



HAL
open science

Relaxation and contact angle dynamics during the coalescence of different sized vertically aligned water drops in different silicone oil viscosities

Mostafa Brik, Souad Harmand, Ibrahim Zaaroura

► To cite this version:

Mostafa Brik, Souad Harmand, Ibrahim Zaaroura. Relaxation and contact angle dynamics during the coalescence of different sized vertically aligned water drops in different silicone oil viscosities. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 2021, 629, pp.127429. 10.1016/j.colsurfa.2021.127429 . hal-03534128

HAL Id: hal-03534128

<https://uphf.hal.science/hal-03534128>

Submitted on 16 Oct 2023

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial 4.0 International License

Relaxation and contact angle dynamics during the coalescence of different sized vertically aligned water drops in different silicone oil viscosities

Mostafa Brik*, Souad Harmand, Ibrahim Zaaroura

LAMIH UMR CNRS 8201 - Département de Mécanique, UPHF, Le Mont Houy, Valenciennes, France
mbrik@uphf.fr

Abstract

This paper reports an experimental investigation for the impact of the head drop size as well as the outer phase viscosity on the coalescence time of two water drops vertically aligned in silicone oil. Two types of silicone oil with different viscosities have been used in this work (47v350 and SilOil M40.165). A specific configuration of a sessile drop in direct contact with another drop placed above has been studied. The coalescence time is depending mainly, in this work, on the relaxation of the resulted drop together with its contact angle evolution during the merger process. For both liquid-liquid (LL) systems, the first stage of the coalescence process was dominated by the inertial forces induced by capillarity where the redefined Reynolds number Re is found to be much greater than the unity ($Re \gg 1$). It's depicted that the smaller the head drop volume the faster the merger process. The viscosity of the outer phase has also found to have a main impact on the coalescence time and therefore on the relaxation of the resulted drop as well as its contact angle. It's found that increasing much more the outer viscosity provokes a long coalescence time accompanied with an elimination of the harmonic movement characterizing the merger drop relaxation as well as a damping of the contact angle dynamics. Focusing on the first instants of the merger process where a liquid bridge linking the drops is formed, it's shown that for the different head drop volumes used in the study that the variation of the dimensionless capillary pressure P^* in function with the dimensionless time t^* is the same for each LL system and it follows a power scaling law presented in the last section.

1. Introduction

With the high development of technologies in the recent decades, drops dynamics is seen to be involved in different industries. From the micro to the large scale of industrial applications, numerous researches have been carried out around this subject. In biology and micro-biology, mixing substances inside drops which are considered as micro-reactors is used to accelerate the validation of physical properties of new molecules [1-4]. Recently and with the evolution of the pandemic situation of COVID-19, the dynamics of respiratory drops in evaporation is utilized to determinate their drying time on infected subject to better understand and control the corona-virus circulation [5]. Drops dynamics and coalescence is amply present as well in pharmaceutical, cosmetics as well as food industries where the emulsification processes are primordial [6-10]. In petroleum engineering, researchers have focused on the study of the droplets coalescence and how to accelerate the destabilization of the water drops-in-oil emulsion [11-20]. The coalescence process is complex and could incorporate more and more physics that could impact its dynamics (heat and mass transfer, electrostatics, electromagnetic...etc). The first instants of the initially stationary drops merger process are generally characterized by the building of a capillary liquid bridge where its formation regime is determinate by a scaling law [21-24]. Numerous factors could impact the development of the capillary liquid bridge. For free surfactant gradient, the increasing in the outer phase viscosity is found to decelerate the formation of the liquid bridge while maintaining a symmetrical coalescence [25-26]. On

the other side, researcher [25] found that in the presence of a surfactant gradient between two interfaces, the coalescence becomes asymmetrical. Focusing on the coalescence of a sessile drop with a same fluid drop placed above (vertically aligned water drops) in silicone oil, Brik et al., [27] proposed an experimental and numerical investigation for the impact of the outer phase viscosity on the liquid bridge building as well as the development speed and the external capillary pressure generated during the formation of the bridge. For a deposited sessile drop where the measured contact angle was about $\theta = 89^\circ$ they illustrated that for a given dispersed phase viscosity, the more the surrounding viscosity is large, the lower the rate of the liquid bridge growth, the lower the earlier radial velocity of the bridge, and the higher the external capillary pressure generated around the bridge. For gas-liquid systems, Gungen et al., [28] studied numerically the impact of the superhydrophobic texture of the surface on the recoiling of the resulted drop during the coalescence of vertically aligned water drops in air. Somwanshi et al., [29] have studied the coalescence dynamics of vertically aligned water drops in air over a superhydrophobic surface. Researchers [29] illustrated that for unequal drops recoil is seen earlier but with a small recoil height. Furthermore, the relaxation of the resulted drop took particular attention due to its importance on the study of the dynamics of the contact line and its impact on the convection generated inside the coalesced drop and so the enhancing performance of the mixing [30-31]. Zheng et al., [32] studied the relaxation of the liquid bridge formed during the merging of two initially stationary sessile drops in air and deposited on an organic glass surface where they used water with different quantities of glycerin in order to investigate the impact of the dispersed phase viscosity. They found that the liquid bridge undergoes a harmonic damped movement. They illustrated as well that the more the dispersed phase viscosity, the more the damping effect due to the viscous dissipation. For the same configuration and the most principle for changing the working fluid viscosity (adding glycerin to water), Lee et al., [33] proved that for low and moderate viscosities the high inertia of the impacting droplet provokes the most spreading of the coalesced drop. For the glycerin drops (very viscous working fluid), researchers found that the spreading length doesn't change appreciably because of the huge resistance generated by the dispersed phase viscosity. Khandekar et al., [34] studied more deeply the relaxation phenomena of vertically aligned drop in air where the sessile drop was deposited on a superhydrophobic surface. In such gas-liquid systems, self-propulsion of the resulted drop could be obtained under surface texture and initial velocity conditions [35-38]. Nowadays, the electronic devices which are more and more compact are found to release huge quantities of heat and an efficient cooling and thermal management for such devices is crucial to avoid their deterioration. For this purpose, fluid drops-in-dielectric oil emulsions are used. In such application, the specific configuration of two vertically aligned drops where one is deposited on a surface is more likely to be encountered. For that, in order to contribute to the understanding of the coalescence process where the first stage is dominated by inertial regime ($Re \gg 1$) which is widely encountered in different thermal applications as well as petroleum and pharmaceutical engineering, we propose herein an experimental investigation for the impact of the drop size as well as the viscosity of the outer phase, on the coalescence time and so the relaxation and the contact angle dynamics during the merging process between vertically aligned water drops in different silicone oil viscosities; silicone oil "SilOil M40.165" and silicone oil "47v350". At first, we investigate the impact of the head drop volume on the time spent to a complete stabilization of the resulted drop. Then, the impact of the outer phase viscosity is studied by comparing the behavior of the relaxation of the resulted drop as well as the dynamics of its contact angle and the evolution of the drop tip velocity when used two different viscosities of silicone oil. At the end, we present a scaling law, observed for the LL systems studied in the work, characterizing the variation of the dimensionless capillary pressure P^* of the liquid bridge in function with the dimensionless time t^* .

2. Methods

In this work, we've investigated the influence of the drop sizes on the dynamics of the coalescence of vertically aligned water drops submerged in silicone oil. As cited above, the first stage of the merger process is dominated by inertial regime for both LL studied systems ($Re \gg 1$). Physical properties of the fluids used in the different experiments are presented in table 1. The different densities and viscosities of the distilled water and silicone oils are obtained from the technical details of the products, while the surface tensions are obtained at a temperature $T \approx 25^\circ\text{C}$ from our lab experiments using the KRÜSS advance system.

Fluids	Density [kg/m^3]	Viscosity [$\text{Pa}\cdot\text{s}$]	Surface tension [N/m]
Distilled water	1000	0.001	-
Silicone oil M40.165	920	0.01	≈ 0.038
Silicone oil 47 v 350	970	0.35	≈ 0.037

Table 1: physical properties of different fluids used in the study

Table 2 summarizes the details (initial radiuses of the head drops and their corresponding capillary pressures) of the LL systems studied in this work.

