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The effect of geometrical confinement on coalescence efficiency of droplet pairs in shear flow

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Abstract

Droplet coalescence is determined by the combined effect of the collision frequency and the coalescence efficiency of colliding droplets. In the present work, the effect of geometrical confinement on coalescence efficiency in shear flow is experimentally investigated by means of a counter rotating parallel plate device, equipped with a microscope. The model system consisted of Newtonian droplets in a Newtonian matrix. The ratio of droplet diameter to plate spacing ($2R/H$) is varied between 0.06 and 0.42, thus covering bulk as well as confined conditions. Droplet interactions are investigated for the complete range of offsets between the droplet centers in the velocity gradient direction. It is observed that due to confinement coalescence is possible up to higher initial offsets. On the other hand, confinement also induces a lower boundary for the initial offset, below which the droplets reverse during their interaction, thus rendering coalescence impossible. Numerical simulations in 2D show that the latter phenomenon is caused by recirculation flows at the front and rear of confined droplet pairs. The lower boundary is independent of Ca , but increases with increasing confinement ratio $2R/H$ and droplet size. The overall coalescence efficiency is significantly larger in confined conditions as compared to bulk conditions.

Keywords

Coalescence
Droplet
Confinement
Shear flow
Blends
Coalescence efficiency

1. Introduction

Multiphasic materials consisting of liquid droplets dispersed in a liquid matrix can be encountered in a diverse range of products and applications such as foods, plastics, pharmaceuticals, oil recovery, etc. The properties of these emulsions not only depend on the characteristics of the components and the concentration, but also on the droplet size and its distribution. In general, the latter results from a dynamic equilibrium between droplet breakup and coalescence. To gain fundamental insight in the effects of parameters such as flow intensity, flow type, material parameters and droplet size on these dynamic processes, model flows, such as shear flow, are frequently used [1,2]. In addition breakup and coalescence are often studied separately [1,3,4]. The relations and models derived from such investigations can in principle be combined to describe and predict the evolution of the droplet size in complex flows such as during processing [3–5].

Per definition, droplet coalescence requires the interaction of at least two droplets. Because not every collision leads to coalescence, the coalescence frequency in emulsions is determined by the product of the collision frequency of sheared droplets and the coalescence efficiency of colliding droplets [8]. An equation for the collision frequency of equal-sized spherical droplets in simple shear flow was first derived by Smoluchowski [9], assuming that the streamlines are not affected by the presence of the droplets. In this case, all droplets in different shear planes for which the distance between their undisturbed trajectories in the vorticity or in the velocity gradient direction is less than the sum of the droplet radii, will collide. The coalescence efficiency provides the percentage of these droplets that will coalesce [8]. The coalescence process is typically described as a three-stage process [10]. First, the droplets approach each other during flow starting from a large separation distance. Once the droplets have collided, the film of matrix fluid between them starts to thin. This second stage is the film drainage. As the film thickness decreases, the distance between the two droplet surfaces can become small enough for the Van der Waals forces to destabilize the film, leading to film rupture, which occurs almost instantaneously.

Both during the approach and drainage, the droplet behavior can deviate from that of non-interacting spheres, which reduces the coalescence efficiency. First, due to hydrodynamic interactions, the droplet trajectories will deviate from the undisturbed ones causing only a fraction of the droplets for which the distance between their undisturbed trajectories in the vorticity or in the velocity gradient direction is less than the sum of the droplet radii to collide. By using trajectory analysis, Wang et al. [11] theoretically calculated the collision efficiency for spherical droplets in simple linear flows, which is the percentage of droplets that would still collide taking into account the hydrodynamic interactions between the two droplets. This trajectory analysis assumes that the droplets are nondeformable. In reality, the contact area between the droplets will flatten when the pressure in the gap between the droplet interfaces exceeds the Laplace pressure. This flattening will hinder film drainage [10]. In addition, droplets can deform due to the external flow field [2]. To model the drainage process of flattened droplets, hydrodynamic scaling theories for the film drainage of flat interfaces are often used [10,12]. The drainage rate largely depends on the interfacial mobility, which defines the boundary conditions for the flow in the film [10]. Based on these drainage models, the coalescence efficiency can be determined by comparing the time needed for drainage with that available during droplet collision [10,13]. A comparison of the coalescence efficiencies determined with the different drainage models and the trajectory analysis can be found in the work of Mousa et al. [14]. Finally, by combining the trajectory analysis with droplet deformability, more accurate expressions for the coalescence efficiency of unconfined droplets can be obtained [15].

Several authors experimentally determined the coalescence efficiency by performing systematic studies on isolated droplet pairs. For glancing collisions, the droplets only coalesce during a gentle collision [16]. Typically the flow intensity is expressed by the capillary number Ca ($= \eta_m \cdot \dot{\gamma} \cdot R / \Gamma$, with η_m the matrix viscosity, $\dot{\gamma}$ the shear rate, R the droplet radius and Γ the interfacial tension). Hence, a critical Ca , below which coalescence occurs, can be defined [17]. In linear flows, this critical Ca decreases with increasing droplet diameter [17,18]. These observations

