MASTER

Chaotic mixing in the extended periodic cavity flow

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Abstract

In this report chaotic mixing in an extended, periodic cavity flow is studied. This flow is based upon the regular periodic cavity flow, which has been extensively studied in the past, for instance by Leong and Ottino (1989). The extension consists of a central placed cylinder, that can be rotated. The central placed cylinder can have a significant influence on the flow, especially when the cylinder is fixed or rotating in the opposite direction of the main fluid flow. When the cylinder rotates in the same direction of the fluid flow, the streamlines resemble those of the regular cavity flow.

Experiments with a Newtonian fluid are performed in which the deformation of an initial circular blob of tracer material is followed in the extended periodic cavity flow. Deformation patterns are simulated using an analytical solution of the velocity field (Galaktionov, 1995). These resulting deformation patterns show a good resemblance with those from the experiments. Further, simulations have been performed using a numerically computed velocity field. These deformation patterns are (nearly) identical to those using the analytical velocity field. Numerically computed velocity fields can also be obtained for flows for which analytical solutions are not available. A brief study on the influence of generalized Newtonian fluid behaviour shows that the mixing capabilities of the flow decrease.

The resulting mixtures are analysed and the quality of mixing is measured using several methods: the correlation coefficient and the power spectrum, the intensity of segregation, the square density and the entropy of the mixture. Although the results obtained from experimental and simulated mixtures do not correspond for these measures, it is seen that trends are followed. The protocol used to process the images of the experiments seems an important factor.
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Chapter 1

Introduction

1.1 Mixing

In polymer processing mixing plays an important role. Mixing can have a large influence on the resulting material properties. Extrusion is one example of polymer processing in which mixing can take place. An extruder is not just a way to transport polymers, also mixing takes place when more than one polymer is processed. Many different extruder screws have been designed to optimize the mixing capabilities of extruders (Meijer et al., 1992). However, to optimize mixing, one must first find an answer to the question: What is mixing?

Mixing can be defined as follows: “A mixture is a combination of two or more substances, and mixing is an operation whose purpose is to increase the spatial homogeneity of a mixture” (Tucker, 1991). In this report, only mixtures of fluids are considered.

Two kinds of mixing are distinguished: distributive mixing and dispersive mixing. The goal of distributive mixing is to distribute one fluid homogeneously among another. Material is deformed, but does not break. Dispersive mixing, however, aims to reduce the size of droplets which are present in the continuous phase of the bulk component.

In this report, we will only consider mixing of two immiscible fluids (no diffusion) without inter-facial stress (deformed material does not break up). Mixing of fluids without molecular diffusion appears in many processes in industry and nature. For example in the already mentioned mixing of two polymers and the geophysical flows in oceans.

Suppose that one fluid is present in the continuous phase or bulk material as a drop or filament. It is clear that mixing implies deformation of the material, that leads to an increase in the inter-facial area between the two components. To analyse the efficiency of mixing, we first regard the stretching rate and the stretching efficiency. The stretching rate \( \dot{\varepsilon} \) is defined as follows (Ottino, 1989)

\[
\dot{\varepsilon} = \mathbf{D} : \mathbf{n} \mathbf{n}
\]  

(1.1)

with \( \mathbf{D} \) the rate of deformation tensor and \( \mathbf{n} \) the orientation vector of an infinitesimal filament \( d\varepsilon \) (\( \mathbf{n} = \frac{d\varepsilon}{|d\varepsilon|} \)). The stretching efficiency \( e_f \) is defined as the ratio of the stretching rate and its upper bound \( |\mathbf{D}| \) (Ottino, 1989)

\[
e_f = \frac{\mathbf{D} : \mathbf{n} \mathbf{n}}{\sqrt{\mathbf{D} : \mathbf{D}}}
\]  

(1.2)
Consider a two-dimensional extensional flow (figure 1.1):

\[
\frac{dx}{dt} = \dot{\varepsilon} x ; \quad \frac{dy}{dt} = -\dot{\varepsilon} y \tag{1.3}
\]

At \( t = 0, x = X, y = Y \) and we define \( \vec{X} = X \vec{e}_x + Y \vec{e}_y \) and the orientation vector \( \vec{m} = d\vec{X}/|d\vec{X}| \). Then

\[
x = X e^{\dot{\varepsilon} t} ; \quad y = Y e^{-\dot{\varepsilon} t} \tag{1.4}
\]

The lineal stretch \( \lambda \) related to \( \vec{m} \) is given by

\[
\lambda^2 = m_x^2 e^{2\dot{\varepsilon} t} + m_y^2 e^{-2\dot{\varepsilon} t} \tag{1.5}
\]

and the specific rate of stretching of \( \lambda \) by

\[
\frac{D \log \lambda}{Dt} = \frac{\dot{\varepsilon}(m_x^2 e^{4\dot{\varepsilon} t} - m_y^2)}{m_x^2 e^{4\dot{\varepsilon} t} + m_y^2} \tag{1.6}
\]

where \( \frac{D}{Dt} \) denotes the material derivative and \( \log \lambda \) the natural strain. The magnitude of \( \textbf{D} = \dot{\varepsilon} \vec{e}_x \vec{e}_x - \dot{\varepsilon} \vec{e}_y \vec{e}_y \) is equal to \( \sqrt{2}\dot{\varepsilon} \). The long time value of the efficiency \( e_f = (D \log \lambda / Dt)/(\textbf{D} : \textbf{D})^{1/2} \) is equal to \( \dot{\varepsilon}/(\sqrt{2}\dot{\varepsilon}) = \frac{1}{2} \sqrt{2} \approx 0.707 \), unless \( m_x = 0 \). The efficiency reaches a constant value with time. A flow is efficient with regard to stretching if the time averaged value of \( D \log \lambda / Dt \) does not decay to zero.

In a bounded flow however, it is not possible to obtain infinite exponential stretching in one direction and at most linear stretch is possible. Therefore, in a stationary bounded flow the typical stretching efficiency \( e_f \) decays proportional to \( t^{-1} \). To mix and stretch efficiently in a bounded flow, it is necessary to reorientate the material. This becomes clear when we consider the 'baker's transformation' (figure 1.2), that stretches, cuts and fuses material (read 'dough'). From figure 1.2, it is clear that the inter-facial area increases exponentially. In a continuous flow, it is in general not possible to cut and fuse. We can come close to the
Introduction

‘baker’s transformation’ by stretching and folding as shown in figure 1.3. In this case it is
important not to lose too much material during folding (white in the picture). This kind of
transformation is referred to as a horseshoe map (Ottino, 1989).

The presence of horseshoe maps in a flow indicates chaos. More generally, a flow displays
chaos if it satisfies one of the following three conditions (Ottino, 1989):

1. the flow is sensitive to initial conditions (mathematical: the flow has a positive Liapunov
   exponent);
2. the flow has transverse homoclinic and/or heteroclinic points;
3. the flow produces horseshoe maps.

These definitions are discussed extensively in the book by Ottino (1989).

1.2 Objectives and outline of the thesis

In this report a chaotic flow is studied in which stretching and folding of material occurs: the
periodic cavity flow. The cavity flow can be regarded as a simple two-dimensional representa-
tion of a part of the three-dimensional flow in an extruder (see figure 1.4). The rotation of the
screw as shown in the first picture of figure 1.4, causes a rotation and a translation in axial
direction of the material between the screw and the static wall. Neglecting rotation, the walls
of the screw in the rectangular box seem to move from left to right. This can be represented
Chapter 1

as a translation of the top wall in the opposite direction, as is shown in the second picture. This situation is also obtained for the cavity flow when the top wall moves from right to left.

![Flow in an extruder and the cavity flow](image)

**Figure 1.4: Flow in an extruder and the cavity flow**

For the periodic cavity flow, simulations of deformation patterns, based on the analytical velocity field provided by Meleshko and Peters (1996) shows good agreement with existing experimental results (Leong and Ottino, 1989). In this report it is investigated whether simulations on a cavity flow with a central placed cylinder (the extended periodic cavity flow) agree with experimental results. Extending the setup with a central cylinder makes the flow more complex. Simulations based upon a velocity field computed by numerical methods are compared to simulations based upon the analytical velocity field, in order to determine the usefulness of the methods for arbitrary two-dimensional flows and time dependent and/or non Newtonian fluid behaviour.

In the extended periodic cavity flow stretching and folding of material takes place as well. For the interpretation of these mixtures it is necessary to investigate measures that define the quality of the mixture and determine these measures for both simulations and experiments. Therefore, pictures taken from the experiments have to be digitized and processed.

The outline of this report is as follows: chapter 2 shortly discusses previous work on the periodic cavity flow and simulations based on an analytical and a numerical velocity field are shown. Experiments and simulations for the extended cavity flow are presented in chapter 3, for Newtonian fluids. In chapter 4 the influence of generalized Newtonian fluid behaviour on mixing in the extended periodic cavity flow is discussed. The quality of mixing and measures for the mixed state are described in chapter 5. Finally, in chapter 6 conclusions and recommendations are presented.
Chapter 2

Cavity flow

2.1 Experimental setup

A typical example of stretching and folding is obtained in the 'cavity flow' (Leong and Ottino, 1989, Ottino, 1989). The cavity is a rectangular cell, filled with fluid. Two opposing walls are fixed, while the two other opposing walls can move. The setup used in our experiments is similar to the one used by Leong and Ottino (1989). It can be divided in three functional units: the belt units (the moving walls), the static walls and the supporting structure (see figures 2.1 and 2.2).

![Figure 2.1: Schematic of the cavity flow apparatus](image)

Two belt units, each consisting of two knurled rollers (50 mm diameter, 180 mm long) and a rubber belt (150 mm deep, 2 mm thick) function as moving walls. These belt units are
Chapter 2

driven by an electromotor (VDO, type: M42x25/I) with gear reduction. The motor drives one roller directly with a tooth shaped belt. The second roller is driven by the rubber belt. A time clock (Kaiser, automatic timer cpd 4212) turns the two electromotors on and off. The velocity is changed by regulation of the voltage supplied to the electromotors. A tachometer (Servo-Tek Products Co, type: SA-740A-2) is connected to the second roller. Using gear wheels, the number of revolutions is amplified by a factor 25 to increase the signal of the tachometers. Additional tension to the belt is supplied by an adjustable dancer. The dancer also suppresses creeping of the belt due to friction drive of the rollers and the belt.

The two static walls consist of PMMA plates (46.5 mm wide, 6 mm thick and 185 mm deep) which are connected to shafts. These shafts are attached to a supporting plate. The edges of the PMMA plates are tapered, which minimizes the contact with the belt. The supporting frame consists of two aluminium plates, connected with each other by four aluminium bars (215 mm long). The rollers and dancers are connected to the upper and lower aluminium plate by bearings. The electromotors and tachometers are adjusted to the upper plate.

