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Coalescence and Non-coalescence Phenomena in Multi-material Problems and Dispersed Multiphase Flows: Part 1, A Critical Review of Theories

Marcello Lappa [1]

Abstract: The manuscript deals with a presentation of the most reliable theories introduced over the years to model particle coalescence and non-coalescence phenomena at both macroscopic and microscopic length scales (including historical developments and very recent contributions) and moves through other macrophysical mechanisms that can cause spatial separation of the fluid phases (liquid-liquid or liquid-gas) in multi-material problems, while providing a rigorous theoretical framework for deeper understanding of how drop (or bubble) migration due to gravity and/or Marangoni effects can interact cooperatively with coalescence to significantly affect the multiphase pattern formation, its evolutionary progress as well as the final quality of the material in its solid state.

1 Introduction

The physical properties of many materials strongly depend on the multiphase morphology which is controlled to a great degree by particle-particle interaction in the liquid phase during the related processing.

The possibility that inclusions (drops or bubbles) collide and coalesce with each other as they move (under various natural forces, e.g., buoyancy or thermal Marangoni effects) has to be regarded as a significant and relevant part of the problem (these aspects often are referred to as collision and coagulation events).

Owing to the experimental difficulties in investigating the fluid-dynamics of nontrasparent liquids (e.g., metal alloys are opaque) a number of mathematical models and methods have appeared over the last few years for a numerical/theoretical analysis of these aspects when they are disjoint or partially combined.

It is known that the dynamics of an immiscible fluid-fluid system subjected to the action of gravity and of surface tension forces is characterized (categorized in terms of relative importance of different effects) in principle by the following non-dimensional numbers:

The capillary number:

$$Ca = \frac{\mu V_{ref}}{\sigma} \tag{1}$$

where, μ is the dynamic viscosity of the external (majority) liquid, V_{ref} a reference velocity and σ is the surface (or interfacial) tension between the two fluid phases. This number is an important measure of the dynamic deformability of the free surface as it represents the relative effect of viscous forces and surface tension. If $Ca \rightarrow 0$ (i.e. $Ca << 1$) dynamic surface deformation can be neglected.

The Bond number:

$$Bo = \frac{\Delta\rho g a^2}{\sigma} \tag{2}$$

where $\Delta\rho$ is the density jump between the liquid phases, "a" a reference length (e.g., the radius of the droplet or bubble). The parameter Bo represents the ratio of internal hydrodynamic pressure to surface-tension force. If this number is sufficiently small the fluid/fluid interfaces behave (approximately) not much differently with respect to the case of zero-g.

The well-known Reynolds number:

$$Re = \frac{\rho V_{ref} a}{\mu} \tag{3}$$

where ρ is the density of the external (majority) liquid and V_{ref}, for instance, can be the average particle sedimentation (or rise) velocity or its migration velocity induced by Marangoni effects, established during the phenomena under consideration (since in many applications, these velocities are not known *a priori*, an equivalent definition can be based on the so-called *scaling* velocities for these phenomena, i.e. the buoyancy velocity $V_g = \frac{g\Delta\rho a^2}{\mu}$

[1] MARS (Microgravity Advanced Research and Support Center), Via Gianturco 31 - 80146, Napoli, Italy, E-mail: marlappa@marscenter.it, marlappa@unina.it

for gravity-dominated processes or the Marangoni velocity $V_{Ma}^{T} = \frac{\sigma_T \Delta T}{\mu}$ for thermally surface-tension driven processes, ΔT being a reference temperature difference).

The Prandtl number:

$$\Pr = \frac{\nu}{\alpha} \tag{4}$$

represents the ratio of molecular momentum to molecular thermal transport, ν and α being the kinematic viscosity and the thermal diffusivity of the majority phase, respectively. The Marangoni number can be defined by a combination of the Re and Pr numbers, i.e. Ma=Pr·Re.

Other relevant non-dimensional parameters are given by the ratios of the minority phase (drop or bubble) density, viscosity, thermal conductivity, specific heat, etc. to the ones of the outer fluid.

2 Earlier studies for solid spheres

Earlier studies in the case of spherical solid spheres are due to Bossis and Brady (1984), Brady and Bossis (1985), Fortes et al. (1987), Feng et al. (1994). See also Lamb (1932) for potential flow solutions and Happel & Brenner (1965), Batchelor (1972), Davis and Acrivos (1985) and Kim & Karrila (1991) for Stokes flow solutions. In general, the central theme of these studies has been calculation of the average sedimentation velocity and/or the drag coefficients for two fundamental modes of interactions (i.e., spheres moving in tandem or side-by-side) and comparison of the results with the corresponding ones for a solitary sphere (Hasimoto, 1959; Sangani and Acrivos, 1982; Zick and Homsy, 1982).

3 Coalescence as an ideal collision process

Embryonic theoretical studies for the case of liquid droplets in simple shear flow (linear flow field) can be tracked back to Smoluchowski (1917), who treated coalescence as an ideal collision process with particle interaction being neglected (a simple ballistic model). In the light of the studies of Lin et al. (1970) and Batchelor and Green (1972) who calculated the hydrodynamic interactions between solid spheres and their effect on particle trajectories under flow, Zeichner and Schowalter (1977) and Wang et al. (1994) added a coalescence "efficiency" to Smoluchowski theory to account for possible hydrodynamic interactions (Lyu et al., 2002).

Smoluchowski's theory, in an analogy with ideal molecular collision theory, simply reflects the main mechanism of coalescence. However, it neglects all particle-particle interactions that exist in real coalescence processes. Without hydrodynamic interaction Smoluchowski-case droplet trajectories are simply straight lines. In such a case a collision occurs for all the small drops that move towards a large one.

