Mixing of non-Newtonian fluids in time-periodic cavity flows

Patrick D. Anderson*, Oleksiy S. Galaktionov, Gerrit W.M. Peters, Frans N. van de Vosse, Han E.H. Meijer

Materials Technology, Dutch Polymer Institute, Eindhoven University of Technology,
P.O. Box 513, WH 0.117, 5600 MB Eindhoven, The Netherlands

Received 25 October 1999; received in revised form 7 March 2000

Abstract

Fluid mixing in two- and three-dimensional time-periodic cavity flows as a function of rheological fluid parameters is studied. Computational methods are applied to obtain accurate descriptions of the velocity fields, which form the basis of the mixing analysis. In addition to some classical techniques, like Poincaré maps and the analysis of periodic points, a recently developed mapping method is used to determine mixing efficiency over a wide range of different flow parameters. Within this framework, different mixing protocols can be evaluated with respect to their long term mixing behaviour and compared quantitatively. It is shown that local ‘optimal’ parameter settings for mixing of Newtonian fluids can result in a considerable worse mixing behaviour for non-Newtonian fluids, and vice versa, illustrating the importance of taking the rheology of the fluid into account. © 2000 Elsevier Science B.V.

Keywords: Mixing; Chaotic advection; Cavity flow

1. Introduction

Many fluids in nature and industrial processes show a non-Newtonian fluid behaviour. The viscosity can depend on the shear-rate, or the stresses can depend on the history of the deformation, or both. Despite the fact that the importance of non-Newtonian fluids in industrial processes is recognised in a wide range of studies, only a few studies have been published on mixing of non-Newtonian fluids. Early ventures to describe and understand mixing processes are by Spencer and Wiley [28], Danckwerts [9], and Bigg and Middleman [6]. Recently, mixing of viscous liquids has been associated with the chaotic behaviour of a dynamical system. Aref [4] studied the chaotic advection of passive tracers by the flow of two alternatively rotating point vortices, called the blinking vortex flow. During mixing in such deterministic chaotic flows, complex structures of folds are observed which could be well-defined by considering the...
flow as a Hamiltonian dynamical system. Soon these ideas were adopted for other types of flow, as for example the journal bearing flow [7], and the lid-driven cavity flow [8].

The studies involving non-Newtonian fluids are roughly divided into two parts; one concerning purely elastic non-Newtonian fluids, the other dealing with inelastic non-Newtonian fluid behaviour. Niederkorn and Ottino [22] reported on chaotic advection in the journal bearing flow for shear-thinning fluids, and Ling and Zhang [18] examined the influence of the deformation-rate-dependent viscosity on mixing windows — regions in the parameter space where mixing systems are nearly chaotic. These studies describe a decrease in the quality of mixing for the inelastic fluids compared to Newtonian fluids, both in rate and in extent. Noteworthy is that already significant effects are observed with less than 5% difference in the velocity field (in the $L_\infty$ norm) compared to the Newtonian results. It was observed that shear-thinning reduces the strength of vortices and weakens the link between high and low shear-rate regions. Hence, the mixing performance degrades.

For a particular class of viscoelastic fluids some results have been presented in the literature. If the relaxation time of the fluid is small compared to the characteristic time of the period of a time-periodic flow, the Deborah number is small and quasi-stationary flow can be assumed. Leong and Ottino [16] experimentally studied the effect of addition of polymer to a Newtonian fluid undergoing chaotic advectios in a two-dimensional time-periodic cavity flow. Two different mixing protocols were applied, one flow that is discontinuous in time consisting of an alternating moving top and bottom wall, the other a continuous (sinusoidal) wall movement. In both cases they observed large effects of the Deborah number on the mixing behaviour at relatively low Deborah numbers (defined as the ratio of fluid relaxation time and duration of a flow period) in the order of 0.5. Niederkorn and Ottino [21] presented experiments and simulations in the journal bearing flow involving planar creeping flow between eccentric cylinders. Each cylinder boundary has an angular velocity that behaves like a square wave as a function of time, creating rectangular pulses, and thus the flow possesses a characteristic period. Their experiments show that the area of the flow domain containing chaotic fluid particle trajectories can either increase or decrease, compared to the Newtonian case, depending on the periods associated with the boundary motion. These effects occur for a Weissenberg number (defined as the product of a fluid relaxation time and a characteristic shear rate) as low as 0.04. Another more recent study on chaotic advection in creeping flow of viscoelastic fluids in the journal bearing flow is by Kumar and Homsy [15]. They used perturbation theory for low levels of elasticity to determine semi-analytically the viscoelastic correction to the Newtonian flow field, based on the Oldroyd-B constitutive model. Their goal was to predict how elasticity affects chaotic advection in quasi-steady flows. It was found that elasticity can act to either increase or decrease the area over which chaotic advection occurs, depending on the boundary motion.

For more realistic viscoelastic mixing flows, the analyses are more complicated because of the memory of the fluids. In general, for viscoelastic flows, the transition of the flow between time intervals can not be neglected when external excitations are changed. Generally, viscoelastic computations in complex flows at high Weissenberg numbers has proven to be a tremendous challenge, in particular for systems where singularities are present. Examples include cavity flows with a steadily moving lid, and only a limited number of computational methods provide satisfactory results [5]. The development of stable algorithms which can handle time-dependent problems (with or without singularities) is even a harder task; see for example, the work of Grillet et al. [11] who studied elastic effects in cavity flows. Another issue of importance in viscoelastic flow computations is the choice of a proper constitutive model, which describes the fluid well both under shear and elongation. Although a variety of constitutive models are available, the choice of the model and the choice of the linear and non-linear fluid parameters is a difficult task
Due to these difficulties associated with viscoelastic flow modelling, this paper only considers fluids with an inelastic non-Newtonian rheological fluid behaviour, and future work will cope with the venture of viscoelastic mixing flows at high Weissenberg numbers.

