 2.6 $[\text{H}_3\text{O}^+] = 10^{-\text{pH}}$</td>
</tr>
<tr>
<td>3</td>
<td>$[\text{OH}^-]$ is calculated from 2.5 $[\text{OH}^-] = K_d / [\text{H}_3\text{O}^+]$</td>
</tr>
<tr>
<td>4</td>
<td>$[\text{NH}_4^+]$ is calculated from ion balance (2.9) $[\text{NH}_4^+] = [\text{OH}^-] - [\text{H}_3\text{O}^+]$</td>
</tr>
<tr>
<td>5</td>
<td>$[\text{NH}_3]$ is calculated from 2.4 $[\text{NH}_3] = K_z [\text{NH}_4^+] / [\text{H}_3\text{O}^+]$</td>
</tr>
<tr>
<td>6</td>
<td>$[\text{NH}_3]_0$ is calculated from 2.7 See 2.7</td>
</tr>
<tr>
<td>7</td>
<td>Conversion is calculated from 2.8 See 2.8</td>
</tr>
</tbody>
</table>

A table is constructed, in which pH values between 7 and 14 are stored, see table 2.2.2.

**Table 2.2.2 Table to calculate conversion of NH$_3$ into NH$_4^+$**

<table>
<thead>
<tr>
<th>pH</th>
<th>10.64</th>
<th>10.66</th>
<th>10.68</th>
<th>10.7</th>
<th>10.72</th>
<th>10.74</th>
</tr>
</thead>
<tbody>
<tr>
<td>$[\text{H}_3\text{O}^+]$ Mole/l</td>
<td>2.29E-11</td>
<td>2.19E-11</td>
<td>2.09E-11</td>
<td>2E-11</td>
<td>1.91E-11</td>
<td>1.82E-11</td>
</tr>
<tr>
<td>$[\text{OH}^-]$ Mole/l</td>
<td>0.000437</td>
<td>0.000457</td>
<td>0.000479</td>
<td>0.000501</td>
<td>0.000525</td>
<td>0.00055</td>
</tr>
<tr>
<td>$[\text{NH}_4^+]$ Mole/l</td>
<td>0.000437</td>
<td>0.000457</td>
<td>0.000479</td>
<td>0.000501</td>
<td>0.000525</td>
<td>0.00055</td>
</tr>
<tr>
<td>$[\text{NH}_3]$ Mole/l</td>
<td>0.019965</td>
<td>0.012023</td>
<td>0.013183</td>
<td>0.014454</td>
<td>0.015849</td>
<td>0.017378</td>
</tr>
<tr>
<td>$[\text{NH}_3]_0$ Mole/l</td>
<td>0.011401</td>
<td>0.01248</td>
<td>0.013661</td>
<td>0.014956</td>
<td>0.016374</td>
<td>0.017928</td>
</tr>
<tr>
<td>$\xi$ %</td>
<td>3.82865</td>
<td>3.662644</td>
<td>3.503573</td>
<td>3.351171</td>
<td>3.205178</td>
<td>3.065343</td>
</tr>
</tbody>
</table>

The $[\text{H}_3\text{O}^+]$ is calculated at every pH and stored in the table. The concentration of the other ions and the ammonia concentration follow directly from the acidity. In the experiments, presented in §4.1, the total ammonia concentration, $[\text{NH}_3]_{\text{tot}}$, in the condensate is determined by titration. The conversion of ammonia in ammonium is determined by linear interpolation from the table. Errors resulting from this procedure are less than 0.1%. Conversion rates are presented graphically versus the total amount of ammonia in fig. 2.2.4.
Example

An example is used to show how the total amount of ammonia that can dissolve in a solution is calculated. Take a drop of water in a gas mixture at 60°C with a partial pressure of ammonia of 100 Pa (10^{-3} bar).

At 60°C, the Henry constant of ammonia is 4.11 bar, see fig. 2.1.3. The equilibrium ammonia mass fraction in the condensate is

\[ P_{NH_3}/H = \frac{0.001}{4.11} = 2.43 \times 10^{-4} \]

1 litre (1000 grams) of liquid contains \(1000 \times 2.43 \times 10^{-4} = 0.243\) g of \(NH_3\). The molar mass of ammonia is 17 g/mole: The ammonia concentration \([NH_3]\) is thus \(0.243/17 = 14.3 \times 10^{-3}\) mole/l.

The amount of dissociated ammonia is found by interpolation of the data presented in table 2.2 between pH=10.68 and 10.70:

\[
\begin{align*}
\xi &= 0.121 \quad \text{(between pH=10.68 and 10.70)}
\end{align*}
\]

The equilibrium amount of dissolved ammonia, \([NH_3]_{tot}\), is \(14.3 \times 1.034 = 14.8\) mmole/l=25.1 mg/l.

Now assume that the measured concentration of ammonia in the solution is 8 mg/l. Clearly, the equilibrium concentration is higher than the amount of adsorbed ammonia, showing that mass transfer limits adsorption of ammonia.
Calculation of drop temperature

The solubility of ammonia depends on temperature, as shown in the previous paragraph. Unfortunately, the temperature of the condensate in the heat exchanger has not been measured. Therefore, a method is presented here to estimate the average condensate temperature in the heat exchanger. The temperature of the condensate is calculated using the heat flux from gas mixture to coolant and average coolant temperature. The assumptions that are made to perform this calculation are graphically presented in fig. 2.2.5.

It has been observed that drops are nearly hemispherical and cover 40-50% of the plate, see e.g. fig. 2.4.13. It is assumed that drops cover 50% of the plate.

Furthermore, it is assumed that the heat flux can be calculated by assuming that the drop height is constant, the temperature of the plate surface is constant and that negligible amounts of heat are transferred to the plate through the air in between the drops. Consequently, all water condenses on the upper surface of the drops. The heat flux Q through drop and plate are given by substituting the proper temperature gradient and heat transfer coefficient in eq. 2.2.10:

\[ Q = \lambda_{\text{H}_2\text{O}} \frac{T_{\text{gl}} - T_{\text{ls}}}{d_{\text{plate}}} \frac{A_{\text{drop}}}{A_{\text{plate}}} \]  

\[ Q = \lambda_{\text{H}_2\text{O}} \frac{T_{\text{ls}} - T_{\text{cool}}}{d_{\text{plate}}} \]  

From 2.10 and 2.11, we find:

\[ T_{\text{ls}} = T_{\text{cool}} + \frac{d_{\text{plate}}}{\lambda_{\text{PVDF}}} Q \]
Because the heat flux is constant, dT/dh is constant in the modelled cylindrical drop. The temperature in the drop, averaged over the drop height, is thus given by:

\[ T_{\text{drop}} = \frac{T_{\text{gl}} + T_{\text{ls}}} {2} = T_{\text{cool}} + Q \left( \frac{d_{\text{plate}}}{\lambda_{\text{PVDF}}} + \frac{h_{\text{drop}}}{\lambda_{\text{H2O}}} \right) \]  

2.2.14

Coolant temperature and heat flux are not constant in the heat exchanger. Therefore, also \( T_{\text{gl}} \) will vary in the heat exchanger. The temperature that is calculated, based on the average heat flux and coolant temperature in the heat exchanger, will give a reasonable representation of the temperature \( T_{\text{gl}} \), averaged of the entire heat exchanger.

The average coolant temperature \( T_{\text{cool}} \) is calculated by the average of the coolant inlet and outlet temperature:

\[ T_{\text{cool}} = \frac{T_{\text{cool,in}} + T_{\text{cool,out}}} {2} \]  

2.2.15

2.2.16

2.3 Experimental set-up: The compact PVDF heat exchanger test rig.

2.3.1 The compact PVDF heat exchanger test rig

The pilot plant test rig consists of a wind tunnel in which a compact plate PVDF heat exchanger is mounted. The wind tunnel has been adapted to allow measurement and visual investigation of condensate re-entrainment and measurement of the adsorption of a water-soluble component, ammonia, in the condensate. A Schematic of the pilot plant test rig is presented in fig. 2.3.1.

Ambient air is blown into the wind tunnel by two 880 W electrical fans. At the outlet, a third fan draws air from the test rig. The air stream is heated by an adjustable
120kW heat exchanger, which is fed with dry steam of 160°C and 7.8 bar. Dry steam of 160°C/7.8 bar is flashed into the air flow, increasing the relative humidity and temperature of the air. Steam is produced by a 957kW steam generator, manufactured by EMK Belgium.

Water of 80°C, containing a mass fraction of 6.7% NH₃ can be injected into the gas stream through a spray nozzle. Observations have shown that all drops are evaporated as the gas mixture enters the measurement section. The air mixture flows through a smooth tube with a length of 6m and an inner diameter D of 200 mm. An orifice with an inner diameter of 150 mm is placed in the tube 4m downstream of the tube inlet. The orifice is used to calculate the mass flow rate of the air mixture.

The air flows through a flow straightener into the measurement section, see fig. 2.3.2.

**Fig. 2.3.2. Schematic of the measurement section of the pilot plant test rig (side view) and PVDF heat exchanger (front view). Sizes not to scale.**

Symbols: 4T = 4 Pt100 thermometers, Hᵣ = relative humidity sensor, ∆p = nanometer

**The measurement section**

The measurement section consists of two sections. A PVDF heat exchanger is placed in each section. In most experiments described here, one section is sealed off with a brass plate, allowing higher vapour flow rates through the other heat exchanger. Both compartments are used only when the adsorption of ammonia is investigated. The superficial air mixture velocities in the heat exchanger during these adsorption experiments were 3.5±0.2 m/s. All other experiments, where air mixture velocities exceeds 6m/s, are performed with one section open and one closed.
A PVDF heat exchanger is placed in each measurement section. A measurement section consists of two chambers, one upstream and one downstream of the PVDF heat exchanger. The heat exchanger is constructed of 48 plates, 400mm high, 76.3mm long and 2mm thick. Every plate contains 42 channels of 1.37x1.47mm² through which cooling water flows, see fig. 2.3.3.

Cooling water flows downward through 24 plates into a header where the cooling water mixes. From the header, cooling water flows upward through the remaining 24 plates. Cooling water temperature is measured before the heat exchanger, in the header and after the heat exchanger. The chamber upstream of the heat exchanger is 0.85m long, 247mm wide and 693mm high. The chamber downstream of the heat exchanger is 0.3m long, 247mm wide and 693mm high. Temperature of the air mixture in both the upstream and the downstream chamber are measured at four heights using class A Pt100 thermoresistors, accuracy 0.1°C. The thermometers are calibrated between 20 and 95°C. The temperature sensors are located at heights of 90, 260, 430 and 600mm. The average temperature of the vapour flowing through the test section is determined from the velocity weighted average at multiple positions in the test section:

\[ T = \frac{\sum_{i=1}^{4} T_i v_i}{\sum_{i=1}^{4} v_i} \]

with \( v_i \) the time-averaged velocity in downstream direction at position \( i \). The weighting factors \( 4v_i/\Sigma v_i \) depend on the position, upstream or downstream of the heat exchanger, and are typically between 0.6 and 1.5.

In experiments with large entrainment rates, condensate drops have been observed to impact on the lower end of the thermometer. The drops stick to the hydrophilic stainless steel thermometer and spread out, forming a thin film that evaporates. This locally reduces the temperature. When these phenomena are observed, temperature is determined from the velocity weighted average of the upper 3 sensors, which are rarely wetted. The average temperature at one position is determined from 100 samples during 10 min. The impact of a drop is revealed by a decrease in temperature during 1-5 samples. Measurements are ignored when
2.3 Experimental set-up

the temperature deviates more than ten times the standard deviation from the average temperature.

Relative humidity in both chambers are determined using Vaisala HMP 135Y sensors with an accuracy of 1%. Humidity sensors are calibrated using demineralised and saturated solutions of Sodium Chloride and Lithium Chloride in demineralised water.

In the experiments with one measurement section, a humidity sensor is placed at the air inlet to determine temperature and humidity of the ambient air flowing into the test rig. A fourth humidity sensor is placed near the ammonia solution spray nozzle, about 7m upstream of the PVDF heat exchanger. Both sensors have not been used for analysis of heat exchanger performance, but make it easier to achieve isothermal conditions.

In the experiments with two measurement sections, humidity sensors are placed in each section, both upstream and downstream of the PVDF heat exchanger.

A droplet catcher has been installed in the open measuring section, 0.4m downstream of the heat exchanger. Condensate is collected from three drains, see fig. 2.3.2. The first drain collect condensate that drains down from the heat exchanger. The second drain collects condensate that settles in the compartment between heat exchanger and droplet catcher. The third drain collects condensate from the droplet catcher. The absence of drops in the air stream leaving the droplet catcher has been confirmed by visual observation.

Condensate mass flow rates have been determined by measuring weight and volume of condensate collected from each drain during 2-5 min. Accuracy is better than 0.5%.

2.3.2 Ammonia injection and measurement equipment

The adsorption rate of water-soluble components in the condensate has been determined by injecting ammonia into the gas stream and measuring ammonia concentrations in condensate and air mixture before and after the heat exchanger. The injection system is presented in fig. 2.3.4.
A 20 litre stainless steel storage vessel is filled with a 6.7% (mass fraction) ammonia solution and pressurised to 10 bar using oil-free pressurised air. The ammonia solution leaving the storage vessel flows through a 3 m long ∅6 mm stainless steel tube submerged in a 80 ºC water bath. The pressure is reduced to 2.5 bar, after which the ammonia solution is fed to a spray nozzle, type Teejet TG SS2. The spray nozzle is located at the entrance of the tube in which the measurement flange is fitted, about 7m upstream of the heat exchanger. The mass flow rate through the spray nozzle has been determined at pressure drops over the spray nozzle between 1 and 9 bar, by weighting the amount of liquid that flows out of the nozzle during 5 min. Reproducibility is better than 0.5%. Furthermore, the amount of ammonia solution that has been injected is determined by measuring the change in amount of solution in the storage vessel during 15 min. Differences in fluxes given by both methods are less than 1%. Presented fluxes in this report are calculated by averaging the two fluxes.

Full evaporation of all drops is confirmed prior to the experiments by increasing the ammonia solution flow rate by a factor five and reducing the temperature from 80ºC to 75ºC at constant absolute humidity. Visual inspection showed that no unevaporated drops reach the measurement section even at the low-temperature condition.

During each experiment, two condensate samples, each about 500ml, from drain I are collected to determine ammonia concentration. The ammonia concentration of the injected solution and the condensate have been determined by titration with a
2.3 Experimental set-up

hydrogen chloride solution. Phenolphthalein is used as pH indicator. Reproducibility of
the ammonia concentration is within 0.2%. Partial pressure of ammonia upstream
and downstream of the heat exchanger is determined by feeding a small amount of
air mixture from the measurement section through an adsorption flask in which a
hydrochloric acid solution with phenolphthalein is present. Vapour is withdrawn from
the measurement section using a water jet pump. Air mixture flow rate is
determined using a rotameter constructed by Brooks Instruments, type R-2-15-AA,
accuracy 1%. When the amount of ammonia that is adsorbed in the adsorption
flask equals half the amount of hydrochloric acid present in the adsorption flask,
the pH indicator changes from colourless to purple. The solution changes colour
within 1 sec, which happens after 2-5 min after opening of the valves. The
accuracy of the analysis is determined by the accuracy of the rotameter, which is
1%. The analysis is performed in the following way.

Before the experiment is started, adsorption flask I and II, see fig. 2.3.5, are filled
with 50 ml of a 11.8 mmol/l HCl-solution.

Adsorption flask III is filled with demineralised water. A single drop of 10 g/l
phenolphthalein is added to each flask. When the temperature of the air stream
leaving the PVDF heat exchanger is stationary, ammonia solution is injected into
the test rig. After 5 min, partial pressure of the ammonia upstream of the heat
exchanger is determined. Valves 1 and 4 are opened, after which the water jet
pump is turned on. Valves 2a and 2b are opened, immediately followed by closing
valve 1.
As long as valve 1 is open no air flows through the adsorption flasks. Immediately after valve I is closed, air is drawn in from the test rig. The time it takes until the solution in vessel I changes colour is determined. This time is corrected for by the time it takes for the air mixture from the heat exchanger to reach the adsorption flasks, which is about 1s. Adsorption vessel III has a special function; it contains demineralised water and the pH indicator. A small amount, less than 0.1% of the amount of ammonia that is adsorbed in vessel I, would suffice to change the pH in this vessel enough for the solution to change to purple. The fact that this does not happen proves that no ammonia passed vessel I without being adsorbed. The experiment is repeated for the air drawn downstream of the PVDF heat exchanger by opening valves 3a and 3b.

During the determination of the partial pressure of ammonia upstream and downstream of the heat exchanger, two batches of 500-1000 ml of condensate are collected from drain 1 and stored in leak-tested PE containers. Concentration of both samples is determined after switching off the pilot plant test rig.

2.3.3 Calculation of air mixture mass flow rate

Mass flow rate of the air mixture is determined from the pressure drop over an orifice, which is measured with an inclined manometer, manufactured by Airflow Developments LTD Lancaster, England (type Mk 4&5). Accuracy of the manometer is 0.1% of full scale (1 kPa). The orifice is constructed according to DIN 1952. Mass flow rate is calculated using DIN 1952:

\[
\dot{m} = \alpha_{\omega} \varepsilon \frac{\pi}{4} \beta^2 D^2 \sqrt{2 \Delta P} \rho \nu
\]

\[2.3.2\]

\(\beta\) is the ratio of the inner tube diameter to orifice diameter, \(d/D\). The orifice discharge coefficient \(\alpha_{\omega}\) is given by DIN 1952:

\[
\alpha_{\omega} = 0.5959 + 0.0312 \beta^{2.1} - 0.1840 \beta^8 + 0.0029 \beta^{2.5} \left(\frac{10^6}{\text{Re}}\right)^{3/4} \frac{1}{\sqrt{1 - \beta^4}}
\]

\[2.3.3\]

The expansion coefficient \(\varepsilon\) is given by:

\[
\varepsilon = 1 - \left(0.41 + 0.35 \beta^4 \left(\frac{\Delta P}{k \rho_0}\right)\right)
\]

\[2.3.4\]

Where \(\kappa\) is the isentropic expansion coefficient of the air-vapour mixture.
2.4 Results of adsorption and re-entrainment measurements.

Three phenomena have been studied in the compact PVDF heat exchanger: heat transfer, ammonia adsorption and re-entrainment. Re-entrainment is presented in the first paragraph of this section. The second paragraph treats heat transfer during dropwise condensation of water-air mixtures. The adsorption of ammonia in the condensate is treated in the third paragraph.

2.4.1 Entrainment during dropwise condensation

In this paragraph, entrainment during dropwise condensation is presented. A picture, taken from downstream direction, of the heat exchanger is presented in fig. 2.4.1.

![Fig. 2.4.1 Drop entrainment at the downstream end of a compact plate heat exchanger with spacers; single frame of a video recording. On the video, the downward motion of drops on the downstream edge is clearly visible. Conditions: \(T_{in}=80.9^\circ C\), \(H_{in}=49\%\), \(v_{out}=11.5 m/s\), \(m_c=12.2 g/s\), \(m_{cool}=0.47 kg/m^2s\).](image)

The picture is a single frame of a video recording, taken with a video camera of 1028x1028 pixels. Experimental conditions were chosen in such a way that the percentage of condensate that is re-entrained into the gas stream is high, 13%. Condensate drops are visible on the plate downstream edge, sliding down. The width of the drops is limited by the plate thickness, 2mm. Drop length (in downward direction) varies between 1.5 and 2 times the plate thickness (3-4 mm). Drop height (in downstream direction) varies between 0.5 and 1.5dplate (1-3 mm). One drop, marked in fig. 2.4.1 as “drop during break-up”, is pinching off from the heat exchanger plate. Another large drop, sliding down over the downstream edge of the fourth plate from the left, has been observed to pinch off 60ms later. Small
drops do not break up but drain down and collect on the spacers. Most fluid drains drown past the spacers, but some drops pinch off from the downstream edge of spacers. Drops of various sizes are visible in the gas mixture leaving the heat exchanger. Estimated sizes of re-entrained drops range from 0.25 to 1.5mm in diameter.

It is expected that the fraction of condensate that is re-entrained into the air stream depends mainly on the ratio of disintegrating hydrodynamic forces to cohering surface tension forces, which is represented by the Weber number with respect to the droplet, $\text{We}_{\text{drop}}$:

$$
\text{We}_{\text{drop}} = \frac{\rho v_d^2 d_{\text{drop}}}{\sigma} = \frac{\rho v_d^2 d_{\text{plate}}}{\sigma} \frac{d_{\text{drop}}}{d_{\text{plate}}} = \text{We}_{\text{plate}} \frac{d_{\text{drop}}}{d_{\text{plate}}}
$$

In the remainder of this chapter, the Weber number with respect to the plate, $\rho v_d^2 d_{\text{plate}}/\sigma$, is referred to as the Weber number. Entrainment rates are measured at temperatures of 80, 82 and 90°C. The relative humidity of the inlet air mixture has been varied between 24 and 50%. Coolant mass flow rate per unit heat exchanger area were 0.47 or 0.28 kg/m²s. In fig. 2.4.2 and 2.4.3, percentages re-entrained liquid, collected from drain II and III, are presented versus the Weber number with respect to the plate.

Drain III collects the drops that are collected by the droplet catcher. Drain II collects drops that settle between the drop catcher and the heat exchanger. As shown in fig. 2.4.2 and 2.4.3, the percentage re-entrained condensate collected from each
2.4 Adsorption and re-entrainment

drain depends only on the Weber number and total amount of condensate in the heat exchanger.

With increasing Weber number, the percentage of condensate that is re-entrained increases. Increasing shear forces causes smaller drops to pinch off, increasing the amount of re-entrained condensate. At low Weber numbers, most re-entrained condensate is collected from the compartment before the drop catcher. With increasing Weber number, the fraction of re-entrained condensate collected from the droplet catcher increases. This has two causes. As the Weber number increases, shear forces working on a drop on the heat exchanger downstream edge increase. As will be shown in the next chapter, pinch-off at higher Weber number results in the formation of smaller drops, which take longer to settle. At this point however, accuracy of the optical equipment is insufficient to determine the drop size distribution after pinch-off.

Furthermore, drops are transported downstream by the gas mixture leaving the heat exchanger. As the gas mixture velocity increases, both the Weber number and the distance over which a drop is transported increase. Increasing the Weber number thus increases the fraction of entrained drops that are collected by the droplet catcher, even when the drop size distribution would not change.

When more condensate is formed, the size of drops on the heat exchanger plate remains constant, but the number of drops that slide over the plate and arrive at the downstream edge increase. This reduces the average distance between two drops that are draining down over the downstream edge, and thereby increases coalescence. Coalescence increases the average drop size; increasing

![Graph showing the percentage of total condensate collected from drain II.](image-url)
condensate fluxes thus increase the average size of drops that slide down over
the downstream edge, even though the average drop size on the plate remains
constant.

Because only large drops on the downstream edge break up and re-entrain
condensate into the gas stream, increasing the condensate flux increases the
amount of re-entrained fluid.

2.4.2 The effect of vapour and coolant inlet conditions and inserts on heat
transfer during dropwise condensation

The goal of this paragraph is to:
- present effects of changes in coolant mass flow rate, air mixture
temperature and humidity on heat transfer in the PVDF heat exchanger for
typical operating conditions.
- determine the effect of PAL inserts on heat transfer and condensate
formation

The effect of inserts and coolant flow rate on heat transfer at low gas velocity

Heat transfer has been determined at a gas mixture inlet temperature of 79.7± 3°C,
air mixture relative humidity of 45±2% and coolant mass flow rates between 10 and
50 kg/m²s. The air mixture contains 0.6±0.1/o/o NH₃(volume fraction). Ammonia
concentrations in the condensate varied between 0.24 and 0.36 g/l. It is assumed
that the ammonia mass fractions in vapour and liquid are too low to influence heat
transfer. Ganzevles [14] has shown that the presence of small amounts of water-
soluble components in the gas mixture has indeed no significant effect on the heat
transfer from gas mixture to coolant. Heat transfer rates at low gas velocities and
varying coolant mass flow rates are presented in fig. 2.4.4.

![Fig. 2.4.4 Heat transfer rates at varying coolant mass flow rates](image)

\[ T_{cool, in}=25.1\pm0.1°C, m_{in}=0.26\pm0.02kg/s, v_{in}=3.5\pm0.2m/s, H_r=45\pm2\%, \text{ pressure drop (with inserts): } 295\pm5Pa, \text{ pressure drop (without inserts): } 39\pm1Pa \]
Experiments have been performed both with and without PAL inserts. The experiments show that at coolant mass fluxes below 0.2 kg/m²s, the amount of transferred heat depends strongly on coolant mass flow rate. At coolant mass flow rates exceeding 0.3 kg/m²s, the effect of changes in coolant mass flow rate on transferred heat levels off. The experiments also show that heat transfer at low coolant flow rates is increased by the presence of the inserts, while at higher coolant flow rates the presence of the inserts decreases the heat transfer. The increase in heat transfer by the inserts at low coolant flow rate can be caused by changes in gas flow. The inserts work as static mixers, mixing cool air with a low absolute humidity near the wall with warmer more humid air in the centre of the slit between the two heat exchanger plates. At higher coolant flow rates more condensate is formed. As argued by Ganzevles et al [14], the inserts hinder drainage of drops from the heat exchanging surface, thus increasing the average drop size. Larger drop have a higher heat transfer resistance, decreasing the heat transfer rate.

The effect of gas velocity and inlet temperature, coolant flow rate and gas mixture humidity on heat transfer at high gas velocities

Heat transfer has been determined at gas mixture inlet temperatures between 70 and 90°C, inlet relative humidities between 20 and 50%, inlet absolute humidities between 9 and 32%, coolant mass flow rates between 10 and 50 kg/m²s.

Fig. 2.4.5 shows the heat flux measured on the gas side, $Q_{\text{gas}}$, versus the heat flux measured on the coolant side, $Q_{\text{cool}}$.

![Fig. 2.4.5 Heat flux, measured on the gas side, versus heat flux, measured on the coolant side of the heat exchanger. Dotted lines give 10% deviation measured heat fluxes Legend format: $T_{\text{g,in}}$ (°C) / $H_r$ (%) / $m_{\text{cool}}$ (m³/h) / $m_c$ (g/s)
Re-entrainment in compact heat exchangers

\[ Q_{\text{gas}} = \frac{1}{A_{\text{HTXR}}} \left[ \dot{m}_{\text{gas}} c_{p,\text{gas}} (T_{\text{gas,in}} - T_{\text{gas,out}}) + H_{\text{vap}} \dot{m}_{\text{cond}} \right] \quad 2.4.2 \]

where \( \dot{m} \) is a mass flow (kg/s), \( c_p \) is the specific heat of the gas mixture and \( H_{\text{vap}} \) is the heat of vaporisation of water.

\[ Q_{\text{cool}} = \frac{1}{A_{\text{HTXR}}} \left[ \dot{m}_{\text{cool}} c_{p,\text{cool}} (T_{\text{cool,in}} - T_{\text{cool,out}}) \right] \quad 2.4.3 \]

For all measurements, \( Q_{\text{cool}} \) and \( Q_{\text{gas}} \) differ less than the experimental error margins, which are determined by the accuracy of \( m_{\text{cool}} \) (±0.02) and the accuracy of \( m_{\text{gas}} \) (±5%).

In fig. 2.4.6, the water mass flux into the heat exchanger, \( m_{H_2O,in} \) is plotted versus the water flux out of the heat exchanger:

\[ m_{H_2O,in} = \frac{H_{a,in} p_{H_2O,in}}{H_{a,in} p_{H_2O,in} + (1 - H_{a,in}) p_{air,in}} m_{tot,in} \quad 2.4.3 \]

\[ m_{H_2O,in} = \frac{H_{a,out} p_{H_2O,out}}{H_{a,out} p_{H_2O,out} + (1 - H_{a,out}) p_{air,out}} m_{tot,out} + m_{\text{cond}} \quad 2.4.4 \]

Differences between measured amounts of water entering and leaving the heat exchanger are less than 5%.

![Fig. 2.4.6 measured water mass flow rates into the heat exchanger versus the water mass flow rate out of the heat exchanger. Dotted line give 5% deviation between measured water fluxes T_{g,in} (°C) / H_r (%) / m_{cool} (m^3/h) / m_c (g/s) ]

Heat fluxes at varying gas velocities in the PVDF heat exchanger, operated without inserts, are presented in fig. 2.4.7 and 2.4.9. In fig. 2.4.7, four measurement series
are presented. Coolant flow rate is constant (0.47 kg/m²s), gas mixture inlet temperature and composition are varied.

**Fig. 2.4.7 Heat transfer rates versus gas mixture velocity for various gas mixture inlet temperatures (T) and inlet absolute humidity (Ha)**

At $T_{\text{gas,in}}=80^\circ$C and $T_{\text{gas,in}}=90^\circ$C/$Ha=17\%$, heat transfer increases slightly with gas velocity. This is contributed to two effects. First of all, convective heat transfer rates increase with increasing vapour velocity, as argued by Ganzevles [14]. This is further discussed later in this paragraph.

Secondly, more gas mixture flows into the heat exchanger at higher gas velocities. Increase in gas velocity decreases the reduction in gas temperature and humidity in the heat exchanger. Thus, average difference between gas mixture and coolant temperature is higher and a higher heat flux is found.

The measurements at $T=90^\circ$C/$Ha=26.5\%$ have been performed over a range of gas velocities that is too limited to reveal a change in heat transfer rate. Comparing the heat transfer rates at constant gas velocity, gas temperature and coolant flow rate but at different temperatures reveals the effect of changes in temperature on the heat transfer rate. The heat transfer increases with $0.17\pm0.01$ kW/m²ºC when the gas inlet temperature is increased from $82^\circ$C to $90^\circ$C at a constant absolute humidity of 17%.

The effect of changes in humidity at constant temperature and coolant flow rate on heat transfer rate has been determined at $82$ and $90^\circ$C. When the absolute humidity is increased, heat transfer rates increase with $0.73\pm0.04$ kW/m² % at $82^\circ$C and $1.12\pm0.09$ kW/m² % at $90^\circ$C. The large effect of humidity on heat transfer is understood from the importance of heat released by condensation. For all experiments presented in fig. 2.4.7, 55% up to 75% of the heat that is transferred to the coolant is produced by condensation. Increasing the inlet water vapour mass fraction of the gas mixture thus strongly increases the heat transfer rate. $F_{\text{cond}}$ is the ratio of heat released by condensation to the total amount of transferred heat:
Re-entrainment in compact heat exchangers

\[ F_{\text{cond}} = \frac{H_{\text{vap}} m_{\text{cond}}}{A_{\text{HTXR}} Q_{\text{gas}}} \times 100\% \] \hspace{1cm} 2.4.5

\[ F_{\text{conv}} \] is the convective part of the transferred heat, defined by 2.4.6:
\[ F_{\text{conv}} = 100\% - F_{\text{cond}} \] \hspace{1cm} 2.4.6

The convective part of the heat released on the gas side, \( F_{\text{conv}} \), is presented versus gas mixture inlet velocity in fig. 2.4.8.

Fig. 2.4.8 shows that the convective heat transfer increases with decreasing absolute humidity, increasing temperature and increasing vapour velocity.

Fig. 2.4.9 presents the effect of gas velocity on heat transfer rate, measured at two temperatures and two coolant mass flow rates.

As expected, heat transfer rates increase with increasing temperature and increasing coolant mass flow rate. Increasing the gas velocity at 90°C increases...
the heat transfer. At 82°C, error margins are too large to conclude on the correlation between gas mixture velocity and heat transfer rate. Concluding, we have found that the heat flux from gas mixture to coolant increases with increasing gas inlet temperature, increasing absolute humidity and increasing gas and coolant mass fluxes. The effect of absolute humidity on heat flux is large compared to the effect of the other factors, which is the result of the large contribution of the heat released by condensation.