are in agreement with the predictions of the drainage models [10,14,15]. Based on these drainage theories, Jaeger et al. [13] also predicted that coalescence will only occur when the initial offset in the velocity gradient direction between the centers of mass of the droplets is smaller than a critical offset. Taking into account the fact that the local collision frequency increases linearly with the offset, and assuming that the offset in the vorticity direction is zero, the coalescence efficiency can be expressed as the dimensionless offset squared [13]. By systematically varying the offset between the droplets, Mousa et al. [19] investigated the effects of droplet size, matrix viscosity and ratio of the droplet radii on the coalescence efficiency in shear flow. Leal and coworkers determined the critical offset as a function of Ca for different droplet sizes and viscosity ratios in mixed linear flows with different values of the flow-type parameter [17]. For systems with a low viscosity ratio, the critical Ca decreases monotonically with increasing offset [17,20]. Yang et al. [20] constructed a master curve of critical Ca versus offset for various droplet sizes by scaling the critical Ca with $R^{0.84}$. However, when the viscosity ratio is larger than 0.1, an increase of the critical Ca occurs at larger offsets [17,21]. The latter is caused by coalescence in the extensional quadrant, which is the region in which the droplets are pulled apart by the external flow.

The results on coalescence efficiency for droplet pairs proved valuable for the prediction of the coalescence kinetics in concentrated blends and emulsions. Vinckier et al. [22] modeled their experimental data on coalescence kinetics in polymer blends with dispersed phase volume fractions up to 20% with the drainage theory for partially mobile interfaces. However, good agreement with the theoretical predictions was only obtained after introducing an adjustable critical film thickness. Lyu et al. [23] used both the trajectory theory and the drainage model for partially mobile interfaces, but also needed an adjustable coalescence rate parameter to be able to describe their experimental data. Burkhart et al. [24] showed that the theory of Rother and Davis [15] substantially improves the predictions of the coalescence kinetics as compared to the trajectory analysis. Due to continuous improvements in algorithms and computational power, numerical studies of droplet coalescence and film drainage are appearing [25–27]. By using a boundary-integral method, Yoon et al. [27] were able

to qualitatively match their experimental data for the critical Ca -numbers and critical offsets for systems with a low viscosity ratio. Nevertheless, there is still no consensus in literature on how to predict a realistic coalescence rate for a given flow type, capillary number and viscosity ratio [25].

The use of microfluidic devices is emerging in a variety of applications [28–31]. In many cases multiphase fluids have to be transported through these devices [31,32]. In multiphase microfluidic applications the size of the droplets can become comparable to the dimensions of the channel. Hence, deviations from bulk behavior can be expected [33]. To understand the underlying physics, morphology development in simple confined geometries, such as channel flow [34] or shear flow [33] has been studied as a model type problem. In shear flow the effect of confinement on droplet deformation [35–37] and droplet breakup [38] is already well documented. Recently, a first experimental study on the effects of confinement on the coalescence of two equal-sized droplets in pure shear flow was performed for droplet-matrix systems with a viscosity ratio of 1 [39]. For confinement ratios (ratio of droplet diameter to gap spacing) larger than 0.2 and an intermediate value of the offset, the critical Ca -number up to which coalescence occurs, increases with confinement. Thus, confinement can clearly promote coalescence. However, this study was limited to a single initial relative position of the droplets. Hence, the coalescence efficiency could not be determined from these results. In the present study, the effect of confinement on droplet coalescence is systematically investigated for a wide range of initial offsets between the droplet centers in the velocity gradient direction. Based on this experimental study, the effect of geometrical confinement on the coalescence efficiency in shear flow is determined.

2. Materials & Methods

2.1. Materials

Polydimethylsiloxane (PDMS Rhodorsil V1000 from Rhodia) with a viscosity η_d of 0.95 Pa·s at the experimental temperature of 27 °C was used as the droplet phase. Polyisobutylene (PIB Glissopal V-190 from BASF) with a viscosity η_m of 10.5 Pa·s at 27 °C was used as the matrix phase; the resulting

viscosity ratio λ being 0.095. In the shear rate range of the experiments both materials are Newtonian and there is no measurable elasticity. Both materials are transparent and their refractive indices are substantially different, resulting in good optical contrast. It has been shown that PIB is slightly soluble in PDMS, whereas the solubility of PDMS in PIB is negligible [40]. While using PDMS as droplet phase, no change in droplet size was observed. Consequently, the system can be considered to be completely immiscible. The interfacial tension Γ was determined by fitting deformation data of a single droplet at low capillary numbers with the small deformation theory of Greco [41], which resulted in a value for the interfacial tension of 1.8 mN/m. The density difference between PDMS and PIB is small (0.08 g.cm^{-3}) [42]. Hence buoyancy effects are expected to be negligible. The temperature of the sample was monitored with a thermocouple. For all experiments the temperature was kept constant at 27 °C by controlling the room temperature.

2.2. Experimental setup and protocol

For studying interacting droplets, the relative position of the droplets with respect to each other is essential. Fig. 1 shows a schematic of two colliding droplets, which defines the parameters that characterize the droplet positions. The x-direction is the velocity direction, the y-direction is the velocity gradient direction. The relative trajectory of the two droplets will be described by $\Delta X = (X_2 - X_1)$ and $\Delta Y = (Y_2 - Y_1)$, with (X_1, Y_1) and (X_2, Y_2) the coordinates of the droplet centers. $\Delta X/2R$ is the dimensionless distance between the two droplets in the velocity direction. $\Delta Y/2R$ is the dimensionless distance in the velocity gradient direction, also referred to as the offset. The orientation angle θ is the angle between the velocity direction and the line connecting the droplet centers. The distance between the centers of mass of the two droplets will be referred to as d .