Liquid-Liquid system	R_{head} [mm]	$R_{s-eq} \left(\frac{R_1^2 + H^2}{2H_1} \right)$ [mm]	$P_{head} (2\sigma/R_{ab})$ [Pa]
Water drops in silicone oil SilOil M40.165	0.5 (0.53 μl)	1.5	152
	1 (4 μl)		76
	1.56 (16 μl)		48,72
Water drops in silicone oil 47 v 350	0.5 (0.53 μl)	1.5	148
	1 (4 μl)		74
	1.56 (16 μl)		47,43

Table 2: initial drops radiuses with their corresponding capillary pressures

In the table "head" and "s - eq" refer to the head drop and the equivalent sessile drop radius, respectively.

3. Experimental setup

Figure 1 illustrates the experimental configuration used to study the coalescence of a sessile water drop with another drop placed above. Both drops are immersed in a cube fill of silicone oil ($80mm \times 80mm \times 20mm$) where the temperature of the system is found to be varied between 23 and 25°C in different experiences carried out in this study. Focusing on this slight temperature variation, no impact is remarked on the obtained results. Using the KRÜSS Advance system, the first drop is generated precisely with one pumping before directed and getting contact with the bottom surface of the cube to form a sessile drop where the measured contact angle is $\theta = 78^\circ \mp 5^\circ$, the spread radius is $R_1 \approx 1,45mm$ and the height is $H = 1.1mm$ which gives an equivalent radius $R_s \approx 1.5mm$. After waiting a while to guarantee that the sessile drop reaches its equilibrium shape (due the fact that the drop is surrounded by oil, its equilibrium takes a couple of milliseconds), the second droplet is then generated and directed very slowly to get in contact with the sessile drop. The reason for bringing the head drop with such a very low speed is to avoid the impact of the velocity approaching on the coalescence. Coupling the KRÜSS system with a high-speed camera (Keyence VW-9000E), the coalescence

process is directly recorded at a frame rate 10000s-1. In order to check the reproducibility of the experimental results, experiments have been repeated three times.

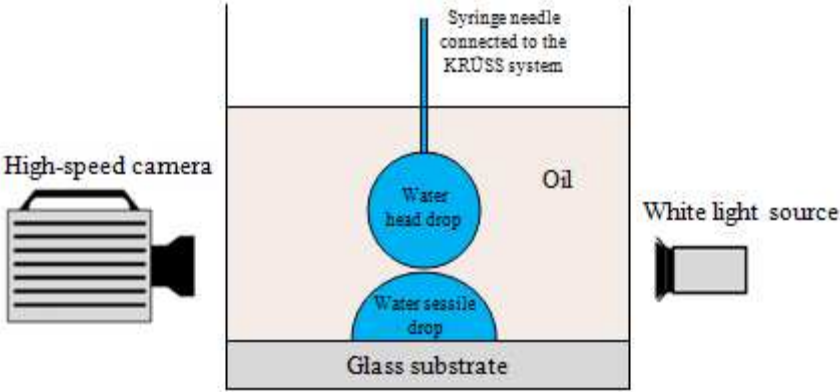
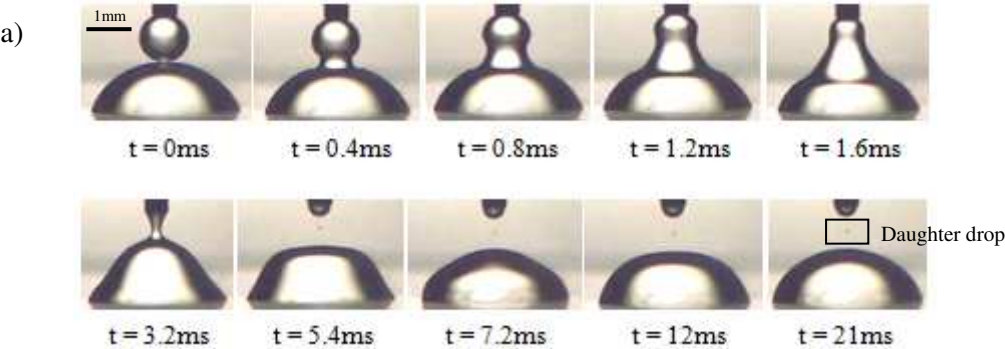


Figure 1: schematic illustration of the coalescence experiment

4. Impact of the head drop radius

Figure 2 presents experimental snapshots for the coalescence between vertically aligned drops on a bottom glass surface of a cube filled with silicone oil M40.165 for different head drop sizes. It's observed from these first series of experiments that the coalescence process is faster for small sized head drops comparing to the cases where the head drops are larger. This is caused mainly to the time spent by the coalesced drop for its relaxation before stabilizing in its final shape as discussed bellow. In the different results presented hereafter, the relaxation time of the coalesced drop is presented as the time variation of the drop tip position. From figures 2a, 3a and 4a, It's illustrated, respectively, that for the small head drop volume ($V_{head\ drop} = 0.54\mu l$) the resulted drop spends almost 0.02s before stabilizing in its final shape which is faster comparing to the case where the head drop volume is equal to that of the sessile one ($V_{head\ drop} = 4\mu l$) where the time is found to be nearly to 0.05s. When the head drop volume is significantly larger than that of the sessile drop ($V_{head\ drop} = 16\mu l$) it's observed that the resulted drop takes much more time to stabilize in its final shape, about 0,15s, which is remarkably greater than other previous cases. This time is spent mainly by the resulted drop in its relaxation around its final stabilized shape as shown in figure 2b, 3b and 4b, respectively. From the latter, it's remarked that in the first instants the drop tip is kept at the same position and doesn't vary with time. This is observed in all the other series of experiments and refers principally to the fact that the resulted drop stays sticking on the needle tip for the earliest instants of the drops coalescence. These instants are generally characterized by the formation of a liquid bridge linking the drops.



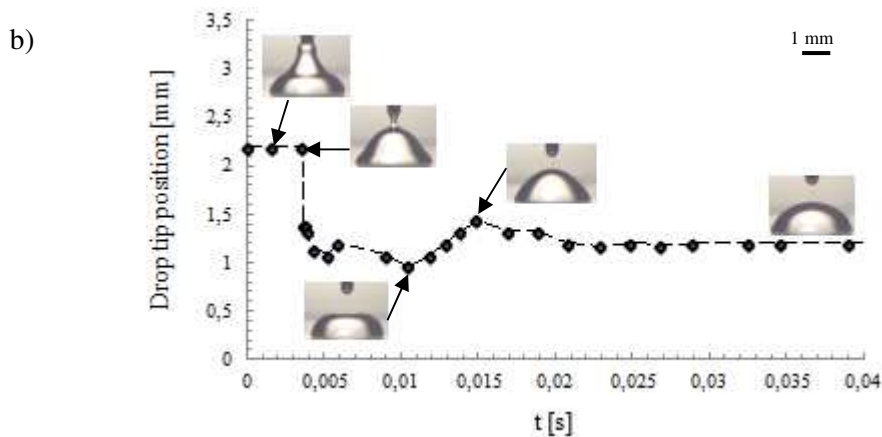


Figure 2: a) experiments snapshots for the coalescence of water drops in silicone oil (SilOil M40.165);
 b) time variation of the drop tip position of the coalesced drop. $V_{sessile\ drop} = 4\mu l$ ($R_{eq} = 1,5mm$);
 $V_{head\ drop} = 0.54\mu l$ ($R_{h-d} = 0,5mm$).