2.4.3 Adsorption of ammonia in condensate
The adsorption of ammonia (NH₃) has been studied to determine how suitable this PVDF heat exchanger is for use as a gas cleaning device. Adsorption experiments have been performed by running the heat exchanger at constant gas velocity, air mixture temperature and composition. To study the effect of changes in the amount of formed condensate, coolant mass flux has been varied between 0.10 and 0.47 kg/m²s. An aqueous ammonia solution has been injected 7m upstream of the heat exchanger using a spray nozzle. Evaporation of the solution results in a gas mixture entering the heat exchanger with an ammonia partial pressure of 60±8Pa. Measured ammonia concentrations in the condensate are presented in fig. 2.4.10.

![fig. 2.4.10](image_url)

Fig. 2.4.10 Ammonia concentrations in the condensate versus coolant mass flow rates $T_{\text{cool}, i}=25.1\pm0.1^\circ\text{C}, m_{\text{fr}}=0.26\pm0.02\text{kg/s}, v_{\text{in}}=3.5\pm0.2\text{m/s}, H_r=45\pm2\%$

The ammonia concentration in the condensate increases with coolant mass flow rate. Furthermore, ammonia concentration in the condensate collected from the heat exchanger in which no inserts are used is higher than the ammonia concentration in the concentrate collected from the heat exchanger in which no inserts are present. The equilibrium ammonia concentration in the concentrate depends on temperature, see fig. 2.4.11.
The temperature of the condensate on the plate has not been measured. Therefore, a method has been constructed in section 2.2 to calculate the temperature of the condensate on the heat exchanger plate, averaged over the plate and the drop height:

$$T_{\text{drop}} = \frac{T_{\text{g}} + T_{\text{ls}}}{2} = T_{\text{cool}} + Q \left[ \frac{d_{\text{plate}}}{\lambda_{\text{PVDF}}} + \frac{h_{\text{drop}}}{\lambda_{\text{H2O}}} \right]$$ \hspace{1cm} 2.2.15

Estimated condensate temperatures in the heat exchanger and the corresponding ammonia concentrations in the condensate are presented in fig. 2.4.12.

**Fig. 2.4.11** Equilibrium NH$_3$ concentration, based on a partial ammonia pressure of 60Pa in the gas mixture. Formation of NH$_4^+$ is included

**Fig. 2.4.12** Estimated temperatures of 0.5 mm drops on heat exchanger plates. $T_{\text{in}}=79.6\pm0.3^\circ\text{C}$, $T_{\text{cool,in}}=25.1\pm0.1^\circ\text{C}$, $m_{\text{cool,in}}=0.26\pm0.02\text{kg/s}$, $v_{\text{in}}=3.5\pm0.2\text{m/s}$, $H_r=45\pm2\%$. 

![Graph](image-url)
2.5 Re-entrainment and efficiency

For the measurements that have been performed without the PAL inserts, the difference between measured ammonia concentration and equilibrium concentration based on average drop temperature are less than 5% for all measurements. When inserts are present in the heat exchanger, measured ammonia concentration in the condensate are lower than the concentration, based on 2.25.

This is understood from the liquid distribution in the heat exchanger when inserts are present, see fig. 2.4.13.

The large reflections in the lower part of this image reveals the presence of condensate drops that are suspended in between the threads of the insert. The temperature of the suspended drop will be about identical to the local gas mixture temperature, because the drops do not touch the heat exchanger plate. The difference between measured ammonia concentration and the calculated concentration is less than 5%, when we assume that 20% of the condensate is suspended in the insert.

Concluding, it has been found that the concentration of a non-condensable water-soluble component in the condensate can be calculated using thermodynamic equilibrium, based on the temperature of drops in the heat exchanger. This is consistent with experimental results by Ganzevles [14]. In paragraph 2.6, this will be used to estimate the adsorption of hydrogen fluoride in an industry-based example.

Fig. 2.4.13 dropwise condensation in PVDF heat exchanger. The large reflections reveal the presence of drops hanging in between the threads of the PAL insert. Conditions: see fig. 2.4.9

2.5 The impact of re-entrainment on the efficiency of heat exchangers

As stated in chapter 1, re-entrainment might have 3 negative effects:

- re-entrainment lowers the thermal efficiency of heat exchanger cascades
- re-entrainment reduces the amount of absorbed pollutants
- re-entrainment might result in damaging of downstream equipment

The first and second effect are treated in paragraphs 2.5.1 and 2.5.2. The third effect are of a different nature, because this requires a mechanical rather than a thermodynamic approach. Furthermore, structural damaging is a long term effect, which depends strongly on design details. Thus, to determine how much faster
downstream equipment wears downs as a result of the continued impingement of drops falls outside the means of this study.

### 2.5.1 The impact of condensate re-entrainment on heat exchanger thermal efficiency

When drops are entrained from the heat exchanger and evaporate, the extraction of water-soluble components decreases. To show to what extent re-entrainment effects the heat transfer efficiency of heat exchangers operating in cascade a case study is presented in this paragraph. The dependence of heat flux on gas mixture inlet temperature and humidity, which has been determined in §2.4.2, is used to determine the effect of re-entrainment.

Suppose that two heat exchangers are operated in a cascade arrangement, see fig. 2.5.1.

![Diagram of two heat exchangers in a heat exchanger array](image)

*Fig. 2.5.1 Schematic of two heat exchangers in an heat exchanger array. Calculation (see main text) shows that re-entrainment reduces effective heat and mass transfer rates*

Gas leaving the upstream (number II) heat exchanger has a temperature 82°C and an absolute humidity of 17%. Coolant flow rate though both heat exchangers is 0.47 kg/m²s. The gas mixture mass flux through both heat exchangers is 0.26 kg/m²s.

When this gas mixture enters the downstream heat exchanger (number III), the amount of condensate that is formed is 4.35 g/m²s. We assume that an identical amount of condensate is formed in the heat exchanger number II, and that 10 % of this condensate is re-entrained into the gas stream. Evaporation of this liquid increases the absolute humidity of the gas mixture by 0.22%, which increases the
heat flux from gas to coolant in heat exchanger III. However, the overall heat transfer rates in the downstream heat exchanger decreases, because the temperature of the gas entering the downstream heat exchanger is lowered. This will now be calculated.

The change in temperature of the vapour entering heat exchanger III is calculated assuming adiabatic evaporation of water. The heat that is released upon evaporation is:

\[ Q = Q_{\text{vap}} \cdot d m_{\text{vap}} \]  \hspace{1cm} 2.5.1

where \( Q \) is the amount of released heat, \( Q_{\text{vap}} \) is the heat of vaporisation of water and \( dm_{\text{vap}} \) is the mass flux of a small amount of water that evaporates. The change in gas temperature is given by 2.4.2:

\[ Q = c_{p,\text{gas}} \cdot m_{\text{gas}} \cdot dT \]  \hspace{1cm} 2.5.2

where \( c_{p,\text{gas}} \) is the specific heat of the gas mixture, \( m_{\text{gas}} \) is the gas mass flux and \( dT \) is the decrease in gas temperature.

Because the amount of released heat in 2.5.1 and 2.5.2 are the same, we find:

\[ \frac{m_{\text{gas}} \cdot dT}{dm_{\text{vap}}} = \frac{dT}{dx_{\text{gas}}} \cdot \frac{Q_{\text{vap}}}{c_{p,\text{gas}}} \]  \hspace{1cm} 2.5.3

The ratio \( dm_{\text{vap}}/m_{\text{gas}} \) is the change in mass fraction water vapour in the gas \( (dx_{\text{gas}}) \) due to evaporation of \( dm_{\text{vap}} \). The absolute humidity is the volume fraction water vapour.

\[ x_{\text{gas}} = \frac{H_a}{H_a + (1 - H_a)} \cdot \frac{P_{\text{air}}}{P_{\text{H2O}}} \]  \hspace{1cm} 2.5.4

The quotient of the densities of air and pure water vapour is 1.6. At an absolute humidity of 17%, \( dH_a/dx \) equals 0.713, or 71.3% \( dT/dH_a \) follows from the ratio of the specific heat of the gas stream and the heat of vaporisation of the water:

\[ \frac{dT}{dH_{a,\text{in}}} = \frac{Q_{\text{vap}}}{c_{p,\text{gas}}} \cdot \frac{dx_{\text{in}}}{dH_{A,\text{in}}} \bigg|_{p,T} = -31.7^\circ C / \% \]  \hspace{1cm} 2.4.5

This means that the temperature of the gas mixture at 80°C decreases with 3.17°C when adiabatic evaporation of water results in an increase in absolute humidity of 0.1%.

The change of heat transfer rate is calculated using:
Re-entrainment in compact heat exchangers

\[
\Delta Q = \left[ \frac{\partial Q}{\partial H_{\text{in}}} + \frac{\partial Q}{\partial T_{\text{m}}} \right] \Delta H_{\text{in}}
\]

The change in heat transfer rate with changing temperature and humidity have been determined in paragraph 2.5.2.

substitution of \[ \frac{\partial Q}{\partial H_{\text{in}}} \bigg|_{T_{\text{m}}, m_{\text{cool}}} = 0.73 \text{kW/m}^2\% \] , \[ \frac{\partial Q}{\partial T_{\text{m}}} \bigg|_{H_{\text{in}}, m_{\text{cool}}} = 0.17 \text{kW/m}^2\circ C \] gives:

\[
\Delta Q = -4.66 \cdot \Delta H_{\text{in}} \text{[kW/m}^2]\]

substitution of the change in absolute humidity (0.22%) gives: \( \Delta Q = -1.02 \text{kW/m}^2\)

The measured heat transfer rate at a temperature of 82\(^\circ\)C and an absolute humidity \( H_{\text{a,in}} \) of 17% is 14.7kW/m\(^2\); the heat transfer rate is thus reduced by 7%.

Re-entrainment from heat exchanger II also increases the re-entrainment from heat exchanger III. The increased vapour humidity in the downstream heat exchanger will increase the amount of condensate that is formed by about 8%. Because the amount of re-entrained condensate increases with increasing condensate flow rates, even more condensate will be entrained from the downstream edge of heat exchanger III. It is quite common in industry to operate heat exchangers in cascade. Re-entrainment can thus seriously decrease overall heat transfer rates.

2.5.2 The impact of re-entrainment on the use of PVDF heat exchanger for gas cleaning

As shown in paragraph 2.4.1, drops are entrained out of the heat exchanger when both gas velocity and the amount of condensate in the heat exchanger are large. In this paragraph, a test case is presented in which the adsorption of an industrial pollutant, HF, in the condensate in a PVDF heat exchanger is calculated. As in paragraph 2.4.4, we assume that 10% of the condensate is re-entrained. Again, we calculate the amount of hydrogen fluoride (HF) in the condensate. This example shows the importance of re-entrainment on the gas cleaning performance of the PVDF heat exchanger.

The concentration of water-soluble non-condensable gases is based on typical conditions of a waste gas stream leaving the furnace of a ceramics factory, as presented by C.Simons [39]. The partial pressure of hydrogen fluoride in the gas stream should be reduced from 14 to 1Pa.

A graphical presentation of the test case is given in fig. 2.5.2.
In this example, inlet temperature and humidity of the heat exchanger are comparable to the conditions in paragraph 2.4.1: \( m_{\text{g, in}} = 0.26 \text{ kg/m}^2\text{s}, T_{\text{in}} = 82^\circ \text{C}, H_a,\text{in} = 16.5\%, T_{\text{cool, in}} = 25^\circ \text{C}, m_{\text{cool}} = 0.47 \text{ kg/m}^2\). These conditions are not too different from operational conditions that could be encountered in an industrially operated heat exchanger (C. Simons).

Measured outlet gas temperature, humidity and condensate flow rate are:

\( T_{\text{out}} = 65.6^\circ \text{C}, H_a,\text{out} = 14.66\%, m_c = 6.3 \text{ g/s} \)

At these conditions, hydrogen fluoride inflow should be reduced from 37.9 mg/m\(^2\)s to 2.71 mg/ m\(^2\)s.

Estimated average droplet temperature in the heat exchanger is determined in paragraph 2.4.3, 48\(^\circ\)C. The solubility of hydrogen fluoride is determined using a low concentration approximation for dissociating vapour. The amount of non-dissociated hydrogen fluoride is given by the classical Henry equation:

\[
p_{\text{HF}} = H \cdot x_{\text{HF, nondiss}}.
\]

where \( p_{\text{HF}} \) is the partial pressure (in bar) over a saturated aqueous hydrogen fluoride solution, \( H \) is the Henry constant of hydrogen fluoride and \( x_{\text{HF, nondiss}} \) is the (non-dimensional) mass fraction of non-dissociated Hydrogen fluoride.

The amount of dissociated HF is given by 2.5.2:

\[
x_{\text{HF, diss}}^2 = C_{\text{diss}} x_{\text{HF, nondiss}}.
\]

where \( C_{\text{diss}} \) is the dissociation constant of HF in water.

Combination of 2.5.1 and 2.5.2 gives 2.5.3:

\[
p_{\text{HF}} = H \cdot C_{\text{diss}} \cdot x_{\text{HF, diss}}^2.
\]

For diluted solutions \( (x_{\text{HF}} < 0.01) \), the amount of dissociated HF is much (at least 100 times) larger than the amount of non-dissociated HF. This gives:

---

*Fig. 2.5.2 Schematic of gas cleaning test case. Hydrogen fluoride partial pressures are calculated later in this paragraph.*
Re-entrainment in compact heat exchangers

\[ P_{HF} = H^* x_{HF} \]  \hspace{1cm} 2.5.4

where \( P_{HF} \) is the partial pressure of hydrogen fluoride, \( H^* \) is the pseudo Henry coefficient (\( H^* \)) and \( x_{HF} \) is the mass fraction of dissociated HF in solution.

\( H^* \) is temperature dependant. Saturation pressures of hydrogen fluoride at various mass concentrations and temperatures are given by Kirk-Othmer [23].

Equation 2.5.5 is determined from a least squares fit. Difference between reported and calculated partial pressures are less than 5%.

\[ \log H^* = a + bT + cT^2 \]  \hspace{1cm} 2.5.5

with \( a = 13.958 \) Pa, \( b = 0.887 \) Pa/\( ^{\circ} \)C and \( c = -0.0246 \) Pa/\( ^{\circ} \)C\(^2\).

The amount of grams of hydrogen fluoride per cubic meter gas is calculated using the ideal gas law:

\[ m = M(n/V) = M(p/R(T + 273.1)) \]  \hspace{1cm} 2.5.6

where \( M \) is the molar mass of hydrogen fluoride (HF), 20 g/mole, \( n/V \) is the molar concentration in the gas mixture and \( R \) is the universal gas constant, 8.314 J/(mole K).

At 48\(^{\circ} \)C, the pseudo-Henry constant \( H^* \) of hydrogen fluoride (pseudo, because dissociation is taken into account) equals 2710Pa

The amount of hydrogen fluoride in the vapour and in the condensate is calculated, assuming equilibrium concentrations in the condensate. The validity of this assumption has been shown in paragraph 2.4.3.

The hydrogen fluoride mass balance and the Henry equation are solved simultaneously, which gives the HF concentration in the condensate and the partial pressure in the air mixture leaving the heat exchanger. Calculated values are:

\[ C_{HF} = 6.0 \times 10^{-3} \]
\[ m_{\text{CHF}} = 37.7 \text{ mg/s} \]
\[ p_{HF, \text{out}} = 60 \text{ mPa} \]
\[ m_{\text{air,HF, out}} = 0.26 \text{ mg/s} \]

percentage extracted hydrogen fluoride: 99.3%

The partial pressure of HF in the air mixture leaving the heat exchanger is 60 mPa. This shows that the PVDF heat exchanger is very well suited to clean this gas stream. When 10% of the re-entrained condensate evaporates, the partial pressure in the gas stream increases with 10% of the amount of hydrogen fluoride in the condensate to 1.4Pa, and the emission reduction goal of hydrogen fluoride to a partial pressure in the air mixture of 1Pa is no longer achieved.
Chapter 3: Pinch-off of a water drop from a horizontal hydrophobic plate

3.1 Introduction

3.1.1 Background
In the previous chapter, heat and mass transfer in a PVDF compact plate heat exchanger have been treated. Although both the equations on heat and mass transfer and substance properties like thermal conductivity of PVDF, water and air are well known, it proved to be impossible to fully predict overall mass and heat transfer rates. The reason for this is twofold. First of all, water condenses dropwise on a PVDF plate. Drops on heat exchanger plates limit heat transfer, and drop topology influences the efficiency with which water-soluble substances (in our experiments ammonia) dissolve. Because the size distribution of the drops and the wetted area cannot be predicted, it is impossible to calculate the heat transfer rate a priori.

Secondly, drops break off the heat exchanger plates, causing re-entrainment of water into the gas stream. Size, location and initial velocity of the drops determine whether the re-entrained drops settle within a short distance after re-entrainment, are swept out of the heat exchanger and settle in the next bend or restriction, or evaporate completely. This makes it impossible to predict composition and temperature of the gas leaving the heat exchanger.

The second problem, the re-entrainment of liquid in a plate heat exchanger, will be investigated in this chapter.

Re-entrainment
Drops that are formed during dropwise condensation drain from the plate and are sometimes re-entrained into the gas stream. This phenomenon has been observed in the experiments presented in the previous chapter. Re-entrainment of drops lowers the thermal efficiency of the heat exchanger and possibly damages downstream equipment.

Furthermore, re-entrainment of drops greatly reduces the efficiency with which water-soluble pollutants are extracted from the gas entering the heat exchanger. Re-entrained drops are heated by the gas leaving the heat exchanger and evaporate when remaining in the gas long enough. As drops are heated, the solubility decreases. The solubility of hydrofluoric acid decreases for example by 70% between 40 and 60°C. When the drop evaporates all dissolved pollutants are released into the gas stream. A case study presented in paragraph 3.5 and 3.6 shows that 10% entrainment results in 7% loss in heat transfer rate and increases the amount of hydrogen fluoride in the gas stream leaving the heat exchanger from
0.06 to 1.4 Pa. In a compact plate heat exchanger, re-entrainment is thus a highly undesirable phenomenon.

The effect of re-entrained fluid on process efficiency and downstream equipment depends on the amount of fluid and the size distribution of the drops that leave the heat exchanger. In general, smaller drops are harder to collect and evaporate faster, making small drops less desirable.

The ideal situation in the heat exchanger consists of drops of minimal size on the plates and no fluid re-entraining into the gas stream. When fluid does re-entrain, ideally the drops should be as large as possible.

3.1.2 Goals
No correlation of drop size after re-entrainment from hydrophobic plates by gravity and vapour shear forces has been found in literature, nor by which mechanism liquid is re-entrained in the gas stream. Experiments have been performed to answer the following questions:
- What is the governing mechanism of droplet formation during re-entrainment in the pilot plant test rig?
- What is the size of re-entrained drops?
- Can modifying the shape of the downstream edge of the heat exchanger plate change the average size of re-entrained drops?

3.1.3 Structure of this chapter
In section 3.2, a method is constructed to calculate the forces acting on a drop. A single plate mock up to investigate pinch-off from hydrophobic plates is described in section 3.3. Experimental results are presented in section 3.4. In the first paragraph (3.4.1), observations on the pinch-off of sliding drops are presented. Observations on the pinch off of a slowly growing drop are presented in paragraph 3.4.2. Various forces acting on a drop during pinch-off of three different plates are calculated and presented in paragraph 3.4.3 - 3.4.6. These are used to calculate drag force coefficients of drops during pinch-off. The results of the experiments using the three edge shapes are presented in paragraph 3.4.7.

3.2 Calculation of forces acting on a drop on a hydrophobic plate.
The various forces that act on a drop on a heat exchanger plate will be calculated in this paragraph. The results are used in paragraphs 3.4 and 4.3, where observed shapes 0and velocities of drops in various situations are presented. These situations are:
1. drop, sliding over a plate
2. drop, breaking off from the downstream edge of a plate
3. pendant drop in quiescent vapour
Various forces acting on a drop are calculated to gain insight into these processes. The goal of this chapter is to construct an equation to describe deformation of drops that rest on, slide over or break off a hydrophobic flat surface. The plate orientation is free; it can be both horizontal, vertical or at an arbitrary angle with respect to the earth’s gravitational field. First, equations describing the drop shape are constructed. Three variables are used to define drop shape: height of the drop, radius of the drop foot and (dynamic) contact angle. Independent variables are for example drop volume and mass density, orientation of the plate, vapour velocity and liquid and vapour viscosities.

Using this shape, forces working on the drop are calculated. Liquid and vapour shear forces, pressure forces, gravity and surface tension are taken into account. The Navier-Stokes equation is used to construct a relationship based on these forces for acceleration of the drop:

\[
\frac{dv_{\text{drop}}}{dt} = f(\text{drop volume & shape, plate orientation, } v_{\text{drop}}, v_{\text{v}}, \cdots) \tag{3.2.1}
\]

where \(v_{\text{drop}}\) is the average drop velocity. Application of this equation to specific situations will allow e.g. calculation of the maximum size of a pendant drop in quiescent vapour (§3.2.8) or hydrodynamic forces from experimental data (§3.4).

In the next paragraph (§3.2.1), the shape of a drop on a plate is determined. This analysis is based on results published by Stauffer and Chatterjee. In §3.2.2, The Navier stokes equation and the equation of continuity are used to construct an equation of the form of 3.2.1. The unknowns of this equation are the forces working on the drop. These are calculated in §3.2.3-3.2.7. In section 3.2.8, the maximum size of a pendant drop is calculated. In the last section (§3.2.9), the shape and velocity of the drop are used to calculate the following components of the drop free energy: potential energy, surface free energy and kinetic energy.

Throughout this chapter, the following assumptions will be used:
1. Mass density of both the liquid in the drop and the vapour outside the drop are constant
2. Viscosity of liquid and vapour are constant
3. Total mass of the drop is constant in time
4. Surface tension coefficient is constant

### 3.2.1 Drop shapes

An equation defining the contour of a drop on a horizontal flat plate in quiescent vapour has been derived by Stauffer [C.E. Stauffer, J. Phys. Chem. 69(6) (1965) 1933-1938]:

\[
\frac{2\pi}{b} = \sigma \left[ \sin(\phi) + \frac{d\sin(\phi)}{dx} \right] + \Delta \rho g z \tag{3.2.2}
\]
where \( x \) is the horizontal co-ordinate of a point on the surface of a drop, \( \sigma \) is the surface tension, \( b \) is the curvature at the tip of the drop, \( \sin \phi/x \) and \( d/dx(\sin \phi/x) \) are the inverses of the curvatures of the drop in direction of observation (perpendicular to the paper) and perpendicular to it. \( \Delta \rho = (\rho_l - \rho_v) \) is the mass density difference between the fluids inside (\( \rho_l \)) and outside the drop (\( \rho_v \)) and \( z \) is the height to the top of the drop. The gravitational constant \( g \) is negative, since gravity points downward. To derive equation 3.2.2, it has been assumed that no vapour flows around the drop. A graphical impression of the drop and the defining quantities is given in fig. 3.2.1.

\[
\cos(\alpha) = \frac{\sigma_{sg} - \sigma_{sl}}{\sigma_{lg}}
\]

where \( \alpha \) is the contact angle between liquid-vapour interface and a solid surface, measured in the liquid, and the subscripts \( sg \), \( sl \) and \( lg \) refer to the solid-gas, solid-liquid and liquid-gas interfaces.

Combining eq. 3.2.2 and 3.2.3, and taking into account that the contact angle of water on PVDF equals 90º, fully defines the shape of a stationary drop in quiescent vapour.

A method by Chatterjee [J.Chatterjee, Colloids and Surfaces A 178 (2001) 249-263] allows explicit calculation of the drop contour, based on a Bond number with respect to the curvature of the drop apex. According to Chatterlee, only a small error is made when the contour is estimated by an ellipse, using:
3.2 forces acting on a drop

\[ y(x) = \frac{1}{B} \sqrt{1 - \left(1 - B(x/b)^2\right)} \]  

where B is the shape factor, which is a function only of the Bond number with respect to the curvature of the drop apex (Bo_{tip}):

\[ Bo_{tip} = \frac{b^2 \Delta \rho g}{\sigma} \]  

Using an error minimisation technique, the dependence of shape factor on Bond number (Bo_{tip}) has been determined by Chatterlee, resulting in a graphical presentation of B(Bo_{tip}). The disadvantage of this method is, that the curvature of the drop apex is not known a priori. Thus, the shape of the contour has to be determined to calculate the curvature at the apex. Only after this, the shape of the contour can be calculated.

To overcome this problem, the shape factor is determined as a function of the drop volume. First, the average drop radius is defined as the radius of a perfect hemispherical drop of identical volume:

\[ r_{eq} = \frac{3V}{2\pi} \]  

The drop volume is given by:

\[ V = \frac{2}{3} \pi r_f^2 h = \frac{2}{3} \pi b^3 / B^2 \]  

where h is the height of the drop and \( r_f \) is the radius of the foot of the drop. h and \( r_f \) follow from 3.2.4:

\[ r_f = b / \sqrt{B} \]  
\[ h = b / B \]  

substitution of 3.2.6 to 3.2.8 in 3.2.5 gives:

\[ b = r_{eq} B^{2/3} \]  
\[ r_f = r_{eq} B^{1/6} \]  
\[ h = r_{eq} B^{-1/3} \]  

the Bond number with respect to the drop volume is defined by:

\[ Bo = \pm \frac{r_{eq}^2 \Delta \rho g}{\sigma} \]
where Bo is positive when the drop rests on a plate and negative when the drop is pendant. Using 3.2.4 and 3.2.9, the Bond number with respect to the drop volume is calculated:

\[ Bo = \pm Bo_{tip} B^{-4/3} \] 3.2.13

In fig. 3.2.2, the inverse of the shape factor B, calculated by Chatterjee, is plotted against the Bond number, calculated using eq. 3.2.13.

Equation 3.2.14 gives an approximation of B with a relative error of less than 5% for Bond numbers between \(-3\) and 2. The error of 5% is an estimate of the error of the method of Chatterlee, not from lack of fit of Chatterlee's data using 3.2.14.

\[
B = \begin{cases} 
1 + 2Bo & \text{Bo} > 0 \\
1/(1 - 2Bo) & \text{Bo} < 0 
\end{cases}
\] 3.2.14

For \(Bo=0\), shape factor (B) equals unity and height and foot radius are identical. For larger absolute values of the Bond number (larger drops), the radius of the drop foot (r) and height (h) are given by 3.2.9 and 3.2.10.

Pendant drops, for which Bond numbers are negative, have a larger height than radius. Drops, resting on a plate, have a height that is less than the radius of the drop foot.

When both drop height and the radius of the foot are known, the shape of the drop is fully fixed:

\[
\left(1 - \frac{y}{h}\right)^2 + \left(\frac{x}{r_{eq}}\right)^2 = 1
\] 3.2.15
Some typical predicted drop shapes at various Bond numbers are presented in fig. 3.2.3

![Fig. 3.2.3 Shape of pendant or resting drops at various Bond numbers. Negative Bond numbers represent pendant drops.](image)

The method that has just been described cannot be applied when other forces besides gravity and surface tension work on the drop, or when gravity is not working perpendicular to the plate surface. This is, for example, the case when a drop is placed in flowing vapour or during break-up. Here, the following approach is followed.

When a force is exerted on a drop, the interface is deformed. An example of a stationary drop that is deformed by vapour shear forces is given in fig. 3.2.4.

![Fig. 3.2.4 Schematic of the shape of a drop on a horizontal hydrophobic plate. Drop is deformed by shear forces of vapour flowing over the plate](image)

Vapour shear forces change the contour of the liquid-vapour interface. As a result, the apparent contact angle no longer satisfies Young's law. A generally excepted explanation [1,2,3] for this phenomenon is that variations in contact angle are caused by inhomogeneities of the solid surface. Phenomena that can cause surface inhomogeneity are:

- roughness
non-constant composition due to:
- reactions at the surface
- mobility of hydrophilic and hydrophobic chains (in polymers)
- preferent (inhomogenous) orientation of polymer chains

For a PVDF surface, roughness is expected to be the main contributor to the difference between observed contact angles and the contact angle given by Young's equation. When the contact line is stationary, surface roughness allows for a difference between macroscopic and microscopic contact angle (fig 3.2.5).

Fig. 3.2.5 microscopic and macroscopic contact angles difference caused by surface roughness

Macroscopically, the contact angle is a free variable even when on a microscopic scale the contact angle is fixed.

At maximum deformation, contact angle at the upstream side of a stationary drop (when the drop is deformed by vapour shear forces) is 0°, while at the downstream side the contact angle is 180°. These dynamic contact angles have been observed and are presented in §3.4.3.

A second effect of surface roughness is that it pins down the contact line. Thus, when vapour shear forces start working on a drop, this will not change the shape of the foot of a drop. It is therefore safe to assume that the radius of a stationary drop in flowing vapour can still be calculated using eq. 3.2.6 and 3.2.7. The height of the tip of the drop can change as a result of lift forces, working on the drop. The change in height must be small, because drop radius and volume are constant. We can thus assume that equations 3.2.10 and 3.2.11 correctly predict the radius and height of a stationary drop in flowing vapour.
3.2 forces acting on a drop

3.2.2 Equations on conservation of mass and momentum

In this paragraph, the well-known equations of continuity, mass and momentum transfer are used to construct an equation to calculate drop movement. This equation will have the form of 3.2.1:

$$\frac{d\bar{v}_{\text{drop}}}{dt} = f(\text{drop volume \& shape, plate orientation}, \bar{v}_{\text{drop}}, \bar{v}_v, \cdots)$$

3.2.1

Mass conservation

The equation of continuity is given by [19,25]:

$$\frac{\partial \rho}{\partial t} = -\nabla \cdot (\rho \bar{v})$$

3.2.16

both inside and outside a droplet, we assume that density is constant. In this way, we find:

$$\nabla \cdot \bar{v} = 0$$

3.2.17

At the liquid-vapour interface, density changes from the density of the liquid to the density of the vapour. Because the density is not constant at the interface, 3.2.17 is not valid here. Two different approaches can be followed. In reality, density changes fast but continuously in the interface. Density gradients in the interface have been predicted by e.g. Van der Waals using a diffuse interface method. This method will be applied in chapter 10 on experiments done in the critical pressure test rig. In this paragraph, a different method is applied.

Here, it is assumed that the liquid-vapour interface is 2-dimensional, i.e. has zero thickness. Surface tension works in this plane, contracting the interface. Substance properties like density and viscosity change step-wise at the interface.

An arbitrary part of an interface is drawn in fig. 3.2.6.

![Fig. 3.2.6 Schematic of a moving liquid-vapour interface. Arrows denote fluid and interfacial velocity.](image)
Fluid velocity at the interface is presented using solid arrows. When the fluid moves normal to the interface, fluid velocity and interfacial velocity are identical. When the fluid moves tangent to the interface, the interface is stationary. The interfacial velocity is given by:

\[ v_{\text{int}} = \hat{n}(\mathbf{v}_l \cdot \hat{n}) \]  \hspace{1cm} 3.2.18

where \( \hat{n} \) is the unit vector normal to the liquid vapour interface, facing in the direction of the vapour.

**Momentum conservation**

Momentum transport is given by the Navier-Stokes equation. (3.2.19)

\[
\frac{\partial \mathbf{v}}{\partial t} = -\nabla (\rho \mathbf{v} \mathbf{v}) - \nabla \rho + \mathbf{\tau} - \mathbf{\tau} + \rho \mathbf{g}
\]  \hspace{1cm} 3.2.19

The first term is the local change in momentum. The second term incorporates momentum transport with respect to a fixed point in space. Third and fourth term account for pressure and shear stresses, working on the fluid and the fifth term accounts for body forces. Here, gravity is the only body force that is considered.