The entire setup is placed in a container of glass (265 x 215 x 230 mm) filled with fluid. Under the glass container a mirror is placed at an angle of 45°, to make pictures of the cavity from the bottom side.

Optionally, a cylinder (14 mm diameter, 275 mm long) can be placed in the cavity. The cylinder can be fixed or driven by an electromotor (Maxon, type: RE025-055-35EBA201A). The cylinder and motor are mounted on a small aluminium plate, that can translate over two parallel beams on the upper plate of the frame, parallel to the moving walls.

To study the mixing process, the deformation of a blob of tracer is examined. The tracer is injected with a capillary tube, connected to a syringe, about 2 – 10 mm beneath the free surface of the fluid (glycerine, Lamers & Pleuger, 2LZ250003), at a preselected location. In
our studies a passive tracer is used. The tracer is a fluorescent dye, made by dissolving fluorescent powder (Aldrich, fluorescein salt) in the glycerine. The diffusion coefficient of the tracer is negligible (Leong and Ottino, 1989) and its density is close to that of glycerine. It is supposed that the presence of the dye has no influence on the flow. The dye is excited by two ultraviolet lamps (UVP, model B-100 AP). These are placed on both sides of the static walls to minimize shadow effects. When excited, the dye colours yellow, while the normally colourless glycerine appears blue due to some UV-absorption.

The experiments are recorded on video tape, using a camera (Panasonic, F15 or Pulnix, TM-765), a VHS recorder (Panasonic, AG-6200) and visualized on a monitor (Philips, V6821/10). Experiments are compared with simulations, using a Panasonic WJ-MX10 video mixer and a VGA-PAL interface board, or using image processing systems (TIM, Difa Measuring Systems, and Corel PHOTO-PAINT, Corel Corporation).

2.2 Cavity flow

Various experiments and simulations are reported in literature on the cavity flow. The most reported dimension of the cavity (see figure 2.1), that is considered here also, is a ratio of width \(W\) to height \(H\) of the cavity of 1.67 (Leong and Ottino, 1989, Leong and Ottino, 1990, Ottino, 1990, Liu et. al., 1994, Meleshko and Peters, 1996, Galaktionov et al. 1996).

Flow or motion is represented by the point transformation

\[
\vec{x}(t) = (x(t)\vec{e}_x, y(t)\vec{e}_y) = \Phi_t(\vec{x}_0)
\]

If we consider an incompressible Newtonian fluid, then the Navier-Stokes equations hold

\[
\rho \frac{\partial \vec{v}}{\partial t} + \rho \vec{v} \cdot \nabla \vec{v} = -\nabla p + \eta \nabla^2 \vec{v} \tag{2.2}
\]

\[
\nabla \cdot \vec{v} = 0
\]

in which \(\rho\) denotes the density of the fluid, \(\vec{v} = u\vec{e}_x + v\vec{e}_y\) the velocity, \(t\) the time, \(p\) the pressure and \(\eta\) the dynamic viscosity. Gravity effects are not taken into account. If we consider the dimensionless form of the Navier-Stokes equation 2.2, we obtain

\[
Sr \frac{\partial \vec{v}'}{\partial t'} + \vec{v}' \cdot \nabla' \vec{v}' = -\nabla' p' + \frac{1}{Re} \nabla'^2 \vec{v}' \tag{2.3}
\]

The accents indicate the dimensionless parameters. \(Sr\) denotes the Strouhal number, which gives the ratio of the instationary inertial forces to the stationary inertial forces, and \(Re\) the Reynolds number, which gives the ratio of stationary inertial forces to viscous forces. For the cavity, we define these dimensionless numbers (Leong and Ottino, 1989)

\[
Sr = \frac{1}{TVW} \frac{H^2}{W} \tag{2.4}
\]

\[
Re = \frac{\rho V H^2}{\eta W} \tag{2.5}
\]

where \(T\) is the time of one period (see next section) and \(V\) is a characteristic velocity (i.e. the velocity of a moving wall).

If we first regard the stationary situation, then \(Sr = 0\) \((T \to \infty)\). If furthermore inertial
forces are considered to be small compared to viscous forces \((Re \ll 1)\), the stationary Stokes flow is obtained and equation 2.2 can be reduced to

\[
\eta \nabla^2 \vec{v} = \nabla p
\]  

(2.6)

To describe a two-dimensional incompressible flow, one can use the streamfunction \(\psi\). The streamfunction is related to the velocity field through

\[
v_x = \frac{\partial \psi}{\partial y} \quad \quad v_y = -\frac{\partial \psi}{\partial x}
\]  

(2.7)

with \(\vec{v} = (v_x \hat{e}_x, v_y \hat{e}_y)\). Combination of equation 2.6 and equation 2.7, leads to the following relation for the streamfunction

\[
\nabla^2 \nabla^2 \psi = 0
\]  

(2.8)

An analytical solution for \(\psi\) is obtained by Meleshko and Peters (1996) using the Lamé superposition method. The velocity field in the cavity can also be computed using numerical techniques, such as finite and spectral element methods. On all the boundaries the velocity of the wall is prescribed, following the no-slip condition. The streamlines of continuous stationary flows are shown in figure 2.3, for the case that only the top wall is moving, for the case that the top and bottom wall are moving in opposite directions and for the case the top and bottom wall are moving in the same direction.

\[
D = \frac{|d_{\text{top}}| + |d_{\text{bot}}|}{W}
\]  

(2.9)

in which \(d_{\text{top}}\) and \(d_{\text{bot}}\) are the displacements of the top and bottom wall respectively and \(W\) is the width of the cavity. In the following simulations, \(D\) is equal to 6.24.

\[\text{Figure 2.3: Streamline pattern of the cavity flow for the top wall moving (left), two walls moving in opposite directions (centre) and two walls moving in the same direction (right)}\]
Cavity flow

Figure 2.4: Deformation patterns of a line on the y-axis from \(-0.98H\) to \(0.98H\) using the velocity field of the Lamé superposition method and 8th order spectral element method. \(D = 6.24\), the period increases with \(\frac{1}{2}\) from left to right. In every picture, the line on the y-axis is depicted.

In figure 2.4 the deformation patterns are compared of initial lines on the y-axis for the velocity field computed by the analytic superposition method and a 8th order spectral element method (SEM) for two periods. The mesh used for the SEM computations is shown in figure 2.5. The computed simulations show a good agreement.

In figure 2.7 a simulation of the deformation pattern of a circular blob is presented for four periods. The simulations are based upon the analytical solution of the 2-D Stokes flow. The center of the tracer is located at \((x, y) = (1.15, 0.0)\), a hyperbolic point (Meleshko and Peters, 1996) and has a radius of 0.05.

From figure 2.4 and figure 2.7 it is seen that folds emerge. This can be described as follows: During each half of the period the streamline pattern is constant. At the switching of the walls (at period \(\frac{1}{2}, 1, 1\frac{1}{2}, 2\), etc.) however, the streamline pattern flips upside down. Locally, the flow direction changes and the stretched line is folded, thus forming horseshoe maps.

One of the conditions for a flow to display chaos is producing horseshoe maps (stretching and folding, Ottino, 1989). In the introduction it was explained that the forming of horseshoe maps is highly relevant for effective mixing. The emerging of folds indicates chaotic flow with efficient mixing.

If a material point returns exactly to its initial position after one period, but not before, we say the point is a periodic point of order 1. Similar, a material point is a periodic point of order \(n\) if it returns exactly to its initial position after \(n\) periods, but not before.

Periodic points are important for understanding mixing processes in the periodic cavity flow. There are three kinds of periodic points: elliptic points, hyperbolic points and parabolic points.
points, according to the deformation of the fluid in the neighbourhood of the point. If the point is elliptic, the fluid rotates around the point, but there is no exchange with the rest of the fluid. Elliptic points will form islands. Hyperbolic points have two invariant regions of inflow and outflow. Material in the neighbourhood of the hyperbolic point is stretched and folded. Therefore, hyperbolic points are centers of efficient mixing. Parabolic points are points that have only outflow or inflow. They can be the center of efficient mixing, but they do not occur in the cavity flow (Leong and Ottino, 1989). In figure 2.6 the physical representation of an elliptic (E) and hyperbolic (H) point are shown.
Figure 2.7: Simulation of a periodic cavity flow with $D = 6.24$. The period increases with $\frac{1}{4}$ from left to right. In every picture, the circular initial blob is depicted.
Chapter 3

Extended cavity flow

3.1 Cavity flow with central cylinder

In this chapter, the influence of a central placed cylinder in the cavity (see figure 3.1) on the flow and mixing properties is studied. This extension makes the flow more complex. The diameter \( d \) of the cylinder is 14 mm. The ratio of the diameter \( d \) of the cylinder to the height \( H \) is \( 14/46.5 = 0.3 \). This cylinder can be either fixed or rotated clock- or counterclockwise. Unless mentioned otherwise, the \( x \)- and \( y \)-coordinates are normalized by dividing \( x \) and \( y \) by \( H/2 \) such that the left and right wall are now located on the lines \( x = \pm 1.67 \) and the top and bottom wall on the lines \( y = \pm 1 \).

As we study mixing processes in the extended cavity flow, we consider a discontinuous operation of the top and bottom wall. When one wall is moving, the other one is fixed. In all experiments the top wall moves in positive \( x \)-direction, and the bottom wall in negative \( x \)-direction. Without the central cylinder, the fluid would rotate clockwise. Therefore we define the cylinder co-rotating when it turns clockwise, and counter-rotating when it turns counter-clockwise.

In figure 3.2 to 3.4 streamline patterns are shown of a flow with a continuous moving top wall and a fixed cylinder, a co-rotating cylinder and a counter-rotating cylinder. The tangential velocity of the cylinder \( U \) is equal to that of the moving wall (\( V_{\text{top}} \) and \( V_{\text{bot}} \)).
Comparison of the extended cavity flow with the cavity flow as discussed in chapter 2 shows that a central placed, fixed cylinder has a great influence on the streamline pattern. From figure 3.2 it is seen that one saddle point $S$ (point with inflow from two opposite regions and outflow to two opposite other regions), two vortices $V$ and two steady parabolic points $P$ on the cylinder occur. When the cylinder is counter-rotating (figure 3.3) a second saddle point $S$ occurs below the cylinder, while the parabolic points on the cylinder disappear. The strength of the vortices is increased compared to the flow with the fixed cylinder. Most of the streamlines pass the cylinder over the top. The vortices disappear when the cylinder is co-rotating with a sufficiently high velocity $U$ ($U \approx 0.2V_{\text{top}}$, see figure 3.4 and 3.5). These streamline patterns resemble that of the regular cavity flow most (see also figure 2.3). Now the flow is fully co-rotating and there are no saddle points. Notice that this does not mean that there are no periodic hyperbolic points.