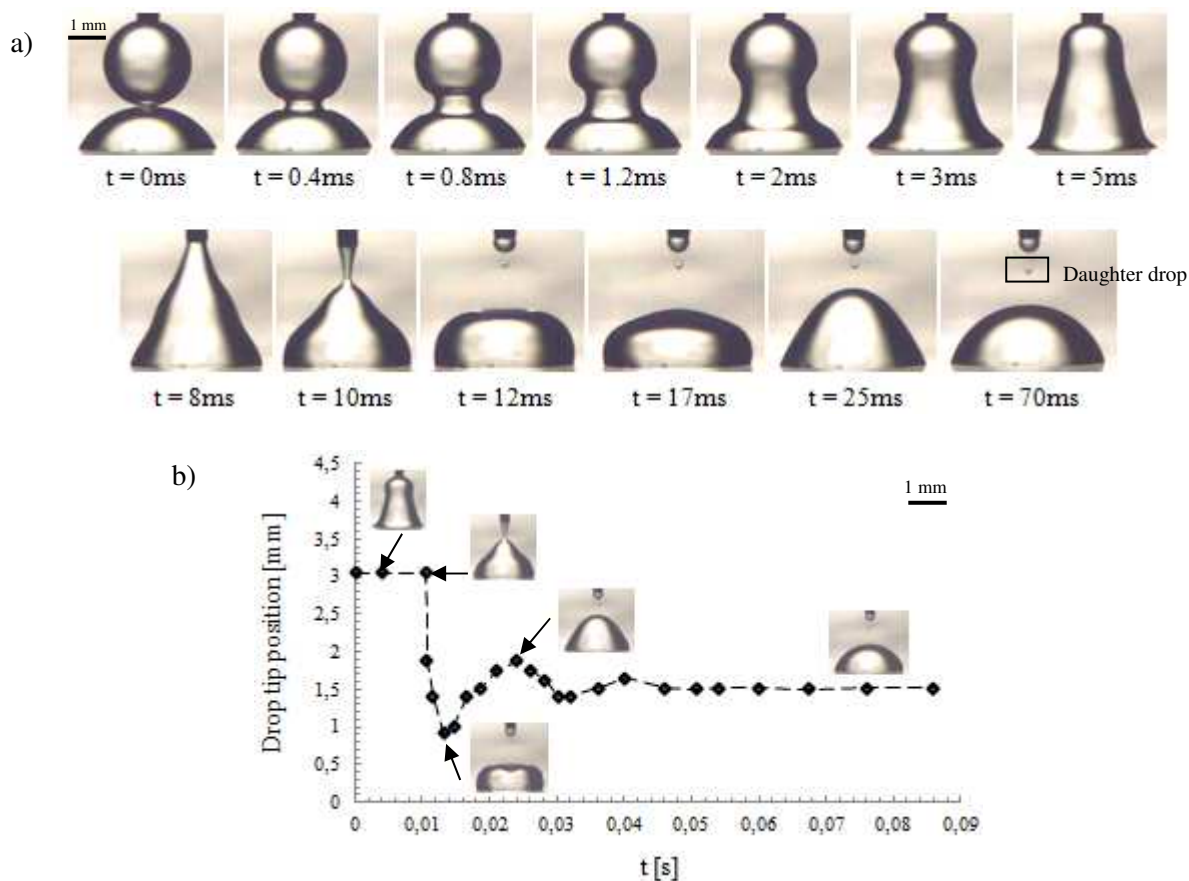


Figure 3: a) experiments snapshots for the coalescence of water drops in silicone oil (SilOil M40.165);
 b) corresponding time variation of the drop tip position of the coalesced drop. $V_{sessile\ drop} = 4\mu l$
 ($R_{eq} = 1,5mm$); $V_{head\ drop} = 4\mu l$ ($R_{h-d} = 1mm$).

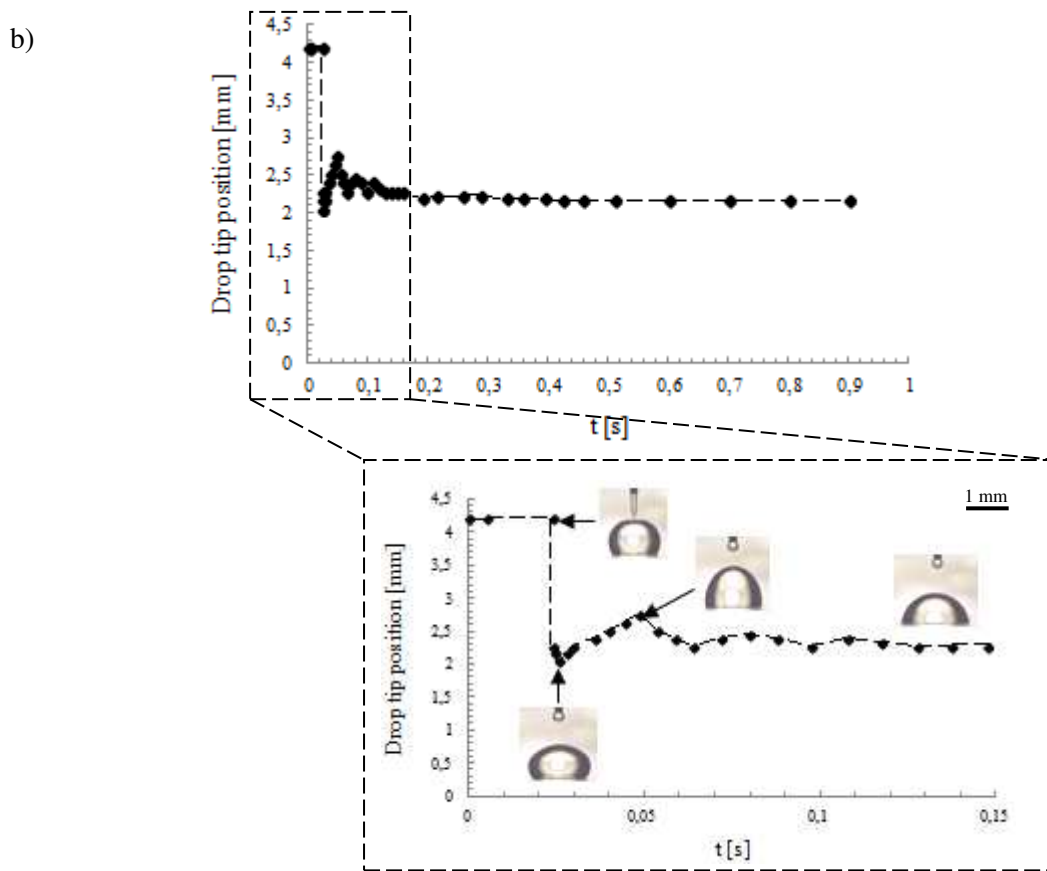
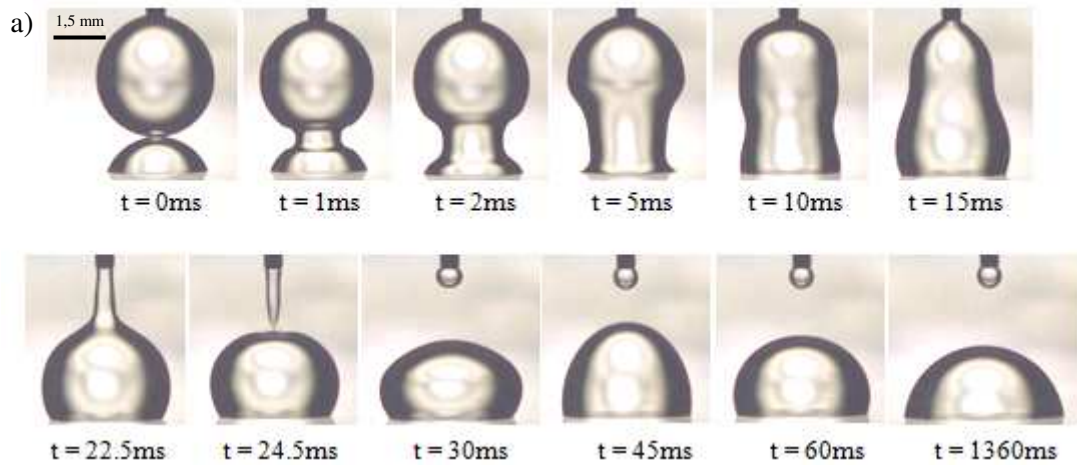


Figure 4: a) experiments snapshots for the coalescence of water drops in silicone oil (SilOil M40.165);
 b) time variation of the drop tip position of the coalesced drop. $V_{sessile\ drop} = 4\mu l$ ($R_{eq} = 1,5mm$);
 $V_{head\ drop} = 16\mu l$ ($R_{sd} = 1,56mm$).

