Phase inversion in a dispersed oil-water and oil-water-gas flow
Phase inversion in a dispersed oil-water and oil-water-gas flow

PROEFSCHRIFT

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Summary

Phase inversion in a dispersed oil-water and oil-water-gas flow - K. Piela

Oil is the most used energy source on our planet. It is present in subsurface reservoirs. Very often such reservoirs contain not only oil; but oil, water and gas. So when oil is produced, also water and gas can be present in the production pipes. To predict the production rate it is essential to know the properties of such oil-water and oil-water-gas flows and to be able to model them. In our research we have restricted ourselves to dispersed two-phase and three-phase flows with a continuous liquid (oil or water), in which drops of the other liquid (and gas bubbles) are present. Dispersed flow occurs only at high mixture velocities. Special attention was given to the phenomenon of phase inversion, whereby the mixture switches from an oil-continuous phase with water drops (and gas bubbles) to a water-continuous phase with oil drops (and gas bubbles), or vice versa. During phase inversion the pressure gradient of the flow in the pipe increases strongly. So it is important to know at what conditions phase inversion occurs, in order to find out whether in practice it can be avoided or its effect on the pressure drop minimized. To that purpose we investigated first phase inversion in an oil-water flow through a horizontal pipe. Thereafter the influence of gas bubbles on phase inversion was studied.

Continuous, direct and discontinuous experiments were carried out. A continuous experiment was started by flowing one of the liquids at a certain velocity through the pipe loop and then gradually increasing the concentration of the other liquid by injecting it (in the form of drops) at a certain flow rate into the continuous liquid. In the case of a three-phase flow injection of liquid was stopped (when the desired dispersed phase volume fraction was reached) and injection of gas was started. It was found that for continuous experiments the critical concentration (at which phase inversion occurs) is very dependent on the injected phase volume fraction (the ratio of the volume rate of injected liquid and the volume rate of the mixture in the pipe). At low values of the injected phase volume fraction phase inversion can be postponed to a very high dispersed phase volume fraction in the pipe (even as high as 90%). During inversion the concentration of drops of the (originally) dispersed phase becomes so high, that they coalesce at certain places in the flow field and form relatively large, rather complex, morphological structures. With a further increase in concentration of the (originally) dispersed phase these morphological structures grow in size and start to form the new continuous phase in which again complex structures are present, but this time consisting of the (originally) continuous phase. It was found, that gas bubbles (that are always present in the continuous liquid phase) do not have a significant effect on the critical concentration and pressure gradient at phase inversion. However, phase inversion has a strong
influence on the bubbles. The dispersed drop concentration at phase inversion is high (much higher than 0.5). Therefore, just before inversion a dispersed bubble flow is not possible. Large bubbles are present and the flow pattern is elongated bubble flow. During the inversion process the bubbles break-up quickly and as the dispersed drop concentration is then much lower than before inversion a dispersed bubble flow is generated.

The direct experiments started with pumping oil and water from separate tanks into the pipe at a constant concentration. Both liquids mixed and flowed downstream. At an oil volume fraction of $0.5 - 0.6$ a strong increase in pressure drops was observed and phase inversion occurred during subsequent experiments with increasing dispersed liquid volume fraction. Visual observation showed similar morphological structures as observed during the continuous experiments. Again the effect of gas bubbles on phase inversion was negligible (at low gas concentration).

Also discontinuous oil-water experiments were performed. During a discontinuous experiment a pure water phase was pumped through the pipe, and at a certain moment it was changed to a pure oil phase (or vice versa). Somewhere in the mixing zone between the two liquids phase inversion occurred, similarly to the direct experiments. The simple discontinuous experiments have given us much insight in the mixing and phase inversion process. The distinction between the different stages of the processes was easy. The observations made in the mixing zone of discontinuous experiments confirmed the existence of (multiple) drops, pockets and regions as also found during continuous and direct experiments. During the passage of the mixing zone the local friction factor increased very considerably.

The experiments showed that a so-called ambivalence region exists, in which both liquids may be found to be continuous. Only outside this region one liquid is always continuous and the other is always discrete. Within the ambivalence region either one of the two liquids can be continuous and the exact value at which phases invert depends on the operating conditions. Two different theoretical models were developed to predict the ambivalence region in case of an oil-water flow (without gas). One model is based on a mean-field theoretical description. The values of two parameters in the model are determined by fitting predictions to experimental data. This model can then successfully be applied to a range of conditions with different values of the injected phase volume fraction of the dispersed phase during continuous experiments. The other model is based on the assumption of minimum dissipation rate. It gives also encouraging results.
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Phase inversion in a dispersed oil-water and oil-water-gas flow - K.Piela

Olie is de meest gebruikte bron van energie op onze planeet. Het is aanwezig in ondergrondse reservoirs. Dikwijls bevatten deze reservoirs niet alleen olie; maar olie, water en gas. Om de productie snelheid te voorspellen is het essentieel om de eigenschappen van zulke olie-water en olie-water-gas stromingen door productiepijpen te kennen en in staat te zijn ze te modelleren. In ons onderzoek hebben we ons beperkt tot gedispergeerde twee-fasen en drie-fasen pijpstromingen met een continue vloeistof (olie of water) waarin druppels van de andere vloeistof (en gas bellen) aanwezig zijn. Een gedispergeerde stroming treedt alleen bij hoge mengselsnelheden op. Tijdens ons onderzoek werd speciale aandacht geschonken aan het verschijnsel van de fase inversie, waarbij het mengsel omschakelt van een olie-continue fase met water druppels (en gas bellen) naar een water-continue fase met olie druppels (en gas bellen), of omgekeerd. Gedurende fase inversie neemt de drukgradient van de mengselstroming in de pijp sterk toe. Het is belangrijk om te weten bij welke condities fase inversie optreedt, om na te gaan of fase inversie in de praktijk voorkomen kan worden of de invloed ervan op de drukgradient verkleind kan worden. Met dat doel voor ogen hebben we eerst fase inversie in een olie-water stroming door een horizontale pijp bestudeerd. Daarna is de invloed van gas bellen op fase inversie onderzocht.

Continue, directe en discontinue experimenten zijn uitgevoerd. Een continu experiment werd gestart door één van de vloeistoffen met een zekere snelheid door de pijp te laten stromen en dan geleidelijk de concentratie van de andere vloeistof te laten toenemen door die vloeistof (in de vorm van kleine druppels) met een bepaalde snelheid in de continue vloeistof te injecteren. In het geval van een drie-fasen stroming werd de injectie van vloeistof gestopt (wanneer de vereiste volume fractie van de gedispergeerde vloeistof was bereikt) en de injectie van gas (in de vorm van bellen) gestart. Voor de continue experimenten werd gevonden, dat de kritische concentratie waarbij fase inversie optreedt zeer afhankelijk is van de volume fractie van de geïnjecteerde vloeistof (de verhouding van de volume snelheid van de geïnjecteerde fase en de volume snelheid van het mengsel in de pijp). Voor lage waarden van de volume fractie van de geïnjecteerde vloeistof kan fase inversie tot zeer hoge waarden van de volume fractie van de gedispergeerde vloeistof in de pijp uitgesteld worden (zelfs tot boven de 90%). Gedurende fase inversie wordt de concentratie van druppels van de (oorspronkelijke) gedispergeerde vloeistof zo hoog, dat de druppels op bepaalde plaatsen in hetstromingsveld coalesceren en relatief grote, tamelijk complexe, morfologische vloeistofstructuren vormen. Met een verdere toename van de (oorspronkelijke) gedispergeerde vloeistof groeien deze morfologische structuren en beginnen de nieuwe continue
Samenvatting

fase te vormen waarin weer complexe structuren aanwezig zijn, maar dit keer bestaande uit de (oorspronkelijke) continue vloeistof fase. We vonden dat gas bellen (die altijd alleen in de continue vloeistof fase voorkomen) geen aanzienlijke invloed hebben op de kritische concentratie en druk gradient bij fase inversie. Fase inversie heeft echter wel een grote invloed op de bellen. De concentratie aan druppels bij fase inversie is hoog (veel hoger dan 0.5). En dat is de reden, dat vlak voor fase inversie een gedispergeerde bellen stroming niet mogelijk is. Er zijn dan tamelijk grote bellen en het stromingspatroon is de z.g. "elongated bubble flow". Gedurende het inversie proces breken de bellen snel op in veel kleinere bellen en (mede omdat na inversie de concentratie aan vloeistof druppels veel lager is dan voor inversie) is daarom na inversie een gedispergeerde bellen stroming aanwezig.

De directe experimenten werd gestart met het pompen van olie en water (met constante concentratie) vanuit afzonderlijke voorraadvaten in de pijp. Beide vloeistoffen mengden in het beginstuk van de pijp en stroomden gemengd stroomafwaarts. Bij een olie volume fractie van 0.5 – 0.6 werd een sterke toename van de drukval geconstateerd en fase inversie trad op gedurende achtereenvolgende experimenten met een toenemende volume fractie van de gedispergeerde vloeistof fase. Visuele waarnemingen lieten soortgelijke morfologische structuren zien zoals waargenomen tijdens de continue experimenten. Bij lage gas concentratie was de invloed van bellen op fase inversie weer verwaarloosbaar.

Ook werden discontinue olie-water experimenten uitgevoerd. Gedurende zo’n experiment werd een pure water fase door de pijp gepompt, en op op een bepaald moment werd die vervangen door een pure olie fase (of omgekeerd). Ergens in de mengzone van de twee vloeistoffen trad fase inversie op, ongeveer zoals bij directe experimenten. Deze relatief eenvoudige discontinue experimenten hebben ons veel inzicht verschaft in het mengproces en in fase inversie. Het onderscheid tussen de verschillende stadia van het inversie proces was goed waar te nemen. De waarnemingen die in de mengzone gemaakt zijn, bevestigden het bestaan van (meervoudige) druppels en grotere vloeistofstructuren zoals die ook gedurende de continue en directe experimenten gevonden werden. Gedurende de passage van de mengzone nam de wrijvingsfactor zeer aanzienlijk toe.

De experimenten lieten ook een z.g. hysteresis gebied zien, waarin beide vloeistoffen de continue fase kunnen zijn. Alleen buiten dit gebied is één van de vloeistoffen altijd continu en de andere vloeistof gedispergeerd. Binnen het hysteresis gebied kan elk van de twee vloeistoffen de continue fase zijn en de precieze conditie waar fase inversie optreedt hangt van de operationele omstandigheden af. Twee verschillende theoretische modellen zijn ontwikkeld om het hysteresis gebied voor het geval van een olie-water pijp stroming (zonder gas) te voorspellen. Eén model is gebaseerd op een "mean-field" theoretische beschrijving. De waarden van twee parameters in het model werden gevonden door theoretische voorspellingen aan te passen aan experimentele resultaten. Het model kan dan daarna succesvol gebruikt worden voor voorspellingen van experimentele resultaten bij andere omstandigheden. Het andere theoretische model is gebaseerd op de veronderstelling van minimale dissipatie. Het geeft ook bemoedigende resultaten.
Chapter 1

Introduction

1.1 Motivation

The use of energy from fire was an important development for improving human live. For a long time wood was the only source of energy. About two hundred years ago wood was replaced by coal and during the last century coal was replaced more and more by oil (37% of the world energy is produced from oil *). Oil has become one of the most important sources of energy, because of its high flexibility in many applications. To answer the high demand for oil new exploration and production techniques are needed.

Very often subsurface reservoirs contain not only oil; but oil, water and/or gas. So when oil is produced, also water and/or gas can be present in the production pipes. To predict the production rate it is essential to know the properties of such oil-water and oil-water-gas flows and to be able to model them. The production pipes in reservoirs are nowadays often horizontal. So in our study we have concentrated on oil-water and oil-water-gas flows in horizontal pipes. This study is not only relevant for horizontal production pipes in subsurface reservoirs. Often pipelines that connect offshore well heads with production platforms rest for a considerable length on the bottom of the sea. In such cases a two-phase or three-phase mixture is transported through a horizontal (or inclined pipeline) for many kilometers.

1.2 Flow pattern map

Much information exists on the flow pattern map for oil-water pipe flows. This flow pattern depends on the superficial velocities of oil and water. At low velocities the flow is stratified with a water layer underneath an oil layer. When the velocities are increasing a mixing between the two layers starts and a new dispersed layer between the oil layer and water layer is created. When the velocities become high enough the flow becomes fully dispersed. In that case one of the liquids is dispersed in the form of drops in the other liquid. When the water volume fraction is high, the flow is water continuous. At high oil volume fractions the flow is oil continuous.

*http://www.wikipedia.org/wiki/World_energy_resources_and_consumption
Two-phase water-gas flows have a different flow pattern map than oil-water flows. At low superficial velocities, similar to the oil-water case, the flow is stratified with a water layer underneath a gas layer. When the liquid velocity increases, the gas layer is broken-up by instabilities at the interface and a transition to elongated bubble/slug flow occurs. At high liquid velocity a dispersed bubble flow is created. However when the gas velocity increases, a transition to annular flow occurs. In annular flow the gas is traveling in the center of the pipe and the pipe walls are wetted by a thin liquid film. Dispersed bubble flow is only possible at a high liquid velocity and low gas fraction.

There have been many studies on oil-water and water-gas flows, but oil-water-gas flows did attract less attention because of the complicated nature of this flow. We have restricted ourselves to dispersed three-phase flow in a horizontal pipe with a continuous liquid (oil or water), in which drops of the other liquid and gas bubbles are present. Such a dispersed flow occurs only at sufficiently high values of the mixture velocity.

### 1.3 Phase inversion

At high mixture velocities an oil-water flow through a pipe is dispersed. So one of the liquids is the continuous phase and the other liquid is present in the form of drops. When the flow condition is changing (for instance the dispersed phase fraction is increasing) phase inversion can occur. During phase inversion the dispersed phase becomes the continuous one, and vice versa. During this process the effective viscosity of the mixture becomes very large, which leads to a high pressure drop over the pipe or to a low flow rate.

The phase inversion phenomenon has been studied for many years (see, for instance, Becher (2001)), but there are still many questions about the inversion process. Some authors assume that a sudden coalescence of drops can encapsulate parts of the continuous phase, which causes drop formation from the (originally) continuous phase (Yeo et al. (2002a); Brauner and Ullmann (2002)). Others observed the creation of multiple drops (small droplets of the continuous phase inside the drops) prior to inversion (Pacek et al. (1994); Pacek and Nienow (1995); Pal (1993); Sajjadi et al. (2000, 2002, 2003); Liu et al. (2005, 2006)). According to some authors inversion is a rather rapid process, called catastrophic by them, (Dickinson (1981); Smith and Lim (1990); Rondón-Gonzaléz et al. (2006); Tyrode et al. (2005); Vaessen et al. (1996); Binks and Lumsdon (2000)). However, during our experiments we found that this is not strictly true as also shown by Liu et al. (2006). Experimental results (with pictures) derived during phase inversion are described in chapter 2, 3 and 5.

### 1.4 Parameters influencing phase inversion

There are many parameters that influence phase inversion. Physical parameters (density, viscosity) determine the flow field and the mixing between the two phases. Also the chemical properties (for instance the presence of a surfactant) can have strong influence. Surfactants are surface active agents which even at a very low concentration can change the behavior of the
1.5 Modeling of phase inversion

Although phase inversion has been a problem for more than forty years, little is known about it and, for the design of industrial equipment, engineers have relied mainly on empirical correlations. For instance, to compute the critical water hold-up \( \varepsilon^I_w \) at which phase inversion takes place in oil-water flow, Arirachakaran et al. (1989) proposed, as a best fit of their experimental data, the following relation

\[
\varepsilon^I_w = 0.5 - 0.1088 \log_{10} \left( \frac{\mu_o}{\mu_r} \right),
\]

where \( \mu_r = 1 \text{ mPa}\cdot\text{s} \). This equation allows to estimate the critical water fraction \( \varepsilon^I_w \), at which phase inversion occurs as a function of the oil viscosity \( \mu_o \). Another empirical correlation to compute the critical water hold-up \( \varepsilon^I_w \) was suggested by Nädler and Mewes (1997)

\[
\varepsilon^I_w = \frac{1}{1 + k_1 \left( \frac{C_o}{C_w} \left( \frac{\rho_o}{\rho_w} \left( \frac{\mu_o}{\mu_w} \right) \left( DU_m \right) \right) ^{n_o-n_w} \right) ^{1/k_2}},
\]

where \( D \) is the pipe diameter, \( \rho_o, \rho_w \) is the oil or water density, \( \mu_o, \mu_w \) the oil or water viscosity, \( U_m \) the mixture velocity; \( C_o, C_w, n_o, \) and \( n_w \) are the parameters of the friction factor correlation \( CRe^{-n} \) for pure oil and pure water pipe flow; \( k_1 \) and \( k_2 \) are empirical parameters. All the details on the
application of Blasius-like relations to two-phase flows can be found in Brauner and Ullmann (2002).

Both empirical relations provide a single value of the hold-up at which inversion takes place once the fluids’ properties are known. However, it is found experimentally that an ambivalent range of hold-up values exists, in which both phases may be found to be continuous. Only outside this range one phase is always continuous and the other is always discrete. Within the ambivalent range, either one of the two phases can be continuous and the exact value at which phases invert depends on the operating conditions, on surface tension, and on pipe wettability. If the prediction of ambivalent range is neglected, the critical hold-up depends mainly on the viscosity ratio of the two fluids. Even if empirical correlations are widely used, some attempts to predict theoretically the critical hold-up for phase inversion have been carried out. Several mechanisms have been postulated to predict both phase inversion and the existence of the ambivalent range. The most important are based on

- instability between droplets coalescence and break-up;
- minimization of the total free energy content (including both gravitational potential energy and interfacial energy);
- effects of dynamic forces.

Phase inversion was explained as an instability between break-up and coalescence of dispersed drops. Phase inversion takes place when there is a large coalescence frequency and the break-up mechanism is insufficiently fast. Up to a certain volume fraction, break-up and coalescence processes can reach a dynamical balance; near the inversion point, instead, this balance cannot be sustained anymore and the coalescence rate overcomes the break-up rate. Droplets grow and their shapes change from spherical to cylindrical to lamellae and, eventually, a complex structure is reached, leading to the formation of the new continuous phase.

Since phase inversion is a phenomenon that occurs spontaneously, other authors proposed that its prediction can be based on a criterion of minimization of the total energy content, including interfacial energy, of the flowing system. Such an approach was originally proposed by Luhning and Sawistowski (1971) and Tidhar et al. (1986). Recently, following this line, Brauner and Ullmann (2002) proposed a new model to predict phase inversion. According to this model, the critical oil hold-up at which phase inversion occurs can be computed as

\[
\varepsilon_o' = \frac{\left[\frac{\sigma}{d_{32}}\right]_{w/o} + \frac{s}{d_{32}} \cos \theta}{\left[\frac{\sigma}{d_{32}}\right]_{w/o} + \left[\frac{\sigma}{d_{32}}\right]_{o/w}}
\]

where \( \theta \) is the water wettability angle, \( \sigma \) the oil-water interfacial tension, \( d_{32} \) the Sauter mean diameter, and \( s \) the wetted perimeter. The prediction of the Sauter diameter \( d_{32} \) involves extensive modelling for the break-up/coalescence of droplets. Because in general the application of the minimum free energy approach relies on a model able to characterize the drop size, several attempts have been made in the literature to predict the typical drop size, for example by means of a population balance equation – Hu and Angeli (2006) – or a Monte Carlo simulation, Yeo et al. (2002b).
The minimum energy criterion per se cannot predict the presence of the ambivalent range, nevertheless Yeo et al. (2000) were able to predict it by calculating the drop size using different relations in o/w and w/o dispersions. On the contrary, Brauner and Ullmann (2002) claimed that a possible reason for the hysteresis (ambivalent) range is the time needed after phase inversion for the new continuous phase to completely wet the pipe wall. The understanding and the prediction of the ambivalent range require further investigations, both experimental and theoretical.

Yeh et al. (1964) suggested another mechanism by which dispersed flow can be regarded as the flow of three thin layers: the dispersed phase, the interfacial phase, and the continuous one. Dynamic forces are considered to play a major role in the determination of the dispersion type. Assuming no shear at the interface, they derived the concentration ratio of dispersed to continuous phase at inversion as a function of pure liquid viscosities. Assuming all the layers in the model are laminar, the relation obtained by Yeh et al. can be written as

$$
\varepsilon'_w = \frac{1}{1 + \left(\frac{\mu_o}{\mu_w}\right)^{0.5}}.
$$

All experiments (described in the literature) carried out to study phase inversion in a pipe flow were made by means of direct experiments, i.e. oil and water were injected simultaneously into the pipe at prescribed superficial velocities. The experiments then consisted in observing if the injected emulsion was either water continuous or oil continuous. The empirical relations as well as the theoretical models just presented predict the critical hold-up at phase inversion for these experiments reasonably well. However continuous experiments in stirred vessels (reported in the literature) and our own continuous experiments in a pipe showed, that phase inversion can be postponed to very high dispersed phase fractions causing a wide ambivalence region. This effect cannot be explained by the above mentioned models. Therefore, in this thesis we propose two theoretical models to explain this phenomenon. First we use a model based on a minimum dissipation assumption (chapter 6). It predicts that phase inversion can be postponed to a high value of the dispersed phase fraction. However, the influence of the injected phase volume fraction is not included in the model and from our experiments it is clear that the width of the ambivalence region depends on it (described in chapter 2 and 3). Our second model is based on a mean-field theoretical description of phase inversion. It takes into account the influence of the injected phase volume fraction. The model is described in chapter 7.

## 1.6 Three-phase flow

There are not many papers dealing with three-phase flow through a pipe, but recently this subject is receiving more attention because of the industrial relevance. The flow pattern as function of the flow conditions was studied (Acikgöz et al. (1992); Hewitt et al. (1997); Wu et al. (2001); Spedding et al. (2005)). Also the influence of the inclination angle of the pipe on the flow was investigated (Oddie et al. (2003)). It has been shown that the pressure drop of the three phase oil-water-gas system is of the same order of magnitude as the pressure drop of a two-phase system consisting of the gas and the dominating liquid phase (Nádlér and Mewes (1995)). Lahey Jr et al.
(1992) applied the drift-flux technique and validated it against volume fractions data for such flows. Furthermore, Hewitt (2005) showed the importance of the mixing process on the flow behavior.

In our research we have restricted ourselves to dispersed three-phase flow with a continuous liquid (oil or water), in which drops of the other liquid and gas bubbles are present. Experimental results from our oil-water-gas experiments are described in chapter 8.

1.7 Aim of this work

In our work we paid special attention to the phenomenon of phase inversion, whereby the mixture switches from an oil-continuous phase with water drops (and gas bubbles) to a water-continuous phase with oil drops (and gas bubbles), or vice versa. During phase inversion the pressure gradient of the flow in the pipe increases strongly. So it is important to know at what conditions phase inversion occurs, in order to find out whether in practice it can be avoided or its effect on the pressure drop minimized. Hopefully this study will contribute to solving practical problems in the (oil) industry. Main research questions were:

- At what dispersed phase volume fraction does inversion in an oil-water flow in a pipe occur? In stirred vessels it has been observed, that inversion can be postponed to high values of the dispersed phase volume fraction. Is that also possible in pipe flows?
- What are the parameters influencing phase inversion?
- How can phase inversion be modeled?
- What is the pressure drop in case of an oil-water dispersed pipe flow? How does it depend on the dispersed phase volume fraction and on properties of the continuous phase?
- What is the influence of gas bubbles on the inversion process?
- Is the presence of drops influencing the behavior of gas bubbles?
- What is the pressure drop in case of an oil-water-gas dispersed flow? How does it depend on the volume fractions of the dispersed phases?

1.8 Outline of the thesis

This thesis is based on papers published or submitted for publication in international scientific journals during the last four years. As a consequence, each chapter can be read on its own. Short sections are added between the chapters to show the connections with previous chapters. Because the chapters are (independent) journal publications, there is sometimes some overlap between them.

In chapter 3 first results are given for an oil-water pipe flow during continuous experiments (during which the dispersed phase volume fraction is gradually increasing by injecting it in the
1.8. Outline of the thesis

Continuous liquid). Special attention is paid to the question, whether it is possible to postpone phase inversion to high values of the dispersed phase volume fraction (similar to continuous experiments in a stirred vessel). To that purpose we varied (for instance) the injected phase volume fraction (volume rate of injected liquid to volume rate of the mixture). In chapter 3 results of continuous experiments are compared with the results of direct experiments (during which the two liquid are pumped separately into the pipe and mix in the entrance region). A high-speed camera was used to visualize the inversion process and pictures taken during the two types of experiments were compared. It is known from earlier experiments that a so-called ambivalence region exists, in which both liquids may be found to be continuous. Only outside this region one phase is always continuous and the other is always discrete. Within the ambivalence region either one of the two phases can be continuous and the exact value at which phases invert depends on the operating conditions. Therefore we construct an inversion map based on our experiments. The inversion map is specific for a certain oil-water mixture and can be modified by adding a surfactant. To show this effect in chapter 4 oil-water experiments are described with added surfactant. In chapter 5 experiments are reported on the phase inversion in the mixing zone between a water flow and an oil flow through a pipe. In chapter 6 a theoretical model is developed for predicting the critical concentration of the dispersed phase at which phase inversion occurs. It is based on a minimum dissipation assumption. A second theoretical model based on a mean-field theoretical description is developed in chapter 7. The experimental results of oil-water-gas pipe flows are given in chapter 8. The effect of gas bubbles on an oil-water flow is described. There is much attention for the influence of bubbles on the inversion process and vice versa.
Chapter 1. Introduction
We started our study with continuous water-oil experiments. To predict the behavior of dispersed water-oil flow it is crucial to know which phase is the continuous one and when inversion occurs. So we paid much attention to the critical concentration at which inversion takes place. Also the pressure gradient (at inversion) as function of the operating conditions was determined. During the experiments a microscope was used to observe the morphology of the dispersed liquid phase before and after inversion.
Chapter 2

Experimental investigation of phase inversion in an oil-water flow through a horizontal pipe loop

An experimental study was made of phase inversion in an oil-water flow through a horizontal pipe loop. The experiments started with the flow of a single liquid through the pipe loop; thereafter the second liquid was gradually added (using different injectors and different injection flow rates) until inversion took place. It was found that in this way the point of inversion could be postponed to high values of the dispersed phase volume fraction (> 0.8). Samples were taken from the flowing mixture and inspected with the aid of a microscope. Multiple drops consisting of oil droplets in water drops were observed, but multiple drops consisting of water droplets in oil drops were never found. The significance of these observations for the phase inversion mechanism is discussed.

