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Effect of confinement and viscosity ratio on the dynamics of single droplets during transient shear flow

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Synopsis

The deformation and orientation of droplets during transient shear flow is studied in a counterrotating device using microscopy. The effect of the degree of confinement and viscosity ratio is systematically investigated. The system consists of polydimethylsiloxane droplets of varying sizes and viscosities dispersed in a polyisobutylene matrix. The observations are compared with the predictions of an adapted version of the Maffettone and Minale model [Maffettone, and Minale, J. Non-Newtonian Fluid Mech. 78, 227–241 (1998)] which includes confinement effects [Minale, Rheol. Acta 47, 667–675 (2008)]. For flow start-up at low capillary numbers, the deformation of confined droplets and their orientation towards the flow direction are increased with respect to the unconfined situation for all viscosity ratios under investigation. The confined model results for start-up and the experimental data at low capillary numbers are in good agreement both showing similar monotonous transients. At high degrees of confinement and high shear rates, one or more overshoots in the droplet deformation are experimentally observed, depending on the viscosity ratio. In addition, droplets become sigmoidal when highly confined. Under these conditions, the confined Maffettone and Minale model, which assumes an ellipsoidal droplet shape, cannot be used to predict the droplet behavior. The relaxation of confined droplets upon cessation of steady-state shear flow is also studied. It is experimentally observed that confinement only affects the relaxation at degrees of confinement above 60% of the gap spacing. Highly confined droplets experience a slightly slower relaxation with respect to bulk conditions. The relaxation predictions of the confined model are in rather good agreement with the experimental data. © 2008 The Society of Rheology. [DOI: 10.1122/1.2978956]

I. INTRODUCTION

The analysis of the dynamics of droplets dispersed in an immiscible fluid started in the 1930s with the well known studies of Taylor (1932, 1934). Ever since, a great deal of theoretical, experimental, and numerical work has been performed to characterize two-phase systems during flow [Rallison (1984); Stone (1994); Ottino et al. (1999); Tucker and Moldenaers (2002); Guido and Greco (2004)]. It was shown that in the absence of buoyancy and inertia effects, the behavior of a Newtonian droplet in a Newtonian matrix during shear flow is determined by two nondimensional parameters: the capillary number $Ca = \eta_m R \dot{\gamma} / \Gamma$, where $\eta_m$, $R$, $\dot{\gamma}$, and $\Gamma$ denote, respectively, the matrix viscosity, the

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droplet radius, the shear rate, and the interfacial tension) and the viscosity ratio \( \lambda \) (\( = \eta_d/\eta_m \), in which \( \eta_d \) is the droplet viscosity). In shear flow for \( \lambda \) close to 1, a Newtonian droplet deforms monotonically towards a steady-state shape and rotates simultaneously towards a specific orientation, as long as \( \text{Ca} \) does not exceed the critical value for breakup. For Newtonian systems, this critical capillary number only depends on the type of flow and the viscosity ratio \( \lambda \) [Grace (1982)]. Various models have been proposed to describe the deformation, orientation and breakup of droplets subjected to a flow field [Rallison (1984); Stone (1994); Guido and Greco (2004)]. Many among these determine the droplet deformation assuming small deviations from sphericity so that a perturbation expansion could be adopted [e.g., Cox (1969); Frankel and Acrivos (1970); Rallison (1980)]. Other models start from the assumption that the droplet shape is ellipsoidal at all times [Guido and Greco (2004)]. A widely used model in this respect is the Maffettone and Minale model [Maffettone and Minale (1998, 1999)]. This simple phenomenological model is capable of predicting the time dependent axes and the orientation angle of ellipsoidal Newtonian droplets in a Newtonian matrix in any type of flow field.

Recently, a growing trend towards miniaturization is observed in the chemical processing industry. This is reflected in a broad field of applications such as the design of microreactors, micromixers, and other microfluidic applications [Thorsen et al. (2001); Link et al. (2004); Stone et al. (2004); Utada et al. (2005)]. This growing trend has raised the need to analyze fluids in a confined environment. In emulsification or blending processes, the proximity of one or two walls can alter the deformation and breakup behavior of single droplets during flow [for an overview, see Vananroye et al. (2006c)]. In such confined blends, several morphological transitions are perceived, ranging from a completely disordered state to layered structures with pearl necklaces and strings [Migler (2001)]. For example, for blends consisting of polyisobutylene (PIB) and polydimethylsiloxane (PDMS) with equal viscosities, these observations were presented in a morphology diagram in the parameter space of shear rate and concentration [Pathak et al. (2002)].