Experiments and simulations are performed to study the mixing behaviour of the extended cavity flow with a fixed, a co-rotating and a counter-rotating cylinder. The blobs of tracer are placed on the positive $x$-axis, around to hyperbolic points. In all simulations and experiments, the top wall starts moving for half a period and then the bottom wall moves for half a period.
3.2 Simulations

The velocity field in the cavity is computed using the analytical solution by Galaktionov et al. (1996) and a spectral element method (SEM). The analytical solution is obtained using the Lamé superposition method. In figure 3.6 the analytically obtained velocity on the top wall is shown for the case that only this wall is moving (figure 3.2). It is seen that closer to the corners the velocity diverges from the prescribed constant value. However, the maximum error is still less than 3%. With the SEM, the stationary Stokes equation is solved using 8th order spectral elements and an approximate predictor corrector scheme. This scheme belongs to the class of continuous projection schemes. Therefore the degree of approximation for the velocity and the pressure may be taken the same and there is no need to fulfill the Brezzi–Babuška condition (Timmermans et al., 1996). The mesh used for the numerical computations is shown in figure 3.7. It is refined near the top wall, where shear rate gradients are highest.

Both the analytical superposition method and the SEM solution describe the velocity field accurate. The largest differences between the computed velocities are present in the regions near the top corners, where the shear rate gradients are the highest and numerical singularities exist. In figure 3.8 and figure 3.9 the absolute value of the difference between the analytically and numerically computed velocities divided by the velocity of the wall \( \frac{|v_{an} - v_{num}|}{V_{top}} \) is shown. The velocity in the singular corner points is explicitly prescribed. In figure 3.8 the velocity in the singular points is set equal to the velocity of the moving top wall. In this situation numerical inflow and outflow on the side walls near the top corners occurs. In figure 3.9 the velocity in the singular nodal points is set to zero. In this situation slip on the moving top wall near the top corners is present. The maximum difference is approximately 30% of the wall velocity. The difference between the computed velocities is the largest near the top corners. When using the slip condition, it is seen that the peak in the difference is slightly higher, while when using the in- and outflow condition the peak is wider. Except for these small regions near the top corners, the agreement between the analytical and numerical solution is extremely good. The difference between the computed velocities for a co- or counter-rotating cylinder gives similar results: the highest differences between the analytical superposition
Figure 3.8: Difference between analytical and numerical solution: The top corners have the same velocity as the top wall.

Figure 3.9: Difference between analytical and numerical solution: The top corners have the same velocity as the side walls (fixed).

The velocity fields computed by the analytical method and the SEM are used for computation of deformation patterns. In figures 3.10 and 3.11 the results are shown for the deformation patterns of initial lines on the positive and negative y-axis respectively. The patterns are computed by integration of the velocity field in time. With the numerical solution of the velocity field, the velocity in an arbitrary point in the cavity is computed by interpolation of the velocity in the surrounding nodal points, as is also performed by Ling (Ling, 1994). The analytical solution is available for any arbitrary point inside the cavity.

The tracer is represented by a finite number of points, that are placed on a line or on the boundary of a blob. The position of these points is calculated by integration of the velocity field in time, thus giving the boundary of the deformed tracer. The velocity of the top or bottom wall, the velocity of the cylinder and the time during which this velocity is applied is measured in the experiments and used for the simulations.

As the periodic cavity flow is a chaotic flow, points can separate exponentially no matter how close they were located initially. Therefore the locations of the points after integration are analysed. If the distance between two adjacent points exceeds a prescribed value (due to stretching), or if the angle between three adjacent points becomes less than a prescribed value (due to folding), a point is inserted on the initial boundary. This point is integrated using the velocity field to the current time. Using this algorithm, a smooth deformation pattern is obtained. Points are inserted on the initial boundary only where they are required. Therefore the computations do not have to be started with many points.

Simulations of deformation patterns of a circular blob are shown in figures 3.12 and 3.13 for a dimensionless wall displacement of $D = 5.0$ and $D = 7.5$ respectively. The initial tracers are circles with a radius of 0.05 with their centers at $(0.63, 0.00)$ and $(1.25, 0.00)$. These tracers are located around periodic hyperbolic points.

The initial location of the periodic points can be found using figure 3.14 (Galaktionov, 1995). It can be proved that since the streamlines are symmetric about the y-axis, points which are located on the y-axis at period $\frac{1}{4}$ and at period $\frac{3}{4}$, are periodic points of period 1.
Figure 3.10: Deformation patterns of a line on the positive y-axis from 0.32H to 0.98H using the velocity field of Lamé's superposition method and the spectral element method. $D = 5$, the cylinder is fixed. The period increases with $\frac{1}{2}$ from left to right.

(Meleshko and Peters, 1996). In figure 3.14 the location of these points at period $\frac{1}{2}$ is shown. From these locations, the positions of periodic points at period 0 can be computed. Whether a periodic point is elliptic or hyperbolic, depends on the deformation of the neighbourhood of the periodic point. By examination of 4 points lying around the periodic point, the deformation of the surrounding area and thus the kind of the periodic point can be determined. From these pictures it becomes clear that an originally circular blob can be stretched and folded extensively in the periodic cavity flow.
Figure 3.11: Deformation patterns of a line on the negative $y$-axis from $-0.32H$ to $-0.98H$ using the velocity field of Lamé's superposition method and the spectral element method. $D = 5$, the cylinder is fixed. The period increases with $\frac{1}{2}$ from left to right.
Figure 3.12: Deformation pattern of a circular blob for a discontinuous operation of top and bottom wall and a fixed cylinder. $D = 5$, center of the initial blob $(0.638, 0.0)$ with radius of 0.05. The period increases with $\frac{1}{4}$ from left to right.

Figure 3.13: Deformation pattern of a circular blob for a discontinuous operation of top and bottom wall and a fixed cylinder. $D = 7.5$, center of the initial blob $(1.25, 0.0)$ with radius of 0.05. The period increases with $\frac{1}{4}$ from left to right.
Figure 3.14: *The location of the periodic points on the y-axis (x = 0) after a quarter period, as function of the dimensionless displacement D. The circles indicate elliptic periodic points, the crosses hyperbolic periodic points* (Galaktionov, 1995)
3.3 Comparison of experiments and simulations

In figures 3.15 to 3.17 pictures are shown of experiments compared with simulations for a fixed cylinder, a co-rotating cylinder and a counter-rotating cylinder respectively. The white areas in the pictures are the deformed patterns from the experiments, the black lines represent the simulations of the boundary of the tracer. For the fixed and co-rotating cylinder the results are shown for three periods. For the counter-rotating cylinder only two periods are shown, due to numerical problems after two periods. The dimensionless wall displacements $D$ are based on measured velocities. The values of $D$ for figure 3.15, 3.16 and 3.17 are shown in table 3.1. The tangential velocity of the cylinder is 0.93 times the average velocity of the top and bottom wall for the case that the cylinder is co-rotating, and 0.92 times the average velocity of the top and bottom wall for the case the cylinder is counter-rotating. Results from comparison between experiments and simulations are also available on video tape.

Table 3.1: Cumulative dimensionless displacement in experiment and simulation

<table>
<thead>
<tr>
<th>Period</th>
<th>fixed cylinder (figure 3.15)</th>
<th>co-rotating cylinder (figure 3.16)</th>
<th>counter-rot. cylinder (figure 3.17)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\frac{1}{2}$</td>
<td>2.6</td>
<td>2.4</td>
<td>2.6</td>
</tr>
<tr>
<td>1</td>
<td>5.1</td>
<td>4.8</td>
<td>5.0</td>
</tr>
<tr>
<td>$1\frac{1}{2}$</td>
<td>7.7</td>
<td>7.2</td>
<td>7.6</td>
</tr>
<tr>
<td>2</td>
<td>10.2</td>
<td>9.6</td>
<td>10.0</td>
</tr>
<tr>
<td>$2\frac{1}{2}$</td>
<td>12.8</td>
<td>11.9</td>
<td>--</td>
</tr>
<tr>
<td>3</td>
<td>15.3</td>
<td>14.3</td>
<td>--</td>
</tr>
</tbody>
</table>

3.4 Discussion

First the agreement between the experiments and the simulations (figure 3.15 to figure 3.17) is discussed. From figure 3.15 it is seen that the simulation resembles the experiment well for the discontinuous operation of the top and bottom wall with a fixed cylinder. At period $P = 1\frac{1}{2}$ (picture 3) however, a fold is present above the cylinder, which is not found in the simulation. When the cylinder is co-rotating (figure 3.16), it is seen that the tracer is stretched further in the experiments. The simulation shows a good agreement with the experiment during the first two periods, but starts to diverge afterwards. The simulations resemble the experiments the least in the case when the cylinder is counter-rotating. In the second picture we see a long striation of the tracer near the right and bottom wall, which is found in the simulation, while in the simulation striations are present around the cylinder that are not observed in the experiment.

There are several causes which underlie the differences between simulation and experiment. First the dimensions of the cavity can slightly deviate from the values used in the simulations due to errors in the positioning of the static walls and the central cylinder. However, these errors are estimated to be less than 1%. Moreover pressure gradients due to the flow in the cavity can deform the rubber belts. Since the velocity in the experiments is small ($V \approx$
Chapter 3

Figure 3.15: Simulations and experiments for a discontinuous operation of top and bottom wall with a fixed cylinder. The period increases with $\frac{1}{D}$ from left to right. $D \approx 5.1$, center of the initial blob (0.63, 0.05) with a radius of 0.10. The initial blob is depicted in every picture.
Figure 3.16: Simulations and experiments for a discontinuous operation of top and bottom wall with a co-rotating cylinder. The period increases with $\frac{1}{2}$ from left to right. $D \approx 4.8$, center of the initial blob $(0.91, 0.04)$ with a radius of 0.10. The initial blob is depicted in every picture.
Chapter 3

Figure 3.17: Simulations and experiments for a discontinuous operation of top and bottom wall with a counter-rotating cylinder. The period increases with $\frac{1}{2}$ from left to right. $D \approx 5.0$, center of the initial blob (0.87, 0.07) with a radius of 0.10. The initial blob is depicted in every picture.
19 mm/s) and the belts are under tension by the adjustable dancers, this effect can be neglected. A third cause might be the inflow and outflow which can occur if the static walls do not perfectly connect to the moving rubber belt.

In the simulations we consider the flow is stationary and inertial effects are neglected. From numerical simulations by Anderson (Anderson, 1996) it followed that it takes less then 0.2 s before a steady Stokes flow is reached. With a typical time of wall movement of approximately 10 s, the time necessary to reach a steady state at the begin and end of the movement is maximal 4 % of this typical time. Also the bottom can have an influence on the two-dimensional cavity flow. This influence is however negligible in the experiments (Anderson, 1996).

