April 1, 2024

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. 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 catching this. We have modified the definitions of Re and Ca accordingly; it was an accident that we had previously defined these quantities in terms of the dispersed material. 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 (z direction); albeit, we prescribe zero gravity in our simulations. The rationale for setting gravity equal to zero is given in more detail in our response to item 9. The purpose of our study is to do controlled numerical experiments on the effects of the density and viscosity ratios. We can isolate the dependencies on these two ratios by prescribing zero gravity. Although [3] and [4] do not address the transition to break-up, they address a very much related aspect to bubble motion in shear flow, so now we cite them in the introduction.

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:
   We agree that there are several differences between the previous study by Renardy and Cristini [8] and our present study. We claim that the major cause of the differences is that Renardy and Cristini [8] used a grid size of $\Delta x = R/8$ whereas in our case, the effective fine grid resolution had a grid size of $\Delta x = R/16$. We have made this more clear in the Figure 2 caption.
   We have made *quantitative* comparisons for the Taylor deformation parameter D in Table I (drop deformation). In the revision, we have added another table with *quantitative* comparisons. Please refer to our newly added Table II (bubble deformation). Note that it took 8 months to compute the results reported in our new Table II. Comparisons of our results against past studies (summarized in Tables I and II) support the validity of our bubble (and drop) 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:

In response to the question "In the present work, only two levels of refinement have been used, why?", we added the following text to our revision, (under the heading "selecting the appropriate grid size")
The grid size and adaptive meshing strategy that we adopt is chosen in order to answer the research question as to the conditions which determine whether a bubble in shear flow will break-up or not. In such a case, we must accurately capture the balance of forces with respect to the (non-local) force exerted from the wall driven flow acting against the interfacial surface tension force. The accuracy of the "Critical Reynolds Number" depends on the largest Taylor Deformation parameter D that is supported by the grid (see e.g. Figures 8 and 11). As we report here, we have found that as long as the grid size is fine enough to support a Taylor Deformation parameter $D < 0.95$, then the transition region (i.e. "Critical Reynolds number") (see Figures 3 and 12) will be captured with a tolerance of three percent. The simulation time becomes impractical if we were to try to further improve the "critical Reynolds number" accuracy. A smaller tolerance would necessitate a larger supported Deformation parameter D which would in turn necessitate a higher aspect ratio computational domain, increased droplet surface area at break-up, increased number of time steps, and higher resolution for representing the drop/bubble at its thinnest point.
We make the distinction between our present research, and the research found in the work of Zang, Ni, and Magnaudet[47,48] on predicting the conditions for bubble mergers. Even in the most extreme cases for mergers, the largest Deformation parameter never exceeds 0.4 in [47]. In summary, our gridding requirements necessitate grid points distributed relatively evenly throughout the computational domain when a bubble is stretched to a 0.9 Deformation, whereas in [47] the gridding strategy necessitates a more localized strategy.
In response to the enquiry "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.", we have modified Figure 3 to report results for three different grid resolutions. Figure 3 illustrates that we capture the proper break-up time as the grid is refined, and at the point where the critical Reynolds' number is prescribed. In Figure 3, we point out that the time step was $O(10^{-5})$s for the elongating bubble! ($\Delta x = R/16$) Further refinements reduce the time step according to the surface tension time step constraint (see section III(A)). We now report in the revision the temporal discretization of our method along with the timestep constraint. Since surface tension is discretized explicitly in time, the surface tension time step constraint is much more restrictive than the classical CFL time step constraint of $\Delta x/U$. (again see section IIIA).

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.

   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: $Re_{\text{channel}} \simeq 6000$. 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 $\mathit{\Gamma} = 2V/H = 2V/6R$, $V = 3\mathit{\Gamma} R = \frac{3\sigma Ca}{\mu_{\text{m}}} = 1.1$ m/s. Also, we obtain Fr (Froude number) $= \frac{\mathit{\Gamma} R}{\sqrt{gR}} = 1.7$.
   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 isolate only the effects of the 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 $Re_{\text{channel}} = \frac{\rho_{\text{m}} 2VH}{\mu_{\text{m}}} = 3300$ for a simple linear shear-flow. As expected, $Re_{\text{channel}} = 3300$ 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:
    We have made a new figure (Figure 8: shear stress profile) for the cases of (i) $Re = 50$ and $Ca = 0.3$, and (ii) $Re = 93$ and $Ca = 0.3$. Our new Figure 8 is a part of our new Section IV.C. The purpose of this new material is to gain further insight on the unsteady dynamics of bubble elongation. What we discovered was that for the Reynolds number sufficiently below the critical value, a relatively quick unsteady elongation period gives way to a steady state (with no break up). On the otherhand for Reynolds number *close* to the critical Reynolds number, there is a prolonged, unsteady, elongation period, in which periodic motion is observed and the deformation parameter D is close to one. It is agreed, that the "vacillating" behavior cannot last forever, ultimately (perhaps stochastically!), the bubble will either settle down or break. We assert that regardless of the outcome, this vacillating behaviour will always occur in close proximity to the critical Reynolds' number. In otherwords, regardless of the outcome, we claim that one is assured of being within 3 percent of the critical Reynolds number (see Figure 3). In fact, we hypothesize that there will *always* be "vacillating" behavior if one is sufficiently close to the critical Reynolds number. i.e. given an almost infinite supply of computational resources, as one hones in closer and closer to the critical Reynolds number, a "tug of war" will be observed between the surface tension force trying to pull the bubble together versus the wall driven shear stress trying to pull the bubble apart. This discussion is added to the paper.

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. 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 other reviewer (reviewer 1), we have added a drop breakup critical curve ($\lambda = \eta$ =1.0) to Fig. 12 (formerly Fig. 11). The difference in Re_{c} number between the bubble and the drop is clearer in the new Fig. 12. In future work, we will determine the Re_{c} for a variety of high Ca numbers in order 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