Also the deformation of confined Newtonian droplets in concentrated blends was investigated [Pathak and Migler (2003); Vananroye et al. (2006a); Tufano et al. (2008)]. It was shown that for individual droplets in 1% and 5% blends, bulk behavior still prevails up to a confinement ratio—defined as the ratio of droplet diameter \( 2R \) to gap spacing \( d \)—of 0.4.

In order to exclude concentration effects, single droplet experiments were conducted to study the deformation, orientation, and breakup during confined shear flows [Vananroye et al. (2006b, 2007), Sibillo et al. (2006)]. These single droplet experiments clearly revealed specific effects of confinement on the breakup behavior: when \( \lambda < 1 \), confinement suppresses breakup whereas for \( \lambda > 1 \), breakup is enhanced [Vananroye et al. (2006b)]. In addition, droplet breakup can also occur in a confined shear flow for viscosity ratios exceeding the critical value for breakup in unconfined conditions (\( \lambda_{\text{crit}} = 4 \)). For viscosity ratios ranging from 0.3 to 5, it was shown that confinement induces both an increase in steady-state deformation and an increased orientation towards the flow direction [Sibillo et al. (2006); Vananroye et al. (2007)]. For viscosity ratios both below and above unity, the steady-state deformation parameter is in agreement with the predictions of the analytical theory by Shapira and Haber (1990) for confined droplets [Sibillo et al. (2006); Vananroye et al. (2007)]. However, this theory, which resulted in an expression for the droplet deformation that consists of the small deformation result of Taylor (1932, 1934), corrected with a term depending on the degree of confinement, is limited to small deformations. In addition, it only yields the steady-state deformation, and a constant orientation angle of 45° is predicted for all capillary numbers and confinement ratios. To overcome these drawbacks, the phenomenological Maffettone–Minale model for bulk flow was recently adapted to include confinement effects [Minale (2008)].
TABLE I. Zero-shear viscosities at 24 °C and activation energies of the blend components; viscosity ratios and critical capillary numbers of the blends at 24 °C.

<table>
<thead>
<tr>
<th>Grade</th>
<th>η₀(24 °C) (Pa s)</th>
<th>Eₐ (kJ/mole)</th>
<th>λ = ηₚDMS / ηₚPIB (24 °C)</th>
<th>Cₐcrit(λ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PIB Parapol 1300</td>
<td>101</td>
<td>64.4</td>
<td>Matrix</td>
<td>Matrix</td>
</tr>
<tr>
<td>PDMS Silbione 70047V30000</td>
<td>30</td>
<td>12.6</td>
<td>0.30</td>
<td>0.48</td>
</tr>
<tr>
<td>PDMS Rhodorsil 47V100000</td>
<td>103</td>
<td>12.9</td>
<td>1.02</td>
<td>0.48</td>
</tr>
<tr>
<td>PDMS Rhodorsil 47V200000</td>
<td>200</td>
<td>12.6</td>
<td>1.98</td>
<td>0.69</td>
</tr>
</tbody>
</table>

Data on the transient dynamics of confined droplets are scarce in literature. For example, for a viscosity ratio of 1, Sibillo et al. (2006) studied drop deformation in shear flow. The main effects reported were complex oscillating transients and very elongated droplet shapes. For a viscosity ratio of unity, numerical simulations—either using a boundary integral method or a volume-of-fluid method—have been performed [Janssen and Anderson (2007); Renardy (2007)]. Vananroye et al. (2008) demonstrated that the experimental transient and steady-state results were in excellent agreement with the boundary integral simulation results. In addition, the simulations are capable of predicting the complete shape of highly confined droplets, a feature that is not present in the models of Shapira and Haber (1990) and Minale (2008). In the present work, the transient dynamics of single Newtonian droplets in a Newtonian matrix is systematically studied during shear flow. Both the start-up and the relaxation behavior are determined for a series of blends with variable viscosity ratios and degrees of confinement. The results are compared with the adapted version of the phenomenological model of Maffettone and Minale that includes confinement effects [Minale (2008)].

