Research article

Effect of temperature and surfactants on evaporation and contact line dynamics of sessile drops

Rachid Bennacer\textsuperscript{a,b,1}, Xiaoyan Ma\textsuperscript{b,c,1}

\textsuperscript{a} International Centre in Fundamental and Engineering Thermophysics, Tianjin University of Commerce, Guangrong Rd 409, Beichen District, Tianjin 300134, China
\textsuperscript{b} Université Paris-Saclay, ENS Paris-Saclay, CNRS, LMPS, Gif-sur-Yvette, France
\textsuperscript{c} Beihang Hangzhou Innovation Institute Yuhang, Xixi Octagon City, Yuhang District, Hangzhou 310023, China

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ABSTRACT

Researches on droplet evaporation have a vast range of applications in different fields. The mechanisms of droplets evaporation have been a topic of investigation for decades. The droplet after spreading on the substrate exhibits constant evaporation rate for a constant base diameter and decreasing contact angle; followed by another evaporation mode with constant contact angle and decreasing base diameter; and finally, evaporation with decreasing of both contact angle and base diameter. In the present study, the evaporation rate and mechanisms influenced by surfactant and temperature effects are analysed. The evaporation was demonstrated to be related to the dynamics of the sessile droplet spreading ability and the depinning event. It remains complex to predict the evaporation of the sessile droplet due to the continuous change in surfactant local concentration. Moreover, the temperature change modifies the surface tension and the induced inner flows. These two questions find answers in the triple line dynamic which controls the evaporation rate. The forces controlling the depinning with surfactant was quantified. A clear constant force was found following depinning. Such dynamic equilibrium demonstrates also the effect of the friction, surface roughness and moreover the limit of surfactant concentration in the triple line direct vicinity. The evaporation rate shows two particularities: one is the classical decrease of the evaporation rate after depinning; the second in case of surfactant, there is an enhancement appearing at later droplet lifetime, which could be a consequence of film evaporation contributing to the evaporation near the triple line. The local evaporation does not change significantly despite the change in the contact angle with respect to surfactant concentration. The substrate temperature enhances evaporation rate. The coupled temperature and surfactant effect show a local evaporation rate decreasing with surfactant concentration increase at ambient temperature (around 20 °C) and enhancement with surfactant concentration at higher temperature.

1. Introduction

Researches on droplet evaporation mechanisms and interfacial phenomena have been a popular topic for decades, for the fact that evaporation of liquid droplets (generally in ambient air) is related to many basic physicochemical processing and it has a wide range of applications in different fields.

Droplet evaporation must absorb heat from the surrounding environment to evaporate, thus inducing a relative cooling effect. Therefore, droplet evaporation is applied in spray cooling [1, 2, 3], fire extinguishing [4, 5, 6], heat exchangers [7], air humidifying [8, 9, 10, 11], electronic chip cooling [12], and more recently in high-density microelectronic cooling [13]. Furthermore, the evaporation of droplets and the deposit patterns can be applied in broader fields such as coating and printing processes [14, 15], spotting of DNA microarray data [16, 17], and medical diagnosis [18], etc. Therefore, it is important to understand the heat transfer mechanisms from the heated surface substrate to deposited droplets, in order to increase the spray cooling efficiency and improve water utilization, which has led to extensive research.

In the field of pesticides spraying, one of the most essential problem to be solved is how to achieve the most effective wetting while prolonging evaporation time. If the coverage surface of pesticide droplets on crops increases, the used quantity of pesticides decreases [19]. In addition, the...
duration time of evaporation (lifetime of the droplet) of pesticide liquid drops spreading on crop surfaces directly determines whether the active ingredients of pesticides could crystallize and be fully absorbed. Droplet size, crop leaf surface property (waxy or plunger) and surfactants are main factors affecting the wetting and evaporation of pesticide droplets. Studies shows also surfactants reduce the surface tension of water, thereby improving the wetting of water droplets on the surface of the leaves [20]. Therefore, the enhancement of wetting and evaporation of pesticide droplets is beneficial to improving crop production. Other applications concern the cleaning and cooling of hot surfaces. Another hot topic is related to the intensive surface cooling (electronics components or PV concentrators). Enhancing heat transfer can be achieved by convection, boiling or droplet spreading.