The paper discusses the influences of a shear-rate-dependent viscosity on mixing in two- and three-dimensional prototypical mixing flows, and the Carreau model is used. Results presented here can be used in the future as a reference for viscoelastic mixing flows exhibiting shear-thinning. The goal of the paper is two-fold. First, details of mixing are studied using Poincaré maps, periodic points, and advection of fluid elements. Second, optimisation of the mixing protocol is investigated using a ‘mapping’ method. In all cases, the influence of rheological parameters (the power-law index and the Carreau number) on the mixing process is studied.

2. Mathematical formulation and numerical techniques

The rheological model used throughout this paper is the Carreau model with zero infinite-shear-rate viscosity

\[
\eta = \eta_0 \left(1 + (\lambda \dot{\gamma})^2 \right)^{(\alpha-1)/2},
\]

where \(n\) is the power-law parameter, \(\lambda\) the time constant and \(\eta_0\) is the zero shear-rate viscosity. This model has advantages over the more simple power-law model, since most fluids exhibit a low shear-rate Newtonian plateau and a transition region into the power law regime. The Carreau model accounts for both aspects. The shear-rate \(\dot{\gamma}\) used in Eq. (1) is defined as \(\dot{\gamma} = \sqrt{\mathcal{I}_2 D}\), where \(\mathcal{I}_2 D = (1/2)(\text{Tr}^2(D) - \text{Tr}(D^2))\) is the second invariant of the rate of deformation tensor \(D = (1/2)(\nabla u + (\nabla u)^T)\). The following dimensionless numbers characterise the shear-thinning nature of the flow:

- the shear-thinning index or power-law parameter \(n\),
- the Carreau number, \(Cr = \lambda U/L\), which can be regarded as a dimensionless shear-rate.

The dimensionless form of the conservation equations read

\[
\frac{\partial u}{\partial t} + u \cdot \nabla u = -\nabla p + \frac{1}{\text{Re}} \nabla \cdot 2\eta^* D, \quad \eta^* = \frac{\eta}{\eta_0}, \quad \nabla \cdot u = 0.
\]

The Reynolds number, \(\text{Re} = \rho UL/\eta_0\), is defined here using the zero shear-rate viscosity \(\eta_0\), and \(\text{St} = L/UT\) is the Strouhal number. \(L\) is the characteristic length, \(U\) the characteristic velocity, \(T\) the characteristic time, and \(\rho\) is the density of the fluid.

The time-periodic flows considered are such that \(\text{Re}\) and \(\text{St}\) are small and, as a result, the transient behaviour of the velocity field during switching of cavity wall motions can be neglected, see Fig. 1. The flows are, therefore, approximated as piece-wise steady, and to obtain a solution of Eq. (2) with appropriate boundary conditions, the quasi-stationary Stokes equations are solved to obtain the velocity field. Since no analytical solutions are available for the generalised Newtonian Stokes flow in a lid-driven cavity, numerical techniques are required, like boundary integral methods, finite difference, or finite element methods. As small changes in the velocity field will give rise to different advection patterns, the numerical integration of chaotic particle trajectories requires an accurate approximation of the velocity field. For this reason a spectral element method [19] has been used. The main advantage of a high-order spectral method is that, in comparison with classical low-order finite element, finite volume, and finite difference methods, far less degrees of freedom are required to achieve a desired level of high accuracy.
The time discretisation is based on an approximate pressure correction scheme for Newtonian fluids [29], and was extended for shear-rate dependent fluids [1]. It essentially reformulates the original problem into a Helmholtz equation for the velocity, and a Poisson equation for a pressure-related quantity.

3. Mixing in two-dimensional cavity flows

The key to effective mixing lies in producing repetitive stretching and folding, an operation referred to as a horseshoe map in the mathematics literature. The existence of horseshoe maps indicates that the system is chaotic (see [23] for definitions of chaos in dynamical systems). If we consider a two-dimensional steady velocity field, then the velocity field is integrable and the mixing cannot be chaotic [12,23]. Fluid travels over closed streamlines and the mixing is thus poor: stretching is linear and no folds are created. On the other hand, if the velocity field is time-dependent, for example time-periodic, there is a good chance that the system is chaotic; particles initially close are spread through the whole flow domain. A necessary condition for chaos is the crossing of streamlines. A well-known example of a two-dimensional chaotic flow is a time-periodic cavity flow generated by alternately moving the upper and lower wall for a fixed time period.

The objective here is to analyse the influence of shear-thinning on mixing in a two-dimensional cavity with a time-periodic motion using the so-called TB protocol, see Fig. 1, thus applying a consecutive movement of the top (front) and bottom (back) walls. This flow configuration is extensively studied, both experimentally and theoretically, in the literature for Newtonian fluids, see for example [2,17,23], is used as a prototype flow. In Section 4 a three-dimensional extension of the two-dimensional cavity flow is considered, see Fig. 1. Since a quasi-static steady velocity field is assumed, only one single velocity field computation for the motion of a single wall is required, and the other field, as a result of a constant motion of the opposite wall, is simply found by a coordinate transformation.