2.1 Introduction

The flow of two immiscible liquids often occurs as a dispersed flow, where one liquid is present in the other liquid in the form of drops. Dispersions are widely applied in the petrochemical -, food -, chemical - and pharmaceutical industries. For our investigation we used oil and water without a surfactant. Both water-in-oil and oil-in-water dispersions were possible. Conversion between these two types of dispersion is called phase inversion. Which type is present, depends on several parameters such as the volume fraction of the two phases, the viscosities of the liquids, the inter-facial tension and the turbulence in the flowing mixture. An often used model for phase inversion takes into account two opposite processes: break-up and coalescence of drops (Arashmid and Jeffreys (1980); Nienow (2004)). Usually the break-up of drops is described by comparing the drop deformation due to turbulent pressure fluctuations

with the deformation-restoring effect due to the inter-facial tension. The coalescence of drops is calculated by modeling the collision between drops. Phase inversion is then assumed to take place, when a certain criterion is satisfied. For instance, it is assumed that phase inversion occurs when the rate of breakup is not equal to the rate of coalescence, or that the viscosity of the mixture grows larger than a certain limiting value (Vaessen et al. (1996); Ioannou et al. (2004)). In another model phase inversion is assumed to take place, when the inter-facial energy of the water-in-oil dispersion and the inter-facial energy of the oil-in-water dispersion are equal (Yeo et al. (2002a); Brauner and Ullmann (2002)). For the calculation of the inter-facial energy it is necessary to know the drop size distribution and, therefore, again to model the break-up and coalescence processes.

Another way of explaining phase inversion has been presented by Groeneweg et al. (1998) and Bouchama et al. (2003). They observed that for certain conditions the drop size increases considerably, before phase inversion takes place. This is explained by assuming the inclusion of small droplets of the continuous phase into the dispersed phase drops. They describe this as the formation of a multiple dispersion; for instance oil droplets in water drops in a continuous phase of oil are formed. As the drops grow in size, their collisions and coalescence become more probable which leads to phase inversion. In the publication by Bouchama et al. (2003) an interesting experiment is reported, which seems to confirm (in an indirect manner) the formation of a multiple dispersion as a mechanism for phase inversion. They carried out experiments in a stirred vessel where inversion was detected by a jump in dispersion conductivity. Their system contained paraffin oil, water and a surfactant. The dispersed phase fraction at phase inversion was compared for two emulsification routes for phase inversion, namely the direct route and the wash-out route. In the direct route emulsions were prepared at a certain composition by the direct mixing of the proper oil and water volumes. In the wash-out route the vessel was filled with oil, the impeller switched on and water was added in small volumes to the vessel at a constant flow rate until the same composition was reached as for the direct route. The occurrence of phase inversion was found to be strongly different for both cases. In particular for the wash-out route the way of dilution appeared to be an important parameter. According to the authors this can only be explained on the basis of the role of multiple-dispersion formation in phase inversion. In the wash-out experiments the generated drops were very small, therefore their Laplace pressure was high and multiple drop formation due to entrainment of droplets from the continuous phase was not possible. During the direct experiments the drops formed during the mixing process were much larger and entrainment of small droplets was possible, leading to a multiple dispersion.

The aim of our research, presented in this publication, is to study phase inversion in an oil-water flow through a horizontal pipe. For that purpose we carried out wash-out type experiments in a pipe loop by starting with the flow of a single liquid through the loop and then gradually adding a second liquid using different types of injectors and applying different injection flow rates. We performed experiments starting with water and then adding oil (water-to-oil experiments) and the other way around (oil-to-water experiments). Additional experiments were done in which the injection was stopped before the point of inversion was reached, after which the mixture continued to flow through the pipe loop at the same mixture velocity until inversion had occurred. Much attention was given to the possibility of postponing phase inversion to higher values of the dispersed phase volume fraction (like in the experiments of Bouchama et al. (2003)) and
2.2. Experimental set-up

The experiments were carried out in the Laboratory for Aero- and Hydrodynamics of the Delft University of Technology. The set-up is shown in Fig. 2.1. It is an (acrylic) pipe loop with an inner pipe diameter of 16 mm, consisting of two straight parts of 6 m connected via two bends. The two immiscible liquids used in our experiments were tap water and Shell Macron EDM 110 oil. The oil properties are given in Table 2.1.

to the possibility of multiple drop formation. Particular attention was paid to the influence of these phenomena on the inversion mechanism. In publications by Ioannou et al. (2004) and Ioannou et al. (2005) also attention is given to phase inversion in dispersed liquid-liquid pipe flows. Contrary to our experiments they performed direct-route type of experiments, as in their case the two liquids are mixed in the pipe at certain composition from the start of the experiment.

2.2 Experimental set-up

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Chapter 2. Experimental investigation of phase inversion in an oil-water flow through a horizontal pipe loop

<table>
<thead>
<tr>
<th>Density</th>
<th>794 kg/m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kinematic viscosity</td>
<td></td>
</tr>
<tr>
<td>- at 20°</td>
<td>3.9 mm²/s</td>
</tr>
<tr>
<td>- at 40°</td>
<td>2.4 mm²/s</td>
</tr>
<tr>
<td>Oil-water inter-facial tension</td>
<td>0.045 N/m</td>
</tr>
</tbody>
</table>

Table 2.1: Properties of Shell Macron EDM 110 oil.

<table>
<thead>
<tr>
<th>Water</th>
<th>Re</th>
<th>Fr</th>
<th>We</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 m/s</td>
<td>15936</td>
<td>6.4</td>
<td>354</td>
</tr>
<tr>
<td>2 m/s</td>
<td>31872</td>
<td>25.5</td>
<td>1418</td>
</tr>
<tr>
<td>3 m/s</td>
<td>47808</td>
<td>57.3</td>
<td>3190</td>
</tr>
<tr>
<td>1.34 m/s</td>
<td>21354</td>
<td>11.4</td>
<td>637</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Shell Macron EDM 110</th>
<th>Re</th>
<th>Fr</th>
<th>We</th>
</tr>
</thead>
<tbody>
<tr>
<td>2 m/s</td>
<td>6515</td>
<td>25.5</td>
<td>1129</td>
</tr>
<tr>
<td>3.5 m/s</td>
<td>11401</td>
<td>78</td>
<td>3458</td>
</tr>
<tr>
<td>1.35 m/s</td>
<td>4398</td>
<td>11.6</td>
<td>515</td>
</tr>
</tbody>
</table>

Table 2.2: Dimensionless numbers for water and oil at different superficial velocities. For the calculation of the Froude number the density difference between water and oil is used and for the calculation of the Weber number the inter-facial tension between the two liquids.

During an experiment first one of the liquids (water or oil) was taken from tank 2 in Fig. 2.1 by means of pump 3 and recirculated through the pipe loop by means of pump 1. Pump 1 is a positive displacement pump, chosen to minimize the pumping effect on the dispersion morphology. Pump 3 pressurized the system, so that no entrainment of air into the pipe loop could occur. After recirculating the liquid for a few minutes to ensure that the pipe walls are wetted by the liquid, injection started by pumping (using pump 2) the other liquid from tank 1 via an injector into the pipe loop. Flow meter 1 (a Krohne Optimass 7000 mass flow meter with a measuring error lower than 0.26% of the measured value) measured the density and the flow rate of the mixture in the pipe loop. The flow meter is validated for oil-water mixtures. All experiments were performed at such high Reynolds numbers and Froude numbers (see Tab. 2.2), that always a fully dispersed flow was present according to Brauner (2001). The dispersed phase fraction could therefore be calculated from the measured density. The flow rate and density measurements made with flow meter 1 were used to control pump 1 via a feedback system. The mixture velocity in the pipe loop was kept constant during the experiment. (This was achieved in the following way: Flow meter 2 measured the injection flow rate through pump 3. Pump 3 is a gear pump, which ensured that the injection flow rate was constant during the experiment. During the injection valve 1 was opened and the same volume of dispersion liquid was removed from the pipe loop as the volume that was injected.) We used as injector a cylindrical container (placed around...
the pipe loop) with a number of holes in it. The oil was pumped through the holes into the pipe loop. Three different types of injector were used, viz. a container with 2 holes of 2 mm diameter, a container with 8 holes of 2 mm diameter, and one with 100 holes of 3 mm diameter (see Fig. 2.2). The pressure was measured at 5 m downstream of the injector (see Fig. 2.1) over a distance of 1m. Differential pressure transducers (with a measuring error lower than 1% of the measured value) were used, that sampled signals with a frequency of 50Hz. Averaging over 50 samples was applied. Conductivity measurements (at a frequency of 50Hz) were done with a cell consisting of four electrodes (4mm diameter) mounted in the pipe wall: two in the vertical direction (at the top and bottom of the pipe) and two in horizontal direction (in the side walls). When the flowing mixture was fully dispersed the measured conductivity in vertical and horizontal direction were equal. Conductivity measurements are not presented in this paper. They were used as a confirmation of phase inversion found from pressure measurements. Also temperature measurements were carried out during the experiments. To inspect the dispersion morphology samples were taken from the mixture and photographed. The samples were taken via a small hole in the pipeloop, injected in a pipe with a flowing liquid (the same liquid as the continuous phase in the pipeloop) and then inspected under a microscope (of a magnification of 50) where also pictures were taken. A sketch of the sampling technique is given in Fig. 2.1. Perhaps this procedure has some influence on the drop size distribution. However, the most important reason for taking samples was to study the composition of the drops, in particular to see whether or not multiple drops were formed. To study the influence of the length of the pipe loop (and of pump 1) also experiments were performed for which, each of the two straight parts of the loop was reduced to 4.5 m. Of course during these experiments the mixture flows more often through the pump. However, we observed no differences with the experiments carried out in the longer pipe loop. Each result presented in this paper is from a single experiment, but all experiments were conducted at least two times and the reproducibility of the experiments was good.

2.3 Experimental results

2.3.1 Water-to-oil experiments

A water continuous to oil continuous (water-to-oil) experiment starts with pumping water through the pipe loop. Thereafter, injection of oil begins and an oil-in-water dispersion develops. With continuing oil injection the volume concentration of drops increases and at a certain moment in time inversion to a water-in-oil dispersion takes place. Pictures have been taken at different stages of the inversion process (see Fig. 2.3). The left picture shows the dispersion before inversion. Only ”clean” oil drops are present; so multiple drops (consisting of small water droplets in larger oil drops) are not formed before inversion. The middle picture shows the dispersion just after inversion. As can be seen during the inversion process multiple drops (consisting of small oil droplets in larger water drops) are generated. The right picture shows the dispersion a few minutes after inversion. Multiple drops are still present, but they contain less oil droplets compared to the situation just after inversion. It appears that oil droplets have escaped from the
Chapter 2. Experimental investigation of phase inversion in an oil-water flow through a horizontal pipe loop

Figure 2.3: Dispersion morphology during water-to-oil experiments. From left to right: before inversion, just after inversion and a few minutes after inversion.

<table>
<thead>
<tr>
<th>injector type</th>
<th>jet exit velocity</th>
</tr>
</thead>
<tbody>
<tr>
<td>100x3mm</td>
<td>0.07 m/s</td>
</tr>
<tr>
<td>8x2mm</td>
<td>2 m/s</td>
</tr>
<tr>
<td>2x2mm</td>
<td>8 m/s</td>
</tr>
</tbody>
</table>

Table 2.3: Jet exit velocities at a mixture velocity of 2 m/s and an injection flow rate of 12.5%.

We studied the influence of different injector types on the phase inversion process. As mentioned, we used three different types of injector, viz. a container with 2 holes of 2 mm diameter, a container with 8 holes of 2 mm diameter, and one with 100 holes of 3 mm diameter. These three types of injector were chosen to investigate the influence of injection velocity (and therefore different drop sizes) on the phase inversion mechanism. We kept the oil injection flow rate the same during the experiments. It is evident, that for the same oil flow rate the jet exit velocity of the oil decreases with increasing number of holes. During the experiments we kept the mixture velocity in the pipe loop at 2 m/s (so a highly turbulent flow - see Table 2.2) by using the information from flow meter 1. The injection flow rate was 12.5% of the mixture flow rate in the pipe loop. The exit velocities of the oil jets issuing from the injector holes (for the three types of injectors) are given in Table 2.3.1. We made an estimate of the size of the oil drops as generated by the jet break-up and as generated by the turbulence in the flowing mixture at the start of the experiments. For all conditions the drop size was determined by the turbulence. The results of the experiments are given in Fig. 2.4, which shows the scaled pressure drop (ratio of the actual pressure drop and the pressure drop at the start of the experiment with the single-phase flow of only water) over a part of the pipe loop as function of the oil volume fraction for the three different injectors. Inversion occurs at an oil volume fraction between 0.8 and 0.85. The figure shows that the inversion process is independent of the injector type. Of course the drop size distribution depends on the break-up of the jets issuing from the injector holes (which depends on the jet exit velocity). However, as can be seen on Fig. 2.4, this effect has hardly any influence on the inversion process for the injectors that we used.
2.3. Experimental results

Next we investigated the influence of the injection rate. For that purpose we carried out experiments with injection rates of 5%, 15% and 26.5% of the mixture flow rate in the pipe loop using one injector type (100x3mm). The mixture velocities for these experiments was 1 m/s and the flow was still fully dispersed. The results are given in Fig. 2.5. As can be seen in this figure (giving the scaled pressure drop as function of the oil volume fraction for the three different injection rates) the development towards inversion appears to be independent of the injection rate. However, the oil volume fraction at the point of inversion decreases with increasing injection rate.

Inversion does not take place at all locations in the pipe loop at the same time. It starts at a certain location with the formation of an inverted pocket which grows as it flows downstream. This explains, that during inversion the pressure drop over the pipe loop fluctuates considerably. This can be seen in Fig. 2.6, in which the scaled pressure drop (not an averaged signal) is shown as function of time (for a mixture velocity of 2 m/s and the injection rate of 12.5%). The inversion process starts at about \( t = 270s \), when the pressure drop strongly decreases. Thereafter the pressure drop increases due to the arrival of a non-inverted pocket of fluid and when this has passed the pressure taps, the pressure drop again decreases. After one circulation through the pipe loop the non-inverted pocket still exists, causing another peak in the pressure drop. When the inverted pocket passes the injector, it is stimulated to grow due to newly injected oil. Usually after two (sometimes more) passes through the pipe loop the complete mixture has inverted. So inversion is a kind of statistical process. At higher injection rates the probability of fast growing inverted pockets increases and so (on average) the oil volume fraction at the point of inversion decreases.

We also investigated the influence of the mixture velocity on the inversion process. For that
Chapter 2. Experimental investigation of phase inversion in an oil-water flow through a horizontal pipe loop

Figure 2.5: Scaled pressure drop as function of oil volume fraction for water-to-oil experiments at a mixture velocity of 1 m/s and for three different injection rates.

purpose we carried out water-to-oil experiments at mixture velocities of 1 m/s, 2 m/s and 3 m/s (with Reynolds numbers ranging from 15000 to 50000 - see Table 2.2) for various injection rates. As can be seen in Fig. 2.7 the influence of the mixture velocities on the inversion process is negligible in the range of mixture velocities that we studied. The influence of the injection flow rate on the oil volume fraction at the point of inversion for mixture velocities of 2 m/s and 3 m/s is similar to the case of 1 m/s.

2.3.2 Oil-to-water experiments

An oil continuous to water continuous (oil-to-water) experiment starts with pumping oil through the pipe loop. Then injection of water begins and a water-in-oil dispersion develops. With continuing water injection the volume concentration of drops increases and at a certain moment in time inversion to an oil-in-water dispersion takes place. Also in this case pictures have been taken at different stages of the inversion process (see Fig. 2.8). These pictures have been taken during an experiment as indicated in Fig. 2.9. In this experiment the mixture flow rate was 3.5 m/s and the injection rate was 5%. The injection of water was stopped before inversion occurred at \( t = 220 \text{s} \), when the water volume fraction reached 0.64. The mixture continued to flow through the pipeloop without further water injection, although the pressure drop continued to increase (see Fig. 2.9) and finally at \( t = 450 \text{s} \) inversion took place. The left-top picture of Fig. 2.8 shows a sample of the mixture taken at \( t = 160 \text{s} \), the right-top picture at \( t = 280 \text{s} \), the left-bottom picture at \( t = 400 \text{s} \) and the right-bottom picture at \( t = 465 \text{s} \). It is evident, that in this case multiple drops (oil droplets in water drops) were formed before inversion. The volume fraction of oil inside the water drops increased with time. After inversion only "clean" oil drops were present; so no multiple drops were formed after the inversion process.
For the oil-to-water experiments we also investigated the influence of the injection rate. For that purpose experiments were carried out for a mixture velocity of 3.5 m/s and injector flow rates of 7%, 10% and 15%. The Reynolds number at the start of the experiments was 11000. For safety reason (too high pressure inside the loop) we could not go to larger values of the Reynolds number in this case. The results are given in Fig. 2.10, showing the scaled pressure drop as function of the water volume fraction for the three values of the injection flow rate. As is the case for the water-to-oil experiments, also for the oil-to-water experiments the development towards inversion is rather independent on the injection flow rate. However, the (critical) water volume fraction at the point of inversion depends on the injection rate (see Fig. 2.11 in which also the results for other injection rates are shown). The explanation for the decrease of the critical water volume fraction with increasing injection rate at large values of the injection rate is the same as for the water-to-oil experiments. However, the small value of the critical volume fraction at low values of the injection rate is new. The explanation is the fact (based on our observations), that at low injection rates the water drops have more time to entrain oil droplets and grow to a larger size than at high injection rates. So, a smaller amount of injected water is necessary for the inversion process to occur. As for the case of the water-to-oil experiments we did not find for the oil-to-water experiments any significant influence of the injector type on the inversion process at a mixture velocity of 3.5 m/s.

We also performed oil-to-water experiments at a mixture velocity of 2 m/s. As the oil viscosity is significantly larger than the water viscosity the Reynolds number of the flow is for this case significantly smaller than for the water-to-oil experiments carried out at 1 m/s, 2 m/s and 3 m/s. It is only 6500 at the start of the experiment. So for this case we could expect an influence of the injector type, as the turbulence of the mixture is not so dominant. The results are given in Fig. 2.12, showing the scaled pressure drop as function of water volume fraction for three types of
Chapter 2. Experimental investigation of phase inversion in an oil-water flow through a horizontal pipe loop

We carried out more experiments similar to the one shown in Fig. 2.9, where the injection of water was stopped before inversion took place. After stopping the water injection the mixture continued to flow through the pipe loop at the same mixture velocity. All the conditions were the same as for the experiment of Fig. 2.9, only the water volume fraction at which the water injection was stopped was varied. The results are given in Fig. 2.13, which shows the scaled pressure drop as function of the water volume fraction for the different experiments. As can be seen the maximum pressure drop at the point of inversion is always nearly the same. Similar behavior was observed by Tyrode et al. (2005) in a rheomixer. In Fig. 2.14 we give the time that is needed after stopping the water injection at a certain water volume fraction to reach the inversion point. This time decreases with increasing value of the water volume fraction at which the water injection was stopped. The reason is that, at a high water volume fraction (at which the water injection was stopped) less time is needed for the multiple drops to grow and achieve a volume fraction large enough for inversion than at a low water volume fraction (at which the water injection was stopped). In all these experiments we observed the same phenomenon as shown in Fig. 2.8. Multiple drops (oil droplets in water drops) were formed before inversion. The volume fraction of oil inside the water drops increased with time. After inversion only "clean" oil drops were present; so no multiple drops were formed after the inversion process.

At this point we want to discuss the possible influence of the length of the pipe loop on our oil-to-water experiments. Inversion is for a significant part determined by the volume fraction of the dispersed phase. During our experiments this volume fraction increases with time because of two mechanisms: injection of the dispersed phase liquid and growth of the multiple droplets by taking-up more oil droplets. Apart from the experiments with a very low injection flow rate, for all other experiments the first mechanism is dominant. For these experiments the equilib-
2.4 Discussion

Figure 2.8: Dispersion morphology for oil-to-water experiment as described in Fig. 2.9; left-top picture is taken at point 1 in Fig. 2.9, right-top picture at point 2, left-bottom picture at point 3 and right-bottom picture at point 4.

Equilibrium drop size distribution and volume fraction of the dispersed phase establish themselves very quickly and there is no influence of the length of the pipe loop. We checked that by reducing the length of the loop and finding the same results. Also pressure drop measurements taken at various distances from the injector show the same results.

Of course, there is also no influence of the length of the pipe loop when only the second mechanism (growth of multiple drops) is present, as in that case there is no injection of liquid (and we know that the pump has no influence on the morphology of the dispersion). The growth of the multiple droplets is a slow process. In the experiment when injection was stopped at a water volume fraction of 0.54, it took 730s for inversion to occur. This means that the dispersion traveled for $2.5km$ in the pipe loop before inversion.

So only at very low injection flow rates, when the two mechanisms become of the same importance, we can expect an influence of the length of the pipe loop.

2.4 Discussion

For the case of water-to-oil experiments multiple drops (water droplets in oil drops) were not observed before inversion. Only after inversion multiple drops (oil droplets in water drops) were present. For the oil-to-water experiments the behavior was the other way around. Multiple
Chapter 2. Experimental investigation of phase inversion in an oil-water flow through a horizontal pipe loop

Figure 2.9: Scaled pressure drop as a function of time for an oil-to-water experiment at a mixture velocity of 3.5 m/s and an injection flow rate of 5%. Injection stopped before inversion at a water volume fraction of 0.64.

drops (oil droplets in water drops) were observed before inversion; no multiple drops (water droplets in oil drops) were present after inversion. Similar behavior was observed by Pacek and Nienow (1995). They observed this non-symmetrical behavior even when the viscosities and densities of water and oil were nearly the same. A possible explanation for this phenomenon is, that the chemical properties of the two-phase system are important for multiple drop formation. Tyrode et al. (2005) introduces the so-called $HLD$-factor (hydrophilic-lipophilic deviation from an optimum formulation) to indicate the chemical formulation. When $HLD < 0$ the surfactant exhibits a stronger affinity to oil than to water, for $HLD > 0$ it is the other way around. Tyrode et al. (2003) and Tyrode et al. (2005) shows that for $HLD < 0$ two types of dispersion are possible: a dispersion with multiple drops consisting of oil droplets in water drops in oil, and a dispersion with clean oil drops in water. When $HLD > 0$ there are again two dispersion types: a dispersion with multiple drops consisting of water droplets in oil drops in water, and a dispersion with clean water drops in oil (see also Fig. 2.15 of Tyrode et al. (2005)). In our experiments we observed the following two types of dispersion: a dispersion with multiple drops consisting of oil droplets in water drops in oil, and a dispersion with clean oil drops in water. So we assume that for our case $HLD < 0$. We plan to carry out experiments at $HLD > 0$ to check this explanation.

These observations let us conclude, that for our experiments the inversion mechanism for water-to-oil experiments is different from the mechanism for oil-to-water experiments. For water-to-oil experiments multiple drops are not formed before inversion; so the effective dispersed phase fraction is equal to the actual oil volume fraction. The volume fraction of the oil drops increases until concentration becomes so high, that phase inversion occurs. The value of the dispersed volume fraction at the point of inversion depends on the injection rate. At higher injection rates the inversion takes place at lower values of the dispersed volume fraction than at lower injection rates. As explained earlier the reason is likely, that at higher injection rates
2.4. Discussion

Figure 2.10: Scaled pressure drop as a function of the water volume fraction for the oil-to-water experiments at a mixture velocity of 3.5 m/s and three different injection flow rates.

the probability of fast growing inverted pockets increases. In this case the dispersion is very stable, even pumping it through the pipe loop for a long time did not change the pressure drop or dispersion morphology. Even at a very high dispersed phase volume fraction multiple collisions did not lead to encapsulation of water as reported by Núñez et al. (1996). The explanation is in our opinion, that encapsulation is determined by chemical properties of the system and that in their case $HLD > 0$.

For oil-to-water experiments water drops are present, that continuously entrain small amounts of oil. So the effective dispersed phase volume fraction is larger than the water volume fraction. The multiple droplets grow continuously in size, until their volume fraction is so large that inversion takes place. Entrainment of oil into the water drops and the possible escape of oil droplets from the water drops are the two mechanisms, that determine the growth rate of the drops (Groeneweg et al. (1998); Liu et al. (2005); Sajjadi et al. (2000, 2002)). Based on this reasoning it is to be expected, that the dispersed phase volume concentration at inversion is lower for the oil-to-water experiments than for the water-to-oil experiments. This is in agreement with our results, although the effect for our experiments is not very strong because also other influences are important (for instance the injection flow rate). It is important to emphasize at this point, that the dispersed phase volume fraction at inversion for (all) our wash-out route type experiments is much higher than the dispersed phase volume fraction at inversion for the direct route type of experiments performed by Ioannou et al. (2004) and Ioannou et al. (2005), where inversion occurs typically for an oil volume fraction around 0.7. In our experiments inversion was delayed up to 0.75 for the water volume fraction for oil-to-water inversion and up to 0.90 for the oil volume fraction for the water-to-oil inversion.