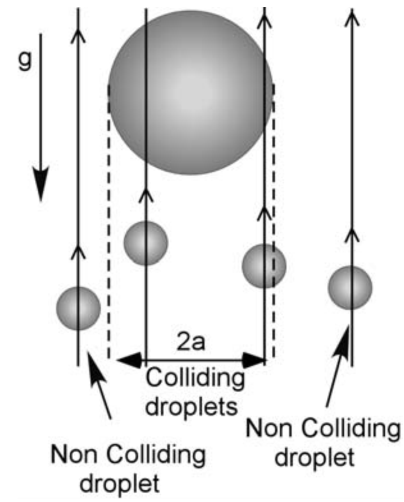


Figure 1 : Coagulation events as predicted by the Ballistic model (solid lines represent small droplets relative trajectories, arrows along solid lines indicate relative motion with respect to the falling large drop).

Consider, for instance, a large drop of radius a sedimenting under the effect of gravity in a liquid matrix containing other droplets of smaller size; since the sedimentation velocity is an increasing function of the radius (Clift et al., 1978), the larger drop will move with larger velocity with respect to the other smaller ones. According to the ballistic model, all the droplets totally or partially located in a region of amplitude 2a under the drop, will collide (and coalesce) with it (Fig. 1).

The example above could also be used as a paradigm case for the Marangoni migration phenomena; it is well-known (see, e.g., Lappa, 2005a), in fact, that the migration velocity is an increasing function of the radius. Thus, according to Smoluchowski's theory a large drop would capture all the droplets totally or partially located in a region of amplitude 2a along the migration direction.

4 The trajectory theory

However, in reality, hydrodynamic interactions cause the trajectories of particles to deviate from straight lines. Only those droplets in a reduced region will collide with

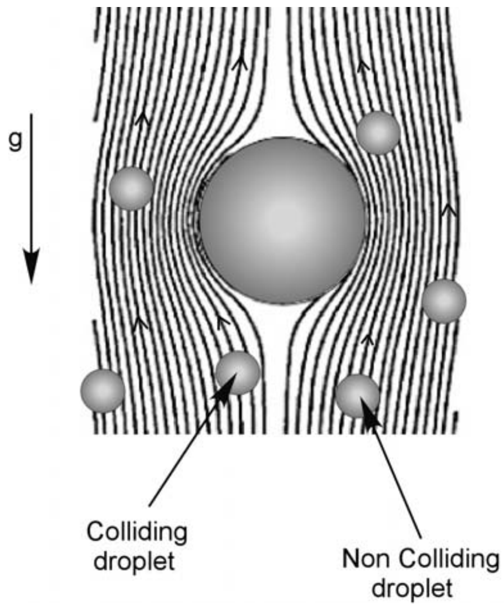


Figure 2 : Coagulation events as predicted by the Trajectory theory (solid lines represent small droplets relative trajectories, arrows along solid lines indicate relative motion with respect to the falling large drop).

the large one. Other droplets will follow the streamlines around the large drop and pass by without colliding with it (Zhang and Davis, 1991), see, e.g., Fig. 2.

As mentioned before, Zeichner and Schowalter (1977) improved Smoluchowski's theory by considering the hydrodynamic interaction that is related to the trajectories of particles (leading to the so-called "trajectory theory"). Recently, Wang et al. (1994) have calculated the coalescence efficiency within the framework of this theory in the canonical reference case of simple shear flow. This theory assumes the drops to be nondeformable.

Both Smoluchowski and trajectory theories deal with macrophysical kinematic aspects leaving aside deforming fluid/fluid interfaces and thinning fluid films between drops in near-contact motion.

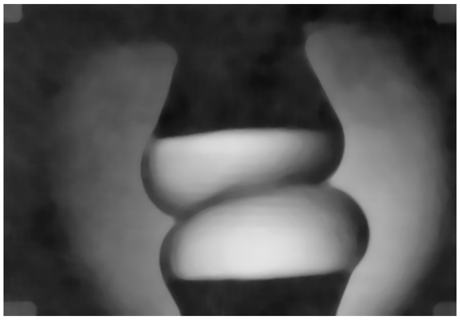
Coalescence of droplets in a liquid matrix, however, cannot be reduced to a mere matter of macroscopic dynamics. It, in fact, has been also defined as the process in which during the mutual interaction of two drops the liquid immiscible film formed between them drains out to a thickness at which it ruptures. Accordingly, coalescence rates and efficiencies between droplets generally depend on the local dynamics of the fluid drainage in the near contact region between the two approaching fluid interfaces. In this region larger pressures can develop (as a result of relative motion of the droplets and of the so-called related "squeeze flow" that is established in the gap between them) and resist and/or retard coalescence sometimes preventing it altogether.

