

Research Article

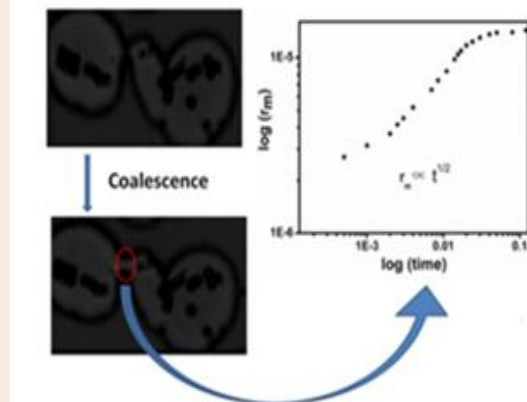
Beads-assisted Investigation of Self-Coalescence of Sessile Water Drops

Qiyu Huang, Lili Zhang, Younian Liu*

College of Chemistry and Chemical Engineering, Central South University, Changsha, Hunan 410083, P. R. China

Abstract

A simple strategy was proposed to investigate the self-coalescence of impurity water droplets at the early stage under quasi-equilibrium conditions. In this strategy, the droplets were allowed to coalesce on a cover glass in a closed cell to eliminate the effects of gravity and evaporation. Moreover, indicator beads were deliberately added into water to construct bead-containing droplets hence tracking the process of the fast coalescence of the water drops. In addition, surfactant was added in this coalescence system to decrease the viscosity of the water, which could slow down the coalescence process, making it possible to be captured by a fast camera. The surfactant was able to induce the formation of asymmetric droplets that are of irregular morphologies. Interestingly, such droplets of asymmetric shapes exhibited similar coalescing process, during which the relationship was obtained between the radius (r_m) of the small bridge formed and time (t) consumed. It follows a scaling law of $r_m \propto t^{1/2}$, similar to that of symmetric coalescence reported by previous researchers.



Keywords: self-coalescence, indicator beads, surfactant, asymmetric droplets

***Correspondence**

Younian Liu,
Email: liuyounian@csu.edu.cn

Introduction

When two liquid drops come into contact at a negligible velocity, surface tension effects is accepted in control of the initial character of flow motion[1]. Meanwhile, van de Waals force drives them into shapes with smaller surface area, during which a tiny bridge are consequently formed [2, 3]. The study of coalescing behaviors of fluid droplets is critical in understanding some industrial processes such as printing, sintering, etc.[4-6] and natural phenomena[7-11]. Note worthily, the early-time behavior of coalescence between two droplets is of particular interest, especially for the formation of the bridge, whose radius change with respect to time has received tremendous attention. Many theoretical and experimental studies on coalescence were concentrated on evolution of the r_m . For example, Eggers et al. have studied viscous regime, within which Reynolds number $Re \ll 1$ and then the r_m was shown to follow the scaling law of $r_m \propto t \ln(t)$. However, in the inviscous regime, Eggers pointed out that the r_m grows much faster and follows a scaling law of $r_m \propto t^{1/2}$ [12, 13]. Their results were confirmed later by Duchemin through extensive simulations.[5] In addition, they extended the conclusions to three dimensional situations providing that r_m was much smaller than the droplet radius (R). In fact, the results would vary in terms of different experiment situations such as velocity and contact angle of the droplets, the substrates utilized for the droplets to grow and etc.[14-16]. Herein, it is worth noting that most experimental studies were restrained in high-viscosity liquids due to their relatively low motion rate and moreover they mainly concentrated on symmetric droplets. Less attention has been paid to visualize the self-coalescence of low viscous droplets and asymmetric droplets[1, 17-19]. Meanwhile, it still remains a challenge to get some coalescing details such as motion direction and collision force, especially for low-viscosity fluids due to their relative high coalescence rate[20]. Hence, it is essential to develop simple strategies which can facilitate the observation of coalescence process of low-viscosity droplets, particularly for the less-studied asymmetry coalescence.

Herein, we proposed a simple approach via adding beads as indicators into water for studying the coalescence process. Through this method we successfully observed the evolution of the bridge between two adjoined drops lying on horizontal glass surface. Moreover, the collision force which directly drove the coalescence between free drops was also estimated by recording the motion of indicator beads. Some surfactants were subsequently added to optimize the observation of the coalescence process by slowing down the coalescence behavior. Notably, the added surfactants can also lead to the occurrence of asymmetric water drops, making available the study of asymmetric droplet coalescence.