The pressure drop increase at inversion compared to the pressure drop at the start of the experiment is for oil-to-water experiments equal to about 2, where as for the water-to-oil experiment
it is about 4 (so considerably larger). This is likely caused by the difference in drop size for the two experiments: for the oil-to-water experiments the drops are significantly larger (because of multiple drop formation) than for the water-to-oil experiments. According to Pal (2000) the effective viscosity of a dispersion with drops of a certain size is larger than the effective viscosity of a dispersion with larger drops, when the dispersed volume fraction is the same for both cases. Moreover, at inversion the dispersed volume fraction for water-to-oil experiments is larger than for oil-to-water experiments.
2.4. Discussion

Figure 2.12: Scaled pressure drop as function of water volume fraction for oil-to-water experiments at a mixture velocity $2 \text{m/s}$, an injection rate of 12.5% and three different injectors.

Figure 2.13: Scaled pressure drop as function of water volume fraction for oil-to-water experiments at a mixture velocity of $3.5 \text{m/s}$ and for several values of the water volume fraction at which injection was stopped.
Chapter 2. Experimental investigation of phase inversion in an oil-water flow through a horizontal pipe loop

Figure 2.14: Time to inversion after injection stop for oil-to-water experiments for a mixture velocity of 3.5 m/s as function of the water volume fraction at which injection was stopped.

Figure 2.15: Composition map for different hydrophilic-lipophilic deviation values.
In the next chapter the continuous experiments from the previous chapter are compared with direct experiments (again with much attention for the critical concentration). The inversion map for the continuous experiments will be given and the position of the direct experiments in this map is explained. Again experimental results for the pressure gradient for both types of experiments will be given. High-speed camera pictures help significantly with the interpretation of the measured results.
Chapter 3

On the phase inversion process in an oil-water pipe flow*

An experimental study of the phase inversion process in an oil-water flow through a pipe was carried out. Special attention was paid to the critical concentration of the dispersed phase at which phase inversion occurs and to the change in morphological structures during inversion. To that purpose two different types of experiments were performed: (1) continuous experiments during which the dispersed phase fraction was gradually increased and (2) direct experiments whereby oil and water were injected simultaneously into the pipe at a certain concentration. During the experiments detailed pictures were taken of the phase inversion process and simultaneously the electrical conductivity of the mixture was measured to determine which liquid formed the continuous phase and which the dispersed phase. Also the pressure gradient over several parts of the pipe was measured.

For continuous experiments the critical concentration was found to depend on the injection phase volume fraction. The critical concentration was significantly higher for continuous experiments than for direct experiments. The change in morphological structures during phase inversion was the same for the two types of experiments. During inversion the concentration of drops of the (originally) dispersed phase becomes so high, that they coalesce at certain places in the flow field and form relatively large, rather complex, morphological structures. With a further increase in concentration of the (originally) dispersed phase these morphological structures grow in size and start to form the new continuous phase in which again complex structures are present, but this time consisting of the (originally) continuous phase.

3.1 Introduction

The flow of two immiscible liquids often occurs as a dispersed flow, where one liquid is present in the other liquid in the form of drops. Dispersions are widely used in the petrochemical-, food-, chemical- and pharmaceutical industries. Handling and controlling dispersion properties is of a

key interest for practical applications. Special interest has to be given to the phase inversion phenomenon, whereby the dispersed phase becomes the continuous one, and vice versa. During this process the effective viscosity of the mixture becomes very large, which leads to high pressure drops or low flow rates.

The phase inversion phenomenon has been studied for many years (see, for instance, Becher (2001)), but there are still many questions about the inversion process. Some authors assume that a sudden coalescence of drops can encapsulate parts of the continuous phase, which causes drop formation from the (originally) continuous phase (Yeo et al. (2002a); Brauner and Ullmann (2002)). Others observed the creation of multiple drops (small droplets of the continuous phase inside the drops) prior to inversion (Pacek et al. (1994); Pacek and Nienow (1995); Pal (1993); Sajjadi et al. (2000, 2002, 2003); Liu et al. (2005, 2006)). According to some authors inversion is a rather rapid process, called catastrophic by them, (Dickinson (1981); Smith and Lim (1990); Rondón-Gonzaléz et al. (2006); Tyrode et al. (2005); Vaessen et al. (1996); Binks and Lumsdon (2000)). However, during our experiments we found that this is not strictly true as also shown by Liu et al. (2006).

Most of the experiments reported in the literature were performed in a stirred vessel and usually water and oil were used. They were often continuous experiments during which the dispersed phase was gradually added to the continuous phase. For this type of experiments it was found, that phase inversion could be postponed to a high value (> 0.8) of the dispersed phase volume fraction. Also a wide ambivalent volume fraction region existed where the mixture could be either water continuous or oil continuous (Vaessen et al. (1996); Groeneweg et al. (1998); Deshpande and Kumar (2003); Mira et al. (2003); Tyrode et al. (2003)). During direct experiments in a stirred vessel the two liquids were mixed at a certain concentration (Quinn and Sigloh (1963); Tyrode et al. (2005)) and inversion usually occurred at a value of the dispersed phase fraction close to 0.5 (dependent on the properties of the liquids) and no ambivalence region was observed. It is important to point out, that in the above mentioned papers some are without an added surfactant (for instance Deshpande and Kumar (2003); Pacek and Nienow (1995); Liu et al. (2005, 2006)) and others with an added surfactant (for instance Binks and Lumsdon (2000); Rondón-Gonzaléz et al. (2006); Tyrode et al. (2005)). The presence of a surfactant can have a significant influence on the inversion process; they tend to favor one type of dispersion over the other. In our experiments we have not added a surfactant. However, in the oil (that we used) there were small concentrations of substances that were acting like a surfactant, as water was favored as the continuous phase.

Only a few phase-inversion experiments were carried out in a pipe. Direct experiments were done by Liu et al. (2006) in a vertical pipe and by Pal (1993); Nädler and Mewes (1997); Ioannou et al. (2005); Chakrabarti et al. (2006) in a horizontal pipe. They paid particular attention to the pressure drop increase during phase inversion. We performed continuous experiments in a horizontal pipe - see Piela et al. (2006). We measured also a strong increase in the pressure drop during the inversion process. Moreover we made pictures of the change in morphological oil-water structures during inversion. As for the stirred vessel experiments the comparison between the direct experiments and the continuous experiments showed, that also in the case of a pipe flow the critical concentration of the dispersed phase fraction at inversion was very different for the two types of experiments. Dependent on the experimental conditions the critical concentration
can be significantly higher for continuous experiments than for direct experiments. For practical applications this result is very important, as it opens the opportunity to avoid or postpone phase inversion (causing a high pressure drop or low flow rate) by gradual injection of the dispersed phase into the continuous one. Therefore we decided to carry out a new set of experiments to study the critical concentration for continuous experiments as function of some parameters (such as the injection phase volume fraction). To get additional information also detailed pictures were taken during phase inversion for both continuous and direct experiments. It is our hope and expectation, that the information about the dependence of the critical concentration on some relevant parameters and the detailed pictures of the structures occurring during phase inversion will lead to a better control of the phase inversion process during practical applications.

The novelty of this publication beyond that of Piela et al. (2006) is: the comparison between continuous experiments and direct experiments, the detailed observation of the inversion process and the influence of the injection phase volume fraction on the width of the ambivalence region. In section 3.2 of this paper the experimental facilities used for the continuous and direct experiments are described. Then results from the continuous experiments are presented in section 3.3 and from the direct experiments in section 3.4. Section 3.5 describes in detail important interactions taking place during phase inversion. In section 3.6 particular attention is paid to the ambivalence region and the critical concentration for continuous experiments. Finally, in section 3.7 concluding remarks are made.

3.2 Experiments

3.2.1 General information

The details of the pipe configurations used for the two types of experiments will be given when these experiments are discussed. An acrylic pipe with an inner pipe diameter of 16 mm was used. The two immiscible liquids used were tap water and Shell Macron EDM 110 oil (density \(794\, \text{kg/m}^3\), kinematic viscosity at 20° \(3.9\, \text{mm}^2/\text{s}\) and oil-water interfacial tension 0.045 N/m).

All experiments were performed at high Reynolds and Froude numbers to make sure that always a fully dispersed flow was present (according to Brauner (2001) a mixture velocity of 1.35 m/s is sufficient for our experimental conditions). Under such conditions the slip velocity is negligible and the dispersed phase volume fraction could therefore directly be calculated from the measured density. In both types of experiments the mixture velocity in the pipe was kept constant.

To make a detailed study of the dispersion morphology samples were taken from the flow by means of a 7 mm inner diameter sampling tube and led through a visualization cell (which consists of 2 glass plates 1 mm apart; the length of the cell is 20 mm). The visualization cell was illuminated from one side with a 500W halogen lamp and a high speed camera took images at the other side. The camera was operated at a frame rate of 50 Hz and 500 Hz. A sketch of the sampling technique is given in Fig. 3.1. To check whether this observation procedure had some influence on the dispersion morphology we performed experiments with various distances between the visualization cell and the pipe and with various sample flow rates. We always achieved the same results. We also changed the distance between the glass walls from 1 mm to
Chapter 3. On the phase inversion process in an oil-water pipe flow

Figure 3.1: Sketch of the visualization method.

5 mm and again the results were the same.
Pressure drops were measured at different locations over a distance of 1 m. Differential pressure transducers (Validyne DP-15, measuring error < 3%) were used and the pressure signal was sampled at a rate of 2 kHz. Data were averaged over 2000 samples. Conductivity measurements (at the same frequency) were done with a cell consisting of two (0.2 mm diameter) wire electrodes mounted in the pipe: one in the vertical direction and one in the horizontal direction. The distance between the electrodes in the center of the pipe was 2 mm. During the experiment we also monitored the temperature with a thermocouple mounted in the pipe wall. Most of the experiments were conducted more than two times and the reproducibility of the experiments was good.

3.2.2 Continuous experiments

The sketch of the set-up used for the continuous experiments is shown in Fig. 3.2. The symbols used in the sketches have the following meaning: V: valves, F1: Krohne Optimass 7000 flow meter (measuring error < 0.26%), F2: Krohne Corimass E flow meter (measuring error < 0.4%), C1: conductivity cell and T1: thermocouple. During the continuous experiments one of the liquids (water or oil) was taken from the continuous phase tank (see Fig. 3.2) and recirculated through the pipe loop by means of pump 1. Pump 1 is a positive displacement pump (lobe pump), chosen to minimize the pumping effect on the dispersion morphology. After recirculating the liquid for a few minutes to ensure that the pipe walls were wetted by the liquid, injection of the other liquid started by pumping (using pump 2) the dispersed phase through the injector into the pipe loop. During the injection valve 2 was opened and the same volume of dispersion liquid was removed from the pipe loop as the volume that was injected. Flow meter 1 measured the density and the flow rate of the mixture in the pipe loop. The flow meter was calibrated for oil-water mixtures. The mixture velocity was kept constant during the experiment by an electronic feedback system (pump 1 was controlled based on measurements of flow meter 1). To study the influence of the length of the pipe loop also continuous experiments were carried out, whereby the pipe loop was significantly reduced in length. During these experiments the mixture flows more often through the pump. However, we observed no difference in the results when compared
with the results of the experiments carried out in the longer pipe loop. A cylindrical container (placed around the pipe loop) with a number of holes in it was used as injector. The oil was pumped through the holes into the pipe loop. Three different types of injector were used, viz. a container with 2 holes of 2 mm diameter, a container with 8 holes of 2 mm diameter, and one with 100 holes of 3 mm diameter (for more details, see Piela et al. (2006)). The experimental results were independent of the injector type. The pressure drop was measured just after the injector, 2 m (125d) downstream of the injector and 5 m (313d) downstream of it.

3.2.3 Direct experiments

During the direct experiments water and oil were taken from the two tanks and injected simultaneously by means of pump 1 and pump 3 into the pipe (see Fig. 3.3). The mixture was injected at a constant concentration for at least 40s. Flow meter 1 measured the density and the flow rate of the mixture in the pipe. The oil volume fraction was calculated from the density measurement. The pressure drops were measured at six different positions: immediately downstream of the inlet and at distances of 2.0 m (125d), 5.0 m (313d), 11.7 m (731d), 18.7 m (1169d) and 26.5 m (1656d) from the inlet.
3.3 Results for continuous experiments

A continuous experiment starts by flowing the first liquid at a certain velocity through the pipe loop and then gradually increasing the concentration of the second liquid by injecting it (in the form of drops) at a certain flow rate into the first liquid. In our description of the morphological structures we will use the following three terms: drops, pockets and regions. A drop is the smallest part of the dispersed phase, usually smaller than 1 mm. A pocket is a larger unit of one of the two phases, that contains several drops of the other phase (see Fig. 3.4); it is usually of the order of a few millimeters up to 1 cm. Finally a region is a still larger part of one of the two phases in the flow field, that encloses several pockets and drops of the other phase and is of the order of 1 cm or larger. In a few cases we will also use the term multiple drop, which is a drop containing small droplets of the other phase. A pocket is larger than a multiple drop and the surface tension is not strong enough to give the pocket a spherical shape, whereas a multiple drop is (nearly) spherical.

3.3.1 Phase inversion mechanism

During a continuous experiment the dispersed phase fraction gradually increases and also the effective viscosity increases (see Pal (2000)). At a certain critical concentration of the dispersed phase inversion occurs. Fig. 3.4 shows the different stages of the phase inversion process for a water-continuous flow to an oil-continuous flow (water-to-oil experiment) for a mixture velocity of 1 m/s and an injection phase volume fraction (ratio of the injection flow rate and the mixture flow rate in the pipe) of 0.125. The top-left picture shows the start of the inversion process at an oil volume fraction of 0.84. Some larger oil-continuous pockets (containing water droplets) are formed in a water-continuous region due to coalescence of the original oil drops. After 16 s (bottom-left picture) the number of these oil-continuous pockets has increased due to continuous oil injection via the injector. With further oil injection isolated water-continuous pockets are created embedded in oil-continuous regions. After 43 s (top-right picture) the oil-continuous pockets in water-continuous regions and water-continuous pockets in oil-continuous regions are about the same in number and size. With further oil injection the oil-continuous pockets disappear and only water-continuous pockets in an oil-continuous region remain. After 82 s (bottom-right picture) the phase inversion is almost completed. There are still some isolated water-continuous pockets, that break up into drops.

The phase inversion process as described above does not take place at the same time at all locations in a cross-section of the pipe flow. This can clearly be observed in Fig.3.5, which shows the complete side view of the pipe for a continuous experiment at a mixture velocity of 1 m/s and an injection phase volume fraction of 0.25. The top-left picture shows the dispersion before inversion. Because of the high oil volume fraction the dispersion is not transparent. After 6 s (center-left picture) the dispersion close to the pipe walls inverts (from water continuous to oil continuous) and the mixture becomes transparent in these regions (as the concentration of the dispersed phase is lower). After 22 s (bottom-left picture) inverted regions can also be observed in the center of the pipe. After 33 s (top-right picture) large non-inverted water-continuous regions are separated by the new oil-continuous phase. 4 s later the pump was stopped and non-
3.3. Results for continuous experiments

Figure 3.4: Inversion from a water-continuous flow to an oil-continuous flow during a continuous experiment. Starting from the top-left picture, the bottom-left picture is taken after 16 s, the top-right after 43 s and bottom-right after 82 s. Inverted water-continuous regions settle to the bottom of the pipe due to their negative buoyancy (center-right picture). It can clearly be observed that the volume fraction of non-inverted (water-continuous) regions is still large. It takes another 39 s to get fully separated layers (bottom-right picture).

3.3.2 Dimensionless parameters

We made an attempt to describe phase inversion during the continuous experiments by means of dimensionless parameters. To that purpose we introduce four dimensionless groups, viz. the Reynolds number \( Re = \frac{\rho ud}{\mu} \), the Froude number \( Fr = \frac{u}{\sqrt{gd}} \), the Weber number \( We = \frac{\rho u^2 d}{\sigma} \), and the injection phase volume fraction \( \chi \). \( \rho \) is the fluid density, \( u \) the average (mixture) velocity, \( d \) the pipe diameter, \( \mu \) the kinematic viscosity, \( g \) the gravity acceleration and \( \sigma \) the water-oil surface tension.

In Fig.3.6 the measured results for the friction factor \( f = \frac{2 \Delta P d}{\rho u^2 L} \) (\( \Delta P \) is the pressure drop over a distance \( L \)) are given for water-to-oil continuous experiments at values of \( \chi \) of 0.03, 0.125 and 0.18, as function of the dispersed phase volume fraction. (The values of \( Re \), \( Fr \) and \( We \) at the start of these experiments are given in Table 1). As can be seen the friction factor increases with increasing dispersed phase volume fraction, until it reaches its maximum during phase inversion after which it decreases significantly. When we repeated these experiments for the same value of \( \chi \) at different (but sufficiently large) velocities \( u \) (and hence different values of \( Re \), \( Fr \) and \( We \)) the results did not change. Fig.3.6 shows the significant influence of the injection phase volume fraction \( \chi \) on the phase inversion process. Both the dispersed phase volume fraction and the friction factor at the point of phase inversion depend on \( \chi \).

To study the dependence on the velocity with which the dispersed phase was injected into the
Chapter 3. On the phase inversion process in an oil-water pipe flow

Figure 3.5: A side view of the pipe during the inversion from a water-continuous flow to an oil-continuous flow during a continuous experiment. Starting from the top-left picture the center-left picture is taken after 6 s, the bottom-left picture after 22 s, the top-right picture after 33 s, the center-right picture after 38 s and the bottom-right picture after 77 s.

<table>
<thead>
<tr>
<th></th>
<th>Re</th>
<th>Fr</th>
<th>We</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 m/s</td>
<td>1.6 \times 10^4</td>
<td>6.4</td>
<td>354</td>
</tr>
<tr>
<td>2 m/s</td>
<td>3.2 \times 10^4</td>
<td>25.5</td>
<td>1418</td>
</tr>
<tr>
<td>3 m/s</td>
<td>4.8 \times 10^4</td>
<td>57.3</td>
<td>3190</td>
</tr>
<tr>
<td>1.34 m/s</td>
<td>2.1 \times 10^4</td>
<td>11.4</td>
<td>637</td>
</tr>
</tbody>
</table>

dispersed phase boundary - Brauner (2001)

<table>
<thead>
<tr>
<th></th>
<th>Re</th>
<th>Fr</th>
<th>We</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shell Macron EDM 110</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2 m/s</td>
<td>0.7 \times 10^4</td>
<td>25.5</td>
<td>1129</td>
</tr>
<tr>
<td>3.5 m/s</td>
<td>1.1 \times 10^4</td>
<td>78</td>
<td>3458</td>
</tr>
<tr>
<td>1.35 m/s</td>
<td>0.4 \times 10^4</td>
<td>11.6</td>
<td>515</td>
</tr>
</tbody>
</table>

dispersed phase boundary - Brauner (2001)

Table 3.1: Dimensionless numbers for water-continuous or oil-continuous experiments at different superficial velocities. For the calculation of the Froude number the density difference between water and oil is used and for the calculation of the Weber number the interfacial tension between the two liquids. "Dispersed phase boundary" indicates the transition from the dispersed flow pattern to another flow pattern according to Brauner (2001).
3.3. Results for continuous experiments

mixture also water-to-oil continuous experiments were carried out for several values of the injection velocity at the same value of the injection phase volume fraction $\chi$. This was achieved by using different numbers of holes in the injector. In Fig. 3.7 the friction factor for these experiments is given as function of dispersed phase volume fraction for three values of the injection velocity. It can be concluded that the friction factor and dispersed phase volume fraction at phase inversion are not very dependent on the injection velocity at a constant value of $\chi$.

In the literature it is sometimes stated that at inversion the friction factor and the dispersed phase volume fraction depend on $Re$, $Fr$, $We$ and $\chi$ (see Deshpande and Kumar (2003); Ioannou et al. (2005); Mira et al. (2003)). We found that the dependence on $Re$, $Fr$ and $We$ disappears for sufficiently large values of these parameters. This is very likely due to the fact that (at high values of $Re$) the turbulence in the pipe flow dominates the mixing between the two liquids. So for our experiments the results only depend on the injection phase volume fraction $\chi$.

3.3.3 Friction factor and conductivity

During each continuous experiment the pressure drop was measured at three different positions along the pipe (see section 3.2.2) and the local friction factor was calculated. Also the conductivity of the mixture was measured. The friction factor and the (dimensionless) conductivity for a water-to-oil experiment at a mixture velocity of 1 m/s (the mixture velocity was sufficiently large to sustain a dispersed flow regime) and a injection phase volume fraction of 0.125 are shown on Fig. 3.8. (Only measurements during the last stage of the inversion process are shown.) For each downstream location the time is shifted to the time at the inlet by using the mixture velocity. In this way it is possible to study the development in time of a certain flow structure.

As during the experiment oil is gradually added to the mixture, the dimensionless conductivity (1 for water and 0 for oil) is decreasing. The conductivity starts to oscillate at 270 s due to the creation of oil-continuous pockets with a lower conductivity than the water-continuous region.
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With further oil injection more and more oil-continuous pockets appear and the oscillations in conductivity increase. Around 320 s the conductivity rapidly decreases, because there are more oil-continuous regions than water-continuous regions and finally inversion takes place.

In Fig. 3.8 also the result for the friction factor as function of injection time is shown. Before inversion the friction factor at 2 m (p2) and at 5 m (p3) downstream of the injector are the same. So the flow is fully developed already at 2 m downstream from the injector. At 270 s after the start of oil injection the creation of oil-continuous pockets causes local regions with a higher effective viscosity and (as with the conductivity) oscillations in the friction factor occur. With further injection oil-continuous pockets in water-continuous regions and water-continuous pockets in oil-continuous regions become about the same in number and size, causing a high effective viscosity (as can be seen from the increase of the friction factor) and large pressure fluctuations (and hence large fluctuations of the friction factor). At about 320 s after inversion the friction factor decreases and it seems as if the phase inversion process has been completed. However, upstream of this large inverted region there is still a large non-inverted region (causing a high pressure and conductivity peak at about 320 s). It takes another passage through the injector (at this speed it takes the mixture 15 s for a complete cycle through the pipe loop) with additional oil injection to cause a phase inversion also in that region before the phase inversion process is completed for the total pipe flow.

3.3.4 Critical concentration (dispersed phase volume fraction at inversion)

We have performed water-to-oil experiments and oil-to-water experiments for different mixture velocities. Water-to-oil experiments were carried out at mixture velocities of 1 m/s, 2 m/s and 3 m/s, and an oil-to-water experiment only at a mixture velocity of 3.5 m/s. (As oil is more viscous than water, the flow was for the oil-to-water experiment only at a velocity of 3.5 m/s sufficiently turbulent to make the results independent of the injection method.) For each mixture velocity it was found, that the volume fraction of the dispersed phase at inversion (the critical concentration)
3.3. Results for continuous experiments

Figure 3.8: Friction factor and dimensionless conductivity for a water-to-oil experiment at 1 m/s and injection phase volume fraction of 0.125 as a function of time.
increases with decreasing injection phase volume fraction $\chi$ (see Fig. 3.9 and Piela et al. (2006)). This is likely due to the fact that with decreasing $\chi$ the flow is less disturbed and phase inversion is postponed to higher values of the dispersed phase volume fraction. For both cases (water-to-oil) and (oil-to-water) the critical concentration depends almost linearly on $\chi$. Only for oil-to-water experiments at very low injection rates $\chi$ the inversion occurs at a somewhat higher oil volume fraction than expected based on a linear relationship. That is likely due to the creation of multiple drops (small droplets of the continuous phase in larger drops of the other phase), which increases the effective dispersed phase volume fraction. At low injection rates there is more time for the development of multiple drops (for more details see Piela et al. (2006) and Tyrode et al. (2003)).

### 3.4 Results for direct experiments

In a direct experiment the oil and water are simultaneously injected by means of two pumps into the pipe at a certain (constant) mixture fraction. Inside the pipe both liquids mix and the flow becomes water-continuous or oil-continuous dependent on the concentration of the two phases. Similar experiments in a horizontal pipe were carried out by Ioannou et al. (2005). They observed that at the point of phase inversion the pressure drop as well as the conductivity were very unstable. Pal (1993) observed a strong increase in effective viscosity at phase inversion during direct experiments in a pipe. The emphasis of our direct experiments was on the change in morphological structures at the point of inversion and how that change influences the pressure drop.
3.4. Results for direct experiments

3.4.1 Phase inversion mechanism

During a direct experiment the phase fractions were kept constant in time. Since the two phases are well mixed the dispersed phase volume fraction in the pipe is equal to the dispersed phase volume fraction at the point of injection at the entrance to the pipe loop. Close to the point where the flow changed from a water-continuous to an oil-continuous flow or vice versa, strong variations in the morphological structures of the dispersion were observed. For a mixture velocity of 2 m/s these changes occur at oil volume fractions between 0.5 and 0.6. Figure 3.10 shows the dispersion morphology for three different values of the oil volume fraction. The top pictures show the dispersion at an oil volume fraction of 0.5. In the left top picture water is the continuous phase with (multiple) oil drops and oil pockets. In the right top picture there is an oil-continuous region at the top with water drops, and a water-continuous region at the bottom with (multiple) oil drops and oil pockets. At this oil volume fraction of 0.5 most of the flow is water continuous with some oil-continuous regions. At an oil concentration of 0.56 (see bottom-left picture) the flow consists of many (water-continuous) pockets in oil-continuous regions (dark regions), and (multiple) oil drops in water-continuous regions (see, for instance, center part of the picture). At a still higher concentration of 0.58 (see bottom-right picture) most of the flow field is oil continuous.