After a complete building of the liquid bridge, the capillary wave starts propagating which provokes later the detachment of the resulted drop from the needle tip. But just before, the later keeps linked to the drop by a liquid satellite as shown in different figures bellow. It's remarked from that different figures that the larger is the drop volume ratio ($\frac{V_{h-d}}{V_{s-d}}$) the longer is the formed continuous satellite. This is due principally to the higher capillarity that generated when the volume of the head drop linked to the needle tip is larger. This capillarity resists the downward fluid drainage during the merging which provokes the generation of the continuous satellite where the length is depending on the volume of the

head drop. After its rupture with the resulted drop, the satellite is pulled up promptly to the needle tip. Like illustrated in figure 2a and figure 3a, an extremely small daughter droplet is generated after the continuous satellite rupture. This remark is missed for the coalescence of the sessile drop with the large volume head drop as presented in figure 4a. The generation of the daughter drop is explained to the fact that her volume was trapped between two opposite bulk flows. The first is due to the downward movement of the merger drop centroid, and the second is referring to the upward movement of the very small quantity that tends to return back and stick on the needle tip. The balance between these forces will cause a rupture of the interface and provokes the generation of the secondary daughter drop. Afterward, the drop tip starts a damped harmonic movement before stabilizing at a final position as shown in figures above. This harmonic behavior is referring principally to the variation of the gravitational energy that causes a downward movement of the head drop centroid from its initial position (just at the starting the merger process) before reaching its smallest value at the first minimal peak where it will gain a maximal kinetic energy that pushes its upward to reach it first maximal peak. The time transformation between these two different energies is damped by the intermediate of the viscous dissipation until the resulted drop reaches its final equilibrium shape. Calculating the difference between the smallest and the largest peak (first amplitude) for each harmonic movement presented in figures 2b, 3b and 4b, respectively, it's found that the amplitude characterized the coalescence of the equal volume drops (figure 3) is the highest comparing to the two other cases.

- **Impact of the continuous phase viscosity**

In order to investigate the impact of the higher outer viscosity on the coalescence dynamics of different sized vertically aligned water drops, a series of experiments have been carried out. Comparing to the first experiments, only the continuous phase have been changed where more viscous silicone oil is chosen (47v350). Physical properties of the latter are cited in table 1. As used in the first experiments, there too the constant volume sessile drop ($V_{sessile\ drop} = 4\mu l$) has been deposited on a non-treated surface where the measured contact angle is $\theta = 72^\circ \mp 5^\circ$. Figures 5, 6 and 7 present snapshots for the coalescence process of vertically aligned water drops in silicone oil (47v350) for different head drop volumes ($V_{h-d} = 0,53\mu l, 4\mu l$ and $16\mu l$) as well as the time evolution of the drop tip position corresponding to each case. As found for the results above, the merger process is faster for small head drops volume than for other cases where the head drop volume is relatively larger. Comparing the coalescence time found for the two LL systems studied herein, it's found that the process in the water drops in low viscosity silicone oil (SilOil M40.165) is faster than the second LL system where the continuous phase (47v350) is highly viscous. The histogram shown in figure 8 illustrates more details about time spent by the merger process for each LL system in function with the head drop volume. This significant difference in the coalescence speed is due to the viscous resistance provoked by the viscosity of the outer phase. This resistance is generated during the different coalescence stages starting from the building of the liquid bridge to the relaxation of the resulted drop passing through the propagation of the capillary wave. Focusing on the relaxation of the resulted drop, it's depicted that for the water drops in high viscous silicone oil system (47v350) the damped harmonic movement is missed and the drop tip position is found to be damped directly after the rupture of the continuous liquid satellite.

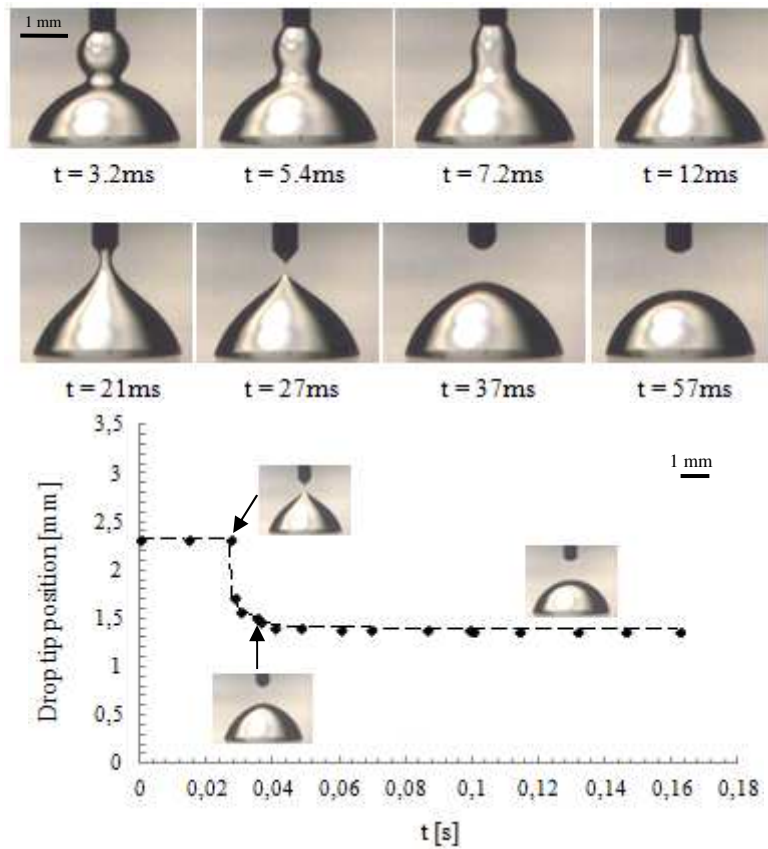
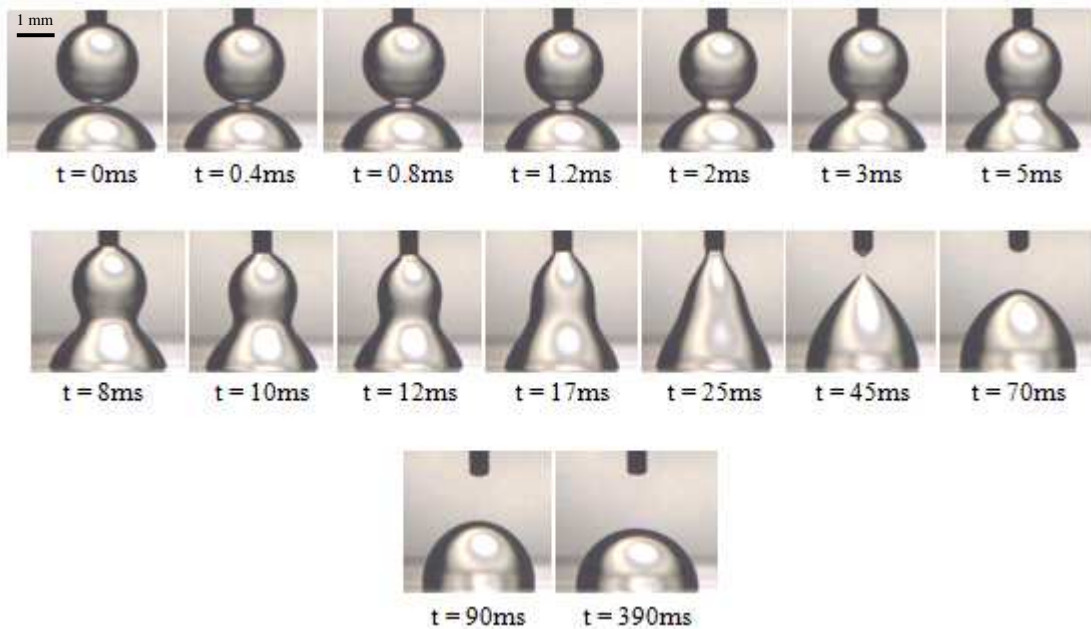


Figure 5: a) experiments snapshots for the coalescence of water drops in silicone oil (47v350); b) time variation of the drop tip position of the coalesced drop. $V_{sessile\ drop} = 4\mu\text{l}$ ($R_{eq} = 1,5\text{mm}$); $V_{head\ drop} = 5,3\mu\text{l}$ ($R_{sd} = 0,5\text{mm}$).



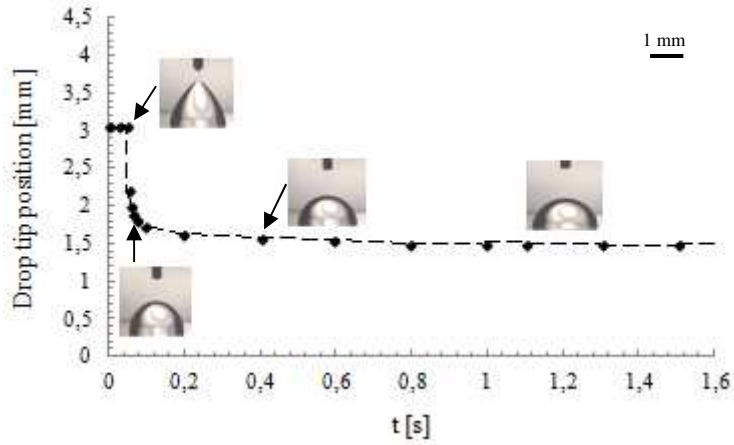


Figure 6: a) experiments snapshots for the coalescence of water drops in silicone oil (47v350); b) time variation of the drop tip position of the coalesced drop. $V_{sessile\ drop} = 4\mu l$ ($R_{eq} = 1,5mm$); $V_{head\ drop} = 4\mu l$ ($R_{sd} = 1mm$).