Experiment

The objective droplets were generated in a closed sample cell to prevent the effect of evaporation (see **Figure 1**). The sample cell consists of two cover glasses with a thickness of 130-160 μm (Fisher 12-542-B22X22-1), which was modified to be hydrophilic. A piece of parafilm (Fisher: 100 μm in thickness) with a fine cylinder hole ($\phi=1$ cm) was laid on the first cover glass. Spherical beads with a diameter of 3 μm were chosen as indicator to indicate the direction of water flow and estimate the collision force qualitatively. Beads suspension of 100 μL (Polybead Amino, CAT#17145, Polysciences, Inc.) was washed by deionized water (MilliQ system, 18.2 $\text{M}\Omega$ cm) for three times according to centrifuge-ultrasonic method[21] before adding into 10 mL deionized water. One drop of bead-containing water (100 ~ 200 μL) was injected into the center of the hole, then the second cover glass was set over the parafilm. The edges of the cell were carefully glued and a light source was placed under the cell to provide bright view. All experiments were done at room temperature.

A fast CCD camera (FastCam super 10K with Kodak Motion corder analyzer Ps-220, Video Kommunikation GMBH) was mounted on the microscope to record the details of the self-coalescence process. To ensure a high image resolution at high recording rate, a light was set under the cell to provide high-bright field. The maximum camera speed of the setup is 3000 pictures/s.

Result and Discussion

The experimental difficulties involved in this type of studies lie in the control of the liquid droplets to contact against each other freely and slowly, and keeping the whole coalescence motion within the exact focal plane of a high-resolution imaging device. To fulfill such purposes, two sessile water droplets were located horizontally between two cover glasses, as shown in **Figure 1**.

This approach allows us to investigate the coalescence of sessile droplets in a simple way. Because of the hydrophilic character of the glass, the boundary of the added droplet started to expand slowly and split into many tiny drops and they began to coalesce thereafter. In this case, we could get an insight into the coalescence process of both symmetric and asymmetric droplets.

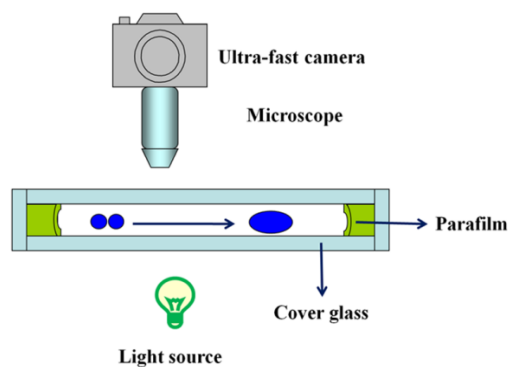


Figure 1 Setup used to approach the free coalescence

Coalescence between symmetric droplets

It is well known that the fusion process of two water drops is extremely fast due to their relatively low viscosity. High speed camera is necessarily required to capture the coalescence process. **Figure 2** shows the evolution of the coalescence between two symmetric water droplets recorded at a speed of 30 pictures/s, *i.e.*, the two droplets approached mutually, came into contact (see **Figure 2a**), and started to coalesce (see **Figure 2b-2d**). At the early stage of the coalescing process, all drops kept their original shapes and all indicator beads stood still at their original positions (see **Figure 2b**). Interestingly, the droplets became smaller with three beads rushing to the bridge (see **Figure 2c**). It seems that the mass flow moved from the smaller droplet to the bigger one through the bridge labeled with an arrow in **Figure 2b**. This flow hit the nearest beads in the big drop but was not able to push them away. Therefore, some beads had to concede to get away from the whole aggregates to release the collision energy (see **Figure 2c**). On the other hand, the three separated beads inclined to move backward to reduce the collision energy from the assault of the mass flow (see **Figure 2d**).

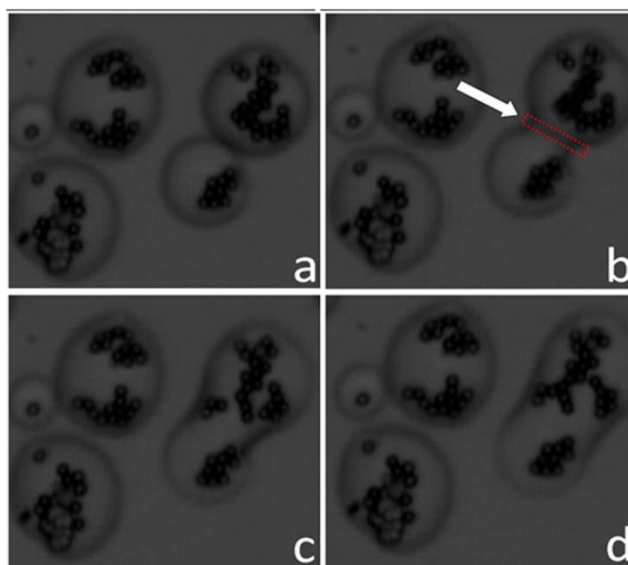


Figure 2 Evolution of coalescence between two symmetric drops at a recording speed of 30 pictures/s, time runs in alphabetical order

