EVAPORATION OF DMF SESSILE DROPS ON STATIONARY AND VIBRATING SUBSTRATES

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ABSTRACT

Contact and interaction between droplet and solid surface is a fundamental transport phenomena problem, with ubiquitous presence in various applications. In this paper, we study the effect of imposing vertical and horizontal ultrasonic vibration on dynamics and evaporation of sessile droplets of dimethylformamide (DMF), a pure volatile model solvent. Droplet contact angle and contact radius are the two main parameters that may change during evaporation. Hence, droplet evaporation may be categorized into different modes: constant angle (CA), constant radius (CR), and a complex combination of CA and CR modes. Imposing substrate vibration affects the evaporation rate and mode by changing the thermodynamics and hydrodynamics of the sessile droplet on the substrate. The former happens by changing the heat transfer coefficient and the latter by pinning or unpinning the droplet from the substrate. Experimental analysis using an optical tensiometer has been conducted for a small DMF sessile drop on a Teflon substrate. Among our results, it is observed that the DMF droplet on a Teflon substrate evaporates in the CR mode until it reaches its receding contact angle. Then, its contact radius recedes to the next equilibrium position. Imposing vertical ultrasonic vibration pins the droplet to the substrate and reduces the receding contact angle, while horizontal ultrasonic vibration unpins it. Furthermore, imposing vibration accelerates the evaporation rate more than 5 times higher than that of the natural convection. The increase is more significant for the horizontal vibration.

INTRODUCTION

Numerous studies have been performed on spreading, deposition, and surface wetting of evaporating liquid droplets on a substrate, e.g. Anderson and Davis [1], Burelbach et al. [2], and Ajaev [3], among others. One influential study that has shed light on the research path in evaporation of sessile droplets is a study conducted by Picknett and Bexon [4], in which two extreme modes of evaporation are identified: constant contact angle (CA) and constant radius (CR) evaporation. Based on their investigation, droplet mass varies linearly with time in the CR mode, while it changes with a power law in the CA mode. In addition, droplet lifetime in the CR mode is shorter unless the contact angle is greater than 140° [4], i.e. the droplet lifetime is affected by the hydrophobicity of the substrate[5]. In reality, droplet evaporation proceeds based on a combination of multiple CR and CA modes, also called stick-slip modes, where stick refers to the CR mode and slip usually denotes the CA mode [6]. The duration and sequence of the CA and CR modes in stick-slip evaporation mechanism is hard to predict and varies with the choice of the liquid and substrate, as well as the ambient conditions [7]. In other words, liquid properties, substrate physiochemical characteristics, as well as the substrate temperature are important parameters that identify the wetting, evaporation, and other phenomena associated with sessile droplets [8-12]. In some cases, after a period of CR evaporation, the droplet may quickly slip or jump to a new position, in lieu of the CA mode. Therefore, the so-called CA, CR, and stick-slip modes only provide qualitative description of sessile droplet evaporation.

Erbil [13] reviewed and discussed some works pertinent to the basic theory and some special topics related to droplet evaporation. Erbil et al. [14] investigated the CA mode of evaporation of sessile droplets of various organic solvents, and Birdi et al. investigated evaporation of sessile droplets of water on smooth substrates, which was found to follow the CR mode [15]. The evaporation rate varies with droplet size, however more recent studies consider other effects, such as substrate temperature and substrate thermal properties [16, 17], and the choice of the ambient gas and pressure [18]. Sefiane et al. [18] showed that by reducing the ambient pressure, water vapor diffusion coefficient increases leading to an increase in evaporation rate [6].

As mentioned above, the CR, CA, or stick-stick modes of droplet evaporation could be quite complex. A theoretical work has analyzed modes of water droplet evaporation on a hydrophobised substrate [19], and some experimental works have demonstrated the existence of these complex modes in most real cases, e.g. [20-22]. For instance, Strauber et al. [22] showed that the lifetime of droplet may not be always constrained by the lifetime in the extreme modes. In other words, since the time of evaporation in CA and CR modes are different, and since the real stick-slip behavior of an evaporating droplet is a combination of CA and CR extreme modes, one may conclude that the lifetime of an evaporating droplet would be in between of those of the extreme modes. However, Strauber et al. showed that this may not be necessarily the case, simply because an evaporating

droplet may not necessarily evaporate according to the extreme evaporation modes.