The aspect ratio of the two-dimensional cavity is chosen to be the same as in previous studies by Leong and Ottino [17] and others, and equals 5:3. The flow domain $\Omega$, depicted in Fig. 1, is $h < x < h$, $(h = 5/3)$, $-1 < y < 1$, is subdivided into $15 \times 9$ spectral elements of 12th order, the mesh being

![Fig. 1. Schemes of the two- and three-dimensional cavity. In both protocols during the first half of the period the top (back) wall is moved from left to right, and in the second half of the period the opposite (front) wall is moved from right to left. This protocol is denoted as the TB protocol.](image-url)
refined near the moving top wall. The Newtonian velocity field is computed and taken as an initial condition, and the time integration of the pressure correction scheme is continued until a steady state for the non-Newtonian fluid is reached. The criterion used to terminate the integration is based on the relative difference $\epsilon = \|u^{i+1} - u^i\|_\infty / \|u^{i+1}\|_\infty$, where $u^i$ denotes the velocity field after $i$ iterations, and $\|\ldots\|_\infty$ denotes the maximum norm. The tolerance $\epsilon$ to terminate the integration was set equal to $10^{-6}$.

Mixing in this cavity is studied for $n = 0.2, 0.4, 0.6, 0.8, \text{ and } 1.0$ in Eq. (1), leading to four shear-thinning fluids and the Newtonian fluid ($n = 1$), which spans the range of shear-thinning behaviour for practical fluids. In the remainder of this section the Carreau number, $C_r$, is fixed at $C_r = 5$, meaning that shear-thinning behaviour is clearly present in the flow, and the fluid behaves like a power-law fluid. First, the influence of the shear-rate-dependent viscosity on the velocity field is studied. Second, the impact on mixing is analysed using Poincaré maps, chaotic advection of fluid elements, and determination and diagnosing first-order periodic points. Finally, the mapping method is used to study mixing for a wide range of dimensionless wall displacements.

### 3.1. Velocity field

Fig. 2 shows the velocity profiles ($Re = 0$) along two lines in the cavity, when the top wall has a constant dimensionless velocity $u = 1$. The lines on the left show the result for the $x$ component of the velocity at $x = -1.5$; the other lines are results for the velocity at the centre line $x = 0$. Only small deviations from the Newtonian velocity field are detectable near the stationary side walls of the cavity, but in the centre of the cavity the differences are more significant and increase as $n$ decreases: shear-thinning reduces the influence of the moving top wall in the core of the cavity. A typical viscosity distribution in the cavity for the Carreau fluid with parameters $n = 0.6$ and $C_r = 5$ is given in Fig. 3, showing the symmetry of the viscosity and the range of viscosities spanned.

### 3.2. Poincaré maps

The Poincaré map is one of the most simple methods to analyse chaotic mixing flows and is a powerful way to reveal zones of regular and chaotic motion. Once the velocity field has been computed, the analysis

![Fig. 2. The $x$ component of the velocity along the lines $x = -1.5$ and $x = 0$.](image-url)
of the chaotic dynamics is based on integration of material points coordinates

$$\frac{d\mathbf{x}}{dt} = \mathbf{u}(\mathbf{x}, t),$$

and the time dependence of $\mathbf{u}$ is the source of any chaotic behaviour in the system. The integration of Eq. (3) with the initial condition $\mathbf{x} = \mathbf{X}_0$ at $t = 0$ for a time length $T$ gives the position of particle $\mathbf{x}$ at $t = T$. The flow can be represented by the motion

$$\mathbf{x} = \Phi_t(\mathbf{X}), \quad \mathbf{X} = \Phi_{t=0}(\mathbf{X}),$$

mapping particle $\mathbf{X}$ to the position $\mathbf{x}$ after a time $t$. The Poincaré section allows a systematic reduction in complexity of problems by reduction of the number of dimensions [12]. Successive intersections of orbits of a point $\mathbf{x}_1$ after a time $t = T$ define a map $\mathbf{x}_{n+1} = \Phi(\mathbf{x}_n)$. The point $\mathbf{x}_1$ is mapped to $\mathbf{x}_2 = \Phi(\mathbf{x}_1)$, the point $\mathbf{x}_2$ is mapped to $\mathbf{x}_3 = \Phi(\mathbf{x}_2) = \Phi^2(\mathbf{x}_1)$, and so on. The Poincaré map is constructed by plotting all intersections. Note that this viewpoint is purely kinematic and all the complexities in solving the dynamical system are associated with obtaining $\mathbf{u}(\mathbf{x}, t)$. The dynamical system (3) is numerically integrated using an adaptive Runge–Kutta scheme, see [25], and errors in the trajectories of particles are mainly imposed by discretisation errors of the numerical velocity field [27]. It should be noted that the particles are passive markers within the fluid, i.e. there is no diffusion of particles.

In Fig. 4 six examples of Poincaré sections for a Newtonian fluid in a time-periodic cavity flow, induced by consecutive motion of top and bottom wall, are presented for a dimensionless wall displacement $D = 3$, 4, 6.24, 15, 20, and 25, defined as

$$D = \frac{\|d_{\text{top}}\| + \|d_{\text{bot}}\|}{2h},$$

$\quad$
where $d_{\text{top}}$ and $d_{\text{bot}}$ are the displacement of the top and bottom, respectively, while $2h$ denotes the width of the cavity. The Poincaré maps in Fig. 4d and f show global mixing, at least unmixed areas are relatively small. The other plots in Fig. 4 all show multiple large and small unmixed regions. The Poincaré map for $D = 3$ shows that the flow domain mainly is occupied by regular mixing zones, which are surrounded by chaotic mixing zones. Further on in Section 3.5.2, the rate of mixing and mixing efficiency is considered and a larger class of mixing flows is compared. First, the influence of the power-law parameter on the Poincaré sections is considered.