Since density of vapour and liquid are constant, we may write:

\[
\frac{\partial \mathbf{v}}{\partial t} = -\nabla (\rho \mathbf{v} \mathbf{v}) - \frac{1}{\rho} \mathbf{v} \mathbf{p} + \frac{1}{\rho} \mathbf{\tau} + \mathbf{g}
\]  \hspace{1cm} 3.2.20

The interest here is to describe movement of the drop. Thus, we follow the fluid, using the substantial derivative:

\[
\frac{D\mathbf{v}}{Dt} = -\frac{1}{\rho} \mathbf{v} \mathbf{p} + \frac{1}{\rho} \mathbf{\tau} + \mathbf{g}
\]  \hspace{1cm} 3.2.21

where \( \frac{D}{Dt} \) is defined by:

\[
\frac{D}{Dt} = \frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla
\]

Integrating 3.2.21 over the entire drop gives:

\[
\int_V \frac{D\mathbf{v}}{Dt} dV = -\frac{1}{\rho} \int_A \mathbf{p}dS - \frac{1}{\rho} \int_A \hat{n} \cdot \mathbf{\tau} + \mathbf{g}V
\]  \hspace{1cm} 3.2.22

\( \frac{D\mathbf{v}}{dt} \) is the change of fluid velocity in time, moving along with the fluid. Averaging this over all liquid fluid, this equals the change in average liquid velocity:

\[
\int_V \frac{D\mathbf{v}}{Dt} dV = \frac{d}{dt} \int_V \mathbf{v} dV = \mathbf{V} \frac{d\mathbf{v}_m}{dt}
\]  \hspace{1cm} 3.2.24
3.2 forces acting on a drop

where the average drop velocity is defined by:
\[
\bar{v}_m = \frac{1}{V} \int \bar{v} dV
\]  \hspace{1cm} \text{3.2.25}

The subscript m refers to the centre of mass of the drop: the average drop velocity equals the velocity of the centre of mass of the drop. The volume \( V \) in the \( 1/V \) and the integral refers to the drop volume. Substituting 3.2.24 in 3.2.21 gives 3.2.26. Note that 3.2.26 has the form of 3.2.1.

\[
V \frac{d\bar{v}_m}{dt} = -\frac{1}{\rho} \int_{\mathcal{A}} \mathbf{p} n dS - \frac{1}{\rho} \int_{\mathcal{A}} \bar{n} \cdot \tau + gV
\]  \hspace{1cm} \text{3.2.26}

The first term in 3.2.26 is the change in average velocity of the drop. Second term is the total pressure working on the drop. This term vanishes when the pressure outside the drop is constant. The third term is the total of stresses working on the drop surface. The last term gives gravitational forces due to the drop mass.

3.2.3 Forces working on drops resting on a flat solid surface

To use equation 3.2.26, the forces that work on a drop have to be calculated. Three different types of forces are taken into account: body forces, pressure forces and shear- and surface tension related stresses. It is assumed that the only body force that works on a drop is gravity; its value and direction are given by multiplying the drop mass with the gravitational vector \( \mathbf{g} \). Pressure forces and stresses work on the outer surface of the drop. To correctly calculate these forces the system boundary must be defined explicitly. This is done in the first part of this paragraph. After this, the pressure forces working on the system boundary are calculated. Next, the reversible stresses (surface tension) and the irreversible stresses (shear stresses) are calculated.

Definition of system boundary and unit vectors

A schematic of the 3-dimensional shape of a drop on a flat plate over which vapour flows has been presented in fig. 3.2.4. Drop height and the radius of the foot of the drop do not need to satisfy 3.2.10 and 3.2.11. The contact angle is not constant but may be a function of position at the contact line.

To calculate forces working on a drop, a convenient system boundary is chosen, see fig. 3.2.6. At the liquid-vapour interface, the system boundary falls just outside the drop. At the foot of the drop, the system boundary cuts through the fluid at short distance from the plate.

Two unit vectors, presented in fig. 3.2.7, are \( \bar{n} \) and \( \bar{m} \).
\( \hat{n} \) is the unit vector normal to the system boundary, pointing out of the enclosed volume. \( \hat{m} \) is the unit vector normal to the plate, facing out of the plate. At the foot of the drop, \( \hat{n} \) and \( \hat{m} \) point in opposite direction. The co-ordinate system is defined with respect to the plate, see fig. 3.2.8.

The height \( h(x,z) \) is defined at any position in the foot of the drop as the distance from a point at the liquid-vapour interface at \( (x,z) \) to the plate.
3.2 forces acting on a drop

Three other unit vectors will also be used in this chapter. These are:

- \( \mathbf{k} \) unit vector in downstream (x) direction
- \( \mathbf{i} \) unit vector in the x-y plane, tangent to the contact line.
- \( \mathbf{j} \) unit vector in y-direction

### 3.2.4 Pressure working on system boundary

To calculate the pressure working on the system boundary, the boundary is split into 2 parts: the liquid-vapour interface and the liquid-plate interface:

\[
\int_{A} p \, dS = \int_{A_{lv}} p_{lv} \, dS + \int_{A_{sl}} p_{sl} \, dS  \tag{3.2.27}
\]

First, the pressure at the liquid-vapour boundary will be calculated. Next, the pressure at the solid-liquid boundary is calculated.

#### Pressure outside the liquid-vapour interface

The pressure working on the system boundary at the liquid-vapour interface is the pressure outside the drop. Far upstream of the drop, influence of the flow around the drop can be ignored and the vapour velocity is parallel to the plate. Since the vapour velocity normal to the plate is zero, pressure gradients in a cross-section through the slit, normal to the plate, depend only on density gradients.

At the outside of the liquid-vapour interface, pressure differs from an upstream position at the same height due to the hydrodynamic dissipation over the channel and flow restriction by the drop. When the distance between the "upstream point" and the drop is not too large, the streamwise pressure gradient due to flow restriction by the drop is much larger than the pressure gradient due to hydrodynamic dissipation of the well-developed flow.

The pressure at the liquid-vapour system boundary, \( p_{lv} \), is thus given by:

\[
\hat{p}_{lv}(\hat{x}_{lv}) = p_{0} + \rho_{v}(\hat{x}_{lv} \cdot \hat{g}) + \Delta p_{r,\text{out}}(\hat{x}_{lv})  \tag{3.2.28}
\]

For a pendant drop, \( \hat{x}_{lv} \cdot \hat{g} > 0 \). When the drop lies on a plate, as in fig. 3.2.9, \( \hat{x}_{lv} \cdot \hat{g} < 0 \) where \( p_{0} \) is the pressure at the "upstream point".

#### Pressure in the drop near the liquid-solid interface

Next, a relationship for the pressure in the drop near the liquid-solid interface is determined, based on the pressure outside the drop. The pressure on the inside of the liquid-vapour interface is higher than the pressure at the liquid-vapour boundary, outside the interface, by \( \alpha(1/r_{1}+1/r_{2}) \), where \( r_{1} \) and \( r_{2} \) are the local radii of curvature of the interface and normal stresses due to viscous flow are ignored. The average radius of curvature is defined by:
We shall now show that it is possible to calculate the pressure difference between two points in the fluid \( \tilde{x}_1 \) and \( \tilde{x}_2 \) with a small error using only the static pressure difference, \( \rho_l (\tilde{x}_2 - \tilde{x}_1) \cdot g \). The error is caused by hydrodynamic friction in the drop.

Pressure gradients due to frictional forces are given by:

\[
\nabla p_f = \nabla \cdot \tau = \nabla \cdot (\eta \nabla v)
\]

To show that the pressure difference due to hydrodynamic friction can be ignored, take a water drop of 1 mm in diameter on a horizontal plate. Assume that velocity differences in the drop are large, say 10 cm/s. The vertical pressure difference over the drop due to gravity equals:

\[
\Delta p_g = \rho_l g \Delta x = 1000 \cdot 10 \cdot 0.001 = 10 \text{ Pa}
\]

The maximal pressure difference due to viscous flow is:

\[
\Delta p_{f,\text{max}} = \eta \frac{\Delta v_{\text{max}}}{\Delta x} = 0.001 \cdot \frac{0.1}{0.001} = 0.10 \text{ Pa}
\]

The pressure difference due to viscous flow will be much smaller than the value, calculated in 3.2.31, because liquid flows both up and downward. In fig. 3.2.9, a circulating liquid is drawn schematically in the drop.

---

**Fig. 3.2.9 Schematic of recirculating liquid flow in a drop.** \( h_l \) is the height of the liquid-vapour interface at \((x,y)\). Gravity can work in arbitrary direction.
To the left of the drop, liquid flows upward and the pressure at the foot is underestimated. At the right side of the drop, liquid flows down and the pressure at the foot is overestimated. When the external force that works on the foot is calculated by integrating the pressure over the foot, these terms partially cancel out, reducing the influence of viscous flow even further.

The above example shows that even when liquid circulation velocities in a 1mm drop are as high as 10 cm/s, vertical pressure differences due to viscous flow can be ignored when the pressure at foot of a drop is to be calculated.

Resuming, the pressure at the solid-liquid boundary \( (p_{sl}) \) is calculated from the pressure at a point on the liquid-vapour boundary by:

\[
p_{sl}(x_{sl}) = p_{lv} + \rho_l (x_{sl} - x_{lv}) \cdot g + 2\sigma / r_{lv}
\]

where \( r_{lv} \) is the mean radius of curvature at the point \( x_{lv} \). For convenience, we choose the point \( x_{lv} \) on the liquid-vapour boundary in such a way that both the \( x \) and \( y \)-co-ordinates of \( x_{sl} \) and \( x_{lv} \) are identical:

\[
x_{sl} - x_{lv} = -h_{lv} m
\]

Substitution of eq. 3.2.33 and 3.2.28 in eq. 3.2.32 allows calculation of the pressure at any point on the liquid-solid boundary:

\[
p_{sl}(x_{sl}) = p_0 + \rho_v (x_{lv} \cdot g) + \Delta p_{lv, out} (x_{lv} \cdot g) - \rho_l h_{lv} (\tilde{m} \cdot g) + 2\sigma / r_{lv}
\]

where \( x_{lv} \) is the point in the liquid-vapour interface defined by:

\[
x_{lv} = x_{sl} + h_{lv} \tilde{m}
\]

Equations 3.2.28 and 3.2.34 are expressions for the pressure at every point on the liquid-vapour boundary and the solid-liquid boundary. These enable us to compute the forces working on the drop due to pressure differences outside the drop. Substitution of 3.2.28 and 3.2.34 in 3.2.27 gives:

\[
\int_{A} p dS = \int_{A_{lv}} [\tilde{n}[p_0 + \rho_v (x_{lv} \cdot g) + \Delta p_{lv, out} (x_{lv} \cdot g)] dS + \int_{A_{sl}} [\tilde{n}[p_0 + \rho_v (x_{lv} \cdot g) + \Delta p_{lv, out} (x_{lv} \cdot g) - \rho_l h_{lv} (\tilde{m} \cdot g) + 2\sigma / r_{lv}] dS
\]

where \( A \) is a closed surface around volume \( V \).

At this point, the force due to the non-uniform pressure at the system boundary is fully determined. The remainder of this paragraph is used to transforming eq 3.2.35 into a form that is easier to use.

First we use the well-known fact that the integral over a closed surface of a constant vector normal to the interface is zero. Furthermore, eq 3.2.35 yields:
Substituting eq. 3.2.36 in eq. 3.2.35 gives:

\[
\int_A \tilde{n}(\bar{x}_v \cdot \tilde{g}) \, dS + \int_A \tilde{n}(\bar{x}_v \cdot \tilde{g}) \, dS = \int_A \tilde{n}(\bar{x} \cdot \tilde{g}) \, dS + \int_A \tilde{n}(\bar{m} \cdot \tilde{g}) \, dS = V \tilde{g} - \bar{m}(\bar{m} \cdot \tilde{g}) V 3.2.36
\]

where \( \bar{m} \) is the unit vector perpendicular to the plate, pointing into the liquid drop. Substituting eq. 3.2.36 in eq. 3.2.35 gives:

\[
\int_A \rho \, dS = \int_A \tilde{n}(\bar{x}_v \cdot \tilde{g}) \, dS + \int_A \tilde{n}(\bar{m} \cdot \tilde{g}) \, dS + \int_A \tilde{n}(\bar{m} \cdot \tilde{g}) \, dS = V \tilde{g} - \bar{m}(\bar{m} \cdot \tilde{g}) V 3.2.37
\]

The following abbreviations are used for the first and second term of the right-hand-side expression in eq. 3.2.37:

\[
R^{-1} = \frac{1}{A_{ls}} \int_A r^{-1}_v \, dS 3.2.38
\]

\[
\bar{F}_{p, out} = -\int_A \tilde{n}(\Delta \tilde{p}_{f, out}(\bar{x}_v)) \, dS 3.2.39
\]

where \( R \) is the average curvature of the drop and \( \bar{F}_{p, out} \) is the force working on the drop due to pressure differences at the liquid-vapour interface.

Substitution of 3.2.38, 3.2.39 in 3.2.37 gives:

\[
\int_A \rho \, dS = -\bar{m} \frac{2\sigma}{R} A_{sl} + \bar{F}_{p, out} + \rho_v [\tilde{g} - \bar{m}(\bar{m} \cdot \tilde{g})] V + \bar{m} \rho_l (\bar{m} \cdot \tilde{g}) V 3.2.40
\]

The vector product \( \bar{m}(\bar{m} \cdot \tilde{g}) \) has some very useful characteristics. When the plate is placed horizontal, \( \bar{m} \) and \( \tilde{g} \) point in identical (or opposite) direction, and \( \bar{m}(\bar{m} \cdot \tilde{g}) = \tilde{g} \). When the plate is positioned vertically, \( \bar{m} \cdot \tilde{g} = 0 \). This makes 3.2.40 a useful equation to calculate the total force due to pressure differences around the drop.

A comparable result is found by van Helden et al [11], who calculated a correction for the buoyancy force on a bubble in flowing liquid that is detaching from a capillary.

**3.2.5 Surface tension working in the liquid-vapour-solid contact line**

In this paragraph we will calculate the stress tensor working on the system boundary. Three different stresses are identified: vapour shear forces on the liquid-
3.2 forces acting on a drop

vapour interface, surface tension forces on the contact line and liquid shear forces at the plate surface due to liquid flow in the drop.

\[
- \int_{A} (\mathbf{n} \cdot \mathbf{t}) dS = - \int_{A_{c}} \mathbf{n} \cdot \mathbf{t} dS - \int_{A_{s}} (\mathbf{n} \cdot \mathbf{t}) dS + \int_{l_{cl}} (\mathbf{F}_{\sigma}) d\mathbf{l}
\]

3.2.41

where \( l_{cl} \) indicates the contact line between liquid-vapour interface and solid plate. The \( \mathbf{t} \) in surface tension force is used to indicate that this is a force per unit length. We will first calculate surface tension force. On every point along the contact line, the magnitude of this force equals the surface tension coefficient (\( \sigma \)). Surface tension contracts the liquid-vapour interface, thus working perpendicular to the surface normal (\( \mathbf{n} \)), while the direction of this force is in the plane of the surface and normal to the contact line. Thus, this force is given by:

\[
\mathbf{F}_{\sigma} = \sigma \mathbf{i}\mathbf{n}
\]

3.2.42

where \( \mathbf{i} \) is the unit vector in the direction of the contact line.

Schematic presentations of the force along the contact line in downstream direction (x) and normal to the plate (z) are given in fig. 3.2.10

For convenience, the force is presented against radial position along the contact line. At the front of the drop, \( \theta = 0 \). The integral of the y-component around the contact line vanishes as a result of symmetry in the y=0-plane.

Integrating in x direction yields the total surface tension force working on the drop in streamwise direction:

\[
F_{\sigma,x} = \int_{l_{cl}} (\mathbf{k} \cdot \mathbf{F}_{\sigma,x}) d\mathbf{l} = \int_{0}^{2\pi} k \cdot \sigma (\mathbf{i} \mathbf{n}) r(\theta) d\theta = r_{f} \int_{0}^{2\pi} F_{\sigma,x} d\theta
\]

3.2.43

The drop radius (\( r_{f} \)) is the distance from the middle of the wetted area to the contact line. Here we assume that the drop radius is constant along the contact line. The subscript cl refers to the contact line. We will now construct an estimate of
the streamwise component of the surface tension force. At four points along the contact line, both $F_{\sigma,x}$ and $d/d(\theta F_{\sigma,x})$ are known.

The force in streamwise direction working on the front ($\theta=0$) and back ($\theta=\pi$) of the drop equals the horizontal component of the surface tension, $\sigma \cos(\alpha_{\text{front}})$ and $\sigma \cos(\pi - \alpha_{\text{back}})$. At the sides of the drop ($\theta=\frac{1}{2} \pi$ and $\theta=\frac{3}{2} \pi$) no surface tension forces work in streamwise (x) direction. The first derivative of the force in streamwise direction ($dF_{\sigma,x}/d\theta$) is zero at both the front, sides and back of the drop ($\theta=0$, $\frac{1}{2} \pi$, $\pi$ and $\frac{3}{2} \pi$).

As reported by Sadhal et al [38], the direction of the surface tension force on a rough surface is not well-defined. Brown et al investigated the shape of a drop on an inclined plate [11] and, using a finite element method, found that the contact angle at the front and the back of the drop deviate approximately equally much from the static contact angle, given by Young’s equation. At the front of the drop, the contact angle is larger than the static contact angle, whereas at the back, the contact angle is smaller. The static contact angle of PTFE is $\pi/2$. Thus we find:

$$\cos(\alpha_{\text{back}}) = \cos(\pi - \alpha_{\text{front}}) = \cos(\alpha_{\text{front}}) = \cos(\alpha)$$  \hspace{1cm} 3.2.44

This allows us to formulate an approximate equation for $F'_{\sigma,x}$:

$$F'_{\sigma,x} \approx \sigma \cos(\alpha) \left[ \frac{1}{2} + \frac{1}{2} \sin(2\theta) \right]$$  \hspace{1cm} 3.2.44

Substituting 3.2.44 in 3.2.43 gives the force working on the drop by the surface tension:

$$F_{\sigma,x} = \int_0^{2\pi} F'_{\sigma,x} d\theta = \sigma \pi r_1 \cos(\alpha)$$  \hspace{1cm} 3.2.45

Using an identical method, we find for the force normal to the plate:

$$F'_{\sigma,z} \approx \sigma \left[ \sin \alpha + (1 - \sin \alpha) \left( \frac{1}{2} - \frac{1}{2} \cos 2\theta \right) \right]$$  \hspace{1cm} 3.2.46

$$F_{\sigma,z} = \int_0^{2\pi} F'_{\sigma,z} d\theta = \sigma \pi r_1 \left[ 1 + \sin \alpha \right]$$  \hspace{1cm} 3.2.47

### 3.2.6 Shear stresses working on system boundary

The second part of 3.2.44 that has to be calculated is the vapour shear force working on the drop. This cannot be done explicitly, because the vapour flow around the drop is not known. We therefore simply write:

$$\bar{F}_{\tau_{lv}} = - \int (\mathbf{n} \cdot \tau_{lv}) dA$$  \hspace{1cm} 3.2.48
The liquid shear forces working on the foot are calculated assuming laminar liquid flow in the drop near the plate:

$$ F_{\text{ls}} = - \int_A (\hat{n} \cdot \tau_{\text{s,l}}) \, dA = - \eta k \frac{\partial V_x}{\partial z} A_{\text{foot}} \quad 3.2.49 $$

Here, $\partial V_x / \partial z$ is to be interpreted as the average velocity gradient at the drop foot. In chapter 8, calculated values of $\partial V_x / \partial z$ will be compared to measured quotients of drop velocity and height, $v_{\text{drop}}/h_{\text{drop}}$.

### 3.2.7 Summary of the forces working on a drop

Relationships for the pressure and stresses working on the system boundary have now been determined.

A relation for the pressure working on the system boundary is given by eq. 3.2.40, while relations for the stress tensor are given by eq. 3.2.45, 3.2.47 and 3.2.49. Substituting these in eq. 3.2.40 gives the total force working on the drop (eq. 3.2.50):

$$ \rho \frac{dv_m}{dt} = - \frac{1}{\rho_i} \int_A p \, dS - \frac{1}{\rho_i} \int_A \hat{n} \cdot \tau + gV $$

$$ \int_A p \, dS = -m \frac{2\sigma}{R} A_{\text{foot}} - F_{p,\text{out}} + \rho_l [g - m(\ddot{m} - \dot{g})]V + m \rho_l (m \cdot \ddot{g})V $$

$$ - \int_A (\hat{n} \cdot \tau) \, dA = F_{\sigma} + F_{l,v} + F_{s,l} \quad 3.2.50 $$

$$ \rho_i V \frac{\partial \ddot{V}_m}{\partial t} = \left( m \frac{2\sigma}{R} A_{\text{foot}} + \ddot{F}_{\sigma} \right) + \left( \rho_l - \rho_v \right) \left[ \ddot{g} - m(\ddot{m} - \dot{g}) \right]V $$

$$ + \left( \dddot{F}_{p,\text{out}} + \dddot{F}_{l,v} \right) + \dddot{F}_{s,l} \quad 3.2.51 $$

The first term (left hand side) in 3.2.51 is the rate of change of momentum of the drop.

The second term encompasses all surface tension related forces, both those that working directly on the contact line and those that result from pressure differences due to surface tension. For a perfectly hemispherical drop, both terms work normal to the plate, are equally big and are oppositely directed. Thus, for a perfectly hemispherical drop these terms cancel each other. As shown in paragraph 3.2.1, this is the case for relatively small drops on a vertical plate in the absence of flowing vapour. The third term takes gravitational and buoyancy forces into
account. For a drop, hanging under a horizontal plate or placed on it, 
\( \dot{m}(\mathbf{m} \cdot \ddot{\mathbf{g}}) = \ddot{\mathbf{g}} \) and this term cancels out. The fourth term accounts for the forces due

to restriction of the vapour flow by the drop. The sum of \( \mathbf{F}_{p,f,\text{out}} \) and \( \mathbf{F}_{r,\text{in}} \) equals the
combined lift and drag force working on the drop. The drag force is the component
of the forces working on the drop by the vapour flowing around it that works in
downstream direction. The lift force is the component normal to the plate:

\[
\mathbf{F}_D = k [ \dot{\mathbf{F}}_{p,f,\text{out}} + \mathbf{F}_{r,\text{in}} ] \quad 3.2.52
\]

\[
\mathbf{F}_L = \dot{m} [ \dot{\mathbf{F}}_{p,f,\text{out}} + \mathbf{F}_{r,\text{in}} ] \quad 3.2.53
\]

Drag forces of many objects have been determined experimentally. In general
(Perry, Janssen), the drag force is calculated using:

\[
\mathbf{F}_D = k C_D \frac{1}{2} \rho v |\Delta v|^2 A_D \quad 3.2.54
\]

where \( A_D \) is the cross sectional area of drop in streamwise direction. This follows
from 3.2.8:

\[
A_D = \frac{\pi}{2} r_h h = \frac{\pi}{2} \frac{1}{2} \sqrt{\frac{r}{r}} r_{eq} \quad 3.2.55
\]

The fifth and last term in 3.2.54 accounts for the frictional forces due to liquid flow
inside the drop.

Equation 3.2.51 will be used in sections 3.4 and 3.6 to calculate the hydrodynamic
forces working on drops in the single plate mock ups. The acceleration of a drop is
determined from the drop position. Surface tension, gravitational and buoyancy
force are calculated from the drop shape. This allows calculation of hydrodynamic
forces working on the drops, for example during break-up.

3.2.8 The maximum size of a drop, hanging under a horizontal flat plate
In the previous paragraphs, an equation to calculate drop movement has been
constructed. To construct this equation, a variable but well-defined drop shape has
been used. This drop shape has been determined using results by Stauffer and
Chatterlee (§3.2.1). The question whether or not this drop shape can exist has not
been answered in the previous paragraphs. In addition, the analysis in §3.2.2-3.2.7
cannot be used to calculate changes in drop shape, because only forces working
on the entire drop are calculated.

In this paragraph, the maximum size of a drop under a horizontal flat plate will be
calculated. The following method is used.

First, it is assumed that a stable drop satisfies equation 3.2.15:

\[
\left(1 - \frac{y}{h}\right)^2 + \left(\frac{x}{r_f}\right)^2 = 1 \quad 3.2.15
\]
where \( h \) is the drop height and \( r_f \) is the radius of the foot of the drop. Such a drop is drawn in fig. 3.2.11.

The maximum size of a pendant drop in quiescent vapour is determined from the pressure inside the drop. The pressure inside a drop near the foot is calculated through two paths which are presented in figure 3.2.11.

The first path crosses the liquid-vapour interface at the drop apex. The second path crosses the liquid-vapour interface at the drop foot.

When the pressure at the drop foot, calculated via path 1, is larger than the pressure calculated via path 2, liquid flows from the centre of the drop to the side, stabilising the drop, until the pressure at the drop foot is constant. When, on the other hand, the pressure via path 2 is higher than the pressure via path 2, liquid flows from the edge of the drop to the centre, increasing the height of the drop. This increases the pressure difference between \( p_3 \) and \( p_6 \) further, resulting in larger flow of liquid to the drop centre, until the drop is very sling and part of the drop pinches off.

**Calculation of pressure at the foot of the drop**

First, the pressure difference over the interface is calculated, assuming that the interface contour is given by an ellipse. Combination with the static pressure allows calculation of the pressure via paths 1 and 2, see eq. 3.2.63 and 3.2.64. Using that the pressure via path 1 and 2 are identical gives an expression of drop size, quantified with the Bond number, as a function of drop shape, quantified with the shape factor \( B: Bo=Bo(B) \). As shown in paragraph 3.2.1, the shape factor depends on Bond number: \( B=B(Bo) \). This gives two equations with two unknowns, which will be solved iteratively.
As argued in §3.2.3, the pressure on the inside of a liquid-vapour interface is higher by a factor of \(2\alpha/r\), where \(r\) is the radius of curvature of the interface. When the radii of curvature are different, the pressure difference is given by:

\[
\Delta p = \alpha \left( \frac{1}{r_1} + \frac{1}{r_2} \right)
\]

3.2.56

The shape of the drop contour, given by equation 3.2.15, is that of an ellipse. A picture of an ellipse with principle axis \(x_A\) and \(x_B\) is given in fig. 3.2.12.

The radius of curvature of the contour in the point \(x_A\) is given by:

\[
r_A = \frac{x_B^2}{x_A}
\]

3.2.57

The two radii of curvature at the drop apex are identical and equal the radius of the contour in the apex, which is given by:

\[
r_{1,tip} = r_{2,tip} = \frac{r_f^2}{h}
\]

3.2.58

At the foot of the drop, the radii of curvature are different. The first radius of curvature equals the radius of the drop foot, while the second radius of curvature equals the radius of curvature of the contour:

\[
r_{1,f} = r_f
\]

3.2.59

\[
r_{2,f} = \frac{h}{r_f^2}
\]

3.2.60
Over paths $2 \to 3$ and $4 \to 5$, pressure changes due to the density of the fluid. The pressure difference is given by:

\[
\Delta p_{23} = \rho_1 (\Delta x_{23} \cdot \hat{g}) = -\rho_1 gh \tag{3.2.61}
\]

\[
\Delta p_{45} = \rho_v (\Delta x_{45} \cdot \hat{g}) = -\rho_v gh \tag{3.2.62}
\]

Using 3.2.59, 3.2.61 and 3.2.64, the pressure at (the centre of) the foot of the drop equals:

\[
p_3 = p_1 + \frac{2gh}{r_f} - \rho_1 gh \tag{3.2.63}
\]

And, using 3.2.59, 3.2.62, 3.2.63 and 3.2.65, the pressure at (the side of) the foot of the drop equals:

\[
p_6 = p_1 + \sigma \left( \frac{1}{r_f} + \frac{r_f^2}{h} \right) - \rho_v gh \tag{3.2.64}
\]

The height and foot radius of a drop have been calculated in paragraph 3.2.1:

\[
r_f = r_{eq} B^{1/6} \tag{3.2.65}
\]

\[
h = r_{eq} B^{1/3} \tag{3.2.66}
\]

Where $B$ is the shape factor, that is a function of Bond number only. Substitution of these relationships in 3.2.63 and 3.2.64 gives:

\[
p_3 = p_1 + \frac{2}{B^{2/3}} \frac{\sigma}{r_{eq}} - \frac{1}{B^{1/3}} \rho_1 gr_{eq} \tag{3.2.67}
\]

\[
p_6 = p_1 + \left( \frac{1}{B^{1/6}} + B^{2/3} \right) \frac{\sigma}{r_{eq}} - \frac{1}{B^{1/3}} \rho_v gr_{eq} \tag{3.2.68}
\]

For a stationary drop, $p_3 = p_6$:

\[
\left( \frac{2}{B^{2/3}} - \frac{1}{B^{1/6}} - B^{2/3} \right) \frac{\sigma}{r_{eq}} = \frac{1}{B^{1/3}} (\rho_1 - \rho_v) gr_{eq} \tag{3.2.69}
\]

\[
Bo = \frac{-\Delta p r_{eq}^2}{\sigma} = \left( \frac{2}{B^{1/3}} - B^{1/6} - B \right) \tag{3.2.70}
\]

Where the minus sign results from the definition of the Bond number. The Bond number and the shape $B$ are also coupled via the shape of the drop:

\[
B = 1/(1 - 2Bo) \tag{3.2.5}
\]
The size of a drop at the moment of pinch off is calculated by solving 3.2.5 and 3.2.69 simultaneously. This gives two solutions. The first solution is the trivial solution of a hemispherical drop of zero volume: \( Bo=0, B=1 \). The second solution is:

\[
Bo_{\text{max}} = -2.752 \\
B_{\text{max}} = 0.1537
\]

\( p_3 \) is larger than \( p_6 \) when the Bond number is larger (less negative) than \(-2.752\), resulting in a stable drop. The drop is not stable when the Bond number is less than \(-2.752\). The height to diameter ratio of such a drop is:

\[
\left( \frac{h}{r_f} \right)_{\text{max}} = \frac{1}{\sqrt{B}} = 2.55
\]

3.3 Experimental set-up: the 3 mirror test rig

3.3.1 Design specifications
The 3 mirror test rig has been built to investigate condensate re-entrainment from hydrophobic heat exchanger plates. Experiments in the pilot plant test rig have shown that re-entrainment occurs at high vapour velocities, but have not revealed by which mechanism drops are formed, see e.g. §2.3.1.

Furthermore, variation of plate geometry in the pilot plant heat exchanger is impractical.

The design specification of the 3 mirror test rig are:
- geometry equal or scalable to downstream edge of pilot plant heat exchanger plate
- liquid-solid-vapour contact angle of the plate 90°
- 3 dimensional visualisation of droplet pinch off possible
- investigation of effect of geometry of downstream edge possible.

Fluid properties of liquid and vapour (mass density, viscosity, surface tension) should be about the same as fluid properties in the pilot plant heat exchanger. Experiments have been performed with demineralised water containing an ionic fluorescent (Fluorescine) and air at ambient conditions.

The plate is constructed from PTFE, which is chemically similar to PVDF, and has a water-air contact angle of 90°.

3.3.2 Description of the 3 mirror test rig
The 3 mirror test rig consists of a recirculation loop filled with air at ambient conditions. The main components of the test rig are a recirculation loop, a test section, a pump and a mirror array.
3.3 Experimental set-up

The recirculation loop is made of \( \odot 22 \) mm PVC tube positioned in a horizontal plane, fig. 3.3.1.

Fig. 3.3.1 Schematic of 3 mirror test rig (top view) sizes in mm

A 250W radial flow pump is used to sustain the air flow in the recirculation loop. Maximum vapour velocity over the test plate is 30 m/s. The vapour velocity can be changed by adjusting a butterfly valve downstream of the pump exit. A gate valve has been built to allow fast (within 10 ms) opening of the flow channel. In the gate valve, the flow channel can be blocked by a plate. The plate is removed using a spring, thus opening the valve.