Surfactants are surface-active chemicals, which contain groups of opposite polarity and solubility tendencies. A surfactant modifies the interaction properties on interfaces by forming micelles and oriented mono-layers. A number of studies have proved that the addition of surfactants into aqueous solutions can enhance the ability of spreading and wetting by reducing the surface tension, therefore imposing an effect on evaporation, [21, 22]. The decrease in surface tension will consequently increase the ability of wetting on a solid surface or substrate. Surfactants modify the evaporation process as what was studied in the boiling process by promoting smaller separation vapor bubbles and the early occurrence of nuclear boiling [23, 24, 25]. In addition to the concentration and related adsorption dynamics of surfactants, studies show that temperature also plays a significant role in heat and mass transfer mechanisms and phase change performance [26, 27]. Although the spreading dynamics of droplets with aqueous surfactant has been investigated over the last few decades, the surfactants influence on wetting spreading and evaporation is not still thoroughly understood.

Many descriptions have reported the evaporation process of a sessile droplet in literatures. With regard to pure water droplets evaporation on an ideal smooth surface under a partially wetting condition, it is generally acknowledged that the contact angle remains constant while the droplet base decreases [28, 29]. On the other hand, evaporation on a real surface tends to maintain a pinned contact line in the initial period of evaporation, which means the base diameter of the droplet remains constant for some duration. More generally, the whole evaporation process of a droplet after being deposited on substrates may consist of four stages: i. Spreading stage; ii. Evaporation stage with constant base diameter and decreasing contact angle; iii. Evaporation stage with constant contact angle and decreasing base diameter; iv. Final stage of evaporation with decreasing of both contact angle and base diameter until the disappearance of the sessile droplet [30, 31]. During the first stage, it is a transient process with contact angle and base diameter changing simultaneously until reaching the static advancing contact angle and static diameter. We neglect the evaporation process of this stage (spreading) because the characteristic time scale is short enough in comparison to the whole evaporation lifetime and the estimated variation in volume during this stage is less than 5% [32, 33].

Therefore, in the present study, aiming at exploring evaporation mechanisms influenced by surfactants and temperature, we mainly focus on the last three stages which contribute to the majority to the evaporation process. To describe the contact angle relationship for the three phases (vapor, liquid, solid) near the triple line at equilibrium state, the Young–Dupre equation is adopted. It can be derived from the force balance at the contact line of the droplet, as shown in Eq. (1).

\[ \cos \theta = \frac{(\sigma_s - \sigma_d)}{\sigma_d} \]  

(1)

In which, \( \theta \) is the contact angle under equilibrium state, \( \sigma_s, \sigma_d \) and \( \sigma_l \) are the surface tension between solid-vapor, liquid-vapor and solid-liquid, respectively. The dynamics of the contact line is very important since it controls the evaporation rate. Literature shows that the evaporation rate is almost proportional to the diameter of the droplet and it remains constant as long as the drop is pinned on the substrate. However, the evaporation rate departs from this tendency the instant de-pinning begins to occur. Consequently, when the evaporation is far from its position of equilibrium state, it is essential to quantify the force acting on the triple line. Eq. (2) provides the relationship of the force on the triple line as function of contract angles and surface tension.

\[ F = \sigma_v (\cos \theta_0 - \theta_l) \]  

(2)

where \( F \) is the force acting on the triple line of the evaporating droplet, \( \theta_0 \) is the dynamic contact angle during droplet evaporation. It is worth noting that this equation is valid under the quasi-static state, and for fast evaporating droplet in a transient state, evaporation velocity needs to be considered, the modified expression are shown in literature [34]. It is acknowledged that the resistance to capillary driving forces is largely due to the heterogeneity property of surfaces and viscous effects [34]. The two distinguished approaches to describe the wetting phenomenon are the kinetic theory proposed by Blake and Haynes [35, 36], and the hydrodynamic approach [37].

With regard to the temperature effect, however, in many reported studies, it is not well controlled [38, 39], which has the potential to influence measurement accuracy of surface tension and contact angle. In the case of temperature imbalance, convection inside the droplet will be inevitably induced and therefore leads to extra motion near the interface, which complicates the interaction between droplet evaporation and molecular adhesion, and also alters the interface energy balance [40, 41]. Therefore, it is quite important to consider temperature effects on the interfacial properties and evaporation process, as well as the surfactant aqueous solution under thermal equilibrium state.

Based on the discussion above, it is obvious that the evaporation of surfactants laden droplets is rather complex due to the continuous change in surfactant local concentration, and due to the change in temperature that modifies the surface tension and the induced inner flows. These two questions find answers in the triple line dynamic which controls the evaporation rate. In the present work, we performed droplet evaporation experiments with pure water as a reference, and surfactant laden solutions under variable substrate temperature. To elucidate the temperature effect on spreading as well as evaporation mechanism, temperature of substrate is controlled and monitored during the evaporation lifetime. Contact angle and base diameter are recorded under different experimental conditions, and finally the Young unbalanced force near the contact line are calculated and compared for different experimental conditions.