For the non-Newtonian cases the dimensionless displacement parameter $D$ is fixed throughout this section at 6.24, and $d_{\text{top}} = d_{\text{bot}}$. Initially, 25 points are distributed in the cavity and tracked for 15,000
periods. The positions of these points after each period are plotted in the figure. These (asymptotic) pictures show that for increasing $D$ the regions of chaotic and regular mixing change. A comparison of the Poincaré map for the Newtonian fluid, presented in Fig. 4c, and the Carreau fluid, presented in Fig. 5 with $n = 0.8$, shows that the main island is still present, but its size has decreased. For two lower values of the power law parameter $n$, i.e. $n = 0.6$ and $0.4$, the asymptotic pictures show more global mixing. For $n = 0.2$ a large number of islands, mainly around high-order elliptic periodic points (see also forthcoming sections), is observed. These results appear to show that the mixing is improved for stronger shear-thinning fluids when $D = 6.24$. However, while the parameters $n = 0.6$ and $0.4$ display a more uniform asymptotic mixture, it will be shown that the rate of mixing is considerably lower than for either the Newtonian fluid or the Carreau fluid with $n = 0.8$. More details of the chaotic behaviour of the flow are revealed by analysing the chaotic advection of fluid elements and studying the periodic points. This is the subject of the following paragraphs.

### 3.3. Chaotic advection

Further indication of the rate of mixing can be obtained by examining the advection of a fluid strip in the cavity. Tracking of strongly deforming fluid elements is efficiently performed by application of an adaptive front tracking technique [10,31]. An initial strip, shown in Fig. 6, is tracked for two periods for
the Newtonian fluid and the four shear-thinning fluids. The advection results are presented in Fig. 6. The increase in interfacial length (length stretch) is defined as $l_t/l_0$, where the interfacial length between the two fluids at time $t$ be denoted as $l_t$, while the initial length of the interface is defined as $l_0$.

For fluids with different shear-thinning behaviour the increase in interfacial length is given in Table 1. The results show that, for decreasing $n$, a decrease of area generation is observed. For $n = 0.2$ the length stretch is five times less than for the Newtonian fluid, while only a minor difference between the Newtonian fluid and the Carreau fluid with $n = 0.8$ is observed. From the Poincaré sections, it was expected that for $n = 0.6$ and 0.4 a more global mixture would appear, but the rate of stretching is significantly less than for $n = 1.0$ and 0.8. The results in Fig. 6 and Table 1 depend on the position of the initial strip, and a more extensive chaotic advection analysis could consider different locations of the initial strip. A periodic point analysis of the time-periodic flow is independent of any initial condition and is the subject of the next paragraph.

3.4. Periodic points

The important characteristics of chaotic flows are determined by the location and nature of the periodic points. Analysis of the behaviour of mixing protocol parameters on the existence of periodic points provides insight in the physics of mixing. Periodic points are points which return to their original position after one or more periods, and they are classified according to the nature of the deformation at that point.

<table>
<thead>
<tr>
<th>$n$</th>
<th>1.0</th>
<th>0.8</th>
<th>0.6</th>
<th>0.4</th>
<th>0.2</th>
</tr>
</thead>
<tbody>
<tr>
<td>$l_t/l_0$</td>
<td>10.04</td>
<td>9.37</td>
<td>6.71</td>
<td>3.82</td>
<td>2.03</td>
</tr>
</tbody>
</table>

*a As a reference the result for a Newtonian fluid is also given.
Using the definition of the mapping in Eq. (4), a periodic point \( P \) of order \( k \) is defined as

\[
\Phi_T^k(P) = P, \tag{6}
\]

\[
\Phi_T^l(P) \neq P \quad \text{for} \quad l < k, \quad k, l \text{ are integers,} \tag{7}
\]

where \( T \) is the duration of one period of motion. Elliptic periodic points are at the centre of non-mixing regions, that are called islands, clearly visible in the Poincaré maps shown in Figs. 4 and 5. The islands translate, stretch and contract periodically and undergo a net rotation after \( k \) periods of motion. Hyperbolic periodic points are centres of stretching in the flow, since in their neighbourhoods the fluid is compressed in one eigendirection and stretched in the other eigendirection. The folding of fluid elements is a result of interactions of elliptic and hyperbolic points [3].

The periodic points in this flow could be located using a general computational method presented in [2]. However, a more elegant way to find periodic points in this particular TB flow is to use the symmetry present in the flow, see [20]. Fig. 7 displays two plots: the streamlines for the flow with a moving top, and the trajectory of a first-order periodic point. Meleshko and Peters [20] showed that the trajectory of any first-order periodic point for the TB protocol intersects with the vertical centre line at the times \( t = 1/4T \) and \( 3/4T \), where \( T \) is the duration of a period. To find periodic points, the centre line \( x = 0 \) is tracked from \( t = 1/4T \) to \( 3/4T \), and the number of intersections between the original and deformed lines equals the number of periodic points. Their actual position can be calculated by tracking these intersections from \( t = 3/4T \) to \( T \). The type of the point, elliptic or hyperbolic, can be found by evaluating the displacement gradient matrix \( \mathbf{F}_x = (\nabla_x \Phi(X))^c \) at the periodic points [2]. Note that this algorithm to search for periodic points will also find periodic points which are not located on any line of symmetry. An extension of the algorithm to locate higher-order periodic points is presented in [20]. In this paper we limit ourselves to first-order points since they usually have the largest effect on the rate of mixing (an example where higher-order points are more important is presented in [13]).