A pitot tube is used to measure the air velocity in the recirculation loop. Temperature, pressure and humidity of the air are measured outside the test rig. Temperature is measured using a calibrated class A Pt100, calibrated between 0 and 40°C, accuracy ±0.01°C. Pressure is calibrated using a mercury manometer, accuracy ±13 Pa (0.1 mmHg). The relative humidity is measured using a Vaisala HMP 135Y sensor, accuracy 1% of full scale.

Several measures have been taken to avoid influence of pump vibrations on droplet break-up in the test rig. First of all, pump and test section have been connected to different support plates. Both support plates have rubber shock absorbers and are placed on separate tables. Part of the piping between pump and test section is made of a \( \odot 24 \) mm flexible tube.

The test section consists of a rectangular Perspex housing in which 3 inserts are placed. The housing acts as a structural support, while the inserts determine the flow field. The experiment is performed in the second (middle) insert, while the
other inserts facilitate a smooth transition between the 10x40mm test channel and the 22mm pipe. The first insert is 200 mm long, has a 20x20mm entrance and an 10x40mm exit. The third insert is 100 mm long, has a 10x40mm entrance and an 20x20mm exit.

A side view of the inner geometry of the middle insert is presented in fig. 3.3.2.

![Fig. 3.3.2 Detail of 3 mirror test rig: side view of flow geometry in middle insert. Width: 40 mm](image)

The flow channel is 10 mm high. A PTFE (poly tetra fluor ethylene, \(\text{C}_2\text{F}_4\),\text{n}) plate is placed in the centre of the channel, allowing air to flow over and under it. A small drop can be formed on the surface of the plate by injecting fluid through a capillary tube. The tube is positioned 30 mm upstream of the downstream edge of the plate, see fig. 3.3.2.

Drops are formed on the upper surface of the test plate by injecting liquid through a 1.2 mm hole at 30 mm distance from the downstream edge. A liquid feed system (fig. 3.3.3) has been used to allow continuous production of drops, about \(\frac{1}{4}\text{cm}^3\) per minute.

![Fig. 3.3.3 Schematic of liquid feeding system. Multiple needle valves allow control over liquid flow](image)
3.3 Experimental set-up

**Camera and optics**

Break-up of the drop from the downstream edge is recorded using a high speed camera and a mirror array. The mirror array is placed over the middle insert. Its aim is to allow 2-sided visualisation of droplet pinch-off. (fig. 3.3.4).

The mirrors are placed in such a way that the upper part of each video frame shows the top view of the plate downstream edge, while the lower part of the same image shows the side view. The optical paths between droplet and lens are equally long, allowing simultaneous focusing of both parts of the image. Standard glass mirrors have been used. All mirrors are placed parallel to each other, at an angle of 45±1º with respect to test plate.

A Kodak Ektapro high speed motion analyser (model 4540) has been used to visualise the droplet break-up. The lens is a Navitor zoom 6.5 (cat. nr. 6000). The camera produces 3000 pictures of 256x256 frames of 256 grey values. The camera was placed on a tripod at close distance from the test rig. Measurement frequencies of 500-2250 Hz have been used. Two adjustable 200W light sources, with infrared filters to minimise heating, have been used for lighting. The droplet is illuminated from above, with the lamps positioned to the left and the right of the mirror array. The lamps are used during short intervals only (~10 s) to avoid heating of the measurement section.

Recordings of single droplets pinching off have been stored as a series of pictures (*.tif-files). Systematic naming has been used, referring to year-month-day, and the number of the experiment that day. Thus, the 14th experiment on September 12th, 2001 is called 01091214. (01 for 2001, 09 for September, etc). The tif-files are stored on cd in a directory with the systematic name of the experiment. When necessary, grey values have been adapted using dedicated Matlab® procedures, which have been written for this purpose.
3.3.3 Operation and Experimental Procedures

Cleaning
Since small amounts of impurities can influence surface tensions (and thus the experimental results), a thorough cleaning procedure of the test rig has been followed.

Test section and piping are cleaned by thorough rinsing with demineralised water. The pump is cleaned using oil-free compressed air.

The plate is cleaned by submerging it for at least 10 min. in a 1% HNO₃-solution, followed by thorough rinsing with demineralised water. Contact of all inner parts of the test rig with hands has been avoided after cleaning.

Performing experiments
Experiments have been performed in two different ways. The first type of experiment is meant to record the break-up of drops that slide off the plate. The second method is used to record the break-up of drops that slowly grow on the downstream edge of the plate.

In the first method, a drop of predefined volume is placed 30 mm upstream of the plate downstream edge. The gate valve is closed and the pump is switched on. Within 2 sec., Illumination is turned on, camera is turned on and the gate valve is opened, recording the break-up of a drop sliding off the downstream edge of the hydrophobic plate. Because the experiment takes only about 2 seconds, vapour velocity is measured afterwards.

In the second method, the gate valve is always open. Vapour velocity is set by switching on the pump and choosing a butterfly valve setting. Vapour velocity is measured using the pitot tube and adjusted when necessary.

The liquid feed velocity to the test rig is set by pressurising the storage vessel and adjusting the needle valve settings. The average liquid flow is about ¼cm³ per minute.

Illumination is turned on and camera position, lens magnification and focal distance are adjusted to yield a high quality recording. Because liquid is continuously fed to the system, droplets are formed on the surface of the test plate at regular intervals. These drops slide over the plate surface and collect at the downstream edge of the plate, forming a large drop. When the drop reaches a critical size, part of the drop pinches off, which is recorded. Several test experiments are performed to chose recording frequency. Droplet break-up should be covered by at least 50 frames to allow calculation of drop shape and acceleration of the drop during pinch-off.
3.4 Results 3 mirror test rig

The 3 mirror test rig has been built to investigate droplet break-up by air shear forces and gravity from a hydrophobic plate. In the pilot plant test rig, this proved to be impractical for two reasons. First of all, the visual accessibility of the downstream edge is not satisfactory. Secondly, geometric adaptation requires rebuilding of the entire heat exchanger. This would be a very inefficient solution.

The 3 mirror test rig has been described in the previous section. Conditions in the test rig have been chosen in such a way that they are comparable to conditions in the pilot plant heat exchanger.

Experiments have been performed in two different ways. The experiments that are described in the next paragraph are performed with a drop of predefined volume, resting on the upper surface of a horizontal hydrophobic plate. Air velocity is increased rapidly by opening a gate valve. The drop moves over the plate and breaks up at the plate downstream edge. These experiments are performed to analyse the pinching off of drops that slide off the plate, which has been observed in the pilot plant heat exchanger at relatively high gas velocities and condensate fluxes.

Next, experiments will be presented where liquid is injected onto the upper surface of a hydrophobic plate, see §3.4.2. During these experiments, air velocity is constant. The liquid forms small drops on the upper surface of the plate, which move towards the downstream edge. There, consecutive drops coalesce, yielding a single large drop. Break-up of this drop is recorded. These experiments mimic the break-up of slowly growing drops on the downstream edge of condenser plates and spacers in the small scale heat exchanger.

3.4.1 Observation of break-up of a sliding liquid drop of predefined volume at the downstream edge of a hydrophobic plate

A water drop, which contains 40 mg/l of fluorescein to enhance visibility, is placed on a horizontal 3mm thick plate, 30 mm upstream of the downstream edge. After opening a valve, air flows through the slit with an average velocity of 25±3 m/s. The drop on the plate is deformed by the flowing air and moves towards the plate downstream edge. Break-up of drops of different size at the downstream edge of three plates has been recorded.

In this way, break-up of drops of different size sliding off a

![Fig. 3.4.1 different shapes of downstream edge of test plates in the three mirror test rig](image)
hydrophobic horizontal plate has been investigated. PTFE plates (poly tetra fluoro ethylene, -[CF2-CF2]-) with three different downstream edge shapes (fig. 3.4.1) have been used. Initial volume-averaged drop radii varied from 0.5 to 3 mm (0.5-80 µl).

**Sliding of drops**
The velocity with which drops slide over the surface has been determined just before the plate downstream edge (30 mm downstream of the drop release point). The sliding velocity of drops over a dry plate are presented in fig. 3.4.2 versus volume-averaged drop radius, \( r_{eq} \). The velocity increases with size, from 0.16 m/s for 2 µl drops (\( r_{eq}=1.0 \) mm) to 1.2 m/s for 80 µl drops (\( r_{eq}=3.4 \) mm).

![Graph showing velocity of drops](image)

**Fig. 3.4.2** Velocity with which a drop slides over the upper surface of a horizontal PTFE plate. \( V_{gas}=25 \pm 3 \) m/s. \( T=21.7^\circ \text{C}, ~p=105 \text{ kPa} \) \( d_{all}=3.5 \) mm. Error bars give experimental error. \( Re=\rho \omega_d \sqrt{\frac{d_{drop} \eta_v}{v}} \)

\[ r_{eq} = \sqrt{\frac{3V}{2\pi}} \]  

3.2.6

The velocity with which a drop slides over the plate increases nearly linearly with drop radius:

\[ v = 0.44 \cdot 10^3 (r_{eq} - 0.7 \cdot 10^{-3}) \]  

3.4.1
A schematic of the shape of sliding drops is presented in fig. 3.4.3.

Fig. 3.4.3 Schematic of the observed shape of sliding drops on a horizontal PTFE plate. Vapour velocity: 25 m/s. Shape has been recorded 25 mm downstream of release point. The shape of drops of 50µl or more is changing at the point of observation.

Sliding drops with a volume less than 5 µl, are hemispherical in shape. A tail, which length is up to twice the drop diameter, is present behind the drop.

Drops with a volume exceeding 10 µl spread out, forming a sheet, thickest and broadest at the front. At the end, the sheet ends in a tail, which is about as long as the sheet. The front of the drop is irregularly shaped with 2 to 5 lobes protruding forward. The fronts of the larger drops (20-80µl) all but touch the upper surface of the slit, which is 3.5 mm high.

Break-up at the downstream edge of the plate with a flat edge
Drops with a volume of 0.5 µl or less do not detach from a plate with a flat edge (see fig, 3.4.1, top figure). A drop of 2µl detaches without break-up, while a 5 µl drop breaks into two drops. Drops with a volume between 10 and 30 µl sliding over the PTFE plate have a broad and flat shape, see fig. 3.4.4.
Pinch-off from a horizontal plate

The drop slides off the plate, while the back of the drop is still connected to the tail on the plate. The flat and thin front contracts, resulting in a compact drop, connected to the plate by a liquid bridge. The drop moves down, after which it is stretched by shear forces of the gas, flowing under the plate. The top of the drop is flattened and pushed upward by the gas stream. Length-wise stretching of the drop results in break-up. This break-up process results in a limited amount (under 10) relatively large drops. The volume-averaged diameter of the largest drops is about 2 mm.

When drops of 40-80 µl reach the downstream edge of the plate, the drop front is between 20 and 30 mm wide. The drop detaches without changing much in height or width. After detachment of the front of the drop, surface tension forces contract the drop sideways, resulting in a thicker drop that is less wide. At the same time, variations in the thickness of the liquid sheet grow, resulting in formation of holes. The holes grow and combine, resulting in rupturing of the sheet. In this process, about 20 drops, all under 1 µl in volume, are formed. The remaining fluid contracts forming an elongated drop, approximately equally wide and high. The drop is stretched by vapour shear forces and breaks up, forming about 10 drops of 1-3 µl each.

**Break-up at the downstream edge of the plate with a round edge**

Drops with a volume of 5 µl or less do not detach from a plate with a round edge. Drops between 20 and 30 µl break-up, very much like the drops of the same size

---

*Fig. 3.4.4. Break-up of a 20µl drop at a flat downstream edge of a horizontal 3 mm thick plate liquid: water+40g/l fluorescine, vapour: air gas velocity: 25±3 m/s, T=21.7°C, p=105 kPa*
from a plate with a flat edge. The largest drops that are formed after pinch-off are smaller, about 1.5 mm in diameter.

40-80µl drops detach from a round edged plate without changing much in height or width, forming a thin sheet. Holes are formed in the sheet which grow and combine, resulting in rupturing of the sheet. In this process, about 20 drops, all under 1µl in volume, are formed. The remaining fluid contracts forming a elongated drop, approximately equally wide and high. The drop is oscillates perpendicular to the plate forming a large amount of drops, under 1µl each. 20±5% of the liquid remains behind on the downstream edge.

**Break-up at the downstream edge of the plate with a sharp edge**

Drops with a volume of 5µl or less do not detach from a plate with a round edge. Drops between 20 and 80 µl slides off the plate edge, following the plate surface, see fig. 3.4.5

As a result, the drop moves both downstream and downward. The drop hits the air stream under the plate and is stretched, while violent up-and downward movement of the drop causes it to break into many drops, usually under 1 µl. After break-up, a drop of about a third of the liquid remains behind on the downstream edge.

**Size of drops after pinch off**

In chapter 2, it has been shown that the formation of drops has a negative effect on the performance and durability of the heat exchanger and downstream equipment. Large drops evaporate relatively slowly and settle within a short distance of the
heat exchanger, limiting the negative effects. Consequently, the formation of large drops is preferred over the formation of smaller ones.

The experiments have shown that both the drop size before pinch-off and the shape of the downstream edge have an influence on the size distribution of drops that are formed by pinch off. The experiments have been performed twice for every drop size and plate type. As a result, size distribution after pinch-off has been determined with a very low accuracy. Still, some trends have been observed, which are presented in fig. 3.4.6

![Graphs showing drop size distribution](image)

*Fig. 3.4.6. Estimated drop size distributions after pinch off of drops of 2, 5, 30 and 50 µl from a PTFE plate with flat, round or sharp downstream edge. Accuracy of the vol. averaged maxima of the distribution is ±0.2mm.*

Break-up of drops from a plate with a flat edge produces mainly drops of about 1.5 mm, which is half the plate diameter. This is nearly independent of the initial drop size. The larger the initial drop, the higher the fraction of small satellites that are formed.

Break-up of drops from a plate with a round edge produces drops of a large variety of sizes. The average drop size after pinch off decreases as the initial drop size increases.

The average drop size after break-up of a drop from a sharp edged plate appears to be independent of initial drop size. Break-up of larger drops gives a slightly larger spread in drop size.

A plate with a flat edge produces a smaller fraction of small drops than a plate with a round or a sharp edge. Changing the shape of the downstream edge from flat to round or sharp in the pilot plant heat exchanger will thus result in an increased re-entrainment, which is undesired.
3.4 Results three mirror test rig

3.4.2 Break-up of a slow growing liquid drop from the downstream edge of a horizontal hydrophobic plate

Pinch-off of a drop from the downstream edge of a hydrophobic horizontal plate has been studied by continuously feeding liquid through a hole in the plate upper surface. The experiments have been performed with three plates with different edge shapes, four times with each plate. Air at ambient conditions flows over the plate at a velocity of 25 m/s. Small drops, about 0.5 mm (0.03±0.01µl) in size, slide over the plate and collect on the downstream edge, where a large drop is eventually formed.

Break-up at the downstream edge of a flat edged plate

A drop behind the downstream edge of a plate with a flat edge grows until its volume is 25±3µl. The height and length of the drop are then equal to the plate diameter to within 5% and the width is about 1.6±0.05 times the plate thickness. The drop moves periodically in the vertical plane with a frequency of 16±2Hz. The amplitude of the oscillatory motion of the tip of the drop equals the plate thickness to (±10%). Pinch-off starts after a final coalescence of a 0.03µl droplet with the drop. Apparently, the volume increase results in a drop volume that is too large to be stable. In 50-150 ms, the drop increases in length from 3 to 12±1 mm, while both thickness and width of the drop decrease to about 2 mm (fig. 3.4.7).

![Fig. 3.4.7. Pinch-off of a drop from the flat downstream edge of a horizontal 3 mm thick plate liquid: water+40g/l fluorescine, vapour: air vapour velocity: 25±3 m/s, T=21.7°C, p=105 kPa Top: Top view Time between consecutive frames: 7.11 ms](image)
At the same time, both the frequency and the amplitude of the vertical oscillation increase. Two droplets of $0.7\pm0.3\mu l$ ($d_{\text{drop}} = 1.1\pm0.15\text{mm}$) pinch-off from the tip of the main drop. The remainder of the fluid ($23\pm3\mu l$) contracts, forming a drop that is slightly smaller and almost identical in shape to the drop before pinch-off. The continued feeding of liquid causes the drop to grow, until liquid pinches off again. Some aspects of the pinch-off process proved to be highly reproducible, whereas others varied between consecutive experiments. The oscillation frequency before pinch-off and the drop volume at pinch-off are reproducible. The spread is determined by experimental error. The pinch-off time, i.e. the time between coalescence and pinch-off, varied by a factor of 3. In all four experiments, two drops pinched off, both about 1.1mm in diameter.

**Break-up at the downstream edge of a round edged plate**

Similar experiments have been performed using both a round and a flat edged plate, see fig. 3.4.8.

---

**Fig. 3.4.8.** Pinch-off of a drop from the round downstream edge of a horizontal 3 mm thick plate; liquid: water+40g/l fluorescine; vapour: air; $T=21.7^\circ\text{C}$, $p=105\text{kPa}$
When a round-edged plate is used, a drop grows at the downstream edge until its volume is $6.7 \pm 0.5 \mu l$. Before pinch-off, the drop moves irregularly. Although the drop is not stationary, no periodic movement has been observed. Length scale of the drop movement is small: the tip of the drop remains within an area of $1 \times 1 \times 1 \text{ mm}$. ($1/4d_{plate}$). After coalescence, the drop moves down until it protrudes under the plate. No sharp edges restrict contact line movement. As a result, the lowest point at the contact line moves upstream of the downstream edge as the drop moves down, and the highest point at the contact line moves to the centre of the downstream edge. Vapour shear forces push the drop up and increase the drop length. In its highest position, the drop protrudes $1 \frac{1}{2} \text{ mm}$ out of the recirculation zone behind the plate. The length of the drop decreases and the drop moves down. The downward velocity is much higher now, and the drop all but touches the bottom of the test rig. Vapour shear forces stretch the drop and accelerate it upward, resulting in break-up of the drop in a large amount of drops. Part of the break-up occurred more than $9 \text{ mm}$ downstream of the plate edge, outside of the field of vision of the camera. The drops that are formed within the field of vision are all under $1 \text{ mm}$ in diameter ($0.5 \mu l$), while most of the drops are about $0.5 \text{ mm}$ in diameter ($0.1 \mu l$).

**Break-up at the downstream edge of a sharp edged plate**

When a sharp edged plate is used, the drop grows at the downstream edge until its volume is $40 \pm 5 \mu l$.

The drop is $1.5 \text{ mm}$ long (measured from the tip of the downstream edge), $3 \text{ mm}$

![Image](image-url)

*Fig. 3.4.9. Pinch-off of a drop from the sharp downstream edge of a horizontal 3 mm thick plate. Liquid: water+40g/l fluorescine, vapour: air

$T=21.7\degree C$, $p=105 \text{ kPa})*
high and 10 to 12 mm (3-4 $d_{plate}$) wide. Before pinch-off, the drop is almost stationary: the tip of the drop remains within an area of 0.6x0.6x0.6 mm, ($1/4d_{plate}$). About half of the liquid of the drop consists of broad and thin sheets above and under the sharp parts of the edge. After coalescence with a last drop small arriving at the downstream edge, the liquid moves down until it protrudes 0.5-1 mm under the plate. As with the flat edged plate, we see that contact line movement is restricted by the sharp corners at the downstream edge of the plate. With the sharp edge, the upper part of the contact line follows the drop movement over the upper part of the downstream edge, whereas the lower contact line follows the drop movement along the lower part of the downstream edge.

When the liquid protrudes from under the plate, vapour shear forces push the drop up and increase the drop length. In its highest position, the drop protrudes 1-1/2 mm out of the recirculation zone behind the plate. The length of the drop decreases and the drop moves down. At the same time, the sheets of liquid are drawn off the plate, forming a large amount of fluid protruding about 3 mm under the plate. This fluid is stretched to a sheet, increasing in length and decreasing in width. 3 processes contribute to the break-up of the sheet: lengthwise stretching, rupturing and violent vertical movement. A large part of the sheet breaks up outside the field of vision of the camera.

The drops that are formed within the field of vision are all under 1 mm in diameter (0.5 µl), while most of the drops are about 0.5 mm in diameter (0.1 µl).

**Size of drops after pinch-off**
The above-described experiments show that the size of the largest drop is about 1 mm, independent of the edge shape. This is a third of the thickness of the plate. When a drop breaks up from a flat edged plate, no satellites are formed. After pinch-off from a round or sharp edged plate, many satellites are formed. The volume fraction of satellites could not be determined because drop break occurs partially out of the field of vision. Figure 3.4.8 and 3.4.9 show that the fraction of fluid that is entrained as small drops is certainly not small compared to the total volume of entrained fluid.

As shown in chapter four, entrainment of small drops is an undesired phenomenon. It is therefore advisable to construct heat exchanger plates with flat downstream edges for condensation at relatively high vapour velocities.

**3.4.3 Pinch off of sessile drops from the downstream edge of horizontal PTFE plates**
Observation of break up of drops that grow on the downstream edge of a horizontal plate (§3.4.2) has shown that the edge shape strongly influences drop break-up. The size up to which a drop grows differed from 6.7µl for a round edged plate to 40µl for a sharp edged plate. Also size and number of re-entrained drops varied. Although paragraph 3.4.2 showed the influence of edge shape on drop break-up, it was not explained why this is so. In this paragraph, drop movement will be
analysed, calculating the forces working on a drop, as well as the increase in kinetic and surface free energy, during break-up.

To analyse drop deformation during pinch off, the location of the drop is determined in time. First, a general description of the drop is given, giving the drop contour as a function of initial drop shape and tip position during pinch off. This is used to calculate the centre of mass off the drop during pinch off. Application of the force balance, derived in chapter 6, allows calculation of the hydrodynamic forces on the drop. Next, the components of the drop energy (surface tension energy, kinetic energy and potential energy) are estimated to determine the power input into the drop during pinch off.

Description of the drop
It is assumed that the drop shape, during deformation, can be regarded as a deformed hemisphere of constant volume. Length of the drop is denoted l, its height h and the width b. (fig. 3.4.10)

![Fig. 3.4.10. Schematic of drop behind heat exchanger plate and variables to determine its size and shape](image)

The origin is positioned in the middle of the downstream edge of the plate. The position of the tip of the drop is \((x_{\text{tip}}, y_{\text{tip}}, z_{\text{tip}})\). Before pinch off, length, l, height, h, and width, b, of the drop are determined.

It is assumed that drop height and width have a constant ratio. This allows calculation of the drop shape at any time. The position of the drop foot is defined from the location of the highest and lowest point on the contact line:

\[
\begin{align*}
\hat{x}_{\text{foot}} &= \frac{1}{2}(x_{cl,1} + x_{cl,2}) \\
l &= |\hat{x}_{\text{tip}} - \hat{x}_{\text{foot}}| \\
b &= b_0 \sqrt{\frac{l_0}{l}} \\
h &= h_0 \sqrt{\frac{l_0}{l}}
\end{align*}
\]
where $l$, $b$ and $h$ refer to the drop length, width and height, respectively. Integration yields the centre of mass of the drop:

$$
\bar{x}_m = \frac{1}{V} \int \bar{x} \, dV
$$

3.4.6

It is shown in appendix A that the centre of mass is given by:

$$
\bar{x}_m = \frac{5}{6} \bar{x}_{\text{foot}} + \frac{3}{8} \bar{x}_{\text{tip}}
$$

3.4.7

**Forces working on the drop**

The forces working on a drop have been derived in chapter 3:

$$
\rho V \frac{d\bar{V}_m}{dt} = \left( \frac{2\sigma}{R} A_{\text{foot}} + \bar{F}_\sigma \right) + \left( \rho_l - \rho_v \right) g - \bar{m}(\bar{\dot{m}} \cdot \bar{g}) + \bar{F}_l
$$

3.2.51

$$
+ \left( \bar{F}_{p,f,\text{wall}} + \bar{F}_{\tau,\text{NL}} \right) + \bar{F}_{\tau,\text{sl}}
$$

where $\bar{m}$ is the unit vector, normal to the plate downstream edge. For a flat edged plate, $\bar{m} \cdot \bar{g} = 0$. For a round or sharp edged plate, $\bar{m}$ is taken normal to the line between the highest and lowest point on the foot of the drop. The fifth term of 6.51 consists of liquid shear forces near the wall and forces due to flow induces pressure differences in the drop. Velocity gradients near the wall are expected to be very low. As a consequence, $F_{\tau,\text{ls}} \approx 0$ and 6.51 simplifies to:

$$
\rho V \frac{d\bar{V}_m}{dt} = \left( \frac{2\sigma}{R} A_{\text{foot}} + \bar{F}_\sigma \right) - \rho_l \bar{g} V + \bar{F}_H
$$

3.4.8

where $\bar{F}_H$ are the hydrodynamic forces, working on the drop. Acceleration of the drop is calculated from the position of the centre of mass of the drop. To determine the force due to surface tension, the circumference of the drop foot, the average curvature of the drop and the area of the drop foot are calculated. The method by which the forces due to surface tension and gravity are calculated is presented in appendix B. The hydrodynamic force is calculated from 3.4.8, since all other forces and the total force are determined from the experiment.

In the following sections, drop pinch-off from a flat, a sharp and a round edged plate are investigated. The position of the tip of the drop is measured during pinch-off and used to calculate hydrodynamic, surface tension, gravitational and total force. Forces during pinch-off of drops from plates with different edge shapes are compared. The hydrodynamic force is used to find an estimate of the drag force coefficient of drops that protrude from behind a plate during pinch-off.
3.4.4 Droplet break-off from a flat edged plate

Break-up of two representative drops from a horizontal flat edged PTFE plate have been analysed at a vapour velocity of 25m/s. Tip positions are followed during the stretching of the drops and are used to calculate forces on the drop and the sum of kinetic, potential and surface free energy, $E_{tot}$. Tip position and estimated hydrodynamic force during 2 experiments are presented in fig. 3.4.11.

On a flat edged plate, a drops grows to a size of $23 \pm 2 \mu l$. The drop oscillates with a frequency of $16 \pm 2Hz$ in the vertical plane. The amplitude of the top of the drop (3.1 $\pm$ 0.2 mm) equals the plate thickness. Before break-up, amplitude and frequency of the oscillation increase. After this, the drop stretches lengthwise and 2 drops pinch off. The last point in fig. 3.4.14 is the tip of the drop at the moment of break up.

The arrows in fig. 3.4.11 give an estimate of the hydrodynamic forces on the drops. Before break-up is initiated, the tip of the drop remains behind the plate ($-1\frac{1}{2} < z < 1\frac{1}{2}$). Since the drop does not protrude above or under the plate, drag forces working on the drop are small. This can be explained from the presence of a recirculation zone behind the plate (fig. 3.4.12).
Recirculation zones have been reported for a wide range of experimental conditions behind objects that are comparable to the downstream edge of a plate, like backward-facing steps. In the critical pressure test rig, described in section 9, a recirculation zone has been observed behind the plate of 5-8 plate diameters long, at Reynolds numbers between 50 and 6000. The flow geometry of the measurement section of this test rig and the critical pressure test rig are identical. Thus, we can also expect that a recirculation zone is present behind the downstream edge of the plate in this test rig.

Because the vapour velocities and velocity gradients in the recirculation zone are much smaller than in the turbulent flow (Re = 5500) further away from the plate, drag forces on the drop are expected to be small when the entire drop remains in the recirculation zone. This is exactly what is found in the experiments reported in fig. 3.4.11. In the first recording (left picture), the tip of the drop is out of the recirculation zone when the drop breaks up. As a result, the drag force is high. In the second recording (right picture) the tip of the drop is in the recirculation zone during the final moments of break up. Consequently, drag forces on the drop are much smaller.

When the forces and trajectories in the left and right graph of 3.4.11 are compared, it must be remembered that the independent variables (vapour velocity, test rig geometry, viscosities, densities etc) are practically the same. Still, different trajectories of the drop tip and different forces are found. This shows that a stochastic component governs drop pinch-off. Amplitude (±5%) and frequency (±10%) of oscillation before pinch off, as well as the drop volume (±10%) when pinch off occurs are reproducible within the error margins.
The difference between the two recordings is explained from two effects. The first is a timing difference the moment of coalescence of the small drop arriving at the downstream edge with the small pendant drop and the phase of oscillation of the pendant drop. The second effect is an irreproducibility in the drop mass after coalescence.

Magnitudes of the forces working on the drop during pinch off are presented in fig. 3.4.13.

![Force chart](image.png)

**Fig. 3.4.13. Absolute value of various forces working on a drop during pinch off. Left and right pictures show the result of two experiments with identical plates and gas velocities.**

- gas velocity: 25 m/s
- gas: air
- liquid: water+40 mg/l fluoresceine
- Estimated errors: gravitational force: ±10%, total force: ±20%, surface tension: ±10% hydrodynamic force: ±25%

The picture to the left corresponds to the left picture in 3.4.11

Figure 3.4.13 presents the magnitude of the different forces, as well as the total force. Because the drop mass is constant, gravitational force is not time dependant. Furthermore, gravitational force is smaller than surface tension or hydrodynamic forces. This limits the relevance of gravity on droplet break-up. Surface tension forces are strongly time-dependant. At the onset of pinch-off, the drop is almost hemispherical and the total force due to surface tension is nearly zero. Deformation of the drop results in a smaller average radius of curvature, thus increasing $F$. When the drop is stretched, a decrease in length of the contact line results in a smaller force.

Hydrodynamic forces are also strongly time-dependant. In the first measurement (00050804/01) the hydrodynamic force has maxima at 6, 21 and 37 ms. At these moments, the tip of the drop has just been at its maximum distance, reaching out of the recirculation zone. In the second measurement, the hydrodynamic force has maxima at 40, 70, 105 and 125ms. The maximum at 70 mm is very broad (about
30 ms). Again, this corresponds well with the maximum tip positions at 39, 80 and 119 ms. Between 60 and 85 ms, the tip of the drop is out of the recirculation zone, corresponding to the broad maximum at 70±15 ms.

From this, we conclude that the hydrodynamic force on a drop is large when the drop protrudes from behind the plate, and are small when the drop is in the recirculation zone. Consequently, hydrodynamic forces at break-up in the first experiment are large. In the second experiment, the drop breaks up in the recirculation zone, resulting in lower hydrodynamic forces.

This explains the difference in forces between the first and second measurement, working on the drop at the moment of pinch off. In the first measurement, the drop breaks up when it is in the recirculation zone. Large differences in the hydrodynamic forces acting on the drop breaking up in the recirculation zone (1.2 mN) and outside the recirculation zone (6 mN) can be seen.

**Determination of an estimate of the drag force coefficient**

The determination of the hydrodynamic force that acts on the drop, emanating from above or under the plate, allows us to make an estimate of the drag force coefficient, since drop shape and vapour velocity have been measured. The magnitude of the drag force is calculated using eq. 6.54:

\[
F_D = A_D C_D \frac{1}{2} \rho_v v^2
\]

where \( A_D \) is the frontal area of the drop, which is estimated by:

\[
A_D \approx \frac{\pi}{6} b(x_{\text{top}} - \frac{1}{2} d_{\text{plate}})
\]

\( b \) is the width of the drop and \( (x_{\text{top}} - d_{\text{plate}}) \) is the distance over which the drop protrudes into the vapour flow.