5 The lubrication theory: the thin film

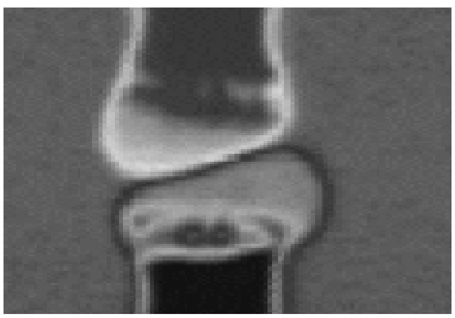
In practice, pressure increase in the thin domain between two droplets is driven by viscous stresses induced by relative motion of the droplets. Along these lines, favorable conditions for occurrence of coalescence were initially investigated within the framework of the lubrication theory by Davis et al. (1989). The hydrodynamic force resisting the relative motion of two unequal drops was determined for Stokes flow conditions and in the absence of the surface Marangoni effect. The drops were assumed to be in near-contact and to have sufficiently high interfacial tension that they remain spherical. The squeeze flow in the narrow gap between the drops was analyzed using the aforementioned lubrication theory. Depending on the ratio of drop viscosity to that of the continuous phase, and also on the ratio of the distance between the drops to their radii, different possible flow situations were pointed out, corresponding to nearly rigid drops, drops with partially mobile interfaces, and drops with fully mobile interfaces. In particular, the hydrodynamic resistance was predicted to be weaker than that for two colliding rigid spheres in near contact due to the mobility of the drop interfaces in the study of Barnovski and Davis (1989).

The shape of droplets was assumed undeformable in these analyses ($Ca << 1$). In reality, the related interaction can cause drop deformation even in the case of negligible convective transport. In turn, such a deformation can result in a greater hydrodynamic interaction, causing the coalescence rate to decrease (as shown by Yiantsios and Davis, 1991). Drops with high surface tension, in fact, produce a film of a small area that drains quickly whereas more deformable drops tend to trap a large amount of fluid in a film with large area (in many circumstances, drop deformation can be also responsible for other important effects; for instance, it can alter the Marangoni migration and sedimentation velocities).

The shape deformation was considered in the so-called "film drainage theory" introduced later by Chesters (1991). Chesters (1991) made a different correction to Smoluchowski's theory by considering particle deformation and squeezing flow of matrix fluid, known as "film drainage".



(a)



(b)

Figure 3 : Non coalescence of two drops of silicone oil in the presence of an imposed temperature difference (after Monti et al., 2000): (a): CCD visualization; (b): Infrared image.

As mentioned before, two drops approaching each other trap a thin film of the continuous phase between their interfaces. At small enough gaps the hydrodynamic forces overcome capillarity and the drop interfaces deform and often acquire a dimpled shape that traps more fluid thus opposing coalescence (Yiantsios and Davis, 1990). In flow-driven drop interactions, coalescence occurs for capillary numbers smaller than a critical value such that the drop interaction time is larger than the drainage time for the fluid trapped in the gap (Chesters, 1991). For sub-critical capillary numbers, at sub-micron separations, van der Waals forces become dominant leading to rapid coalescence (film rupture).

It was theoretically shown that the critical thickness h_c at which the matrix film between the droplets automatically ruptures is $h_c = (AD_m/16\pi\sigma)$ where A is the Hamaker constant, D_m is an average drop diameter defined as $D_m=2(1/D_i+1/D_j)^{-1}$. Coalescence efficiency was then calculated for three ranges of viscosity ratio called "mobile", "partially mobile", and "rigid interfaces" in the original article. These results have been recently refined by Bazhlekov et al. (2000).

6 Permanent non-coalescence and static configurations

The theory related to the existence of the lubrication film under dynamic conditions (drops in relative motion) retarding coalescence and preventing it in some circumstances has enjoyed an outstanding success also in explaining non-coalescence phenomena induced by Marangoni effects at the drop surface.

It is known (Dell' Aversana et al., 1996, 1997; Monti and Savino, 1997; Monti et al., 2000; Neitzel and Dell'Aversana, 2002; Dell'Aversana and Neitzel, 2004) that thermocapillarity can be used to prevent a pair of drops of liquid from coalescing in a stable permanent way in air (all that is found for drops in air holds for drops submerged in an immiscible liquid matrix; the situation is more complex but the explanation is basically the same). Consider, for instance, the photograph shown in Fig. 3. Two drops of silicone oil are attached to copper rods and subjected to a temperature difference as they are pressed together.

The upper drop is hotter than the lower one, resulting in a region near the point of apparent contact which is either colder (upper drop) or hotter (lower drop) than the bulk liquid in the drop, and hence, the majority of the free-surface. The existence of a surface-temperature gradient and the temperature dependence of surface tension provide a liquid flow toward the contact region in the upper drop and away from it on the lower one, as illustrated in the sketch provided in Fig. 4.

The liquid along the upper drop surface is directed towards the symmetry axis, dragging the nearby external fluid with it by viscous forces. Thus the external fluid is entrained between the two surfaces and this entrainment effect is responsible for the presence of a stable fluid film. For coalescence to occur this thin lubrication channel must be removed or become sufficiently thin so that the liquid at the opposed sides can come into molecular contact.

In practice, in the lower drop the Marangoni flow along the contact region is instead directed from the symmetry axis towards the exit of the channel so that it opposes

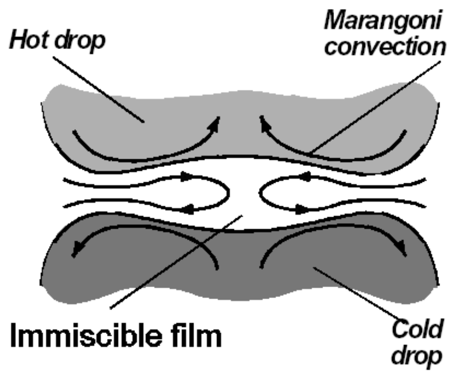


Figure 4 : Sketch of flow motion inside the lubrication film between two drops as they are pressed together under an imposed temperature (hot drop on the top, cold drop on the bottom).