Fig. 1. Schematical representation of two colliding droplets in shear flow.

A counter rotating parallel plate shear flow device (based on an Anton Paar MCR 300 rheometer) is used for the coalescence experiments. A detailed description of this device is given elsewhere [38,43]. In brief, the device consists of two parallel quartz plates with a diameter of 50 mm. The two plates can rotate in opposite direction, thus creating a stagnation plane in between the plates. Droplets in this stagnation plane can easily be visualized with a non-moving microscope setup. The optical train consists of a stereo microscope (Wild M5A) in combination with a digital camera (Basler A631fc). The resulting resolution is 1.34 pixels/ μm . Images of the interacting droplet pair are taken in the velocity-velocity gradient plane using Streampix Digital Video Recording Software (Norpix) and are analyzed using ImageJ Software. Images taken in the velocity-velocity gradient plane are slightly elongated in the velocity direction due to the curvature of the cup surrounding the bottom plate (see schematic of device in Vananroye et al. [38]) and need to be rescaled in the velocity direction before image analysis. The edges of the droplets are detected with a Laplace operator. When the two droplets are in apparent contact, the images of both droplets should be separated from each other to enable the fitting of an ellipse on the droplet contours of both droplets. For this separation the watershed algorithm, which generates a white line between the cusps on both sides of the doublet, is used.

For each experiment, a PDMS droplet of the desired size is injected in the PIB matrix by means of a homemade injection device. A strong shear flow is then applied to break up this droplet into two daughter droplets with a difference in size of less than a few percent. Subsequently, a low shear rate is applied in the other direction. As a result, the two droplets approach each other and start rotating over each other. When during the rotation the desired vertical offset is reached, the shear rate is reversed again until the desired distance in the flow direction is obtained. With this tedious procedure both the initial vertical offset and initial distance in the velocity direction could be controlled. After relaxation of the droplets to their spherical shape, the coalescence experiment can begin. To verify that the droplets are in the middle of the gap during the complete experiment, the rotational velocities of the top and bottom plate were monitored, while keeping the droplet pair in the stagnation plane.

In this work, a gap spacing H of 3 mm is used to represent bulk conditions and a gap spacing H of 0.5 mm is used to represent confined conditions; droplet sizes of 165 μm and 210 μm are used throughout. For the droplet diameter of 165 μm , this results in a confinement ratio $2R/H$ of 0.055 for the bulk conditions and a confinement ratio $2R/H$ of 0.33 for the confined conditions. For the droplet diameter of 210 μm , this results in a confinement ratio $2R/H$ of 0.07 for the bulk conditions and a confinement ratio $2R/H$ of 0.42 for the confined conditions. In this study the effects of varying the initial dimensionless offset $\Delta Y_{in}/2R$, the capillary number, the droplet size and the confinement ratio ($2R/H$) are investigated. The initial dimensionless distance in the velocity direction $\Delta X_{in}/2R$ was fixed at 1.6 for all experiments. This parameter space allows for the unambiguous assessment of the effects of confinement on the coalescence efficiency of sheared droplets. A low value of the Re number (order 10^{-7}) and a high value of the Pe number (order 10^7) indicate that inertia and Brownian motion can be neglected.

2.3. Numerical simulations in 2D

Numerical simulations of two interacting droplets in shear flow are performed in 2D. The computational domain is a rectangular channel, with the upper and lower wall moving in counter directions with a velocity U , resulting in a shear rate $\dot{\gamma} = 2U/H$. The gap height H is taken either 3 mm or 0.5 mm. The length of the channel is chosen to be at least 30 times the droplet radius and it has been verified that this length does not affect the results. The droplets are initially spherical and at the inlet and outlet of the channel a linear velocity profile, which is characteristic for shear flow between two parallel plates, was imposed as the boundary condition. The liquids are density-matched and incompressible and their viscosities are taken equal to the experimental values. The interfacial tension equals the experimental value and the conditions at the droplet interfaces are continuity of the velocity (u) across the interface and a balance between interfacial tension and normal forces at the interface:

$$u_m = u_d \quad (1)$$

$$n \cdot (T_m - T_d) = \Gamma(\nabla \cdot n)n \quad (2)$$

where n is the normal vector at the interface and T is the total stress in each fluid:

$$T = -pI + \eta(\nabla u + (\nabla u)^T) \quad (3)$$

The governing equations that are solved in each fluid domain are the continuity equation and the momentum balance:

$$\nabla \cdot u = 0 \quad (4)$$

$$\rho \left(\frac{\partial u}{\partial t} + u \cdot \nabla u \right) = \nabla \cdot (-pI + \eta(\nabla u + (\nabla u)^T)) \quad (5)$$

These equations are discretized by means of a finite element method in the package COMSOL Multiphysics. A triangular mesh with an extra fine mesh distribution along the droplet interfaces and a boundary layer of quadrilateral mesh elements at the moving walls is used. It has been verified that either a further refinement of either the overall size of the mesh elements or an increase of the number of mesh elements along the droplet interface, does not significantly affect the results. Because in this approach the mesh is fixed to the interface and moves with it, the simulations can

handle deformations but no topological changes of the interface, such as coalescence. Hence, in the present work, the simulations will only be used to provide streamlines and droplet trajectories. The initial dimensionless distance between the droplets in the velocity direction $\Delta X_m/2R$ is taken to be 1.2 in all simulations. The lower initial dimensionless distance between the droplets as compared to the experimental one is selected due to practical limitations of long-time simulations of shear flow between parallel plates by means of the moving mesh method. It has been verified that this shorter initial distance between the droplets in the velocity direction does not affect the droplet trajectories and streamlines around an interacting droplet pair. The initial dimensionless offset $\Delta Y_m/2R$ has been varied over a wide range to cover the different qualitative behaviors of interacting droplet pairs in shear flow.