There is an important difference between a continuous experiment and a direct experiment. In a continuous experiment the inversion process starts with the coalescence of drops leading to the formation of larger drops, pockets and regions by encapsulation of parts of the continuous phase. Finally this process causes the disappearance of regions of the originally continuous phase. In a direct experiment the two liquids are mixed from the start at a constant concentration of the phases and there is no preliminary structure. Depending on the concentration, the liquids mix as a water-continuous mixture or an oil-continuous one. However, at a concentration of about 0.5-0.6 both water-continuous and oil-continuous regions are created during the same experiment (because of the non-homogeneous mixing, in particular in the T-junction and the entrance region of the pipe). At the concentration of around 0.5-0.6 none of the regions is sufficiently strong to dominate the other one, and so both regions flow downstream. These regions interact, entrap parts of the other continuous phase, break-up and coalescence (see section Appendix A) causing a large pressure gradient over the pipe.

3.4.2 Friction factor and conductivity

At a low concentration of either of the two phases the friction factor and conductivity are for a direct experiment the same as for a continuous experiment. However, for a direct experiment with a concentration of around 0.5 for both phases an increase in the friction factor was observed and the conductivity started to oscillate. As discussed at such conditions strong interactions between oil-continuous regions and water-continuous regions take place, which cause a significant increase in the effective viscosity of the mixture (as can be concluded from the strong increase of the friction factor). The friction factor, dimensionless conductivity and oil volume fraction for a direct experiment at a mixture velocity of 3 m/s and an average oil volume fraction of 0.52 are shown in Fig. 3.11. The friction factor was determined for 6 different positions downstream.
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Figure 3.10: Direct experiments at a mixture velocity 2 m/s. Pictures taken at different concentrations. Top-left and top-right pictures are taken at an oil volume fraction of 0.5, the bottom-left picture at 0.56 and the bottom-right picture at 0.58.
3.4. Results for direct experiments

Figure 3.11: Friction factor, dimensionless conductivity and oil volume fraction for direct experiment at mixture velocity of 3 m/s and an averaged oil volume fraction of 0.52. The time is shifted by using the average mixture velocity.

of the injector. For each downstream location the time is again shifted to the time at the inlet by using the mixture velocity. Fig. 3.11 shows that just after the inlet (p1) the friction factor is nearly constant, as during the experiment the two liquids entered the pipe as separate layers and mixing of these layers still had to take place. 2 m downstream (p2) the friction factor is increasing, because the flow was developing and mixing had started. 5 m downstream (p3) the friction factor is already much higher and peaks are visible. Still further downstream (p4, p5 and p6) the peaks remain nearly the same. An interesting point is, that the peaks in the friction factor coincide with steep gradients of the conductivity. As can be seen in Fig. 3.11 the conductivity is changing because the oil volume fraction is not constant during the experiment (bottom graph on Fig.3.11). The changing pressure in the inlet causes small oscillations in the oil volume fraction (the flow rate through the pumps is not completely stable). When the oil volume fraction is lower than 0.52, there are more water-continuous regions in the flow (conductivity value is closer to conductivity of water continuous flow). When the oil volume fraction is higher, there are more oil-continuous regions.
3.4.3 Critical concentration

It is hard to decide, what the critical concentration is in a direct experiment. At values of the oil volume fraction between 0.5 and 0.6 the flow is unstable, which means that oil-continuous regions and water-continuous regions coexist and interact flowing downstream. Fig. 3.12 shows the average (averaged over at least 40 s) friction factor for the three locations at p4, p5 and p6 for mixture velocities of 2 m/s and 3 m/s. It can be seen that at low and high oil volume fractions the friction factor is small and equal to the friction factor of the single phase (0.024 for pure water and 0.035 for pure oil at 2 m/s). The (average) friction factor has a maximum around an oil volume fraction of about 0.54. At this point the interaction between oil-continuous regions and water-continuous regions is strongest (effective dispersed phase fraction is highest, hence effective viscosity has a maximum). The width of the curve for the friction factor as function of the oil volume fraction seems more narrow for a mixture velocity of 3 m/s than for 2 m/s. The reason is likely, that at a higher velocity the interaction between oil continuous and water continuous regions is stronger. The mixing process is faster. So the strong increase in friction factor during this development period takes place over a shorter length of the pipe and the average friction factor over the complete length of the pipe is lower.

For a direct experiment one could argue, that there is no phase inversion comparable to the phase inversion in a continuous experiment. In a continuous experiments there is a constant injection of one of the phases. This causes an increase in the number of drops of the injected phase, which leads to the formation of pockets and regions of the injected phase due to coalescence. These pockets and regions of the injected phase start to interact with pockets and regions of the other phase, and with a further increase in the concentration of the injected phase inversion occurs. So the constant injection of one of the phases is crucial for phase inversion. In a direct experiment at an oil volume fraction between 0.5 and 0.6 water-continuous regions and oil-continuous regions are directly formed due to mixing of the phases downstream of the entrance to the pipe. So certain parts of the mixture flow are oil continuous and other parts water continuous. Below this critical concentration region between 0.5 and 0.6 the flow is always water continuous and above it the flow is always oil continuous (the measured conductivity does not fluctuate). The mixing of the two phases is crucial. However, in the concentration region between 0.5 and 0.6 the interactions between water-continuous regions and oil-continuous regions (leading to high friction factors and oscillations in conductivity) are very similar to the ones observed for a continuous experiment.

3.5 Drop interactions during phase inversion process

In the preceding paragraphs we have given results for two types of experiments: the continuous experiment and the direct experiment. During both experiments there is a transition between a water-continuous phase and an oil-continuous phase (or vice versa). We will loosely call all these transition processes phase inversion, although one could argue that real phase inversion only occurs in a continuous experiment. During phase inversion there is strong interaction between water-continuous regions and oil-continuous regions. These regions have developed due to coalescence of drops and pockets (continuous experiment) and due to turbulent mixing of the
originally separate phases (direct experiment). The interaction of the water-continuous regions and oil-continuous regions lead to a high effective viscosity and hence a large friction factor. In the appendix we discuss in some detail the coalescence -, break-up - and escape processes as observed during the experiments.

3.6 Discussion

As mentioned in the introduction many scientists studied the phase inversion process by means of stirred-vessel experiments. They found, for instance, that an ambivalence region exists. Dependent on the experimental conditions (turbulence level, presence of a surfactant, etc.) the flow can be oil continuous or water continuous (Tidhar et al. (1986); Arashmid and Jeffreys (1980); Deshpande and Kumar (2003)). So the critical concentration at which phase inversion takes place, depends then on the experimental conditions.

In our pipe flow experiments we also paid attention to the region between the maximum and minimum value of the dispersed phase fraction where phase inversion can take place (for a certain set of conditions). During the continuous experiments we found that the critical volume fraction of the dispersed phase at the point of inversion is not so much dependent on \( Re, Fr, We \) and also not on the injection velocity of the dispersed phase, as long as the mixture velocity is sufficiently large \((\geq 2\, \text{m/s})\). However, we found that the injection phase volume fraction \( \chi \) had a significant influence on the critical concentration. This makes it possible to represent the critical volume fraction as measured during the experiments as function of \( \chi \) (see Figure 3.13). The upper line gives the oil volume fraction at the point of phase inversion as measured during water-to-oil continuous experiments; the lower line represents the oil volume fraction at inversion measured during oil-to-water continuous experiments. The region in between the two lines is the ambivalence region, where both oil and water can be the continuous phase (depending on the ‘history’ of the experiment). As can be seen the width of the ambivalence region decreases with increasing injection phase volume fraction \( \chi \). The direct experiment can be considered as the limiting case
Chapter 3. On the phase inversion process in an oil-water pipe flow

Figure 3.13: Inversion map. The upper line gives the oil volume fraction at the point of inversion as measured during water-to-oil continuous experiments. The lower lines represent the oil volume fraction at inversion as measured during oil-to-water continuous experiments. The region in between the two lines is the ambivalence region, where both oil and water can be the continuous phase. Also the oil volume fraction at inversion as measured during the direct experiments is indicated.

of a continuous experiment, with a large injection rate of the dispersed phase and with inversion taking place before one cycle through the pipe loop is completed. The injected phase volume fraction is then equal to the ratio of the injected volume rate of one of the two phases to the volume rate of the mixture. The phase with the lowest volume rate is considered as the injected phase. As can be seen in Figure 3.13 with this interpretation the measured critical concentration at inversion for the direct experiment fits well with the results of the continuous experiments.

As indicated before during the oil-to-water experiments the formation of multiple drops (oil droplets in water drops in a continuous phase of oil) was found. Due to the inclusion of water droplets in the oil drops the effective dispersed phase volume fraction is larger than as calculated starting from the assumption that only pure oil drops are present. This effect explains the deviation of the measured critical volume fraction for the oil-to-water experiments at low values of $\chi$. (This effect was found to be unimportant for water-to-oil experiments. Multiple drops consisting of water droplets in oil drops in a continuous phase of water were very unstable and did not live long.)

3.7 Conclusions

An important conclusion of our work is, that also for pipe flows the critical concentration can be significantly higher for continuous experiments than for direct experiments. By careful injection of the dispersed phase during continuous experiments phase inversion can be postponed to very high values of the dispersed phase volume fraction. For practical applications this result is important, as it opens the opportunity to avoid or postpone phase inversion (causing a high pressure drop or low flow rate).

Another conclusion of our work is that although the critical volume fraction of the dispersed phase at phase inversion was found to be very different for the two types of experiments, the
change in morphological structures was always the same. Far away from the point of inversion the dispersed phase consists of (nearly) spherical drops. However close to inversion the situation changes drastically. Larger morphological structures appear in the form of pockets and regions of the dispersed phase. With increasing dispersed phase volume fraction the concentration of these isolated pockets and regions of the (originally) dispersed phase becomes so high, that they coalesce at certain places in the flow field where isolated pockets and regions of the (originally) continuous phase are formed. With a further increase in volume fraction of the (originally) dispersed phase the balance between the two types of pockets and regions changes in such a way, that the (originally) dispersed phase becomes the continuous one and the (originally) continuous phase the dispersed one. With a still further increase in volume fraction the dispersed phase consists again of (nearly) spherical drops.

3.8 Appendix. Drop interactions during phase inversion process

During our experiments we observed three types of drop interactions: coalescence, break-up and escape.

- Coalescence occurs when two units (drops, pockets or regions) stay in contact for a sufficiently long time. The film between them ruptures and a larger unit is created. Fig. 3.14 shows two examples of coalescence. On the left-hand side a large (multiple) drop and a region can be observed (top-left picture). After 0.01 s the multiple drop and region collide (center-left picture). After only 0.016 s (bottom-left picture) they have merged and a new surface is created. Remains of the multiple drop are now present inside the region. On the right-hand side an elongated region collides with another region. The top-right picture shows them before collision. The center-right picture was taken after 0.004 s and the regions have already almost completely merged. The bottom-left picture shows the situation after 0.008 s. Another form of coalescence between two regions occurs, when a part of one of the regions is entrapped in form of a drop in the other region. Fig. 3.15 shows two examples of entrapment. On the left side a water-continuous region is enclosed between two oil-continuous regions (top-left picture). The water continuous region elongates (second picture from the top on the left-hand side, taken after 0.004 s) and becomes eventually so thin, that coalescence between the two oil continuous regions occurs (third picture from the top on the left-hand side, taken after 0.006 s). That leads also to entrapment of part of the water-continuous region inside the new oil-continuous region (bottom-left picture, taken after 0.016 s). On the right-hand side of Fig. 3.15 the process of entrapment of a part of a water-continuous region inside an oil-continuous region is shown. After 0.004 s (second picture from the top on the right-hand side) the thickness of the water film is decreasing. After 0.006 s (third picture from the top on the right-hand side) it breaks and merging of the two oil regions occurs. And after 0.01 s (bottom-right picture) part of the water-continuous region is entrapped as a water pocket inside the new extended oil-continuous region.
Chapter 3. On the phase inversion process in an oil-water pipe flow

Figure 3.14: Two examples of coalescence between a multiple drop and a region.

Figure 3.15: Two examples of coalescence which leads to entrapment.
3.8. Appendix. Drop interactions during phase inversion process

- Break-up occurs when the fluctuating hydrodynamic force on the surface of a drop or pocket exceeds the surface tension. Fig. 3.16 shows the break-up of two pockets. The top picture shows two elongated pockets. The pocket on the right-hand side breaks after 0.004 s. The pocket on the left-hand side is further elongated and breaks after 0.008 s.

- Escape occurs when the film between an entrapped drop or pocket present inside a certain region and another outside region breaks. Fig. 3.17 shows the escape of a multiple drop into an outside region. The top-left picture shows the structure before the escape process starts. After 0.002 s (bottom-left picture) the film ruptures. After 0.004 s (top-right picture) droplets originally present inside the multiple drop have been released into the outside region. After 0.006 s (bottom-right picture) also the surface of the inner region is adapting to the new condition.
Figure 3.17: Escape of the multiple drop from the region.
In the preceding chapter the inversion map was introduced. This map is specific for a certain oil-water mixture. When a surfactant is added it will change. In the coming chapter we report about phase inversion experiments with a surfactant added to the oil-water mixture and study the influence on the inversion map.
Chapter 4

Influence of a surfactant or salt on phase inversion in a water-oil pipe flow

Phase inversion experiments have been performed on a water-oil flow through a pipeloopt to study the effect of the addition of a surfactant or of salt on the critical dispersed phase volume fraction (at the point of phase inversion). The addition of a surfactant caused a small change in the critical volume fraction, but the general form of the ambivalence region in the inversion map remained the same. The influence of salt was negligible.

4.1 Introduction

The flow of two immiscible liquids often occurs as a dispersed flow, where one liquid is present in the other liquid in the form of drops. Dispersions are widely used in the petrochemical-, food-, chemical- and pharmaceutical industries. Handling and controlling dispersion properties is of a key interest for practical applications. Special interest has to be given to the phase inversion phenomenon, whereby the dispersed phase becomes the continuous one, and vice versa. During this process the effective viscosity of the mixture becomes very large, which leads to a high pressure drop or a low flow rate. The phase inversion phenomenon has been studied for many years (see, for instance, Becher (2001)), but there are still many questions about the inversion process. Some authors assume that a sudden coalescence of drops can encapsulate parts of the continuous phase, which causes drop formation from the (originally) continuous phase (Yeo et al. (2002a); Brauner and Ullmann (2002)). Others observed the creation of multiple drops (small droplets of the continuous phase inside the drops) prior to inversion (Pacek et al. (1994); Pacek and Nienow (1995); Pal (1993); Sajjadi et al. (2000, 2002, 2003); Liu et al. (2005, 2006)). According to some authors inversion is a rather rapid process, called catastrophic by them, (Dickinson (1981); Smith and Lim (1990); Rondón-Gonzaléz et al. (2006); Tyrode et al. (2005); Vaessen et al. (1996); Binks and Lumsdon

*K.Piela, E.Djojorahardjo, G.J.M.Koper and G.Ooms. Influence of a surfactant or salt on phase inversion in a water-oil pipe flow. Submitted to Chemical Engineering Research and Design
Two types of phase inversion experiments are reported in the literature: continuous experiments and direct experiments. During a continuous experiment the dispersed phase is gradually added to the continuous phase and so the volume fraction of the dispersed phase increases with time. During a direct experiment the two liquids are mixed from the start at certain values of the phase volume fraction for the two phases, which remain constant with time. In figure 4.1 a sketch is given of a direct and of a continuous experiment. Most of the experiments reported in the literature were performed in a stirred vessel and usually water and oil were used. For continuous experiments in a stirred vessel it was found, that phase inversion could be postponed to a high value (> 0.8) of the dispersed phase volume fraction. Also a wide ambivalence region for the dispersed phase volume fraction existed, where the mixture could be either water continuous or oil continuous (Vaessen et al. (1996); Groeneweg et al. (1998); Deshpande and Kumar (2003); Mira et al. (2003); Tyrode et al. (2003)). During direct experiments in a stirred vessel inversion usually occurred at a value of the dispersed phase volume fraction close to 0.5 (dependent on the properties of the liquids) and no ambivalence region was observed (Quinn and Sigloh (1963); Tyrode et al. (2005)). Similar results were found by Angelia and Hewitt (1999); Pal (1993); Ioannou et al. (2005) for direct experiments in a pipe flow.

We carried out continuous and direct experiments in a pipe loop (for details see Piela et al. (2006, 2008)). In our experiments we paid attention to the region between the maximum and minimum value of the dispersed phase volume fraction where phase inversion can take place (for a certain set of conditions). During the continuous experiments we found that the critical volume fraction of the dispersed phase at the point of inversion is not dependent on the Reynolds number, Froude number and Weber number of the mixture flow and also not on the injection velocity of the dispersed phase, as long as the mixture velocity is sufficiently large ($\geq 2m/s$). However, we found that the injection phase volume fraction (ratio of the rate of injection of the dispersed phase and the rate of the mixture flow through the pipe) had a significant influence on the critical volume fraction. This made it possible to develop an inversion map, in which the critical volume fraction as measured during the experiments is plotted as function of the injection phase volume fraction (see figure 4.2). The upper line gives the oil volume fraction at the point of phase inversion as measured during water-to-oil continuous experiments; the lower line represents the oil volume fraction at inversion measured during oil-to-water continuous experiments. The region
4.2 Influence of surfactants

Figure 4.2: Inversion map. The upper line gives the oil volume fraction at the point of inversion as measured during water-to-oil continuous experiments. The lower lines represent the oil volume fraction at inversion as measured during oil-to-water continuous experiments. The region in between the two lines is the ambivalence region, where both oil and water can be the continuous phase. Also the oil volume fraction at inversion as measured during the direct experiments is indicated.

in between the two lines is the ambivalence region, where both oil and water can be the continuous phase (depending on the 'history' of the experiment). As can be seen the width of the ambivalence region decreases with increasing injection phase volume fraction. The direct experiment can be considered as the limiting case of a continuous experiment, with a large injection rate of the dispersed phase and with inversion taking place before one cycle through the pipe loop is completed. The injected phase volume fraction is then equal to the ratio of the rate of injection of one of the two phases and the rate of the mixture flow through the pipeloop. The phase with the lowest injection rate is considered as the injected dispersed phase. As can be seen in figure 4.2 with this interpretation the measured critical volume fraction at inversion for the direct experiments fits well with the results of the continuous experiments. All our experiments discussed above were conducted with the same water (tap water) and oil (Shell Macron EDM 110). The chemical and physical properties of the liquids were kept constant. In this brief communication we report on experiments where a surfactant or salt was added to the liquids prior to the experiments. The purpose was to study the influence of these additives on the ambivalence region shown in figure 4.2.

4.2 Influence of surfactants

During our previous experiments (Piela et al. (2006, 2008)) no surfactant was added. We noticed, however, that the results of the first experiments (after starting with a clean water tank with fresh tap water and a clean oil tank filled with new oil) showed different results than the later experiments (for which water and oil had been stored together in a settling tank for some time in order to separate them). Figure 4.3 shows the location of the upper line of the inversion map of figure 4.2 as function of time. As can be seen the experimental results change during the first days and remain stable thereafter. Our previous experiments were conducted for more than a
year with the same oil and water and no changes in the results were found after a few days. The 
changes during the first days are likely due to the presence of surface active substances in the 
liquids. The results shown in figure 4.2 are from the stable period.

![Graph showing location of the upper line of the inversion map as a function of time.]

Figure 4.3: Location of the upper line of the inversion map of figure 4.2 as function of time. The location changes 
during the first days (after starting with fresh water and oil) and become stable thereafter.

There are many papers dealing with the influence of surfactants on phase inversion (for a review 
see Salager et al. (2000)). He presents a formulation-composition map for phase inversion, which 
shows how the composition of the mixture at which the dispersion inverts is changing with sur-
factant affinity difference (SAD). Zambrano et al. (2003); Mira et al. (2003); Tyrode et al. (2003); 
Sajjadi et al. (2003); Tyrode et al. (2005); Rondón-González et al. (2006) show how the HLD-
factor (hydrophilic-lipophilic deviation from an optimum formulation) which is a less complex 
concept than SAD, influences phase inversion. When \( HLD < 0 \) the surfactant exhibits a stronger 
affinity to oil than to water, for \( HLD > 0 \) it is the other way around. Tyrode et al. (2003) and 
Tyrode et al. (2005) show that for \( HLD < 0 \) two types of dispersion are possible: a dispersion 
with multiple drops consisting of oil droplets in water drops in oil, and a dispersion with clean oil 
drops in water. When \( HLD > 0 \) there are again two dispersion types: a dispersion with multiple 
drops consisting of water droplets in oil drops in water, and a dispersion with clean water drops 
in oil (see also figure 4.5 of Tyrode et al. (2005)). In our previous experiments we observed the 
following two types of dispersion: a dispersion with multiple drops consisting of oil droplets in 
water drops in oil, and a dispersion with clean oil drops in water. So we assume that for our case 
\( HLD < 0 \).

Hydrophilic-lipophilic deviation (HLD) is a generalized expression which includes not only sur-
factant HLB, but also the influence of salinity, oil nature, etc. HLD is a deviation from the 
optimum formulation of HLB, but one unit of \( HLB \) is not exactly equivalent to one unit of \( HLD \) 
(for more details see Rondón-González et al. (2006)). In our experiments we decided to influ-
ence HLD by adding surfactants and by adding salt. Our aim was to observe the influence on 
the inversion map of figure 4.2. We decided to use a surfactant with a high \( HLB \)-value, which 
increases the \( HLD \) of the system.

Tween 20 was used as a surfactant. The \( HLB \) of Tween 20 is 16.7 according to the manufacturer. 
In the first experiments we added 10 grams of Tween 20 to the tank with 80 liters of oil and 80 
liters of water. The oil and water had been in contact in the tank for a sufficiently long time,
4.2. Influence of surfactants

so that a stable situation was present (see our earlier discussion about this point). After adding the surfactant both liquids were mixed for a long time. Thereafter, a series of experiments was carried out. As during the previous experiments (reported by Piela et al. (2006, 2008)) water-to-oil experiments were performed at a mixture velocity of 2 m/s and different values of the injected phase volume fraction, as well as oil-to-water experiments at a mixture velocity of 3.5 m/s. The results from these experiments are shown on Fig. 4.4. The inversion lines for the systems with surfactant addition are shifted downward slightly with respect to the inversion lines for the case without surfactant addition. The direction of the shift is as expected from Fig. 4.5. As the $HLD$ increases as a result of the surfactant addition, the water-in-oil dispersion becomes more stable and phase inversion is delayed. The oil-in-water dispersion becomes less stable. The reproducibility of the experiments was good. An important outcome of the experiments is, that the form of the ambivalence region in the inversion map remains the same.

The preferred morphology is not changed by the surfactant addition. A dispersion with multiple drops consisting of oil droplets in water drops in oil and a dispersion with clean oil drops in water remained. This implies that the $HLD$-value is still lower than 0. For a change in the preferred morphology this value must be larger than 0. Addition of surfactants influenced separation rate of oil-water mixture. Even a small addition of surfactants created an oil-water layer in the separation tank which was stable for a few hours. Experiments with a higher surfactant concentration in the mixture failed. At a higher surfactant concentration the separation rate of water and oil in the separation tank decreased dramatically. A stable emulsion was created, even after one week no separation was observed. That made impossible to continue experiments in the existing configuration.
4.3 Influence of salt

After the experiments with surfactants the oil-water emulsion was removed from the experimental set-up, the set-up was cleaned and finally filled again with fresh water and oil. A few continuous experiments were carried out in order to achieve the stable condition discussed before. Then a series of phase inversion experiments were performed with salt added to the water. The influence of the salt concentration on the critical oil volume fraction (at phase inversion) was investigated. Salt is changing the chemical properties of the dispersion. Also the density of water is changing due to the salt addition. Results for water-to-oil experiments and oil-to-water experiments at constant injected phase volume fraction are shown in Tab. 4.3. As can be seen salt has no influence on the critical concentration. This means that even a high concentration of salt (8%) does not change the HLD-value sufficiently enough for a significant change in the phase inversion process. Most likely surfactants present in the oil-water mixture are non-ionic and higher concentration of ions do not change the surface conditions. Also the increase in density difference between oil and water from 200 kg/m$^3$ to 285 kg/m$^3$ has no influence. The mixture velocity is still large enough to cause a fully dispersed flow.

4.4 Discussion and conclusions

In our previous experiments we observed a linear dependence of the critical volume fraction (at phase inversion) on the injected phase volume fraction. This result was independent of other flow parameters, such as the Reynolds number, Froude number and Weber number. For this reason it was possible to develop an inversion map, which was unique for the oil-water system used in these previous experiments. However the question remained: Is this inversion map similar for other oil-water systems? To answer this question we carried out new experiments with the same oil and water, but with a surfactant or with salt added to the mixture. The new results showed, that the location of the inversion lines shifted, but the general form of the ambivalence region
## 4.4. Discussion and conclusions

Water-to-oil continuous experiment, 2 m/s, injected phase volume fraction 0.1

<table>
<thead>
<tr>
<th>Salt concentration</th>
<th>Critical oil volume fraction</th>
</tr>
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<tbody>
<tr>
<td>0.0%</td>
<td>0.85</td>
</tr>
<tr>
<td>1.5%</td>
<td>0.83</td>
</tr>
<tr>
<td>3.0%</td>
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</tr>
<tr>
<td>6.0%</td>
<td>0.83</td>
</tr>
<tr>
<td>8.0%</td>
<td>0.84</td>
</tr>
</tbody>
</table>

Oil-to-water continuous experiment, 3.5 m/s, injected phase volume fraction 0.05

<table>
<thead>
<tr>
<th>Salt concentration</th>
<th>Critical oil volume fraction</th>
</tr>
</thead>
<tbody>
<tr>
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<td>0.31</td>
</tr>
<tr>
<td>1.5%</td>
<td>0.29</td>
</tr>
<tr>
<td>3.0%</td>
<td>0.33</td>
</tr>
<tr>
<td>6.0%</td>
<td>0.33</td>
</tr>
<tr>
<td>6.0%</td>
<td>0.32</td>
</tr>
<tr>
<td>8.0%</td>
<td>0.28</td>
</tr>
</tbody>
</table>

Table 4.1: Critical oil volume fraction at phase inversion for water-to-oil continuous experiments at a mixture velocity of 2 m/s (injected phase volume fraction 0.1) and critical oil volume fraction at phase inversion for oil-to-water continuous experiments at a mixture velocity of 3.5 m/s (injected phase volume fraction 0.05) as a function of the salt concentration.

