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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.

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, pharmaceutics, 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

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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 in 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 inalgorithms 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 multiphasic 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 and 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 and the y-direction is the velocity gradient direction.