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Modelling of immiscible liquid-liquid systems by Smoothed Particle Hydrodynamics

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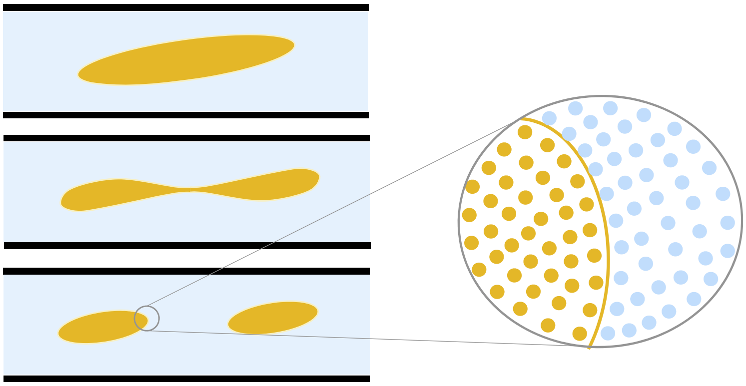
GRAPHICAL ABSTRACT



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ABSTRACT

Immiscible fluid systems are ubiquitous in industry, medicine and nature. Understanding the phase morphologies and intraphase fluid motion is often desirable in many of these situations; for example, this will aid improved design of microfluidic platforms for the production of medicinal formulations. In this paper, we detail a Smoothed Particle Hydrodynamics (SPH) approach that facilitates this understanding. The approach includes surface tension and enforces incompressibility. The approach also allows the consideration of an arbitrary number of immiscible phases of differing viscosities and densities. The nature of the phase morphologies can be arbitrary and change in time, including break-up (which is illustrated) and coalescence. The use of different fluid constitutive models, including non-Newtonian models, is also possible. The validity of the model is demonstrated by applying it to a range of model problems with known solutions, including the Young-Laplace problem, confined droplet deformation under a linear shear field, and a droplet falling under gravity through another quiescent liquid. Results are also presented to illustrate how the SPH model can be used to elucidate the behaviour of immiscible liquid systems.

1. Introduction

Processing of immiscible liquid-liquid dispersions occurs widely in the manufacture of foods [1], pharmaceuticals [2], cosmetics [3], paints [4], and oil [5]. Examples of processes include emulsification [4,6], encapsulation [7,8], multiphase reaction

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Abbreviations: CSF, continuum surface force; DPD, Dissipative Particle Dynamics; LBM, Lattice Boltzmann method; LGA, Lattice Gas Automata; LJ, Lennard-Jones; PPE, Pressure Poisson Equation; SPH, Smoothed Particle Hydrodynamics; VOF, Volume of Fluid.

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systems [9,10], electrochemical processes [11], bioprocesses [12], separation processes [13] such as liquid-liquid extraction [14], polymer blending [15], and oil recovery [16]. Processes involving immiscible liquids are also found beyond industry, including in environmental clean-up [17], artificial oxygen carriers [18], oil spills [19], and pyroclastic flows [20].

There is an increasing need to understand and exploit the link between liquid-liquid phase morphology and fluid dynamics within the phases on the one hand and the conditions that lead to them on the other. For example, the droplet size distribution of an emulsion produced through agitation is a function of the balance between droplet breakup and coalescence, which can be controlled by the surfactant and stabilizer concentration and relative velocities of the phases [21–23]. Microfluidic production of encapsulates provides a further example: in this case, encapsulate morphology can be varied through the nature of the flow-focusing in the microchannels and, amongst other things, the continuous-to-dispersed phase flow rate ratio [24,25]. The challenges faced in experimentally elucidating these types of relationships are significant, however. For example, the visualization of the morphologies of the phases and flow fields therein are still very much in their infancy [26–29]. Models that treat the phases and interfaces between them explicitly have, therefore, a key role to play in building understanding of liquid-liquid systems and exploiting this understanding in a systematic way.

Models of liquid-liquid systems in which the individual phases and interfaces between them are treated explicitly are long-standing [30–33]. The earliest models, which focus on droplets in a continuous phase, include those of Taylor [34,35], Mason and co-workers [36,37], Cox and co-workers [38,39], and Acrivos and co-workers [40–42] amongst others. Whilst these models were important in building understanding, they are limited by a good number of simplifying assumptions [43], including negligible inertial effects (*i.e.* small Reynolds numbers), small viscosity ratio ranges, and regular droplet shapes (*e.g.* spheres; ellipsoids). Phenomenological approaches have been adopted to overcome some limitations associated with wholly analytical models. For example, Maffettone and Minale [44] used such an approach, determining the model parameters by ensuring it matches analytical results in appropriate limits. Such models are, however, also limited by underpinning assumptions such as, for example, specific droplet shapes and the absence of any connection between the model parameters and underlying fundamentals.