The begin and end of the tracer can be longer or shorter due to some mismatch of the placement of the blob. To investigate this fault, a simulation is performed for the periodic cavity flow with a co-rotating cylinder with a blob which has a radius that is 50 % larger than the matched blob from figure 3.16. Now the initial blob from the experiment is certainly located inside the blob used for the simulation. The results of this simulation are shown in figure 3.18. It is seen that compared to figure 3.16 the simulation of the experiment is indeed better during the first period, but that after three periods the agreement has not improved. Furthermore an error is introduced at the measurement of the wall displacement. The maximum difference between the computed number of rotations of a roller and the imposed number of rotations was 6 %. The signal measured from the tachometers is corrected for this difference. Finally, high stretching of the tracer in the cavity flow can result in extremely thin striations. As in the experiments the blob is a sphere, the thickness of the striations is not constant. These very thin striations are not observed in the experiments.

Secondly consider the structures of the mixtures that are obtained. Regard the fourth picture of figure 3.15. It is seen that the tracer is stretched and folded. However, for this structure, the deformed pattern is restricted to a relatively small area and striations lay close to each other.

From examination of the deformed pattern after two periods of the mixing experiment with a co-rotating cylinder (picture 4, figure 3.16), it is clear that the tracer material is now spread over a larger area with more space between successive striations.

After two periods, the deformed pattern is spread over the largest area when the cylinder is counter-rotating in the mixing experiment (picture 4, figure 3.17). Considering the simulation, it is seen that the material is extremely stretched and folded. Some repetitions of striations lay close to each other, then followed by more space between the successive striations.

Besides the kind of flow, the shape and spread of the deformation patterns are also dependent on parameters as initial location and dimensionless wall displacement. Therefore it is hard to compare the resulting deformation patterns with those of the regular periodic cavity flow which is discussed in chapter 2. Nevertheless it is clear that the presence of a central (rotating) cylinder has a great influence on the shape and spread of the tracer. Agreement in shape and spread with the deformation patterns shown in figure 2.7 is present for mixing with a co-rotating cylinder. When the cylinder is fixed (shown in figure 3.12, 3.13 and 3.15) more folds appear and the striations are more curved. These effects are even more pronounced in the deformation patterns obtained with a counter-rotating cylinder.
Figure 3.18: Simulations and experiments for a discontinuous operation of top and bottom wall with a co-rotating cylinder. The period increases with $\frac{1}{2}$ from left to right. $D \approx 4.8$, center of the initial blob $(0.91, 0.04)$ with a radius of 0.15. The initial blob is depicted in every picture.
Table 3.2: Estimation of the number of folds in the deformation pattern for mixing with a fixed cylinder, a co-rotating cylinder and a counter-rotating cylinder

<table>
<thead>
<tr>
<th>Period</th>
<th>fixed cylinder</th>
<th>co-rotating cylinder</th>
<th>counter-rotating cylinder</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>1</td>
<td>0</td>
<td>2</td>
</tr>
<tr>
<td>1</td>
<td>2</td>
<td>1</td>
<td>3</td>
</tr>
<tr>
<td>1 1/2</td>
<td>5</td>
<td>2</td>
<td>7</td>
</tr>
<tr>
<td>2</td>
<td>8</td>
<td>4</td>
<td>&gt; 10</td>
</tr>
</tbody>
</table>

It is already mentioned in the introduction, that stretching and folding is relevant to efficient mixing. In figure 3.19 the relative inter-facial length and the area of the blob are shown against the number of periods. Also the inter-facial length and area of the blob for mixing in the regular cavity flow are depicted for the case of figure 2.7. Notice that for this last situation, the dimensionless wall displacement $D = 6.24$, while for mixing in the extended cavity flow, in all cases $D \approx 5$. The inter-facial length increases exponentially with time. After two periods, the inter-facial length has grown the most for the mixing operation with a counter-rotating cylinder, followed by the fixed cylinder and the co-rotating cylinder. The inter-facial length of mixing in the regular cavity flow nearly corresponds to the case of mixing with a co-rotating cylinder. If we try to count the number of folds in figure 3.15 to 3.17, (which is somewhat arbitrary as it is difficult to distinguish a ‘fold’ from a ‘curved line’), we see that the number of folds seems to increase exponentially as well (see table 3.2). Thus all considered periodic flows satisfy the conditions for efficient mixing. Moreover it seems that, based on the inter-facial length, the mixing capabilities are increased in the extended cavity flow.

From figure 3.19 also follows that the conservation of area holds for the simulations with a fixed, a co-rotating and no cylinder. The deviations of the area are less than 1.5 %. When the cylinder is counter-rotating, this deviation is maximal 5 %.

Figure 3.19: ––: fixed cylinder, •--: co-rotating cylinder, ⋯: counter-rotating cylinder, $D \approx 5$; – regular cavity flow, $D = 6.24$
Chapter 4

Generalized Newtonian fluid

4.1 Non–Newtonian cavity flow

Most research on the periodic cavity flow is performed using viscous fluids. Leong and Ottino (1990) performed experiments with viscoelastic fluids in the periodic cavity flow. They used ‘Boger fluids’: fluids exhibiting viscoelastic behaviour and having a constant shear viscosity.

The importance of elasticity in a viscoelastic flow can be represented by the Deborah-number $De$, defined as

$$De = \frac{\lambda}{T_{flow}}$$

in which $\lambda$ represents the relaxation time of the fluid, and $T_{flow}$ a characteristic time scale for the flow. Both the inverse of the shear rate $H/V$ or the time period $T$ can be taken as the characteristic time of the flow. If the experiments are carried out at constant velocity $V$ and dimensionless wall displacement $D = 2VT/W$, these characteristic times are proportional to each other.

Leong and Ottino found that the chaotic behaviour decreased with increasing Deborah-number (with increasing elasticity). The mixing process is slower and the islands become larger. They also found that the streamlines were hardly distinguishable from the streamlines for a Newtonian cavity flow. Therefore they stated that differences in dye patterns are due to variations in the velocity along the streamlines. Viscoelasticity influences the magnitude of the velocity, but barely influences the direction of the velocity compared with the Newtonian flow.

Niederkorn and Ottino (1993) also found that in general the regularity increases with increasing $De$. Like Leong and Ottino (1990) they also used Boger fluids, but a different chaotic flow: a periodic flow between eccentric cylinders.

Ling and Zhang (1995) performed simulations of the periodic cavity flow, to study the behaviour of generalized Newtonian fluids. The viscosity $\eta$ is a function of the shear rate $\dot{\gamma}$, but viscoelasticity is not taken into account. They studied the effect of shear thinning, using the Powell model

$$\eta = \frac{\eta_0}{1 + (\dot{\gamma}/\dot{\gamma}_0)^{1-n}}$$

(4.2)
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The main conclusion of their work is that shear thinning fluid behaviour degrades the mixing in the corotating discontinuous cavity flow, as shear thinning reduces the link between the high and low shear rate regions.

4.2 Extended cavity flow

Figure 4.1: Mesh used for FEM computations

Figure 4.2: Absolute value of the difference between the velocity fields between a Newtonian and a generalized Newtonian flow. Only the top wall is moving.

Figure 4.3: Streamline pattern of the generalized Newtonian fluid for a moving top wall and a fixed cylinder.

In this section the influence of generalized Newtonian fluid behaviour in the extended cavity flow is discussed. Problems that arose using viscoelastic fluids are listed in appendix C. The dimensions of the cavity and cylinder are the same as in chapter 3 (figure 3.1). Simulations with shear thinning fluids are performed using the finite element method (FEM). The velocity field is computed using the Carreau-Yasuda model for the viscosity

\[ \eta = \eta_{cy} \frac{1}{(1 + (\lambda \sqrt{t})^\alpha)^{1-\alpha}} \]  (4.3)
In which $II_D$ denotes the second invariant of the rate of deformation tensor $II_D = \frac{1}{2}((\text{tr } D)^2 - \text{tr } (D^2))$. The material parameters ($\eta_c$, $\lambda$, $n$, $a$) used for the simulations can be found in appendix A. The stationary Stokes solution is solved using quadratic quadrilateral elements (figure 4.1) and an iterative process in which the equations are solved using a direct Gaussian elimination method. The velocity of the top wall $V_{\text{top}} = \frac{1}{2} \cdot 10^3 \cdot W$, to be sure that shear thinning behaviour is present in the simulations (in practice it is not realistic to solve the stationary Stokes equation with that high wall velocity). The absolute value of the difference between the velocity fields normalized by $V_{\text{top}}$ for a Newtonian flow, computed by the analytical superposition method, and the generalized Newtonian flow, is plotted in figure 4.2. In figure 4.3 the streamline pattern of the generalized Newtonian flow is plotted. Comparison with the streamlines for the Newtonian flow (figure 3.2) shows that despite of the evident difference in absolute velocities, the streamlines are barely identical to the Newtonian flow.

Figure 4.4: Deformation patterns of a line on the positive y-axis using the velocity field of the Lamé superposition method for a Newtonian flow and the finite element method for a generalized Newtonian flow. $D = 5$, the cylinder is fixed. The period increases with $\frac{1}{2}$ from left to right.
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Deformation patterns of lines on the $y$-axis are compared to the deformation patterns of viscous fluids for three periods (figures 4.4 and 4.5). The stretching of the initial line is regarded in figures 4.6 and 4.7. From these pictures it becomes clear that the deformation of the line, and thus the mixing properties of the flow, decreases with generalized Newtonian fluid behaviour. This conclusion agrees with the results of Ling and Zhang (1995) for the corotating discontinuous cavity flow.

Figure 4.5: Deformation patterns of a line on the negative $y$-axis using the velocity field of the Lamé superposition method for a Newtonian flow and the finite element method for a generalized Newtonian flow. $D = 5$, the cylinder is fixed. The period increases with $\frac{1}{2}$ from left to right.
Figure 4.6: Relative stretch of the deformed lines in figure 4.4

Figure 4.7: Relative stretch of the deformed lines in figure 4.5
Chapter 5

Quality of a mixture

5.1 Correlation coefficient and power spectrum

The correlation function $R(\vec{r})$ of a mixture is determined by examining how the compositions $c(\vec{x})$ at two points, separated by a vector $\vec{r} = r \vec{e}_r$, differ from the average composition of the mixture (Danckwerts, 1952)

$$R(\vec{r}) = \langle [c(\vec{x} + \vec{r}) - \bar{c}][c(\vec{x}) - \bar{c}] \rangle \text{ with } \bar{c} = \langle c(\vec{x}) \rangle$$ (5.1)

The angle brackets denote an average over the entire mixture. When $\vec{r} = \vec{0}$ the correlation function describes how the composition at each point differs from the average composition. This is termed the variance in composition.

$$R(\vec{0}) = \langle [c(i) - \bar{c}]^2 \rangle = \sigma_c^2$$ (5.2)

The variance is used to normalize the correlation function and obtain the correlation coefficient.

$$\rho(\vec{r}) = \frac{R(\vec{r})}{\sigma_c^2}$$ (5.3)

The graph of $\rho(\vec{r})$ is called a correlogram. An alternative description that contains exactly the same information is a spectral description: the power spectrum $P(\vec{n})$, with $\vec{n} = n \vec{e}_r$. It is defined as the Fourier transform of the correlation function:

$$P(\vec{n}) = \int_{-\infty}^{\infty} R(\vec{r}) e^{-i(2\pi nr)} d\vec{r}$$ (5.4)

Here $i$ denotes the imaginary number. The variable $n$ is termed the wave number and has unit of length$^{-1}$. Similar, $\vec{n}$ is termed the wave number vector. Roughly speaking, the spectrum turns the correlogram inside out: correlations at short distances influence the spectrum at large wave numbers, and correlations at long distances influence the spectrum at small wave numbers.