The vertical centre line \( (x = 0) \) is tracked from a quarter of a period to three quarters of a period for the shear-thinning fluids and the Newtonian fluid, see Fig. 8. The number of first-order periodic points follows from the number of intersections with the line of symmetry \( x = 0 \). For the Carreau fluid with \( n = 0.8 \) the number of intersections, and thus the number of periodic points, is equal to 5, which is the same as for the Newtonian fluid. For the Carreau fluids with a lower parameter \( n \), the line deforms less...
Fig. 8. Deformation of the line $x = 0$ from $t = 1/4T$ to $3/4T$ for five different fluids. The number of intersections with the original line reveals the number first-order periodic points. Note that, for computational reasons, the initial centre line does not touch the upper and lower walls.

and, as a result, the number of periodic points is only three. Obviously periodic points ‘collapse’ if $n$ is decreased from $n = 0.8$ to $0.6$. For the flow with fluid parameter $n = 0.2$ only one single (elliptic) periodic point remains. The decrease in number of first-order periodic points, caused by reduction in shear-rate magnitude, usually results in a decrease of local mixing efficiency, since hyperbolic periodic points, which account for repetitive high stretching in mixing flows, may be lost.

A similar effect as shown in Fig. 8 is observed when the rheology of the fluid is fixed and the dimensionless displacement parameter $D$ is decreased. However, the differences in mixing behaviour for the Newtonian and shear-thinning fluids cannot be all attributed to a reduction of an effective dimensionless displacement parameter. This will become clear in the following Section 3.5.

3.5. Application of the ‘mapping’ method

The methods applied in the previous sections discuss details of mixing, and have proven to be elegant tools to analyse the chaotic behaviour of mixing flows. In principle it is possible to obtain all the relevant information on the behaviour of a mixing protocol, and analyse whether it may lead to (global) chaotic mixing. Since these techniques are relatively expensive, it is computationally impossible to qualitatively or quantitatively compare a large number of different mixing protocols, or even a single mixing protocol for different dimensionless displacements. Optimisation of mixing is thus impractical. To answer this question a more flexible and efficient technique is necessary and introduced in the next sub-section.

3.5.1. The technique

The ‘mapping’ method is proposed based on the original ideas of Spencer and Wiley [28], and the idea is not to track each material volume in the flow domain separately, but to create a discretised mapping from a reference grid to a deformed grid. Instead of tracking the boundaries of material volumes a set of distribution matrices are used to advect concentration in the flow. Within the mapping method a flow domain $\Omega$ is subdivided into $N$ non-overlapping sub-domains $\Omega_i$ with boundaries $\partial \Omega_i$. The boundaries
Fig. 9. (Left) Initial sub domain discretisation (10 × 6 grid) and (right) deformed grid after displacement of the top wall by two times its length. During the actual computations a finer 200 × 120 grid was used.

∂Ωi of these sub-domains, represented by polygons, are tracked in a flow from \( t = t_0 \) to \( t_0 + \Delta t \) using an adaptive front tracking model [10], and result in deformed polygons. The subdivision of \( \Omega \) into \( \Omega_i \)'s is not related to any finite differences, or finite element discretisation used to solve the velocity field in \( \Omega \). Next, a discretised mapping from the initial grid to the deformed grid is constructed, see Fig. 9. The distribution of, for example, fluid concentration in the original grid is then mapped to obtain a new distribution.

The area of the intersections of the deformed sub-domains with the original ones, determine the elements of the mapping (or distribution) matrix \( \Psi \), where \( \Psi_{ij} \) equals the fraction of the deformed sub-domain \( \Omega_j \) at time \( t = t_0 + \Delta t \) that is found in the original \( t = t_0 \) sub-domain \( \Omega_i \)

\[
\Psi_{ij} = \frac{\int_{\Omega_i} \int_{\Omega_j} \text{d} \Omega}{\int_{\Omega_j} \text{d} \Omega}.
\]  

(8)

The polygonal descriptions of the sub-domains are used to determine the matrix elements \( \Psi_{ij} \), and the accuracy of these elements is determined by the accuracy of the velocity field and the error tolerances defined in the adaptive front tracking procedure. Note that the matrix \( \Psi \) is essentially sparse if \( \Delta t \) is not too large [14].

The computation of the matrix elements \( \Psi_{ij} \) is time consuming (more than 100 h using a 12 CPU multiprocessor 225 MHz R10K Silicon Graphics\textsuperscript{TM}) for the shear-thinning examples presented later in this section. However, once the matrices are obtained, matrix–vector multiplications are extremely fast (less than 1 s on a single CPU 100 MHz Pentium\textsuperscript{TM}). The combination of a number of matrices for different wall displacement parameters \( D \) are used in [14] to compare different Newtonian mixing protocols. For potential ‘optimal’ mixing protocols, mixing efficiency diagrams are created, which can be used to choose an optimal dimensionless displacement parameter. Here this technique is applied to investigate the influence of shear-thinning behaviour on mixing for a wide range of the flow parameter \( D \).