Adoption of wholly numerical approaches can, in principle, overcome limitations faced by analytical and related models. Some of the earliest numerical models include those of Acrivos and co-workers [43,45,46], who studied the deformation and rupture of a viscous droplet suspended in another liquid within extensional and shear flow fields. More recent examples include the works of Loewenberg, Hinch and Davis [47–53], which are based on a boundary-integral approach. Interface tracking techniques such as Volume of Fluid methods (VOF) [54,55] and level set methods [56,57] have been used even more recently. Whilst these works represent major advances in the field, they have two significant disadvantages that lead to algorithmic complexity, computational expense and numerical stability challenges [33,58]: (1) the need to adjust the mesh as the phases deform; and (2) the need to track the interfaces between the different phases. Additionally, the treatment of coalescence and rupture of the interfaces between the phases is extremely challenging if not impossible in many of these techniques [59,60].

Particle-based approaches have more recently been used as a remedy to the issues faced by mesh-based approaches. The most widely-adopted is the Lattice Boltzmann method (LBM) [61,62]. This method as well as its less widely-used antecedent, Lattice-Gas Automata (LGA) [63,64], are also not without their problems and limitations, however. For example, Galilean invariance is lost in LGA, leading to a spurious density-dependent factor multiplying the inertial term in the momentum conservation equation whose effect cannot be removed [65]. LBM-based simulation of immiscible multiphase fluid flows, on the other hand, tend to suffer from stability problems [66]. An alternative approach is Dissipative Particle Dynamics (DPD) [58,65,67], which has been used to study the rheology of complex fluids [68]. A related but computationally far cheaper approach [69] of much longer standing is Smoothed Particle Hydrodynamics (SPH) [70]. In addition to avoiding all the issues associated with mesh-based methods, SPH has the advantages that it is derived directly from the underlying equations of continuum mechanics and, hence, can be easily extended to any non-Newtonian matter.

In addition to being used to model multiphase fluid systems *via* the two-fluid model [71,72], SPH has been used extensively to model such systems in which the interfaces are explicitly resolved. Many of these ignore surface tension (*e.g.* [73–78]), making them of limited value when interest lies in liquid-liquid systems where it plays a key role in dictating behaviour. Some have, however, sought to include surface tension effects. The first broad approach is that initially proposed by Morris [79], which embeds the continuum surface force (CSF) approach [80] within SPH. This approach is disadvantaged by the need to evaluate the curvature of interfaces, a computationally challenging task that also leads to numerical instabilities. Adams and co-workers [81] addressed this issue, although computational complexity and expense is still an issue. The CSF-based SPH approach has since been extended to systems involving solid surfaces of desired wettability [81,82] and strictly incompressible systems [83–90]. A second broad approach sees SPH combined with the Cahn-Hilliard model [91,92]; this diffuse interface approach is yet to see wider use, however. The final broad approach to including surface tension within SPH is *via* incorporation of inter-particle forces that lead to phase separation. The earliest example of this approach was due to Nugent and Posch [93], whose inter-particle force was inspired by the intermolecular interaction term that arises in the van der Waals equation of state used to close their compressible SPH model. This particular model, which is far simpler and cheaper than the two alternative approaches mentioned above, has seen some use since (*e.g.* [94,95]) along with similar models based on other inter-particle force models, including some inspired by DPD [96,97] along with various other more arbitrary forms [98], including trigonometric [99] and inverse square law [100]. In all these cases, however, strict incompressibility has not been imposed; this has only been done once previously [101] using the trigonometric interaction model.

In this paper, we report incorporation of surface tension within strictly incompressible SPH framework by including a Lennard-Jones (LJ) interaction between particles. The adoption of the LJ interaction to bring about surface tension has the advantage that it mirrors the almost universally used approach to incorporating solid boundaries within SPH, thus opening the way to unifying the treatment of interfaces in SPH. The paper first provides details of the SPH model and how it is parametrized to yield the desired surface tension. The new method is then validated by comparing its predictions to analytical results and experiments for a number of model problems, including deformation of a liquid droplet in a linear shear field and the same for a freely falling droplet in a quiescent continuous phase.

2. The method

2.1. Governing equations

Smoothed-particle hydrodynamics builds on the Navier–Stokes equations expressed in the Lagrangian frame, which for an incompressible, isothermal system are of the form