Apart from such natural effects as oscillations due to capillary waves induced by inclined surface or self-excitation [23, 24], droplet excitation for instance by imposed mechanical vibration is a fundamental research topic because excitation influences dynamics and evaporation of sessile droplets. Therefore, the behavior of sessile droplets could be altered and manipulated by excitation. Considering the fact that in reality solid surfaces are rough and the roughness could affect the wetting phenomena, two different states have been realized and studied in the framework of non-sticky Cassie and sticky Wenzel models. Wetting transition between the Cassie and Wenzel states has been observed by implementing mechanical substrate vibration [25, 26]. Boreyko et al. used vertical vibration to overcome the energy barrier for transition from Wenzel to Cassie state [27]. The dynamics of sessile droplet under vertical vibration has been studied theoretically [28] and a computational fluid dynamics (CFD) analysis has been performed to capture the behavior of horizontal vibration on a droplet [29]. The competition between inertial and Laplace forces causes the formation of capillary waves on the droplet surface. Imposing vertical vibration on a sessile drop at low amplitudes may only result in surface waves, while the droplet is pinned. By increasing the vibration amplitude, the energy barrier (friction force) preventing contact line motion may be overcome, which results in triggering of the stick-slip behavior and therefore the occurrence of both surface oscillations and contact line motion [30, 31]. The excitation by mechanical vibration has some application, as well. Celestini et al. [32] used it to reduce hysteresis of contact angle and to increase the mobility of sessile droplets. Furthermore, contact angle measurement on rough surfaces has been studied by monitoring the droplet roundness under vertical vibration [33]. There are many other works that have employed vibration to manipulate droplet motion, contact angle, and so on, e.g. [34-37]. However, comprehensive studies on the effect of vertical and horizontal vibration on droplet evaporation are still lacking or scarce. The effect of vertical and horizontal vibration on drop-cast liquid films was studied in our previous work [38]. In the present work, to understand the evaporation mechanism of sessile drops on solid substrates subjected to horizontal and vertical ultrasonic vibration, we have conducted a set of experiments. The stick-slip behavior has been captured in all three cases of drop evaporation under natural convection (substrate with no vibration), and the drop excited by horizontal and vertical vibration, through imposed vibration on the substrate. Dimethylformamide (DMF) was used as the model solvent.

THEORETICAL BACKGROUND

We assume that the drop size is smaller than the capillary length, to assure than the gravity force is insignificant and the balancing forces arise from the three binary interfacial tensions, according to the Young's equation. Figure 1 shows some aspects of the stick-slip evaporation mechanism of a sessile drop, where Figure 1a represents the CR extreme mode. Considering the equality of the initial contact angle to equilibrium contact angle θ_0, the droplet may start to evaporate in a pinned manner until its contact angle reaches the receding contact angle, θ^*. Then, its radius decreases (slip), while the contact angle increases but it may or may not reach the initial contact angle (Figure 1b), because under the influence of the surface friction, the drop may stop slipping before reaching the initial equilibrium contact angle. This figure and the terminology associated with it will be used to explain the theory and formulations.

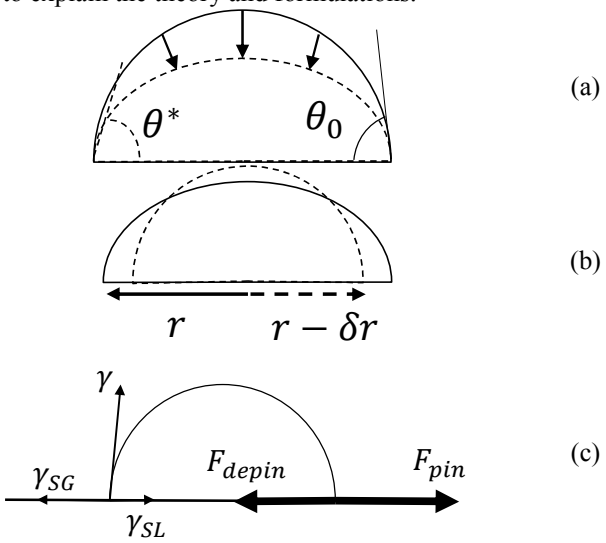

Figure 1. Schematic representation of (a) sessile droplet slipping, b) CR mode of evaporation, and its receding contact angle, and (c) contact forces affecting the droplet pinning.