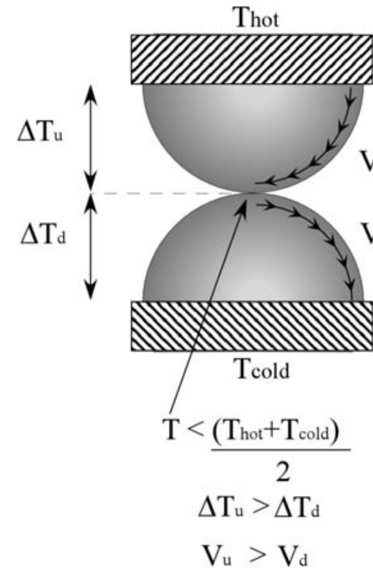


Figure 5 : Sketch of flow motion inside the drops and related temperature gradients established along their surface as they are pressed together under an imposed temperature gradient.

the aforementioned entrainment effect (the ambient fluid is convected outside the film); however, because of the smaller velocity along the lower drop surface compared to the one along the upper drop surface (in practice the temperature established in the intermediate region tends to be lower than the average temperature of the two-droplet system, see Fig. 5), the entrainment effect prevails.

Thus the pressure in the film changes from the ambient value to a maximum value in correspondence of the axis of symmetry that can be used to obtain stable non-coalescence (Dell'Aversana et al., 1997; Monti et al., 2000). A similar effect, of course, can be also effective under dynamic conditions strongly retarding or preventing coalescence.

Surfactants (through Marangoni stresses, surface viscosity, Gibbs elasticity, surface and/or bulk diffusivity and intermolecular forces) can also have a significant effect and stabilize emulsions by increasing deformation and causing surface tension gradients (solutal Marangoni stresses) that resist radial flow in the gap and interface–interface approach thus preventing coalescence (see, e.g., Chester and Bazhlekov, 2000 and the recent review of Cristini and Tan, 2004).

7 Hydrodynamic interactions under buoyancy forces

With regard to the case of the interaction of drops undergoing a buoyancy-driven motion numerous recent numerical studies revealed a rich variety of interaction patterns of deformable drops depending on the Bond number and the initial configuration of the system (see, e.g., Manga and Stone, 1993; Rother et a., 1997; Cristini et al., 1998).

The deformation and motion of interacting droplets was investigated by these authors in the framework of boundary-integral techniques (with mesh adaptation and stabilization) applicable to the case of slow viscous motion.

As a recent example of such computations the reader may consider the axisymmetric buoyancy-driven interaction of a leading drop and a smaller trailing drop treated by Davis (1999). He demonstrated that the trailing drop elongates considerably due to the hydrodynamic influence of the leading one and that later, depending on the governing parameters, the drops may either separate and return to a spherical shape or the trailing drop may be captured by the leading one (coalescence), or one of the drops may break up. In particular, it was found that when the Bond number is small, interfacial tension keeps the drops nearly spherical, and they separate with time. At higher Bond numbers, however, deformation is significant and the trailing drop is stretched due to the flow created by the leading drop; it may form one or more necks and break when one of these pinches off. The leading drop is flattened due to the flow created by the trailing drop; it may form a depression on its underside which evolves into a plume that rises through its center. Moreover, at sufficiently high Bond numbers, the larger leading drop does not leave the trailing drop behind, but instead may entrain and engulf it within the aforementioned depression or plume.

Cristini et al. (1998) carried out a similar study for the more general case of three-dimensional (3D) interactions and Bo=O(1) (initial conditions corresponding to a small leading drop and a larger trailing drop not aligned along the gravity direction, see Fig. 6). Lubrication stresses between the deformed drops were found to prevent coalescence; they observed the smaller drop to slide past the larger one, then to become highly stretched in the straining flow behind the large drop, then to continue to stretch under the action of buoyancy (surface tension being too weak for the drop to recover) and finally to form a neck and to pinch off under the action of surface tension.

8 Hydrodynamic interactions under Marangoni forces

For the case of motion induced by thermal surface tension forces, most of the available studies of the interaction of bubbles and droplets in the course of their Marangoni migration were performed under the assumption of nondeformable drops (zero capillary number). The literature on the thermocapillary motion of deformable drops, in fact, is limited in contrast to the problem concerning the interaction of drops undergoing a buoyancy-driven motion discussed before.

For the Marangoni motion, the effect of deformability was studied mostly by a perturbation technique assuming small deformations; some recent analyses within the framework of boundary integral methods are due to Zhou and Davis (1996), Berejnov et al. (2001) and Rother et al. (2002).

Berejnov et al. (2001), in particular (in the case of axisymmetric interaction), found that for equal-sized drops, the motion of a leading drop is retarded while the motion of the trailing one is enhanced compared to the undeformable case. The distance between the centers of equal-sized deformable drops decreases with time. They also illustrated that when a small drop follows a large one, two patterns of behavior may exist: for moderate or large initial separation the drops separate; however, if the initial separation is small there is a transient period in which the separation distance initially decreases and only afterward the drops separate.

Rother et al. (2002), by means of 3D results, pointed out that deformation increases the minimum separation and inhibits coalescence but is not important enough for appropriate physical parameters to induce the capture or breakup behaviors observed in buoyancy.

These analyses and methods, although very interesting, however, do not provide an exhaustive picture of the possible interaction mechanisms. Mutual interplay of different droplets, in fact, is not limited to hydrodynamic influences; also thermal (wake) effects can play a crucial role in these dynamics especially when dealing with surface tension driven motion.

