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Numerical simulation of bubble deformation and breakup under simple linear shear flows
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The deformation and breakup of a single bubble in a liquid undergoing simple shear flow is examined. A coupled level set/volume-of-fluid (CLSVOF) method is used. The scheme is tested by comparing its predictions to previous numerical studies for drop deforming in another immiscible liquid. It is predicted that the bubble breakup requires stronger shear rates than the drop breakup, as already discussed in [1] and [2], references which should be included and discussed by the present authors. Moreover, in certain cases the bubble, unlike the drop, does not attain a steady shape in the proximity of the critical Reynolds number. The results are interesting and novel, but there are too many and serious issues that need to be addressed by the authors, while revising their manuscript and before resubmitting it for further review.

More important issues

1. Page 3: The CLSVOF method is not an interface tracking one (in such methods the interface is calculated simultaneously with the velocity and pressure fields), but an interface capturing method (in such methods the interface is reconstructed after the velocity field is calculated at each time step). Although this is a matter of nomenclature, using the wrong name can be confusing because the interface tracking algorithms, such as ALE, are completely different from CLSVOF.

2. Page 3: The definitions of the dimensionless numbers, *Re*, and *Ca*, are not appropriate. These numbers are defined here using the properties of air (the dispersed material), whereas in the related references, which are used for validation, they are defined using the properties of the continuous phase. This is not so important in the validation tests where viscosity and density ratios between the two phases are set equal to one, but it is important for the new results. In general, the deformation of the inclusion is caused by the forces exerted on it by the fluid in the matrix, and even more so when the inclusion is just an air bubble, which has negligible density and viscosity. Moreover, the capillary number is the ratio of viscous forces over capillarity and the Reynolds number is the ratio of inertia over viscous forces, and the viscous forces are negligible for air in comparison to viscous forces in the liquid.

3. Page 3: The direction of gravity is not specified. Is it acting along the z-direction? If so, see point #9 below. If not, in what direction does it act? In other studies, an air bubble was considered under the action of both gravity and velocity gradient in the same direction, ref. [3] and [4]. Interesting variations arise from the occurrence of a lift force on a deformable bubble. Again, these additional works need to be cited and discussed.

4. Page 4, top: "The singular Heaviside gradient term in the right hand side of equation (5) is a body force…". Although this term arises along with gravity as a body force, in fact, it presumably acts only on the fluid/fluid interface.

5. Page 4: Eq. (7a) determines the evolution of *F*, but the need to introduce *F* in addition to the colour function required by VOF (which is determined by eq. (7b)) is not discussed. *F* was not even mentioned before eq. (7). The authors should also reference previous works where the need for this distinction has been explained.

6. Page 5: "The results shown in Figure 2 verify that our numerical approach can reproduce the same drop breakup behavior presented in [8]." The comparison is only qualitative, not quantitative. One can readily see several differences in all three cases presented. The comparison could be improved either by superimposing the plots or even better by extracting numerical values for the velocity field or the drop shape. The latter may be more involved, which makes addressing issue #7 below even more important.

7. Page 6: It is well-known that interface capturing methods have drawbacks in terms of accuracy, which in this problem could lead to uncertainty or inaccuracy concerning the breakup conditions and the related bubble shapes. Indeed, the topological changes may affect the small characteristic size of the filament formed by the distorted drop or bubble, when it becomes comparable with the grid size. For instance, in their study of two bubbles rising in line (using VOF and Basilisk as a solver), Zang and Magnaudet, ref. [5], found that refinement up to R/272 (i.e. the local grid size is 272 times smaller than the bubble radius) is required in the proximity of the interface to properly capture the topological changes of millimetric bubbles, since only then the characteristic size of the grid is smaller than the average film thickness in typical coalescence conditions. Instead, the authors use R/24 local grid size at the most. Moreover, the Adaptive Mesh Refinement (AMR) technique is usually employed for 3-4 consecutive levels of refinement, at least. In the present work, only two levels of refinement have been used, why? The mesh convergence study seems quite incomplete. A more complete study with figures to verify it should be included in an appendix concerning the mesh and time step independence of the results. The type of time discretization used is not stated; is it explicit and which algorithm is used?

8. Several experimental studies concerning bubbles in shear flow exist (for example, ref. [6]) so alternatively one could compare the numerical results with experiments and avoid the comparison with other numerical results, which are older and may not have achieved convergence due to lack of access to the software and hardware we have today.

9. The following is the most disturbing issue with this study: It is fine to examine conditions leading to bubble breakup or other flow instabilities, but the range of parameter values investigated must correspond to existing materials and achievable flow conditions. The authors have given the governing equations in dimensional form and defined dimensionless numbers incorrectly as stated in point #2, but also assigned them values that may not be physically relevant. Moreover, the bubble size is nowhere stated.