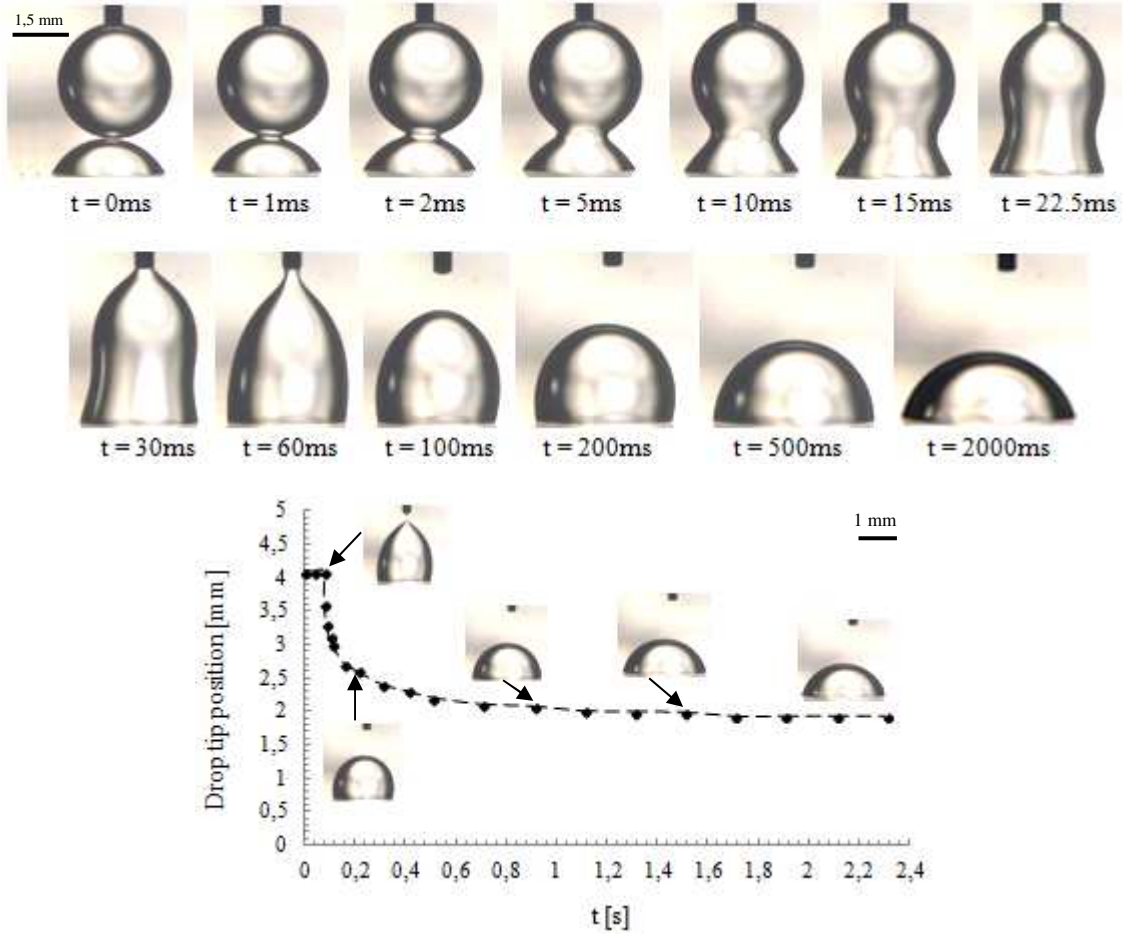


Figure 7: a) experiments snapshots for the coalescence of water drops in silicone oil (47v350); b) time variation of the drop tip position of the coalesced drop. $V_{sessile\ drop} = 4\mu l$ ($R_{eq} \approx 1,5mm$); $V_{head\ drop} = 16\mu l$ ($R_{sd} = 1,56mm$).

This behavior is due mainly to the high viscosity of the surrounded phase (47v350 silicone oil). The latter have resisted the gravitational energy and provoked, in contrast with the first LL system, a one stage downward movement of the merger drop centroïd until the equilibrium shape. Although, despite

the absence of the relaxation time, the merger process takes much more time to stabilize in its final shape (figure 8). The time evolution of the coalesced drop tip velocity is presented in figure 9 for both LL systems used in this work. As found for the evolution of the drop tip position, it's depicted that the variation of the velocity is characterized by a harmonic damped movement when the surrounded phase has a low viscosity contrary to the case where the outer viscosity is much higher and in this case it's remarked that the time evolution velocity starts from a higher velocity and stabilized without passing through harmonic movement.

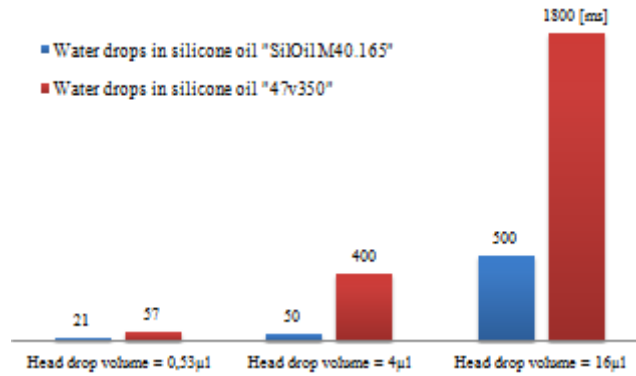
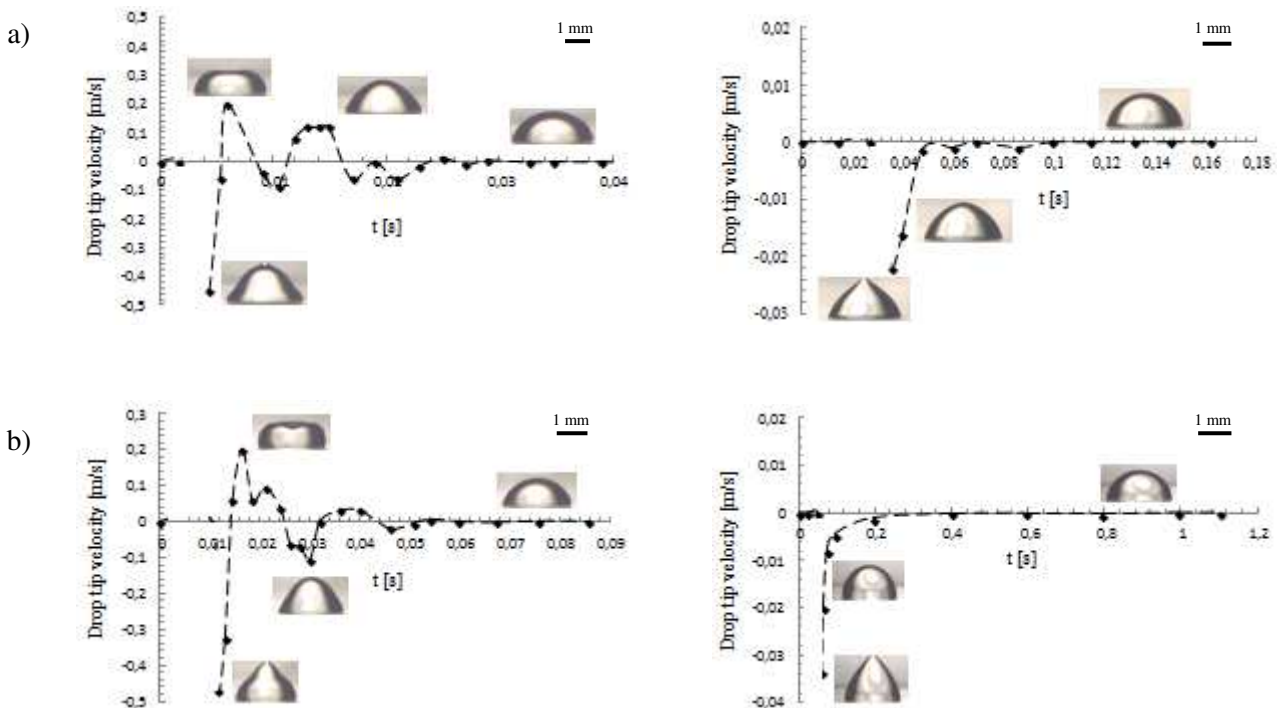