II. MATERIALS AND METHODS

A. Materials

The two-phase system consists of a PIB liquid as the matrix phase (Parapol, obtained from ExxonMobil Chemical, USA) and PDMS oils as the droplet phase (Rhodorsil and Silbione, obtained from Rhodia Chemicals, France). All pure components are transparent liquids at room temperature and have a constant viscosity up to the highest shear rates used in the present experiments. Since elasticity effects are fairly low, the pure materials can be considered as Newtonian under the measurement conditions [Vinckier et al. (1996)]. Gravitational effects can be omitted since the densities of the pure materials are nearly identical (ρₚIB = 890 kg/m³ at 20 °C and ρₚDMS = 970 kg/m³ at 20 °C) [Minale et al. (1997)]. In Table I, the measured zero-shear viscosities η₀ at 24 °C and the activation energies Eₐ of the components are summarized together with the viscosity ratios λ, and a calculation of the critical capillary numbers Cₐcrit according to de Bruijn (1989). The interfacial tension Γ of the PDMS/PIB system, measured by Sigillo et al. (1997), is 2.8 mN/m and is independent of the molecular weight of PDMS for grades with relatively high molecular weight, as is the case here [Kobayashi and Owen (1995)].
B. Methods

The dynamics of droplets is studied in a counterrotating parallel plate flow cell (Paar-Physica). The advantage of using a counterrotating device is the possibility to create a stagnation plane in the flow field, which facilitates continuous observation of a droplet during flow. More details about the setup are given elsewhere [Vananroye et al. (2006b)]. For practical reasons, the gap spacing $d$ between the parallel plates is chosen to be 1 mm. The degree of confinement is varied by injecting droplets with different sizes (diameter $2R$ ranging from 200 to 900 μm) in the matrix fluid. The droplets are carefully positioned at the center plane between the two plates and remain there for the duration of the tests due to the close matching of the densities of PIB and PDMS in combination with a high matrix viscosity. Droplets are observed by means of a Wild M5A stereo microscope and a Basler A301f camera. Both microscope and camera are mounted on vertically translating stages such that droplets can be visualized in the velocity-vorticity plane as well as in the velocity-velocity gradient plane [Figs. 1(a) and 1(b)]. Images are recorded using Streampix Digital Video Recording Software (Norpix) and analyzed using Scion-Image Software. During flow start-up, images are first taken in the velocity-vorticity plane until steady-state is reached. Then, the flow is stopped and after relaxation of the droplet, the same experiment is performed while capturing images in the velocity-velocity gradient plane. By fitting an equivalent ellipse to the drop contour in the velocity-vorticity plane, $L_p$ and $W$ are obtained at each time instant. From the sideview images, the height $L_v$ of the droplet is measured at the corresponding times. Using the volume preservation condition, the projection equations for an ellipsoid on a plane, and the measured dimensions $L_p$, $W$, and $L_v$, the two remaining droplet axes $L$ and $B$ and the orientation angle $\theta$ of a droplet during transient flow are determined.

The experimental results are compared with the predictions of the phenomenological model proposed by Minale (2008) which is an adapted version of the bulk model of Maffettone and Minale (1998) (MM model) to include confinement effects. The bulk MM model assumes that during flow, the droplet displays an ellipsoidal shape which can be expressed by a symmetric, positive-definite, second rank tensor $S$ with eigenvalues describing the square semiaxes of the ellipsoid. The evolution equation of $S$ is given by

$$\frac{dS}{dt} - \Omega \cdot S + S \cdot \Omega = -\frac{f_1}{\tau} [S - g(S)I] + f_2(E \cdot S + S \cdot E).$$

In Eq. (1), $t$ represents the absolute time, $\tau = \eta_m R / \Gamma$ is a characteristic emulsion time, $I$ is the second rank unit tensor, and $E$ and $\Omega$ are the deformation rate and vorticity tensors of

FIG. 1. Scheme of a deformed droplet with the geometrical parameters in shear flow; (a) velocity-vorticity plane; (b) velocity-velocity gradient plane.
the appropriate flow field. For simple shear flow, \( E \) and \( \Omega \) are given by

\[
E = \frac{1}{2} \begin{bmatrix}
0 & \dot{\gamma} & 0 \\
\dot{\gamma} & 0 & 0 \\
0 & 0 & 0
\end{bmatrix}
\]

and

\[
\Omega = \frac{1}{2} \begin{bmatrix}
0 & \dot{\gamma} & 0 \\
-\dot{\gamma} & 0 & 0 \\
0 & 0 & 0
\end{bmatrix}
\]