   In the following, calculations are presented of the relevant quantities considering the pair water / air, which is the most often studied, to examine the consistency and relevance of the values used for the parameters and material properties in this study. The surface tension will be given the well-known value of 0.072 N/m. The density ratio at ambient temperature is $\lambda_\rho = \frac{\rho_{air}}{\rho_{water}} = 1.19 \cdot 10^{-3} \approx O(10^{-3})$, while the viscosity ratio is $\lambda_\mu = \frac{\mu_{air}}{\mu_{water}} = \frac{1.8 \cdot 10^{-5}}{8..9 \cdot 10^{-4}} \approx 2 \cdot 10^{-2}$, which is 20 times larger than the one used by the authors. This difference will affect the values of the relevant dimensionless quantities because all of them should include the viscosity of the external fluid. The viscosity ratio obviously plays a significant role because it indicates how resistant the internal phase will be to the deformation induced by the viscous stresses acting on the interface. So, it is imperative to define an alternative capillary number as $Ca = \frac{\mu_m U}{\sigma}$. Using these values for the properties and correctly defining the often used in this study values for Re=92 and Ca=0.3, we find:

$$Ca = 0.3 = \frac{\mu_m U}{\sigma} \rightarrow 0.3 = 8.9 \cdot 10^{-4} \cdot \frac{U}{7.2 \cdot 10^{-2}} \rightarrow U = 24.2 \frac{m}{s}$$

$$Re = 92 = \frac{\rho_m U R}{\mu_m} \rightarrow R = 3.37 \cdot 10^{-6}\, m$$

   The velocity of the plates (V) is calculated as:

$$\dot{\gamma} = \frac{2V}{H} = \frac{U}{R} \rightarrow V = 4U \approx 100\, m/s$$

$$\dot{\gamma} = \frac{2V}{H} \approx 10^7 \left(\frac{1}{s}\right)$$

   This is an extremely high value of shear rate. The velocity of the plates is even less realistic, since we are considering a domain, whose height is just $8R \approx O(10^{-5})\, m$. Moreover, the Froude number, becomes:

$$Fr = \frac{U}{\sqrt{gR}} \approx 4200$$

This large value makes the implied assumption of negligible gravity realistic, because the terminal velocity according to the Hadamard expression is:

$$V_{buoyancy} = \frac{\frac{2}{3} gR^2 \Delta\rho}{\mu_m} * \frac{\mu_m + \mu_b}{2\mu_m + 3\mu_b} = \frac{2}{3\mu_m} gR^2 \Delta\rho \frac{1 + \lambda_\mu}{2 + 3\lambda_\mu} \approx 4.2 \cdot 10^{-5} \frac{m}{s}$$

Since the channel has a width of 8R, the total distance from the surface of the bubble at time zero and the wall is $3R \approx 10^{-5}\, m$. This implies that in about 0.24 s the bubble would touch the upper wall. The dimensionless time defined in the paper is $T = \dot{\gamma} t$ and the calculations stop at $T = 40$, or at a dimensional time of $O(10^{-5})\, s$. This justifies the fact that gravity does not affect the bubble motion, and this should be stated explicitly.

Only a few Newtonian liquids have a viscosity such that $\lambda_\mu = 10^{-3}$. As a second example and in an effort to approach this ratio used by the authors, one could consider ethylene glycol (Perry's Handbook), a liquid that would give, in combination with air at ambient temperature, a viscosity ratio of 10^{-3} and a density ratio of 10^{-3}. Its density is 1100 kg/m^3 and its viscosity is $2 \cdot 10^{-2}\, Pa \cdot s$. Its interfacial tension with air is $\sigma \approx 0.0415\, N/m$. The latter affects the value of the capillary number. So, for the pair ethylene glycol / air, we would have:

$$\rho_m = 1100 \frac{kg}{m^3}, \mu_m = 2 \cdot 10^{-2}\, Pa \cdot s \rightarrow \lambda_\rho = 10^{-3}, \lambda_\mu \approx 10^{-3}, \sigma = 0.0415 \frac{N}{m}$$

Following the above procedure, we find:

$$U = 0.63 \frac{m}{s}, \qquad R = 2.7\, mm$$

The height of the channel would be:

$$H = 8R = 2.15\, cm$$

The shear rate would be:

$$\dot{\gamma} = \frac{U}{R} = \frac{2V}{H} = 233 \left(\frac{1}{s}\right)$$

The velocity of the plates:

$$V = 4U = 2.34\, m/s.$$

All these values are easier to attain. However, the Froude number decreases to:

$$Fr = \frac{U}{\sqrt{gR}} \approx 3.83$$

This is quite close to unity, which means that gravity effects are not negligible anymore. The rising velocity can be calculated via the Hadamard expression:

$$V_{buoyancy} = 0.0012\, m/s$$

The time required to reach the upper wall is $t_{buoyancy} = \frac{3R}{V_{buoyancy}} \approx 7\ (s)$

While the simulations stop at $t_{fini} \quad = 0.18\ (s)$.

So, it seems reasonable to neglect gravitational effects again. However, the authors should specify at least the value of the surface tension, because from two dimensionless numbers one cannot deduce more than two parameters (radius and velocity, or radius and shear rate).