Figure 8: comparing between the times spent for the merger process for different head drops sizes



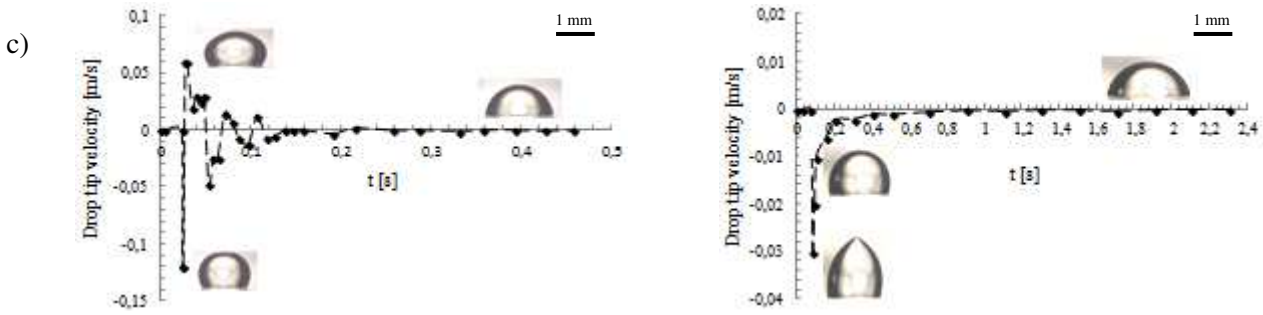


Figure 9: time variation of the drop tip velocity for different head drops sizes. Water drops in SilOil M40.165 (left) and water drops in 47v350 (right). $V_{sessile\ drop} = 4\mu l$ ($R_{eq} \approx 1,5mm$); a) $V_{h-d} = 0,53\mu l$ ($R_{head\ drop} = 0,5mm$); b) $V_{h-d} = 4\mu l$ ($R_{head\ drop} = 1mm$); c) $V_{head\ drop} = 16\mu l$ ($R_{h-d} = 1,56mm$).

The relaxation of the resulted drop does not involve just vertical motions; it also provokes a variation of the dynamic contact angle which is as well found to be impacted by the size of the head drop as well as the viscosity of the outer phase. Figure 10 illustrates the time evolution of the contact angle of the resulted drop during the coalescence process for both studied LL systems. It's observed that the first instants of the coalescence process are characterized by a no change of the contact angles. This period is referring to the initial moments of the liquid bridge building where the capillary wave starts propagating and located around the contact point between the two drops and so doesn't impact the bottom levels of the sessile drop. It's remarked that this initial period where the contact angle doesn't change is impacted by the head drop volume for water drops in higher viscous silicone oil (47v350) where it's found that the smaller the head drop volume the longer the no change period of the merger drop contact angle (7ms and 10ms for $4\mu l$ and $16\mu l$ head drop volumes, respectively). For the water drops in lower silicone oil (SilOil M40.165) system, the initial period (no change contact angle) is found to be almost the same (2ms). The viscosity of the outer phase has also an impact on the no change period of the resulted drop contact angle. It's depicted that for higher outer phase viscosity, the period where the merger drop contact angle doesn't vary is longer than what was observed when the surrounded viscosity is low. This is provoked mainly by the external capillary pressure generated from the outer to the inner phase and that resists the propagation of the capillary wave during the drops coalescence. This resistance action is more important within higher viscous oils than in lower viscous ones. A special case is observed for the small volume head drop ($0.53\mu l$) in contact with the sessile drop in the high viscous silicone oil (47v350). It's found that the merger drop contact angle doesn't change during all the coalescence process. This is due to the combination of the fact that the head drop is small and put in highly viscous outer phase which provokes a permanent no change of the contact angle as shown in figure 10a (right). After the no change period, the contact angle starts moving for both LL systems. It's depicted that for a low viscosity outer phase, the contact angle undergoes a damped harmonic movement where the first amplitude ($\theta_{max} - \theta_{min}$) is found to be increasing with the increasing of the head drop volume. In contrast, for the higher outer phase viscosity the contact angle variation is more stable where the harmonic movements are missing. It's depicted that the more the head drop volume increases the more intense is the first variation of the contact angle before getting its final value without undergoing the harmonic movement. This is due to the relatively higher gravitational energy caused by the increasing of the head drop volume that pushes the centroid downward and could provoke a slight variation in the contact angle before getting damped by the intermediate of the high viscous resistance.

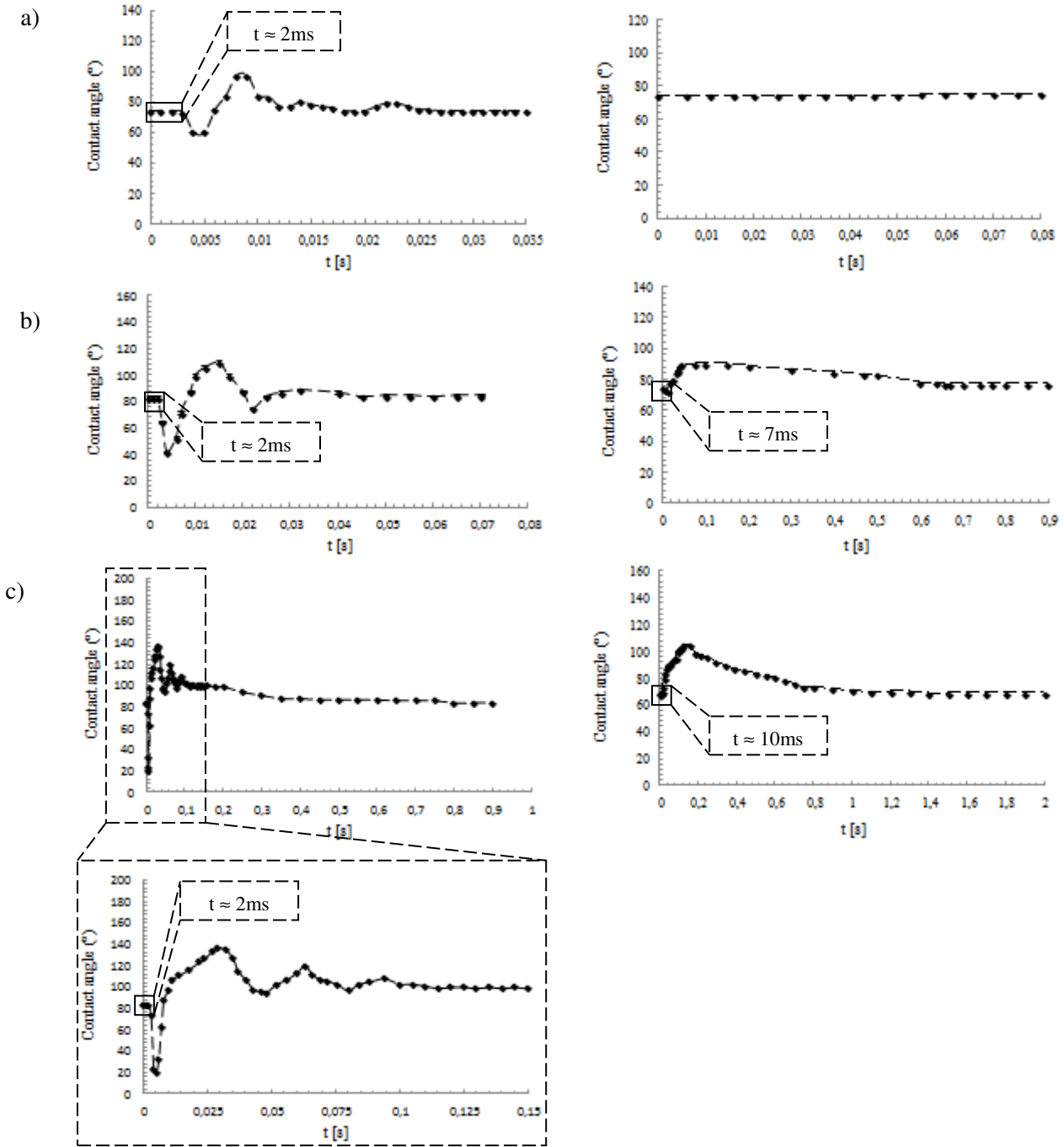


Figure 10: time variation of the contact angle for different head drops sizes. Water drops in SilOil M40.165 (left) and water drops in 47v350 (right). $V_{sessile\ drop} = 4\mu l$ ($R_{eq} \approx 1,5mm$); a) $V_{h-d} = 0,53\mu l$ ($R_{head\ drop} = 0,5mm$); b) $V_{h-d} = 4\mu l$ ($R_{head\ drop} = 1mm$); c) $V_{head\ drop} = 16\mu l$ ($R_{h-d} = 1,56mm$).