In the inversion map remained the same. The critical concentration is still linearly dependent on the injected phase volume fraction and independent of the other flow parameters. This is an important conclusion for comparing and predicting phase inversion experiments. Because of practical reasons we were not able to change the sign of the $HLD$-factor from negative to positive to study the influence of the $HLD$-factor on the preferred morphology of the suspension.
The previous chapters gave experimental results from continuous and direct experiments. The next chapter presents the results of a different type of experiment: the discontinuous experiment. In a discontinuous experiment a pure water phase is pumped through the pipe, and at a certain moment it is changed to a pure oil phase (or vice versa). Somewhere in the mixing zone between the two liquids phase inversion will occur similarly to, for instance, the direct experiments. The purpose of this chapter is to report about the results of these experiments and to see whether the observed morphological structures are identical to the ones found in the continuous and direct experiments.
Chapter 5

Phase inversion in the mixing zone between a water flow and an oil flow through a pipe

5.1 Introduction

The flow of two immiscible liquids often occurs as a dispersed flow, where one liquid is present in the other one in the form of drops. Water-oil dispersions can occur as oil drops in a water-continuous phase or as water drops in an oil-continuous phase. Phase inversion is the phenomenon by which the dispersed phase becomes the continuous one, and vice versa. The phase inversion phenomenon has been studied for many years (see, for instance Becher (2001)). Recently we (see Piela et al. (2006, 2008)) carried out continuous experiments and direct experiments to study phase inversion in a pipe flow. During the continuous experiments we started with the flow of one of the liquids and gradually injected the other liquid, while keeping the mixture velocity constant. During the direct experiments the two liquids are injected from the start simultaneously into the pipe with certain concentrations. Detailed pictures were taken of the phase inversion process and also the electrical conductivity of the mixture was measured to determine which liquid formed the continuous phase and which the dispersed phase. Although the concentration at inversion was significantly higher for continuous experiments than for direct experiments, the change in morphological structures during phase inversion was the same for the two types of experiments. At inversion the concentration of drops of the (originally) dispersed phase becomes so high, that they coalesce at certain places in the flow field and form relatively large, rather complex, morphological structures. With a further increase in concentration of the (originally) dispersed phase these morphological structures grow in size and start to form the new continuous phase in which again complex structures are present, but this time consisting of the (originally) continuous phase.

Recently we realized that there is still another type experiment possible during which phase inversion takes place: discontinuous experiments. In a discontinuous experiment a pure water phase is pumped through the pipe, and at a certain moment it is changed to a pure oil phase (or

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vice versa). Somewhere in the mixing zone between the two liquids phase inversion will occur similarly to, for instance, the direct experiments. The purpose of this brief communication is to report about the results of these experiments and to see whether the observed morphological structures are identical to the ones found in the continuous and direct experiments.

In section 5.2 of this communication the experimental facility used for the experiment as well as the visualization method are described. Then results found during water-to-oil and oil-to-water discontinuous experiments are presented in section 5.3. Finally, in section 5.4 some conclusions are drawn.

5.2 Experimental set-up

A sketch of the pipe facility used for the experiment is shown in Fig. 5.1. An acrylic pipe with an inner pipe diameter of 16 mm was used. All the experiments were carried out at such a high mixture velocity, that a fully developed dispersed flow was present in the pipe. We realize that the results of these experiments are likely not representative for dispersed flow in a large diameter pipe, as wall effects have proved to be more significant for inversion behavior in small diameter pipes than in large diameter ones. (It was shown by Ioannou et al. (2005) that the wetting properties of the pipe wall can influence the concentration at which phase inversion occurs.) Moreover, in a small-diameter pipe what happens near the wall is quickly transported to the centre of the pipe. Furthermore, in a large diameter pipe a higher mixture velocity is required for fully developed dispersed flow than in a small diameter one (see Brauner (2001)).

The two immiscible liquids were tap water and Shell Macron EDM 110 oil (density 794 kg/m$^3$, kinematic viscosity at $20^\circ$ 3.9 mm$^2$/s and oil water interfacial tension 0.045 N/m). An experiment started by injecting one liquid (water or oil) by means of pump 1 (see Fig. 5.1) into the pipe. After several seconds the injection of the first liquid was stopped and the other liquid was injected by turning valve 1. Because pump 1 is a positive displacement pump, the second liquid was injected at nearly the same volume flow rate. Pump 1 was controlled by the feedback system from flow meter F1, which measured the density and mass flow rate (KROHNE OPTIMASS 7000, error < 0.26%). Pressure drops were measured at different locations over a distance of 1 m. Differential pressure transducers (Validyne DP-15, error < 3%) were used and the pressure signal was sampled at a rate of 2 kHz. Data were averaged over 2000 samples. The pressure drops were measured at three different positions: 8.5 m (530 d), 15.2 m (950 d) and 22.2 m (1390 d) from the inlet valve V1 (d is the pipe diameter). So the distance between the pressure drop measurement points p1 and p2 is 420 d and between p2 and p3 is 440 d. The pressure taps were located upstream of the bends. They were also rather close to the bends (see Fig. 5.1) to ensure a long development length for the flow before the measurements were taken. We found no significant influence of the pipe bends (downstream of the pressure taps) on the results.

Conductivity measurements (the signal was also sampled at a rate of 2 kHz) were made with a cell consisting of two (0.2 mm diameter) wire electrodes mounted in the pipe: one in the vertical direction and one in the horizontal direction. The distance between the electrodes in the center of the pipe was 2 mm. During the experiment we also monitored the temperature. The experiments were performed more than once and the repeatability of the experiments was good.
To make a detailed study of the dispersion morphology samples were taken from the flow by means of a 7 mm inner diameter sampling tube at a distance of 25.5 m downstream of the inlet valve V1 (see Fig. 5.1) and led through a visualization cell (two glass windows that were 1 mm apart). The visualization cell was illuminated from one side with a 500 W halogen lamp and a high speed camera took images at the other side. The camera was operated at a frame rate of 50 Hz. A sketch of the sampling technique is given in Fig. 5.2. To check whether this observation procedure had some influence on the dispersion morphology we performed experiments with various distances between the visualization cell and the pipe. We always achieved the same results. We also changed the distance between the glass walls from 1 mm to 5 mm and again the results were similar. Moreover in our earlier studies (see Piela et al. (2006, 2008)) we compared the results with non-intrusive observations from the side of the pipe. The results were in agreement with those from the visualization cell.
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5.3 Results

During the study of the mixing zone between the two liquids three types of morphological structures were observed (similarly to the continuous and direct experiments): drops, pockets and regions. A drop is the smallest part of the dispersed phase, usually smaller than 1 mm. A pocket is a larger unit of one of the two phases, that contains several drops of the other phase; it is usually of the order of a few millimeters up to 1 cm. Finally a region is a still larger part of one of the two phases in the flow field, that encloses several pockets and drops of the other phase and is of order of 1 cm or larger. At some conditions also multiple drops were observed, which are drops containing small droplets of the other (continuous) phase. A pocket is larger than a multiple drop and the surface tension is not strong enough to give a pocket a spherical shape, whereas a multiple drop is (nearly) spherical.

5.3.1 Observations during experiments

We started with pumping, for instance, water at a constant velocity through the pipe and at a certain moment (by switching the manual valve) oil was pumped at the same superficial velocity. The valve is constructed in a such a way that a flow of pure water is followed by a flow of pure oil, or vice versa. Of course some disturbance is created by switching the valve. To study possible inlet effects on the mixing zone we repeated the experiments several times and at different velocities. The results were always the same. So the inlet effects are negligible. In the first part of the mixing zone the flow was water continuous (with oil drops) and in the second part the flow was oil continuous with water drops. Between these two parts there is a zone where both water-continuous regions and oil-continuous regions coexist next to each other. Some results for the morphological structures inside this mixing zone are given in Fig. 5.3. It shows some pictures taken as function of time at a certain position downstream of the entrance to the pipe. The top-left picture in Fig. 5.3 shows a continuous-water phase with oil drops in the first part of the mixing zone. The center-left picture shows the morphology after 0.06 s (further downstream into the mixing zone, but still in its initial part) and a number of oil pockets has been formed. After 0.16 s (bottom-left picture) there are many oil pockets and (multiple) drops. After 0.38 s (top-right picture) oil-continuous regions occur with water drops and water pockets. The flow is almost completely oil continuous after 0.5 s (center-right picture). Still some larger water regions exist. However, they break-up into smaller drops. After 0.86 s (bottom-left picture) the flow is oil continuous.

Pictures taken during a similar experiment only with oil as the initial phase and water as the final one are shown in Fig. 5.4. Multiple drops (oil droplets inside water drops in a continuous phase of oil) can be observed at the start of the mixing zone (top-left picture). After 0.94 s (center-left picture) the concentration of water drops has increased and larger pockets of water have been formed. The concentration of these water pockets seems lower than the concentration of oil pockets at the same stage of the transition for the water-to-oil discontinuous experiment. After 4.9 s (bottom-left picture) and 5.64 s (top-right picture) the flow consists of both water-continuous regions and oil-continuous regions. At time 6.1s (center-right picture) the flow is water continuous with multiple oil drops. The existence time of these multiple oil drops (water
5.3. Results

Figure 5.3: Transition from a water-continuous flow to an oil-continuous flow during a discontinuous experiment. The flow velocity was kept constant at 1 m/s.

droplets in oil drops in a continuous phase of water) is considerably shorter than the existence time of multiple water drops. Multiple oil drops break-up easily and the water droplets escape from their inside into the continuous water phase. After 8.2 s only pure oil drops in water remain.

5.3.2 Friction factor and conductivity

Before the phase inversion experiments single phase pressure drops were compared with Blasius friction factor values for the pressure drop and they compared well. Thereafter the two-phase experiments started. During phase transition an increase of the friction factor was observed. The friction factor is defined as

\[ f = \frac{2\Delta P d}{\rho u^2 L}, \]

where \( \Delta P \) is the pressure drop over a distance \( L \), \( d \) the pipe diameter, \( \rho \) the density and \( u \) the average mixture velocity. The density \( \rho \) and mixture velocity \( u \) were measured by means of the flow meter F1. (Using the mixture velocity the results could be shifted from the location of the flow meter to the location of the pressure measurement points.) Fig. 5.5 shows the friction factor and conductivity for a water-to-oil discontinuous experiment at a mixture velocity 2 m/s. As can be seen the friction factor is strongly increasing during the passage of the mixing zone. The difference between the friction factors at p1 and p2 suggests that the flow is definitively not fully-developed. So even at p2 (15.2 m downstream of the entrance to the pipe) the flow is still developing. There is also still a noticeable difference between the friction factors at p2 and p3. So the flow appears to be developing all along the pipe. The conductivity measurement shows that in the first part of the mixing zone (between 61s to
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pipe

Figure 5.4: Transition from an oil-continuous flow to a water-continuous flow during a discontinuous experiment. Flow velocity was kept constant at 1 m/s.

64s) a water-continuous dispersion exists. With the passage of the mixing zone the concentration of oil drops increases, which causes a decrease in conductivity. Between 64 s and 64.5 s inversion takes place. Both water-continuous regions and oil-continuous regions coexists. Finally (after 64.5 s) an oil-continuous dispersion is present. The concentration of water drops is decreasing and finally a pure oil flow is observed.

Results from an oil-to-water experiment are presented in Fig. 5.6. The mixture velocity is significantly larger than in the preceding case, viz. 4 m/s. There is again a significant difference between the signals at p1, p2 and p3, although the difference is less pronounced than as shown in Fig. 5.5. So also in this case the flow is still developing. Between 125 s and 126 s there is remarkable decrease in the friction factor (much more pronounced than in the preceding case). This is in agreement with our previous observations (Piela et al. (2006)) as well as with observations made by Ioannou et al. (2005). In that part of the mixing zone oil is still the continuous phase, but the increase in conductivity implies that water-continuous pockets are already present. Between 126 s and 126.3 s both water-continuous and oil-continuous regions coexist. Thereafter a water-continuous dispersion is present, which is finally followed by a pure water flow.

The width of the mixing zone is determined by the initial mixing due to the valve operation and by the subsequent mixing due to the turbulence in the pipe. However from the pressure measurements in Fig. 5.5 and certainly from Fig. 5.6 it can be seen that already after 8.5 m the width of the mixing zone is established. By repeating the experiments we confirmed that the width and the observed structures inside the mixing zone are independent of the exact details of
Figure 5.5: Friction factor and dimensionless conductivity for a discontinuous water-to-oil experiment at a mixture velocity of 2 m/s. For each downstream location the time is shifted to the time at the inlet by using the mixture velocity. In this way it is possible to study the development in time of the mixing zone.

Figure 5.6: Friction factor and dimensionless conductivity for a discontinuous oil-to-water experiment at a mixture velocity of 4 m/s. For each downstream location the time is shifted to the time at the inlet by using the mixture velocity. In this way it is possible to study the development in time of the mixing zone.
the initial condition due to the valve operation.

In Fig. 5.7 the conductivity as measured during water-to-oil discontinuous experiments at 1, 2 and 4 m/s is given. This figure shows the surprising result, that the length of the mixing zone (found by multiplying the mixture velocity with the time duration of the mixing zone) is constant (about 6 m). (Although the part of the trace with a steep gradient shows about 1 m length for 2 m/s and 2.5 m length for 4 m/s.) The same length is found for oil-to-water discontinuous experiments. So, obviously, the length of the mixing zone seems independent of the location in the pipe, independent of the mixture velocity and of the type of experiment (water-to-oil or oil-to-water).

We are not certain about the explanation for this independence of the mixing length zone from (some of) the flow conditions, although we checked experimentally that this length is not influenced by the valve operation. We do not know, how general this result is and whether the mixing length is perhaps dependent on other flow conditions. The fact that the mixing length is independent of the mixture velocity indicates, that the time necessary for inversion is not constant and decreases with increasing velocity. This points out in the direction, that the length of the mixing zone is determined by the details of the mixing process of water and oil in this zone. With increasing velocity the (turbulent) mixing of (multiple) drops, pockets and regions will increase and likely cause a decrease of the time needed for the inversion process. It is clear, that this point needs further study.

5.4 Conclusion

The simple discontinuous experiments have given us much insight in the mixing and phase inversion process. The distinction between the different stages of the processes was easy. The
5.4. Conclusion

observations made in the mixing zone of discontinuous experiments confirmed the existence of (multiple) drops, pockets and regions as also found during continuous and direct experiments. With the passage of the mixing zone the concentration of drops (of the last-injected liquid in the continuous first-injected liquid) becomes so high, that they coalesce at certain places in the flow field and form relatively large, rather complex, morphological structures. With a further increase in concentration of the last-injected liquid these morphological structures grow in size and start to form the new continuous phase in which again complex structures are present, but this time consisting of the first-injected liquid. During the passage of the mixing zone the local friction factor increases very considerably.
In the following chapter a model is developed to predict the inversion map for an oil-water pipe flow. It is based on the minimum dissipation assumption. The model is compared with our experimental results.
Chapter 6

Interpretation of phase inversion in liquid-liquid flows by minimal dissipation rate approach*

Phase inversion refers to the phenomenon whereby a small change in operational flow conditions causes an oil-in-water dispersed flow pattern to suddenly switch to a water-in-oil flow pattern, and vice versa. This paper proposes an interpretation of phase inversion in terms of minimal dissipation rate. To this end, the dissipation rate is computed by a simple homogeneous model together with available correlations for effective viscosity in dispersed flows. It is shown that the data available in the literature can be reasonably interpreted as a manifestation of minimal dissipation rate. Furthermore, if the assumed effective viscosity correlations take into account pipe wettability, the minimal dissipation rate approach is capable to interpret also the so-called ambivalent range (hysteresis effect) and correlate the available data.

6.1 Introduction

The flow of two immiscible liquids is often encountered in industrial applications such as food, pharmaceutical, and oil industries. The possibility to predict the flow characteristics – such as flow pattern or pressure drop – is essential both to design and to maintain industrial facilities. Dispersed flow is a common flow pattern which consists of a continuous phase in which the other phase is dispersed in the form of droplets, Brauner (1998). A water-in-oil dispersion ($D_{w/o}$) is a dispersion of aqueous phase in the organic phase. Conversely, oil-in-water dispersion ($D_{o/w}$) is a dispersion of organic phase in the aqueous phase. At a particular value of the hold-up $\varepsilon$ – *in situ* oil or water fraction – phase inversion occurs; phase inversion refers to the phenomenon where, with a small change in the operational conditions, the continuous phase and the discrete phase invert. So, for instance, a water-in-oil dispersion ($D_{w/o}$) suddenly switches into an oil-in-water dispersion ($D_{o/w}$).

dispersion ($D_{o/w}$).

Phase inversion plays a major role in the design of industrial equipment since it affects mass, momentum, and heat transfer efficiency and hence it needs to be accounted for. For instance in petroleum industry where water and oil are frequently transported together, the flow pattern strongly affects the pressure drop; furthermore, corrosion, and hence inhibition agents injection, strongly depends on the wetting phase. In settling-tanks, where the settling time depends on the viscosity of the continuous phase, the correct prediction of the flow pattern ($D_{w/o}$ or $D_{o/w}$) proves essential for a proper design of the process. In petrochemical plants, where for the refinery process oil-water mixtures pass through heat exchangers, the knowledge of which phase is the continuous one is essential for a correct prediction of the heat transfer.

Studies on phase inversion have been carried out in stirred tanks, batch mixers, continuous mixers, and pipe flows (see Yeh et al. (1964) and references therein) in the attempt to establish a relation that allows to predict the hold-up at phase inversion (i.e., the critical hold-up, $\varepsilon^I$) as a function of the fluid properties (viscosity ratio, material wettability, and surface tension). In this paper we focus our attention on phase inversion in pipe flow. However the approach we propose has a broader validity and can be easily extended and applied to different geometries such as stirred vessels.

Although phase inversion has been a problem for more than forty years, little is known about it and, for the design of industrial equipment, engineers have relied mainly on empirical correlations. For instance, to compute the critical water hold-up $\varepsilon^I_w$ at which phase inversion takes place in oil-water flow, Arirachakaran et al. (1989) proposed, as a best fit of their experimental data, the following relation

$$\varepsilon^I_w = 0.5 - 0.1088 \log_{10} \left( \frac{\mu_o}{\mu_r} \right),$$

(6.1)

where $\mu_r = 1$ mPa·s. Equation 6.1 allows to estimate the critical water fraction $\varepsilon^I_w$ at which phase inversion occurs as a function of the oil viscosity $\mu_o$. Another empirical correlation to compute the critical water hold-up $\varepsilon^I_w$ was suggested by N"adler and Mewes (1997)

$$\varepsilon^I_w = \frac{1}{1 + k_1 \left[ \frac{C_w \rho_o^{1 - n_o} \mu_o}{C_w \rho_u^{1 - n_u} \mu_u} \right] \left( DU_m \right)^{n_u - n_o} \left( DU_m \right)^{1/k_2}}$$

(6.2)

where $D$ is the pipe diameter, $\rho_{o,w}$ is the oil or water density, $\mu_{o,w}$ the oil or water viscosity, $U_m$ the mixture velocity; $C_o$, $C_w$, $n_o$, and $n_w$ are the parameters of the friction factor correlation $C Re^{-n}$ for pure oil and pure water pipe flow; $k_1$ and $k_2$ are empirical parameters. All the details on the application of Blasius-like relations to two-phase flows can be found in Brauner and Ullmann (2002).

Relations 6.1 and 6.2 provide a single value of the hold-up at which inversion takes place once the fluids’ properties are known. However, it is found experimentally that a so-called ambivalent range of hold-up values exists, in which both phases may be found to be continuous. Only outside this range one phase is always continuous and the other is always discrete. Within the ambivalent range, either one of the two phases can be continuous and the exact value at which phases invert depends on the operating conditions, on surface tension, and on pipe wettability. If the prediction
of ambivalent range is neglected, the critical hold-up depends mainly on the viscosity ratio of the two fluids. Even if empirical correlations are widely used, some attempts to predict theoretically the critical hold-up for phase inversion have been carried out. Several mechanisms have been postulated to predict both phase inversion and the existence of the ambivalent range. The most important are based on

- instability between droplets coalescence and break-up;
- minimization of the total free energy content (including both gravitational potential energy and interfacial energy);
- effects of dynamic forces.

Phase inversion was explained as an instability between break-up and coalescence of dispersed drops. Phase inversion takes place when there is a large coalescence frequency and the break-up mechanism is insufficiently fast. Up to a certain volume fraction, break-up and coalescence processes can reach a dynamical balance; near the inversion point, instead, this balance cannot be sustained anymore and the coalescence rate overcomes the break-up rate. Droplets grow and their shapes change from spherical to cylindrical to lamellae and, eventually, a complex structure is reached, leading to the formation of the new continuous phase. Recently, detailed Laser Induced Fluorescence (LIF) experiments have shown the appearance of a complex flow structure at the inversion point. The presence of secondary dispersions is observed both in stirred vessels – see Liu et al. (2005) – and in pipe flow – Piela et al. (2006). Experiments – Yeo et al. (2000) – have shown that all the factors that affect droplet break-up/coalescence rate (liquid properties, electrostatic interactions, wettability, and shear rate) influence the phase inversion as well as the ambivalent range. So, phase inversion can be regarded as an instability in the dynamic balance between break-up and coalescence. The ambivalent range corresponds to the difference in the onset of this instability in the aqueous and organic continuous dispersions.

Since phase inversion is a phenomenon that occurs spontaneously, other authors proposed that its prediction can be based on a criterion of minimization of the total energy content, including interfacial energy, of the flowing system. Such an approach was originally proposed by Luhning and Sawistowski (1971) and Tidhar et al. (1986). Recently, following this line, Brauner and Ullmann (2002) proposed a new model to predict phase inversion. According to this model, the critical oil hold-up at which phase inversion occurs can be computed as

$$
\varepsilon'^o = \frac{\frac{\sigma}{d_{32}}_{w/o} + \frac{s}{d_{32}} \cos \theta}{\frac{\sigma}{d_{32}}_{w/o} + \frac{\sigma}{d_{32}}_{o/w}}
$$

(6.3)

where $\theta$ is the water wettability angle, $\sigma$ the oil-water interfacial tension, $d_{32}$ the Sauter mean diameter, and $s$ the wetted perimeter. The prediction of the Sauter diameter $d_{32}$ involves extensive modelling for the break-up/coalescence of droplets. Because in general the application of the minimum free energy approach relies on a model able to characterize the drop size, several attempts have been made in the literature to predict the typical drop size, for example by means
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The minimum energy criterion per se cannot predict the presence of the ambivalent range, nevertheless Yeo et al. (2000) were able to predict it by calculating the drop size using different relations in o/w and w/o dispersions. On the contrary, Brauner and Ullmann (2002) claimed that a possible reason for the hysteresis (ambivalent) range is the time needed after phase inversion for the new continuous phase to completely wet the pipe wall. The understanding and the prediction of the ambivalent range require further investigations, both experimental and theoretical.

Yeh et al. (1964) suggested another mechanism by which dispersed flow can be regarded as the flow of three thin layers: the dispersed phase, the interfacial phase, and the continuous one. Dynamic forces are considered to play a major role in the determination of the dispersion type. Assuming no shear at the interface, they derived the concentration ratio of dispersed to continuous phase at inversion as a function of pure liquid viscosities. Assuming all the layers in the model are laminar, the relation obtained by Yeh et al. can be written as

$$\varepsilon^I_w = \frac{1}{1 + \left(\frac{\mu_o}{\mu_w}\right)^{0.5}}.$$  \hspace{1cm} (6.4)

All the experiments carried out to study phase inversion in pipe flow are made in the so-called direct way, i.e. oil and water are injected simultaneously into the test section at prescribed superficial velocity. The experiments then consisted in observing if the injected emulsion is either water continuous or oil continuous. The empirical relations as well as the theoretical model just presented describe in a reasonable way the phenomenon providing a quite good estimated of the critical hold-up for phase inversion.

Recently, Piela et al. (2006) performed a new type of experiments in pipe flow: the so-called wash-out experiments. In such kind of experiments – already performed in stirred vessels – the pipe is filled with one of the fluids (either oil or water) and then the other is gradually added up to phase inversion. In that case, the experiment consists in the injection of small quantities of oil in a continuous way in order to register at what hold-up phase inversion occurs.

Piela et al. (2006) observed that the critical hold-up for phase inversion in wash-out experiments is much higher than for the direct experiments; the reason for that is that, probably, during wash-out experiments multiple dispersion is formed. Due to multiple drop formation, if the theoretical models are used, the critical hold-up for phase inversion is largely underestimated.

The aim of this paper is to provide an alternative tool to interpret inversion data arising from wash-out experiments. In Sect. 6.2, we propose our interpretation for phase inversion based on minimal energy dissipation rate reasoning. In Sect. 6.3 we interpret the experimental results recently published by Piela et al. (2006) in terms of minimal energy dissipation rate. Conclusions are given in Sect. 6.4.