In general, the power spectrum is the fastest way to compute the correlation function, if the fast Fourier transform (FFT) algorithm can be used. $P(\vec{n})$ is the power spectrum of the concentration field $c(\vec{x})$ as well. To compute it directly from concentrations one first defines $a(\vec{x})$, the local deviation of the concentration from the average:

$$a(\vec{x}) = c(\vec{x}) - \bar{c}$$ (5.5)
Chapter 5

and then takes the Fourier transform of $a(x)$

$$Q(\vec{n}) = \int_{-\infty}^{\infty} a(\vec{x}) e^{-i(2\pi n x)} d\vec{x}$$  \hspace{1cm} (5.6)

$Q(\vec{n})$ is a complex function, having real and imaginary parts at each wave number. $P(\vec{n})$ is the square magnitude of these numbers:

$$P(\vec{n}) = Q(\vec{n})Q^*(\vec{n}) = |Q(\vec{n})|^2$$  \hspace{1cm} (5.7)

where the asterisk denotes the complex conjugate. Now, one can calculate $R(\vec{r})$ by taking the inverse transform:

$$R(\vec{r}) = \int_{-\infty}^{\infty} P(\vec{n}) e^{i(2\pi n r)} d\vec{n}$$  \hspace{1cm} (5.8)

The (discrete) computation of the correlation coefficient is discussed in more detail in appendix B. The correlation function is considered to give a complete description of the texture. Several other measures can be derived from the correlation function (Tucker, 1991). Two examples of derived measures are described in the next section.

5.2 Intensity and scale of segregation

The intensity and scale of segregation are measurement techniques that can be computed from the correlation function (Danckwerts 1952). The intensity of segregation refers to the difference between the composition of the mixture at a certain moment and the (desired) uniform distribution. It is determined by examining how the composition at each point differs from the average composition of the mixture. The variance in composition is a measure of how much the concentration $c(x)$ varies from the average (equation (5.2)). If the concentration $c(x)$ is either 1 or 0 everywhere (no diffusion), the variance $\sigma_c^2 = c(1-c)$. This is used to normalize the variance and obtain the intensity of segregation

$$I = \frac{\sigma_c^2}{c(1-c)} = \frac{R(0)}{c(1-c)}$$  \hspace{1cm} (5.9)

So without molecular diffusion or chemical reaction, the intensity $I$ will always be 1. A value of $I$ equal to zero means no intensity, and a complete uniform distribution is obtained. The concentration at each point is equal to the average concentration $c(x) = \bar{c}$.

As no diffusion is present in our experiments, the intensity of segregation is always 1. Now consider the concentration $C$ at a small unit of area $\delta A$ instead of the concentration $c$ at a point $x$. This concentration $C$ is referred to as the coarse grain density. Using the coarse grain density, the intensity of segregation will decrease under mechanical mixing. The concentration at the scale of these units of area $C \leq 1$ and cannot be 1 over the entire cavity. Similar, the coarse grain intensity of segregation decreases from $I = 1$ when all the units of the cavity area $\delta A$ have a concentration $C = 1$ or $C = 0$, to $I = 0$ when all units of the cavity area have the same concentration $C = \bar{C}$.  

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Quality of a mixture

The scale of segregation is a measure, that has its origin in the measurement of 'clumpy mixtures': mixtures that consist of clumps of two components that are (statistically) similar in shape, but have no regular arrangement or regular order. The scale of segregation is a measure of the size of the clumps. When \( \tau \) exceeds a certain value, the relationship between the two concentrations will become a random one (provided that there is no large scale regularity or regular periodicity in the mixture) and \( \rho(\tau) \) will fall to 0. The value of \(|\tau|\) for which \( \rho(\tau) \) falls to zero is termed \( \xi \). Moreover, the correlation coefficient will not become less than zero, as in a clumpy mixture no regularity exists. So \( 1 \geq \rho(\tau) \geq 0 \) for \( 0 \leq |\tau| \leq \xi \), and \( \rho(\tau) = 0 \) for \(|\tau| > \xi\).

In the notation used here, \( \tau \) is a vector, but reported correlograms usually treat \( \tau \) as a scalar quantity. This is strictly correct only if the mixture statistics are isotropic. In practice however, it is common to ignore this anisotropy (Tucker, 1991). If this approach is taken then \( \rho(\tau) \), where \( \tau = |\tau| \), should be the average of \( \rho(\tau) \) over all possible directions of \( \tau \).

There are two ways in which the correlogram is used to define a scale of segregation. First the linear scale of segregation \( S_L \), which is defined as the area under the correlogram

\[
S_L = \int_0^\infty \rho(\tau)d\tau = \int_0^\xi \rho(\tau)d\tau
\]

and second the volume scale of segregation \( S_V \), which is defined as \( 2\pi \) times the area under the curve \( r^2 \rho(r) \) versus \( r \)

\[
S_V = 2\pi \int_0^\infty r^2 \rho(r)dr = 2\pi \int_0^\xi r^2 \rho(r)dr
\]

\( S_L \) is a measure of the size of the clumps, and \( S_V \) represents a measure of the volume of the clumps.

The two measures, intensity and scale of segregation, are complementary, which is shown in figure 5.1. Intensity of segregation is affected by molecular diffusion, which can take place without bulk deformation. And the scale of segregation can decrease without changing the intensity, for example in mixing without molecular diffusion.

The limitation of scale of segregation is that it is not defined for mixtures with enough order to have negative values in the correlogram or in which long distance correlation occurs.

5.3 Coarse grain square density and entropy

Consider again a mixture of two materials of which one can be imagined to be coloured to distinguish it from the other one. We restrict ourselves to two dimensional cases. The coarse grain density \( C \) of the coloured material has been defined as the ratio of the area of coloured material \( \delta A_c \) in an unit \( \delta A \) and the total area of the unit \( \delta A \)

\[
C = \frac{\delta A_c}{\delta A}
\]

Integration of this density over the cavity space \( A \) will give the total area of coloured matter, which for the cavity flow should be constant.

\[
\sum_A C \delta A = \sum_A \delta A_c = A_c
\]
However, if we consider the square density $C^2$ we obtain the relation

$$F = \sum_A (C)^2 \delta A = \sum_A C \delta A_c < A_c$$

(5.14)

since $C \leq 1$ and can not be 1 in the entire cavity. The square density $F$ decreases with an increasing number of units of the cavity space that contain dyed material, and the density per occupied unit of area decreases. The minimal value of square density will be reached when all units of area contain the same density, and a 'uniform' distribution (at least on the dimension of the unit of area) is reached.

It is possible to use the analogy of entropy ($E = -C \log C$) instead of the square magnitude of the density for the statistical measure of the quality of mixing

$$E = -\sum_A C \log C \delta A$$

(5.15)

When the quality of the mixture improves, the entropy of the cavity will increase. The maximum value for the entropy that can be reached, corresponds to the best possible mixed state (uniform distribution).

$$E_{\text{max}} = -\sum_A \bar{C} \log \bar{C} \delta A = -\bar{C} \log \bar{C} A$$

(5.16)

where $\bar{C}$ denotes the average coarse grain density of $C$ defined by $A_c/A$. 

Figure 5.1: Examples of scale and intensity of segregation
5.4 Quality of the obtained mixtures

5.4.1 Image processing

Video recordings from the experiments are processed using the TIM image processing system (Difa Measuring Systems, 1993). These images are represented by a two dimensional array of integers, between 0 and 255. The value 0 denotes that the corresponding pixel is totally black (no intensity), the value 255 corresponds to a white pixel (maximum intensity). Values between 0 and 255 denote the intermediate gray levels. Before the quality of the mixture can be computed, it is necessary to edit the image to get information whether a pixel is occupied by dye, partly occupied by dye or not occupied by dye. Several protocols are used to digitize and process the video images. These protocols are:

1. **Threshold an image**: In this method, a threshold is applied to the image. This results in a two dimensional array, containing the values 1 (the pixel is occupied by dye) when the intensity \( h \) is higher than the threshold, or 0 (the pixel is not occupied by dye) when the intensity is less than the threshold. It is supposed that the background gives a fairly uniform signal. The threshold is chosen at a intensity \( h_{\text{thres}} \) slightly above this reference signal.

2. **Threshold with intensity**: This protocol is based on the definition of the coarse grain density. Difference with protocol 1 is that after the threshold is applied, a pixel with intensity \( h \) above the threshold, gets its original value divided by the overall maximal pixel value. So the new value \( h_2 \) is \( h/h_{\text{max}} \) for \( h \geq h_{\text{thres}} \) and \( h_2 = 0 \) for \( h < h_{\text{thres}} \). The underlying reason for this, is that pixels which are partly occupied by dye (because the width of a striation is less than that of the pixel or because the boundary of the tracer does not follow the boundary of the pixels) will get a value proportional to the area of the pixel that is occupied by the tracer. Strictly this is only correct when the intensity of the pixel is proportional to the amount of light which is emitted, and if the amount of light which is emitted is proportional to the part of the pixel which is occupied by dye.

However, besides of the amount of dye, the intensity in the image is also dependent on other factors. When a pixel is partly occupied by a relatively wide striation, the intensity will be higher due to the light emitted in the surrounding area, while when the pixel is occupied by a small striation in a dark background, the amount of light and thus the intensity in that area will be considerably less. A second factor is that the tracer intensity is also dependent on the height of the tracer (in the direction of the cylinder axis) and the amount of UV-light. As the blobs resemble more to spheres than to cylinders, it is clear that the height is not constant over the entire blob. The obtained concentrations are therefore not accurate, as the intensity of the pixels is not only dependent on the area of the pixel which is occupied by dye, but also by the height of the tracer, the amount of UV-light and the presence of tracer in the neighbourhood.

