December 28, 2023

Physical Review Fluids
Professor Eric S. G. Shaqfeh, Editor

Dear Prof. Shaqfeh,

Please find enclosed the revised version of our manuscript (FZ10233): "Numerical Simulation of Bubble Deformation and Breakup under Simple Linear Shear Flows" We thank the reviewers for their comments and thoughts regarding improvement of our paper. We believe that we have addressed all of the reviewers' concerns; the changes are itemized in detail below.

NOTE: all references to equations and figures below are with reference to the numbering scheme of the revised version of the paper, not the original version. Also, changes are hi-lighted in red.

Changes made in response to comments of Reviewer 2:

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.

   Response:
   We agree with the reviewer's opinion. We have changed the "interface tracking" wording to "interface capturing method".
   Please see the 1st line from the top in "Ⅲ. A. Numerical method and governing equations".

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.

Response:
Thank you very much for your suggestion. We have modified the definitions of Re and Ca because we had typos in the definitions of Re and Ca. The computations presented were performed using Re and Ca defined by the density and the viscosity of the surrounding liquid.
Please see Eq.(2).

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.

   Response:
   We have added the direction of gravity in Fig. 1. Regarding the setting of gravity in this study, we will describe it in our response for Question #9.

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.

   Response:
   Following to reviewer's comment, we have provided an additional explanation "The surface tension force expressed by the singular Heaviside gradient term acts only on the gas-liquid interface."
   Please see the 5th line from the top of Page 4.

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.

   Response:
   We agree with reviewer's comment. We have added details of the CLSVOF method at the beginning of Section "A. Numerical method and governing equations."

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.

Response:
Though there are differences between previous studies and our study, we think a major cause of differences is attributable to the grid size (Renardy: $\Delta x = R/8$, our study: Renardy: $\Delta x = R/16$) as we mentioned in the paper. We present a new quantitative comparison, in terms of the Taylor deformation parameter D as a function of Ca, in Table II for bubble deformation. Note that it took several months to obtain additional computational results for bubble deformation. Comparisons of our results against past studies (summarized in Table II) support the validity of our bubble deformation computations.

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?

   Response:
   First, we would like to emphasize to the reviewer that very long computational times are required for drop/bubble deformation and breakup in shear flow. For bubble simulations, in particular, even longer computational times are needed because the time step is much smaller than that for the drop. In our study, time steps smaller than values determined theoretically from various time-step constraints were used: the time step was $O(10^{-4})$ s for the drop and $O(10^{-5})$ s for the bubble. In addition, it is especially difficult to solve stably two-phase flows with λ (density ratio) $\simeq 0$ and η (viscosity ratio) $\simeq 0$. In some cases, computational times over a few months long were required. To overcome the problem of very long computational times, various computational countermeasures were

employed in past studies. For example, in Renardy's study (Phys. Fluids, 2000) a domain size with W (width (y-directional length)) $= 2D$ (D: diameter of drop/bubble) was employed to reduce the number of computational grid points. As shown in Table III in this study, using a width of $W = 2D$ accentuates drop deformation. Regarding the grid size, relatively coarse discretizations have been used in recent studies: $\Delta x = R/48$ (Komrakova et al., CES, 2015), $\Delta x = 2R/15, R/15, R/25, R/30$ (Hernandez and Rangel, Comput. Fluids, 2017), $\Delta x = R/15$ (Amania et al., CES, 2019), $\Delta x = R/30$ (Zhang et al., CES, 2021).

Komrakova et al. (CES, 2015) and Zhang et al. (CES, 2021) used a LBM method for two-phase flows, Hernandez and Rangel (Comput. Fluid., 2017) used a VOF method and Amania et al. (CES, 2019) applied a level set method to computations. Although Komrakova et al. (CES, 2015) set a higher grid resolution ($\Delta x = R/48$, which corresponds to R=48 lattice units in the LBM method), they performed computations over a quarter of the full domain, which is inappropriate for high Re conditions. In the study by Hernandez and Rangel (Comput. Fluids, 2017), it was shown that the grid resolution should be $\Delta x \leq R/15$ for approximately a 5% error. Zhang et al. (CES, 2021) used a higher grid resolution (R=60 lattice units), but the height of the computational domain was $4R$ for 3D-computations. Additionally, the LBM method (Komrakova et al., CES(2015), Zhang et al., CES(2021)) allows the use of higher grid resolutions due to shorter computational times that result from not solving the continuity equation simultaneously; the decreased computational times come at the expense of diminished accuracy.
Our simulations were performed under stringent computational settings, in terms of domain sizes and grid resolutions, selected to guarantee a minimum accuracy in our numerical results. The critical Re numbers found from the bubble simulations presented in this study will provide a minimum standard for future studies. Anticipating future increased computational resources and the development of new numerical algorithms, we have added the following comment: Additional grid refinements may be desirable to perform further accuracy evaluations for bubble deformation and breakup. (see the last line at the section "C. Consideration of domain and grid sizes", Page 9)
Regarding the refinement level of AMR, the use of 3-4 consecutive refinement levels is not always preferred. A small refinement level is rather suitable in terms of stable computations and we found that our mesh setting for this purpose was reasonable.
We briefly described the type of time discretization, the temporal discretization, and the algorithm of our numerical method. Also, we have added information on our paper as a reference to obtain more details about the numerical method used in this study.

8. Several experimental studies concerning bubbles in shear flow exist (for ex-

ample, 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.