The parameters \( f_1 \) and \( f_2 \) are nondimensional, non-negative functions of \( \lambda \) and \( Ca \), chosen to recover the small deformation limits of Taylor (1932, 1934) for the droplet shape. \( g(S) \) is a specific function to preserve the volume of the droplet during flow

\[
g(S) = \frac{3 \text{III}_{S}}{\text{II}_{S}},
\]

with \( \text{III}_{S} \) and \( \text{II}_{S} \) the third and second invariants of \( S \). For bulk conditions, the model is well capable of predicting the evolution of the three main axes of a droplet and its orientation in an arbitrary flow field [Maffettone and Minale (1998)]. In the adapted model proposed by Minale (confined MM model) the same evolution equation for \( S \) is used, but new parameters \( f'_1 \) and \( f'_2 \) are defined in a way that the droplet axes recover the analytical limits of the small deformation theory of Shapira and Haber (1990) for confined droplets in shear flow. However this way, only the ratio of \( f'_1 \) and \( f'_2 \), which now becomes a function of the degree of confinement, could be imposed. The remaining degree of freedom in the choice of \( f'_1 \) and \( f'_2 \) was assigned with a best fit through the experimental confined steady-state data of Vananroye et al. (2007) and Sibillo et al. (2006). More information about this model and expressions for \( f'_1 \) and \( f'_2 \) can be found in Minale (2008). So far, the results of this model are not yet validated for transient shear flow.

III. RESULTS AND DISCUSSION

A. Start-up of shear flow

In a first series of experiments, the effect of confinement on the start-up behavior of single droplets is studied. Two start-up conditions are explored; in a first part, the start-up behavior at a relatively low capillary number is investigated. In a second part, the behavior at near-critical conditions for breakup is observed. For both cases, viscosity ratios ranging from 0.28 to 2.2 are explored.

1. Low capillary number (Ca=0.2)

For the three studied viscosity ratios between 0.28 and 2.2 no breakup is expected to occur at an imposed Ca of 0.2, even for the most confined droplets [Vananroye et al. (2006b)]. Figure 2 shows the evolution of the dimensionless axes \( (L/2R, B/2R, \text{and} \ W/2R) \) and the orientation angle \( (\theta) \) of three droplets \( (\lambda=1) \) as a function of the dimensionless time for three degrees of confinement. The time \( t \) is made dimensionless with the characteristic emulsion time \( \tau \) of the droplet, and \( t/\tau=0 \) corresponds to the start-up of the flow. The full lines in the figure are the predictions of the confined MM model for \( \lambda =1 \), calculated from Eq. (1), using the adapted parameter values \( f'_1 \) and \( f'_2 \) [Minale
The experimental data in Fig. 2a show that confinement has little or no effect on the transient deformation during the first part of the start-up transient ($t/\tau < 3$), which agrees with the model results. For longer dimensionless times, it is observed that the deformation [Fig. 2(a)] of confined droplets ($2R/d > 0.3$) further increases whereas non-confined droplets have already reached steady state. Consequently, the steady-state deformation of confined droplets increases and is reached after a longer shearing time with respect to bulk conditions. As can be observed in Fig. 2(b), the orientation angle decreases with increasing degree of confinement, i.e., the droplet is more oriented towards the flow direction. Both the measured deformation [Fig. 2(a)] and orientation angle [Fig. 2(b)] of the least confined droplet ($2R/d = 0.27$) are in good agreement with the predictions of the confined MM model. At this degree of confinement, the confined MM model hardly differs from the original unconfined MM model. This indicates that bulk theories can still be used for $2R/d < 0.3$. For increasing degrees of confinement, the confined MM model predicts the correct trend in droplet deformation. The orientation angle is also nicely described by the model for all degrees of confinement. The microscopic images of

![FIG. 2. Dynamics of three droplets with $\lambda = 1$ and varying degrees of confinement during start-up of shear flow: comparison of experimental data with the confined MM model for Ca=0.2; (a) dimensionless axes $L/2R$, $B/2R$, $W/2R$; (b) orientation angle $\theta$. Microscopic images for $2R/d=0.60$.](image-url)
The droplet with $2R/d = 0.6$ [inset in Fig. 2(b)] show that at this specific confinement ratio, the droplet still has an ellipsoidal shape and deforms monotonically towards its steady state. The highly confined droplet on the other hand ($2R/d = 0.91$), showed a slightly sigmoidal shape upon start-up of flow (not shown here). To enable comparison with the less confined data and the confined MM model, the droplet axes were still obtained with the method described in the Sec. II, which assumes an ellipsoidal droplet shape. For this case, an absolute match between experimental results and model predictions is thus not expected. It can be seen, however, that the increasing time needed to reach steady state is nicely predicted. Whereas it was demonstrated that the breakup criterion of single droplets with $\lambda = 1$ is hardly influenced by the degree of confinement [Vananroye et al. (2006b)], the present results clearly show that confinement has a considerable effect on the transient and steady-state deformation and orientation of droplets with $\lambda = 1$.