Vapour density and velocity are constant: \( \rho_v = 1.27 \text{ kg/m}^3 \) and \( v_v = 25 \text{ m/s} \). The width of a drop behind a flat edged plate is about 4.8 mm. The force is determined using fig. 3.4.15, at the moment that the drop protrudes from above the plate, and the distance between downstream edge of the plate and tip of the drop is small. This is at 22 ms in the left graph and 70 ms in the right graph. The distances over which the tips emanates are 2.7-1.5=1.2 mm (left) and 2.0-1.5=0.5 mm (right). The magnitude of the hydrodynamic forces is in fig. 3.4.16: 1.4 mN (left) and 0.75 mN (right). Another "useful" point is found in the right figure when the drop protrudes from under the plate 1.1 mm into the gas stream at t=104 ms (\( F_D = 1.2 \) mN).

The corresponding values of \( C_D \) are: 1.3 (left), 1.4 (right, 70 ms) and 1.2 (right, 104 ms). Estimated errors of the drag forces coefficients are 30%.
Gravitational, kinetic and surface free energy of the drop during break-up

The time evolution of total energy and its components during break-up are presented in fig. 3.4.14.

When we look at the first measurement (left graph), we see that the energy of the drop starts to increase when the drop protrudes from the recirculation zone (5, 20 and 35 ms). The energy increases during about 5 ms, during which time hydrodynamic forces are large and the tip moves in downstream direction. When the drop is in the recirculation zone, the total energy decreases, possibly by energy dissipation in the recirculation zone. The increase in surface free energy, which is the result of increase in surface area of the drop, is $2.5 \pm 0.2 \, \mu J$. This is more than necessary to account for the free surface energy of 2 drops of 0.7 $\mu l$, which is about $0.4 \, \mu J$ each.

The last 30 ms of the second recording show the same trend: The free energy increases when the tip of the drop reaches out of the recirculation zone. The first 100 ms of the second experiment however show a different trend. Drop velocity and deformation are small, yielding relatively small values of kinetic and surface free energy. Here, variations in potential energy mainly contribute to changes in the total free energy.
3.4.5 Analysis of break up of drops from a round-edged plate

Break-up of two representative drops from a round-edged plate have been recorded. Tip position and estimated hydrodynamic force are presented in fig. 3.4.15.

Several similarities and differences are observed. In both experiments, the tip of the drop moves non-periodically in a small part of the space downstream of the plate. In the first experiment (left picture of 3.4.15), the average height of the tip before coalescence of the two drops is 0.9 mm under the middle of the downstream edge of the plate. In the second experiment, this is −0.2 mm. The difference in tip position prior to break-up is thought to be caused by a small difference in drop mass, with the drop mass in the first experiment the biggest.

After final addition of the small droplet, the tip of the drop hanging from the downstream edge moves down and upstream. The downward movement is caused by gravitational forces on the drop. The upstream movement is caused by the round shape of the downstream edge. As the drop moves down, it follows the contour of the plate edge. Because the edge is round, a downward moving drop must move upstream. In its lowest position, almost the entire drop protrudes from under the plate. This results in large hydrodynamic forces acting on the drop, accelerating the drop in downstream direction. The tip of the drop makes a counter-clockwise rotation, moving downstream when the drop protrudes into the gas flow under the plate and upstream when the drop protrudes into the gas above the plate.

As the drop emanates for the second time from under the plate, the vertical velocity is much higher than the first time. In the lowest position, vertical distance from the
3.4 Results three mirror test rig

tip of the drop to the middle of the plate is over 4 mm in both experiments; the drop extends far out of the recirculation zone. As a result, hydrodynamic forces are large enough to stretch and break up the drop.

Initial differences in tip position between the first and second experiment are relatively large. The minimum tip height is \(-3\pm0.1\) mm in the first, and \(-1.9\pm0.1\) mm in the second experiment. After about 40 ms, differences between tip trajectories become smaller. Just before break-up, tip trajectories are almost the same.

Hydrodynamic, gravitational, surface tension and total force acting on the drops are presented in fig. 3.4.16.

Fig. 3.4.16. Absolute value of various forces acting on a drop during pinch off. Left and right pictures show the result of two experiments with identical plates and gas velocities. (round edged plate) gas velocity: 25 m/s, T=21.7°C, p=105 kPa gas:air, liquid:water+40 mg/l fluorescine

Estimated errors: gravitational force: \(\pm10\%\), total force: \(\pm20\%\), surface tension: \(\pm20\%\) hydrodynamic force: \(\pm25\%\)

Gravitational, surface tension and total force are determined from the shape, position and acceleration of the drop. The gravitational force is small, (66 \(\mu\)N) because the mass of the drop is small. Both surface tension and hydrodynamic force show large values (over 2.5 mN) at the first 50 ms of the droplet break-up. This is caused by mismatch of the drop shape and has no physical nature. The drop shape is chosen in such a way that it can describe the drop shape using 2 variables: the tip position and the middle of the foot of the drop. This method works well for elongated drops, but fails for very flat drops with a curved foot.

During the last 50 ms, calculated forces are reliable within 20\% for both experiments. In both recordings, hydrodynamic forces are large when the drop protrudes from under the plate and the distance between drop tip and plate is small.
The drag force coefficient is estimated when the drop protrudes from under the plate prior to break up (85ms left graph, 70 ms right graph), using the method presented in the previous paragraph. For a drop behind a round edged plate, b=3.9 mm. Estimated values of the drag force coefficient are $0.8 \pm 0.3$ (left) and $1.0 \pm 0.3$ (right).

**Gravitational, kinetic and surface free energy of the drop during break-up**

In fig. 3.4.17, the surface free energy, potential energy, kinetic energy and the total energy of the drop are presented. For both experiments, potential, surface free and kinetic energy all contribute significantly to the total energy.

In the first recording, the free energy increases between 40 and 60 ms. In this period, the drop protrudes from under the plate and is accelerated and stretched by vapour shear forces. Between 60 and 85 ms, the total energy decreases. In this period, the drop remains in the recirculation zone behind the plate or protrudes above the plate, moving upstream. Because drop and vapour movement are oppositely directed, the drop is decelerated, lowering the kinetic energy of the drop. The decrease in total energy is large (from 80 to 16 μJ), because the drop protrudes far out of the recirculation zone.

In the second recording, the total energy increases between 35 and 50 ms. As in the first recording, the drop protrudes from under the plate during the increase in free energy. Between 50 and 70 ms, the energy decreases from 55 to 30 μJ. The decrease in total energy is much smaller than in the first recording. This is understood from the tip trajectory. Because the tip does not protrude far out of the recirculation zone, deceleration of the drop in the second recording is limited.

**3.4.6 Droplet break-off from a sharp-edged plate**

The break-up of two representative drops from a sharp-edged plate has been analysed. Tip positions are measured during the stretching of the drops and are
used to calculate forces on the drop and the total energy. Tip position and estimated hydrodynamic force during 2 experiments are presented in fig. 3.4.18.

Using a sharp edged plate, the drop grows to a size of $40 \pm 5 \mu l$. Prior to break-up, the position of the tip of the drop is fixed. No oscillating movement (as with the flat edged plate) or irregular movement (as with the round edged plate) has been observed.

When pinch-off starts, the drop moves down, until the tip of the drop protrudes $1/2$ to 1 mm into the gas under the plate. In the lowest position, the drop moves downstream, followed by a counter clockwise rotation of the tip of the drop: downstream in the lowest position and upstream when the drop emanates from above the plate. As the drop moves down again, it protrudes 2 mm into the gas stream flowing under the plate, out of the recirculation zone. Relatively large hydrodynamic forces stretch the drop until it breaks up.

The magnitude of the various forces acting on the drop are presented in fig. 3.4.19. Because the drop mass is constant, gravitational force is not time dependant. The magnitude of the gravitational force is small compared to hydrodynamic forces, even though the drop is relatively large (40 mg).
In both recordings, hydrodynamic forces are large when the drop protrudes into the gas stream under the plate. This is at 11 ms and 56 ms in the first recording and at 16 and 56 ms in the second recording. A weaker maximum is observed when the drop protrudes from above the plate. Forces due to surface tension are also small, except at 10-15 ms in the second recording, when the tip of the drop is at a short distance to the plate.

Fig. 3.4.19. Magnitudes of the various forces acting on a drop during pinch off. Left and right pictures show the result of two experiments with identical plates and gas velocities. (round edged plate) gas velocity: 25 m/s, T=21.7°C, p=105 kPa gas: air, liquid: water+40 mg/l fluoresceine

In both recordings, hydrodynamic forces are large when the drop protrudes into the gas stream under the plate. This is at 11 ms and 56 ms in the first recording and at 16 and 56 ms in the second recording. A weaker maximum is observed when the drop protrudes from above the plate. Forces due to surface tension are also small, except at 10-15 ms in the second recording, when the tip of the drop is at a short distance to the plate.

Fig. 3.4.20. Total energy of a drop and its components during pinch off. Left and right pictures show the result of two experiments with identical plates and gas velocities. Sharp edged plate gas velocity: 25 m/s, T=21.7°C, p=105 kPa gas: air, liquid: water+40 mg/l fluoresceine
Changes in free energy are presented in fig. 3.4.20. During the first 50 ms, the largest component of the total free energy is the potential energy. Surface free energy increases strongly to become the largest component of the drop free energy only in the last 20 ms before the drop breaks up.

The drag force coefficient is estimated when the drop protrudes from under the plate prior to break up (90 ms left graph, 71 ms right graph), using the method presented in the paragraph 3.4.4. For a drop behind a sharp edged plate, b=12 mm. Estimated values of the drag force coefficient are 1.1±0.3 (left) and 0.9±0.3 (right).

### 3.4.7 Comparison between break-off of drops from round, flat and sharp edged plate

Break-up of a pendant drop from the downstream edge of hydrophobic plates with three different edge shapes has been analysed in the previous paragraphs. Gas velocity has not been varied. Drop size at the moment of break-up depends on edge shape, as does the size distribution of the drops formed by break-up. A 40 µl drop (lxbxh=2.4x10.6x3 mm³) is formed on the downstream edge of the sharp-edged plate, while drops of 23 (3x4.8x3 mm³) and 6.7 µl (1.1x3.9x3 mm³) are formed on the downstream edges of the flat and sharp edged plates. Although the volumes and shapes of the drops prior to break-up from a sharp or round edged plate are very different, the process of break-up showed much agreement.

When drops break off a sharp or round edged plate, only a small amount of fluid remains behind on the plate. Most of the fluid (75-90%) breaks up, resulting in a broad drop size distribution with drops of up to 0.5 µl. When a drop pinches off from a flat edged plate, both two drops pinch off, both about 0.5 µl in size. After pinch-off, the remainder of the fluid contracts, forming a 22 µl drop. The drops that pinch off contain only about 5% of the mass of the pendant drop.

At the beginning of break-up, hydrodynamic, surface tension and gravitational forces all but cancel out. As a drop is stretched and the tip protrudes into the gas stream flowing over or under the plate, hydrodynamic forces increase, until the drop breaks up.

The drag force coefficients of drop that protrude into the gas stream has been determined for drops on a flat edged, a round edged and a sharp edged plate. The drag force coefficient of a drop behind a flat edged plate is 1.3±0.4. The drag force coefficients of drops behind a plate with a round and a sharp downstream edge are 0.9±0.3 and 1.0±0.3. This is of the same order of magnitude as the drag force coefficient of a cylinder, which is 1 at Re=6000.

The potential energy of a drop cannot be ignored, although the gravitational force is relatively small compared to the other forces. This is the result of the fact that gravity always acts in the same direction (down), while surface tension and hydrodynamic forces act in directions which change in time. The kinetic energy of
the drop is small at the onset of break-up and large as the first drops pinch off, but it does not increase continuously. Surface free energy is initially small only to become relatively large during the final moments of break-up.

3.5 Conclusions on the pinch-off from horizontal plates

The aim of this chapter has been to answer the following questions:

- What is the governing mechanism of droplet formation during re-entrainment in the pilot plant test rig?
- What is the size of re-entrained drops?
- Can modifying the shape of the downstream edge of the heat exchanger plate change the average size of re-entrained drops?

It is assumed that the results, obtained in the single plate mock-up, can be used for the small scale heat exchanger because:

- Contact angle and surface structure of the heat exchanger plate and the single plate mock-up are nearly identical (PVDF versus PTFE).
- Geometry of the heat exchanger plates and the single plate mock-up are nearly identical (slit and plate 50% larger in the single plate mock up).
- Comparable gas properties (although differences in temperature and relative humidity results in differences in gas density and viscosity of up to 30%).
- Comparable liquid properties (although differences in temperature and the presence of fluorescine has a small influence on surface tension).

An important difference between the single plate mock-up and the small scale heat exchanger is the orientation with respect to the gravitational field; the heat exchanger plates are oriented vertically, while the small scale mock-up is placed horizontally.

It is found that water drops on a hydrophobic plate in flowing vapour do not break-up by vapour shear forces, but slide to the plate edge. Relatively small drops stick to the edge, while larger drops slide off the plate. It is expected that in the small scale heat exchanger, almost no drops detach from the downstream edge for the following reason.

The smallest drop that slides over the plate in the single plate experiment has a volume of 0.03 µl. The smallest drop that pinches off is 2 µl. This means the drop, sliding over a heat exchanger plate, must increase its volume 70 times to be large enough to pinch-off. This is very unlikely, because the heat exchanger plate is 76mm wide and drops in the heat exchanger slide off the plates when their diameter is about 1 mm.

As larger drops slide off a plate, break-up results in the formation of many smaller drops. The larger drops are about half the plate thickness in diameter, although the size distribution after break-up depends on the initial drop size and the edge shape.
Small drops that do not break off the plate coalesce with drops that previously arrived until a large drop is formed which becomes unstable and pinches off. The size of the drop at the moment of pinch-off depends on the shape of the downstream edge of the plate.

The forces acting on a drop during pinch-off have been calculated using shape and acceleration of the drop. Gravity plays an important role during pinch-off, although gravitational forces acting on the drop during break-up are small. The reason for this is that gravity pulls the drop down, out of the recirculation zone behind a horizontal plate into the gas stream under the plate. As a result, drop pinch-off from a vertical plate is probably quite different from the pinch-off of an equally big drop from a horizontal plate. Measured drop sizes can thus not be used to predict the size of condensate drops which are re-entrained from vertical heat exchanger plates.

Drag force coefficients of drops protruding from behind the downstream edge have been calculated for a sharp, a round and a flat edged plate. These can be used to calculate hydrodynamic forces on drops behind vertical plates, because drag force coefficients are independent from the orientation of the gravitational field.
Chapter 4: 
Drop pinch-off at near-critical conditions

4.1 Introduction

4.1.1 Goals & why experiments are performed at near-critical conditions
Drop pinch-off will be investigated to improve our understanding in drop pinch-off from hydrophobic plates, as occurring in the pilot plant heat exchanger (see e.g. paragraph 2.4.1). The experiments, presented in this chapter, are performed at elevated pressure. Most experiments are performed using ethane at near-critical conditions which will allow validation of numerical simulations on droplet pinch-off using the diffuse-interface model. A second use of these experiments is to construct a semi-empirical correlation to predict drop size after pinch-off. Additional measurements using water and ethanol have been performed to achieve a larger range of experimental conditions and to allow comparison between pinch-off from a vertical and a horizontal plate.

Drop pinch-off is studied at near-critical conditions for two reasons. The first reason is that this yields experimental data to validate numerical simulations at conditions where gravity, friction nor surface tension can be ignored. An interface between two phases at near-critical conditions is much thicker than the size of the molecules in both phases. As a result, experiments of droplets that pinch-off at near-critical conditions are useful for validation of numerical simulations using a diffuse interface model. When the measurements are performed at temperatures that differ very little from the critical point, the difference between the thickness of the interface and drop diameter is small, and no numerical scaling of interfacial thickness is necessary. The numerical simulation is treated in section 4.4.

Secondly, small changes in temperature at near-critical conditions result in large changes in surface tension and composition of both phases, without affecting e.g. chemical characteristic of the components or viscosity. Changing the temperature at near-critical conditions thus allows changing the Bond number of a system of constant composition.

This will prove to be a valuable characteristic for the construction of a correlation to predict drop size after pinch-off. These results are presented in section 4.3.

4.1.2 The choice of the experimental system
To investigate droplet pinch-off, two or more phases must be present in the test rig. A two-phase system can be used to perform all needed experiments. The presence of more phases complicates analysis of the experiments and the applicability for comparison with numerical simulations. Thus, a 2-phase system is used to perform the experiments.
Both single component (gas-vapour), two component (gas-vapour or liquid mixtures) and multi-component two-phase systems can be investigated.

The following paragraphs explain why a single component system at near-critical conditions is used to investigate drop pinch-off.

A two-phase system can contain many components. When substance concentrations at the liquid-vapour interface are not constant, surface tension gradients are present, causing fluid movement near the interface. This phenomenon is better known as Marangoni flow, which effects droplet pinch-off. Because the influence of Marangoni flow on drop break-up falls outside the scope of this study, no two phase systems which can have gradients in component concentration at the two phase interface will be investigated. Only three different types of systems remain; single component systems at liquid-vapour equilibrium conditions, multi-component systems where all but one component are non-condensable gases and 2 component systems, where both phases are liquids at thermodynamic equilibrium.

The second goal of this study is to obtain measurements to validate numerical simulations on drop pinch-off, using the diffuse-interface method. This method calculates shear stresses in an interface of non-zero thickness. The thickness of a liquid-vapour interface of a single component system increases with temperature and becomes infinite at the liquid-vapour critical point. At temperatures, which are several Kelvin under the critical point, direct comparison between experiment and simulation without scaling of interfacial thickness should be possible. Therefore, experiments will be performed at near-critical conditions.

Performing experiments at near-critical conditions has a second advantage. Changing the temperature at near-critical conditions allows variation of surface tension over several orders of magnitude. This will be important for the validity range of a semi-empirical correlation to be constructed.

Two different types of critical points are known. The best known type is the liquid-vapour critical point. This is the point in pVT-space (pressure, specific volume, and temperature) at which the difference between vapour and liquid of a single component system disappears. At temperatures above the critical temperature, only a single phase is present. Two-component liquid mixtures can also have a critical temperature, above which only one single phase can be present. At lower temperatures, two phases can be present, dependent of the mass ratio of the components. Both phases are liquid mixtures. This critical point is in many respects similar to the liquid-vapour critical point: with increasing temperature, the composition of the two phases becomes more similar and surface tension drops to zero. Two component liquid mixtures could thus be used to study drop pinch-off at near-critical conditions. However, when liquid mixtures are investigated, differences in mass densities of the two phases are usually small, whereas the difference between condensate and vapour density in the pilot plant heat exchanger is quite large (1000 vs. 1 kg/m³). Secondly, mixing of liquids is mostly investigated using
polymer blends. Because of their high viscosities, only creeping flow is accessible. The pilot plant heat exchanger (like almost any heat exchanger) is not operated in the creeping flow regime. Drop pinch-off of polymer blends will thus have limited value for understanding drop formation in hydrophobic heat exchangers.

Concluding, we have chosen to investigate drop pinch-off of a single component system near its liquid-vapour critical point, because it better simulates pinch-off actually taking place in a heat exchanger.

Choice of component

Transport properties of many pure components near their critical point do not differ much. Thus, for interpretation of the experimental results, the choice of component is of small importance, as long as vapour and liquid viscosities are not relatively large (over 1 mPas) and thermodynamic and transport properties are known. Component choice can thus be based on experimental accessibility.

The liquid-vapour critical point of no known substances is at near-ambient conditions. The critical pressure of most components is over 50 bar. Some components, like Helium, have a low critical pressure (2.2 bar) but the critical temperature is only 5 K, which makes it experimentally nearly inaccessible. Thus, to perform experiments at near-critical conditions, a high pressure test rig will have to be build. Ideally, the critical temperature is as low as possible and the critical temperature just over ambient temperature (about 22°C).

The following criteria have been used to choose a chemical substance for the experiments (in order of relevance):

Primary criteria
- (chemically) stable at near-critical conditions
- critical pressure as low as possible, (less than 100 bar to allow easy construction)
- critical temperature should be well accessible (preferably between 0 and 100°C)
- thermodynamic properties of liquid and vapour at near-critical conditions have to be available in literature

Secondary criteria
- not poisonous/aggressive
- not combustible (explosive)
- readily and economically available as a pure substance
- physical transport properties, such as viscosity, and surface tension should be available in literature
- easy-to-use
Physical properties have been obtained by scanning relevant chemical property data banks (Coulson and Richardson [1983] p765-785, Perry [1997] p2/136-2/139, Platzer [1990], Talcott) for substances that match these criteria. Twenty-five substances have been found that match the primary criteria. Critical temperature and pressure of these components are presented in fig. 4.1.1. Most are refrigerants (e.g. CBrF₃, C₂ClF₅, C₄F₈) or small hydrocarbons (e.g. ethane, ethene, acetylene, propane), but also carbon dioxide, hexafluor sulfide and xenon can be used.

Because of the easy accessibility of the critical point (Tₐ=32.3°C, Pₐ=48.8 bar), its well-documented thermodynamic properties, easy use and good availability, ethane has been chosen. Its combustibility and the lack of well documented liquid and vapour viscosity correlations are the greatest drawbacks.

4.1.3 Structure of this chapter

In this paragraph, the rationale behind the choice of ethane at near-critical conditions to investigate drop pinch-off is presented. The test rig, experimental procedures and methods to calculate vapour velocity and viscosity from drop trajectories are presented in section 4.2.

Experimental results are presented in section 4.3. Paragraph 4.3.1 presents the measured saturation curve of ethane and surface tension and vapour viscosity between 20 and 31°C. Pinch-off of ethane drops from a vertical plate without forced convection is treated in paragraph 4.3.2. Pinch-off of ethane drops in forced convective flow is treated in paragraph 4.3.3. One of these experiments is used to as a case to validate a numerical simulation. Additional experiments on the pinch-
off of water and ethanol drops are presented in paragraphs 4.3.4 (no forced convection) and 4.3.5 (forced convective flow). A semi-empirical equations to calculate the size of the largest drop after pinch-off is constructed. In paragraph 4.3.6, results are compared to measured drop sizes after pinch-off from a horizontal plate. This shows that re-entrainment from horizontal plates results in smaller drops.

A numerical simulation of drop pinch-off in forced convective flow using the diffuse interface method is presented in section 4.4. An introduction of the numerical method is given in paragraph 4.4.1. Thermodynamic background of the diffuse interface method is presented in paragraph 4.4.2. Paragraph 4.4.3 presents the Equations governing fluid flow, while paragraph 4.4.4 presents the numerical method. In paragraph 4.4.5, results are presented and compared to the observed pinch-off, presented in paragraph 4.3.3.

4.2 Experimental set-up: Critical pressure test rig
This section is organised in the following way. In the first paragraph, the design criteria of the test rig are presented. The design of the critical pressure test rig, which is described in paragraph 4.2.2, is based on these criteria. The experimental procedures are given in paragraph 4.2.3. Vapour velocity, viscosity and surface tension are calculated from visual observations. These methods and their accuracy are presented in paragraph 4.2.4.

4.2.1 Test rig design criteria
The critical pressure test rig has been build to study drop pinch-off from a vertical plate at elevated pressure. The test rig should contain a section in which the detachment of a single droplet from the downstream edge of a vertical plate can be studied. Comparison with results obtained using the pilot plant heat exchanger, the 3 mirror test rig and numerical work should be possible.

To increase our understanding in the influence of various forces, like shear forces, gravity and surface tension on droplet pinch-off, control over governing dimensionless numbers, in particular the Reynolds, Weber and Bond number, is required. This implies control over vapour velocity, liquid and vapour mass density, vapour viscosity and surface tension.

Furthermore, it must be possible to have liquid and vapour ethane at equilibrium at near-critical conditions. From this, the following design criteria have been distilled:

- Operating pressure up to 55 bar
- Operating temperature in the range of 10 to 40°C
- Vapour velocity up to 1 m/s (pressure: 40 bar, vapour density: 100 kg/m³)
- Characteristic sizes in test section equal (or scalable) to those in 3 mirror test rig.
- Well-defined geometry in the test section
4.2 Experimental set-up

- possibility to place a droplet on the downstream edge of the plate
- Test section geometry should be easily adaptable
- good optical accessibility of the test section
- Safe handling of ethane at pressures up to 55 bar and temperatures between 0 and 40°C.
- Easy to use
- Fast and low cost manufacturing of the test rig

4.2.2 Description of the critical pressure test rig

The critical pressure test rig (fig. 4.2.1.) consists of a circulation loop containing a pump and a test section.

![Schematic of critical pressure test rig](image)

*Fig. 4.2.1 Schematic of critical pressure test rig (not to scale)*

The loop is constructed using seamless high pressure tubing, designed to be used at pressures up to 217 bar (DIN 50049-3.1B). The internal tube diameter is 18 mm. A pressure sensor is connected to the circulation loop (reproducibility: 0.1 bar, accuracy: 0.25 bar). The flow velocity can be determined both optically and using a pitot tube. The optical method (which is based on particle tracking velocimetry (PTV), is described in §4.2.4.
The temperature is controlled in three ways:
1) Adapting coolant flow and temperature through the heat exchanger upstream of the test section
2) Adapting power of the 0-400W electrical heating wire around the test section
3) Submerging the liquid storage vessel in a water bath

A Pt100 thermoresistor (reproducibility: 0.01ºC, accuracy: 0.1ºC) measures the temperature of the vapour entering the test section. A PID controller has been installed but was not used because manual control proved to be better. During experiments, temperature changed less than 0.02ºC.

Detailed drawings of the test rig and its components are presented in appendix C. The test section consists of a steel tube to withstand pressures up to 55 bar and three inserts to realise the desired plate geometry. Side view, front view and cross sectional area of plate and flow channel are presented in fig. 4.2.2.

The cross sectional area upstream of the plate is almost (max difference 10%) constant, minimising fluid acceleration when the fluid enters the test section. The first insert is build to realise a smooth transition from the cylindrical tubing to the rectangular test section. Over a length of 197.2 mm, the flow channel changes smoothly from circular ($\phi 18$ mm, 254 mm²) to a rectangular channel of 7x40 mm (280 mm²). At the end of the insert, the channel widens smoothly from 7 to 10 mm.
The experiment is performed in the middle insert. It contains a channel which is 40 mm wide, 200 mm long and 10 mm high. A 3 mm thick aluminium plate is placed in the middle of the channel. A small groove in the side walls of the insert keeps the plate in place. Liquid is supplied to the lower edge of the plate through a hole, 1.2 mm in diameter, connected to a tube of 3 mm outer diameter. The connection of the tube to the channel in the plate is 35mm upstream of the plate downstream edge. The influence of the flow around the tube on the flow at the plate downstream edge is negligible.

Glass windows allow optical measurements at the downstream edge of the plate from three sides. (front and sides). For each direction, both a thin inner window and a thick outer window are used. A 1 mm thick window is glued to the flow channel in such a way that the top of the window is in the same plane as the wall of the flow channel. As a result, the shape of the channel is undisturbed. The outer window is 20 mm thick. It is connected to the outer tube of the test section. Rubber seals between glass and test tube prevent leakage and from direct contact between glass and steel. The distance between inner and outer window is 20±0.5 mm. All experiments that are presented in this report have been recorded using the side windows. The observation area is limited by the size of the inner side windows, which is 50x10mm.

Gas bottles to supply nitrogen and ethane are connected to the primary gas inlet of the test rig. Both nitrogen and ethane are 99.5% pure. A second gas bottle containing 99.5% pure ethane in placed top-down, allowing the withdrawal of liquid ethane. It is connected to the secondary gas inlet.

The primary gas outlet from the circulation loop is connected in the proximity of the gas inlet, separated from each other by an 18 mm valve. This valve is closed while cleaning the test rig prior to experiments or when zero superficial gas velocity is desired. Otherwise, the valve is open.

A small fluid storage vessel is connected to the circulation loop under the measurement section. A T-connection, connecting the storage vessel to the circulation loop, has been used as a partial liquid/vapour separator. Liquid can be drawn from the storage vessel using a syringe pump, which is used to form a drop at the downstream edge of the test plate.

The recirculation pump
A recirculation pump has been constructed to allow the vapour to flow through the loop. The pump is a mixed flow type. Drawings are presented in Appendix C. It consists of a chamber in which one or more fans are mounted on an axis. Because the required flow velocities were relatively low, only one fan has been used in experiments at elevated pressure. An adjustable 500W motor powers the axis. A stack of 3 high pressure seals prevents leakage where the axis enters the pump. Three bearings keep the axis in place. At atmospheric conditions, vapour velocities of 20 m/s can be realised using five fans and a 1:5 transmission.
Camera and optics
The camera that is used is described in §7.2. The camera was placed on a tripod at close distance from the test rig. Frame rates varied between 500 and 2250 Hz. Two adjustable 200W light sources with infrared filters to minimise heating have been used for lighting. Droplets have been illuminated either through the front window or from behind, with some paper sheets in between lamp and test rig to diffuse the light.

The optical path is perpendicular to the 20 mm thick outer window to minimise optical deformation, see fig. 4.2.3.

A grid with 0.25 mm dots at 1mm spacing on a paper sheet has been used for calibration. Before each measurement series, the test section is opened and the grid is inserted at the centre of the channel. The camera is focussed on the grid and some pictures are taken, after which the grid is removed. After the measurements, the test section is opened and the calibration grid is inserted again. Both calibration recordings have been used to determine magnification, optical deformation and their accuracy.

Recordings of single droplets pinching off have been stored as a series of pictures (*.tif-files). Systematic naming has been used, see §3.3.2 Some recordings have also been stored on video. These recording have been digitised and compressed as mpeg’s.

![Fig. 4.2.3 schematic of optical set-up, front view. Sizes in mm. Mirror and camera are in the horizontal plane.](image-url)
4.2 Experimental set-up

4.2.3 Operation and Experimental Procedures

Leak testing
Before the experiments, the test rig is pressurised to 10 bar using nitrogen or compressed air. Leaks are traced using a non-corrosive leak tester. Special attention is given to the windows (which are opened to insert the calibration grid), recently disconnected flanges and seals (pump axis, thermometer and syringe pump). When no leaks are found, the test rig is pressurised to 20 bar. A pressure drop of 1% in 15 minutes (which equals the accuracy of the pressure sensor) has been chosen as an acceptable value.

Cleaning
Since small amounts of impurities can affect surface tensions (and thus the experimental results), a thorough cleaning procedure of the test rig has been followed. The method described here has been used for the measurements using ethane at near-critical conditions. A similar procedure has been followed for the measurements with water or alcohol droplets in compressed nitrogen.

Since some experiments have been performed using water or alcohol, rust formation can be a problem. In between measurements, the test rig was flushed with dry air to avoid rust formation. A day before the measurements, the test rig was opened and checked for rust or deposits. Components were replaced or cleaned when necessary.

Prior to performing experiments, all air in the test rig is replaced with ethane. Air in the recirculation loop is easily replaced, but air in the capillary leading from the syringe to the test section is not. Thus, first a small amount (but at least 10 times the capillary volume) of ethane was blown through the capillary. The test rig is sealed off and the recirculation valve closed.

Next, both primary inlet and outlet are opened and ethane if fed trough the recirculation loop, consequently replacing the air in the test rig with ethane. The amount of ethane fed trough the test rig was at least 5 times the test rig volume. Next, ethane was fed to the test rig through the secondary inlet (~3 times the test rig volume) to replace air in the storage vessel, followed by flushing from the primary inlet (at least 5 times the test rig volume).

The capillary was cleaned again by flushing with ethane (about 3 times the capillary volume) using the syringe pump. We can assume that no impurities are present in the capillary, but dead ends in the test rig, like pressure sensor, thermometer and pitot tube, can still contain remains of air.

The recirculation valve was opened and the pump was operated at full speed to allow good mixing, thus replacing impurities in the dead ends. Finally, the recirculation valve was closed again and the at least 5 times its volume was fed through the test rig. All air in the test rig is now replaced with ethane.
Installing process conditions
After cleaning the test rig, the recirculation valve is opened and all inlets and outlets are closed. The test rig is pressurised to 10 bar using ethane from the primary inlet and checked for leaks at some critical points (windows, seals, recently opened and closed connections etc.).