The quantity being mapped here, as an example, is the averaged concentration of a marker fluid, which is rheologically identical to the matrix fluid. The distribution of the marker fluid is described by a column vector \( \mathbf{C} \), and its components \( C_i \) are the locally averaged concentrations in each sub-domain \( \Omega_i \). If the
initial distribution at $t_0 = 0$ is described by the concentration vector $\mathbf{C}^0$, the concentration after time $\Delta t$ can be computed as $\mathbf{C}^{\Delta t} = \Psi^0 \mathbf{C}^0$. An initial condition can be a strip of fluid like in Fig. 6, or for example, the cavity is filled with half white and half black fluid. If the same flow is continued (repetitive mixing), the concentration after $k$ steps is $\mathbf{C}^{k\Delta t} = \Psi^k \mathbf{C}^0$. Spencer and Wiley [28] suggested to analyse mixing behaviour by studying $\Psi^k$. However, this is not feasible for high spatial resolution. In this case the matrix $\Psi^k$ will not be sparse for large $k$, because fluid from every sub-domain is advected into most, or even all, of the other sub-domains. Instead of studying $\Psi^k$, the evolution of the vector $\mathbf{C}^k$ is studied. The concentration vector $\mathbf{C}^k$ is computed in a sequence

$$\mathbf{C}^{i+1} = \Psi \mathbf{C}^i,$$  \hspace{1cm} \text{(9)}

without a direct evaluation of $\Psi^k$. A simple quantitative criterion to characterise the degree of ‘mixedness’ is needed to describe the mixed state. A useful characteristic, which is called ‘discrete intensity of segregation’, is defined as

$$I = \frac{1}{\bar{c}(1 - \bar{c})} \frac{1}{A} \sum_{i=1}^{N} (c_i - \bar{c})^2 a_i, \quad \text{where} \quad \bar{c} = \frac{1}{A} \sum_{i=1}^{N} c_i a_i, \quad A = \sum_{i=1}^{N} a_i.$$  \hspace{1cm} \text{(10)}

The sum is computed over all internal cells, $N$ is the number of cells, $c_i$ the concentration of the marker fluid in the cell $i$ that has the area $a_i$, $A$ the total area of all cells taken into account, and $\bar{c}$ is the average concentration of marker fluid in $\Omega$. As the grids used in this work are regular and all their cells have equal volumes, formula (10) can be simplified to

$$I = \frac{1}{\bar{c}(1 - \bar{c})} \frac{1}{N} \sum_{i=1}^{N} (c_i - \bar{c})^2, \quad \text{with} \quad \bar{c} = \frac{1}{N} \sum_{i=1}^{N} c_i.$$  \hspace{1cm} \text{(11)}

The definition of the mixing measure $I$ is similar to the standard intensity of segregation as introduced by Danckwerts [9]. Similar measures were used by Tucker [30]. Unlike the intensity of segregation, the discrete intensity of segregation is defined through the concentration averaged in the finite sized cells, and not via the integral over point-wise values of concentration. Note that $0 \leq I \leq 1$ and for $I = 0$ the mixture is ideal; $I = 1$ represents an unmixed state.

3.5.2. Mapping results for two-dimensional flow

The mapping technique was applied for the top–bottom, i.e. TB mixing protocol as studied earlier. The mapping matrix is constructed on the $200 \times 120$ grid defined on the domain $\Omega_m$: $\delta \times h \leq x \leq \delta \times h, -\delta \leq y \leq \delta$, where the factor $\delta = 0.985$ is used for computational reasons. The geometric corner and boundary singularities of the cavity flow result in infinite polymer stresses, and particle paths close to the corners lead to unacceptable long program runs (if $\delta \rightarrow 1$). Therefore, the excluded domain $\Omega \setminus \Omega_m$ is treated as a single cell in the mapping matrix. Mapping matrices are constructed with $D = 0.25, 1, 2, \text{ and } 4$, where $T$ is again the duration of one period of motion. It should be noted that numerical diffusion is introduced as a side effect of the mapping method, and the amount of diffusion is determined by the grid size, the size of the domain $\Omega, \Omega_m$, and the number of subsequent mapping steps needed. As a result the minimum intensity of segregation is here of the order $10^{-3}$. In [14] some modifications in the computation of the mapping matrix are proposed, which makes it possible to use finer grids and as a result significantly lower values of the intensity of segregation are reached.
The initial distribution of fluid which is mixed is as follows: black fluid ($C = 1$) is placed in the left half of the cavity, while in the other half of the cavity white fluid ($C = 0$) is placed. The fluids are mixed using the mixing protocol TB for a large number of values of the mixing parameter $D$. Discrete intensity of segregation, as introduced in Eq. (11), is used as a mixing measure. For some dimensionless displacements results are presented in Fig. 10 for the Newtonian fluid and the Carreau fluids with power-law parameter $n = 0.6$ and 0.2. The Carreau number, Cr, is fixed and equals 5. The total number of wall movements, a direct measure for the energy input, is equal for all these plots and equals 32, i.e. we compare 16 times a $D = 2$ wall displacement, with eight times $D = 4$, four times $D = 8$, two times $D = 16$, and one step of $D = 32$. Horizontally the influence of the power-law parameter is compared. For this relatively small number of wall movements it is observed that the Newtonian fluid mixes better than the Carreau fluid with $n = 0.6$, which again mixes better than the Carreau fluid with $n = 0.2$. Moreover, further on in Figs. 11 and 12 it is, however, concluded that if more wall displacements are applied, shear-thinning behaviour can lead to better mixing.