In Fig. 3, the transient dimensions of three equally confined droplets ($2R/d = 0.73$), yet with different viscosity ratios ($\lambda = 0.28$, $\lambda = 1.2$, and $\lambda = 1.9$), are shown as a function of the dimensionless time $t/\tau$. For all viscosity ratios, the corresponding predictions of the confined MM model are added for comparison. As can be seen, the magnitude of the deformation of these droplets changes monotonically with time until steady state is reached, as is the case for nonconfined droplets at this Ca. For nonconfined droplets at Ca=0.2, the start-up deformation at viscosity ratios of 0.28, 1.2, and 1.9 are quite similar, and $L/2R$ evolves towards a value of 1.27 (not shown in Fig. 3) for all three viscosity ratios. However, as can be seen in the figure, under confinement, the droplets only behave similarly during the first part of the start-up transient. For $\lambda = 0.28$, it was shown that confinement has little effect on the steady-state deformation [Vananroye et al. (2007)]. As expected, also the transient deformation is hardly changed compared to the bulk behavior. For $\lambda \geq 1$, the experimental steady-state deformation is substantially larger than the bulk deformation [Vananroye et al. (2007)], and as expected, the start-up dynamics of confined droplets at $\lambda \geq 1$ evolve to increased values at longer time scales compared to bulk droplets. Similar to the steady-state results [Vananroye et al. (2007)], little to no difference is seen when comparing the experimental start-up dynamics for $\lambda = 1.2$ and $\lambda = 1.9$. In agreement with the data, the confined MM model predicts monotonous transients and the time scales at which steady state is reached nicely match those of the experimental results. However, the steady-state values obtained by the model slightly differ from the experimental results, especially at the lowest viscosity ratio. The discrepancies might be

**FIG. 3.** Dynamics of three droplets with $2R/d = 0.73$ and varying viscosity ratios during start-up of shear flow: comparison of experimental droplet deformation with the confined MM model for Ca=0.2.
partially due to experimental errors combined with the fact that a limited number of experimental data at high confinement ratios is used to determine the model parameters. This will not be further explored here, however, since the present work focuses on the transient results. From the results of Figs. 2 and 3, it can be concluded that after start-up of flow at \( \text{Ca}=0.2 \), confined droplets deform monotonically towards their steady-state deformation. For \( \lambda<1 \), hardly any deviations from bulk behavior are seen. For \( \lambda \geq 1 \), confined droplets elongate more and orient further towards the flow direction than non-confined droplets, and the increased steady-state deformation and orientation are reached after longer shearing times. The confined MM model is quite capable of describing the confined transient dynamics at low capillary numbers.

2. Near-critical capillary number

In bulk flow, it is known that after start-up of flow at \( \text{Ca}<\text{Ca}_{\text{crit}} \) and for viscosity ratios around unity, a droplet deforms monotonically towards its steady-state deformation even for capillary numbers close to the critical one. When \( \text{Ca} \geq \text{Ca}_{\text{crit}} \), a nonconfined droplet will continuously deform under flow until breakup is achieved [Grace (1982)]. Here, the start-up behavior of highly confined single droplets is studied at high degrees of confinement for several viscosity ratios. As an example, Fig. 4 shows microscopy images of a highly confined droplet (\( 2R/d=0.83 \)) with a viscosity ratio of 0.32 upon start-up of shear flow. It was reported that the critical capillary number for breakup at \( \lambda=0.32 \) increases with increasing degree of confinement [Vananroye et al. (2006b)]. For \( 2R/d =0.83 \), \( \text{Ca}_{\text{crit}} \) is approximately 0.7 at this viscosity ratio. Therefore, a capillary number of 0.6 is chosen to study the near-critical transient dynamics of this droplet. As can be seen
in Fig. 4(a), which are microscopy images taken in the vorticity-velocity direction, a particular behavior is recorded. After starting the flow, the droplet stretches into a fibril. When the fibril reaches its maximum elongation, some necking is seen, though, instead of breaking, the fibril partially retracts. Figure 4(b) shows an image taken in the velocity-gradient direction after $t/\tau = 88.25$. From Fig. 4(b), it can be concluded that the central part of the fibril is cylindrical and completely oriented in the flow direction. The ends of the fibril are blunt and oriented under an angle with respect to the flow direction, giving the fibril its sigmoidal shape. This shape is more prominently observed for $\lambda \leq 1$, and especially at high capillary numbers. The elongation of the droplets is rather large. Nevertheless, no breakup was observed during flow nor after stopping the flow.