Ethane vapour is now fed from the primary inlet to the test rig until the pressure no longer rises. After this, liquid ethane is fed from the secondary inlet into the test rig. When more ethane is fed but the pressure is constant, we know that both liquid and vapour must be present. Next, a droplet is placed on the downstream edge of the plate using the syringe pump, proving that both liquid and vapour are present. The test rig is now ready for measurements at room temperature. To perform measurements at elevated temperatures, more liquid is added to the test rig (through the secondary inlet) and the storage vessel is heated using a 40ºC water bath and electric heating of the test section. When the desired temperature is reached, the water bath is removed and insulation is re-installed. Measurements at near-critical conditions can be performed.

Performing experiments
Using the syringe pump, a small droplet is placed on the plate edge. The lighting is adjusted until a good picture is recorded. After this, the pump is switched on and the desired vapour velocity is chosen by adjusting the recirculation pump settings. Temperature and pressure are recorded, and immediately after this a droplet is produced using the syringe pump. The drop break-off is recorded with the high speed camera. After this, temperature and pressure in the test rig are recorded again. When the difference in pressure before and after the experiment exceeds 0.1 bar, or the difference in temperature exceeds 0.02ºC, the measurement is discarded and a new experiment is performed.

In some experiments, the droplet has been produced by condensation using the top heat exchanger.

For about 2 s, an ice-water mixture of 0ºC is fed through the top heat exchanger. This results in the condensation of ethane above the measurement section. The liquid drains down through the test section, partially over the plate, forming droplets at the lower plate edge. For approximately the first 30 seconds, multiple drops are present under the test plate simultaneously. After about 30 seconds, the amount of draining liquid decreases and a single drop grows under the test plate until it pinches off. At near-critical conditions, this method proved to be more successful than the formation of drops using the syringe pump.

Shutting down the test rig
After all experiments have been performed, the vapour exit is opened for about 20 sec. Temperature of the ethane vapour might decrease by as much as 150ºC, as a result of expansion in the pressure reduction valve. To prevent the valve from freezing solid, the pressure reduction valve and the piping before and after the
valve are heated using a 400W heating wire. After a few minutes, the vapour exit is opened again for 20 sec.

It proved to be impossible to burn the ethane because of its irregular supply. Instead, ethane is mixed with large quantities of air (at least 100 times more) after which no risk of explosion is present, and fed to the exhaust.

After the experiments, the test rig is flushed with nitrogen (to remove ethane remains) and connected to a continuous supply of dry air.

### 4.2.4 Methods to determine vapour velocity, viscosity and surface tension

The average vapour velocity, viscosity and surface tension cannot be determined directly. The vapour viscosity is calculated from the falling velocities of 2 droplets of different size. The surface tension is calculated from the maximum size of a pendant drop. Finally, the maximum vapour velocity is calculated from the falling velocity of small droplets. The methods will be described in this paragraph.

#### A method to determine the surface tension coefficient

The surface tension of ethane is determined from the size of a drop just before pinch-off in quiescent vapour. As described in §4.2.3, a drop of liquefied ethane is formed on the lower edge of the test plate when the saturated ethane in the heat exchanger above the test section is cooled using a water/ice mixture.

The drop grows slowly, typically in about 5 sec, after which the drop stretches and pinches off from the plate. The moment of the onset of pinch-off is determined from the drop shape, which changes slowly when the drop is stable and changes rapidly when the drop is pinching off. Because pinch-off is fast compared to the growth of the drop (typically 50 ms), the error in drop volume that results from a wrong choice of the moment when pinch off starts is very small, about 0.2%. The error of the drop volume is thus determined by the error in the drop size, which is about 5%. A schematic of a pendant drop just before pinch-off is given in figure 4.2.4.
The surface tension is calculated from the mass of the pendant drop, see §3.2.8:

\[ \text{Bo} = \frac{\Delta \rho g r_{eq}^2}{\sigma} = 2.752 \]  

3.2.12

where \( r_{eq} \) is the volume-averaged radius of the pendant drop and \( \Delta \rho \) is the mass density difference between liquid and vapour.

The volume-averaged radius \( r_{eq} \) is calculated from the height of the drop and the radius of its foot, \( r_{foot} \):

\[ r_{eq} = \frac{h}{2 \sqrt{r_{foot}}} \]  

4.2.1

The only unknown in 3.2.12. is the surface tension. The experimental error of the surface tension is twice as high as the error in \( r_{eq} \), 10%.

A method to determine the vapour viscosity.

Vapour viscosity is determined to allow calculation of the Reynolds number of the flowing vapour. Droplets fall through a fluid with a well predictable velocity, the Stokes velocity. For slow-moving small drops the falling velocity depends on the mass density difference between vapour and liquid, the drop size and vapour viscosity. For larger droplets surface tension also effects the terminal falling velocity.

Since all other constants are known, measurement of the terminal falling velocity enables calculation of the vapour viscosity.

High speed footage of falling droplets through vapour allows us to determine the droplet falling velocity and its diameter. By turning off the pump and closing the recirculation valve we assure that the cross-sectional averaged vapour velocity is zero. However, even small temperature differences (of about 0.05ºC) produce density differences, which result in a recirculation flow inside the test section. Local velocities can be as much as 5 cm/s, which is of the same order of magnitude as the terminal falling velocity. Thus, the local fluid velocity has to be determined to calculate the terminal falling velocity.

This method uses 2 drops, which are different in size and flow along (almost) the same path. As a consequence, the droplets experience the same fluid velocity. Large differences in the drop size are desirable, causing a large difference in terminal falling velocity. The size of the biggest drop is limited, because very large drops are no longer spherical or ellipsoid, and terminal faaling velocity relationships are valid only for spherical and ellipsoid drops.

Drops are produces by refreshing the coolant in the heat exchanger above the test section for a short period, typically \( \frac{1}{4} \) s. A few seconds later, several drops of varying sizes fall into the measurement section. Drop movement is recorded and the recordings are screened for suitable pairs. A pair is suitable when:
4.2 Experimental set-up

1. both drops are spherical or ellipsoid
2. diameter of the large drop is at least twice the diameter of the small drop
3. drop position at the moment of velocity measurement differs less than the radius of the large drop.

About 0.5-5% of the drops forms a suitable pair. As a result, several recordings were necessary to find suitable pairs.

Measuring the size and velocity of two drops results in a (solvable) set of 2 equations with 2 unknowns:

\[
\begin{align*}
\nu_1 &= f(\rho_v, \Delta\rho, \eta_v, d_1) + v_v \\
\nu_2 &= f(\rho_v, \Delta\rho, \eta_v, d_2) + v_v
\end{align*}
\]  

4.2.2

As a first estimate, we assume that the vapour velocity equals the falling velocity of the smallest droplet. Assigning subscript 1 to the small droplet, we get:

\[
v_2 = f(\eta_v, d_2) + v_1
\]  

4.2.3

Since this equation contains only 1 unknown, it allows us to give a first estimate of the vapour viscosity.

Substituting the viscosity in the equation of motion of the first droplet results in a better estimate of the vapour velocity:

\[
v_v = v_1 - f(\eta_v, d_2)
\]  

4.2.4

Repeating this procedure several times results in fast convergence.

**Calculation of the terminal falling velocity.**

To determine the viscosity of vapour ethane, the terminal falling velocity of a single drop has to be calculated. A method presented by Janssen [22] is used.

First, 4 dimensionless numbers have to be calculated:

\[
C_D = \frac{4g(\rho_l - \rho_v)d}{3\rho_v v_1^2}
\]  

4.2.5

\[
We = \frac{\rho_v v_1^2 d}{\sigma}
\]  

4.2.6

\[
Re = \frac{\rho_v v_1 d}{\eta_v}
\]  

4.2.7

\[
M_O = \frac{g\eta_0 f(\rho_l - \rho_v)}{\rho_v^2 \sigma^3}
\]  

4.2.8

**Fig. 4.2.5**  Method to calculate the terminal falling velocity. Dimensionless numbers are defined in eq. 4.2.5-4.2.8
Two combinations of these dimensionless numbers are plotted against each other, see fig. 4.2.5. The dimensionless group on the x-axis \((x=\text{C}_0\text{We}/\text{Mo}^{0.15})\) is independent of the droplet velocity: it contains only the drop size and substance properties. The figure allows us to determine the y-value graphically \((y=\text{ReMo}^{0.15})\). Here, y is calculated using the equation:

\[
y = \frac{x}{(2.22 - 18.7 \cdot 10^{-3} x - 4.218 \cdot 10^{-6} x^2)}
\]  

4.2.9

Maximum difference between graph and equation less than 1%.

Now the Reynolds number (and thus the terminal falling velocity) can be calculated.

**A method to calculate the average vapour velocity**

To calculate Reynolds and Weber numbers, the average vapour velocity in the measurement section must be known. Because the accuracy of the velocity measurements using the pitot tube was not satisfactory, a different method has been developed to calculate the average vapour velocity. This method, which uses observed droplet velocities, is presented here.

The local vapour velocity is calculated by subtracting the terminal falling velocity from the observed falling velocity of drops. We can assume that the largest velocity that has been calculated in this way, after observing a large amount of drops, deviates only a small amount from true maximum vapour velocity in the measurement section \((v_{\text{max}})\).

The goal of this section is to calculate the number of drops that has to be observed to give a 95% accuracy interval that deviates less than 2% from \(v_{\text{max}}\).

For low Reynolds numbers \((\text{Re}<2000)\), the velocity profile of a well developed flow between two parallel flat plates is given by:

\[
v = 4v_{\text{max}} \frac{x(d-x)}{d^2}
\]  

4.2.10

Where x is the distance to the wall and d is the width of the slit.

This assumption \((\text{Re}<2000)\), will be justified in chapter 10. Downstream of the plate, the velocity profile is not well developed and this relationship cannot be used. This relationship is, however, valid for the slit between the plate and wall of the test section, see fig. 4.2.6.

Thus, droplet velocities will be determined in this slit.
4.2 Experimental set-up

Integrating this velocity profile and dividing by the slit width gives the average velocity:

\[ v_{\text{avg}} = \frac{1}{d} \int_0^d v \, dx = \frac{2}{3} v_{\text{max}} \quad 4.2.11 \]

To determine the number of drops to be used for estimating the velocity with a good accuracy, we assume that the drop number density is constant in the slit. Consequently, the experimentally determined velocity distribution is the same as the velocity distribution of the drops in the slit after correction for terminal falling velocity. Using eq. 4.2.10 allows us to calculate the normalised velocity distribution:

\[ \frac{v}{v_{\text{max}}} = 4 \frac{x(d - x)}{d^2} \quad 4.2.12 \]

Two new variables are introduced, Θ and φ. φ is the relative difference between velocity \( v^* \) and the maximum vapour velocity.

\[ \phi = \frac{v_{\text{max}} - v^*}{v_{\text{max}}} \quad 4.2.13 \]

Θ(φ) is the fraction of drops for which the difference between the local vapour velocity and maximum vapour velocity is smaller than φ. As an example, we’ll calculate how many drops have a velocity that differs less than 25% (φ=\(1/4\)) from \( v_{\text{max}} \):

\[ v^* = \frac{3}{4} v_{\text{max}} \]

\[ \phi = \frac{1}{4} \]

---

Fig. 4.2.6 schematic of measurement section and test plate. Velocities are determined in the slit, upstream of the plate downstream edge.
In figure 4.2.7, the normalised velocity distribution, given by eq. 4.2.12, is presented for half the slit.

\[ v(x) = v(d-x) \]

All drops which are located in the middle half of the channel have a velocity which is higher than \( \frac{3}{4}v_{\text{max}} \). Because we have assumed that the drop density distribution is homogeneous, 50% of all drops have a velocity which is higher than \( v^* \). The velocity of half of all drops differs less than 25% from the maximum velocity: \( \Theta(0.25) = 0.5 \). In the same way, we see that \( \Theta(1/9) = \frac{1}{3} \) and \( \Theta(1/16) = \frac{1}{4} \):

\[ \Theta = \sqrt[4]{\phi} \]  \hspace{1cm} 4.2.14

After measuring the velocity of \( n \) drops, the chance that all have a velocity smaller than \( \phi \) is:

\[ 1 - \Psi = (1 - \Theta)^n \]  \hspace{1cm} 4.2.15

Where \( \Psi \) is the chance that of \( n \) drops, at least 1 has a velocity equal or larger than \( v^* \).

Since the drop with the highest velocity is used as an estimate of \( v_{\text{max}} \), \( \Psi \) is also the accuracy with which the velocity is calculated.

Substituting eq. 3 in eq. 5 gives:

\[ n = \frac{\log(1 - \Psi)}{\log(1 - \sqrt[4]{\phi})} \]  \hspace{1cm} 4.2.16
The desired accuracy is a 95% accuracy interval with a relative error of less than 2%. This requires the measurements of 20 individual drop velocities:
\[ n = \frac{\log(1 - 0.95)}{\log(1 - \sqrt{0.02})} = 19.6 \]

4.3 Results Critical pressure test rig

The experiments in this chapter serve two goals. Firstly, several experiments using saturated ethane are performed to validate numerical calculations of drop pinch-off in forced convective flow, see section 4.4. Secondly, the experiments are used to determine how drops pinch off from a vertical plate at non-zero vapour velocity. These experiments are performed using saturated ethane, water and alcohol.

In the first paragraph of this chapter (§4.3.1), the saturation curve, surface tension and vapour viscosity of ethane, measured along the saturation curve, will be presented.

The following paragraph (§4.3.2) presents result on pinch-off of ethane drops in quiescent vapour. Pinch-off of ethane drops in downward flow at various experimental conditions is presented in §4.3.3.

Pinch-off of water and ethanol drops has been studied at various pressures and vapour velocities. These experiments are presented in paragraphs 4.3.4 and 4.3.5. A comparison between these experiments and pinch off from a vertical plate (§3.3.4) is made in paragraph 4.3.6.

4.3.1 Measured substance properties of ethane

In this paragraph, the temperature-pressure relationship of saturated ethane at the two-phase curve is presented. Next, surface tension and the vapour viscosity of saturated ethane are presented. Vapour viscosity and surface tension are needed for the analysis of droplet break-up. Experimental data on the liquid-vapour equilibrium curve of ethane is available (Sychev, [1987]).

Liquid-vapour equilibrium measurements and validation of equilibrium conditions

The saturation pressure of ethane between 10°C and the critical temperature has been determined from recorded temperatures and pressures during drop break-up experiments in the critical pressure test rig. Drops have been formed using two different methods:

A droplet injection at room temperature
B droplets formed by condensation
Measured temperature-pressure relation is plotted in figure 4.3.1.

![Graph showing temperature and pressure relation for ethane at near-critical conditions]

*Fig. 4.3.1 Measured temperatures and pressures of 2 phase mixture of ethane at near critical conditions*

The two phase curve, as reported by Sychev [1987], is presented using a solid line. The measurements performed at room temperature (series A) show good agreement between measured pressure and calculated equilibrium pressure.

In series B, droplets are produced by condensation in the heat exchanger above the test section. The continuous presence of both vapour and small droplets in the test section guarantees equilibrium conditions. Some points of this series fall a somewhat (~0.5 bar) under the saturation curve. This is especially clear with measurements performed without forced convection, which is easily explained in the following way. The Pt100 that has been used to measure the temperature is connected to a (well insulated) metal flange of considerable size. Both the heat capacity (~1kJ/K) of the flange and thermal conductivity of the thermometer (25 mW/K) are large. During the condensation experiments (B), ethane temperature was lowered by about 2°C/hour. Thus, the flange was warmer than the ethane 2 phase mixture. At non-zero vapour velocities, vapour flows around Pt100, causing high heat transfer (>25 mW/K). As a result, the measured temperature corresponds to temperature of the vapour. At zero vapour velocities, heat transfer through the vapour is less good and the temperature deviates towards the flange temperature (higher temperatures). The test section is, however, still at equilibrium conditions.

**Surface tension of ethane at near critical conditions**

The surface tension has been determined experimentally, because it is an important quantity for the analysis of the experimental results (§4.3.2-§4.3.5).
4.3 Results critical pressure test rig

Surface tension has been determined from the mass of a pendant drop just before break off.

Measured surface tensions (using both methods) at different temperatures are presented in figure 4.3.2.

![Surface tension vs. temperature graph](image)

*Fig. 4.3.2 Surface tension of ethane, measured using the pendant drop technique. Calculated correlation constant are: $\sigma_0=47.6 \times 10^{-3} \text{mN/m}$, $n=1.207$. Error bar: 2 standard deviation*

The average standard deviation of the experimentally determined surface tension using the pendant drop method was about 1.5% (4 measurements) at room temperature and increased to about 4% (4 measurements) at near critical conditions.

A correlation for surface tension vs. temperature at near critical conditions is reported by Yaws [1999], and Perry [1997]:

$$\sigma = \sigma_0 (T_c - T)^n$$  \hspace{1cm} (4.3.1)

Fig. 4.3.2 shows that this relationship fits the surface tension measurements using the pendant drop method quite well. Calculated constants using least squares method are: $\sigma_0=47.6 \times 10^{-3} \text{mN/K}$, $n=1.207$. This surface tension according to Yaws [1977] is 5% higher.

**Viscosity of ethane at near critical conditions**

The viscosity of ethane will be necessary to analyse the droplet break-up experiments (of ethane) in flowing vapour. Thus, a method has been developed to calculate the viscosity of ethane vapour from the terminal falling velocity of 2...
droplets of different size, which fall through an (almost) quiescent vapour along the same flow line. This method is described on §3.5.3.

Measured vapour viscosities are presented in fig. 4.3.3.

As commonly found for gases and vapours, vapour viscosity increases with temperature. The increase in vapour viscosity with increasing temperature is high at temperatures just under the critical temperature is high, which is also a common phenomenon [44].

In chapter 11, vapour viscosity at a temperature of 25.4°C is needed. It is determined by logarithmic interpolation between the viscosity measured at temperatures of 20.4 and 27.5°C:

$$\eta = 10^{(0.046T-1.16)} \text{ (mPas)}$$ 

Temperature in °C, estimated error (95%) is 10%.

A relationship to calculate viscosity of liquid and vapour ethane was found (Yaws, [1977] and [1999]). It is clear from fig. 4.3.3 that there is disagreement with the experimentally determined viscosity. The following points are reasons to question the validity of Yaws’ equation:

- According to Yaws’ relationships, liquid and vapour viscosity of ethane at the critical temperature are different by a factor of about 2.5, whereas vapour and liquid viscosity are equal at critical conditions.
- For most small hydrocarbons, vapour viscosity is pressure-dependant at near-critical conditions (see e.g. Altunin [1987]). Yaw’s relationship does not capture the dependence of pressure on viscosity.
For most small hydrocarbons, vapour viscosity is strongly temperature-dependent at near-critical conditions (see e.g. Altunin [1987]). Yaw's relationship predicts a weak temperature dependence of viscosity.

No experimental data was presented by Yaws, and the sources that Yaws uses could not be traced. The values reported by Yaws are probably valid at low pressures only, and thus do not hold at near-critical conditions.

### 4.3.2 Pinch-off of ethane drops without forced convection

The goal of this paragraph is to construct a relationship giving the size of the main drop after pinch-off from a vertical plate in quiescent vapour at near-critical temperatures.

Pinch-off of liquid drops in quiescent vapour has been studied using ethane at liquid-vapour equilibrium. 19 experiments have been performed with temperatures ranging from 20°C to 31.2°C. Before the experiments, the test rig is filled with ethane vapour. A small amount of liquefied ethane (10-100 ml) is present in the storage vessel under the test section as part of the measures to ensure equilibrium conditions.

At the start of each experiment, a small amount of liquefied ethane (1-10 ml) is produced by cooling the vapour in the heat exchanger above the measurement section, see chapter 9 for details. The cooling water is turned on for about 1 second, refreshing the cold water in the heat exchanger. As a result, ethane condenses and drains down the test rig. Although most liquid drains down over the outer wall of the test section, a small amount of the liquefied ethane forms drops on the lower edge of the test plate. In the first half minute after refreshing the cooling water, a large amount of droplets are produced, pinching off from various places from the downstream edge of the plate. After some time, the number of droplets growing on the downstream edge of the plate decreases. The camera is turned on when only a single drop is observed to grow, normally after about 30-60 sec.

In about 5 sec, a drop grows to its maximal stable size. Surface tension forces no longer balance the droplet weight, and the drop starts to elongate. The middle of the drop contracts, and three parts of the liquid are identified: a large droplet at the bottom, a slim fluid column (the neck) in the middle and a foot, attached to the plate. A typical example of the subsequent break-up of the droplet is presented in fig. 4.3.4.
The neck contracts resulting in pinch-off at the point where the neck meets the main drop.

At the moment of pinch-off, the distance from plate to the pinch-off point is 2.2 times the volume averaged diameter of the droplet that pinched off. After pinch-off a thin column remains, which contracts. The thickness at the free end increases, while the thickness at the top (where the column is still connected to the foot) decreases. At the same time, an instability propagates upwards, leading to the formation of 6 satellites. The three smallest satellites coalesce, resulting in the overall formation of four satellites.

The shape of the drop after pinch-off is almost spherical. Inertia of the fluid drained from the column causes the droplet to deform. Two different processes are observed. A surface wave is observed travelling around the droplet and the top of the droplet is flattened. 13 ms after pinch-off (at 20.4 °C), a small amount of fluid protrudes at the bottom of the droplet (arrow in fig. 4.3.1). This is thought to be caused by the impulse of the fluid that drained from the column into the droplet. The protrusion retracts into the droplet, after which the droplet starts to oscillate with a frequency of 19±1 ms.
4.3 Results critical pressure test rig

Size of ethane drop after pinch-off
The size of the main droplet after pinch-off in quiescent vapour has been determined at temperatures between 20.4 and 31.2°C. With increasing temperature, the drop size decreases, see fig. 4.3.5.

\[
\Delta = \sigma - \rho g r_v^2
\]

As shown in §4.3.1, surface tension of ethane decreases with increasing temperature. As the surface tension decreases, the size of a pendant drop just before pinch-off decreases.

To find the relationship between drop size before and after pinch-off, we plot the measured radius of the drop after pinch-off versus the calculated radius of the drop before pinch-off \( r_{eq} \), see fig. 4.3.6.
Drop pinch-off at near-critical conditions

\[ r = \frac{C^{-}}{\sqrt{2}} r_{eq} = \frac{C^{-}}{\sqrt{2}} \sqrt{\frac{\rho}{\Delta \rho g}} \approx (1.26 \pm 0.02) \sqrt{\frac{\rho}{\Delta \rho g}} \]

where \( C^{-} \) equals 0.96±0.01

4.3.3 Pinch-off of ethane drops in forced convection

Pinch-off of ethane droplets has been studied at constant temperature (24.5 +/- 0.1°C) and pressure (41 +/- 0.2 bar). Drops are formed by condensing liquid in the heat exchanger upstream of the measurement section. The vapour velocity has been varied between 0 and 0.55 m/s. The vapour velocity that is reported in this chapter is the superficial vapour velocity downstream of the plate downstream edge, as determined in paragraph 9.4.
The pinch-off of 6 drops at vapour velocities under 10 cm/s has been recorded. Reproducibility of the experiments is high. Droplet break-up strongly resembles the pinch-off at zero flow, described in the previous paragraph. As in the experiments with quiescent vapour, the pendant drop is hanging in the centre of the plate. 6 or 7 satellites are formed, of which the smaller ones coalesce, resulting in 4 or 5 satellites. As with pinch-off of ethane drops in quiescent vapour, the main drop oscillates after pinch-off.

The pinch-off of 5 droplets at a superficial gas velocity of 0.16 m/s has been recorded. Many similarities, but also quite some differences with pinch-off in quiescent vapour are observed. A typical example is presented in fig. 4.3.7.

![Fig. 4.3.7. Typical example of drop pinch-off from the middle of the downstream edge of a plate (D=3 mm) at relatively low vapour velocity](image)

Before break-off, the fluid column (the neck) between the main droplet and foot is somewhat longer (about 6$\ell_{aq}$), than the length of the neck at pinch-off in quiescent vapour. After pinch-off, the main droplet is 5±2% smaller. The biggest satellite is 20-50% larger than in the experiments with quiescent vapour. Before pinch-off, small irregularities in the neck shape are found; the neck is not exactly rotationally symmetric, which was the case for pinch-off in quiescent vapour.

Large variations in the number of satellites have been observed, ranging from 1 to over 10. During the measurement, droplets pinched off at intervals of about 5 s. We can therefore assume that temperature, pressure and vapour velocity are constant. Before pinch-off, the pendant drop was not totally stationary, but wobbled a bit. As a result, small differences in the droplet shape at the moment of pinch-off are present. After pinch-off, an instability propagates upward through the neck.
amplifying differences is neck shape. Thus, reproducibility is not as high as with pinch-off in quiescent vapour.

Drop pinch-off has been studied at 5 different superficial gas velocities between 0.30 and 0.55 m/s. At every gas velocity, the pinch-off of at least 4 different drops has been recorded. Two different break-up mechanisms have been observed, which both can occur at the same temperature, pressure and velocity. The first type is identical to the break-up mechanism at lower gas velocities. Before pinch-off, the pendant drop hangs from the middle of the downstream edge of the plate. The drop stretches, and the previously described shape with a main drop, a neck and a foot is observed. At higher gas velocities, the neck shape is less regular and after pinch-off the main drop is smaller, but the general characteristics are identical. This type I will call “centre break up”. The second type will be called “edge break up”.

A typical example of edge break-up is presented in fig. 4.3.8.

![Image](image_url)

**Fig. 4.3.8 Typical example of pinch-off of ethane drop from the edge of a vertical plate, found at vapour velocities exceeding 20 cm/s**

A drop, pendant from the downstream edge of the plate, moves from the middle of the plate towards the plate edge. Here, the droplet is elongated, probably by vapour shear forces, until a filament with a small drop at the end remains. No foot is observed. The filament and the main drop show unsteady irregular motion, while the filament thickness is rather constant over its length. Rupture of the filament results in the formation of one main droplet and 10 or more satellites.

Unlike with centre break-up, the main droplet as well as the satellites have a non-zero horizontal velocity. At vapour velocities that exceed 35 cm/s a third mechanism of droplet formation is observed: large drops break up after pinch-off.
The motion of small satellites reveals that a recirculation zone is present. The length of the recirculation zone increases from about twice the plate diameter at 42 cm/s to four times the plate diameter at 60 cm/s. Shear forces on a drop, that is partially in and partially out of the recirculation zone, are large as a result of the large velocity gradients. This causes the already irregularly shaped main drop to deform further, until it breaks into several smaller ones or some protrusions break off forming separate droplets. At vapour velocities between 35 and 45 cm/s secondary break-up results in the formation of two to five drops. At vapour velocities that exceed 55 cm/s, usually over ten drops are formed.

Size of drops after pinch-off
At vapour velocities under 16 cm/s, all drops pinched off from the centre of the plate. At higher vapour velocities, drops both pinch-off from the centre of the plate as from the plate edge. For both pinch-off mechanisms, the average size of the main drop decreases with increasing vapour velocity. For a given vapour velocity, the droplet that pinched off from the plate edge (edge break-up) is significantly smaller than a droplet that pinched off from the middle of the plate (centre break-up). With increasing vapour velocity, the spread in size of the main drop increases from a standard deviation of 5% at 3 m/s to 15% at 4.2 m/s. The average size of the main drop formed during pinch-off is presented in fig. 4.3.9.

![Fig. 4.3.9. Break up type and average main drop diameter after pinch-off of ethane drop from the downstream edge of a vertical plate. The sizes of the error bars of the drop sizes equal the standard deviation. T=24.4±0.1°C, p=40.9±0.25bar](image)

No edge break-up is observed without forced convection. The fraction of droplets breaking off the edge of the plate increases from 0% at 16 cm/s to 100% at 55 cm/s. A smoothed heavyside function (eq. 4.3.5) has been fitted through the measured values. Since only a limited number (10-20) of droplets pinching off have
been investigated (for one vapour velocity), the accuracy of this correlation is about 10%.

\[ F_c = \left[ \frac{1}{2} - \frac{1}{2} \tanh \left( \frac{v - 0.31}{0.1} \right) \right] \times 100\% \]  

4.3.6

with \( F_c \) the fraction of droplets that pinch-off from the centre of the plate and \( v \) in m/s.

The measured fraction of drops pinching off due to centre break-up and correlation 4.3.5 are presented on the secondary y-axis in fig. 4.3.9

At vapour velocities of 42 cm and higher, little difference can be made between the main drop and the larger satellites. Furthermore, the main drop, as well as the larger satellites tend to break up into smaller droplets in the recirculation zone behind the plate. Also, the spread in drop size is so large, that the number averaged droplet size presents little useful information. Thus, for the higher velocities (\( v=42 \text{ cm/s} \) and \( v=55 \text{ cm/s} \)) the droplet size distribution has been determined (fig. 4.3.10 and 4.3.11) from over 50 drops, both main drops and satellites.

\[ \text{Fig. 4.3.10 Drop size distribution, measured after pinch-off of pendant ethane drop from downstream edge of vertical 3 mm thick plate} \]

\[ T=24.4\pm0.1^\circ\text{C}, \ p=40.9\pm0.25\text{bar}, \ v=42 \text{ cm/s} \]
4.3 Results critical pressure test rig

Because of the relevance of secondary break-up, the drop size distribution has been determined after break-up in the wake behind the plate. Size classes are presented in table 4.3.1.

<table>
<thead>
<tr>
<th>Drop diameter</th>
<th>Name of size class</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 &lt; 0.2 mm</td>
<td>Small</td>
</tr>
<tr>
<td>2 0.2-0.35 mm</td>
<td>Intermediate</td>
</tr>
<tr>
<td>3 0.35-0.7 mm</td>
<td>Large</td>
</tr>
<tr>
<td>4 &gt;0.7 mm</td>
<td>Very large</td>
</tr>
</tbody>
</table>

At superficial vapour velocities of 42 cm/s, the number of small (<0.2 mm), intermediate sized (0.2-0.35 mm) and large drops (0.35-0.7 mm) was of the same order of magnitude. A single drop of 0.71 mm has been detected, which was formed through centre break off. As a result of the larger volume of the big drops, most of the fluid, 78%, is present in large (0.35-0.7 mm) drops.

At superficial vapour velocities of 55 cm/s, the amount of large drops is small (~5%), whereas about equal amounts of drops are smaller than 0.2 mm or have a diameter between 0.2 and 0.35 mm. Most of the liquid volume is present in intermediate sized drops (53% between 0.2 and 0.35 mm), which are large satellites or fragments of broken drops. A slightly smaller amount of fluid is present in large drops (38% between 0.35-0.7 mm). Although half of all drops are smaller than 0.2 mm, the total volume of small drop is only 9%

The error of both the number fraction and volume fraction is proportional to the inverse of amount of droplets of this size, resulting in a substantial error for the large drops.
Secondary break-up
At high vapour velocities, droplets have been observed to break up in the recirculation zone behind the plate. In general, large droplets tend to break up, whereas small droplets tend to stay together. In table 4.3.2, the size of the smallest droplet that broke up and the biggest that did not are presented. One exception has been made. At 0.42 m/s, one (0.71 mm) droplet has been formed by centre break-off.

It was almost spherical in shape, quite unlike all other droplets, which were formed by edge break-off.

This example shows clearly that the not only the droplet size, but also other initial droplet conditions (droplet shape and velocity) influence whether or not a droplet breaks up in the recirculation zone. Because this droplet was not representative for the majority of the droplets (which are at these conditions formed by edge break up), it has been ignored in the analysis of secondary break up.