2. Experiments

The apparatus used for injecting droplets will be introduced in this part. For the creation of constant volume sessile droplets from the syringe to the substrate, an automatic injection pump was used. The injection pump utilized is a Cole-Parmer Company 74900 series one. The pump could deliver droplets of the same volume following calibration. The inner diameter of the syringe was used as a reference to calibrate the pump. For the sake of the investigation of the evaporation of a sessile drop, a volume of the drops was chosen to be 10 \( \mu \)l and the injection rate was 0.25 ml/s.

The droplet evaporation tests were carried out under controlled laboratory room temperature 20 ± 1 °C, and relative humidity 45 ± 5% rh, the ambient pressure is considered as the same as atmosphere pressure.

The experimental equipment used to carry out the test is an FTA200 drop shape analyser. The FTA software quantifies the base diameter, height and contact angle of the sessile drops instantaneously, as showed in Figure 1. The substrate is heated by a resistive heater at constant temperature using a PID controller.

A container with a volume of one litter was used to prepare mixtures. This was cleaned twice with distilled water. Afterwards it was dried with compressed air and weighed on a balance.
Using a syringe filled with Triton-X100, 1 g of the latter was carefully dispersed in the container and diluted with 999 g of water. The solution, with a concentration $10^{-3}$ by weight of Triton-X100, was stirred to dissolve the surfactant thoroughly. Another 1 liter container was used, which was cleaned and weighed in the same way. One gram of the $10^{-3}$ by weight solution was dispersed in the container carefully and diluted with another 999 g of water. That produced a solution with a concentration of $10^{-6}$ by weight surfactant in water with a relative error on the obtained concentration of 0.2%. These solutions are used to generate the droplets to be studied.

The droplets evaporation test was performed on Drop Shape Analyzer (DSA100) by Krüss GmbH. The classical approach was adopted to capture and measure instantaneously contact angle (CA) and base diameter (BD), and the drop volume is calculated according to the spherical cap assumption with the circular segment method. Consequently, the evaporation rate is defined as the volume decreasing rate of the droplet. Uncertainties in measurements of the contact angle and drop base radius are estimated at within ±1%.

3. Results

The experimental results for various tests will be presented in this part. Figure 2 shows the evolution of contact angle (CA) and base diameter (BD) over time for two different temperatures, which are 20 °C and 40 °C. The accuracy on the contact angle ±1° and the base diameter with ±5 × 10^{-2} mm. The volume is deduced from the base and the contact angle so the error will be less than ±7.2 × 10^{-2} μl. These results provide a comparison of the temperature effect during evaporation.

As indicated in Figure 2, we observe the two recognizable stages of constant base diameter (BD, blue color) with a decrease of contact angle (CA, red color) controlled by the evaporation rate and followed by depinning at a given time indicated by the diameter decreasing over time.

The comparison of the two blue-dot curves shows that the de-pinning of the base happens earlier for higher temperature, and the ratio of pinning period over the total evaporation duration is smaller for the evaporation under a higher temperature (40 °C).

The red-triangle curves present a sudden increase of contact angle at the end of the constant BD point (stick and slip phenomenon) for both conditions. However, the effect is less obvious for higher temperature (40 °C). The contact angle decreases linearly with a lower slope after depinning. The slope before depinning for 20 °C is almost the same as that of 40 °C, and the evaporation rate varies after depinning, which will be discussed later.

The comparison of the total duration also shows that by increasing the temperature from 20 °C to 40 °C, the evaporation life time has decreased by almost two thirds, which implies a significant enhancement of evaporation.

Figure 3 presents the evolution of contact angle and base diameter with time for pure water and a solution with surfactant 10^{-2} wt., which corresponds to the surfactant effect.

We can conclude from these results that the surfactant increases spreading, which is deduced from the significant decrease of contact angle and the slight increase in base diameter. In addition, the de-pinning phenomenon almost disappears after adding the surfactant, because the BD remained almost constant after the initial spreading. Furthermore, in comparison with pure water, the droplet with surfactant evaporates with almost a constant base diameter and a continuously decreasing contact angle.
angle until the end of the evaporation. When the evaporation rate is high and contact angle tends to weak values, it is more challenging for the automatic software to identify the external limit of the droplet. As shown in Fig. 3, the maximum error on the diameter is indicated versus time for the worst situation in case of surfactant addition.