9 Thermal interactions under Marangoni forces

Most of the available studies about thermal interaction have been carried out for the case of bubbles. Axisymmetric thermal-wake interaction of two bubbles in a uniform temperature gradient at large Reynolds and Marangoni numbers was studied by Balasubramanian and Subramanian (1999). Their analysis considered the bubbles moving in the direction of the temperature gradient and assumed to interact axisymmetrically via the influence of the thermal wake shedded by the leading bubble on the trailing bubble; in this analysis it was proven that the thermal wake past the leading bubble can induce a nonmonotonic temperature field on the surface of the trailing bubble. The effective temperature gradient on the trailing bubble is weakened and hence its migration speed is reduced compared to the case when it is isolated.

Similar behaviors are also expected in the case of interacting drops. The temperature gradient in the wake will be weaker than the applied gradient. The thermal wake field of the leading drop will wrap around the trailing drop having a significant impact on its motion.

In the aforementioned landmark analysis shape deformation was neglected and results were obtained in the limit as $Ma \rightarrow \infty$ (flow potential theory).

Leshansky et al. (2001) carried out a similar investigation for low values of the Marangoni number and undeformable bubbles. The perturbations to the bubble velocities (with respect to the single bubble case) were

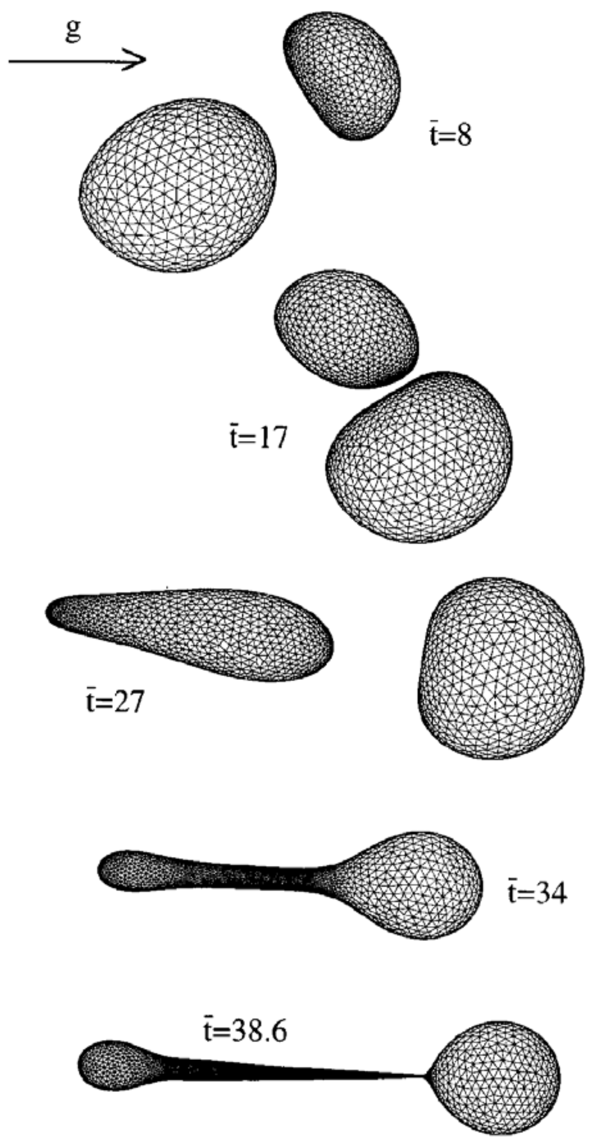


Figure 6 : Three-dimensional interaction between two drops undergoing sedimentation (initial conditions corresponding to a small leading drop and a larger trailing drop not aligned along the gravity direction); from Cristini et al. (1998) (reproduced with permission from the American Institute of Physics).

found to have opposite signs (the motion of the leading bubble is enhanced while the motion of the trailing one is retarded). They also disclosed that equal-sized bubbles, which otherwise would move with equal velocities, acquire a relative motion apart from each other under the influence of convection, whereas for slightly unequal bubbles there are three different regimes of large-time asymptotic behavior: attraction up to the collision, infinite growth of the separation distance, and a steady migration with equal velocities, the steady motion separation distance being a function of the parameters of the problem (the Marangoni number, initial separation and radii ratio).

The thermocapillary interaction of two equal bubbles with an arbitrary orientation relative to an externally imposed temperature gradient was studied by Leshansky and Nir (2001). They analytically demonstrated (in the weakly nonlinear limit of small Ma numbers) the tendency of equal bubbles to line up in a plane perpendicular to the applied thermal gradient.

Recently Lavrenteva and Nir (2003) considered the axisymmetric motion and related thermal wake interaction of two undeformable drops in a viscous fluid (with $Pr > 1$) under the combined effect of gravity and thermocapillarity. The analysis was focused on the case of "spontaneous" thermocapillary motion, i.e. Marangoni effects induced on the trailing droplet by the thermal wake originated from a leading rising drop moving under the effect of buoyancy forces (without imposed temperature gradient in the external liquid, i.e. isothermal liquid matrix and temperature of the leading drop exceeding that of the continuous media). They found that the induced change in the speed of the trailing drop is comparable in magnitude with its (buoyancy) speed when isolated even for large separation distance between the drops where the hydrodynamic interaction is negligible and that in the extreme case of very large Marangoni effect the direction of the trailing drop can be reversed.

10 Population methods

From the foregoing it is evident that current understanding is primarily limited to the motion of single drops and bubbles, or at most two interacting drops and bubbles, which are often assumed to remain spherical (a limit which requires that that interfacial tension forces are large compared to viscous and pressure forces). Moreover, all the studies dealing with the boundary-integral method and deformable drops are still limited to the case of very viscous flow ($Re \rightarrow 0$).