Finally, the Reynolds number of the channel in this case would be:

$$Re_{channel} = \frac{\rho_m 2V_{plate}H}{\mu_m} \approx 6000$$

This high value of Re corresponds to turbulence conditions, see ref. [7] and [8]. Moreover, one can determine the Taylor number for the corresponding flow between two coaxial cylinders, which is the experimental setup used to actually generate this flow. Then the resulting Taylor number $\left(\frac{4\Omega^2 H^4}{\eta^2} \approx 10^7\right)$ is quite higher than the threshold for the well-known vortex instability in circular Couette flow, ref. [9]. This raises more questions with the values of properties and parameters used by the authors.

10. Page 9, Fig. 5 and related discussion: It is quite strange that the bubble does not reach a final steady shape, but seems to expand, contract and then expand again. Here, it is even more important to verify that the simulations have converged (via mesh and time refinement) and to extend them much further in time to determine if this periodic motion will prevail or finally lead to a steady state.

11. Page 13: Fig. 11, which distinguishes the two areas of bubble breakup against just bubble deformation, presents only 4 points and a curve connecting them, although this is an important physical result. Several more points must be included in it to reach a definitive conclusion.

12. End of conclusions section: "*We attribute the large differences in morphology for the bubble undergoing breakup, compared with the drop, to the density and viscosity ratio. The density and viscosity ratio remarkably impacts on bubble/drop deformation and breakup. The bubble deformation and breakup is subject to a synergistic coupling of the density and viscosity ratio, and whose effect will be examined separately in future work*." A similar statement exists in the abstract: "*It is asserted that the differences in morphology for a bubble undergoing breakup, versus a drop in the same process, can be attributed to the density and viscosity ratio of the corresponding two-phase flow systems*." These phrases state the obvious, the densities and viscosities are different between drops and bubbles and, therefore, they cause different dynamics. Instead, it is imperative that the authors give the different physics that the property differences generate. To this end, they should examine thoroughly and in every detail the velocity and stress fields in each arrangement. This may require additional figures and text, but it must be done. Without this examination and its conclusions, this study is more appropriate for a Journal on numerical methods in fluid flows than a physical review Journal. This is the second most important issue with this presentation.

Less important issues

1. Page 2: The symbols for the domain length, width and height are L(x), W(y) and H(z), the variables in parenthesis indicate the direction related to the lengths L, W, and H. However, these are geometric parameters, and could be confused with functions, in the way they are given by the authors here and elsewhere in the text.

2. Page 6: "… depicted in Figure 4(a), uses Ca = 0:3 and Re = 1:1." It should be Figure 4(b).

3. Page 8: "… process is the almost same…" should be "… process is almost the same…"

4. The authors could cite another relevant paper where a similar analysis was undertaken [10]. The main difference is that the present computations are fully 3D, while in this suggested study shear flow until breakup is examined of a 2D bubble. Nevertheless, there are some similarities that should be recognized. The present work is more accurate and extensive.

5. The authors should use D instead of De as the symbol for the parameter introduced by Taylor to determine the extent for bubble deformation, because De stands for the Deborah number.

References

[1] J. R. Grace, T. Wairegi, and J. Brophy, “Break-up of drops and bubbles in stagnant media,” *Can. J. Chem. Eng.*, vol. 56, no. 1, pp. 3–8, (1978).

[2] H. P. Grace, “Dispersion phenomena in high viscosity immiscible fluid systems and application of static mixers as dispersion devices in such systems,” *Chem. Eng. Commun.*, vol. 14, no. 3–6, pp. 225–277, (1982).

[3] D. Legendre and J. Magnaudet, “The lift force on a spherical bubble in a viscous linear shear flow,” *J. Fluid Mech.*, vol. 368, pp. 81–126, (1998).

[4] E. A. Ervin and G. Tryggvason, “The rise of bubbles in a vertical shear flow,” *J. Fluids Eng. Trans. ASME*, vol. 119, no. 2, pp. 443–449, (1997).

[5] J. Zhang, M. J. Ni, and J. Magnaudet, “Three-dimensional dynamics of a pair of deformable bubbles rising initially in line. Part 1. Moderately inertial regimes,” *J. Fluid Mech.*, vol. 920, pp. 1–45, (2021).

[6] N. Müller-Fischer, P. Tobler, M. Dressler, P. Fischer, and E. J. Windhab, “Single bubble deformation and breakup in simple shear flow,” *Exp. Fluids*, vol. 45, no. 5, pp. 917–926, (2008).

[7] B. J. Bayly, S. A. Orszag, T. Herbert, "Instability Mechanisms in Shear-Flow Transition" *Ann. Rev. Fluid Mech*. 20, 359-391, (1988).

[8] S. A. Orszag and L. C. Kells, "Transition to turbulence in plane Poiseuille and plane Couette flow", *J. Fluid Mech*. 96, 159-205, (1980).

[9] N. Tillmark and P. H. Alfredsson, “Experiments on transition in plane Couette flow,” *J. Fluid Mech.*, vol. 235, pp. 89–102, (1992).

[10] Wei, Qian, Xu, “Lattice Boltzmann Simulations of Single Bubble deformation and breakup in Shear Flow:,” *J. Comp. Multiphase flows*, pp. 111–117, (2012).