6.2 Phase inversion model and minimal dissipation rate

In this paper, we want to show that phase inversion can be interpreted in terms of minimal dissipation rate and, accordingly, the flow pattern that develops (i.e., $D_{o/w}$ or $D_{w/o}$) is the one that, for
a given hold-up, minimizes the dissipation rate. As a first step we compute the dissipation rate as a function of hold-up, first assuming the oil to be continuous and the water discrete, and then the converse. Assuming Newtonian flow, the dissipation rate is given in general by the relation

\[ \Phi = 2 \int_{V_1 \cup V_2} \mu s_{ij} s_{ij} dV = 2 \int_{V_1} \mu_1 s_{ij} s_{ij} dV_1 + 2 \int_{V_2} \mu_2 s_{ij} s_{ij} dV_2, \]  

(6.5)

where \( \mu \) is the fluid viscosity and \( s_{ij} \) the shear rate; integration is carried out over the entire two-phase domain \( V_1 \cup V_2 \). In case of pipe flow, the dissipation amounts to

\[ \Phi = \left( \frac{\dot{m}_o}{\rho_o} + \frac{\dot{m}_w}{\rho_w} \right) \Delta p, \]  

(6.6)

where \( \Delta p \) is the pressure drop along the pipe and \( \dot{m} \) are the oil and water mass flow rates respectively. To use Eq. 6.6 it is necessary to estimate the pressure drop \( \Delta p \) where \( \Delta p = L \cdot \frac{dp}{dx} \) and \( L \) is the pipe length. To this end, we use the homogeneous model according to which the pressure gradient \( \frac{dp}{dx} \) can be found as

\[ \frac{dp}{dx} = 2 f_m \rho_m U_m^2 \]  

(6.7)

where \( U_m = 4 (\dot{m}_o/\rho_o + \dot{m}_w/\rho_w)/\pi D^2 \) is the mixture velocity, \( \rho_m = \rho_w \varepsilon_w + \rho_o \varepsilon_o \) is the mean mixture density, \( D \) is the pipe diameter, and \( \varepsilon_w = 1 - \varepsilon_o \). For the mixture friction factor \( f_m \) we assume a Blasius relation

\[ f_m = \frac{C}{Re_m^n}, \]  

(6.8)

where the values of \( C \) and \( n \) depend on the flow regime and the mixture Reynolds number \( Re_m \) is defined by means of an effective viscosity \( \mu_m \),

\[ Re_m = \frac{\rho_m U_m D}{\mu_m}. \]  

(6.9)

If the flow is laminar, i.e. \( Re_m \leq 2300 \), \( C = 64 \) and \( n = 1 \); if the flow is turbulent, i.e. \( Re_m \geq 2300 \), \( C = 0.079 \) and \( n = 0.2 \). Of course, there exist more complex relations to compute \( f_m \) that account for pipe roughness and for the non-Newtonian behaviour of highly concentrated mixtures; here, we restrict our analysis to this simple relation.

For the effective viscosity, \( \mu_m \), of a dispersed system, different relations have been proposed and analysed. For dilute dispersions, the well-known Einstein’s relation can be used; for highly concentrated mixtures, more sophisticated relations need to be used in order to account for droplet-droplet interactions; a collection of these relations is reported by Brauner (1998) and Hu et al. (2005); the simplest relation suitable for highly concentrated mixtures of solid mono-dispersed spheres is proposed by Ball and Richmond (1980)

\[ \mu_m = \mu_c (1 - K \varepsilon_d)^{-5/(2K)}, \]  

(6.10)

where \( \mu_c \) is the continuous phase viscosity, \( \varepsilon_d \) the discrete phase hold-up, and \( 1/K \) can be interpreted as the maximum packing fraction. For a mono-dispersed suspension of rigid spheres the
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Figure 6.1: Example of the application of Eqs. 6.7-6.10 for turbulent flow of oil droplets in water (dashed curve, \(D_o/w\)) and water droplets in oil (dotted curve, \(D_w/o\)). Points represent experimental data: pipe diameter \(D = 20\) mm, water superficial velocity \(2\) m/s, oil superficial velocity \(1.5\) m/s, viscosity ratio \(\tilde{\mu} = 24\), density ratio \(\tilde{\rho} = 0.79\).

maximum packing fraction can be calculated to be 0.58, however it is shown by Pal (1993) that in case of emulsions the maximum packing fraction can be well above 0.58 and up to 0.95. For that reason, in the case of deformable drops, \(1/K\) may be assumed equal to 1.

As already discussed by Poesio and Beretta (2008), relation 6.10 can be modified in order to account for different effects such as pipe wettability and in our case to the formation of multiple drops. The relation that can be proposed is

\[
\mu_m = \mu_c \left(1 - K_1 \varepsilon_d \right)^{-5/(2K_2)},
\]

(6.11)

where the two constants \(K_1\) and \(K_2\) must be found by fitting.

Using Eqs. 6.7-6.11, the pressure drop is computed as a function of the hold-up and shown, for example, in Fig. 6.1. If water is assumed to be the continuous phase for all values of the hold-up, the resulting pressure drop is given by the dashed curve. Similarly, if oil is assumed to be the continuous phase for all values of the hold-up value, the resulting pressure drop is given by the dotted curve. Figure 6.1 shows the two predicted pressure drops compared to the measured values. As long as water is actually the continuous phase, the pressure drop is predicted correctly and the same holds to some approximation when oil is the continuous phase.

Our reading of this result is that the system chooses the flow pattern that, for a given oil hold-up \(\varepsilon_o\), provides the lowest pressure drop, i.e. the minimum energy dissipation. If this is true, as it appears from Fig. 6.1, the value at which the two curves cross each other is the inversion point and the corresponding hold-up \(\varepsilon_o^I\) is the critical value.

So, the construction of Fig. 6.1 involves the following steps:

- the asymptotic behaviours are considered separately; first the pressure drop is computed, using Eqs. 6.7-6.10, under the assumption that an oil-in-water dispersion occurs for all
6.2. Phase inversion model and minimal dissipation rate

values of the oil hold-up (dashed curve). Similarly, the pressure drop is computed assuming that a water-in-oil dispersion obtains for all values of the oil hold-up (dotted line).

- an estimate of the actual pressure drop – the continuous line in Fig. 6.1 – is computed by an asymptotic matching interpolation of the two curves, i.e., the resulting pressure drop is computed by the relation

\[
\left( \frac{dp}{dx} \right)_a^a = \left( \frac{dp}{dx} \right)_{o/w}^a + \left( \frac{dp}{dx} \right)_{w/o}^a,
\]

where the exponent \( a \) was chosen such that \( a = 10 \);

- the value of the hold-up corresponding to the maximum of the interpolated pressure drop curve is an estimate of the critical value, \( \varepsilon^I \). A simpler alternative estimate is the value at which the two curves cross.

The predicted pressure drop curve compares reasonably well with the experimental data. Figure 1 gives us a possible reading key of phase inversion in case of wash-out experiments: let us assume to start with a single phase flow of pure water (\( \varepsilon_o = 0 \)) and then to gradually add oil; since there is a two-phase flow, at each value of \( \varepsilon_o \) the system has to choose between \( D_{o/w} \) and \( D_{w/o} \). For instance, if we look at the case corresponding to \( \varepsilon_o = 0.44 \), the system can be either in state \( a \) (\( D_{o/w} \)) or in state \( b \) (\( D_{w/o} \)); experimentally, it is observed that the system is in state \( a \), i.e. the one of minimal dissipation rate. Similarly, if \( \varepsilon_o = 0.56 \), state \( d \) (rather than state \( c \)) is chosen (the corresponding flow rate being \( D_{o/w} \)) and, again, this is the flow regime for which the dissipation rate is minimal. If we keep adding oil, the pressure gradient increases but the flow pattern does not change up to \( \varepsilon_o \sim 0.64 \) where, a small further increase in oil flow rate causes a dramatic change: the flow pattern suddenly switches from \( D_{o/w} \) to \( D_{w/o} \). Phase inversion has occurred. Figure 1 shows that phase inversion can be interpreted as the spontaneous tendency of the flow to switch to the flow pattern which minimizes the dissipation rate (pressure gradient).

Notice also that because of the homogeneous model assumption and of setting \( 1/K = 1 \) in Eq. 6.10, our simple model yields, for both laminar and turbulent flow and regardless of the values of \( \rho_w, \rho_o, D, \) and \( U_m \), the following expressions

\[
\varepsilon_I^I_w = \frac{1}{1 + (\mu_o/\mu_w)^{2/5}} \quad \varepsilon_I^I_o = \frac{1}{1 + (\mu_o/\mu_w)^{-2/5}}
\]

for the hold-up at inversion. Of course, this is because in this model we neglect wettability and surface tension. The choice of \( K = 1 \) was suggested by Pal (1993) since it gives a good agreement on the pressure drop prediction; if we assume \( K \neq 1 \) then the critical hold-up, in case of both laminar and turbulent flow, is given by

\[
\varepsilon_I^I_o = \frac{1 - \bar{\mu}^{-2/5K} + K\bar{\mu}^{-2/5K}}{1 + \bar{\mu}^{-2/5K}},
\]

where \( \bar{\mu} = \mu_o/\mu_w \).
6.3 Comparison with theoretical models, empirical correlations, and experimental results

In this section, we check if phase inversion occurring in wash-out experiments performed by Piela et al. (2006) can be interpreted in term of minimal dissipation rate, i.e., if the flow pattern that develops (either $D_{o/w}$ or $D_{w/o}$) is indeed the one minimizing the dissipation rate. To this end, we compare, by using the homogeneous model, the dissipation rate associated to each flow pattern and we check if the configuration with the lowest dissipation rate is the one that actually occurs. In Fig. 6.2 the critical oil hold-ups $\varepsilon^*_o$ predicted by different models are compared with the ones measured in the wash-out experiments by Piela et al. (2006).

In most of the theoretical models and empirical relations given in the literature the critical concentration at inversion depends only on the viscosity ratio of the two liquids. Their predictions for the critical concentration do not depend on other experimental conditions, such as the mixture velocity or the injection rate of the dispersed phase. So, as can be seen in Fig. 6.2, the critical concentration is constant and almost the same (about 60%) for all literature models and correlations. However, the experimental results carried out by Piela et al. showed, that much higher critical concentrations are possible (even as high as 90%).

When we applied our minimal dissipation model, a much better agreement was found. In order to be able to apply this model we fitted the predicted pressure gradient as function of the dispersed phase concentration (using equation (6.7) with equation (6.11) for the mixture viscosity) to the experimentally determined pressure gradient. In this way the two constants in equation (6.11) were found for each experiment separately. For instance, the two constants as found for a certain water-to-oil experiment before inversion are also used for the oil-continuous dispersion after inversion. At the intersection of the two lines the critical concentration is found. If then we use the same calculation procedure in the other direction, we find different values for the two constants for the oil-to-water experiment before inversion which are also used for the water-continuous dispersion after inversion. In this way the ambivalence region is calculated and the predicted results are in good agreement with experimental data as can be seen in Fig. 6.2.

6.4 Conclusions

We propose an interpretation of phase inversion in liquid-liquid (oil-water) pipe flow based on minimal dissipation rate. The approach is based on estimating the relation between pressure drop $dp/dx$ and hold-up $\varepsilon$ by means of a homogeneous model based on a simple relation for the effective mixture viscosity. This must be done for both water-in-oil dispersed flow ($D_{w/o}$) and oil-in-water dispersed flow ($D_{o/w}$) obtaining two $dp/dx$ vs $\varepsilon$ curves, each defined for all values of $\varepsilon$. Our model compares reasonably well with experimental data coming from wash-out experiments, as well as with other empirical and theoretical models available in the literature. It assumes that at every value of $\varepsilon$, the flow pattern which actually occurs is the one which minimizes the overall dissipation rate (in pipe flow, this is equivalent to minimizing the pressure gradient $dp/dx$). As a result, the model implies that phase inversion occurs at the value of the hold-up $\varepsilon$ where the two $dp/dx$ vs $\varepsilon$ curves intersect.
Figure 6.2: Comparison between model resent in literature and data from wash-out experiments by Piela et al. (2006). From a to d wash out experiments start with oil and the gradually water is added; in this case the experimental results are compared with a: Arirachakaran et al. (1989); b: with the interpolation curve $\tilde{\mu}_0^{0.22}/(1 + \tilde{\mu}_0^{0.22})$; c: Poesio and Beretta (2008); d: Brauner and Ullmann (2002). Experiments from e to h refer to the case where a water flow is started and oil is gradually added. Those experiments are compared with e: Arirachakaran et al. (1989); f: with the interpolation curve $\tilde{\mu}_0^{0.22}/(1 + \tilde{\mu}_0^{0.22})$; g: Poesio and Beretta (2008); h: Brauner and Ullmann (2002). Finally the comparison of the experiments with the model proposed here is given as i ($o \rightarrow w$) and l ($w \rightarrow o$).
Even very high critical hold-up values ($\varepsilon_o = 90\%$), that were greatly underestimated by the model present in literature, are predicted reasonably well showing that minimal dissipation rate approach can provide a consistent way to interpret, and may be in future predict, phase inversion phenomenon.

The results and approach presented here can be easily extended to account for the influence of other parameters such as pipe roughness, using more complex relations for the friction factor and for the effective viscosity.
The theoretical model presented in the previous chapter cannot predict the dependence of the critical concentration at phase inversion on the injected phase volume fraction. In the coming chapter another model is presented, that is able to describe this dependence. It is based on a mean-field theoretical description and contains two adaptable parameters.
Chapter 7

A phenomenological description of phase inversion

We propose a Ginzburg-Landau model for a description of the ambivalence region associated with the phenomenon of phase inversion observed in dispersed water-oil flow through a pipe. In analogy to the classical mean-field theory of phase transitions, it is shown that a good quantitative representation of the ambivalence region is obtained by using the injected phase volume fraction and a friction factor as the appropriate physical parameters.

7.1 Introduction

The flow of two immiscible liquids often occurs as a dispersed flow, where one liquid is present in the other one in the form of drops. Dispersions are widely used in the petrochemical-, food-, chemical- and pharmaceutical industries. Handling and controlling dispersion properties is of key interest for practical applications. Oil-water dispersions can occur as oil drops in a water continuous phase or water drops in an oil continuous phase. Phase inversion is the phenomenon by which the dispersed phase becomes the continuous one, and vice versa. During this process the effective viscosity of the mixture becomes very large, which leads to a high pressure drop or low flow rate. Prediction of the critical concentration where phase inversion occurs, is important for two main reasons. It can help in designing processes in such a way, that a high-pressure build-up due to inversion is avoided. It can also help in designing processes, in which a dense dispersion is necessary for achieving the correct product property (for instance in the food or pharmaceutical industry).

The phase inversion phenomenon has been studied for many years (see, for instance Becher (2001); Yeo et al. (2000)). Several models proposed in the literature for predicting the critical concentration are based on a description of a balance between break-up and coalescence of the drops forming the dispersed phase (see for instance Arashmid and Jeffreys (1980) and Vaessen et al. (1996)). In Yeo et al. (2002a) and Brauner and Ullmann (2002) it is assumed that inversion

Chapter 7. A phenomenological description of phase inversion

occurs, when the surface energy of the oil-in-water dispersion is equal to the surface energy of the water-in-oil dispersion and the surface energy is calculated based on the drop size which is again calculated from a balance between break-up and coalescence.

Experimentally it has been found that a so-called ambivalence region exists, in which both phases may be found to be continuous. Only outside this region one phase is always continuous and the other is always discrete. Inside the ambivalence region either one of the two phases can be continuous and the exact value at which phase inversion occurs depends on the operating conditions, surface tension, pipe wettability and chemical properties. Based on some of the aforementioned models attempts have been made to predict the ambivalence region. For instance in Yeo et al. (2002a), an attempt has been made to predict the ambivalence region by calculating the drop size using a different relation for an oil-in-water dispersion than for a water-in-oil dispersion. In Brauner and Ullmann (2002) it was suggested that a possible reason for the hysteresis (ambivalence) region is the time needed after phase inversion for the new continuous phase to completely wet the wall.

Most of the experiments reported in the literature were performed in a stirred vessel and usually water and oil were used. They were often ”continuous experiments” during which the dispersed phase was gradually added to the continuous phase. For this type of experiments it was found, that phase inversion could be postponed to a high value (> 0.8) of the dispersed phase volume fraction. Also a wide ambivalent volume-fraction region existed where the mixture could be either water continuous or oil continuous (Vaessen et al. (1996); Groeneweg et al. (1998); Deshpande and Kumar (2003); Mira et al. (2003); Tyrode et al. (2003)). Alternatively, during ”direct experiments” in a stirred vessel the two liquids were mixed at a certain concentration (Quinn and Sigloh (1963); Tyrode et al. (2005)) and inversion usually occurred at a value of the dispersed phase fraction close to 0.5 (dependent on the properties of the liquids) and no ambivalence region was observed.

Only a few phase-inversion experiments have been carried out in a pipe. Direct experiments in a vertical pipe have been reported in Liu et al. (2006) and direct experiments in a horizontal pipe in Pal (1993); Nádler and Mewes (1997); Ioannou et al. (2005); Chakrabarti et al. (2006). In these experiments particular attention was paid to the pressure-drop increase over the pipe during phase inversion. Recently, we have carried out continuous phase-inversion experiments in a pipe (Piela et al. (2006)). During these experiments we started with a flow of one of the liquids and gradually injected the other liquid, while keeping the mixture velocity constant. We measured also a strong increase in the pressure drop during the inversion process. Depending on the injected phase volume fraction $\chi$ (the ratio between the injection rate of the dispersed phase and the flow rate of the mixture in the pipe), it was found that phase inversion could be postponed up to high values of the dispersed phase. With decreasing values of $\chi$ the critical concentration increased, even up to values of 0.9. Hence, the ambivalence region for these experiments was very wide at small values of $\chi$ and it became narrower with increasing $\chi$. At a value of about $\chi = 0.5$ the ambivalence region disappeared and the critical concentration became equal to the one found during direct experiments (see Piela et al. (2008)), whereby the two liquids are injected from the start simultaneously into the pipe with certain concentrations.

It is not evident to us, how this important result from the experiments reported in Piela et al. (2006) can be explained with one of the aforementioned models. Therefore, we have devel-
7.2. Experimental observations

opened a framework for the interpretation of the phase-inversion experiments adopting an analogy with the classical Ginzburg-Landau mean-field theory of phase transitions in thermodynamics (Chaikin and Lubensky (1995)). By considering the friction factor $f$ as function of the injected phase volume fraction $\chi$, we have succeeded to obtain a realistic description of the ambivalence region for a range of conditions with different values of the injected phase volume fraction during continuous experiments.

In section 7.2 we briefly review our experimental results (Piela et al. (2006, 2008)). In section 7.3 we adopt a Ginzburg-Landau model for the interpretation of our phase inversion experiments. In section 7.4 it is shown how this model yields a quantitative representation of the ambivalence region by using the injected phase volume fraction $\chi$ and a measured friction factor $f$ as the characteristic physical variables of the phase-inversion process. In section 7.5 we show how the model can be used in the absence of experimental data for the friction factor by relating the measured friction factor $f$ to an effective viscosity. Our results are summarized in section 7.6.

7.2 Experimental observations

In our experimental studies (Piela et al. (2006, 2008)) many phase-inversion experiments were performed on an oil-water flow through a pipe. Two types of experiments were performed: continuous experiments and direct experiments.

The sketch of the set-up used for the continuous experiments is shown in Fig. 7.1. The symbols used in the sketches have the following meaning: $V$: valves, $F_1$: Krohne Optimass 7000 flow meter (measuring error $< 0.26\%$), $F_2$: Krohne Corimass E flow meter (measuring error $< 0.4\%$), $C_1$: conductivity cell and $T_1$: thermocouple. During the continuous experiments one of the liquids (water or oil) was taken from the continuous phase tank (see Fig. 7.1) and recirculated through the pipe loop by means of pump 1. After recirculating the liquid for a few minutes to ensure that the pipe walls were wetted by the liquid, injection of the other liquid started by pumping (using pump 2) the dispersed phase through the injector into the pipe loop. During the injection, valve 2 was opened and the same volume of dispersion liquid was removed from the pipe loop as the volume that was injected. Flow meter 1 measured the density and the flow rate of the mixture in the pipe loop. The mixture velocity was kept constant during the experiment by an electronic feedback system (pump 1 was controlled based on measurements of flow meter 1). During continuous experiments the dispersed phase was gradually added to the flowing mixture in the closed pipe loop. The dispersed phase volume fraction was slowly increased and at a certain moment inversion occurred. During these experiments it was noticed that the dispersed phase fraction at which inversion occurs, depends nearly linearly on the injected phase volume fraction $\chi$. By starting an experiment with pure water and gradually adding oil and by starting with pure oil and gradually adding water the ambivalence region was measured. It was found that the ambivalence region is almost independent of the Reynolds -, Froude - and Weber numbers and also independent of the injection velocity of the dispersed phase (as long as the mixture velocity is sufficiently large ($\geq 2$ m/s)). However, it was found that the injection phase volume fraction $\chi$ had a significant influence on the width of the ambivalence region. As can be seen in Table I all experiments were conducted at high $Re$, $We$ and $Fr$ numbers. According to Brauner
Chapter 7. A phenomenological description of phase inversion

Figure 7.1: Sketch of the experimental set-up for continuous experiments.

<table>
<thead>
<tr>
<th>water</th>
<th>$Re$</th>
<th>$Fr$</th>
<th>$We$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 m/s</td>
<td>$1.6 \cdot 10^4$</td>
<td>6.4</td>
<td>354</td>
</tr>
<tr>
<td>2 m/s</td>
<td>$3.2 \cdot 10^4$</td>
<td>25.5</td>
<td>1418</td>
</tr>
<tr>
<td>3 m/s</td>
<td>$4.8 \cdot 10^4$</td>
<td>57.3</td>
<td>3190</td>
</tr>
<tr>
<td>1.34 m/s</td>
<td>$2.1 \cdot 10^4$</td>
<td>11.4</td>
<td>637</td>
</tr>
<tr>
<td>Shell Macron EDM 110</td>
<td>$Re$</td>
<td>$Fr$</td>
<td>$We$</td>
</tr>
<tr>
<td>2 m/s</td>
<td>$0.7 \cdot 10^4$</td>
<td>25.5</td>
<td>1129</td>
</tr>
<tr>
<td>3.5 m/s</td>
<td>$1.1 \cdot 10^4$</td>
<td>78</td>
<td>3458</td>
</tr>
<tr>
<td>1.35 m/s</td>
<td>$0.4 \cdot 10^4$</td>
<td>11.6</td>
<td>515</td>
</tr>
</tbody>
</table>

Table 7.1: Dimensionless numbers for water-continuous - or oil-continuous experiments at different superficial velocities. (The superficial velocity of a certain fluid is defined as the ratio of the volume flow rate of that fluid and the pipe cross-section.) For the calculation of the Froude number the density difference between water and oil is used and for the calculation of the Weber number the interfacial tension between the two liquids. "Dispersed phase boundary" indicates the transition from the dispersed flow pattern to another flow pattern according to Brauner (2001).

(2001) a mixture velocity of 2 m/s is sufficiently large to be sure of a dispersed pipe flow during our experiments.

For direct experiments the set-up shown in Fig. 7.1 was modified. The pipe loop was opened and two additional straight pipe sections were added. During the direct experiments the two liquids were injected (from separate tanks) simultaneously into the pipe by means of pump 1 and pump 2. The two liquids flowed through the pipe to the large separation tank, which was situated at the end of the last pipe section. The mixture was injected at a constant concentration for at least 40 s. Flow meter 1 measured the density and the flow rate of the mixture in the pipe. The pressure gradient was measured at six different positions: immediately downstream of the inlet and at distances of 2.0 m ($125d$), 5.0 m ($313d$), 11.7 m ($731d$), 18.7 m ($1169d$) and 26.5 m ($1656d$) from the inlet, where $d$ is the pipe diameter.

The phase-inversion process was determined by the mixing between oil and water at and after
7.2. Experimental observations

Figure 7.2: Inversion map. The upper line gives the oil volume fraction at the point of inversion as measured during water-to-oil continuous experiments. The lower line represents the oil volume fraction at inversion as measured during oil-to-water continuous experiments. The region in between the two lines is the ambivalence region, where both oil and water can be the continuous phase. Also the oil volume fraction at inversion as measured during the direct experiments is indicated.

The observed ambivalence region for the case of continuous experiments is shown in Fig. 7.2. As can be seen from this figure, the width of the ambivalence region depends strongly on the injected phase volume fraction $\chi$. The experimental results lie on two lines, one for the water-to-oil experiments and one for the oil-to-water experiments. (Two experimental data points for the oil-to-water experiments do not coincide with the line. The explanation for this deviation was given in Piela et al. (2006): entrapment of the continuous phase (oil) into the dispersed phase (water) increases the effective dispersed phase fraction. If the concentration of oil present as oil droplets inside the water drops would be subtracted from the total oil concentration, the experimental data points would fit the line. These two points will be omitted in the remainder of this publication.)

In Fig. 7.3 the friction factor $f$ (calculated from the measured pressure drop by means of $f = 2\Delta P d / \rho u^2 L$, where $\Delta P$ is the pressure drop over a distance $L$, $d$ the pipe diameter, $\rho$ the density and $u$ the average mixture velocity) is shown as function of the oil volume fraction $\phi$ for water-to-oil continuous experiments presented in Fig. 7.2. As can be seen, the friction factor $f$ strongly increases when the concentration of the dispersed phase exceeds 0.6. The friction factor does
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Figure 7.3: Friction factor for three water-to-oil experiments at a mixture velocity of 2 m/s and 3 m/s and different values of the injected phase volume fraction (0.03, 0.125, 0.18).

Figure 7.4: Inversion from a water-continuous to an oil-continuous flow during a continuous phase-inversion experiment.

not depend on the injected phase volume fraction prior to the occurrence of phase inversion. (Although the critical concentration at which phase inversion occurs, depends on $\chi$). The friction factor was also found to be independent of the mixture velocity. A similar behavior was observed for oil-to-water continuous experiments.