To obtain better time resolution, the position resolution had to be sacrificed. **Figure 3** shows a similar coalescence process recorded at a speed of 250 pictures/s. In such a recording situation, the droplets looked more like two dimensional with unclear boundaries. Focus adjustment did not work well in getting a clear drop boundary but fortunately we could still concentrate the light on the beads very well. Actually, the images were the same as those showed in **Figure 2** if the recording speed was turned back to 30 pictures/s. In detail, the coalescence occurred and the beads near contacting interface between the droplets interacted mutually and quickly as shown in **Figure 3b**. It is clear that bead 1 moved upward for a certain distance. Noticeably, only one bead should be situated in either position 5 or 6 since the total number of beads is constant during the interacting process. However, beads could not be found in either position 5 and 6 in the view of microscope. We suppose it was due to the ultra fast moving of bead 2 between position 5 and 6. Therefore, bead 3 and bead 4 might derive from bead 7 and bead 8, respectively. Then all the beads near the bridge were settled down at 8 ms as shown in **Figure 3c**. Obviously, the backward moving speed of bead 1 was relatively slow on account of the fact that they moved shorter distance within given time period (see **Figure 3a-3b** and **3d-3e**). However, the recording speed of 250 pictures/s can still not meet the requirement of a qualified analysis. Even though a camera speed of 3000 pictures/s was equipped for more precise study of coalescing process, the time resolution was still not high enough to afford a high position resolution (see **Figure 4**). Hence, surfactant SDS (sodium dodecyl sulfate) (0.01mg/mL) were added to decrease the surface tension and slow down the

evolution speed of two coalescing droplets to get detailed information about the forming of the bridge. Moreover, since our beads are of negative charge, negative-charged SDS could prevent the beads from aggregation. Interestingly, in the presence of the surfactant, many asymmetric droplets appeared on the surface of the cover glass.

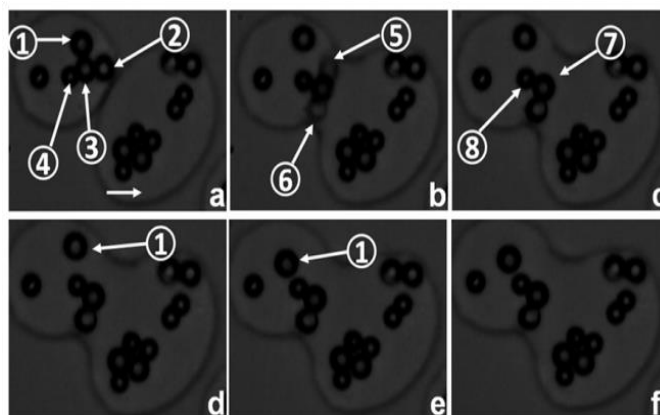


Figure 3 Evolution of coalescence recorded at 250 pictures/s. The time points in alphabetical order are 0, 0.004, 0.008, 0.024, 0.124, and 0.200, respectively

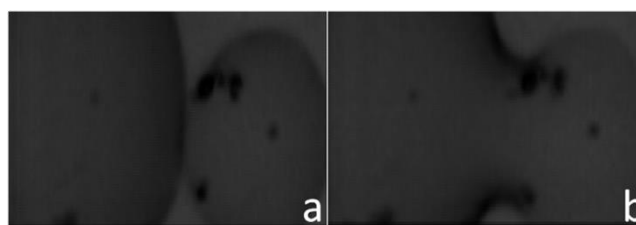


Figure 4 Evolution of coalescence between two symmetric drops at a recording speed of 3000 pictures/s