3. **Filter an image**: This protocol is developed to distinguish the areas which light up due to a large amount of dye present in the surrounding area, from the striations. From two images at period 0 (\( A_0 \)) and period \( \frac{1}{2} (A_{\frac{1}{2}}) \) a background image (B) is created by taking the minimum intensity of these two pictures for each pixel (\( B = \min[A_0,A_{\frac{1}{2}}] \)). This background image (B) is subtracted from the original image (A), resulting in image (C
This image is then filtered \( (D = \text{filter}[C]) \) (the intensity at a pixel becomes the average of a number of surrounding pixels) and again subtracted from the original image without background \( (E = C - D) \). The size of the filter has to be chosen small enough (such that the new intensity is a local value), but larger than the local dimensions of the striations (such that the centre of thick striations will not disappear when the filtered image is subtracted). After the subtraction, a threshold is applied to image \( (E) \) resulting in image \( (F) \) that contains only pixels with a value of 1 (occupied by dye) or 0 (pixel belongs to the background).

The periodic cavity flow is supposed to be area conserving. If the concentration of dye is proportional to the measured intensity the number of pixels should remain (approximately) constant. In figure 5.2, 5.3 and 5.4 the ratio of the area of the tracer to the total cavity area (protocol 1 and 3) and the ratio of the total tracer intensity to the maximum possible intensity \( (P2, \text{for protocol 2}) \) is shown, for the three mixing cases regarded in section 3.3. \( (P2 = \frac{1}{\# \text{ pixels}} \sum_{\# \text{ pixels}} h2(j)) \). For protocol 1 and 2, the upper line is the result after a threshold value of 50, and the lower line after a threshold value of 75. We see that the number of pixels, and thus the average concentration of the tracer, increases with the period for all protocols. From all the figures it is obvious that the increase is the least following the second protocol with a threshold of 75. Moreover it is seen that the increase is much less for the case of mixing with a co-rotating cylinder than in the other two cases. When a threshold value of 75 is used in the second protocol for this mixing case, the concentration is yet 1.9 times the reference concentration at period 2. When the first protocol is used with a threshold value of 50, the concentration at period 3 is 11.3 times the reference concentration. In case of mixing with a fixed cylinder, the worst concentration is 21.2 times the reference concentration and for the case of a counter-rotating cylinder this is even 21.7 times.

However, if high threshold values are used, the resulting image quality is reduced. In figure 5.5 the resulting deformation patterns are shown using a threshold value of 50 and 75 (protocol 1) in case of mixing with a co-rotating cylinder after three periods. Although it is clear that in the first picture some striations are missing, the second picture is a very poor representation of the deformation pattern.

Since pixels in the neighbourhood of dyed pixels will also light up, the number of pixels recognized as 'dyed' can increase. The inter-facial length increases exponentially (see figure 3.19). If the number of pixels increases proportionally to the length of the boundary of the tracer (the inter-facial length), the concentration should increase exponentially as well. From figure 5.2 to 5.4 however it is seen that the increase is more a linear one than an exponential one.

5.4.2 Results

The measures described in paragraph 5.1 to 5.3 are used for the computation of the quality of the mixture, except for the scale of segregation \( (S_L \text{ and } S_V) \). In the mixtures, there is some long term regularity and negative values in the correlation coefficient occur. Therefore it has no physical meaning to compute the scale of segregation.

After each full period the correlation coefficients of the mixtures shown in figure 3.16 are computed (see figure 5.6). At \( \bar{r} = 0 \) a peak with value 1 occurs, since the correlation coefficients are normalized by the variance \( \sigma_c^2 = R(0) \). This peak is not shown in this figure,
Quality of a mixture

Figure 5.2: Average concentration for the case of mixing with a fixed cylinder

Figure 5.3: Average concentration for the case of mixing with a co-rotating cylinder

Figure 5.4: Average concentration for the case of mixing with a counter-rotating cylinder

Figure 5.5: Deformation patterns for the case of mixing with a co-rotating cylinder at period 3 using protocol 1 and a threshold of 50 and 75 respectively
Figure 5.6: Three dimensional and contour plot of the correlation coefficient for the obtained mixtures resulting from a discontinuous operation of the top and bottom wall and a co-rotating cylinder (figure 3.16) after one period (upper), two periods (middle) and three periods (lower picture)
Quality of a mixture
to display the development of the correlation coefficient more clearly. In these figures it is seen
that regularity at long distances is present. The contour plots show that correlation occurs
at the vectors between the striations of the tracer in figure 3.16. The regularity becomes
more complicated after three periods, which also follows from the contour plot. From the
pictures at period 2 and 3 it is seen that correlation at short distance exists. Also anisotropic
structures in the mixtures are shown in the plots. These type of pictures become more usefull
for more developed mixtures where the structure is not recognized from the deformation
patterns. Here they are ment to illustrate their possibilities.

The protocol and the parameter $\delta A$ are varied in the computations of the intensity of
segregation $I$, the square density $F$ and the entropy $E$.

First the intensity of segregation $I$ is obtained from the different protocols (figure 5.7).
This figure shows that the correspondence is bad for the case of mixing with a fixed cylinder,
fairly good for protocol 2 and 3 when the cylinder is co-rotating, and also bad in the counter-
rotating case, except for protocol 2.

Figure 5.7: The intensity of segregation $I$ for the case of a fixed cylinder (upper left), a co-
rotating cylinder (upper right) and a counter-rotating cylinder (lower picture), as a function
of the used protocol. For protocol 1 and 2, the upper lines result from a threshold value of
50 and the lower lines from a threshold value of 75
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Figure 5.8 shows the square density $F$. The best agreement between the experimentally obtained results and those from the simulations, is found for protocol 2 with a threshold value of 75. But also the other experimentally obtained computations follow the trend of the computations of the simulations.

![Graphs showing square density $F$ for different protocols](image)

**Figure 5.8:** The square density $F$ for the case of mixing with a fixed cylinder (upper left), a co-rotating cylinder (upper right) and a counter-rotating cylinder (lower picture), as a function of the used protocol. For protocol 1 and 2, the lower lines result from a threshold value of 50 and the upper lines from a threshold value of 75.

The entropy is shown in figure 5.9. The best agreement between the experimentally obtained results and those from the simulations, is found for the case of mixing with a co-rotating cylinder. In this situation it also holds that in general protocol 2 and 3 show the best correspondence.

Secondly the influence of the size of the area $\delta A$ is regarded. Squares with sizes ranging from $\frac{H}{8}$ (small, ⋯⋯⋯⋯) to $\frac{H}{4}$ (medium, ⋯⋯) and $\frac{H}{2}$ (large, ⋯⋯) are evaluated. Only the case of mixing with a co-rotating cylinder will be considered, as for this case the agreement between the resulting measures from experiments and simulations is best.

In figure 5.10 the intensity of segregation $I$ is plotted as a function of the size of $\delta A$. The
Figure 5.9: The entropy $E$ for the case of mixing with a fixed cylinder (upper left), a co-rotating cylinder (upper right) and a counter-rotating cylinder (lower picture), as a function of the used protocol. For protocol 1 the upper lines result from a threshold value of 75 and the lower lines from a threshold value of 50, while for protocol 2 the upper lines result from a threshold value of 50 and the lower result from a threshold value of 75.
Figure 5.10: The intensity of segregation $I$ for protocol 1 (upper left) for protocol 2 (upper right) and protocol 3 (lower picture), as a function of the area $\delta A$ for the case of mixing with a co-rotating cylinder.
Quality of a mixture

Intensity of segregation is shifted down with increasing $\delta A$. There is little agreement between the experimentally obtained and simulated results for all protocols and $\delta A$. Protocol 1 is clearly not useful.

![Graphs showing square density $F$ for protocols 1, 2, and 3 as a function of the area $\delta A$.](image)

Figure 5.11: The square density $F$ for protocol 1 (upper left) for protocol 2 (upper right) and protocol 3 (lower picture), as a function of the area $\delta A$ for the case of mixing with a co-rotating cylinder

The dependence of the square density $F$ on the size of $\delta A$ is shown in figure 5.11. In this case the curves representing the square density are also shifted down with increasing $\delta A$. The square densities obtained from the experiments resemble those obtained from the simulations quite well, and the agreement even seems to improve with increasing $\delta A$. Protocols 2 and 3 show the best resemblance.

In figure 5.12 the entropies are plotted dependent on the size of $\delta A$. The entropy is shifted up with increasing $\delta A$. The agreement between the experimentally obtained results and those from the simulations seems not dependent on the size of $\delta A$.

Overall it can be concluded that there is little resemblance between experimentally obtained values and those from the simulations. Nevertheless trends are clearly followed for all measures, and the experimentally obtained values depend on $\delta A$ in the same way as those
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Figure 5.12: The entropy $E$ for protocol 1 (upper left) for protocol 2 (upper right) and protocol 3 (lower picture), as a function of the area $\delta A$ for the case of mixing with a co-rotating cylinder.
Quality of a mixture

obtained from simulations. The experimentally obtained values give flatter results, when the intensity of segregation is regarded: higher where the value is low and lower where the value should be high. For the square density the values from the experiments are less than those from the simulations. Both for the intensity of segregation and the square density, the most variance occurs during the first period, and is flattened in the following periods. Regarding entropy, the experimentally obtained values are also lower than those from the simulations.
Chapter 6

Conclusions and recommendations

6.1 Conclusions

- For the periodic Newtonian extended cavity flow, the simulations of deformation patterns agree with the experimentally obtained results.

- The presence of a central cylinder can have a great influence on the flow and the resulting deformation patterns. When the cylinder is counter-rotating this influence increases. In case that the cylinder is co-rotating with a sufficiently high velocity, the flow and resulting deformation patterns are more like the regular periodic cavity flow.

- In the extended periodic cavity flow chaotic mixing occurs, since horseshoe maps are formed. The obtained mixtures show an exponential increase of both the inter-facial length and the number of folds.

- Simulations based on a numerically computed velocity field can be performed. For the stationary Stokes solution with a Newtonian fluid, (nearly) identical results are obtained for the resulting deformation patterns and deformation patterns based upon a velocity field computed by the analytical Lamé superposition method. The deviations lie within experimental accuracy.

- Mixing decreases for generalized Newtonian fluid behaviour, although the streamline patterns hardly show any differences. The differences in mixing behaviour are due to variations of speed along the streamlines.

- While processing the images, problems occur concerning the detection of thin striations and area conservation of the tracer. The images resulting from the processing of the experimental results using different protocols differ from those obtained from the simulations for all used protocols.

- Computation of the correlation coefficient can be very useful in the analysis of mixtures. Not only is the correlation coefficient a good measure by itself, it is also the basis for several other mixing measures.

- The values for the intensity of segregation, square density and entropy resulting from the experiments do not correspond well with those computed from the simulations. However trends are predicted reasonably well. The correspondence is the worst for the
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case that only a threshold is used while processing the image. The correspondence is much better when during the processing of the image, a filtered image is subtracted. The best resemblance is found when the coarse grain density is used in the processed image.