Initial progress on the simulation of many drops has been made by combining theoretical results available for neighboring drops (reviewed in the earlier sections) with economical multipole techniques previously used in multiparticle conductivity (see e.g., Zinchenko, 1994). On such a philosophy are based the so-called "population methods" (see, e.g., Davis et al. 1993; Diefenbach et al., 1993 and the more recent contributions Wu et al., 2003 and Zinchenko and Davis, 2003).

With these methods, drops (a large number, i.e. ensemble of droplets) are assumed to be in relative motion due to either gravitational sedimentation or thermal Marangoni migration. Possible collisions are predicted using a "trajectory analysis" to follow the relative motion of pairs of drops (such approach must not be confused with the trajectory theory illustrated in Sect. 4; the macroscopic trajectory analysis at the basis of population methods is a simple ballistic model). The trajectory analysis, in turn, is based on governing equations formulated for conditions of small Reynolds number (negligible inertia) and on estimation of the sedimentation or (Marangoni) migration velocity provided by analytical relationships reported in the literature.

When the drops become sufficiently close, they are assumed to interact with each other due to hydrodynamic disturbances. This hydrodynamic disturbance, as mentioned above, is modeled in the light of information obtained by means of a separate microscopic approach to the problem.

For instance, drops with low viscosity undergoing buoyancy motion are assumed to become aligned and coalesce due to their shape deformations as observed and described by Manga and Stone (1993), whereas drops with modest viscosity become stretched and may break as a result of hydrodynamic interactions and drops with large viscosity tend to be swept around larger ones.

Therefore, these methods rely on fundamental insights provided by previous analyses (lubrication theory for spherical drops and boundary-integral methods for the deformable case) in the case of only two interacting drops. Such information is used by population methods to model interaction of droplets at microscopic scale length (local interaction) from a macroscopic point of view. For this reason droplets pertaining to the initial distribution are categorized according to their size (drop size categories), and as a results of drop collisions and coalescence, the drop size spectrum in a dispersion is allowed to change over time with respect to the initial distribution. In practice, this change in the drop size distribution represents a macrophysical problem that is solved using "population dynamics equations". From a macroscopic point of view drops are treated as isolated, microscopic quantities compared to field variables like temperature, but the aforementioned equations include the kernels which contain the information regarding the interaction at microscopic scale between two drop size categories (this information, as discussed above, being provided a priori by solving separately a microphysical problem involving only a limited number of droplets). Finally coalescence events are handled at macroscopic scale length as instantaneous unions of drops at the center of mass, replacing them with a new drop with their combined volume.

Such a philosophy can be regarded as a very improved and modern form of the Smoluchowski model. It allows a simple and efficient treatment of the problem from a computational point of view and has been applied successfully to situations in which the physical phenomena of interest have a large length scale with respect to the average droplet size. However, vital information is lost about the effective microscopic evolution of the phenomena (drop deformation, shape instabilities, i.e. all those factors dealing with the local history of the shape or other "local" effects). Moreover the applicability of these methods is limited by the assumptions at the basis of the methods and information used to build the aforementioned kernels ($Re \rightarrow 0$).

With regard to all these numerical techniques and the possible approaches to the problem discussed before, it should be pointed out that quasisteady (very viscous) or nonviscous models are often inadequate, and that a fully transient and not simplified analysis would be necessary in order to properly describe and interpret the effective behavior of particles in real experiments and processes. An outstanding need is for new scientific approaches not plagued by any limitation or simplification of the governing model equations, applicable to the case of low, high, as well as intermediate values of the Reynolds number, and allowing the treatment of multiple drops (or bubbles) and related interplay, deformation and coalescence from a "local" point of view without resorting to dual-scale models. The second part of this article seeks to meet the outstanding need by introducing and/or reviewing moving-boundary methods able to predict particle interactions and deformations as well as collision and coagulation events in temperature gradients, with or without gravity being present over a wide range of conditions.

They can capture in a single numerical treatment and without limiting assumptions both macroscopic aspects (i.e. the macrophysical problem, heretofore treated in terms of population dynamics) and microscopic details (i.e. the microphysical problem, heretofore treated within the framework of boundary integral methods and/or under the assumption of nondeformable drops and/or under the assumptions of infinite Reynolds number and fluid-dynamic potential theories).

These methods have only recently been made sufficiently powerful to meet these objectives, including fully three-dimensional simulations of multiple drops or bubbles (Fig. 7). See Part 2 for additional details (Lappa, 2005b).

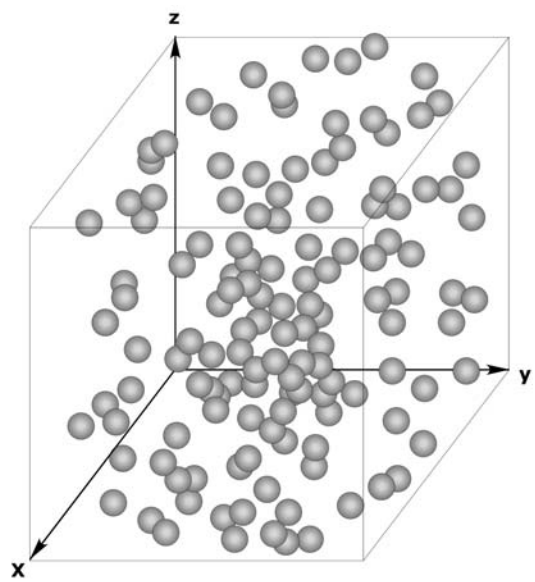


Figure 7 : Example of spatially inhomogeneous population of drops whose dynamics must be solved to predict phase-separation rates in problems related to immiscible organic or metal alloys.