3. Results & Discussion

3.1. Reversing trajectories for confined droplets with a small initial offset

For intermediate values of the offset, it was shown before that geometrical confinement can substantially increase the critical Ca and hence promote droplet coalescence [39]. However, droplets in emulsions and polymer blends generally have random positions with respect to each other. Consequently, the overall effect of geometrical confinement on coalescence efficiency can only be assessed by studying its effects for a wide range of initial offsets between the droplets. Such a study is performed in the present work for droplets with a diameter of 165 μm and 210 μm respectively.

Fig. 2 displays typical time sequences of the interaction of two droplets with a diameter $2R = 165 \mu\text{m}$ at a Ca of 0.006. The left column shows the two possible outcomes of a collision experiment in bulk conditions. For an initial offset $\Delta Y_m/2R = 0.144$ (Fig. 2a-e) the two droplets approach each other and come into apparent contact. Subsequently the two droplets form a droplet pair, or doublet, that starts rotating in the shear flow. During the rotation of the droplets, the matrix film between the droplet interfaces is being drained. When the matrix film is sufficiently thinned, Van der Waals forces cause the film to rupture, resulting in droplet coalescence. On the other hand, for an

initial offset $\Delta Y_{in}/2R = 0.327$ (Fig. 2f-j) the two droplets rotate over each other without coalescing.

This indicates that there is a critical initial offset above which coalescence is not possible. These results are in line with literature data for mixed linear flows [20,21] and shear flow [19] in bulk conditions.

Fig. 2. Time sequence of the interaction of two droplets, $\Delta Y_{in}/2R = 1.6$, $2R = 165 \mu\text{m}$, $Ca = 0.006$.
 Left column: $H = 3 \text{ mm}$, $2R/H = 0.055$; (a-e) $\Delta Y_{in}/2R = 0.144$; (f-j) $\Delta Y_{in}/2R = 0.327$.
 Right column: $H = 0.5 \text{ mm}$, $2R/H = 0.33$; (a'-e') $\Delta Y_{in}/2R = 0.182$; (f'-j') $\Delta Y_{in}/2R = 0.329$; (k'-o') $\Delta Y_{in}/2R = 0.437$.
 Original images without rescaling of the x-axis.

The right column shows the possible outcomes of a collision experiment in confined conditions. In contrast to bulk conditions, for which there are two possible scenarios, in confined conditions three different behaviors were observed. When the initial offset is large, for example $\Delta Y_{in}/2R = 0.437$ (Fig. 2k'-o'), the two droplets rotate over each other without coalescing, because there is not enough time for the matrix film to become sufficiently thin. When the initial offset is smaller than a certain upper critical offset, for example $\Delta Y_{in}/2R = 0.329$ (Fig. 2f'-j'), the two droplets approach each other, start rotating over each other and when the matrix film has sufficiently thinned, coalesce. These two scenarios are the same as in bulk conditions. However, if the initial offset is decreased even further, a different sequence of droplet positions is observed. For example, for an initial offset $\Delta Y_{in}/2R = 0.182$ (Fig. 2a'-e'), it can be seen that during the approach the initial

offset decreases until it becomes zero and the droplets are located next to each other at exactly the same height. Subsequently, rather than rotating over each other, the direction of movement reverses and the droplets separate again. As a consequence, for confined conditions there is not only an upper critical initial offset, above which the droplets do not coalesce, but there is also a lower critical initial offset, below which there is no coalescence. Reversing trajectories have been observed before in literature for a pair of equally charged droplets in shear flow [16] and for a pair of solid particles in shear flow that interact in the presence of inertia [44]. In this study the droplets are however not charged and effects of inertia can be excluded due to the low Re number. Zurita-Gotor et al. [45] investigated numerically the effect of wall proximity on the trajectories of solid particles and also found reversing trajectories. The effects of confinement ratio, droplet size and flow intensity on this lower critical initial offset will be investigated in detail in section 3.2.

To gain insight in the effect of confinement on the critical offsets for coalescence, the droplet trajectories are investigated for droplet pairs with a range of initial offsets. Fig. 3 shows the evolution of the vertical offset $\Delta Y/2R$ as a function of the horizontal distance between the droplets $\Delta X/2R$ for different initial offsets. Fig. 3a shows the trajectories for the unconfined case at a gap of 3 mm. It is clear from Fig. 3a that during approach of the droplets, the offset slightly increases due to hydrodynamic interactions between the droplets. After the droplets make apparent contact ($d/2R = 1$), they start rotating over each other, resulting in a swift increase of the offset. When the initial offset is small enough, the droplets coalesce. When the initial offset is too large, there is not enough time for the film to drain and the droplets rotate over each other and separate again. It can be seen in Fig. 3a that at an initial offset $\Delta Y_{in}/2R = 0.239$, coalescence is not possible anymore and the droplets rotate over each other.