6.2 Recommendations

- The velocity and displacement of the moving walls have to be regulated and controlled instead of being measured for optimal control of the experiments.

- One should excite the tracer material with light of one wavelength instead of UV-light containing a wide spectrum as is used now. When there is only one wave length present in the background, it becomes easier to filter the tracer from the background.

- Instead of recording the experiments on video one should take photographs of the mixing experiments. Photographs give a higher resolution than pictures taken from video, which might be important for the detection of very thin striations and mixing measurement.

- Investigation of the influence of non-Newtonian fluid behaviour by comparison of experiments and simulations for fluids with shear thinning and viscoelastic fluid behaviour, should be conducted.

- The programs used for the integration of points using the numerical computed velocity field have to be changed, such that the determination of the velocity at a point takes less time. A lot of CPU time is spend in finding the element of the mesh in which the point is located.

- The protocol to process the image has to be studied further to increase the agreement between the experimentally obtained results and those obtained from the simulations. A combination of the coarse grain density with correction for the background might be a basis for further development. Better agreement could also be realized by finding a method that is able to translate the simulated results better to experimental results.
Bibliography


Bibliography


Appendix A
Materials

A.1 Glycerine

In all experiments in this study glycerine is used. The glycerine (Lamers & Pleuger, 2LZ250003) is considered a Newtonian fluid. In figure A.1 the shear stress and the viscosity are shown as a function of the shear rate. These data are obtained by a simple shear test at the Rheometrics-RFS-II viscometer with a plate-plate geometry (diameter 50 mm, temperature 22 °C). In table A.1 some material parameters are shown.

![Figure A.1: Glycerine: shear stress (left) and viscosity (right) as a function of the shear rate](image)

Table A.1: The viscosity and density of the glycerine

<table>
<thead>
<tr>
<th>( \eta \ [Pa \ s] )</th>
<th>1.06e+00</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \rho \ [10^3 \ kg \ m^{-3}] )</td>
<td>1.26e+00</td>
</tr>
</tbody>
</table>
Appendix A

A.2 S1' solution

The fluid S1 is a solution of 2.5%(w/w) polyisobutylene (PIB) in 47.5%(w/w) Decalin (decahydronaphtalene) and 50%(w/w) polybutene oil. The S1 regarded here, however, deviates from the S1 discussed by Baaijens (1994). The average molecular weight of the PIB, which is solved in Decalin and polybutene oil, is lower compared to the PIB used by Baaijens. To distinguish, the fluid considered here is called S1'. The PIB used for this solution was Vistanex L-120 (Exxon Chem.), the polybutene oil was Hyvis 10 (BP Chem).

Figure A.2: S1': shear stress as a function of the shear rate (left) and comparison of the steady shear viscosity (o) and the dynamic viscosity (x) (right)

A simple shear test and a dynamic frequency test are performed at the Rheometrics-RFS-II viscometer with a plate-plate geometry (diameter 50 mm, temperature 22 °C). In figure A.2 the shear stress and the viscosity are shown as a function of the shear rate and the dynamic frequency. From the right picture, it can be concluded that S1' does not satisfy the Cox-Merz rule. The real S1 solution does not satisfy the Cox-Merz rule either (Baaijens, 1994).

Table A.2: Maxwell parameter values for 1-mode and 4-mode (m: number of modes) and parameters values of the Carreau-Yasuda equation

<table>
<thead>
<tr>
<th>Maxwell parameters</th>
<th>Carreau-Yasuda parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>m</td>
<td>η [Pa s]</td>
</tr>
<tr>
<td>1</td>
<td>3.42e+00</td>
</tr>
<tr>
<td>4</td>
<td>1.06e+00</td>
</tr>
<tr>
<td>1.50e+00</td>
<td>8.06e-01</td>
</tr>
</tbody>
</table>

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In table A.2 the 1-mode and 4-mode Maxwell-parameters as well as the parameter values of the Carreau-Yasuda model (equation 4.3) are given. In figure A.3 the dynamic viscosity with the fitted Maxwell models are presented. The Carreau-Yasuda fit and the steady shear viscosity are plotted in figure A.4.

**Figure A.3:** $S1^1$: fit of the linear viscoelastic Maxwell model on the complex viscosity for a single mode Maxwell model (left) and for a 4 mode Maxwell model (right)
Figure A.4: $\eta_s$: comparison of the steady shear viscosity with the Carreau-Yasuda fit
Appendix B

Computation of the correlation coefficient and power spectrum

B.1 Discrete analysis

B.1.1 Discrete Fourier transform

It can be proved (Newland, 1993) that if the function \( a(\vec{x}) \) with \( \vec{x} = x \vec{\varepsilon} \) is periodic with period \( H \) in the direction \( \vec{\varepsilon} \), it is possible to write \( a(\vec{x}) \) as

\[
a(\vec{x}) = c_0 + 2 \sum_{k=1}^{\infty} \left( c_k(\vec{\varepsilon}) \cos \frac{2\pi k x}{H} + d_k(\vec{\varepsilon}) \sin \frac{2\pi k x}{H} \right)
\]  

(B.1)

where

\[
c_k(\vec{\varepsilon}) = \frac{1}{H} \int_{0}^{H} a(\vec{x}) \cos \frac{2\pi k x}{H} d\vec{x}
\]

\[
d_k(\vec{\varepsilon}) = \frac{1}{H} \int_{0}^{H} a(\vec{x}) \sin \frac{2\pi k x}{H} d\vec{x}
\]

By defining \( Q_k(\vec{\varepsilon}) = c_k(\vec{\varepsilon}) - id_k(\vec{\varepsilon}) \) we can write the Fourier transform of \( a(\vec{x}) \)

\[
Q_k = Q_k(\vec{\varepsilon}) = \frac{1}{H} \int_{0}^{H} a(\vec{x}) e^{-i(2\pi k x/H)} d\vec{x}
\]  

(B.2)

Often measurements are digitized with some sampling interval \( \Delta \). Consider the case that the continuous function \( a(\vec{x}) \) is digitized with sampling interval \( \Delta_x = \Delta_x \vec{\varepsilon} = H/M \vec{\varepsilon} \). Then

\[
a_m = a_m(\vec{\varepsilon}) = a(m \Delta_x)
\]  

for \( m = 0, 1, 2, \ldots, (M - 1) \)  

(B.3)

Thus \( a(\vec{x}) \) is represented by the M samples of the discrete series \( a_m \). The discrete form of equation (B.2) can be written as

\[
Q_k = \frac{1}{H} \sum_{m=0}^{M-1} a_m e^{-i(2\pi k x/H)(m \Delta_x)} \Delta_x
\]  

(B.4)
Appendix B

Substitution of \( H = M \Delta x \) gives

\[
Q_k = \frac{1}{M} \sum_{m=0}^{M-1} a_m e^{-i\left(2\pi km/M\right)}
\]  

(B.5)

This is the definition of the discrete Fourier transform (DFT). The values \( a_m \) can be regained exactly by the inverse discrete Fourier transform (IDFT)

\[
a_m = \sum_{k=0}^{M-1} Q_k e^{i\left(2\pi km/M\right)}
\]  

(B.6)

Since \( a_m \) is considered to have been derived as a single cycle of a continuous periodic function \( a(x) \), the components \( Q_k \) are an approximation for the coefficients of a Fourier series expansion of \( a(x) \).

B.1.2 Characteristics of the DFT

Consider equation (B.5) and suppose \( k = M + l \), then

\[
Q_{M+l} = \frac{1}{M} \sum_{m=0}^{M-1} a_m e^{-i\left(2\pi km/(M+l)\right)}
\]

\[
= \frac{1}{M} \sum_{m=0}^{M-1} a_m e^{-i\left(2\pi lm/M\right)} e^{-i2\pi m}
\]  

(B.7)

which, since \( e^{-i2\pi m} = 1 \quad \forall \ m \in \mathbb{N} \), gives

\[
Q_{M+l} = Q_l
\]  

(B.8)

The coefficients of \( Q_k \) just repeat themselves for \( k > M - 1 \). Therefore, the range of the Fourier components \( Q_k \) is limited to \( k = 0 \) to \( M - 1 \), corresponding to harmonics of frequency \( \omega_k = 2\pi k/H \).

Furthermore it can be seen that if we consider \( k = -l \)

\[
Q_{-l} = \frac{1}{M} \sum_{m=0}^{M-1} a_m e^{i\left(2\pi lm/M\right)}
\]

\[
= Q_l^*
\]  

(B.9)

Periodicity and symmetry about the zero frequency position, make that the unique part of the graph of \( |Q_k| \) against \( \omega_k = \frac{2\pi k}{M\Delta x} \) occupies the frequency range \( 0 \leq \omega \leq \pi/\Delta x \) (see fig. B.1). Therefore the coefficients calculated by the DFT are only the correct coefficients for frequencies up to

\[
\omega_k = \frac{\pi}{\Delta x}
\]  

(B.10)

that is for \( k \) in the range \( k = 0, 1, 2, \ldots, M/2 \). If frequencies above \( \pi/\Delta x \) are present in the original signal, these introduce a distortion of the graph called aliasing. The high frequency components contribute to the \( a_m \) series and falsely distort the Fourier components calculated by DFT below \( \pi/\Delta x \) rad/s (see figure B.2). The maximum frequency that can be detected from data sampled at spacing \( \Delta x \) is

\[
\omega = \frac{\pi}{\Delta x} \quad \text{or} \quad f = \frac{1}{2\Delta x}
\]  

(B.11)
Computation of the correlation coefficient and power spectrum

Figure B.1: Periodicity of Fourier coefficients

Figure B.2: Aliasing: --: true spectrum; ---: computed spectrum; . . . : distortion

B.2 Computation of the correlation function and spectrum

B.2.1 Circular and linear correlation function

Consider the calculation of the correlation function of the series $a_m$. Clearly we can only estimate this for distances which are multiples of the sampling space $\Delta x$ and, if we write $R_m$ as an estimate for the correlation function $R(r)$ for $r = m\Delta x$, then we can define

$$R_m = \frac{1}{M} \sum_{s=0}^{M-1} a_s a_{s+m} \quad m = 0, 1, 2, \ldots, (M-1) \quad (B.12)$$

where, since $a(x)$ is assumed to be periodic,

$$a_{s+m} = a_{s+m-M} \quad \text{when} \quad s + m \geq M \quad (B.13)$$

Now consider for example the correlation function when $r = 4\Delta x$ and $M = 10$. If we write out the summation of equation (B.12) we obtain

$$R_4 = \frac{1}{10} \left\{ a_0 a_4 + a_1 a_5 + a_2 a_6 + a_3 a_7 + a_4 a_8 + a_5 a_9 \right\} + \frac{1}{10} \left\{ a_6 a_0 + a_7 a_1 + a_8 a_2 + a_9 a_3 \right\} \quad (B.14)$$