Figure 7.4 shows the different stages of the phase inversion process during a continuous experiment of a water-continuous flow to an oil-continuous flow (water-to-oil experiment) for a mixture velocity of 1 m/s and an injection phase volume fraction $\chi = 0.125$. The left picture shows the start of the inversion process at an oil volume fraction $\chi = 0.84$. Some larger oil multiple drops (containing water droplets) are formed in a water-continuous region due to coalescence of the original oil drops. After 16 s (second picture) the number of these oil-continuous pockets has increased due to continuous oil injection via the injector. With further oil injection, isolated water-continuous pockets are created embedded in oil-continuous regions. After 43 s (third picture) the oil-continuous pockets in water-continuous regions and water-continuous pockets in oil-continuous regions are about the same in number and size. With further oil injection, the oil-continuous pockets disappear and only water-continuous pockets in an oil-continuous
7.2. Experimental observations

region remain. After 87 s (right picture) the phase inversion is completed and the mixture is oil-continuous with water drops.

During a direct experiment the phase fractions were kept constant in time. Since the two phases are well mixed the dispersed phase volume fraction in the pipe is equal to the dispersed phase volume fraction at the point of injection at the entrance to the pipe loop. Close to the point where the flow changed from a water-continuous to an oil-continuous flow or vice versa, strong variations in the morphological structures of the dispersion were observed. For a mixture velocity of 2 m/s, these changes occur at oil volume fractions \( \phi \) between 0.5 and 0.6. Figure 7.5 shows the dispersion morphology for two different values of the oil volume fraction. At \( \phi = 0.56 \) (see left picture) the flow consists of many (water-continuous) pockets in oil-continuous regions (dark regions), and (multiple) oil drops in water-continuous regions (see, for instance, the center part of the picture). At \( \phi = 0.58 \) (see right picture) most of the flow field is oil continuous.

There is an important difference between a continuous experiment and a direct experiment. In a continuous experiment the inversion process starts with the coalescence of drops leading to the formation of larger drops, pockets and regions by encapsulation of parts of the continuous phase. Finally this process causes the disappearance of regions of the originally continuous phase. In a direct experiment the two liquids are mixed from the start at a constant concentration of the phases and there is no preliminary structure. Depending on the concentration, the liquids mix as a water-continuous mixture or an oil-continuous one. However, at an oil volume fraction \( \phi \) between 0.5 and 0.6 both water-continuous and oil-continuous regions are created during the same experiment (because of the non-homogeneous mixing, in particular in the T-junction and the entrance region of the pipe). At a value of \( \phi \) between 0.5 and 0.6, none of the regions is sufficiently strong to dominate the other one, and so both regions flow downstream. These regions interact, entrap parts of the other continuous phase, break-up and coalescence causing a large pressure gradient over the pipe.

A more detailed description of the experimental observations of phase inversion during continuous and direct experiments can be found in some of our previous publications (Piela et al. (2006, 2008)). Pictures taken during phase inversion were presented and measurements of the pressure drop and conductivity were reported. The behavior of the pressure drop increase during inversion was explained by visual observations. Also interactions during phase inversion were described. These results showed that phase inversion during continuous and direct experiments, despite the different onset, has a very similar behavior.
7.3 A Ginzburg-Landau model for phase inversion

During phase inversion very complex structures are created (as described in Piela et al. (2008)). Regions, pockets and (multiple) drops interact with each other. The complexity of the structures and their interaction make it very difficult to model phase inversion in detail. However, we know from thermodynamics that phase transitions can be described by a mean-field approach without any need for a knowledge of the detailed molecular interactions. In this paper we propose a similar approach for a description of the phenomenon of phase inversion.

The mean-field theory of phase transitions is based on a classical Landau expansion of an appropriate free-energy potential \( G \) in terms of an order parameter \( s \). The simplest version is a two-term Ginzburg-Landau model of the form \( G(s) = s^4 + as^2 \), where the coefficient \( a \) is a control parameter that is related to the difference between the actual temperature \( T \) and a critical temperature \( T_c \) (Chaikin and Lubensky (1995); Luban (1976)). This model yields a bifurcation at \( a = 0 \) (i.e., at \( T = T_c \)) with a quadratic phase boundary asymptotically close to the critical point (Baker (1980)). To describe our phase-inversion experiments we need a slightly more general Ginzburg-Landau model of the form (Gilmore (1981); Dickinson (1981)):

\[
G(s) = s^4 + as^2 + bs ,
\]

with two control parameters \( a (a \leq 0) \) and \( b \) that will be specified for the process of phase inversion in the subsequent section.

Depending on the values of \( a \) and \( b \) the \( G(s) \)-function has one or two minima. The location of the minima in the \( G(s) \)-domain are found by the following condition

\[
\frac{dG}{ds} = 4s^3 + 2as + b = 0 ,
\]

and the number of minima changes from one to two when the second derivative of \( G \) to \( s \) becomes equal to zero

\[
\frac{d^2G}{ds^2} = 12s^2 + 2a = 0 .
\]

Eliminating \( s \) from Eqs. (7.2) and (7.3) leads to a relation between \( a \) and \( b \) for the change-over from one to two minima

\[
(2a/3)^3 + b^2 = 0 .
\]

The shape of this relation between \( a \) and \( b \) is shown in Fig. 7.6. Asymptotically the two branches merge according to a power-law behavior of the form \( b \propto |a|^{3/2} \). Hence, Eq. (7.1) is also being referred to as a bifurcation model exhibiting a cusp catastrophe (Vaessen et al. (1996); Gilmore (1981); Dickinson (1981)).

When the line M-P-R-Q and the line N-Q-S-P in Fig. 7.6 are followed, the shape of the \( G(s) \)-function changes as sketched in Fig. 7.7. Let us start with the line M-P-R-Q. At point \( M \) only one minimum in the free energy exists and the system is assumed to be in this minimum (indicated by the black spot in Fig. 7.7). Increasing \( b \) in Fig. 7.6 moves the system to point \( P \), where a second minimum starts to appear. At point \( R \) there are two symmetric minima with a barrier between them, so the system remains in the left minimum. At point \( Q \) the barrier disappears and
7.3. A Ginzburg-Landau model for phase inversion

Figure 7.6: Relation between $a$ and $b$ where a change-over takes place from one to two minima in the $G(s)$-function.

Figure 7.7: Changes in the shape of the $G(s)$-function with passage through the line M-P-R-Q and the line N-Q-S-P in Fig. 7.6.
even a small perturbation causes the system to move to the right minimum. Similar behavior is observed for the line N-Q-S-P, which is followed with decreasing value of $b$. As can be seen a hysteresis occurs. When the line M-P-R-Q is followed, a transition from the left minimum to the right one takes place at point Q. When the line N-Q-S-P is followed, a transition from the right minimum to the left one occurs at point P. So between P and Q two states are possible and the actual state depends on the starting condition.

This behavior with a hysteresis effect based on the Ginzburg-Landau model, Eq. (7.1), is very similar to the physical behavior in the ambivalence region observed in our continuous experiments. Also Fig. 7.6, where two states are possible between the two converging lines, looks the same as the ambivalence region shown in Fig. 7.2. So the idea to use an approach for the description of phase inversion similar to the one used for the description of phase transitions looks attractive. In the next section this idea will be worked out in detail.

### 7.4 Application to phase inversion

We submit that the injected phase volume fraction $\chi$ is a correct quantity to be used as a control parameter for the interpretation of the ambivalence region measured during phase inversion experiments in terms of the hysteresis region of the Ginzburg-Landau model. With increasing $\chi$, the injection rate of the dispersed phase into the mixture increases (at constant mixture velocity). This causes a stronger disturbance in the dispersion and the barrier in the $G(s)$-domain can more easily be taken. Therefore, with increasing $\chi$ the width of the ambivalence region will become smaller. Hence, we use $\chi$ as the first physical relevant quantity.

At first sight one might consider the oil volume fraction at inversion $\phi$ as the second relevant physical control parameter. However, there is a better choice. Important for the occurrence of phase inversion is the shear stress in the dispersion. This stress plays a role in the coalescence and break-up of drops that form the dispersed phase. The shear stress is directly related to the pressure gradient over the pipe. As the second correlating quantity should also be non-dimensional, this leads to the friction factor $f$ as the possible second correlating quantity. In Fig. 7.3 the measured friction factor $f$ was shown as function of the oil volume fraction $\phi$. As can be seen, $f$ is rather dependent on the dispersed phase volume fraction (certainly at high values of $\phi$). So it seems reasonable to select $f$ as the second correlating quantity.

For given values of the injected phase volume fraction $\chi$ and the friction factor $f$, the potential $G$ represents the free energy of the system as a function of an order parameter $s$. This parameter $s$ represents the possible mesoscopic conditions of the liquids during their flow through the pipe. With mesoscopic conditions, we mean their mutual distribution (size of drops, presence of multiple drops, etc.). The resulting macroscopic condition will correspond to a minimum of $G(s)$ as function of $s$. To calculate $G(s)$ as a function of $s$ explicitly is difficult. However, as also noted by previous investigators (Dickinson (1981)), the nice property of the Ginzburg-Landau model is that it gives an explicit relation, Eq. (7.4), for the control parameters associated with the macroscopic states of the system that will enable us to characterize the ambivalence region in terms of only two system-dependent parameters.

As mentioned in Section 7.2, we have observed an ambivalence region with a cusp. At the
7.4. Application to phase inversion

Figure 7.8: The oil volume fraction $\phi$ as a function of the injected phase volume fraction $\chi$ at the boundaries of the ambivalence region. The circles and squares indicate the experimental data points and the curves represent the values calculated from the Ginzburg-Landau model.

termination of the cusp (in Fig. 7.6) $a = 0$. The continuous experiments show that the width of the ambivalence region decreases with increasing value of $\chi$. The ambivalence region even disappears at about $\chi = 0.5$. This makes sense, as at $\chi = 0.5$ it does not matter which liquid is in the continuous phase and which liquid is in the dispersed one. So the cusp in the ambivalence region is at about $\chi = 0.5$. Hence, we assume the following linear relation between $a$ and $\chi$

$$a = \chi - 0.5. \quad (7.5)$$

The relation between $b$ and $f$ is assumed to have the following form

$$b = K_{b1}(f - K_{b2}) \quad (7.6)$$

in which $K_{b1}$ and $K_{b2}$ are constants. To determine the values of these constant two experimental data points ($A$ and $B$) were fitted to Eq. (7.4) implied by the Ginzburg-Landau model. During a water-to-oil experiment with an injection phase volume fraction of $\chi_A = 0.05$ phase inversion took place at an oil volume fraction of $\phi_A = 0.9$ and during an oil-to-water experiment with a $\chi_B = 0.05$ phase inversion occurred at $\phi_B = 0.31$. These two points $A(\chi = 0.05, \phi = 0.9)$ and $B(\chi = 0.05, \phi = 0.31)$ (and their related $f$-values) were selected to determine $K_{b1}$ and $K_{b2}$.

Once the constants have been determined, substitution of Eqs. (7.5) and (7.6) into Eq. (7.4) gives the ambivalence region in the $(\chi, f)$-domain. Using the experimentally found relation between $\phi$ and $f$, we can also calculate the ambivalence region in the $(\chi, \phi)$-domain. The result is shown in Fig. 7.8. As can be seen the calculated ambivalence region agrees well with the experimental data. Also the value from the direct experiment is on the calculated line. (The curve in Fig. 7.8 does not terminate in the cusp-like behavior as in Fig. 7.6, since the conversion from $f$ to $\phi$ distorts the curve.)

We used the experimental data points A and B to determine the constants $K_{b1}$ and $K_{b2}$, since these points have the highest accuracy. To investigate the sensitivity of the description to the choise


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<table>
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<tr>
<th>( \chi )</th>
<th>point 1</th>
<th>point 2</th>
<th>predicted ( \phi_A )</th>
<th>predicted ( \phi_B )</th>
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</tr>
<tr>
<td>0.18</td>
<td>( \phi_E=0.82 )</td>
<td>( \phi_F=0.37 )</td>
<td>0.86</td>
<td>0.34</td>
</tr>
</tbody>
</table>

Table 7.2: Predicted critical concentrations \( \phi_A \) and \( \phi_B \) when different pairs of data points (A,B; C,D and E,F) are used for the determination of the constants \( K_{b1} \) and \( K_{b2} \).

of pair of data points, two other pairs were taken at \( \chi = 0.125 \) (pair C,D) and \( \chi = 0.18 \) (pair E,F) and used to calculate the constants and the predicted values of the critical concentrations at \( \chi = 0.05 \) (pair A,B). The results are shown in Table II. As can be seen, the values calculated for the critical concentrations at \( \chi = 0.05 \) are rather insensitive to the pairs of data points selected for the determination of the two system-dependent parameters.

7.5 Application in terms of a theoretical friction factor

In practice experimental data for the friction factor will not always be available. So it would seem that the method presented in the last section can then not be applied. However, there is a solution to this problem. In practice experimental data on the oil volume fraction \( \phi \) at inversion are often available and these data can be used to predict the friction factor \( f \) at inversion by applying an empirical correlation between \( f \) and \( \phi \).

Assuming a homogeneous dispersed flow we can calculate the friction factor by applying the Blasius correlation \((2000 < Re < 10^5)\)

\[
f = \frac{0.316}{Re^{0.25}}. 
\]

The effective viscosity \( \eta \) in the Reynolds number \((Re)\) can be calculated by one of the following correlations

- Mooney equation (Mooney (1951))

\[
\eta = \exp\left(\frac{2.5\phi}{1-k\phi}\right) 
\]

with \( k = 0.8 \)

- Krieger-Dougherty equation

\[
\eta = \eta_0 \left(1 - \frac{\phi}{\phi_c}\right)^{-5/2\phi_c} 
\]

- Pal model 1 (Pal (2000))

\[
\eta \left[\frac{2\eta + 5K}{2 + 5K}\right]^{3/2} = \exp\left[\frac{2.5\phi}{1-\phi/\phi_c}\right] 
\]
7.6 Discussion

As can be seen from Figs. 7.8 and 7.9, a Ginzburg-Landau model can be used to describe phase inversion in an oil-water mixture. This is due to the close similarity between the hysteresis region for first-order phase transitions and the ambivalence region for phase inversion between two liquids in a dispersed flow.
According to our measurements (Piela et al. (2006)), the ambivalence region is unique for a specific water-oil mixture. For such a mixture the ambivalence region depends on the injected phase volume fraction. It is independent of the Reynolds -, Froude - and Weber numbers and also independent of the injection rate, as long as the mixture velocity is large enough. With our method the ambivalence region can be fully determined by carrying out only two experiments. Of course, when the oil type is changed or when a surfactant is added, the ambivalence region will change and two new experiments need to be performed.
The last chapter of this thesis describes our oil-water-gas experiments. The continuous and direct experiments described in the first part of the thesis are now extended with gas injection and a dispersed three-phase flow is studied. Much attention is given to the effect of the gas bubbles on the inversion process and vice versa. Also the influence of the bubbles on the pressure gradient will receive attention.
Chapter 8

Dispersed oil-water-gas flow through a horizontal pipe*

An experimental study of three-phase dispersed flow in a horizontal pipe has been carried out. Pressure drop over the pipe strongly increases with increasing bubble and drop concentration. Due to the presence of drops the transition from dispersed bubble flow to elongated bubble flow occurs at a lower gas concentration. The gas bubbles have no significant influence on the phase inversion process. However, phase inversion has a strong effect on the gas bubbles. Just before inversion large bubbles are present and the flow pattern is elongated bubble flow. During the inversion process the bubbles break-up quickly and as the dispersed drop concentration after inversion is much lower than before inversion, a dispersed bubble flow is present after inversion. (When inversion is postponed to high dispersed phase fractions, the concentration of the dispersed phase can be as high as 0.9 before inversion and as low as 0.1 after inversion.)

8.1 Introduction

In the oil industry oil-water-gas flow often occurs. For instance, when producing oil from a subsurface reservoir it is very well possible that not only oil but also water and gas (present in the reservoir) flow through the production tubing to the facilities at the surface. To predict the production rate it is essential to know the properties of such a three-phase flow and to be able to model it. There are not many papers dealing with three-phase flow through a pipe, but recently this subject is receiving more attention because of the industrial relevance. The flow pattern as function of the flow conditions was studied by Acikgöz et al. (1992); Hewitt et al. (1997); Wu et al. (2001); Spedding et al. (2005). Also the influence of the inclination angle of the pipe on the flow was investigated by Oddie et al. (2003). It has been shown that the pressure drop of the three phase oil-water-gas system is of the same order of magnitude as the pressure drop of a two-phase system consisting of the gas and the dominating liquid phase (Nádler and Mewes (1995)). Lahey Jr et al. (1992) applied the drift-flux technique and validated it against volume

---

fractions data for such a flows. Furthermore, Hewitt (2005) showed the importance of the mixing process on the flow behavior.

In our research we have restricted ourselves to dispersed three-phase flow with a continuous liquid (oil or water), in which drops of the other liquid and gas bubbles are present. Dispersed flow occurs only at high mixture velocities. Special attention was given to the phenomenon of phase inversion, whereby the mixture switches from an oil-continuous phase (with water drops and gas bubbles) to a water-continuous phase (with oil drops and gas bubbles), or vice versa. During phase inversion the pressure gradient of the three-phase flow in the pipe increases strongly. So it is important to know at what conditions phase inversion occurs, in order to find out whether in practice it can be avoided or its effect on the pressure drop can be minimized.

In our previous studies (Piela et al. (2006, 2008)) we carried out experiments to study phase inversion in an oil-water flow through a pipe. Two different types of experiments were carried out (see Fig. 8.1). During a direct experiment water and oil were pumped from separate tanks and injected simultaneously into the pipe at a constant concentration. During a continuous experiment the dispersion was circulated through the pipe loop and the dispersed phase was gradually injected (while the same amount of dispersion was removed). So during a continuous experiment the dispersed phase fraction was slowly increasing up to the inversion point. The continuous experiments showed that phase inversion can be postponed to a high dispersed phase fraction (even as high as 0.9). Phase inversion was also found to be strongly dependent on the injected phase volume fraction (ratio of the volume flow rate of injected liquid and the volume flow rate of the mixture). One of the purposes of this further study was to investigate the influence of the presence of gas bubbles on the earlier results.

In section 8.2 the experimental set-ups are described. Also the visualization technique using a high speed camera and an optical fiber probe will be discussed. In section 8.3 our previous results from the oil-water experiments are briefly summarized. Section 8.4 is devoted to gas-liquid dispersed flow. In section 8.5 the results from the three-phase flow experiments are given with special attention to the influence of gas bubbles on the phase inversion process. Finally section 8.6 presents the most important conclusions.
8.2 Experiments

8.2.1 General information

An acrylic pipe with an inner pipe diameter of 16 mm was used. The two immiscible liquids used were tap water and Shell Macron EDM 110 oil (density 794 kg/m$^3$, kinematic viscosity at 20° $3.9 \, \text{mm}^2/\text{s}$ and oil-water interfacial tension 0.045 N/m). As a gas phase nitrogen was used. All experiments were performed at high Reynolds and Froude numbers to make sure, that always a fully dispersed flow was present (according to Brauner (2001) a mixture velocity of 1.42 m/s is sufficient for our experimental conditions). Continuous and direct experiments were carried out; they will be explained in more detail later on. In both types of experiments the mixture velocity in the pipe was kept constant. Continuous experiments with water as the continuous phase were conducted at a mixture velocity of 3 m/s, which yields a Weber number of 3200. Continuous experiments with oil as the continuous phase were performed at a mixture velocity 3.5 m/s, which gives a Weber number of 3500. The maximum stable bubble size as calculated by the relation of Hinze (1955) is 0.13 mm and 0.12 mm respectively. The details of the pipe configurations used for the two types of experiments will be given when these experiments are discussed.

Pressure drops were measured at different locations over a distance of 1 m. Differential pressure transducers (Validyne DP-15, measuring error < 3%) were used and the pressure signal was sampled at a rate of 2 kHz. Data were averaged over 2000 samples. Conductivity measurements (at the same frequency) were done with a cell consisting of two (0.2 mm diameter) wire electrodes mounted in the pipe: one in the vertical direction and one in the horizontal direction. The distance between the electrodes in the center of the pipe was 2 mm. During the experiment we also monitored the temperature with a thermocouple mounted in the pipe wall. Most of the experiments were conducted more than at least two times and the reproducibility of the experiments was good.

8.2.2 Visualization with high-speed camera

To make a detailed study of the dispersion morphology, samples were taken from the flow by means of a 7 mm inner diameter sampling tube centered in the 16 mm inner diameter pipe loop. The sample tube liquid is led through a visualization cell (which consist of 2 glass plates 1 mm apart, leaving a channel of width 20 mm). The visualization cell was illuminated from the backside using a 500W halogen lamp, while a high-speed camera was used to take images at the front side. The camera was operated at a frame rate of 50 or 500 fps. A sketch of the sampling technique is given in Fig. 8.2; more details are given in our previous paper (Piela et al. (2006)). To check whether this observation procedure had some influence on the dispersion morphology we performed experiments with various tube lengths between visualization cell and pipe, and at various sample liquid flow rates. We found little if any influence. We also changed the distance between the glass plates in the visualisation cell from 1 mm to 5 mm; again the results were qualitatively the same, except that for the wider gap the observed mixture turns more opaque at higher concentrations.
8.2.3 Optical fiber probe

At high mixture velocities the drops and bubbles were very small and the pipe becomes optically dense (opaque). Visualization by means of a (high-speed) camera was then not possible any more. To gather more information about the gas bubble size distribution a single-tip optical fiber probe was used.

The fiber probe consists of a glass rod (of 200 micron diameter), with a rounded tip (of tip radius 60 micron) inserted in the flow (Cartellier (1990); Delfos et al. (2001)). The probe is mounted radially traversable along the vertical (z) axis. The percentage of light led into the fiber reflected back from the tip is detected, and is a measure for the difference in refractive index between glass and the medium wetting the fiber tip. After appropriate thresholding or signal recognition, the actual phase wetting the probe, and related parameters such as bubble frequency or local void fraction can be derived. Probes of this kind for gas-liquid, liquid-liquid and three-phase flows have been in use since the nineties (Cartellier (1992); Hamad (1997); Fordham et al. (1999a,c,b)). In general, they lack the ability to detect phase elements that are small or bounce off. Furthermore, in liquid-liquid flows they tend to suffer most from the wetting phase sticking to the interface, as described by Fordham et al. (1999c,b) and Descamps et al. (2007).

A typical probe signal for an oil-water-gas flow is presented in Fig. 8.3. The refractive index of the gas is much lower than that of the two liquids, hence the gas bubbles can easily be discriminated from the probe signal. The difference in refractive index between the two liquids is much smaller, and only the bigger oil drops (lowest signal) can be separated from the measurement noise, which makes determination of the liquid distribution more questionable. For this reason we used the optical probe only to detect gas bubbles.

We used the fiber probe near the pipe centerline up to halfway the outer radius. The probe signal was sampled at a rate of 30kHz; at a mixture velocity of 3m/s and the presumably rather flat velocity profile, this gives an axial resolution of 0.1 mm. This is sufficiently small, while smaller bubbles will not be detected sufficiently anyway. With the given limitations of the optical probe system, we used the results in a qualitative way only, i.e. to compare bubble sizes between similar experiments (in which only one parameter was changed) and to observe the transition from the dispersed bubble flow pattern to the elongated bubble flow pattern (for which the average bubble size changes significantly). In this way the results of the optical fiber probe contributed well to
8.2. Experiments

Figure 8.3: Signal from the optical probe for an oil-water-gas flow. Water is the continuous phase (gas fraction is 0.07, oil volume fraction is 0.12 and mixture velocity 3 m/s). The presence of gas bubbles and oil drops can be observed.

the interpretation of the measurements.

8.2.4 Continuous experiments

A sketch of the set-up used for the continuous experiments is shown in Fig. 8.4. The symbols used in the sketches have the following meaning: V: valves, F1: Krohne Optimass 7000 flow meter (measuring error < 0.26%), F2: Krohne Corimass E flow meter (measuring error < 0.4%), C1: conductivity cell and T1: thermocouple. During the continuous experiments one of the liquids (water or oil) was taken from the continuous phase tank (see Fig. 8.4) by means of pump 1 and recirculated through the pipe. Pump 1 is a positive displacement pump (lobe pump), chosen to minimize the pumping effect on the dispersion morphology. After recirculating the liquid for a few minutes to ensure that the pipe walls were wetted by the liquid, injection of the other liquid started by pumping (using pump 2) the dispersed phase through the injector into the pipe loop. During the injection valve 2 was open and the same volume of dispersion liquid was removed from the pipe loop as the volume that was injected. When the desired dispersed-phase volume fraction was reached the injection by means of pump 2 was stopped and injection of gas was started. Nitrogen gas was used, which was stored in a gas bottle (pressurized at 180 bar with a reduction to 6 bar). The gas flow was monitored by a rotameter. Flow meter 1 measured the density and the flow rate of the mixture in the pipe loop. The mixture velocity was kept constant during the experiment by an electronic feedback system (pump 1 was controlled based on measurements of flow meter 1). To study the influence of the length of the pipe loop also continuous experiments were carried out, whereby the pipe loop was significantly reduced in length. During these experiments the mixture circulates more often through the pump. However, we observed no difference in the results when compared with the results of the experiments carried out in the longer pipe loop.

A cylindrical container (placed around the pipe loop) with a number of holes in it was used as injector. The dispersed phase was pumped through the holes into the pipe loop. Three different types of injectors were used, viz. a container with 2 holes of 2 mm diameter, a container with 8
holes of 2 mm diameter, and one with 100 holes of 3 mm diameter (for more details, see Piela et al. (2006)). The experimental results were independent of the injector type. The pressure drop was measured 5 m (313d) downstream of the injector.