Fig. 3. Evolution of the vertical offset $\Delta Y/2R$ as a function of position ($\Delta X/2R$) for different initial offsets, $\Delta X_0/2R = 1.6$, $2R = 165 \mu\text{m}$, $Ca = 0.008$. (a) $H = 3$ mm, (b) $H = 0.5$ mm.

Fig. 3b displays the trajectories for the confined case at a gap of 0.5 mm. The trajectories are clearly different as compared to the unconfined case in Fig. 3a: during approach of the droplets, rather than increasing, the offset decreases. Moreover, when the initial offset is very small, it decreases to zero before the droplets can make apparent contact and the direction of movement reverses after which the droplets separate again. It can be seen from Fig. 3b that if the initial offset is smaller than 0.179, these reversing trajectories are present. As in bulk conditions, when the initial offset exceeds a critical value, the droplets rotate over each other without coalescence. It is clear that the critical initial offset, above which no coalescence is possible, is larger in the confined case ($\Delta Y_{\text{crit}}/2R \approx 0.312$) as compared to the bulk case ($\Delta Y_{\text{crit}}/2R \approx 0.239$). A coalescence diagram, mapping out the critical offset boundaries as a function of Ca , $2R$ and $2R/H$ will be discussed in section 3.2.

The trajectory $\Delta Y/2R - \Delta X/2R$ that represents the rotation of a pair of hard spheres has been added to Fig. 3. Since hard spheres do not deform during their interaction, the distance between the centers of the spheres remains constant during the rotation of the doublet. Hence, the trajectory obeys the equation:

$$1 = \sqrt{x^2 + y^2} \quad (6)$$

It is clear from Fig. 3 that, under the present conditions, both the confined and unconfined rotating droplet pairs approximately follow the trajectory of a pair of hard spheres. As a matter of fact, the

observed center-to-center distances are slightly lower than $2R$ because the droplets undergo a small deformation when they are pushed together due to the external flow. Chen et al. [39] studied the deformation of interacting droplets in bulk and confined shear flow and concluded that in the gentle flows that are required for droplet coalescence, the effects of confinement on the droplet deformation are negligible.

The decrease in offset during approach of the droplets in a confined shear flow can be understood by simulating the flow field around the two droplets in shear flow. Fig. 4 shows the streamlines around two colliding droplets, generated with a 2-dimensional simulation of a droplet pair in shear flow. Because the moving mesh method can not handle topological changes of the interface, the coalescence event itself can not be simulated this way. Hence, the aim of the simulations is not to reproduce the experimental results but rather to provide insight in the origin of the observed phenomena. Fig. 4a depicts the streamlines for the unconfined case at a gap of 3 mm and Fig. 4b for the confined case at a gap of 0.5 mm. A clear qualitative difference can be seen. In the unconfined case all the streamlines going to the right, pass above the two droplets and all the streamlines going to the left, pass underneath the two droplets. On the other hand, in the confined case, there is a large recirculation zone at the front and rear of the droplet doublet. Streamlines which start approximately at the height of the droplet pair, do not pass over the droplet pair, but turn before reaching it. As a consequence of the symmetry, the streamlines coming from the left and right show the same behavior. Similar large recirculation zones at the front and rear of a droplet were already reported from 3D numerical simulations of the flow field around a single highly confined droplet [46].

Fig. 4. Simulation in 2D of the streamlines around two colliding droplets. $2R = 165 \mu\text{m}$, $Ca = 0.008$, $\theta = 45^\circ$. (a) $H = 3 \text{ mm}$, (b) $H = 0.5 \text{ mm}$.

This large recirculation zone, present in confined conditions, causes the offset to decrease during approach of the droplets, as can be seen in Fig. 3b. Whereas in bulk conditions all streamlines pull the upper left droplet over the lower right droplet, in confined conditions there is a competition between streamlines pulling the upper left droplet over the lower right droplet and streamlines of the recirculation zone pulling the upper left droplet down. In addition, when the initial offset is small enough, the reversing streamlines cause the offset to decrease to zero, subsequently leading to a reversal of the direction of movement and the separation of the droplets.

3.2. Initial offset boundaries for coalescence

For the determination of the coalescence efficiency and hence the final morphology of a polymer blend, it needs to be assessed which fraction of the colliding droplets will coalesce. The coalescence efficiency is determined by the critical offset boundaries for coalescence [13]. Fig. 5 depicts the upper and lower initial offset boundaries for coalescence as a function of the Ca for droplet sizes of $2R = 165 \mu\text{m}$ and $2R = 210 \mu\text{m}$. Fig. 5a gives the initial offset boundaries at a gap of $H = 3 \text{ mm}$, representing bulk conditions, whereas Fig. 5b gives the initial offset boundaries at a gap of $H = 0.5 \text{ mm}$, representing confined conditions.

Fig. 5. Initial dimensionless offset boundaries as a function of Ca number, $\Delta Y_m/2R = 1.6$. (a) $H = 3$ mm, (b) $H = 0.5$ mm. For the upper boundaries $\blacktriangle$ and $\triangle$ indicate the highest initial offset for which coalescence occurs, while ∇ and $\triangledown$ indicate the lowest initial offset for which the droplets rotate over each other. For the lower boundaries $\blacktriangle$ and $\triangle$ indicate the highest initial offset for which the droplets reverse, while ∇ and $\triangledown$ indicate the lowest initial offset for which the droplets coalesce.