The concentration effect of surfactant with reference to pure water is shown in Figure 4. As shown in Figure 4a, the contact angle decreases continuously for pure water and for all surfactant concentrations, but depinning happens in case of pure water and in case of low concentration (10⁻⁵ wt%) of surfactant (see Figure 4b). While with higher concentration (more than 10⁻⁴) it is less obvious, which is characterized by the continuously decreasing contact angle in Figure 4a. With relative high concentration of surfactant (>10⁻³), the contact angle drops drastically especially in the initial period of evaporation.

Such significant decrease is also attributed to the spread (BD increase) which could be attributed to the local increase of surfactant concentration modifying the surface tension in the vicinity of the triple line. The error bar illustrates the change in base diameter and the local concentration effect remain obvious. We don’t plot the other error bar with lower values in order to keep clear the figures.

Figure 4b shows that wetting diameter begins with a constant value stage for pure water and low concentration of surfactant (<10⁻⁵), while for higher concentration (10⁻⁴ and above) there appears an obvious increase of diameter after injection and followed by a shorter constant diameter stage, which emphasizes the enhancement of spreading induced by the addition of surfactant.

As a short summary of the effects of the two main parameters - temperature and surfactant, we can conclude that the effect of temperature does not significantly affect the dynamics and profile of evolution of the contact line and wetting angle, however it enhances evaporation and reduces the lifetime of the drops. By contrast, surfactants have significant effect on the dynamics of the contact line and contact angle behavior. Surfactants have an effect on evaporation rate and lifetimes because of the significant spreading of the base. The Young unbalanced forces will be calculated for both temperature and surfactants cases. This aims to study the force acting on the contact line and quantify the depinning force.

4. Analysis and discussion

This section analyzes the surfactants effect on the evaporation rate and the dynamics of the sessile droplet. It is well established that the evaporation rate is related to the triple line length (base diameter) and under some conditions the depinning occurs and therefore modifies the phenomena. We will present the different regimes and analyze the related forces inducing the depinning. The effect of the substrate temperature and the surfactant concentration are presented and analyzed as well. We will finalize the discussion by the induced evaporation rate and the surfactant effect on these important phenomena.

4.1. Contact line dynamics

The two accepted stages of constant base diameter (BD) with decreasing contact angle (CA) and followed by depinning is influenced by temperature. De-pinning of the base happens earlier for higher temperature evaporation. However, the sudden increase of contact angle (stick and slip) is less obvious for higher temperature. As revealed in Eq. (1), the Young unbalanced force increases with time as long as the contact line is pinned.

The force in Eq. (2) increases faster for higher temperature because the evaporation is enhanced by the saturated vapor pressure, which increases with temperature [13]. We underline also the effect of the temperature on the surface tension with changes from –72 to –60 mN/m corresponding to 20-80 °C.

As indicated by an arrow for 20 °C, in Figure 5, the depinning event corresponds a small peak in the Young unbalanced force. In the case of surfactants at high concentration, after an initial increase in the force, it levels off because the contact line slides on the substrate. It is illustrated by a plateau appearing after depinning. The resulting real dynamic forces integrate the shear stress and inertia so it is more complex as it affected by the combination factors of local velocity, the layer thickness and the substrate roughness.

The Young unbalanced force applied at a given temperature and different surfactant concentrations is illustrated in Figure 6. It is shown that the force increases faster for higher temperature because the
evaporation is enhanced by the saturated vapor pressure, which increases with temperature.

The depinning event corresponds to a small peak for low concentration and a significant plateau duration in the Young unbalanced force (indicated by horizontal dashed lines in Figure 6).

In the case of surfactants at high concentration, after an initial increase in the force, it levels off because the contact line slides on the substrate. It is clearly illustrated that the plateau level decrease with the increase of concentration. The relationship of surfactant concentration and the surface tension is illustrated by the insert on Figure 6 and the values are included in the evaluated forces. However, the local enrichment in surfactant concentration due to evaporation will modify locally the equilibrium. This is illustrated by the nonlinear observed plateau decreases with concentration as $F_{\text{line}} = 3.6 \ C^{-0.2}$.

The resulting real dynamic is related to the previous mentioned parameters and the evaporation rate effect will be discussed later.

### 4.2 Evaporation rates

In this section we’ll discuss the evaporation rate on different conditions. Figure 7 shows a constant volume decreasing over time due to evaporation and the depinning for the water reference case illustrates clearly the evaporation rate decrease. For the case with surfactant, better wetting confirms the evaporation enhancement due to the spreading. However, we can observe the evaporation increase over time, which could be the consequence of the resulting thin film and the added contribution to the triple line.

In order to extract the increase of the diameter due to the spread, we represent the evolution of volume over base diameter (Volume/ BD) in Figure 8. At early time we can see that the BD increases with surfactant and it induces a volume to this base diameter ratio up to 3.