Coalescence between asymmetric droplets

A typical evolution of the coalescence between asymmetric droplets is shown in **Figure 5**. At a recording speed of 30 pictures/s, a bridge marked with an arrow could be found at the beginning of the coalescence, which was similar to that in symmetric coalescence (see **Figure 5b**). **Figure 5e** shows the detailed information of those beads in transitional state. The extra two bunches of beads were believed to come from the over-recording of a fast moving delayed by a slow camera. Although it has been reported that droplet size and geometry could exert some influence on the coalescence, it still remains a challenge to illustrate why asymmetric droplets coalescence can be better resolved by a fast camera in details[22-23]. **Figure 6** shows an asymmetric coalescence process recorded at the speed of 2000 pictures/s. The droplets firstly connected with each other through a thin bridge. With the continual growth of the bridge, the arrow-marked beads close to the bridge started to move following the mass flow (see **Figure 6e**), whereas it was not the same case for those away from the bridge. The time-dependent growth of the bridge width was listed in **Figure 7**. Although a symmetric coalescence between two droplets grew much faster than that of asymmetric one, the morphological evolutions were similar, *i.e.*, both of them began with forming a tiny bridge and the mass flow moved unidirectional until the growing of the bridge finished. Moreover, other than the contacting area (bridge), the rest of the droplets morphology stayed almost unchanged, during the early period of the coalescing process. This suggests that the local shape of the contact area may exert great influence on the dynamic coalescence behavior[24].

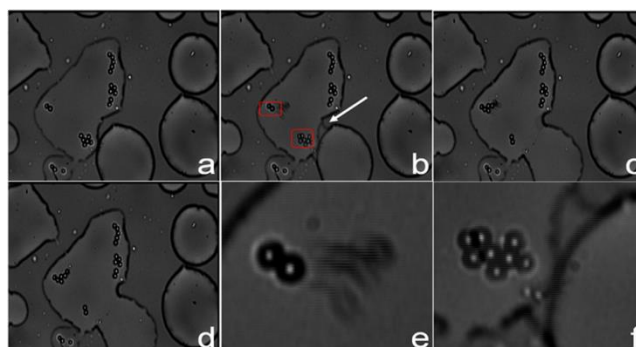


Figure 5 Evolution of coalescence between two asymmetric drops at a recording speed of 30 pictures/s, recording time from a to d: 0s, 0.033s, 0.067s, 0.333s, respectively; e and f show the enlarged views of marked zone in b, e for the top-left one and for the bottom-right one

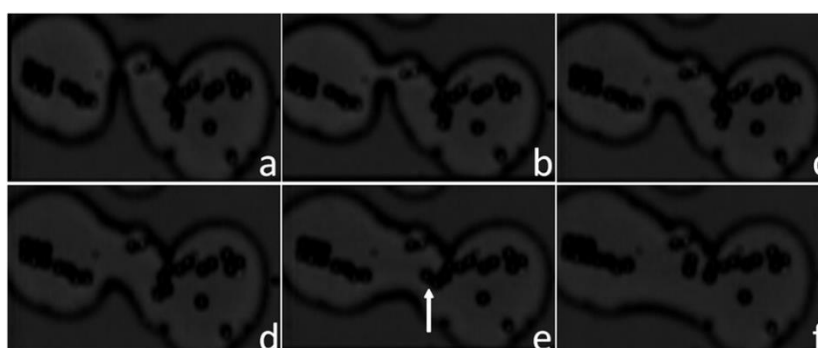


Figure 6 Evolution of coalescence between two asymmetric drops at a recording speed of 2000 pictures/s, time runs in alphabetical order

Finally, the relationship between r_m and t was investigated. As mentioned above, most of the studies on the scaling law of $r_m t$ were for symmetric droplets. In this essay, we present experimental study on the dependence of r_m on t for asymmetric droplets. As shown in **Figure 7**, the $\lg(r_m) - \lg(t)$ curve (see **Figure 7b**), a linear relationship can be obtained from point 3 to 13 with a scaling law of $r_m t^{0.4995}$. This agrees with other studies for symmetric droplets coalescence. However, at the beginning stage as well as the final stage the points did not fit the scaling law. For the last period of the coalescence process, the condition of $r_m \ll R$ is broken with the coalescence growing, hence it will not follow the situation described in the previous studies. On the other hand, at the very beginning, neither of the first two points agreed with the equation, which could be owing to that the 0s we defined was not the real starting point. Although the deviation could occur in the first few points, it can be neglected when time scale is large enough.

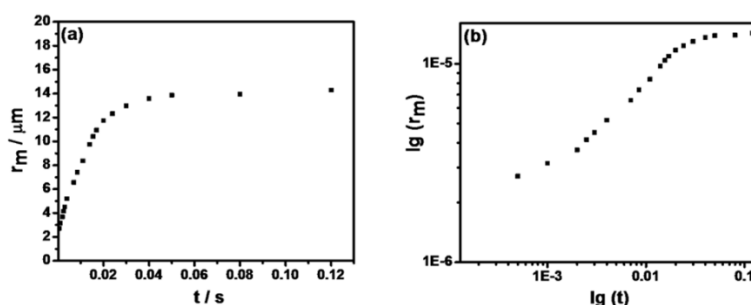


Figure 7 Dynamic curve of r_m via t according to Figure 6

Conclusion

In conclusion, a simple strategy was proposed to investigate the early-time behavior of the water droplets self-coalescence. In this strategy, beads were added to assist the study of the dynamic coalescence process. These beads can act as indicators to visualize the motion direction of the liquid and evaluate the collision force in the coalescence process. Furthermore, surfactant (SDS) is used to slow down the coalescence process and induce the formation of asymmetric droplets. Interestingly, the functional relationship between r_m and t follows $r_m \propto t^{1/2}$, which is similar to that of previously reported symmetric droplets. Finally, it is a promising strategy to investigate other liquids and for further study on later stages of coalescing behaviors.