It is clear from Fig. 5a that the upper boundary, above which the droplets rotate over each other without coalescence, decreases with increasing Ca . There are two reasons for this decrease. First, at the same offset but at a higher shear rate, the rotation speed of the doublet is faster [16]. Hence, there will be less time for film drainage. Secondly, due to the higher flow intensity, the hydrodynamic forces are larger, which causes an increase of the size of the flattened film drainage area and consequently a decrease in the film drainage rate [10,13,20]. As a result of this combined effect, the critical initial offset up to which coalescence occurs, decreases with the Ca . Yang et al. [20] also reported a decrease of the critical Ca with the initial offset for droplet pairs with a viscosity ratio of approximately 0.1 in mixed linear flows. It can also be seen in Fig. 5a that the initial offset boundary continues to decrease and converges towards zero at large values of the Ca .

A comparison of the results for droplet size $2R = 165$ μ m and $2R = 210$ μ m in the unconfined case shows that for the larger droplets the initial offsets up to which coalescence is possible are somewhat smaller. This can be explained by the fact that for a larger droplet size, the film drainage area is larger and hence, the film drainage is slower [10,20]. There are two reasons for this larger film drainage area. First, from Eq. 8 in Yang et al. [20], it can be seen that the hydrodynamic force along the line of centers of an interacting droplet pair in shear flow scales with $\eta_m \dot{\gamma} R^2$, which corresponds

to CaR . Hence, at the same Ca , the hydrodynamic force increases with droplet size. In addition, larger droplets have a smaller capillary pressure, which facilitates flattening of the droplet interface. As a result, the film drainage area scales with CaR^2 , as can be seen from Eq. 11 in Yang et al. [20]. Thus, at a constant value of the Ca , the film drainage area increases substantially with droplet size. However, it must also be noted that for the larger droplets, at the same Ca the shear rate is lower and hence, the rotation speed of the doublet is also lower [16]. It can be concluded from Fig. 5a that the effect of the slower film drainage dominates over the effect of the slower rotation speed and hence, the net result of an increase in droplet size is a decrease of the initial offset boundary at a fixed value of the Ca . Alternatively, at a fixed initial offset, an increase in droplet size results in a decrease of the critical Ca . A similar observation has also been reported for mixed linear flows [18,20,47] and simple shear flow [39].

When comparing between bulk ($H = 3$ mm) and confined ($H = 0.5$ mm) conditions for both droplet sizes, it is clear from Fig. 5 that for all capillary numbers the upper offset boundary for coalescence is higher in the confined case as compared to the bulk case. A partial explanation for this can be given based on the trajectories in Fig. 3. Whereas in bulk flow the offset slightly increases during approach of the droplets, in confined flow it decreases during approach. As a consequence, starting from the same initial offset, the droplets make apparent contact at a lower offset in confined conditions as compared to bulk conditions, which implies that in confined conditions more rotation of the droplet doublet can occur before the droplets separate. In addition, the hydrodynamic force, pushing the droplets together during apparent contact, is smaller at low orientation angles (or offsets) [13]. A smaller hydrodynamic force causes a smaller size of the film drainage area and an increase in the film drainage rate [10,13,20]. As a consequence, droplets in confined conditions which make apparent contact at a smaller offset will initially have a smaller film drainage area and the initial film drainage is expected to be faster. The observed higher initial offset boundary in confined conditions at a fixed Ca is in agreement with the observation of Chen et al. [39], who showed that confinement enables coalescence up to larger capillary numbers at a fixed initial offset.

Those authors also mentioned that geometrical confinement results in additional hydrodynamic wall forces that might affect the film drainage process. As in bulk conditions, also for the confined conditions, the upper offset boundary decreases with the Ca . The reasons for this decrease are similar to those in bulk conditions.

A comparison of the upper boundaries for the two droplet sizes in the confined case is less straightforward as in the unconfined case, since now both the droplet size and the confinement ratio vary. Hu et al. [47] showed that in bulk conditions the critical capillary number for a fixed initial offset varies as $R^{-0.94}$. By using this scaling, Yang et al. [20] obtained a master curve of $Ca \cdot R^{0.94}$ versus offset for three different droplet sizes between 45 and 73 μm . In Fig. 6 the upper boundaries for the initial offset are plotted as a function of $Ca \cdot R^{0.94}$ for both droplet sizes. Fig. 6a demonstrates this scaling for the unconfined case. It can be concluded that the scaling applied by Yang et al. [20] for mixed linear flows also leads to superposition here. Fig. 6b shows the scaled results for the confined case. In order to verify if the scaling is also applicable for confined conditions, the upper boundaries for a droplet size of $2R = 210 \mu\text{m}$ at a confinement ratio of $2R/H = 0.33$ are added to Fig. 6b. It is clear from Fig. 6b that the scaled results for $2R = 210 \mu\text{m}$ and $2R/H = 0.33$ coincide with the scaled results for $2R = 165 \mu\text{m}$ at the same confinement ratio. Consequently, it can be concluded that the scaling of Ca_{crit} with $R^{0.94}$ is also applicable for the confined data in this work. Due to the scaling, the effects of droplet size are eliminated and the effect of confinement ratio can be assessed. By comparing the results for $2R/H = 0.42$ with those for $2R/H = 0.33$, it is clear that for the higher confinement ratio the initial offset boundary is larger. Hence, an increase in the confinement ratio enables coalescence up to higher initial offsets.