For the same three fluids ($n = 1, 0.6, 0.2$) mixing efficiency plots are computed. These plots are constructed by considering fluid mixing for a large number of combinations $k \times D$, where $k$ is the number of wall movements, and plotting the discrete intensity of segregation. The maximum dimensionless displacement was equal to 16, and the number of wall movements is limited to 32. To construct each of the mixing efficiency diagrams presented in Fig. 11, a total of 8192 mixing simulations are performed using the mapping approach. The picture on the top of Fig. 11 shows the results for the Newtonian fluid, while the other two plots shows the results for the Carreau fluids. White regions correspond to parameter subsets leading to efficient mixing; dark regions correspond to poor mixing parameter settings.

The Newtonian results in Fig. 11 show a number of intervals for $D$ of low and high intensity of segregation. For $0 \leq D \leq 4$ the plot shows that the mixing quality is poor, and even for an extreme large number of wall movements, beyond the range of this figure, mixing remains poor. For a somewhat larger dimensionless displacement of $D$, where $4 \leq D \leq 5.5$, mixing becomes considerably more efficient. In the range of $5.5 \leq D \leq 9$ a poorer mixing quality results, but again increasing the parameter $D$ to 10 leads to more efficient mixing zone again. Between $D = 10$ and 12 a third zone of less efficient mixing is visible, which transfers to the chaotic zone $12 \leq D \leq 16$.

As a result of the shear-thinning behaviour of the Carreau fluids $n = 0.6$ and 0.2 shifts in the corresponding mixing efficiency diagrams are observed. Decreasing the power-law parameter $n$ enlarges the minimum dimensionless displacement required to obtain a nearly global chaotic mixing flow. For the strongest shear-thinning fluid, displacement steps of $D = 8$ are required to get good mixing. The zones of good and bad mixing as found for the Newtonian fluid are also observed for the non-Newtonian fluids, but the streaks appear to be wider.

A more clear comparison of the mixing behaviour of the three fluids is given in Fig. 12. Here intensity of segregation is plotted as a function of number of wall displacements for a fixed dimensionless displacement parameter. For $D = 2$ mixing quality is very poor for all three fluids, as expected from the (more qualitative) results shown in Fig. 11. For $D = 6$ it is first observed that shear-thinning fluids can lead to better mixing than in the Newtonian case. Here, the curve for $n = 0.6$ is below the Newtonian curve, which again is below the $n = 0.2$ curve. In this case a large island is present for the Newtonian fluid (see Fig. 4), and obviously also for $n = 0.2$. If $D$ is increased to $D = 8$ results are inverted and the Carreau fluid with $n = 0.2$ leads to the most efficient mixing. Already after 30 wall displacements the intensity of segregation is orders lower than for the fluids with $n = 0.6$ and 1.0. For $D = 12$ the weaker shear-thinning fluid leads to better mixing, where for the strong shear-thinning fluid the intensity of
Fig. 10. Mixing patterns for the Newtonian (left) and the two shear-thinning fluids (middle and right). The initial distribution of fluid is as follows: black fluid ($C = 1$) is placed in the left half of the cavity, while in the other half of the cavity white fluid ($C = 0$) is placed.
Newtonian:

Carreau $n = 0.6$:

Carreau $n = 0.2$:

Fig. 11. Mixing quality plots for a Newtonian (top) and shear-thinning Carreau fluid with $n = 0.6$ (middle) and a Carreau fluid with power-law parameter $n = 0.2$ (bottom). The Carreau number equals five in all cases. Grey values denote different values for $(1 - I)$; black represents bad mixing. The dashed hyperbolic contours represent lines of equal energy input.
Fig. 12. Mixing quality plots for the Newtonian and two shear-thinning fluids for a constant dimensionless wall displacement.

segregation is bounded by approximately $1 \times 10^{-3}$. Increasing to $D = 14$ shows that the Newtonian fluids has the largest rate of mixing, and only after eight wall displacements the lower limit of intensity of segregation is reached. Note that in Fig. 12, the total energy input in the mixing process is, at a fixed number of wall displacements, linearly proportional to $D$.  

4. Three-dimensional cavity flows

As far as known to the authors, no studies have been reported in the literature on the analysis of three-dimensional non-Newtonian mixing flows focusing on the behaviour of periodic points. The mixing protocol studied here is a straightforward extension of the protocol for the two-dimensional cavity used in the preceding section, and consists of a time-periodic motion of the front and back wall. All other walls are stationary. As a result of the lid and bottom of the cavity, three-dimensional effects are expected. The dimensionless displacement $D$ equals 7, similar to the Newtonian results presented in [2].

Similar computational techniques as used for the two-dimensional problem are applied to obtain the velocity field in the three-dimensional cavity. The flow domain is subdivided in $6 \times 8 \times 6$ spectral elements each of sixth order. The computed Newtonian velocity is again taken as an initial field to obtain the shear-thinning velocity fields. In principle it is possible to construct a Poincaré map for a three-dimensional cavity flow, and analyse the mixing behaviour similar as done for the two-dimensional problems. However, the outcome is less simple to use since the resulting set of points is located in the three-dimensional cavity and regions of bad mixing are less easy detected. Therefore, we will go directly to the core of the problem and study the influence a shear-rate dependent viscosity on first-order periodic points for the TB protocol.