Figure 5 quantitatively shows the transient deformation of the confined droplet depicted in Fig. 4. Since the droplet shape at near-critical conditions can no longer be approximated by an ellipsoid, the projections $L_p$ and $W$ directly measured in the velocity-vorticity plane (see Fig. 1), are used to express the deformation of the droplets. The time scales at which the overshoots occur in Fig. 5 are significantly larger than the ones at which steady state is reached at lower capillary numbers (see Fig. 3). Even after a dimensionless time of 150 (approximately 2000 s), steady state has not yet been reached. The corresponding predictions of the confined MM model at $Ca=0.6$ and $2R/d=0.83$ (full lines) are also added to the graph. As can be seen, the model predicts a continuously increasing deformation and thus breakup at $Ca=0.6$. Therefore, an additional comparison is made with the confined MM model at a capillary number of 0.5, which is slightly below the critical capillary number as predicted by the confined MM model ($Ca_{crit} = 0.52$ at $2R/d = 0.83$ and $\lambda = 0.32$). The predictions at $Ca=0.5$ (dashed-dotted lines) show a monotonic deformation towards the steady-state shape which is reached after approximately 100 dimensionless time units. Hence, the model is not capable of predicting the overshoots at near-critical conditions. This was expected since it assumes an ellipsoidal droplet shape, which is clearly not the case anymore.

Figure 6 shows the transient deformation of a highly confined droplet with $\lambda = 1$. Since the critical capillary number for breakup at a viscosity ratio of unity is not affected by the degree of confinement [Vananroye et al. (2006b)], the droplet is expected to breakup at $Ca=0.48$ [Grace (1982), de Bruijn (1989)]. Therefore, a capillary number of 0.43 was chosen to study the near-critical transient dynamics of this droplet. In Fig. 6, the predic-
tions of the confined MM model for \( L_p/2R \) and \( W/2R \) at \( 2R/d=0 \) (dashed-dotted lines), which represents the bulk situation, and at \( 2R/d=0.83 \) (full lines) are also added. The experiments show that around \( t/\tau=60 \), a maximum elongation is reached from which the droplet retracts slowly until steady state is reached. The steady-state value of \( L_p \) is two times larger than the bulk model prediction and it takes about ten times longer to reach it as compared to the unconfined case. The confined MM model at \( 2R/d=0.83 \) again predicts a monotonic evolution of \( L_p \) towards the steady-state deformation. The steady-state value for \( L_p \) overestimates the experimental results, though, compared with the unconfined prediction, the confined prediction is rather good, especially since the shape of this droplet is sigmoidal instead of ellipsoidal. Similar experiments for a viscosity ratio of 1 were performed by Sibillo et al. (2006). They showed the transient length \( L/2R \) of a droplet with a degree of confinement of 1 at \( Ca=0.4 \). Under these conditions, they observed a damped oscillation in droplet deformation with a first maximum \( L/2R=4.5 \) at a dimensionless time of 25. The steady-state deformation was reached after a dimensionless time of 140 \( L/2R=3.5 \). The experiments presented in Fig. 6, which are run at a lower confinement ratio yet at a slightly higher capillary number, are in agreement with the results of Sibillo et al. (2006).

In Fig. 7, the experimental data at near-critical conditions for \( \lambda=2.2 \) and \( 2R/d=0.82 \) are compared with the results of the confined MM model. At a viscosity ratio of 2.2, bulk breakup is seen around a capillary number of 0.66 [Grace (1982); de Bruijn (1989)], whereas for \( 2R/d=0.82 \) breakup is expected around \( Ca=0.45 \) [Vananroye et al. (2006b)]. Therefore, similar to \( \lambda=1 \), also a capillary number of 0.43 is chosen to investigate the droplet behavior at near-critical conditions for \( \lambda=2.2 \). Although the flow conditions are similar, in the case of \( \lambda=2.2 \) the overshoot in deformation is less pronounced than for \( \lambda=1 \) (see Fig. 6). From this, it could be stated that the presence of one or more overshoots at near-critical capillary numbers depends on the viscosity ratio of the system. With increasing viscosity ratio, a transition from damped oscillations towards a single overshoot which becomes less pronounced at higher viscosity ratios, is seen. It is expected that the oscillatory effect will completely disappear above a certain viscosity ratio. As can be observed, the confined MM model at \( 2R/d=0 \) significantly underestimates the experimental deformation at \( 2R/d=0.82 \) and is therefore not capable of predicting con-

**FIG. 6.** Confined dynamics of a droplet with \( 2R/d=0.83 \) and \( \lambda=1 \) during start-up of shear flow: comparison of experimental data with the confined MM model for \( Ca=0.43 \).
fined near-critical dynamics at \( \lambda = 2.2 \). The confined MM model at \( 2R/d = 0.82 \) largely overestimates the experimental results. According to this model, breakup is expected at \( \text{Ca} = 0.44 \) for \( 2R/d = 0.82 \) and \( \lambda = 2.2 \), so at \( \text{Ca} = 0.43 \), the model predictions are just below critical. As a result, the model predicts an extended ellipsoidal droplet shape and a long time before steady state is reached.