References

Balasubramaniam, R. and Subramanian, R. S. (1999): Axisymmetric thermal wake interaction of two bubbles in a uniform temperature gradient at large Reynolds and Marangoni numbers. *Phys. Fluids*, vol. 11, pp. 2856-2864.

Barnovski, G.; Davis, R. H. (1989): The lubrication force between spherical drops, bubbles and rigid particles in viscous flow. *Int. J. Multiphase Flow*, vol. 15, pp. 627-638.

Batchelor, G. K. (1972): Sedimentation in a dilute dispersion of spheres. *J. Fluid Mech.*, vol. 52, pp. 245-268.

Batchelor, G. K.; and Green, J. T. (1972): The Hydrodynamic Interaction of Two Small Free-Moving Spheres in a Linear Flow Field. *J. Fluid Mech.*, vol. 56, pp. 375-400.

Bazhlekov, I. B.; Chesters, A. K. and Van de Vosse, F. N. (2000): The effect of the dispersed to continuous-phase viscosity ratio on film drainage between interacting drops. *Int. J. Multiphase Flow*, vol. 26, pp. 445–466.

Berejnov, V.; Lavrenteva, O. M. and Nir, A. (2001): Interaction of two deformable viscous drops under external temperature gradient. *J. Colloid Interface Sci.*, vol. 242, pp. 202-213.

Bossis, G. and Brady, J. F. (1984): Dynamic simulation of sheared suspension. I. General method. *J. Chem Phys.*, vol. 80, pp. 5141-5154.

Brady, J. F. and Bossis, G. (1985): The rheology of concentrated suspensions of spheres in simple shear flow by numerical simulation. *J. Fluid Mech.*, vol. 155, pp. 105-129.

Chesters, A. K. (1991): The modelling of coalescence in liquid-liquid dispersions:a review of current understanding. *Trans. Inst. Chem. Eng. Res. Des. Part A*, vol. 69, pp. 259–270.

Chesters, A.K. and Bazhlekov, I.B. (2000): Effect of Insoluble Surfactants on Drainage and Rupture of a Film between Drops Interacting under a Constant Force. *Journal of Colloid and Interface Science*, vol. 2302, pp. 229-243.

Clift, R.; Grace, J.R.; Weber, M.E. (1978): *Bubbles, drops and particles*. Academic Press, New York, 1978.

Cristini, V.; Blawzdziewicz, and Lowenberg, M. (1998): Drop breakup in three-dimensional flows. *Phys Fluids*, vol. 10, pp. 1781-1783.

Cristini, V. and Tan, Y.C. (2004): Theory and numerical simulation of droplet dynamics in complex flows: a review. *Lab Chip*, vol. 4, pp. 257-264.

Davis, R.H. (1999): Buoyancy-driven viscous interaction of a rising drop with a smaller trailing drop. *Phys. Fluids*, vol. 11, pp. 1016-1028.

Davis, R. H. and Acrivos, A. (1985): Sedimentation of noncolloidal particles at low Reynolds numbers. *Ann. Rev. Fluid Mech.*, vol. 17, pp. 91-118.

Davis, R.H.; Schonberg, J.A.; Rallison, J.M. (1989):

The lubrication force between two viscous drops. *Phys. Fluids*, vol. 1, pp. 77-81.

Davis, R.H.; Zhang, X.; Wang, H. (1993): Hydrodynamics of Dispersions and Coagulation. pp. 163-182, *in Immiscible Liquid Metals and Organics, L. Ratke Editor, Proceedings of an international workshop organized by the ESA expert working group, "Immiscible alloys" held in the Physikzentrum, Bad Honnef 1992*, Germany, 1993.

Dell'Aversana, P. and Neitzel, G.P. (2004): Behavior of Noncoalescing and Nonwetting Drops in Stable and Marginally Stable States. *Exp. in Fluids*, vol. 36, pp. 299-308.

Dell' Aversana, P.; Banavar, J.R.; Koplik, J. (1996), "Suppression of coalescence by shear and temperature gradients. *Phys. Fluids*, vol. 8, pp. 15-28.

Dell'Aversana, P.; Tontodonato, V.; Carotenuto, L.; (1997), "Suppression of coalescence and of wetting: The shape of the interstitial film. *Phys. Fluids*, vol. 9, pp. 2475-2485.

Diefenbach, S.; Ratke, L.; Prinz, B, Ahlborn, H. (1993), "Marangoni motion During Melting of a Hypermonotectic Alloy - Numerical Simulations for the D2-Experiment IHF04. pp. 291-298, *in Immiscible Liquid Metals and Organics, L. Ratke Editor, Proceedings of an international workshop organized by the ESA expert working group, "Immiscible alloys" held in the Physikzentrum, Bad Honnef 1992*, Germany, 1993.

Feng, J.; Hu, H. H.; and Joseph, D. D. (1994): Direct simulation of initial value problems for the motion of solid bodies in a Newtonian fluid. Part 1. Sedimentation. *J. Fluid Mech.*, vol. 261, pp. 95-134.

Fortes, A. F.; Joseph, D. D.; and Lundgren, T. S., (1987): "Nonlinear mechanics of fluidization of beds of spherical particles. *J. Fluid Mech.*, vol. 177, pp. 467-483.

Happel, J. and Brenner, H. (1965): *Low Reynolds number hydrodynamics*. Prentice-Hall.

Hasimoto, H. (1959): On the periodic fundamental solution of the Stokes equations and their application to viscous flow past a cubic array of spheres. *J. Fluid Mech.*, vol. 5, pp. 317-328.