The volume ($\forall$) and surface area (A) of a sessile drop in the form of a spherical cap (negligible gravity effect) with contact angle and base or contact radius of θ and r, respectively, may be calculated as follows [11]:

$$\forall = \frac{\pi r^3}{3\sin^3\theta}(1-\cos\theta)^2(2+\cos\theta) \quad (1)$$

$$A = \frac{2\pi r^2}{(1+\cos\theta)} \quad (2)$$

In addition, the Gibbs free energy of the droplet, G, due to interfacial free energies, may be written as follows [11]:

$$G = \gamma A + \pi r^2(\gamma_{SL} - \gamma_{SG}) = \gamma\pi r^2\left[\frac{2}{(1+\cos\theta)} - \cos\theta_0\right] \quad (3)$$

where γ, γ_{SL}, γ_{SG} are the liquid-gas, solid-liquid, and solid-gas surface tensions, respectively. Besides, $\gamma_{SL} - \gamma_{SG}$ in the middle part of Eq. (3) was replaced by $\gamma\cos\theta_0$, using the Young's equation for the equilibrium contact angle, θ_0, to arrive at the last part of Eq. (3). Eq. (3) incorporates a contact angle θ, as well as the equilibrium contact angle, θ_0. This implies that the drop may be slightly out of equilibrium (unequilibrated drop), thus the triple line may be under tension due to friction between the

substrate and the pinned drop. The energy associated with this force is not considered in Eq. (3). Based on the aforementioned argument, for the pinned drop shown in Figure 1a, the equilibrium and actual radii are related as $\delta r = r - r_0$, and similarly the equilibrium and actual contact angles are related as $\delta\theta = \theta_0 - \theta$. Using Taylor expansion about the equilibrium state, Shanahan [11] has shown that the excess free energy per unit length of the triple line for this pinning effect may be calculated as follows:

$$\delta\widetilde{G} \approx \frac{\gamma \sin^2\theta_0\,(2+\cos\theta_0)(\delta r)^2}{2r} \tag{4}$$

$$\delta\widetilde{G} \approx \frac{\gamma r(\delta\theta)^2}{2(2+\cos\theta_0)} \tag{5}$$

where Eq. (5) is another form of Eq. (4) obtained by replacing δr by $-rsin\theta\delta\theta/(2+\cos\theta)$ obtained from Eq. (1) at constant drop volume.

On a smooth and homogenous and frictionless (ideal) solid substrate, an evaporating drop would shrink continuously by receding of its contact line to minimize its free energy. However, in many cases it may pin to the substrate due to surface heterogeneity or roughness, creating a corresponding potential energy barrier, U. According to Eq. 5, as the drop evaporates and its actual contact angle decreases, $\delta\theta$ and therefore the excess free energy, $\delta\tilde{G}$, increases until it reaches the potential energy barrier. Then, the droplet depins from the substrate and its radius decreases until it vanishes or pins to another contact radius. This depinning force per unit length of the triple line (Figure 1c) may be related to the excess free energy per unit length of the triple line as follows:

$$F_{depin}\delta r \approx \delta\widetilde{G} \tag{6}$$

The acting forces on the triple contact line are shown in **Error! Reference source not found.**(c), and depinning occurs when the depinning force overcomes the pinning force (F_{pin}), where the depinning force increases as the drop evaporates. Hence, the maximum pinning force per unit length of the triple line occurs at the receding contact angle, θ^*:

$$F_{pin}^{max} = \gamma\cos\theta^* + \gamma_{SL} - \gamma_{SG} \tag{7}$$

In Eq. (7), the potential energy barrier can be equated to the work done by maximum pinning force as follows:

$$U = F_{pin}^{max}\delta r \tag{8}$$

Imposing an external energy or excitation source, such as mechanical vibration, to the substrate would influence the force balance, as well as the rate of evaporation.

MATERIALS AND METHODS

DMF solvent, purchased from Sigma-Aldrich, USA with 99.9% purity, was used as the model liquid. Teflon substrates with dimensions of 50 mm × 50 mm were cleaned with detergent in an ultrasonic bath and dried in a vacuum furnace, as well as in a UV-Ozone cleaner. All substrates were cleaned and treated based on the same procedure to keep the substrate surface energies the same in all experiments. In all cases, the drop initial base radius was about 3.2 mm. A piezoelectric transducer with ultrasonic frequency of 40 KHz installed inside a metal box was used to generate ultrasonic vibration. The substrates were placed and secured atop the vibrating box. The piezoelectric ceramic generates vertical vibration normal to the metal box surface. The direction of vibration was changed to horizontal by titling the metal box by 90º. The vibration power was set to 50 W, using a signal generator, creating vertical vibration amplitude of 56 nm on the substrate [39]. A Theta Lite Optical Tensiometer (Biolin Scientific, Sweden) was used to measure DMF surface tension, as well as the drop base radius and contact angle. Surface energy of the Teflon substrate was measured by Zisman method by measuring the contact angle of two different liquids drops (2-propanol and DMF), using the same substrate. Hence, the solid-gas surface tension of Teflon was obtained as $\gamma_{SG} = 26.715 \pm 0.45$mN/m

RESULTS AND DISCUSSION

Time varying contact angle and base radius of an evaporating DMF sessile drop on Teflon substrate is shown in Figure 2. Consistent with the theoretical considerations, the contact angle decreases as the drop evaporates, while it is pinned to the substrate until its excess free energy exceeds the potential energy barrier at which point the base radius jumps to another position at (receding) contact angle of $\theta^* \approx 39^\circ$. At this point, the contact angle increases and attains a new value that is observed to be much smaller than the initial contact angle. This stick-slip scenario occurs multiple times with varying time duration until the drop vanishes.