Fig. 6. Initial dimensionless offset boundaries as a function of the Ca number scaled by $\frac{\Delta Y_0}{2R} = 1.6$. (a) $H = 3$ mm, (b) $H = 0.5$ mm and $\frac{H}{2R} = 0.636$ mm.

In addition to increasing the upper initial offset boundary, confinement also induces a lower boundary for coalescence. It can be seen from Fig. 5b that this lower boundary is independent of the capillary number for capillary numbers ranging from 0.004 up to the maximum value that allows coalescence. This lower boundary due to confinement could contribute in the formation of pearl necklaces as observed by Pathak et al. [48] by preventing coalescence of interacting droplets with a low offset. Comparing the lower initial offset boundaries for $2R = 165$ μm and $2R = 210$ μm in Fig. 5b shows that the lower initial offset boundary is the largest for the largest droplet size and hence the largest confinement ratio. To assess the separate effects of droplet size and confinement ratio on this lower initial offset boundary, the confinement ratio was varied for both droplet sizes by changing the gap spacing. Fig. 7 provides an overview of the results. It is clear from Fig. 7 that the lower initial offset boundary increases with increasing confinement ratio for both droplet sizes. This is expected, because the reversing droplet trajectories are a result of confinement. Consequently, this lower boundary for the initial offset should go to zero when the confinement ratio becomes small, as is indeed the case in the unconfined experiments. A second observation is that this lower boundary is also dependent on the droplet size: for the larger droplets the initial offset boundary is larger. Hence, the larger lower boundary for the initial offset in Fig. 5b as compared to Fig. 5a is a result of both the larger confinement ratio and the larger droplet size.

Fig. 7. Initial dimensionless offset boundaries as a function of confinement ratio $2R/H$, $\Delta Y_0/2R = 1.6$, $Ca = 0.010$. $\blacktriangle$ and $\triangle$ indicate the largest initial offset for which the droplets reverse. $\blacktriangledown$ and $\triangledown$ indicate the smallest initial offset for which the droplets coalesce.

Numerical simulations in 2D of the approach and interaction of droplets with different initial offsets were performed to relate the dependencies of the lower offset boundary on confinement ratio, droplet size and capillary number to the hydrodynamics of the flow around the droplet pairs. The trajectories of two interacting droplets are given in Fig. 8 for different values of the relevant parameters. Fig. 8a and b show that the offset from which droplets start to rotate over each other rather than reversing their flow direction upon collision, increases significantly when the confinement ratio $2R/H$ is increased from 0.2 (Fig. 8a) to 0.33 (Fig. 8b). It should be noted that the values of the lower offset boundary do not quantitatively match the ones of Fig. 7. This is most probably caused by the 2D nature of the simulations. In addition, for some trajectories, the dimensionless offset $\Delta Y/2R$ decreases with the dimensionless distance $\Delta X/2R$ between the droplets. Hence, the reported initial dimensionless offset depends on the chosen value of the initial dimensionless distance $\Delta X_0/2R$, which is lower in the case of the simulations. However, it is clear that the flow field around the droplet pairs contains a larger zone of recirculating flow when the confinement ratio is increased. When comparing Fig. 8c with Fig. 8b, it becomes evident that the droplet trajectories are not affected by a doubling of the Ca , which results from the fact that both cases are in the creeping flow regime [49]. This explains the insensitivity of the lower offset boundary

to the capillary number, which was shown in Fig. 5. However, Fig. 8c shows that the larger capillary number leads to slightly more deformed droplets, as the trajectories of the droplet doublets deviate more from that of hard spheres as compared to those of Fig. 8b. Finally, Fig. 8d clearly shows that an increase of the droplet size with about 25% does not affect the droplet trajectories. This is expected as none of the dimensionless groups affecting the hydrodynamics around the droplet pair is altered with respect to Fig. 8b. Nevertheless, the experimental results in Fig. 7 show that the range of offsets for which coalescence occurs, extends to lower values for smaller droplet sizes. The latter is caused by the fact that a smaller droplet size facilitates the drainage of the matrix film between the droplets [17]. Hence, even though a droplet moves on a reversing trajectory, it might still coalesce depending on the droplet size, as can be seen in Fig. 3b for $\Delta Y_{in}/2R = 0.186$.

Fig. 8. Droplet trajectories from numerical simulations in 2D: Evolution of the offset $\Delta Y/2R$ as a function of $\Delta X/2R$ for different initial offsets, $\Delta X_{in}/2R = 1.2$.

3.3 Coalescence efficiency

As a consequence of the existence of a lower boundary for the initial offset, the extent of the initial offset range in which coalescence is possible is slightly smaller in confined conditions as compared to bulk conditions, for all capillary numbers under investigation (Fig. 5). However, it must be noted that in the confined case the range of initial offsets for which coalescence is possible lies in the region of higher offsets where the relative velocity between the two droplets is higher. Hence, in a concentrated emulsion there is a larger chance of collisions per unit time between droplets with these offsets. More specifically, the local collision frequency linearly increases with the offset in the velocity gradient direction [13]. To take this into account, the coalescence efficiency is not defined as the maximum initial offset up to which coalescence occurs, but as the square of this maximum offset [13,50]. Since in the confined case there is also a lower critical initial offset below which coalescence is not possible, the coalescence efficiency for the confined case can be extended to:

$$\text{Coalescence efficiency} = (\text{upper critical initial offset})^2 - (\text{lower critical initial offset})^2 \quad (7)$$

The coalescence efficiencies according to this definition are plotted in Fig. 9 for droplet sizes $2R$ of respectively 165 and 210 μm . It can be seen that for both droplet sizes the coalescence efficiency is larger in the confined case. Mainly at low Ca , there is a substantial effect of confinement, with more than a doubling of the coalescence efficiency for $2R = 210 \mu\text{m}$. With increasing Ca , the difference in coalescence efficiency between confined and unconfined cases gradually decreases. The enhancement of the coalescence efficiency due to confinement may facilitate the formation of very large droplets or strings which were observed by several authors for confined, concentrated emulsions [33].