Acknowledgements

The author acknowledges Prof. Yi Zhang for financial support and insightful discussion.

References

- [1] Menchaca-Rocha, A. Martínez-Dávalos, R. Núñez, S. Popinet, S. Zaleski, *Phys. Rev. E.*, **2001**, 63, 046309.
- [2] J. Eggers, *Zamm-Z. Angew. Math. Me.*, **2005**, 85, 400-410.
- [3] M. Wu, T. Cubaud, C.-M. Ho, *Phys. Fluids*, **2004**, 16, L51.
- [4] J. S. Eow, M. Ghadiri, *Chem. Eng. J.*, **2002**, 85, 357-368.
- [5] L. Duchemin, J. Eggers, C. Josserand, *J. Fluid Mech.*, **2003**, 487, 167-178.
- [6] K. Ahn, J. Agresti, H. Chong, M. Marquez, D. A. Weitz, *Appl. Phys. Lett.*, **2006**, 88, 264105.
- [7] O. Navon, V. Lyakhovskiy, *Vesiculation processes in silicic magmas*, Geological Society, London, Special Publications, 1998, 145, p27-50.
- [8] S. K. Cho, H. J. Moon, C. J. Kim, *J. Microelectromech. Syst.*, **2003**, 12, 70-80.
- [9] N. Garnier, R.O. Grigoriev, M. F. Schatz, *Phys. Rev. Lett.*, **2003**, 91, 054501.
- [10] P. Paik, V. K. Pamula, R. B. Fair, *Lab Chip*, **2003**, 3, 253-259.
- [11] S. Espino, H. J. Schenk, *J. Exp. Bot.*, **2011**, 62, 1119-1132.
- [12] J. Eggers, *Phys. Rev. Lett.*, **1998**, 80, 2634-2637.
- [13] J. Eggers, J. R. Lister, H. A. Stone, *J. Fluid. Mech.*, **1999**, 401, 293-310.
- [14] H. J. Maris, *Phys. Rev. E.*, **2003**, 67, 066309.
- [15] R. Ishiguro, F. Graner, E. Rolley, S. Balibar, *Phys. Rev. Lett.*, **2004**, 93, 235301.
- [16] S. Karpitschka, H. Riegler, *Langmuir*, **2010**, 26, 11823-11829.
- [17] D. G. A. L. Aarts, H. N. W. Lekkerkerker, H. Guo, G. H. Wegdam, D. Bonn, *Phys. Rev. Lett.*, **2005**, 95, 164503.
- [18] W. Yao, H. J. Maris, P. Pennington, G. M. Seidel, *Phys. Rev. E.*, **2005**, 71, 016309.
- [19] J. Hernández-Sánchez, L. Lubbers, A. Eddi, J. Snoeijer, *Phys. Rev. Lett.*, **2012**, 109, 184502.
- [20] S. G. Bradley, C. D. Stow, *Philos. Trans. R. Soc. London, Ser. A, Mathematical and Physical Sciences*, **1978**, 287, 635-675.
- [21] Léaustic, R. E. Riman, *J. Non-Cryst. Solids*, **1991**, 135, 259-264.
- [22] J. Kamp, M. Kraume, *Chem. Eng. Res. Des.*, **2014**, 92, 635-643.
- [23] Eddi, K. Winkels, J. Snoeijer, *Phys. Rev. Lett.*, **2013**, 111, 144502.
- [24] J. D. Paulsen, R. Carmigniani, A. Kannan, J. C. Burton, S. R. Nagel, *Nat. Commun.*, **2014**, 5.

© 2015, by the Authors. The articles published from this journal are distributed to the public under "Creative Commons Attribution License" (<http://creativecommons.org/licenses/by/3.0/>). Therefore, upon proper citation of the original work, all the articles can be used without any restriction or can be distributed in any medium in any form.

Publication History

Received	15 th Dec 2014
Revised	13 th Jan 2015
Accepted	24 th Jan 2015
Online	15 th Feb 2015