Periodic points are located for three different Carreau fluids with power-law parameters $n = 0.9, 0.7,$ and 0.6 (this range is considered here, since an interesting transition in mixing quality is observed around $n = 0.65$). The technique applied to locate the periodic points is similar to the two-dimensional case with

---

Fig. 13. Periodic lines from shear-thinning fluids with power-law parameters $n = 1.0, 0.9, 0.7,$ and 0.6.
the difference that here a two-dimensional plane located at the plane of symmetry is tracked from a quarter to three quarters of a periodic. The intersection of the deformed and the original plane determines the set of periodic points. The type of the first-order periodic points is determined by studying the eigenvalues of the displacement gradient matrix at the periodic points.

Similar to results discussed in [2] the sets of periodic points form periodic lines, and the nature of the periodic points can change along each line. The results are displayed in Fig. 13, where the set of lines corresponding to the Newtonian fluid are added as a reference. The thin lines are of hyperbolic type, whereas the thick lines are of elliptic type. Symmetry, which is present for this flow, is also revealed by the periodic points. The planes of symmetry are the mid-plane $z = 0.5$ and the plane $y = 0$. It appears that within this range periodic points collapse (for decreasing $n$) and that the type of a periodic point may change. The front view of Fig. 13 shows that for $n = 0.9$ periodic lines are still present in the left half of the cavity; for the lower $n$ parameters these lines disappear. The side view picture clearly shows that, for a decreasing $n$, the length of the lines decreases and the type changes along the line. For $n = 0.6$ the lines are almost completely elliptic. From these results it is expected that the mixing in this cavity flow will be poorer for shear-thinning fluids. If the power-law index $n$ would be decreased further, the set of three periodic lines would eventually collapse and a single (elliptic) periodic line remains.

To demonstrate the influence of the parameter $n$ on mixing, a blob is advected in the flow for four periods of motion. The initial blob is located such that it touches the periodic lines for $n = 0.6$ and 0.7,

![Position of the initial blob](image1)
![Position of the initial blob relative to the periodic lines](image2)
![Deformation of the blob for $n = 0.6$](image3)
![Deformation of the blob for $n = 0.7$](image4)

Fig. 14. Deformation after four periods of a blob positioned between periodic lines belonging to different shear-thinning parameters. The centre of the blob lies on the periodic line for $n = 0.65$, while the radius of the blob is chosen such that the blob touches the periodic lines for $n = 0.6$ and 0.7.
see Fig. 14. A large difference in the advection of the blob is observed. For the Carreau fluid with $n = 0.7$ the blob is stretched and folded more extensively than for $n = 0.6$. Such an outcome is not surprising, since the periodic points in this region are hyperbolic for $n = 0.7$ but elliptic for $n = 0.6$. These results show that the mixing behaviour can locally be very sensitive to the rheological properties of the fluid. A natural route to extend the three-dimensional analysis would be the development and application of the mapping method, and to study the influence of dimensional wall displacement on the mixing quality for the different shear-thinning fluids. This is the subject of future research.

5. Summary and conclusions

This paper deals with a Carreau shear-thinning viscosity model to study mixing characteristics of non-Newtonian fluids. To obtain the corresponding velocity fields for a two- and three-dimensional cavity flow, computational methods are used to study the inelastic non-Newtonian effects. Even though the deviations from the Newtonian flow of steady-state velocity fields are relatively small, large effects are observed in the chaotically advected patterns in the time-periodic two- and three-dimensional flows. In general, for the systems studied in this work, the overall effect of a shear-thinning viscosity is a decrease in the rate and extent of mixing. The shear-thinning viscosity results in a decrease of stretching in the flow, a primary route to efficient mixing.

For the two-dimensional flows, it is shown that hyperbolic (unstable) periodic points disappear if shear-thinning is increased, or they transform into elliptic (stable) periodic points. For the three-dimensional flows, similar effects are observed for the set of periodic lines present in the cavity. The length of the lines decreases if shear-thinning is increased, and the nature of parts of these lines change from hyperbolic to elliptic. The overall effect is again a decrease in the rate of mixing. Shear-thinning behaviour of the fluid can, however, lead to a more global mixture (also observed by Niederkorn and Ottino [22]), and for the two-dimensional system several examples are provided. Similarly, mixing behaviour in three-dimensional shear-thinning flows can be (locally) very sensitive to rheological fluid parameters, and future work is focused to study global effects on mixing using a three-dimensional version of the mapping method.

The application of the two-dimensional mapping method for different fluids, makes a comparison of mixtures possible for a much larger variation of parameters. In mixing efficiency diagrams the results are easily quantitatively compared and parameter zones of good and bad mixing are separated. From the plots it is immediately clear that zones of good mixing found for a Newtonian fluid, can be zones of bad mixing for a shear-thinning fluid or vice versa. The non-Newtonian behaviour of the fluid not only leads to a shift of these zones, since some of them increase in width, while other zones become more narrow. Hence, the differences cannot be only attributed to a reduced shear-rate magnitude. The results summarised in this paper clearly show the (possible) influence of changes in the rheology of the fluid on the mixing properties. Moreover, examples are given where mixing may become either better or worse compared to the Newtonian system.

Future research will study elastic effects in mixing. For Boger fluids we can study pure elastic effects and compare with the Newtonian results. For viscoelastic flows with shear-thinning behaviour a comparison with the shear-thinning results presented in this paper is possible. Moreover, an experimental setup of a three-dimensional cavity flow is currently built in our laboratory and will be used to validate results presented here.
Acknowledgements

The authors would like to acknowledge support by the Dutch Foundation of Technology (STW), Grant No. EWT44.3453, and the Dutch Polymer Institute (DPI), Project No. 161.

References