Fig. 9. Coalescence efficiency as a function of Ca-number, $H_0/2R = 1.6$. (a) $2R = 165 \mu\text{m}$, (b) $2R = 210 \mu\text{m}$. For the calculations of the coalescence efficiency at $Ca = 0.002$, extrapolated values of the lower critical initial offset have been used.

The experimentally determined coalescence efficiencies for bulk conditions can be compared with model predictions for coalescence efficiency. Based on the partially mobile interface drainage model, Mousa et al. [14] predict a coalescence efficiency of 1 for the Ca, flow number ($Fl = 6\pi \cdot \eta_m \cdot \gamma/A$) and viscosity ratio used in our experiments (the Hamaker constant A used for the calculation of the flow number is taken to be $5 \cdot 10^{-21} \text{ J}$, which is the same order of magnitude as values used in literature [5,14,15]). The large overestimation of the coalescence efficiency by this PMI model can be attributed to the assumptions that were made concerning the lubrication force and the shape of the contact area between the droplets. With the help of trajectory analysis Wang et al. [11] predict a value of approximately 0.59 for the coalescence efficiency of droplets with a viscosity ratio of 0.1. Since the trajectory analysis does not take film drainage into account, this value is an upper limit for the coalescence efficiency. It can indeed be seen on Fig. 9 that the coalescence efficiency in unconfined conditions never exceeds this value. Vinckier et al. [22] used the partially mobile interface model of Chesters [10] to predict the coalescence probability in concentrated blends with the critical film thickness h_c as a fitting parameter. When using this technique to fit the coalescence efficiencies in Fig. 9, it was clear that this very simple model poorly predicts the dependency of the coalescence efficiency on the capillary number (results not shown). Moreover, a value of h_c which is too small to be physically possible, was needed as fitting parameter. The poor

predictions can be attributed to the simplicity of the model, i.e. the assumptions made concerning the force, film geometry and interaction time. Hence, to describe the coalescence efficiency correctly over the complete range of Ca , the full hydrodynamic problem of the flow around the droplet pair and between the droplet interfaces will have to be solved [15]. However, this is beyond the scope of the present work.

4. Conclusions

The effect of confinement on droplet coalescence in simple shear flow is systematically investigated by visualizing two interacting droplets. More specifically, the effect of varying the initial offset between the droplet centers in the velocity gradient direction is studied. It is found that confinement enables coalescence up to higher initial offsets on the one hand. On the other hand, confinement also induces a lower boundary for the initial offset. These differences between bulk and confined droplet pairs can be explained by the changes in the flow field induced by confinement: confinement induces large recirculation zones at the front and rear of the droplet pair. As a consequence of these recirculation zones, for low initial offsets the droplets reverse flow direction upon interaction, which causes the lower boundary for the initial offset. For higher initial offsets, the recirculating streamlines cause a decrease in offset during approach of the droplets. Hence, they make apparent contact at a lower offset. As a consequence, coalescence is possible in confinement up to higher initial offsets.

The effects of Ca , droplet size and confinement ratio on the lower initial offset boundary are investigated in detail. It is observed that this lower boundary is independent of the capillary number, but increases with increasing confinement ratio and droplet size. 2D simulations of the droplet trajectories confirm the independency of this boundary of the capillary number (when remaining in the creeping flow limit) and the increase of this boundary with increasing confinement ratio. When increasing the droplet size, without changing the confinement ratio or Ca , the hydrodynamics of the flow around the droplet pair and hence the droplet trajectories remain the same. Nevertheless,

when there is apparent contact between the droplets, a smaller droplet size facilitates the drainage of the matrix film between the droplets, which results in a lower value of the lower offset boundary. Finally the coalescence efficiencies in confined and bulk conditions are compared. It can be concluded that confinement systematically increases the coalescence efficiency. The latter is expected to have a substantial effect on droplet coalescence in emulsions, which is determined by a combination of collision frequency and coalescence efficiency. The experimental dataset presented here can also serve as a reference data set for the future development of models for coalescence efficiency in confined shear flow.

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$\Delta Y_{in}/2R = 0.437$ - $Ca = 0.006$ - Confined conditions

$\Delta Y_{in}/2R = 0.329$ - $Ca = 0.006$ - Confined conditions

$\Delta Y_{in}/2R = 0.182$ - $Ca = 0.006$ - Confined conditions

Confined conditions

Bulk conditions

Highlights

- Experimental investigation of the coalescence of 2 droplets in confined shear flow.
- Focus on effects of varying the initial offset in the velocity gradient direction.
- Due to confinement coalescence is possible up to higher initial offsets.
- Confinement causes reversing droplet trajectories at low initial offsets.
- Confinement increases the coalescence efficiency compared to bulk conditions.