As cited above, the first instants of the coalescence process obtained for different series of experiments carried out in this work are characterized by the building of a liquid bridge that connects the drops and initiates the production of a single resulted drop (figure 11). The impact of the drop size initially positioned above the sessile one is investigated on the variation of the dimensionless capillary pressure generated around the liquid bridge during its formation.



Figure 11: schematic of the evolution of the capillary liquid bridge during the coalescence of vertically aligned drops.

Results proved that the same behavior was found for different head drop sizes where the external pressure is higher at an earliest stage of the bridge formation and decreases as the liquid bridge building progresses. As proved in the literature [27], this behavior is due mainly to the fact that the radius of the liquid bridge is smaller enough to guarantee a higher capillary pressure at earliest instants of the merging ($P_{l-br} = \frac{\sigma}{R_{l-br}(t)}$) and as the liquid bridge radius grows the capillary pressure generated around it decreases in parallel until it reaches its lowest value when the liquid bridge is almost completely formed. Normalizing the capillary pressure P^* for each head drop size, using the capillary pressure of the head drop before the coalescence P_{head} (table 2), its variation for both systems in function with the dimensionless time t^* for both LL systems is presented in figures 12a and 12b, respectively. Calculating the redefined Reynolds number ($Re = \frac{\sigma r(t)}{\rho \gamma^2}$) for these different LL systems at $t = 0.0001s$, it's found that it is widely larger than the unity ($Re > 3000$ for all the different studied head drop volumes) which explains that the regime of the initial stage of the coalescence process is dominated by the inertial forces induced by capillarity. Thereby, the time normalizing was realized using the inertial-capillary time $\tau_i = \sqrt{\frac{\rho R^3}{\sigma}}$. It's observed for the different sizes of the head drop that the variation of the dimensionless capillary pressure ($P^* = \frac{P(t)}{P_{c-head}}$) with the dimensionless time ($t^* = \frac{t}{\sqrt{\frac{\rho R^3}{\sigma}}}$) is characterized by a power scaling law as following:

$$\frac{P(t)}{P_{c-head}} = A \left(\frac{t}{\sqrt{\frac{\rho R^3}{\sigma}}} \right)^{-B} \quad (1)$$

here σ is the surface tension of the LL system, R and ρ are, respectively, the radius and the density of the head drop.

It's depicted from the figures that the parameters A and B haven't been affected by the head drop size where the same values are found regardless of the head drop volume. In contrast, since the surface tensions of both LL systems are practically the same, these parameters A and B are depending strongly but not only on the viscosity of the outer phase. For the water drops-in-Silicone oil "M40.165" system, parameters A and B are found to be ≈ 0.51 and ≈ 0.41 , respectively. On the other side, these parameters take the value of about $A \approx 0.91$ and $B \approx 0.47$ for the water drops-in-Silicone oil "47v350". In order to get further information about different elements those impact parameters A and B , more experiments and numerical studied have to be carry out for more liquid-liquid and gas-liquid systems.

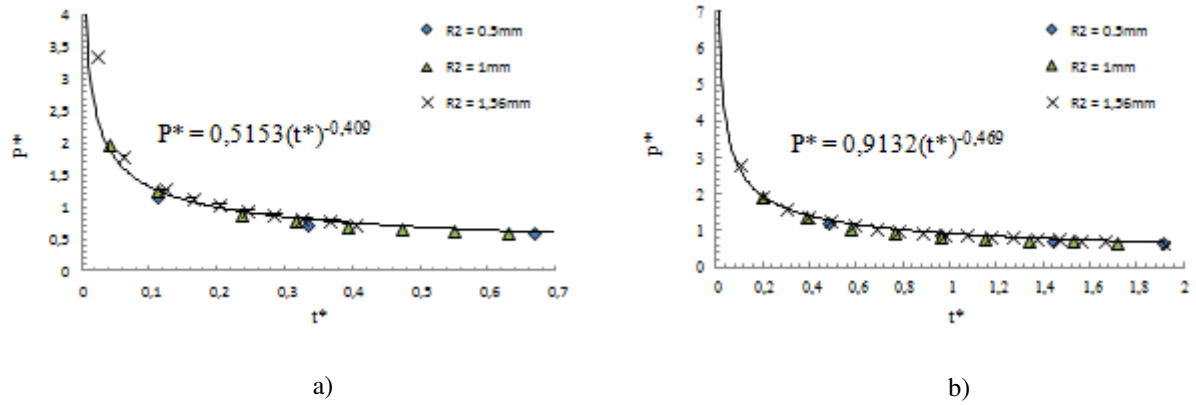


Figure 12: Variation of the dimensionless capillary pressure P^* around the liquid bridge with the dimensionless time t^* for different sizes of the head drop; a) water drops-in-silicone oil "M40.165"; b) water drops-in-silicone oil "47v350".

Conclusion

In this paper, the impact of the outer phase viscosity as well as the size of the head drop have been investigated on the relaxation together with the contact angle of the resulted drop during the coalescence of vertically aligned drops in silicone oil. Two types of silicone oil have been used; Huber SilOil M40.165 and Rhodorsil 47v350. Calculating the redefined Reynolds number ($Re \gg 1$), it was found that the first stages of the coalescence process is dominated by the inertial force for both LL systems. The main results that have been found are as follow:

- For low viscous outer phase, the resulted drop undergoes a damped harmonic relaxation that is referred to the balance between the gravitational energy that pushes the drop centroid downward, the kinetic energy that pushes the centroid upward and the dissipative viscous energy that damped this harmonic movement. Considering that the contact line changes very slowly, this energy balance will impact directly the contact angle as well, which undergoes the same behavior with a damped harmonic variation. It's found that the first amplitude of the contact angle variation increases with the increasing of the head drop volume. Contrary to the vertical relaxation of the resulted drop where it's found that the first amplitude is higher for equal volume drop ($V = 4\mu\text{l}$) than other cases where the aligned drops are not equal.
- For a highest viscosity of the outer phase, it's depicted that the relaxation of the resulted drop is mono-stage and doesn't undergo any harmonic movement. This is explained by the fact that the viscous dissipation generated herein is much higher than the previous case which leads to a sufficient resistance to the gravitational energy that supposed to push the centroid downward what results on the direct damping of the drop tip. Concerning the contact angle, here too the highest viscosity has a main impact on its dynamics. It's found that for very small head drops volume ($V = 0.53\mu\text{l}$), the contact angle doesn't undergo any variation and conserved its initial contact angle during all the merger process. Furthermore, it's found that the more the head drop volume increases the more the contact angle varies slightly but without any harmonic movement comparing to the previous case before coming back to its final equilibrium contact angle.
- Focusing on the first instants of the coalescence process which is characterized by the building of liquid bridge, it's illustrated that the dimensioning of the capillary pressure of the latter

shows that its evolution with the dimensionless time follows a power scaling law presented in eq.1 where new parameters A and B appeared. In order to check the applicability of this power law, a series of other numerical and experimental studies have to be carried out taking into account a large range of liquid-liquid and gas-liquid systems.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