The evaporation rate is the slope of the volume curves in time (Figure 8) for constant BD (illustrated by the grey boxes) for different surfactant concentrations. We can see some specific behavior during the spread for high concentration in the early stage of the droplet or the depinning of the pure water case (0 wt.). However, we can see that the evaporation rate per unit length of contact line is almost constant with the relative increase at the final stage as previously mentioned. The global evaporation is enhanced by the spreading nevertheless evaporation rate per unit length of contact line is also affected by the liquid constitution and especially the surfactant concentration increases over time towards a maximum value. The flow in the vicinity of the triple line and the internal flow on the droplet are the controlling parameters of such evaporation rate per unit length of contact line.

The evaporation rate per unit length of contact line for the different surfactant concentrations and substrate temperatures are summarized in Figure 9. It is obvious that the evaporation increases, as previously mentioned, with the temperature increase. It is interesting also to see that besides the global evaporation increase with the surfactant concentration, we observe also a relative evaporation rate per unit length of contact line decrease with surfactant at ambient temperature (20 °C). For higher temperature an increase is observed and confirm the fact that the underlined evaporation rate per unit length of contact line is function of both the local surfactant concentration and the convective flow induced by the thermal and solutal coupling. After depinning the evaporation rate is also reduced by the smaller diameter. Such depinning phenomenon is coupled with the surface tension and is enhanced with the concentration increase. We can add that for some given temperature around 30 °C the local evaporation rate can appear independent of surfactant concentration. The surfactant affects several underlying phenomena acting in antagonist way on the observed evaporation, for example by increasing the spreading but also facilitating the depinning angle.

As a perspective a μPIV experiments can visualize the eventual complex flows and direction flow change in the droplet bulk and in the vicinity of the triple line. Concerning the surfactant concentration limit near the triple line and the corresponding Marangoni forces a numerical work will be helpful.

### 5. Discussions and conclusions

We analyzed and discussed the temperature and surfactants effects on the global and evaporation rate per unit length of contact line. The
evaporation was demonstrated to be related to the dynamics of the sessile droplet spreading and the depinning event.

The depinning modifies the base diameter and is controlled by local forces. The analysis of such forces confirms the balance controlling the depinning and with surfactant the constant spreading in competition with depinning was demonstrated. A clear constant force was found. Such dynamic equilibrium demonstrates also the effect of the friction, surface roughness and moreover, the limit of surfactant concentration in the triple line vicinity. Such concentration limit controls the apparent surface tension. The reached concentration is controlled by both the evaporation rate on the contact line and the inner flow supplying fresh liquid arriving from the bulk. The inner supplied liquid flow is also controlled by the thermal Marangoni flow (especially on hot substrates) and the solutal Marangoni flow. As mentioned previously, such flows coupled to the local evaporation rate provide the local surfactant concentration limit and the corresponding local surface tension and adsorbed layer affecting the evaporation.

The evaporation rate shows two particularities, one is the classical decrease of the evaporation rate after depinning and the second in case of surfactant, there is an enhancement appearing at later droplet lifetime which could be a consequence of film evaporation contributing to the evaporation process. The evaporation rate per unit length of contact line does not change significantly despite the change in the contact angle versus surfactant concentration. The substrate temperature enhances the global and evaporation rate per unit length of contact line. The coupled temperature and surfactant effect exhibit a local evaporation rate decrease with surfactant increase at ambient temperature (around 20 °C) and enhancement with surfactant at higher temperature. The particular case at temperature around 30 °C with weak effect of surfactant on the evaporation is explained by the complex coupling phenomena and antagonist effects previously underlined.

Last but not least, it is worth noticing that the surface tension is function of temperature for a variety type of liquid and solute. In this work, we control the temperature of substrate, and for all concentrations, droplets were initially at room temperature. There is surely heat transfer between the substrate and the droplet via triple line, meanwhile there is also energy loss by evaporation. Therefore, it is a challenge to quantify the global energy balance and calculate the exact temperature distribution inside the droplet. Concerning the concentration of droplet, it is not constant but increasing during the lifetime. What we are able to control is the initial concentration. The surface tension versus this increasing initial concentration varies from –72 to –37 mN/m and for the temperature effect varies from –72 to –60 mN/m. The effect of concentration on surface tension is more important than the temperature one but both effects remain the same order of magnitude so the two observed effects are coupled.

Declarations

Author contribution statement

Rachid Bennacer: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.
Xiaoyan Ma: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

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Data availability statement

Data will be made available on request.

Declaration of interest’s statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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