### B. Relaxation after shear flow

In a second series of experiments, the relaxation of single droplets after a steady-state shear flow is studied. Similar to the start-up experiments, droplets with varying degrees of confinement and viscosity ratios are investigated. Flow conditions include a low capillary number \( \text{Ca} = 0.2 \) as well as a somewhat higher capillary number \( \text{Ca} = 0.3 \).

1. **Low capillary number (Ca=0.2)**

In Fig. 8, the shape relaxation after shear flow is shown as a function of dimensionless time for \( \lambda = 1.1 \). The relaxation behavior is expressed by means of the dimensionless parameter \( (L_p/2R-1) \), which can be obtained from images taken in the vorticity-velocity plane. The parameter is normalized for each droplet by dividing it by its value at \( t/\tau = 0 \), which is the instant the shear flow is stopped. For a relatively low capillary number of 0.2, it was observed that only highly confined droplets display a sigmoidal shape, whereas droplets at a lower degree of confinement still have an ellipsoidal shape, though with an increased deformation compared to bulk conditions [Vananroye et al. (2008)]. This shape difference in combination with the proximity of the walls could cause a difference in retraction. As can be seen in Fig. 8, the relaxation behavior of all droplets is quasi similar. The experimental observations for the droplets with \( 2R/d = 0.27 \) and \( 2R/d = 0.45 \) nicely coincide over the entire relaxation. Hence, it can be concluded that in these cases bulk behavior still prevails. The droplet with a confinement ratio \( 2R/d = 0.73 \) seems to retract slightly slower. Nevertheless, the relaxation parameter changes as an exponential decay function of time with a single relaxation time for all confinement ratios. It should be reminded that effects of confinement during start-up of flow become already visible at a degree of confinement of 0.3 for \( \lambda = 1 \). In the case of droplet relaxation, however, it can be stated that effects of confinement on the retraction process are...
postponed to higher degrees of confinement with respect to the start-up experiments. This is probably due to the absence of the external flow field during relaxation. These results are in line with recent data on droplets in more concentrated blends at a viscosity ratio of 0.5, where at an average degree of confinement of 0.5, no confinement effects on the relaxation behavior were observed [Vananroye et al. (2006a)]. When comparing the experimental results with the predictions of the confined MM model, it can be concluded that at a capillary number of 0.2 and a viscosity ratio of 1.1, the model is appreciatively capable of predicting the effect of confinement on the relaxation process. The model predictions for the unbounded case ($2R/d=0$) nearly coincide with the results at $2R/d=0.27$ and are omitted for the sake of brevity.

The same type of experiments is conducted for different viscosity ratios. Figures 9 and 10 show the dimensionless relaxation parameters after cessation of shear flow at $Ca=0.2$ for $\lambda=0.33$ and $\lambda=2.2$ at different confinement ratios. As shown by the experimental results in Fig. 9, in the case of $\lambda=0.33$, the effect of confinement on the relaxation of

![FIG. 8. Relaxation of three droplets with varying degrees of confinement for $Ca=0.2$ and $\lambda=1.1$: comparison of experimental data with the confined MM model.](image)

![FIG. 9. Relaxation of four droplets with varying degrees of confinement for $Ca=0.2$ and $\lambda=0.33$: comparison of experimental data with the confined MM model.](image)
droplets with $2R/d<0.6$ is within experimental error. The relaxation data of the three droplets with the lowest degrees of confinement all coincide. The highly confined droplet clearly relaxes slower than the less confined ones. Therefore, it can be concluded that for a viscosity ratio of 0.33, confinement has a clear effect on the relaxation process of single droplets after shear flow at $Ca=0.2$, though only at confinement degrees greater than 0.55. Good agreement is seen between the experimental results and the predictions of the confined MM model, especially at a high confinement ratio of 0.88. However, the model already predicts substantial confinement effects at $2R/d=0.54$, which is not confirmed by the data. For a viscosity ratio of 2.2, as is the case in Fig. 10, again clear differences between the more confined and less confined cases are observed. The droplets with degrees of confinement of 0.66 and 0.91 clearly relax slower than the less confined droplet. Similar to the results at lower viscosity ratios, for both unconfined and substantially confined droplets, experimental and model results are in good agreement.