References

- [1] Sarrazin, F., Prat, L., Casamatta, G., Joanicot, J., Gourdon, C., Cristobal C., (2006) “Micro-drops approach in micro-reactors: mixing characterization,” Université de Toulouse.
- [2] Simon, M., G., and Lee, A. P. (2012) “Microfluidic Droplet Manipulations and Their Applications,” *Microdroplet Technology: Principles and Emerging, Applications in Biology and Chemistry*, Chapter 2.
- [3] Xu, B., Nguyen, N.-T., Wong, T., N., (2011) “Droplet Coalescence in Microfluidic Systems,” *Micro and Nanosystems*, Volume 3, Issue 2.
- [4] Hsieh A., Pan P., Lee, A., (2009) “Rapid label-free DNA analysis in picoliter microfluidic droplets using FRET probes,” *Microfluid Nanofluidics* 6:391–401.
- [5] Bhardwaj R., and Agrawal, A. (2020) “Likelihood of survival of coronavirus in a respiratory droplet deposited on a solid surface,” *Physics of Fluids* 32, 061704.
- [6] Jing, J., Chin, H., O., Dzung, V., D., Nguyen, N.-T., (2017) “Coalescence Processes of Droplets and Liquid Marbles”, *Micromachines*. 8, 336.
- [7] Saboni, A., Gourdon, C., and Chesters, A. K., (1995) “Drainage and Rupture of Partially Mobile Films during Coalescence in Liquid-Liquid Systems under a Constant Interaction Force”, *J. Colloid and Interface Science*. 175. 27-35.
- [8] Dreher, T., Glass, J., O’Connor, A., Stevens, G., (1999) “Effect of rheology on coalescence rates and emulsion stability”, *AIChE*, 45, 1182–1190.
- [9] Bradley, S., Stow, C., (1978) “Collisions between liquid drops. *Philos. Trans. R. Soc. Lond*”. A *Math. Phys. Eng. Sci.*, 287, 635–675.
- [10] Kapur, N., Gaskell, P.H., (2007) ”Morphology and dynamics of droplet coalescence on a surface,”. *Phys. Rev. E Stat. Nonlinear Soft Matter Phys.*, 75, 056315.
- [11] Jing, J., Chin, H., O., Dzung, V., D., Nguyen, N.-T., (2017) “Coalescence Processes of Droplets and Liquid Marbles”, *Micromachines*. 8, 336.
- [12] Brik, B. Ruscassie, R., Saboni, A., (2016) “Droplet deformation and coalescence under uniform electric field,” *Journal of Chemical Technology and Metallurgy*, 51, 6, 649-659.

- [13] Mohammadi, M., Shahhosseini, S., Bayat, M., (2012) "Direct numerical simulation of water droplet coalescence in the oil," *International Journal of Heat and Fluid Flow*, 36, 58-71.
- [14] Anand, V., Juvekar, V., A., Thaokar, R., M. (2019) "Modes of coalescence of aqueous anchored drops in insulating oils under an electric field," *International Journal of Heat and Fluid Flow*, 36, 58-71.
- [15] Degrand, L., Michon, C., A., Bosc, V. (2016) "New insights into the study of the destabilization of oil-in-water emulsions with dextran sulfate provided by the use of light scattering methods," *Food Hydrocolloids*, 52, 848-856.
- [16] Eow, J.S., and Ghadiri, M. (2000) "Electrostatic enhancement of coalescence of water droplets in oil: a review of the technology," *Chem. Eng. J.*, 85, 357-368.
- [17] Charles, G.E., and Mason, S.G. (1960) "The coalescence of liquid drops with flat liquid/liquid interface," *J. Colloid Sci.*, 15, 236-276.
- [18] Chiesa, M., Melheim, J.A., Pedersen, P.A., Ingebrigtsen, S., Berg, G. (2005) "Forces acting on water droplets falling in oil under the influence of an electric field: numerical predictions versus experimental observations," *Eur. J. Mech. B-Fluid*, 24, 717- 732.
- [19] Teigen, K.E., Munkejord, S.T., Bjørklund, E. (2008) "A computational study of the coalescence process between a drop and an interface in an electric field," 6th International Conference on CFD in the Oil & Gas, Metallurgical and Process Industries SINTEF/ NTNU, Trondheim, Norway.
- [20] Helleso, S.M., Atten, P., Berg, G., Lundgaard, L.E. (2015) "Experimental study of electrocoalescence of water drops in crude oil using near infrared camera," *Experiments in Fluids* volume 56.
- [21] Thoroddsen, S. T., Takehara, K., Etoh, T. G., (2005) "The coalescence speed of a pendent and a sessile drop", *J. Fluid Mech.* 527, 85–114.
- [22] Eggers, J., Lister, J. R., Stone, H. A., (1999) "Coalescence of liquid drops," *J. Fluid Mech.* 401, 293–310.
- [23] Eiswirth, R. T., Bart, H.-J., Ganguli, A. A. Kenig, E. Y. (2012) "Experimental and numerical investigation of binary coalescence: Liquid bridge building and internal flow fields", *Phys. Fluids* 24, 062108.
- [24] Duchemin, L., Eggers, J., Josserand, C., (2003) "Inviscid coalescence of drops," *J. Fluid Mech.* 487, 167–178.
- [25] Nowak, E., Kovalchuk, N. M., Che, Z., Simmons, M. J. H. (2016) "Effect of surfactant concentration and viscosity of outer phase during the coalescence of a surfactant-laden drop with a surfactant-free drop," *Colloids Surf., A*, 505, 124 – 131.
- [26] Nowak, E., Xie, Z., Kovalchuk, N. M., Matar, O. K., Simmons, M. J. H. (2017) "Bulk advection and interfacial flows in the binary coalescence of surfactant-laden and surfactant-free drops," *Soft Matter*, 13, 4616 – 4628.

- [27] Brik, M., Harmand, S., Zaaroura, I., Saboni, A. (2021) “Experimental and Numerical Study for the Coalescence Dynamics of Vertically Aligned Water Drops in Oil,” *Langmuir* 37 (10), 3139-3147.
- [28] R. Gunjan, M., Somwanshi, P., Agrawal, A., Khandekar, S., Muralidhar, K., (2015) “Recoil of drops during coalescence on superhydrophobic surfaces,” *Interfacial Phenomena and Heat Transfer*, 3(2): 203-220.
- [29] Somwanshi, P., Muralidhar K., Khandekar S., (2020) “Coalescence of vertically aligned drops over a superhydrophobic surface,” *Phys. Fluids* 32, 052106.
- [30] Sykes, T. C., Castrejon-Pita, A. A., J. Castrejon-Pita, R., Harbottle, D., Khatir, Z., Thompson, H. M., Wilson, M. C. T. (2020) “Surface jets and internal mixing during the coalescence of impacting and sessile droplets,” *Phys. Rev. Fluids* 5, 023602.
- [31] Xia, X., He, C., Yu, D., Zhao, J., Zhang, P. (2017) “Vortex-ring-induced internal mixing upon the coalescence of initially stationary droplets,” *Phys. Rev. Fluids* 2, 113607.
- [32] Zheng, J., Shi, H., Chen, G., Huang, Y., Wei, H., Wang, S., Wen, W., (2016) “Relaxation of liquid bridge after droplets coalescence”, *AIP Advances* 6, 115115.
- [33] Lee, M. W., Kim, N. Y., Chandra, S., Yoon S. S. (2013) “Coalescence of sessile droplets of varying viscosities for line printing,” *International journal of multiphase flow*, 56, 138-148.
- [34] Somwanshi P., Muralidhar K., Khandekar S. (2020) “Coalescence Dynamics of Drops over a Hydrophobic Surface,” *Book: Drop Dynamics and Dropwise Condensation on Textured Surfaces*, 81-129. Mechanical Engineering Series. Springer, Cham.
- [35] Li, S., Chu, F., Zhang, J., Brutin, D., Wen, D. (2019) “Droplet jumping induced by coalescence of a moving droplet and a static one: Effect of initial velocity,” *Chemical Engineering Science*, 211, 115252.
- [36] Chen, Y., Lian, Y. (2018) “Numerical investigation of coalescence-induced self-propelled behavior of droplets on non-wetting surfaces,” *Physics of Fluids* 30, 112102.
- [37] Chen, X., Patel, R. S., Weibel, J. A., Garimella, S. V. (2016) “Coalescence-Induced Jumping of Multiple Condensate Droplets on Hierarchical Superhydrophobic Surfaces,” *Scientific Reports* 6(1):18649.
- [38] Vahabi, H., Wang, W., Mabry, J. M., Kota, A. K. (2018) “Coalescence-induced jumping of droplets on superomniphobic surfaces with macrotexture,” *Sci. Adv.*; 4: eaau3488.