8.2.5 Direct experiments

During the direct experiments water, oil and gas were taken from the three tanks and injected simultaneously by means of pump 1, pump 3 and rotameter into the pipe (see Fig. 8.5). The mixture was injected at a constant concentration for at least 40 s. Flow meter 1 measured the density and the flow rate of the mixture in the pipe. The pressure drops were measured at six different positions: immediately downstream of the inlet and at distances of 2.0 m (125d), 5.0 m (315d), 11.7 m (730d), 18.7 m (1170d) and 26.5 m (1655d) from the inlet (where d is the pipe diameter).
8.3 Oil-water dispersed flow

In our previous experimental studies (Piela et al. (2006, 2008)) many phase inversion experiments were performed in an oil-water flow through a pipe. In this section we briefly summarize the results. We think that this will help in the interpretation of the experiments with gas injection, which will be presented in the coming sections.

In our earlier work results from continuous experiments and direct experiments were compared. As mentioned in the preceding paragraph during continuous experiments the dispersed phase was gradually added to the flowing mixture in the closed pipe loop. The dispersed phase fraction was slowly increasing and at a certain moment inversion occurred. During these experiments it was noticed that the dispersed phase fraction at which inversion occurs, depends almost linearly on the injected phase volume fraction $\chi$ (ratio of the volume flow rate of the injected liquid and the volume flow rate of the mixture).

For continuous experiments a so-called ambivalence region exists, in which both phases may be found to be continuous. Only outside this region one phase is always continuous and the other is always dispersed. Within the ambivalence region either one of the two phases can be continuous and the exact value at which phases invert depends on the operating conditions. By starting an experiment with pure water and gradually adding oil and by starting with pure oil and gradually adding water the ambivalence region was measured. It was found that the ambivalence region is almost independent of the Reynolds-, Froude- and Weber number and also independent of the injection velocity of the dispersed phase (as long as the mixture velocity is sufficiently large ($\geq 2\text{m/s}$)). The measured ambivalence region is shown in figure 8.6. As can be seen the width of the region depends much on the injected phase volume fraction $\chi$. The measured results lie on two lines, one for the water-to-oil experiments and one for the oil-to-water experiments. (Two measurement points for the oil-to-water experiments do not coincide with the line. The explanation for this deviation is given in Piela et al. (2006); entrapment of the continuous phase (oil) into the dispersed phase (water) increases the effective dispersed phase fraction. If the concentration of oil present as oil droplets inside the water drops would be subtracted from the total oil concentration, the measurement points would likely fit the line.)

During the direct experiments the two liquids were injected (from separate tanks) simultaneously into the pipe. The phase inversion process was determined by the mixing between oil and water at and after the inlet. At the critical concentration water-continuous and oil-continuous regions were created simultaneously. These regions interact flowing downstream. The interaction was very similar to the one observed during the inversion process during the continuous experiments. The direct experiment can be considered as a limiting case of the continuous experiment, with a large injection phase volume fraction of the dispersed phase and with inversion taking place before one cycle through the pipe loop is completed. No ambivalence region was observed during the direct experiments.

In Fig. 8.7 the measured results for the friction factor $f = \frac{2\Delta P d}{\rho u^2 L}$ ($\Delta P$ is the pressure drop over a distance $L$) for water-to-oil and oil-to-water continuous experiments and for direct experiments are presented as a function of the dispersed phase volume fraction. As can be seen for continuous experiments the friction factor increases with increasing dispersed phase volume fraction, until it reaches its maximum during phase inversion after which it decreases significantly. During direct
Chapter 8. Dispersed oil-water-gas flow through a horizontal pipe

Figure 8.6: Inversion map. The upper line gives the oil volume fraction at the point of inversion as measured during water-to-oil continuous experiments. The lower lines represent the oil volume fraction at inversion as measured during oil-to-water continuous experiments. The region in between the two lines is the ambivalence region, where both oil and water can be the continuous phase. Also the oil volume fraction at inversion as measured during the direct experiments is indicated.

experiments an increase in pressure drop was observed at a dispersed phase volume fraction between 0.5 to 0.6. At that oil volume fraction water-continuous and oil-continuous regions are simultaneously created during mixing process. Those region flow downstream while interacting, which causes an increase of the friction factor. Similar experiments in a horizontal pipe were carried out by Ioannou et al. (2005). They observed that at the point of phase inversion the pressure drop as well as the conductivity were very unstable. Pal (1993) observed a strong increase in effective viscosity at phase inversion during direct experiments in a pipe.

8.4 Gas-liquid dispersed flow

We started the gas experiments with the injection of nitrogen in a single liquid (water). At the high liquid velocities that we applied, dispersed bubble flow occurred at low enough values of the gas volume fraction. At volume fractions in the range of 0.25 to 0.30 a transition to elongated bubble flow or slug flow was observed, in agreement with literature results (Hewitt et al. (1986); Andreussi et al. (1999)). Figure 8.8 shows the gas fraction and bubble size as function of the vertical distance to the pipe center at three different locations for a water-nitrogen flow with a mixture velocity of 3 m/s. As can be seen from this figure even at a mixture velocity much higher than as used for the oil-water experiments, it is difficult to uniformly disperse the gas in the pipe due to the buoyancy force on the bubbles. At a low gas fraction small bubbles are evenly distributed in the pipe cross-section. However, at higher gas fractions large bubbles are created which tend to accumulate at the top of the pipe (leading to a transition to elongated bubble flow or slug flow).
8.4. Gas-liquid dispersed flow

**Figure 8.7**: Friction factor as a function of oil volume fraction for continuous and direct experiments.

**Figure 8.8**: Gas volume fraction and bubble size in a cross section of the pipe at different average gas volume fractions for a mixture velocity of 3 m/s. "0" indicates the pipe center, "-4" indicates 4 mm below the center, and "+4" is 4 mm above the center.
Chapter 8. Dispersed oil-water-gas flow through a horizontal pipe

8.5 Oil-water-gas dispersed flow

As mentioned in the introduction there is a limited number of studies dealing with three-phase flows. Some studies deal with the establishment of the flow pattern map. In our research we concentrated our effort on the oil-water-nitrogen dispersed flow pattern. The continuous experiments were an extension of our previous oil-water experiments. The direct experiments were similar to the experiments performed by Descamps et al. (2006, 2007) in a vertical pipe. Only in the horizontal pipe configuration (that we used) a higher mixture velocity had to be used to ensure dispersed flow.

Before starting the continuous and direct three-phase flow experiments we tried to find out, in which liquid phase the gas bubbles were present. To that purpose we performed a series of direct experiments, in which a liquid (oil or water) with nitrogen bubbles was injected simultaneously with the other liquid into the pipe. Therefore nitrogen was first dispersed into one of the liquids using an injection needle to ensure the generation of small bubbles. Thereafter this liquid-gas mixture was pumped together with the other liquid into the pipe. Figure 8.9 shows some pictures taken during these experiments. The top-left and bottom-left pictures show the results for an experiment, for which after the mixing process at the entrance to the pipe water has become the continuous phase with oil drops and nitrogen bubbles. In the top-right and bottom-right pictures oil is the final continuous phase with water drops and nitrogen bubbles. In the top pictures the volume fraction of the dispersed liquid phase is 0.07 and in the bottom pictures 0.15. The gas volume fraction is 0.03 for all cases. Because the refractive index between oil and water is small, the oil or water drops have a gray interface. However, the refractive index between oil and gas or between water and gas is much larger, which causes a black gas-liquid interface. This property allows us to easily recognize bubbles and drops in the pictures. We found that in the pipe the gas bubbles are always in the continuous liquid phase after the mixing region in the entrance part of the pipe. Also for the case that the liquid with gas bubbles that is pumped into the pipe, becomes the dispersed liquid phase in the pipe. This is an important result. With increasing concentration of the dispersed liquid phase the gas bubbles tend to grow in size.

8.5.1 Continuous experiments

A continuous experiment was started by flowing the first liquid at a certain velocity through the pipe loop and then gradually increasing the concentration of the second liquid by injecting it (in the form of drops) at a certain flow rate into the first liquid. At a certain moment in time injection of the second liquid was stopped (when the desired dispersed phase volume fraction was reached) and injection of gas was started. The gas concentration was increased up to the moment that a transition to elongated bubble flow took place.

8.5.2 Continuous experiments with water as continuous phase

In the first series of continuous experiments water was the continuous phase with oil injected in it up to a certain volume fraction. As mentioned nitrogen gas was then injected into it up to the point of elongated-bubble flow formation. Figure 8.10 shows the measured friction factor for
Figure 8.9: Gas bubbles are always found in the continuous liquid phase during a three-phase flow through a pipe. In the left pictures water is the continuous phase; in the right pictures oil is the continuous phase.
Chapter 8. Dispersed oil-water-gas flow through a horizontal pipe

Figure 8.10: Friction factor for oil-water-gas flows as a function of the gas volume fraction for five different values (0.0, 0.1, 0.2, 0.36 and 0.55) of the oil concentration. Water is the continuous phase. The mixture velocity was 3 m/s.

these experiments as function of the gas volume fraction for five different values (0.0, 0.1, 0.2, 0.36 and 0.55) of the oil concentration.

As can be seen the friction factor for the water-gas case (oil concentration is 0.0) is steadily increasing with gas concentration up to a concentration of 0.3, at which point a very strong increase of the friction factor occurs. The maximum and average bubble size as measured by means of the optical fibre probe during the water-gas experiments are shown in Fig. 8.11. The maximum bubble size is steadily increasing up to a concentration of 0.27, when large bubbles are created and the flow pattern is changing from dispersed bubble flow to elongated bubble flow. This is in the agreement with other experiments (Hewitt et al. (1986)).

Figure 8.10 and Figure 8.11 show also the friction factor, maximum and average bubble size for water-oil-gas experiments at different oil volume fraction. Even a small volume fraction of oil (0.1) has already a significant influence on the transition of dispersed bubble flow to elongated bubble flow. This transition occurs at a lower gas fraction (around 0.16). This is perhaps surprising, as the friction factor for this case is larger than for the water-gas experiments and, therefore, break-up of bubbles could be expected to be easier than for the water-gas case. Obviously the oil drops avoid this from happening and stimulate the creation of large bubbles. Increasing the oil volume fraction to 0.2 and 0.36 does not change these results very much. The transition to elongated bubble flow was always measured to be close to a gas concentration of 0.16 and we found a further small increase in friction factor and bubble size. Only at a much higher oil volume fraction of 0.55 there was a significant decrease again in the gas concentration where transition to elongated bubble flow took place. It is then almost impossible to create a dispersed bubble flow. Even the injection of a small amount of gas causes transition to elongated bubble flow.

Figure 8.12 shows similar results, only this time the mixture velocity has values of 2 m/s, 3 m/s and 4 m/s and the oil volume fraction is 0.36. For a mixture velocity of 2 m/s the transition of dispersed bubble flow to elongated bubble flow occurs at a low gas concentration of around 0.1. At 3 m/s the transition is shifted to a higher gas fraction of around 0.17. At 4 m/s the influence of the oil drops is qualitatively similar as observed for the case of a mixture velocity of 3 m/s.
8.5. Oil-water-gas dispersed flow

Figure 8.11: Averaged and maximum bubble size for oil-water-gas flows as a function of the gas volume fraction for five different values (0.0, 0.1, 0.2, 0.36, and 0.55) of the oil volume fraction. Water is the continuous phase. The mixture velocity was 3 m/s.

At 4 m/s the transition starts also at a gas fraction 0.17, but the maximum bubble size is smaller than at a mixture velocity 3 m/s. In figure 8.12 also the probability density function of the bubble size for the experiments at 3 m/s is presented. At a low gas fraction of 0.03 most of the bubbles are small. Increasing the gas fraction increases the bubble size. A strong peak in the bubble size is observed for a gas fraction of 0.12 and 0.18. Despite the fact that at a gas fraction 0.18 the maximum bubble size increases strongly, only a small decrease in the probability of the small bubbles is observed. Most of the bubbles are still small (so the flow is still dispersed). However, the creation of even a small number of large bubbles has a strong influence on the flow (see friction factor in Fig. 8.10).

8.5.3 Continuous experiments with oil as continuous phase

Oil continuous experiments started with pumping oil through the pipe loop and then injecting water in it. After achieving the desired water volume fraction, the water injection was stopped and gas injection started. Figure 8.13 shows the friction factor as measured during the oil continuous experiments for four different values of the water volume fraction (0.0, 0.37, 0.51, 0.61) and at a mixture velocity 3.5m/s. The friction factor behaves in the same way as for the water-continuous experiments. It is steadily increasing with increasing gas volume fraction and at the point of transition to elongated bubble flow it becomes very large. Figure 8.14 presents the maximum bubble size and average bubble size for the oil continuous experiments. As in the previous experiments, also here the transition from dispersed bubble flow to elongated bubble flow is influenced by the presence of the injected liquid in the form of drops. Only this time there is a visible difference between the transition at a water volume fraction of 0.2 and 0.37. This transition occurs at a gas volume fraction of 0.25 and 0.15 respectively. At a dispersed phase volume fraction of 0.51 a dispersed bubble flow is almost impossible, similarly to the water continuous experiments. An injection of a small amount of gas already causes the transition to elongated bubble flow. In the
oil continuous experiments the influence of the water drops on the average bubble size is stronger than the influence of the oil drops on the bubble size during the water continuous experiments. Furthermore there is another difference. The concentration of the water drops has a stronger influence on the bubble size than the concentration of the gas bubbles. For the water-continuous flow that was the opposite.

8.5.4 Bubble behavior during phase inversion in a continuous experiment

It is not easy to observe the influence of bubbles on the phase inversion process. During continuous experiments phase inversion only takes place at a high volume fraction of the dispersed phase and as we have shown in the preceding sections dispersed bubble flow is then no longer possible. Another difficulty is the fact, that it is not easy to control the gas volume fraction when injecting liquid close to the inversion point. A way out to these difficulty is to carry out the experiments in the region where multiple drops are formed. As was shown by Piela et al. (2006) when injection of water in an oil continuous flow is stopped at a high water volume fraction (0.54-0.64), production of multiple drops (which entraps parts of the continuous oil phase) increases the effective dispersed phase volume fraction and finally leads to inversion. During such experiments no injection is needed and it is certain that the gas fraction is the same before and after inversion. All experiments were started in the same way as during the continuous oil-water experiments with oil as the continuous phase. The mixture velocity was kept constant at 3.5 m/s. After achieving the desired water volume fraction and gas volume fraction, the injection was stopped and the mixture was circulated through the pipe loop. As with the oil-water experiments the pressure drop was increasing in time up to the inversion point. Figure 8.15 shows the friction factor, maximum and average bubble size during the phase inversion process. Before inversion the water volume fraction is 0.59. At that concentration dispersed phase flow is not possible.
8.5. *Oil-water-gas dispersed flow*

Figure 8.13: Friction factor for oil-water-gas flows as a function of the gas volume fraction for four different values (0.0, 0.37, 0.51, 0.61) of the water volume fraction. Oil is the continuous phase. The mixture velocity is 3.5 m/s.

Figure 8.14: Averaged and maximum bubble size for oil-water-gas flows as a function of the gas volume fraction for four different values (0.0, 0.2, 0.37, 0.51) of the water volume fraction. Oil is the continuous phase. The mixture velocity is 3.5 m/s.
Chapter 8. Dispersed oil-water-gas flow through a horizontal pipe

So the average bubble size is large (around 3 mm) and the largest elongated bubbles are of the order of 20-25 mm. Due to the continuous entrapment of oil into the water drops the friction factor increases with time and at time 535 s inversion occurs. During the inversion process the bubbles are quickly broken into small ones, so that after inversion a dispersed bubble flow pattern is observed.

The explanation is likely due to the difference in dispersed phase concentration before and after inversion. Before inversion the volume concentration of multiple water drops (with increasing oil droplets inside) is very large ($\geq 0.59$) and there is not much space left for the gas bubbles. This causes coalescence of the bubbles and hence creation of large bubbles (slugs). After inversion the volume concentration of oil drops is 0.31 which is much lower (also because oil drops do not easily form multiple drops), which leaves more space and less coalescence for the bubbles in the continuous phase.

During our experiments we observed a strong influence of the bubbles (slugs) on the phase inversion process in the multiple drop formation region. Figure 8.16 shows, that the presence of gas is slowing down the inversion process. The reason is, that the formation and growth of multiple drops takes place at a lower pace. In the formation and growth process of multiple drops the collision between drops plays an important role and the presence of bubbles decreases the chance on such collisions. This result is important for practical applications. We estimated for a typical case, that the slow down of phase inversion causes the inversion to take place over a pipe length of about 10 km in stead of about 1 km for the case without the presence of gas bubbles. This increased phase inversion length causes a considerable increase of the pressure drop over the production tubing.
8.5. Oil-water-gas dispersed flow

8.5.5 Direct experiments

During the direct experiments oil, water and nitrogen gas were simultaneously injected into the pipe at a certain (constant) mixture composition. Inside the pipe both liquids and the gas mix and flow as a three-phase dispersed mixture. As already shown earlier in Figure 8.7 for a two-phase oil-water flow the friction factor is nearly equal to the friction factor of the single phase flow at all dispersed phase concentrations except in the region with an oil volume fraction of 0.5-0.6 where phase inversion occurs. In the phase inversion region the friction factor increases significantly. Figure 8.17 shows the friction factor and maximum bubble size for two-phase oil-water flows as well as for three-phase flows with a gas volume fraction of 0.03. The mixture velocity was 2m/s. As can be seen a small amount of gas does not influence the oil-water flow results. Phase inversion still occurs at the same oil volume fraction and the peak in the friction factor and bubbles size remains more or less the same. So the presence of gas is not influencing the inversion process, but this process has a considerable effect on the bubbles as we have discussed before. Figure 8.18 shows similar results for the friction factor for a mixture velocity 3 m/s. Although the results for this case are somewhat inaccurate, it seems that phase inversion is still taking place at the same oil volume fraction, but the presence of bubbles increases the friction factor.

Figure 8.19 shows the variations in time in the friction factor and bubble size during a single direct experiment at oil volume fractions of 0.53 and 0.61. At an oil volume fraction of 0.53 we are in the phase inversion region, at 0.61 we are outside it. At 0.53 strong variations in the friction factor and the maximum bubble size are observed. At phase inversion water and oil continuous regions are continuously created (for more details see our earlier publications), which causes this effect on the friction factor and bubble size. At the higher oil volume fraction of 0.61 oil is the continuous phase with water drops in it. The friction factor is stable and smaller than the friction factor at phase inversion. Also the bubble size is much smaller than in the inversion region.
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Figure 8.17: Friction and maximum bubble size factor for oil-water and oil-water-gas experiments at a mixture velocity 2 m/s and gas volume fraction of 0.03. The points represent the oil-water flows and the lines the oil-water gas flows.

Figure 8.18: Friction factor for oil-water and oil-water-gas experiments at a mixture velocity 3 m/s and gas volume fractions of 0.07 and 0.13. The points represent the oil-water flows and the lines the oil-water gas flows.
8.6 Conclusions

In this section a number of points are discussed, that are in our opinion important outcomes of our study.

Let us first discuss the influence of drops on a dispersed bubble flow. We found that the presence of drops causes the transition from dispersed bubble flow to elongated bubble flow at a lower gas volume fraction. This can be explained by the fact, that the drops causes an increase in the effective volume concentration of bubbles in the continuous liquid phase. (Gas bubbles are always present in the continuous liquid phase.) This leads to an increase in bubble coalescence and hence a transition to elongated bubble flow at a lower gas concentration. When the volume fraction of drops is larger than about 0.5, dispersed bubble flow is even not possible anymore. There is a difference, however, between the situations that oil is the continuous phase and that water is the continuous one. In an oil continuous phase the effect of water drops on the quickening of the transition is stronger than the effect of oil drops when water is the continuous phase. We cannot explain this difference. The calculated bubble size is nearly the same for both types of experiment: 1.3 mm when water is the continuous phase and 1.2 mm when oil is the continuous phase (in agreement with the experimental observations of 0.15 mm and 0.14 respectively.)

A next point is the influence of the phase inversion process on the gas bubbles during a continuous experiment. It is important to point out, that during such experiments the dispersed drop concentration at phase inversion is high (much higher than 0.5). So just before inversion a dispersed bubble flow is not possible. Large bubbles are present and the flow pattern is elongated bubble flow. During the inversion process the bubbles break quickly and as the dispersed drop concentration is then much lower than before inversion a dispersed bubble flow is generated. We found that the bubbles do not have a significant effect on the critical concentration and pressure gradient at phase inversion, but phase inversion has a strong influence on the bubbles.
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Conclusions and perspectives

9.1 Conclusions

The aim of this study was to investigate experimentally dispersed oil-water and oil-water-gas flows through a pipe. Different types of experiments were conducted: continuous, direct and discontinuous. Different experimental techniques were used. We achieved much insight concerning the inversion process in an oil-water dispersed pipe flow and concerning the influence of gas bubbles on this process. We also made an effort to model phase inversion theoretically.

9.1.1 Experimental techniques

Different experimental techniques were used to investigate the properties and processes occurring in a two-phase and three-phase pipe flow. The following conclusions can be drawn:

- The conductivity probe and differential pressure transducers are good measurement tools to detect phase inversion.
- The visualization cell and high speed camera enables to observe the phase inversion process in time. All inversion stages can be clearly recognized and the observations can explain the results made with other experimental techniques.
- Even a one-tip optical probe can give much information about bubble behavior in a dense dispersion and during phase inversion.
- Our simple and easy-to-modify set-up gave a lot of freedom in performing different type of experiments (continuous, direct and discontinuous). This was crucial for our study.

9.1.2 Dispersed oil-water flow

The first part of our research was dedicated to the study of oil-water dispersed pipe flow. One of the most important processes in such a dispersed two-phase flow is phase inversion. We
performed many experiments to acquire more information about this process. Also two different modeling approaches were proposed. Important conclusions are:

- During a continuous experiment phase inversion can be postponed to a high value of the dispersed phase volume fraction (even higher than 0.9).

- Phase inversion is strongly dependent on the injected phase volume fraction.

- There is an ambivalence region. Which phase is continuous in a dispersed oil-water pipe flow depends on the 'history' of the experiment.

- The pressure drop for an oil-water pipe flow far below the critical dispersed phase volume concentration is nearly equal to the pressure drop of a single phase flow of the continuous phase. The pressure drop at higher concentrations of the dispersed phase (higher than 0.6) is strongly increasing with the dispersed phase volume fraction.

- The inversion map can be predicted with the Ginzburg-Landau model, when two adaptable parameters are fitted to experimental results.

### 9.1.3 Dispersed oil-water-gas flow

The second part of our study was dedicated to the investigation of dispersed oil-water-gas flow through a pipe. The following results were found:

- Gas bubbles are always present in the continuous phase.

- In a dense dispersion it is not possible to disperse gas in the form of small bubbles.

- The bubbles have no significant influence on the phase inversion process. The pressure drop at inversion and the critical dispersed phase volume concentration at which inversion takes place, remain the same.

- However, phase inversion has a strong effect on the gas bubbles. The size of gas bubbles is strongly changing during the inversion process. Just before inversion large bubbles are present and the flow pattern is elongated bubble flow. During the inversion process the bubbles break-up quickly and as the dispersed drop concentration after inversion is much lower than before inversion a dispersed bubble flow is present after inversion.

### 9.2 Perspectives

The complexity of (phase inversion in) three-phase pipe flow made it impossible to pay attention to all interesting phenomena that we encountered. Several important topics remain. Some of them are listed below.
• The influence of the injected phase volume fraction on the inversion map was studied by us. However, the influence of other parameters (density ratio, viscosity ratio, HLB, oil-water surface tension, contact angle) on the inversion map was not investigated. Such an investigation is necessary to establish a more general inversion map (valid for different oil-water systems).

• Inversion in the multiple drop formation region occurs without injection. In the literature two mechanisms were proposed to explaining this phenomenon: 1) deformation of drops due to turbulent forces and 2) multi-drop collisions where part of the continuous phase is trapped between colliding drops. It is important to study these mechanisms in more detail.

• Modeling of the inversion process is difficult. As shown in this work the mean-field theoretical model gives a good description, but only after two parameters in the model have been fitted to experimental results. Further modeling work is needed.

• In a three-phase flow the presence of drops causes the transition from dispersed bubble flow to elongated bubble flow at a lower gas volume fraction. This can be explained by the fact, that the drops causes an increase in the effective volume concentration of bubbles in the continuous liquid phase. (Gas bubbles are always present in the continuous liquid phase.) This leads to an increase in bubble coalescence and hence a transition to elongated bubble flow at a lower gas concentration. When the volume fraction of drops is larger than about 0.5, dispersed bubble flow is even not possible anymore. There is a difference, however, between the situations that oil is the continuous phase and that water is the continuous one. In an oil continuous phase the effect of water drops on the quickening of the transition is stronger than the effect of oil drops when water is the continuous phase. We cannot explain this difference.

• The presence of gas bubbles influences the phase inversion process in the multiple-drop-formation region. The production of multiple drops is becoming slower with increasing gas volume fraction. We have no explanation for this result.

• During inversion in a direct experiment water and oil continuous regions co-exist. We wonder what happens to the gas bubbles in this case. How is the gas moving from one region to another?
Bibliography


List of publications

Journals


Conference Contributions


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Krzysztof Piela, Delft, September 2008
About the author

Krzysztof Piela was born on 6 September 1979 in Żywiec, Poland. He graduated from secondary school at the V Liceum Ogólnokształcące in Bielsko-Biała. After graduation in 1998 he started the Mechanical Engineering study at Warsaw University of Technology. During his studies in 2002 he moved to Goteborg in Sweden for half a year to study at Chalmer University of Technology as part of Erasmus student exchange program. In 2003 he defended his master thesis: *Adaptive mesh refinement for two-dimensional gas detonation simulations*. A few months later he started his PhD study at the Laboratory of Aero & Hydrodynamics at Delft University of Technology which led to this thesis.