2. High capillary number (Ca=0.3)

Next, the relaxation of confined droplets is studied after a shear flow at a higher capillary number of 0.3 to further investigate the effects of a larger initial deformation. In Fig. 11, the dimensionless droplet relaxation parameter $(L_p/2R-1)$ of three retracting droplets with a viscosity ratio of 1.1 is shown as a function of the dimensionless time after a steady-state shear flow at $Ca=0.3$. As can be seen on the figure, the two droplets with the lowest degree of confinement still experience a similar relaxation behavior independent of their degree of confinement. However, it is observed that the highest confined droplet ($2R/d=0.73$) displays a slower relaxation as was also the case at a capillary number of 0.2. Again, the confined MM model nicely predicts the experimental results.

Also experiments at other viscosity ratios are performed. In Fig. 12, the relaxation results are shown for droplets with a viscosity ratio $\lambda$ of 0.33. It is again observed that for the three least confined droplets, the shape relaxation is hardly affected. However, for the highly confined droplet ($2R/d=0.88$), again a slower relaxation is present. When comparing the experimental data of Figs. 9 and 12, no additional effect of increasing the capillary number is visible. Reasonable predictions are made by the confined MM model. In Fig. 13, the relaxation of two droplets with a viscosity ratio of 2.2 is shown at a
capillary number of 0.3. One droplet has a low degree of confinement \((2R/d=0.20)\) where the other one has a more moderate degree of confinement \((2R/d=0.66)\). Similar trends as at \(Ca=0.2\) are seen. Again, the model nicely covers the data for both situations.

All relaxation results after shearing at capillary numbers of 0.2 and 0.3 indicate that the effect of confinement on the retraction of droplets is not as great as its effect during flow. For all viscosity ratios, only for highly confined, highly elongated droplets retraction occurs significantly slower than in bulk flow. From these experiments, one can estimate that the critical confinement ratio for droplet relaxation is situated around a value of 0.55. For all viscosity ratios, the results of the confined MM model are in rather good agreement with the experimental data at both capillary numbers, especially at high confinement ratios.

**FIG. 11.** Relaxation of three droplets with varying degrees of confinement for \(Ca=0.3\) and \(\lambda=1.1\): comparison of experimental data with the confined MM model.

**FIG. 12.** Relaxation of four droplets with varying degrees of confinement for \(Ca=0.3\) and \(\lambda=0.33\): comparison of experimental data with the confined MM model.
IV. CONCLUSIONS

Confined start-up and relaxation dynamics of Newtonian droplets dispersed in a Newtonian matrix subjected to a shear flow are studied in the parameter space of capillary number and viscosity ratio. The experiments are performed in a counterrotating parallel plate cell where single PDMS droplets with specific sizes and viscosities were injected in a PIB matrix confined between the two plates. The experimental results during start-up are compared with the predictions of an adapted version of the Maffettone and Minale model that includes confinement effects. For $\lambda_1$, the effect of confinement on droplet deformation during start-up at relatively low capillary numbers is rather small. For $\lambda_1 \gg 1$, an increased deformation and an increased orientation towards the flow direction is seen for $2R/d > 0.3$. In addition, the steady-state regime is reached after a longer time period with respect to the unconfined case. At relatively low $Ca$, the confined MM model is in good agreement with the experimental results. Both the deformation and orientation angle of confined and unconfined droplets are nicely predicted. At considerably high confinement ratios, overshoots in the droplet deformation during start-up are observed at near-critical conditions. With increasing viscosity ratio, a transition from damped oscillations towards a single overshoot, which becomes less pronounced at higher viscosity ratios, is seen. Under these conditions, the confined MM model becomes less meaningful since sigmoidal droplet shapes are obtained. It is demonstrated that the experimentally observed overshoots are indeed not recovered by the model. The relaxation behavior of confined droplets is less sensitive to the confinement ratio than the start-up transient. The relaxation of droplets up to a moderate degree of confinement ($2R/d < 0.55$) is hardly affected by the presence of the walls. For all viscosity ratios, it was seen that only highly confined droplets ($2R/d > 0.55$), which have greater initial deformations relax slower compared to unconfined droplets. Nevertheless, at high confinement ratios, the relaxation parameter still evolves as an exponential decay function of time with a single relaxation time. Especially at high confinement degrees, the confined MM model predictions for relaxation are appreciative.

FIG. 13. Relaxation of two droplets with varying degrees of confinement for $Ca=0.3$ and $\lambda=2.2$: comparison of experimental data with the confined MM model.
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