Kim, S. and Karrila, S. J. (1991): *Microhydrodynamics: principles and selected applications*. Butterworth-Heinemann.

Lamb, H. (1932): *Hydrodynamics*. Dover, New York.

Lappa, M. (2005a): Assessment of VOF Strategies for the analysis of Marangoni Migration, Collisional Coagulation of Droplets and Thermal wake effects in Metal Alloys under Microgravity conditions. *Computers, Materials & Continua CMC*, vol. 2, pp. 51-64.

Lappa, M. (2005b): Coalescence and non-coalescence phenomena in multi-material problems and dispersed multiphase flows: Part 2, a critical review of CFD approaches. *Fluid Dynam. Mater. Processing*, present Issue.

Lavrenteva, O. M. and Nir, A. (2003): Axisymmetric thermal wake interaction of two drops in a gravity field at low Reynolds and high Peclet numbers. *Phys. Fluids*, vol. 15, pp. 3006-3014.

Leshansky, A. M.; Lavrenteva, O. M. and Nir, A. (2001): Thermocapillary migration of bubbles: convective effects at low Péclet number. *J. Fluid Mech.*, vol. 443, pp. 377-401.

Leshansky, A. M. and Nir, A. (2001): Thermocapillary Alignment of Gas Bubbles Induced by Convective Transport. *J. Colloid Interface Sci.*, vol. 240, pp. 544-551.

Lin, C. J.; Lee, K. J. and Sather, N. F. (1970): Slow Motion of Two Spheres in a Shear Field. *J. Fluid Mech.*, vol. 43, pp. 35-47.

Lyu, S.P.; Bates, F.S; Macosko, C.W. (2002): Modeling of Coalescence in Polymer Blends. *AIChE*, vol. 48, pp. 7-14.

Manga, M. H.; Stone, H. A. (1993): Buoyancy-driven interactions between two deformable viscous drops. *J. Fluid Mech.*, vol. 256, pp. 647-683.

Monti, R.; Savino, R. (1997): Correlation between experimental results and numerical solutions of the Navier-Stokes problem for two non coalescing liquid drops with Marangoni effect. *Phys. Fluids*, vol. 9, pp. 260-262.

Monti, R.; Savino, R.; Paterna, D.; Lappa, M. (2000): New features of drops dynamics under Marangoni effect. *Recent Research Developments in Fluid Dynamics, Transworld Research Network, Trivandrum, India*, vol. 3, pp. 15-32.

Neitzel, G.P.; Dell'Aversana, P. (2002): Noncoalescence and nonwetting behavior of liquids. Annu. *Rev. Fluid Mech.*, vol. 34, pp. 267-289.

Rother, M. A.; Zinchenko, A. Z.; and Davis, R. H., (1997): "Buoyancy-driven coalescence of slightly deformable drops. *J. Fluid Mech.*, vol. 346, pp. 117-148.

Rother, M. A.; Zinchenko, A. Z.; Davis, R. H. (2002): "Three-dimensional Boundary-integral Algorithm for Thermocapillary Motion of Deformable Drops. *J. Colloid Interf. Sci.*, vol. 245, pp. 358-364.

Sangani, A. S. and Acrivos, A. (1982): Slow flow through a periodic arrays of spheres. *Intl J. Multiphase Flow*, vol. 8, pp. 343-360.

Smoluchowski, V. (1917): Versuch einer Mathematischen Theorie der Koagulationskinetik Kollider Losungen Z.. *Phys. Chem.*, vol. 92, pp. 129-168.

Wang, H.; Zinchenko, A. Z. and Davis, R. H. (1994): The Collision Rate of Small Drops in Linear Flow Fields. *J. Fluid Mech.*, vol. 265, pp. 161-188.

Wu, M.; Ludwig, A.; Ratke, L. (2003): Modelling the solidification of hypermonotectic alloys. *Modelling Simul. Mater. Sci. Eng.*, vol. 11, pp. 755-769.

Yiantsios, S. G. and Davis, R. H. (1990): On the Buoyancy Driven Motion of a Drop Towards a Rigid Surface or a Deformable Interface. *J. Fluid Mech.*, vol. 217, pp. 547-573.

Yiantsios, S. G.; and Davis, R. H. (1991): Close Approach and Deformation of Two Viscous Drops due to Gravity and van de Waals Forces. *J. Colloid Interface Sci.*, vol. 144, pp. 412-433.

Zeichner, G. R. and Schowalter, W. R. (1977): Use of Trajectory Analysis to Study Stability of Colloidal Dispersions in Flow Fields. *AIChE. J.*, vol. 23, pp. 243-254.

Zhang, X. G. and Davis, R. H. (1991): The Rate of Collisions of Small Drops due to Brownian or Gravitational Motion of Small Drops. *J. Fluid Mech.*, vol. 230, pp. 479-504.

Zhou, H.; Davis, R.H. (1996): Axisymmetric Thermocapillary Migration of Two Deformable Viscous Drops. *J. Colloid Interf. Sci.*, vol. 181, pp. 60-72.

Zick, A. A. and Homsy, G. M. (1982): Stokes flow through periodic arrays of spheres. *J. Fluid Mech.*, vol. 115, pp. 13-26.

Zinchenko, A. Z. (1994): An efficient algorithm for calculating multiparticle thermal interaction in a concentrated dispersion of spheres. *J. Comput. Phys.*, vol. 111, pp. 120-134.

Zinchenko, A. Z.; Davis, R.H. (2003): Large-scale Simulations of Concentrated Emulsion Flows. *Phil. Trans. R. Soc. Lond.*, vol. 361, pp. 813-845.