The first term in {} brackets is an estimate for $R(r = 4\Delta x)$, but the second term has nothing to do with $R(r = 4\Delta x)$ at all and is instead the estimation for $R(r = 6\Delta x)$. Both terms are biased estimates for $R(r)$, since the denominator would have to be 6 in the first, and 4 in the second case, to find correct mean values in each case. Furthermore it is now easy to see, that $R_6$ will be the same as $R_4$, or more general $R_{M-m} = R_m$. This correlation function is therefore also called the periodic or circular correlation. We can write equation (B.12) in two parts as

$$R_m = \frac{1}{M} \sum_{s=0}^{M-1-m} a_s a_{s+m} + \frac{1}{M} \sum_{s=M-m}^{M-1} a_s a_{s+m-M}$$

$$= \frac{1}{M} \sum_{s=0}^{M-1-m} a_s a_{s+m} + \frac{1}{M} \sum_{s=M-m}^{M-1} a_s a_{s-(M-m)} \quad (B.15)$$
These two summations can be related to estimates of the (continuous) linear correlation function \( R(\tau) \) defined by the sample average

\[
R(\tau) = \lim_{H \to \infty} \frac{1}{H} \int_0^H a(\tilde{x}) a(\tilde{x} + \tau) \, dx
\]

for ergodic processes. If the continuous record of \( a(\tilde{x}) \) is available for the record length \( H \) only, then an approximation for \( R(\tau) \) is

\[
\hat{R}(\tau) = \frac{1}{H - |\tau|} \int_0^{H-|\tau|} a(\tilde{x}) a(\tilde{x} + \tau) \, dx
\]

for \( 0 \leq |\tau| < H \), since the available integration length depends on the lag \( |\tau| \) and clearly it is not possible to make calculations for \( |\tau| \) greater than the record length.

Consider again the finite sequence \( a_m \), obtained by sampling the continuous record at regular sampling intervals \( \Delta x \). The discrete form of equation (B.17) is then

\[
\hat{R}_m = \frac{1}{M - m} \sum_{s=0}^{M-1-m} a_s a_{s+m} \quad \text{for } m = 0, 1, 2, \ldots, (M - 1)
\]

This is the estimate for the true or linear correlation function and is related to the periodic or circular correlation function defined by (B.15) by

\[
R_m = \frac{M - m}{M} \hat{R}_m + \frac{M - (M - m)}{M} \hat{R}_{M-m}
\]

**B.2.2 Addition of zeros**

The main reason to add zeros to a series \( a_m \), is that the computational more efficient fast Fourier transform (FFT) requires a length of the sequence of data points which is a power of 2. But the adding of zeros to a sequence has other advantages as well. Consider again the correlation function when \( m = 4, M = 10 \). \( L = 6 \) zeros have to be added to obtain a sequence of length \( 16 = 2^4 \). Write out the summation to obtain the correlation function

\[
R_4 = \frac{1}{16} \{a_0 a_4 + a_1 a_5 + a_2 a_6 + a_3 a_7 + a_4 a_8 + a_5 a_9\}
\]

\[
= \frac{1}{6} \cdot \frac{1}{6} \{a_0 a_4 + a_1 a_5 + a_2 a_6 + a_3 a_7 + a_4 a_8 + a_5 a_9\}
\]

\[
= \frac{M - m}{M + L} \hat{R}_m \quad \text{for } m \leq L
\]

since \( a_{10} \) to \( a_{15} \) have a value of zero. There will be no overlap from one period of the sequence to the next period for other values up to \( m = 6 \), but for \( m = 7, 8, 9 \) there will be overlap. Then

\[
R_m = \frac{M - m}{M + L} \hat{R}_m + \frac{M - (M - m)}{M + L} \hat{R}_{M+m-L} \quad \text{for } L < m \leq M - 1
\]

In order to separate the two overlapping parts of the linear correlation function completely, the number of additional zeros \( L \) must be sufficient \((L \geq M - 1)\). The circular correlation function is then related to the linear correlation function as follows

\[
R_m = \frac{1}{M + L} \sum_{s=0}^{M-1-m} a_s a_{s+m} = \frac{M - m}{M + L} \hat{R}_m
\]
**B.2.3 Spectrum**

We have already seen in chapter 5 that the power spectrum can be computed by taking the Fourier transform of the correlation function, or by taking the square magnitude of the Fourier transform of the concentration field. So we can write

\[ P_k = \frac{1}{M} \sum_{m=0}^{M-1} R_m e^{-i(2\pi km/M)} \]

\[ = \left| \frac{1}{M} \sum_{m=0}^{M-1} a_m e^{-i(2\pi km/M)} \right|^2 \]  \hspace{1cm} (B.23)

From the previous, it is clear that this is the spectrum corresponding to the circular correlation function. If the spectrum for the linear correlation function is wanted, one has to add \( L \geq M - 1 \) zeros to the record length. Then we obtain

\[ P_k = \frac{1}{M + L} \sum_{s=0}^{M-1} \tilde{R}_s e^{-i(2\pi km/(M+L))} \]

\[ = \left| \frac{1}{M + L} \sum_{m=0}^{M-1} a_m e^{-i(2\pi km/(M+L))} \right|^2 \]  \hspace{1cm} (B.24)

Often one wants to know the power spectrum dependency on the frequency. Often one wants to know the dependence of the power spectrum on the frequency \( \omega \) instead of the wave number \( k \). In that case equation (B.24) can be rewritten as

\[ P(\omega_k) = \sum_{k=0}^{M-1} P_k \delta(\omega - \frac{2\pi k}{H}) \]  \hspace{1cm} (B.25)

Notice that in case of a circular spectrum the frequency \( \omega_k = 2\pi k/(M\Delta_x) \) is higher than in case of the linear spectrum \( \omega_k = 2\pi k/((M+L)\Delta_x) \). Peaks in the spectrum occur at the same value of \( \omega \), but not for the same value of \( k \).

**B.2.4 Summary**

**Circular correlation**

\[ R_m = \frac{1}{M} \sum_{s=0}^{M-1} a_s a_{s+m} \]

\[ P_k = \frac{1}{M} \sum_{m=0}^{M-1} R_m e^{-i(2\pi km/M)} \]

\[ = \left| \frac{1}{M} \sum_{m=0}^{M-1} a_m e^{-i(2\pi km/M)} \right|^2 \]

\[ P_c(\omega_k) = \sum_{k=0}^{M-1} P_k \delta(\omega - \frac{2\pi k}{H}) \]
Appendix B

Linear correlation

\[
\hat{R}_m = \frac{1}{M - m} \sum_{s=0}^{M-1-m} a_s a_{s+m}
\]

\[
\hat{P}_k = \frac{1}{M + L} \sum_{s=0}^{M-1} \hat{R}_m e^{-i(2\pi km/(M+L))}
\]

\[
= \frac{1}{M + L} \sum_{m=0}^{M-1} a_m e^{-i(2\pi km/(M+L))}^2
\]

\[
P_l(\omega_k) = \sum_{k=0}^{M-1} \hat{P}_k \delta(\omega - \frac{2\pi k}{(M + L)\Delta_x})
\]

Notice that power spectrum computed with the linear correlation function in comparison with the circular has lower value of \(\omega_k\) at the same value of \(k\), because the sequence has been extended by \(L\) zeros.

In practice, we deal with two dimensional arrays, containing information about the concentration field in the cavity. In order to obtain the linear correlation function and spectrum, we first zeros to the two dimensional array to an array with a size of 2 times the next power of 2 (for example: if \(M = 10\), the next power of two is \(16 = 2^4\), we must add 22 zeros to obtain a series of \(32 = 2^5\)). A two dimensional Fourier transform on such an array is performed by taking the FFT of each row of the two dimensional array. The results are placed in a two dimensional array such that the FFT of row \(m\) is placed in row \(m\). Now a FFT on each column of that array is performed, and the Fourier transform of the two dimensional array is obtained. The power spectrum is computed by multiplying the transform at each position of the array by its complex conjugate. In the same way, the correlation function is now computed by taking the two dimensional inverse transform of the power spectrum.

B.3 Examples

Some examples of the correlation coefficient and spectrum of some imaginary surfaces. First the signal is shown, then the circular correlation coefficient and the circular power spectrum, and finally the linear correlation function and linear power spectrum. In the following examples \(\Delta_x = 1\). examples.
The signal can be considered as one period with length $H = 32\Delta x$. The spectrum will therefore show a peak at $\omega = \frac{3\pi}{32}$ for the circular and linear spectrum. From the figure it is also clear, that the circular correlation function is mirrored about $k = 16$ and the spectra are mirrored about $\omega = \pi$. 
Figure B.4: The signal consists of four repetitions of a pulse signal. Therefore a peak in the spectrum occurs at $\omega = \frac{2\pi}{8}$ for the circular and linear spectrum.
Figure B.5: The signal offered is a periodic signal with 1 period in one direction and 4 periods in the other direction. This results in peaks at $\omega = (\frac{3\pi}{32}, \frac{2\pi}{8}$ for the circular and linear spectrum. Again we see the circular correlation function is mirrored about the lines $k = (16, 16)$, and the spectra are mirrored about $\omega = \pi$. 
Figure B.6: The periodic signal offered has no evident orientation in one direction. Therefore the correlation function doesn’t show high peaks, but gives a ribbled surface. In the linear correlation the long distance relation however evidently show up again. In the spectrum there isn’t one evident peak.
Appendix C

Viscoelastic fluids

C.1 Experiments

Several problems occurred when chaotic mixing experiments with the S1’ solution (appendix A) were performed. Due to absorption of Decalin from the solution, the rubber belts swell up. Therefore (Viton) belts are necessary, that are resistant to Decalin. A second problem when using the S1’ solution is the visualization of the deformed blob. Since S1’ is an apolar fluid, polar fluorescent powders do not dissolve. A usable apolar fluorescent solvent has not yet been found.

Moreover Decalin in the S1’ solution evaporates relatively fast. Therefore the composition and the mechanical properties of the S1’ vary with time. For experiments with viscoelastic fluids, it is recommended to use a different fluid, e.g. a solution of corn syrup, polyacrylamide (PAA), water and a trace of sodium azide (Na N₃), which constitutes a viscoelastic polar fluid.

C.2 Computations

A finite element program of Selen (1995) is used to compute the velocity of a viscoelastic fluid in the extended cavity flow. The constitutive model used is a linearized Phan-Thien/Tanner model (PTT-b). The solution only converges for very low wall velocities. The computed result of the velocity field then corresponds to a Newtonian flow. For relatively high wall velocities it was not possible to obtain a solution of the velocity field. The main problem for convergence seems to be the combination of the singular corner points and viscoelasticity. This problem can probably be solved by using stress free elements near these corners (Anderson, 1996).