Table 4.3.2. size of droplets that break up in the recirculation zone behind the plate for ethane liquid in ethane vapour at $T=24.4\pm0.1^\circ\text{C}$, $p=40.9\pm0.25\text{bar}$.

<table>
<thead>
<tr>
<th>$v$ (cm/s)</th>
<th>max. drop diameter that does not break-up in the recirculation zone (mm)</th>
<th>min. drop diameter that breaks up in recirculation zone (mm)</th>
<th>probable boundary value for break-up in the recirculation zone (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>37</td>
<td>0.79</td>
<td>0.81</td>
<td>0.80</td>
</tr>
<tr>
<td>42</td>
<td>0.53*</td>
<td>0.69</td>
<td>0.62</td>
</tr>
<tr>
<td>55</td>
<td>0.48</td>
<td>0.42</td>
<td>0.45</td>
</tr>
</tbody>
</table>

* one 0.70 mm droplet has been observed that did not break (see text)

We will now construct a correlation that describes which droplets break-up in the recirculation zone behind the plate and which drops remain intact.

It is well known that in an long tube ($L/D>10$) with laminar simple shear flow, droplets break up when the local Weber number ($W_{ec}=\rho v^2d/\sigma$) exceeds $8/C_D$ (see e.g. e.g. Froth, p15 [2000]). $C_D$ is the friction coefficient of the channel. $\Delta v$ is the velocity difference between droplet and the surrounding vapour. Lefebvre [1989] presents the same equation, using $We=1.18$ for turbulent flows.

In our case, we don’t know the local velocity field behind the plate, and the assumption of a homogeneous flow field is clearly not valid. We do know, however, that (assuming the droplet has little influence on the vapour velocity and fluid velocity is much smaller than the speed of sound) the flow field depends on the Reynolds number and the geometry alone. Since we do not change the geometry, only the Reynolds number determines the flow field and thus the difference between the local velocity and the droplet velocity, $\Delta v$. 
From the above, it follow that the Weber number with respect to the droplet diameter and the average flow velocity \( \text{We} = \frac{\rho v^2 d}{\sigma} \) must be a function of Reynolds alone:

\[
\text{We}_{\text{drop}} = \text{We}(\text{Re}_{\text{plate}}) \quad 4.3.7
\]

with:

\[
\text{We}_{\text{drop}} = \frac{\rho_v v^2 d_{\text{break}}}{\sigma}, \\
\text{Re}_{\text{plate}} = \frac{\rho_v v D}{\eta_v}
\]

\( d_{\text{break}} \) = minimal drop size for homogeneous droplet break-up in the recirculation zone (mm)

\( D \) = plate diameter (m)

\( \rho_v \) = vapour density (kg/m³)

In fig. 4.3.12, the Weber number has been plotted against the Reynolds number. It increases from \( 22.8 \pm 0.7 \) for \( \text{Re}_{\text{plate}} = 118 \pm 6 \) to \( 29.3 \pm 2.1 \) for \( \text{Re}_{\text{plate}} = 176 \pm 9 \).

![Graph showing the relationship between drop size, characterised using the Weber number, and the flow behind the downstream edge of the plate, characterised using the Reynolds number.](image)

A linear fit has been used to estimate the relationship between We and Re:

\[
\text{We}_{\text{drop}} = a + b \cdot \text{Re}_{\text{plate}}
\]

The values of a and b have been determined using a weighted least squares method.

### 4.3.4 Pinch off of water and ethanol drops without forced convection

The goal of this paragraph is to construct a relationship giving the size of the main drop after pinch off of a drop of water or ethanol from a vertical plate in quiescent vapour.
Pinch-off of water or ethanol drops in quiescent nitrogen has been studied at temperatures between 21.7 and 24.5ºC and pressures between 1 and 51 bar.

At the start of each experiment, a small amount of the liquid (~100 ml) is present in the storage vessel under the measurement section. Liquid is drawn from the storage vessel using the syringe pump and used to form a drop on the lower edge of the test plate. Drop pinch-off has been recorded using high-speed footage of 225 up to 1125 frames/s. A typical example of the pinch-off of an ethanol drop is presented in fig. 4.3.13.

![Image of pinch-off of ethanol droplet in quiescent gas](image)

**Fig. 4.3.13 pinch off of ethanol droplet in quiescent gas**
*T=23.5ºC p=50.8 bar

A typical example of both the pinch-off of an ethanol and a the pinch-off of a water drop from a 3mm plate are presented.

An ethanol drop is pendant from the downstream edge of the test plate. As ethanol is added to the drop, the drop size increases until surface tension forces no longer
balance the droplet weight. The drop starts to elongate and the middle of the drop contracts. As with the pinch-off of an ethane drop, three different parts of the drop are identified: a large droplet at the bottom, a slim fluid column (the neck) in the middle and a foot, attached to the plate. The neck contracts resulting in pinch-off of a 29±3µl drop. The neck breaks at the point where the neck meets the main drop. At the moment of pinch-off, the distance from plate to the pinch-off point is 1.5±0.05 times the average diameter of the droplet that pinched-off. After pinch-off a thin column remains, which contracts. The thickness at the free end increases, while the thickness at the top (where the column is still connected to the foot) decreases. At the same time, an instability propagates upwards, leading to the formation of multiple satellites. The satellites coalesce, resulting in the overall formation of a single 1.2µl satellite. No influence of pressure on the pinch-off has been found.

A typical example of the pinch-off of a water drop are presented in fig. 4.3.14.

![Image of pinch-off of water drop](image)

**name**: 02010213  
**T**: 22.6 °C, **p**: 50.9 bar  
**liquid**: water  
**gas**: nitrogen, **frequency**: 225 Hz

*Fig. 4.3.14 Typical example of pinch-off of water droplet in quiescent vapour*

Pinch-off of a water drop resembles the pinch-off of a drop ethanol. A larger drop is formed (65±5µl) while only a single 0.05±0.03µl satellite has been observed. The length of the neck is 0.7 time the volume-average drop diameter after pinch-off.

**Drop diameter after pinch off**

Drop diameter of the drop after pinch-off is calculated by assuming that the amount of fluid that remains on the plate and the volume of the satellites is small compared to the volume of the main drop after pinch-off. The drop volume before pinch-off is (almost) equal to the volume of the drop after pinch-off.

The method to calculate the volume of a drop before pinch-off, presented in paragraph 6.8, cannot be used to calculate the volume of a water or ethanol drop from a 3 mm plate, because the drop touches both edges of the plate. It is therefore not allowed to use the assumption that the plate under which the drop hangs is infinitely large. Here, a different approach is used, assuming that the gravitational forces due to differences in liquid and vapour density are counterbalanced by surface tension forces:
\[ \Delta \rho g V = \pi \sigma D \]  
4.3.8

or:

\[ V = \frac{\pi \sigma D}{\Delta \rho g} \]  
4.3.9

substituting \( V = \frac{3}{2} \pi r^3 \) gives:

\[ r = \sqrt[3]{\frac{3\sigma D}{4\Delta \rho g}} \]  
4.3.10

with \( r \) the volume of the drop after pinch-off.

Substituting \( \Delta \rho = 800 \text{kg/m}^3 \) and \( \sigma = 22 \text{ mN/m} \) for ethanol and \( \Delta \rho = 1000 \text{kg/m}^3 \) and \( \sigma = 72 \text{ mN/m} \) for water gives a drop size for ethanol of 27\( \mu \text{l} \) and a drop size for water of 69\( \mu \text{l} \). Measured drop volumes are 29±3\( \mu \text{l} \) and 65±5\( \mu \text{l} \). Measured and predicted drop volume correspond quite well.

4.3.5 Pinch off of water and ethanol drops in forced convection

The goal of this paragraph is to present the influence of Reynolds and Weber number of the flow on the size of drops of high surface tension after pinch-off.

Pinch-off of water and ethanol drops in quiescent nitrogen has been studied at temperatures between 21.7 and 24.5°C and pressures between 1 and 51 bar. Vapour velocities varied between 0 and 20 m/s.

Both for ethanol and water, three measurements series have been performed: one with a constant vapour velocity of 0.65±0.5 m/s and pressures between 1 and 51 bar, one at atmospheric conditions with vapour velocities between 0 and 20 m/s and one with constant pressure of 51±0.25 bar and vapour velocities between 1 and 1.3 m/s. This will allow us to differentiate between the influence of the Weber number (\( \text{We} = \rho v^2 d/\sigma \)), the Reynolds number of the flow (\( \text{Re}_\text{plate} = \rho v D/\eta_v \)) and the relative drop size (\( d_c/D = 2/D \sqrt{[3\sigma/\Delta \rho g]} \)).

Pinch-off of water and ethanol at a pressure of and 51 bar and vapour velocities between 0 and 1.3 m/s

At low vapour velocities, droplet pinch-off resembles the pinch off in quiescent vapour. This is not surprising: gravitational force is still the dominant force causing break-up. The characteristic shape of the breaking drop, as seen in quiescent vapour, is recognised. The droplet consists of a foot, attached to the plate, a thin column of fluid and a main droplet, which contains the majority of the fluid.
4.3 Results critical pressure test rig

With increasing vapour velocity, the shape of the droplets becomes more irregular and the column between drop and foot becomes thicker, see fig. 4.3.15.

![Image](image.jpg)

**Fig. 4.3.15. pinch off of a water drop from the downstream edge of a vertical plate**

Break-up of the droplet measured from the time the droplet starts to contract until the last satellite is formed, takes considerably longer. As an example, for ethanol in quiescent vapour, pinch off takes about 30 ms. At 1.1 m/s and 51 bar, pinch-off of a drop of ethanol takes about 200 ms.

At vapour velocities over 1 m/s, no clear distinction between foot, column and main droplet is observed. The droplet is stretched to a column, which elongates and oscillates perpendicular to the plate. The main droplet (which is merely the thicker end of a fluid column) becomes very irregular in shape, even before pinch off. Small droplets have been observed to pinch off from the fluid column before the main droplet pinched off. With increasing vapour velocity, more and bigger satellites are formed, quite often over half the main droplet diameter in size. These phenomena have been observed for both water and ethanol. For water, the vapour velocity at which the same phenomena are observed is higher, and both the formed droplets and satellites are bigger.

**Pinch-off of water and ethanol at 0.65 m/s and pressures between 1 and 51 bar**

At constant vapour velocity, but with increasing pressure (vapour density) the same phenomena are observed. At low pressure (1 bar), little difference is observed between droplet pinch off in quiescent and in flowing vapour. As the pressure increases, the difference between foot, column and main drop disappears, and break up takes much longer.
Drop size after pinch-off
The size at which a drop breaks up is determined by the forces that work on the drop. At zero vapour velocity, only gravitational and surface tension forces work on the drop. The size of a drop after pinch-off in quiescent vapour is calculated using 4.3.9.

The results are validated in the previous paragraph and deviation between measured and calculated drop radii were of the same order as the measurement error, which is about 5%.

The following paragraph is used to expand this correlation to droplet pinch-off of drops in downward forced convective flow.

\[ r_{v=0} = \frac{1}{4} \frac{3 \sigma D}{\Delta \rho g} \]

4.3.11

Measured radii after pinch-off of the main drop versus vapour velocity and vapour mass density are presented in figures 4.3.16-4.3.18.

Fig. 4.3.16 and 4.3.17 show that the difference between the drop diameter at zero vapour velocity and the drop diameter at higher vapour velocities is proportional to \( v^2 \).

![Graph showing drop size after pinch-off](image-url)
4.3 Results critical pressure test rig

Fig. 4.3.17. Size of main drop after pinch-off from vertical 3 mm thick plate. Constant temperature and pressure, vapour velocity between 0 and 20 m/s
ethanol: T=21.0 +/- 0.5ºC, p=1+/− 0.05 bar

Fig. 4.3.18 shows that this difference is also proportional to $p^2$.

Fig. 4.3.18. size of main drop after pinch-off from 3 mm thick plate. Constant temperature ad vapour velocity, mass density between 1 and 60 kg/m³
ethanol: T=23.1 +/- 0.6ºC, v=0.65 +/- 0.6 m/s

Combining both, we find that the drop diameter after pinch-off is calculated using:
\[
\frac{d}{d_{v=0}} = 1 - C(\rho_v v_v)^2
\]

where C is \(2.9 \pm 0.3 \cdot 10^{-4} \text{ kg}^2 \text{m}^{-4} \text{s}^{-2}\) for ethanol and \(2 \pm 0.3 \cdot 10^{-4} \text{ kg}^2 \text{m}^{-4} \text{s}^{-2}\) for water.

This leads to the following equation to calculate the drop size after pinch-off:

\[
\frac{r}{r_{v=0}} = 1 - \left(6.2 - 5.1 \frac{r_{v=0}}{D}\right) \times 10^{-4}(\rho_v v_v)^2
\]

This is a rather unexpected result. The expected result is that the drop size after pinch off depends on the Weber number, which scales with \(\rho v^2\). That this is not correct is shown in fig. 4.3.19, where the diameter of ethanol drops after pinch-off are plotted against the Weber number. No agreement is found between the experiments that have been performed at 1 and 51 bar.

The drop diameter after pinch off, calculated using equation 4.3.11, is plotted against measured drop diameter in fig. 4.3.21. Error bars show the errors of the measurements. This shows that eq. 4.3.11 described the measured data quite well. Equation 4.3.11 should be used carefully; because the physical mechanism is not well understood, extrapolation to different conditions is not advised.

### 4.3.6 Comparison between pinch-off at various conditions

The size of drops after pinch-off has been determined at various experimental conditions. In section 3.4, pinch-off of water drops from a horizontal PTFE plate has been observed in flowing air of 1 bar, 21.7°C at a vapour velocity of 25m/s. A
drop grows at the downstream edge of a flat edged plate until the its volume is 23$\mu$l. The drop is stretched to a length of about 10 mm, after which two drops of 0.7$\mu$l (1.1 mm in diameter) pinch-off. The remainder of the liquid remains behind on the downstream edge.

In paragraph 4.3.4 and 4.3.5, pinch-off of water drops from the downstream edge of a vertical 3 mm thick aluminium plate has been investigated at various vapour velocities and pressures. In quiescent vapour, a drop with a volume-averaged diameter of 4.8 mm pinches off, forming a single small satellite. In flowing vapour, drop size decreases with increasing vapour velocity, while the size of the satellite increases. Pinch-off of water at $v=25$ m/s under atmospheric conditions has not been investigated. However, a correlation has been constructed to calculate the size of the main drop after pinch off. The correlation is valid at these conditions, because the Reynolds, Weber and Bond number fall within the experimental range. The predicted diameter of the main drop after pinch-off is 3.8 mm.

The difference between the drop sizes after pinch-off is quite strong. The experiments from section 3.4 and 4.3 differ in two respects: plates have been constructed from materials with different static contact angles, and plate orientation is different. The effect of contact angle is expected to be small for the following reason. At the onset of pinch-off, the drop elongates and a thin neck is formed, which breaks. Break-up takes places relatively far away from the plate, limiting the effect of the solid-liquid-vapour contact line on break-up.

The difference in drop size, both before and after pinch-off, is thought to be caused by the effect of the plate orientation, which will now be explained.

In paragraph 3.4.4 it has been shown that the magnitude of gravitational force, compared to surface tension and hydrodynamic force, working on a drop on the downstream edge is small. The gravitational forces however pulls the drop down, into the gas stream that flows under the plate. This results in a very large hydrodynamic force. A drop under a vertical plate is also drawn down by the gravitational force, but here the drop stays in the recirculation zone behind the plate and hydrodynamic forces are small. In this way, the relatively small gravitational force has a relatively large effect on drop pinch-off.

When we apply the above to compact hydrophobic condensers, see chapter 2, we find that the orientation of heat exchanger plates has a very significant effect on the size of re-entrained drops. The heat exchanger plates, which are mounted vertically, will produce re-entrained drops that are relatively large. The spacers on the other hand are mounted horizontally, and drops pinching off from the spacers will be much smaller.
4.4 Diffuse interface method to simulate drop pinch-off
In cooperation with V.V. Khatavkar, Eindhoven University of technology

4.4.1 Introduction
The diffuse interface method (DIM) is based on a thermodynamic analysis by van der Waals [48] and Cahn and Hillard [11]. According to van der Waals, a liquid-vapour interface has non-zero thickness (ξ) at near-critical conditions, because the mass density between two phases changes smoothly.

Diffuse interface methods are very suitable to simulate flow with changing topology, like drop pinch-off, because no interface tracking is needed. The interface thickness is, however, much smaller that the domain size. As a result, many gridpoints are necessary to simulate the diffuse interface. To speed up numerical calculations, an artificial increment of interface thickness is used.

Verschuren implemented a DIM and developed scaling rules, i.e. how various dimensionless numbers, Bond, Weber, Peclet and Reynolds number, should change when the interfacial thickness is increased in a numerical simulation. To validate these scaling rules, physical experiments are necessary. In literature, validation experiments have been found for low Reynolds number density matched systems, where surface tension and viscous shear are the dominant forces, and pinch-off in quiescent vapour, where surface tension and gravity are dominant.

In the previous chapter, dedicated experiments have been presented which are useful for validating numerical simulations of two-phase systems at near-critical conditions when both gravity, viscous shear and surface tension forces are relevant. All data, both the initial conditions and footage of break-up at various vapour velocities, liquid and vapour mass density, surface tension and vapour viscosity, are summarised to facilitate future comparisons which, for the sake of time, are not given here.

In this chapter, one experiment and the numerical simulation of drop pinch-off from a vertical plate in slow-flowing fluid at identical conditions are compared. To further speed up calculations, numerical calculations using the DIM are performed on a two-dimensional grid, rather than in 3D.

4.4.2 Thermodynamics of interfaces
In this paragraph, we will calculate the thickness of a flat liquid-vapour interface at rest at temperatures between 0ºC and 31.8ºC. It is assumed that the temperature in the interface is constant. Both liquid and vapour consist of pure saturated ethane.

According to van der Waals, the local Helmholtz free energy (f) of a heterogeneous system is, in every point, dependant on temperature (T), the concentration of all substances (c,) and the concentration gradients:
4.4 Diffuse interface method

\[ f = f(T, c_i, \nabla c_i) \]  \hspace{1cm} 4.4.1

In multiphase systems, the concentration of a component near the interface changes rapidly but continuously between the bulk values in the two phases.

Cahn and Hillard [12] have shown that for a single component system, the Helmholtz energy can be written in the following form:

\[ f = f_0(c, T) + \kappa(\nabla c)^2 \]  \hspace{1cm} 4.4.2

where \( f_0 \) is the Landau-Ginzburg free energy, \( c \) is the normalised specific volume, see 4.4.3, and \( \kappa \) is a coefficient, that can depend on temperature and composition. Following Cahn and Hillard and Verschuren, the dependence of composition on \( \kappa \) will be ignored; \( \kappa = \kappa(T) \).

\[ c = \frac{2v - v_l - v_v}{v_l - v_v} \]  \hspace{1cm} 4.4.3

where \( v \) is the specific volume. In the liquid far away from the interface, \( v = v_l \) and \( c = 1 \). In the vapour, \( v = v_v \) and \( c = -1 \).

\( f_0 \) is the classical term of the Helmholtz free energy, which depends on mass density and temperature. This term can be calculated from PVT-data which are readily available for many substances using 4.4.4. The non-classical term \( \kappa(\nabla c)^2 \) is not readily determined but follows from equilibrium conditions of the interface and measured surface tension in a way that is now described. An in-depth treatment of this deduction is given by e.g. Verschuren [47] and Cahn and Hillard [11]. Experimentally determined PVT data of ethane between 270 and 320 K [36] are presented in fig. 4.4.1

![Fig. 4.4.1 Pressure of ethane versus temperature and specific volume [36]. Solid lines: isothermes; dotted line: saturation curve; circles: saturated liquid and vapour. The area under the dotted line is the two-phase region, where pressure is calculated by interpolation. (Limited smoothness of the lines results from the numerical method, not from a physical phenomenon)](image-url)
f₀ is calculated by integrating pressure versus specific volume at constant temperature.

\[ \left. \frac{\partial f_0}{\partial v} \right|_T = -\frac{p}{v} \quad 4.4.4 \]

A schematic presentation of f₀ is given in fig. 4.4.2.

![Schematic presentation of f₀ versus specific volume for a substance at constant sub-critical temperature.](image)

The homogeneous part of the excess helmholz energy f'₀ is given by:

\[ f'0 = f_0(v_i) - p_{sat}(v - v_i) \quad 4.4.5 \]

For true substances, p_{sat}(v-v_i) is much larger than f₀, which is why a schematic is presented in 4.4.2 rather than the true thermodynamic data.
In fig. 4.4.3, $f'_0$ is plotted versus $c$.

![Graph showing $f'_0$ versus $c$ for different temperatures with a legend indicating 270K, 280K, 290K, and 300K. The graph includes a dotted line for the "ideal" curve.]

Profiles are calculated by applying equation 4.4.5 to thermodynamic data of ethane [36] at 270, 280, 290, and 300K. The independent variable is scaled by dividing $f'_0$ by its maximum value ($f_{0,max}$) between $c=-1$ and $c=1$.

According to Cahn and Hillard, the Helmholtz free energy at near-critical conditions is given eq. 4.4.6. Equation 4.4.6 is plotted in fig. 4.4.3 as "ideal". As temperature increases, the difference between the "ideal" curve and the true $f'_0(c)$ curve of ethane decreases.

$$f'_0 = f'_{0,max} \left( c - 1 \right)^2 \left( c + 1 \right)^2$$  \hspace{1cm} (4.4.6)

The chemical potential is given by the variational derivative of the Helmholtz energy:

$$\mu = \frac{\partial f}{\partial c} - \nabla \cdot \left( \frac{\partial f}{\partial \nabla c} \right)$$  \hspace{1cm} (4.4.7)

When 4.4.2 is substituted in 4.4.7 and the substantial derivative is solved, we find:

$$\mu = \mu_0 + \frac{\partial f'_0}{\partial c} - \kappa \nabla \cdot \nabla c$$  \hspace{1cm} (4.4.8)

Substitution of 4.4.5 and 4.4.6 in 4.4.8 gives:
\[ \mu = \mu_0 + 4f_{\text{max}} c^2 (c - 1) - \kappa \nabla \cdot \nabla c \]  
4.4.9

At equilibrium, the chemical potential is constant. A constant chemical potential allows calculation of \( \nabla c \) from 4.4.8 as follows:

\[ |\nabla c| = \sqrt{\frac{f'_0(c)}{\kappa}} \]  
4.4.10

When the curvature of the interface is large compared to the interfacial thickness, the surface tension can be calculated from the integral of the excess Helmholtz free energy over the interface:

\[ \sigma = F_{\text{int}} = \int_{-\infty}^{\infty} f'_0 + \kappa (\nabla c)^2 \, dx \]  
4.4.11

Substitution of 4.4.10 and assuming that the curvature of the interface thickness is much smaller than the interfacial curvature gives:

\[ \sigma = 2 \int_{-1}^{1} \sqrt{\kappa f'_0} \, dc \]  
4.4.12

Equation 4.4.12 allows calculation of \( \kappa \) for ethane, because both the surface tension and the integral \( \int \sqrt{\Delta f'_0} \, dc \) have been determined experimentally:

\[ \kappa = \sigma^2 \left( \frac{1}{2} \int_{-1}^{1} \sqrt{f'_0} \, dc \right)^2 \]  
4.4.13

The thickness of the interface is defined by eq. 4.4.14. The definition is graphically explained in fig. 4.4.4.

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**Fig. 4.4.4** density profile and interfacial thickness of ethane at 25.4°C. The interface thickness is found from the steepest gradient.
4.4.14

Calculated interfacial thicknesses at temperatures between 0 and 32.2°C are presented in fig. 4.4.5.

Equation 4.4.15 gives the interfacial thickness of ethane versus temperature. Difference between the values calculated with 4.4.14 and 4.4.15 are less than 1.5% at temperatures between 0°C and 31.8°C.

\[ d = 2 \sqrt{\frac{K}{f_{0,\text{max}}}} \]  

4.4.14

4.4.3 Equations governing fluid flow

Vapour flow is governed by the Navier-Stokes equation (4.4.16), the continuity equation (4.4.17) and the component continuity equation, 4.4.18.

\[ \rho \left( \frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} \right) = -\nabla p + (\mu v_c - \nabla f) + \eta V^2 \mathbf{v} + \rho g \]  

4.4.16

\[ \frac{\partial p}{\partial t} = -\nabla \cdot (\mathbf{p} \mathbf{v}) \]  

4.4.17

Fig. 4.4.5 Thickness of a flat ethane interface at temperatures between 0°C and the critical temperature, 32.2°C. Points: calculated using eq. 4.4.14 line: eq. 4.4.15
\[ \frac{\partial c}{\partial t} + v \cdot \nabla c = \Lambda V^2 \mu \] 4.4.18

The non-classical term in the Navier-Stokes equation, \( \mu V c - \nabla f \), incorporates the effect of surface tension, distributed over the interface, on fluid flow.

In eq. 4.4.18, \( \Lambda \) is the mobility.

It is assumed that instantaneous acceleration of the fluid (\( \rho \frac{\partial v}{\partial t} \)) can be ignored.

Non-dimensionalising of the Navier-Stokes equation gives:

\[ \frac{\text{We} \, \rho}{\text{Ca}} p^* (\nabla \cdot \nabla \cdot \nabla) = -\nabla \cdot p^* + \frac{1}{\text{Ca} \, \text{Ch}} (\mu^* \nabla \cdot c^* - \nabla \cdot f^*) + \eta^* \nabla \cdot \nabla \nabla \cdot \nabla + 4.4.19 \]

The dimensionless numbers are given by:

- Weber (We) \( \frac{\rho_v v_{max} D}{\sigma} \)
- Bond (Bo) \( \frac{\rho_v g D^2}{\sigma} \)
- Reynolds (Re) \( \frac{\rho_v v_{max} D}{\eta_l} \)
- Capillary (Ca) \( \frac{\eta_v v_{max}}{\sigma} \)
- Cahn (Ch) \( \frac{\xi}{D} \)

D is the channel width and \( v_{max} \) is the maximum velocity at the vapour inlet.

The subscript “n” indicates that the dimensionless numbers are used in the numerical simulation.

The following method is used to non-dimensionalise the variables from 4.4.16:

- \( \rho^* = \frac{\rho}{\rho_v} = 2p_i \)
- \( v^* = v v_{max} \)
- \( p^* = \frac{D}{\eta_l v_{max}} p \)
- \( f^* = \frac{1}{4} c^4 - \frac{1}{2} c^2 + \frac{\text{Ch}^2}{2} \left| \nabla c \right|^2 \)
- \( \mu^* = \frac{\partial \nabla^*}{\partial \nabla c} \bigg|_{\nabla^*, \text{T}} = c^3 - c - \text{Ch}^2 V^2 c \) 4.4.20

\( \bar{g}^* = \bar{g}/g \) (unit vector)
where the shape factor C corrects for the effects of simulating in 2D in a way that is explained below.

Numerical simulations are performed in 2D to reduce computational time. In the 2D simulation, we want the ratio of the two most important forces, gravity and surface tension, to be identical to the ratio in 3D. In the 3-dimensional world, this ratio is:

\[
\frac{F_g}{F_o} = \frac{\Delta p g}{2 \pi r} = \frac{\Delta \rho g r^2}{3 \sigma}
\]

In the two-dimensional simulation, this ratio is:

\[
\frac{F_g}{F_o} = \frac{\Delta p C g}{2 \sigma} = \frac{\Delta \rho C \pi r^2}{4 \sigma} = \frac{C}{4/3 \pi} \frac{\Delta \rho g r^2}{3 \sigma}
\]

This shows that when the shape factor C is \(4/(3\pi)\), the ratio of gravity and surface tension forces working on the drop in the simulation is identical to the ratio in the 3D-experiment.

\[\nabla = \nabla\n\]

\[\nabla = \nabla\n\]

The dynamic viscosity is calculated by assuming a kinematic viscosity which is independent of mass density:

\[n^* = \frac{n}{n_v} = \frac{2n}{n_v(c-1) + n_v(c+1)}\]

Dropping the asterixes for the dimensionless numbers in 4.4.18 gives:

\[
\frac{We_n}{Ca} \rho (\nabla \cdot \nabla) = -\nabla p + \frac{1}{CaCh} (\mu \nabla c - \nabla f) + n^2 v^2 \nabla + \frac{Bo_n}{Ca} \rho g
\]

De-dimensionalising the continuity equation and dropping the asterixes gives:

\[
\frac{\partial c}{\partial t} + \nabla \cdot v c = \frac{1}{Pe} v^2 \mu
\]

Equations 4.4.23 and 4.4.24 are used to solve a numerical flow problem, which is presented in the next paragraph.
4.4.4 The numerical method

The numerical grid
Equations 4.4.19, 4.4.22 and 4.4.23 are solved simultaneously using a 2-dimensional equidistant grid, see fig. 4.4.6.

Shape of the flow field corresponds to 2D cut through the test section of the critical pressure rig. The grid contains 20,544 points. Distance between to adjacent points in the grid is 0.125mm

Boundary conditions
An overview of all boundary conditions that are used is given in fig.4.4.5. At the left and right borders, vapour velocity is zero and Neumann boundary conditions with respect to pressure and $c$ are applied ($\partial c/\partial n=0$; $\partial p/\partial n=0$). At the inflow, Dirichlet boundary conditions are used for chemical potential and $c$ ($\mu=0$; $c=1$). Vapour flow is downward; the velocity profile is parabolic. At the outflow, Dirichlet boundary
conditions are used for chemical potential and \( c (\mu=0;c=1) \). Horizontal vapour velocity is zero at the outflow, the vertical vapour velocity is not prescribed. At the downstream edge of the plate, pressure, chemical potential and c are not prescribed. Fluid velocity at all solid walls are zero.

**Initial condition**
Initially, a hemispherical drop with \( R=D_{plate}/4 =3/4 \) mm hangs from the lower edge of the plate.

The c-profile in the interface is given by:

\[
c = \tanh\left( \frac{r}{\sqrt{2}Ch} - R \right) \tag{4.4.22}
\]

Where \( r \) is the distance to the origin of the drop and \( Ch \) is the Cahn number.

Inside the drop, \( c=1 \). Out of the drop, \( c=-1 \). In the interface, \( c \) changes from 1 to \(-1\), and the interface thickness equals the numerical interface thickness.

**Solver**
First, the velocity field is calculated (forward) from eq. 4.4.22. Next, chemical potential (4.4.19) and concentration \( c \) (4.4.23) are solved (backward) using an implicit biquadratic conjugate gradient method. For details, see: Verschuren [47].

The default dimensionless timestep is 0.01, but the timestep is reduced to increase accuracy just before pinch-off.

**4.4.5 Results**
Pinch-off of a drop from the downstream edge of a vertical plate has been simulated at the following conditions:

<table>
<thead>
<tr>
<th></th>
<th>Numerical</th>
<th>Experimental</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bond number:</td>
<td>( \frac{\Delta p g d_{\text{drop}}^2}{\sigma} )</td>
<td>33</td>
</tr>
<tr>
<td>Viscosity ratio:</td>
<td>( \frac{\eta_l}{\eta_v} )</td>
<td>2.5</td>
</tr>
<tr>
<td>Weber number:</td>
<td>( \frac{\rho_v v_{\max}^2 d}{\sigma} )</td>
<td>10</td>
</tr>
<tr>
<td>Reynolds number:</td>
<td>( \frac{\rho_v v_{\max} d_{\text{plate}}}{\eta_v} )</td>
<td>21.6</td>
</tr>
</tbody>
</table>
Simulation of pinch-off of a drop is presented in fig. 4.4.7.