Response:
We added a new quantitative comparison for bubble deformation in Table II to verify the validity of our computational results; in the table we compare against the experimental results of Muller-Fischer et al. (Experiments in fluids, 2008) and Rust and Manga (Journal of Colloid and Interface Science, 2002). The new numerical results for bubble deformation indicate that our computations are consistent with past studies.

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................
Finally, the Reynolds number of the channel in this case would be: $Re_{\mathrm{channel}} \simeq$ 6000. This high value of Re corresponds to turbulence conditions................

Response:
As we have stated in response to your comment #2, correct dimensionless numbers based on the density and the viscosity of the surrounding liquid were used in real computations. Regarding physical properties and conditions in our computations, we will give you an example for the case of Ca = 0.3 and Re =93.

Physical properties (a silicone oil has a very similar physical properties with these values.):
ρ_{m} (density of liquid) = 1000 kg/m^3, μ_{m} (viscosity of liquid) = 0.2 Pa·s, σ (surface tension) = 0.025 N/m, R (bubble radius) = 5.0 mm.
From $\Gamma = 2V/H = 2V/6R,\ V = 3\Gamma R = \frac{3\sigma Ca}{\mu_{\mathrm{m}}} = 1.1$ m/s. Also, we obtain Fr (Froude number) $= \frac{\Gamma R}{\sqrt{gR}} = 1.7$.
In this flow system, the effect of gravity may not be completely negligible. However, in our computations, we set $g = 0$ because we wanted to clearly manifest only the effect of density and viscosity ratios. From another perspective, it can be said that we investigated phenomena in space. At the end of the Problem Description section, we added a detailed explanation about physical properties and conditions.
Also, regarding the channel Re number, we obtain $Re_{\mathrm{channel}} = \frac{\rho_{\mathrm{m}} 2VH}{\mu_{\mathrm{m}}} =$ 3300 for a simple linear shear-flow. As expected, $Re_{\mathrm{channel}} = 3300$ falls

within turbulence conditions for simple linear shear-flow of a single phase fluid. It is important to note that the generation of turbulence is suppressed and turbulent eddy dissipation becomes large if bubbles exist in fluid flows. However, as far as we know, there are no conclusive results on turbulence in a simple linear shear-flow which includes bubbles. We predict that the generation of turbulence is considerably suppressed when bubbles occupy a significant portion of the flow in a channel, as in this study. At present, we believe that our computations based on the assumption of laminar flows are reasonable and not unrealistic.

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.

    Response:
    Regarding Question #12, we have made a new figure (shear stress profile) for the case of $Re = 50$ and $Ca = 0.3$. In the case of $Re = 50$, the bubble finally settles into a steady deformed shape. Additionally, we observed a steady deformed bubble at the $Re = 70$ (not presented in this paper). As discussed in detail in the new Section IV.C, in the limit, as the value of Re approaches the critical Re_c condition, the bubble does not maintain its deformed state but instead alternates in an elongation and contraction process. Moreover, the main object of this study is to find Re_c; we have plans for examining bubble periodic deformation with expansion and contraction in future work. In ongoing and upcoming work, we will examine the effect of λ and η on drop breakup for some combinations of λ and η as follows:

    1. $\lambda = 1.0$, $\eta = 1.0$ (done)
    2. $\lambda \simeq 0.0$, $\eta \simeq 0.0$ (this study)
    3. $\lambda = 1.0$, $\eta \simeq 0.0$
    4. $\lambda = 1.0$, $\eta = 0.1$
    5. $\lambda = 0.1$, $\eta = 1.0$
    6. $\lambda = 0.1$, $\eta = 0.1$
    7. $\lambda = 1.0$, $\eta = 100$
    8. $\lambda = 1.0$, $\eta = 1000$

    After we complete the study of the effect of λ and η, we will work on periodic bubble deformation with expansion and contraction. Due to prohibitely long computation times, additional computing resources and improved numerical algorithms are necessary to simulate multiple periodic cycles in bubble expansion and contraction.

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.

Response:
We appreciate the reviewer's opinion, but it is important to keep in mind that at the start of our study there were no guiding initial results that indicated even potential values for critical Reynolds numbers for a bubble. The critical Reynolds numbers found and presented were obtained after extensive and expensive computations. After lengthy computations, we were able to determine the bubble Re_{c} number for 4 Ca numbers. Nevertheless, we believe that a bubble breakup critical curve drawn based on 4 points will be useful for low Ca numbers. Following the suggestion from the reviewer 1, we have added a drop breakup critical curve ($\lambda = \eta = 1.0$) to Fig. 12 (former Fig. 11). The difference in Re_{c} number between the bubble and the drop is clearer in the new Fig. 12. But, the reviewer's opinion is appropriate. In future work, we will determine the Re_{c} for a variety of high Ca numbers to construct a detailed critical curve for a wider range of Ca numbers.

12. End of conclusions section: .... 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

    Response:
    We agree with reviewer's comment. In response, we have added two new images of the shear stress profile for a bubble for two Re conditions in Fig. 8. A discussion of the new results, in terms of the shear stresses acting on the bubble at the two different conditions, is given in Section IV.C.

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.

Response:
We agree with reviewer's comment. We deleted $(x), (y), (z)$ and will simply express the domain length, width and height as L, W, H.

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

   Response:
   We've fixed the error.

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

   Response:
   We've fixed the typo.

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.

   Response:
   The paper by Wei et al. is now cited in the Introduction.

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.

   Response:
   We have changed the symbol from De to D as suggested.

On behalf of the authors,

Mitsuhiro Ohta