Initially, the drop is hemispherical. The drop elongates and becomes less wide. The middle of the drop contracts, resulting in a neck that grows in length and decreases in thickness. At the moment of pinch-off, the length of the drop equals 24.8 mm ($3.3 \, r_{eq}$). The neck breaks at a distance $0.8 \, r_{eq}$ from the plate. Break-up results in the formation of an elongated drop, 2.5 $r_{eq}$ long and 0.9 $r_{eq}$ wide. No satellites are formed. About 12% of the liquid remains behind on the plate.

The experimentally observed pinch-off of an ethane drop has been presented in fig. 4.3.7. As in the numerical simulation, the drop elongates, forming a neck that breaks. Several differences between experiment and numerical simulation are observed. First of all, the neck breaks at a different position. In the experiment, the neck breaks near the main drop, resulting in the formation of a nearly round droplet. In the numerical simulation, the neck breaks near the plate, resulting in the formation of an elongated drop.
Secondly, the number of satellites that is formed is different. In the simulation, no satellites are formed after pinch-off. In the experiment, two satellites are formed.

The numerical simulation is performed to show the usefulness of the experiments for validation of numerical simulations. The amount of detail has proven to be sufficient to show in which respect the results of the simulation are correct, and in which respect the results differ from what is found in an experiment. Thus, the outcome of the simulation is twofold. First of all, several aspects of the simulation are predicted incorrectly. At this stage, this numerical method should not be used to simulate drop pinch-off. Two changes are proposed to improve the outcome of the simulation. First of all, a smaller interfacial thickness should be used. In this simulation, the interface is 0.1mm (0.13 $r_{eq}$). Details, smaller than 0.1mm are thus not captured. Secondly, performing the calculation in 2D is too much of an oversimplification. Capillary forces in the contracting neck are e.g. not captured in 2D. The second and most important result of this simulation is that it shows that the experiments can be used for their main purpose: to validate numerical simulations of droplet break-up under conditions when gravitational forces, shear forces nor surface tension forces can be ignored.
Chapter 5: Conclusions

This study has been performed to increase the knowledge of the mechanisms and consequences of drop pinch-off, as occurring in hydrophobic polymer heat exchangers. Three aspects have been investigated:

- Re-entrainment has been studied to quantify the effect of drop pinch-off on the overall heat and mass transfer rate.
- The mechanism of drop break-up and the size of drops after pinch-off
- Validation of numerical simulations of drop pinch-off in forced convective flow.

Re-entrainment
Re-entrainment rates of up to 14% have been measured in the small scale heat exchanger. The effect of re-entrainment on heat and mass transfer has been quantified using a case study. At typical industrial conditions, a re-entrainment rate of 10% reduces the thermal efficiency of the next heat exchanger in a heat exchanger cascade by 7%. Furthermore it has been shown that a PVDF heat exchanger can be used to collect hydrogen fluoride from a polluted gas stream, but that 10% re-entrainment reduces the gas cleaning efficiency from 99.3 to 90%.

Break-off mechanism and drop size after break-up
Condensate re-entrainment from heat exchanger plates is caused by pinch-off of drops from the downstream edge. Drops grow due to coalescence until a large drop at the downstream edge is formed. The drop pinches off when its mass reaches some critical size. This size depends on orientation of the plate, shape of the downstream edge, vapour velocity, surface tension and density of vapour and liquid. Of these, plate orientation has the largest influence on drop shape, because the pinch-off mechanism from a vertical plate is different from the pinch-off mechanism from a horizontal plate.

When a drop pinches off a vertical plate in quiescent vapour, lengthwise stretching of the drop results in formation of a neck that breaks. When the surface tension is low, a drop pinches off that is smaller than the plate diameter, forming about 5 satellites. When the surface tension is relatively high, the drop volume is determined by the size of the plate. At the moment of pinch-off, the neck is much shorter, resulting in the formation of less, smaller satellites.

This break-up mechanism has also been found at relatively low vapour velocities. At higher vapour velocities, sideways swaying of an elongated drop results in pinch-off. Vapour shear forces are high, because the drop protrudes out of the recirculation zone behind the plate, where the vapour velocity is relatively small. Smaller drops are formed, while more and larger satellites are formed. As the vapour velocity increases, the main drop becomes smaller and satellites larger, until the main drop and the larger satellites are equally big.
As a drop pinches off a vertical plate with a flat edge, gravity pulls a drop down, into the vapour flowing under the plate. As a result, vapour shear forces on the drop are high, even when the vapour velocity is relatively low. Drops are stretched while the centre of mass oscillates vertically until the tip of the drop is torn off and the remainder (90%) of the fluid contracts.

Drops pinching off plates with a round or a sharp edge oscillate initially mainly perpendicular to the plate, without large lengthwise stretching. As a result, at the moment of break-up, the drop protrudes far into the gas flowing over the plate. This results in formation of a large amount of relatively small drops. A small amount of fluid (~10%) remains behind on the downstream edge.

Various forces acting on a drop during pinch-off have been calculated using shape and acceleration of the drop. Gravity plays an important role during pinch-off, although gravitational forces acting on the drop during break-up are small. The reason for this is that gravity pulls the drop down. When a plate is placed horizontally, gravity pulls a drop out of the recirculation zone behind a horizontal plate into the gas stream under the plate. When a plate is placed vertically, gravity does not draw a drop out of the recirculation zone. As a result, drops pinching off vertical plates are larger than drops pinching off horizontal plates.

Drops that are smaller than the thickness of the downstream edge of a vertical plate have been investigated using ethane. It is found that at low vapour velocities, drops pinch off from the centre of the plate, while at relatively high vapour velocities drops pinch off from the side of the downstream edge. At intermediate vapour velocities, both pinch-off mechanisms are observed. Drops pinching off from the centre of the downstream edge break-up forming a limited number of relatively droplets, while “edge break-up” produces relatively small drops.

Break-up of drops that are larger than the plate is studied using water and ethane. An empirical correlation to predict drop size after pinch-off from a vertical flat edged plate has been constructed, which is valid at a Bond number under 1.5, a Weber number under 60 and Reynolds under 6000. The drop size after pinch-off appears not to be determined by the Weber number, but instead correlates with $v^2d^2$. This is not well understood.

**Validation of numerical simulation**

Recorded pinch-off of an ethane drop at near-critical conditions in downward convective flow has been used to validate a numerical simulation using the diffuse interface model. Discrepancies between experiment and simulation show that the numerical method is –at this point- not capable of correctly simulating pinch-off of drops in flowing vapour.

It also shows that the experimental observation of drop pinch-off at near-critical conditions is suited for its primary use: to allow validation of numerical simulations of systems of changing
References


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   A  **11** pp355-365: fluoride compounds [HF]

   B  **11** pp428-435 : fluoride compounds [SF₆]

   C  **11** pp694-712: : fluoride compounds [PTFE and PVDF]


46. F. Weitz F.A. Ganzevles C.W.M. van der Geld A. Lexmond, *Enhancing heat transfer and gas cleaning efficiency of compact plastic heat exchangers by promoting drainage*, Banff


# Nomenclature

<table>
<thead>
<tr>
<th>symbol</th>
<th>definition</th>
<th>dimension</th>
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<tr>
<td>A</td>
<td>area (constant)</td>
<td>m²</td>
</tr>
<tr>
<td>b</td>
<td>radius of drop apex</td>
<td>m</td>
</tr>
<tr>
<td>B</td>
<td>shape factor</td>
<td>-</td>
</tr>
<tr>
<td>c</td>
<td>concentration (chapter 2,3)</td>
<td>mole/l</td>
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<tr>
<td>c</td>
<td>dimensionless concentration (chapter 4)</td>
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<tr>
<td>C_{diss}</td>
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</tr>
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<td>C_D</td>
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<td>d</td>
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<td>K_d</td>
<td>dissociation constant of water</td>
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</tr>
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<td>radius (constant)</td>
<td>m</td>
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<tr>
<td>R</td>
<td>universal gas constant (8.314)</td>
<td>J/(mole K)</td>
</tr>
<tr>
<td>S</td>
<td>area (variable)</td>
<td>m²</td>
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<tr>
<td>t</td>
<td>time</td>
<td>s</td>
</tr>
<tr>
<td>T</td>
<td>temperature</td>
<td>°C</td>
</tr>
<tr>
<td>v</td>
<td>cross sectional averaged velocity over the plate, def: see eq. 4.2.11</td>
<td>m/s</td>
</tr>
<tr>
<td>v</td>
<td>specific volume (section 4.4)</td>
<td>m³/kg</td>
</tr>
<tr>
<td>v</td>
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<td>m/s</td>
</tr>
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<td>Definition</td>
<td>Dimension</td>
</tr>
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</tr>
<tr>
<td>( V )</td>
<td>Volume</td>
<td>( m^3 )</td>
</tr>
<tr>
<td>( x, y, z )</td>
<td>co-ordinates</td>
<td>( m )</td>
</tr>
<tr>
<td>( \chi )</td>
<td>mass fraction</td>
<td>-</td>
</tr>
<tr>
<td>( a )</td>
<td>contact angle</td>
<td>-</td>
</tr>
<tr>
<td>( a_{dr} )</td>
<td>orifice discharge coefficient</td>
<td>-</td>
</tr>
<tr>
<td>( \beta )</td>
<td>cross sectional area ratio</td>
<td>-</td>
</tr>
<tr>
<td>( \Delta )</td>
<td>difference</td>
<td>-</td>
</tr>
<tr>
<td>( \epsilon )</td>
<td>expansion coefficient (chapter 2)</td>
<td>-</td>
</tr>
<tr>
<td>( \phi )</td>
<td>contact angle</td>
<td>( ^\circ )</td>
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<tr>
<td>( \kappa )</td>
<td>isentropic expansion coefficient (chapter 2)</td>
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<tr>
<td>( \kappa )</td>
<td>diffuse interface constant (chapter 4)</td>
<td>( J m^2/kg )</td>
</tr>
<tr>
<td>( \lambda )</td>
<td>heat transfer coefficient</td>
<td>( W/m^2 )</td>
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<tr>
<td>( \mu )</td>
<td>chemical potential</td>
<td>( Pa )</td>
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<tr>
<td>( \xi )</td>
<td>fraction dissociated ammonia</td>
<td>-</td>
</tr>
<tr>
<td>( \rho )</td>
<td>mass density</td>
<td>( kg/m^3 )</td>
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<tr>
<td>( \sigma )</td>
<td>surface tension coefficient</td>
<td>( N/m )</td>
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<tr>
<td>( \tau )</td>
<td>stress tensor</td>
<td>( N/m^2 )</td>
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<table>
<thead>
<tr>
<th>Subscript</th>
<th>Definition</th>
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<tbody>
<tr>
<td>c</td>
<td>critical point</td>
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<tr>
<td>c</td>
<td>condensate</td>
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<td>conductive</td>
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<td>f</td>
<td>friction</td>
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<td>foot</td>
<td>contact area between drop and plate</td>
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<td>g</td>
<td>gas</td>
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<td>H</td>
<td>hydrodynamic</td>
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<td>hydrogen fluoride</td>
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<tr>
<td>H(_2)O</td>
<td>water</td>
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<td>i</td>
<td>summation integer</td>
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<td>in</td>
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<td>l</td>
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<tr>
<td>m</td>
<td>centre of mass</td>
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<tr>
<td>NH(_3)</td>
<td>ammonia</td>
</tr>
<tr>
<td>out</td>
<td>out</td>
</tr>
</tbody>
</table>
plate | plate 
PVDF | poly vinylidene fluoride 

s | solid 
t | terminal falling velocity 
tot | total 
v | vapour 
vap | vaporisation 
v=0 | without forced convection 

<table>
<thead>
<tr>
<th>Dimensionless numbers</th>
<th>definition</th>
<th>name</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bo (Bo_{drop})</td>
<td>$\pm \frac{r_{eq}^2(\rho_l - \rho_v)g}{\sigma}$</td>
<td>Bond number</td>
</tr>
<tr>
<td>Bo_{n}</td>
<td>$\pm \frac{D^2_{\text{n}}(\rho_l - \rho_v)g}{\sigma}$</td>
<td>&quot; &quot;</td>
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<td>&quot; &quot;</td>
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<tr>
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<td>$\frac{\eta_v V_{\text{max}}}{\sigma}$</td>
<td>Capillary number</td>
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<tr>
<td>Ch</td>
<td>$\frac{d_{\text{interface}}}{D_{\text{numerical domain}}}$</td>
<td>Cahn number</td>
</tr>
<tr>
<td>Re_{drop}</td>
<td>$\frac{\rho_v V_{\text{drop}}}{\eta_v}$</td>
<td>Reynolds number</td>
</tr>
<tr>
<td>Re_{n}</td>
<td>$\frac{\rho_v V_{\text{max}} D_{\text{numerical domain}}}{\eta_v}$</td>
<td>&quot; &quot;</td>
</tr>
<tr>
<td>Re_{plate}</td>
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<td>&quot; &quot;</td>
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<tr>
<td>Re_{ip}</td>
<td>$\frac{\rho_v V_{\text{plate}}}{\eta_v}$</td>
<td>&quot; &quot;</td>
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<tr>
<td>We_{drop}</td>
<td>$\frac{\rho_v V_{\text{drop}}^2}{\sigma}$</td>
<td>Weber number</td>
</tr>
<tr>
<td>We_{n}</td>
<td>$\frac{\rho_v V_{\text{max}}^2 D_{\text{numerical domain}}}{\sigma}$</td>
<td>&quot; &quot;</td>
</tr>
<tr>
<td>We_{ip}</td>
<td>$\frac{\rho_v V_{\text{drop}}^2}{\sigma}$</td>
<td>&quot; &quot;</td>
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Appendix A: centre of mass of a drop behind the downstream edge of a plate

The goal of this appendix is to present how the centre of mass of a drop, pendant behind the downstream edge of a plate, has been determined. This is used in section 3.4 to calculate various characteristics of the drop, such as the volume averaged acceleration and the potential energy.

A graphical presentation of the shape of a such drop is given in fig. 3.4.10. The position of the foot (x_{foot}), tip (x_{tip}), width (b_0) and height (h_0) of the drop are measured.

The position of the centre of the foot is given by eq. 3.4.2:
\[ x_{foot} = \frac{1}{2} (x_{cl,1} + x_{cl,2}) \]  

For convenience, we put a (new) origin at the centre of the foot:
\[ \tilde{x}_{new} = \tilde{x}_{old} - \tilde{x}_{foot} \]

The subscript “new” will be dropped until needed again, at eq. A.12.

It is assumed that the drop is symmetrical with respect to the y=0-plane. The centre of mass of the drop is found by integration:
\[ \tilde{x}_m = \frac{\int \tilde{x} \rho(\tilde{x}) dV}{\int \rho(\tilde{x}) dV} \]

When we assume that the density of the drop is constant, we find eq. 3.4.6:
\( \bar{x}_m = \frac{1}{V} \int \bar{x} dV \)  \hspace{1cm} (3.4.6)

The volume of the drop is easily calculated:

\[
V = \int_V dV = \int_0^{x_{tip}} A(x) dx
\]

With \( A(x) \) the cross sectional area, parallel to the downstream edge, through the drop. \( b(x) \) and \( h(x) \) are the width and height of the drop at downstream distance \( x \) from the plate respectively. These are given by eq. A.4 and A.5:

\[
b(x) = b_0 \sqrt{1 - \left( \frac{x}{x_{tip}} \right)^2} \quad \text{A.4}
\]

\[
h(x) = h_0 \sqrt{1 - \left( \frac{x}{x_{tip}} \right)^2} \quad \text{A.5}
\]

Every cross section through the drop, that is parallel to the plate, is an ellips. The centre of the ellips is given by eq. A.6:

\[
(x, y, z) = (x, 0, x \frac{z_{tip}}{x_{tip}}) \quad \text{A.6}
\]

The \( y \)-coordinate is always zero, because the drop is (by its definition) symmetrical with respect to the \( y=0 \)-plane. The cross sectional area of the ellips is given by eq. A.7:

\[
A(x) = \frac{\pi}{4} bh \quad \text{A.7}
\]

Substitution of A1.4, A1.5 and A1.7 in A1.3 gives the volume of the drop:

\[
V = \int_V dV = \frac{\pi}{4} b_0 h_0 \int_0^{x_{tip}} \left( 1 - \left( \frac{x}{x_{tip}} \right)^2 \right) \left( 1 - \left( \frac{x}{x_{tip}} \right)^2 \right) dx = \\
V = \frac{\pi}{4} b_0 h_0 \int_0^{x_{tip}} \left( 1 - \left( \frac{x}{x_{tip}} \right)^2 \right) dx = \frac{\pi}{4} b_0 h_0 x_{tip} - \frac{\pi}{4} \frac{b_0 h_0}{x_{tip}^2} \int_0^{x_{tip}} x^2 dx =
\]
Appendix A

\[ V = \frac{\pi}{4} b_0 h_0 x_{tip} - \frac{\pi}{4} b_0 h_0 x_{tip}^3 \frac{3}{3} = \frac{\pi}{6} b_0 h_0 x_{tip} \quad \text{A.8} \]

The x-co-ordinate of the centre of mass is found by solving:

\[ x_m = \frac{1}{V} \int y dV = \frac{\pi}{4} b_0 h_0 \int_0^{x_{tip}} x \left( 1 - \left( \frac{x}{x_{tip}} \right)^2 \right) dx \quad \text{A.9} \]

\[ x_m \frac{\pi}{4V} b_0 h_0 \frac{1}{2} x_{tip}^2 - \frac{\pi}{4V} b_0 h_0 \frac{1}{4} x_{tip}^4 = \frac{\pi}{16} b_0 h_0 x_{tip}^2 = \frac{3}{8} x_{tip} \quad \text{A.10} \]

An an identical manner we can solve for \( z_m \) and find:

\[ z_m = \frac{3}{8} x_{tip} \quad \text{A.11} \]

Combining A1.9, A1.10 and the fact that \( y_m = 0 \), we find:

\[ \tilde{x}_m = (x_m, y_m, z_m) = \left( \frac{3}{8} x_{tip}, 0, \frac{3}{8} z_{tip} \right) = \frac{3}{8} \tilde{x}_{tip} \quad \text{A.12} \]

Where we remember that this is the position of the tip with respect to the "new" origin, which is the centre of the foot of the drop. Re-applying A.1 gives the centre of mass with respect to an arbitrary origin:

\[ \tilde{x}_{m, old} = \tilde{x}_{m, new} - \tilde{x}_{m, foot} = \frac{3}{8} (\tilde{x}_{tip} - \tilde{x}_{foot}) - \tilde{x}_{foot} \quad \text{A.13} \]

\[ \tilde{x}_m = \frac{5}{8} \tilde{x}_{foot} + \frac{3}{8} \tilde{x}_{tip} \quad \text{3.4.7} \]
Appendix B: A method to calculate surface tension and gravitational force acting on a drop

In this appendix it will be shown how the gravitational and surface tension component of the forces acting on a drop are calculated. A graphical presentation of a drop behind the downstream edge of a plate is given in fig. 3.4.10, see e.g. Appendix A

The equation of motion, applied to a drop, has been specified in chapter 3, eq. 3.2.51:

\[ \rho_l V \frac{\partial \mathbf{v}_m}{\partial t} = \left( \frac{m}{R} \right) \frac{2 \sigma}{A_{\text{foot}}} + \bar{F}_\sigma + \left( \rho_l - \rho_v \right) \left[ \bar{g} - m \left( \bar{m} \cdot \bar{g} \right) \right] V \]

First, the gravitational force will be calculated. The correction term for the pressure at the foot of the drop will be incorporated in the calculation:

\[ \bar{F}_g = \left( \rho_l - \rho_v \right) \left[ \bar{g} - m \left( \bar{m} \cdot \bar{g} \right) \right] V \]

Where \( m \) is the unit vector normal to the wall. When the downstream edge of a plate is flat, the unit normal \( m \) is (in these experiments) always perpendicular to the gravity vector \( \bar{g} \); the inner product \( m \cdot \bar{g} \) is therefore zero. When the downstream edge has another shape, i.e. is round or sharp, the inner product \( m \cdot \bar{g} \) can be nonzero, and has to be calculated.

Size and direction of the foot of the drop are defined by the upper and lowermost points, \( \bar{x}_{cl,1} \) and \( \bar{x}_{cl,2} \). \( \bar{m} \) is perpendicular to the line \( \bar{k} \) through these points:

\[ \bar{k} = \frac{\bar{x}_{cl,2} - \bar{x}_{cl,1}}{||\bar{x}_{cl,2} - \bar{x}_{cl,1}||} \]

Therefore, \( \bar{m} \) satisfies:

\( (m_1, 0, m_3) = (-k_3, 0, k_1) \)
Substitution in A2.1 gives:
\[
(F_{x,1}, F_{x,2}, F_{x,3}) = \left( \rho_f - \rho_v, \psi \left( k, k_3, 0, (1 - k_1^2) \right) \right)
\]  
B.4

The surface tension related forces are lumped in the second term of eq. 3.2.51:
\[
\vec{F}_{\alpha} = \bar{m} \frac{2\sigma}{R} A_{\text{foot}} + \vec{F}_{\alpha}'
\]  
B.5

To calculate the first part, \(\bar{m}\), the average drop radius, the cross-sectional area of the foot of the drop, \(A_{\text{foot}}\), and the average drop radius, \(R\), have to be calculated. \(\bar{m}\) is calculated using B.3. \(A_{\text{foot}}\) is calculated using equation A.7:
\[
A_{\text{foot}} = A(x = 0) = \frac{\pi}{4} b_0 h_0
\]  
B.6

The average drop radius is calculated by averaging the radius of curvature at every point on ellipses in the \(x=0, y=0\) and \(z=0\)-plane. This works out as:
\[
\frac{1}{R} \approx \frac{1}{5} \left( \frac{2}{b_0} + \frac{2}{h_0} + l \right)
\]  
B.7

This force “pushes” the drop away from the plate, because \(\bar{m}\) is directed in that direction and all other scalars are positive.

The second component of the force due to surface tension effects, \(\vec{F}_{\alpha}'\), is found by multiplying the surface tension coefficient of the liquid (actually, the gas-liquid interface) with the length of the foot of the drop, \(O\). This force “pulls” the drop towards the plate. The direction in which this force works equals the direction in which the drop protrudes, see paragraph 3.2.5:
\[
\vec{F}_{\alpha}' = -\sigma \left( \frac{\vec{x}_{\text{tip}} - \vec{x}_{\text{foot}}}{\vec{x}_{\text{tip}} - \vec{x}_{\text{foot}}} \right) O
\]  
B.8

The circumference of the foot is approximated using:
\[
O \approx \pi \sqrt{\frac{h_0^2 + b_0^2}{2}}
\]  
B.9

Substitution of A2.6-A2.9 in A2.5 gives the surface tension force.
Summary

Compact PVDF heat exchangers are used to recover heat from highly polluted gas streams. Quite often, water vapour condenses in the heat exchanger, forming drops on the cold plates. Pinch-off of these drops from the downstream edge causes condensate to be re-entrained into the gas stream, possibly damaging downstream equipment and reducing thermal efficiency.

The goal of this study is to increase the knowledge of the causes and consequences of drop pinch-off, as occurring in hydrophobic polymer heat exchangers. To this end, three aspects have been investigated.

Firstly, re-entrainment, which is caused by drop pinch-off, has been studied in an small scale heat exchanger to quantify the effect of drop pinch-off on the overall heat and mass transfer rate. For this heat exchanger re-entrainment is found to depend on the total condensate flux and the Weber number of the gas flowing around the drop. A case study is used to quantify the effect of re-entrainment on heat and mass transfer. At typical industrial conditions, a re-entrainment rate of 10% reduces the thermal efficiency of the next heat exchanger in a heat exchanger cascade by 7%. It is also shown that a PVDF heat exchanger can be used to collect hydrogen fluoride from a polluted gas stream, but that 10% re-entrainment reduces the gas cleaning efficiency from 99.3 to 90%.

Secondly, experiments are performed in two single plate mock-ups to find by which mechanism drops break off a heat exchanger plate. Some experiments are performed using ethane at near-critical conditions. Various variables, i.e. temperature, pressure, liquid and vapour density, the shape of the downstream edge, plate orientation, vapour velocity and surface tension coefficient have been varied to observe the effect on drop pinch-off.

During pinch-off from a vertical plate at low to moderate Weber numbers, a drop is stretched until the neck breaks. Drop size and the number of satellites formed depend mainly on Bond and Weber numbers, although no correlation has been found describing all measurements.

During pinch-off from a vertical plate at relatively high Weber number, or pinch off from a horizontal plate, the drop is stretched and oscillates perpendicular to the plate. The amplitude of the oscillation increases, until break-up follows. This break-up mechanism results in relatively small drops after pinch-off.

The shape of water drops during break-up has been used to calculate various forces that act on a drop pinching off a horizontal plate. The magnitude of the surface tension force and hydrodynamic force are much bigger than the magnitude of gravitational forces acting on the drop. However, plate orientation (or the direction of the gravitational force) has a large influence on the process of drop break-up. Changing a plate from vertical to horizontal has, at one condition,
resulted in a decrease in drop volume after pinch-off by a factor 40. This large difference is caused by the difference in break-up mechanism (stretching versus oscillating) between the two situations.

Thirdly, several experiments have been performed at well-defined conditions, which allow validation of numerical simulations of drop pinch-off in forced convective flow. Drop break-up has been simulated using a diffuse interface model. Comparison between numerical simulation and physical experiment has shown that at this moment, the simulation does not correctly describe drop pinch-off. Reasons for this are thought to be the use of a 2D grid and the large grid size. Most importantly, it shows that the measurements can be used for validation of multiphase flow simulations of changing topology without density matching at relatively high vapour velocities.
Samenvatting

Compacte warmtewisselaars worden gebruikt om warmte terug te winnen uit sterk vervuilde industriële gasstromen. Water uit de gasstroom condenseert in de warmtewisselaar en vormt druppels op de koude platen. Het afbreken van deze druppels van de warmtewisselaar platen resulteert in de terugvoer van condensaat aan het gas, waardoor andere apparaten kunnen beschadigen en het thermisch redement van de installatie afneemt.

Het doel van deze studie is de kennis te vergroten omtrent de oorzaken en gevolgen van het afbreken van druppels, zoals dat in hydrofobe warmtewisselaars plaatsvindt. Om dit te bereiken zijn drie aspecten van het afbreken van druppels onderzocht.

Ten eerste is de terugvoer van condensaat aan het gas door het afbreken van druppels onderzocht. Met metingen in een kleinschalige warmtewisselaar is het effect hiervan op de thermische en stofoverdrachts efficiency gemeten. In deze opstelling is gebleken dat de fractie condensaat die met de gasstroom de warmtewisselaar verlaat afhankelijk is van zowel het totale condensaatdebiet als van het Weber getal. Met een case study is de invloed van dit fenomeen op de warmte- en stofoverdracht bij een industriële toepassing afgeschat. Als, bij verder standaard industriële condities, 10% van het condensaat als druppels in de gasstroom de warmtewisselaar verlaat, verlaagt dit de hoeveelheid overgedragen warmte in de volgende warmtewisselaar met naar schatting 7%.

Hiernaast is aangetoond dat een compacte warmtewisselaar, die als condenser ingezet wordt, geschikt is wateroplosbare vervuiling uit de gasstroom te verwijderen. Bij de uitgevoerde case study bleek de reinigingsefficiency door terugvoer van 10% condensaat aan het gas, van 99,3% naar 90% af te nemen.

Ten tweede is het afbreken van druppels van een plaat in twee speciaal hiervoor gebouwde opstellingen onderzocht. Een aantal van deze experimenten is uitgevoerd met ethaan nabij het thermodynamisch kritisch punt. Verschillende grootheden - temperatuur, druk, dichtheid van vloeistof en damp, de vorm van de rand van de plaat, plaatoriëntatie, gassnelheid en oppervlaktespanningscoëfficiënt - zijn gevarieerd om de invloed hiervan op het opbreken van druppels te onderzoeken.

Bij relatief lage gassnelheden breekt een druppel van een verticale plaat doordat de druppel oprek en insnoert. De grootte van de druppel en de grootte en het aantal satellieten zijn sterk afhankelijk van het Bond en het Weber getal. Desondanks is het niet gelukt een correlatie te vinden die alle metingen goed beschrijft.
Als een druppel bij relatief hoge gassnelheden van een verticale plaat of van een horizontale plaat breekt, rekt de druppel op en oscilleert parallel aan de plaat. De amplitude van de oscillatie neemt toe totdat de druppel breekt. Dit opbreekmechanisme resulteert in relatief kleine druppels.

De vorm van druppels tijdens het opbreken van een horizontale plaat is onderzocht om de verschillende krachten die op een druppel werken te kunnen berekenen. Wat grootte betreft is de zwaartekracht vrijwel verwaarloosbaar vergeleken met de oppervlaktespanningskrachten of hydrodynamische krachten. Toch heeft de oriëntatie van de plaat (ofwel de richting van de zwaartekracht) een grote invloed op het opbreekproces. Bij één conditie is bekeken wat het effect van die oriëntatie op de grootte van een druppel na opbreken is. Druppels die afbraken van een verticale plaat hadden een volume dat 40 keer zo groot is als het volume van druppels die van een horizontale plaat afbraken. Dit verschil wordt veroorzaakt door de verschillen in opbreekmechanisme: insnoren versus oscilleren.

Ten derde zijn enkele experimenten uitgevoerd om numerieke simulaties van topologieveranderingen in stromende media te kunnen valideren. Het opbreken van druppels is gesimuleerd met behulp van een diffuus-grenslaagmodel. De vergelijking van de numerieke simulatie met het experiment toont aan, dat de simulatie op dit moment nog niet geschikt is voor het doorrekenen van het afbreken van druppels. De reden hiervoor is waarschijnlijk het gebruik van een 2D-grid of de te grof grid.

Belangrijker is echter, dat de vergelijking heeft laten zien dat de metingen geschikt zijn voor validatie van stromingsberekeningen met topologieveranderingen waarbij zwaartekracht noch schuifkrachten verwaarloosd kunnen worden.
Nawoord

Het begint er nu toch echt naar uit te zien dat “mijn kindje”, waar 5 jaar in geïnvesteerd is, binnenkort het licht zal zien.

Dit proefschrift beschrijft het resultaat van jaren van meten in de hal van de warme werktuigbouw in W-laag. De eerste metingen zijn uitgevoerd in een opstelling die er al van vóór mijn tijd stond, maar al snel is begonnen met de constructie van een aantal kleine schaalmodellen. Het eerste schaalmodel, geconstrueerd van een oude ventilator, wat resten multiplex en een stuk perspex, was na 2 dagen gebruiksklaar. Aan de tweede opstelling heb ik samen met Lambert een maand gebouwd. De constructie van de derde en laatste opstelling heeft meer dan een jaar gekost. Zoals het zo vaak gaat bij experimenteel onderzoek, is het ook nu weer verlopen. Na jaren bouwen volden een paar weken meten, waarna het vervolgens weer jaren gekost heeft om alle resultaten te verwerken.

Zonder de hulp van velen was dit onderzoek nooit tot stand gekomen.

In de eerste plaats wil ik Bert bedanken voor de grote vrijheid die ik gekregen heb, en Cees voor zijn tomeloze enthousiasme en geduld bij het redigeren van de aangeleverde stukken.

Daarnaast wil ik de studenten Frank en Jonathan bedanken voor hun vele inspanningen en mooie resultaten bij metingen aan de marangoni opstelling en Vinny voor de samenwerking die tot sectie 4.4 geleid heeft.


Van alle collega-AIO’s uit de warme werktuigbouw wil ik vooral mijn kamergenoot Paulos bedanken. Naast zijn “schaklessen” en de gezelligheid hebben de vele ongeplande brainstormsessies schijnbaar onmogelijke constructieproblemen helpen oplossen en het onderzoek naar een hoger plan getild.

Tenslotte wil ik Carlijn bedanken voor de vele uren die ze gestoken heeft in dit proefschrift en het geduld dat ze heeft moeten opbrengen, vooral in de afrondingsfase.

Axel