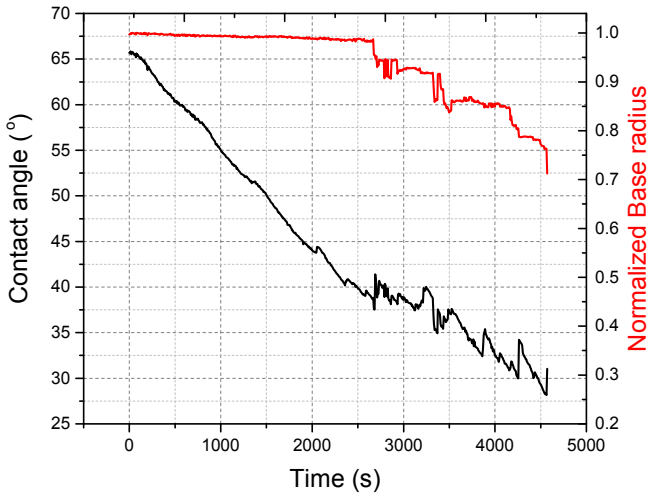

Figure 2. Evaporation behavior of a DMF sessile drop on Teflon substrate subjected to natural convection. The left axis shows the contact angle and the right axis shows the normalized base radius.

Figure 3 displays three consecutive pictures of an oscillating sessile drop subjected to vertical ultrasonic vibration. As a result of the imposed vibration, the height, base radius, and the contact angle oscillate. Figure 4 shows the time variation of the base radius and contact angle of an evaporating sessile drop that is subjected to vertical vibration. The contact angle oscillates with higher amplitudes, compared to that of the base radius and the oscillations damp with time. Figure 4 shows that as the drop evaporates, the contact angle decreases, while it is pinned. A continuous decrease in contact angle finally results in accumulation of excess free energy sufficient to overcome the potential energy barrier at (receding) contact angles of $\theta^* \approx 25^\circ$, and thus the drop slips. By comparing the instant of the occurrence of the first unpinning event in Figs 2 and 4, one may conclude that the presence of the vertical vibration pins the droplet to the substrate more effectively and down to a smaller contact angle compared to the case of natural convection. All these sequential phenomena occur about 5 times faster than that of the natural convection case (Figure 2), thus imposed vertical vibration significantly increases the evaporation rate.

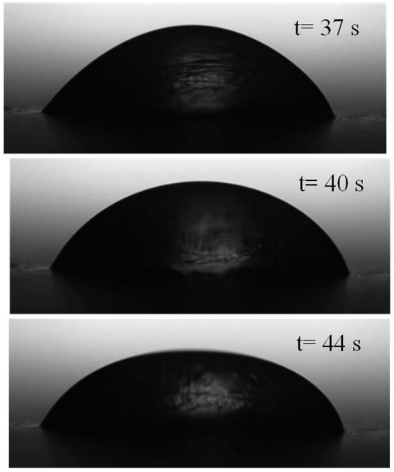

Figure 3. Representative images of drop oscillations, where the substrate is excited by vertical ultrasonic vibration.

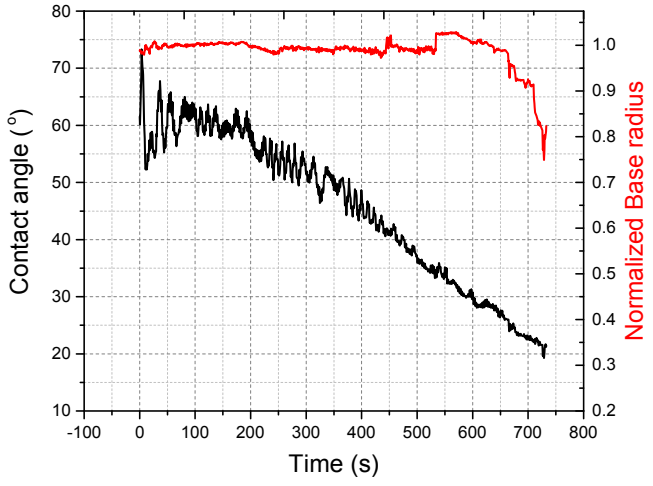

Figure 4. Evaporation behavior of a DMF sessile drop on Teflon substrate subjected to 50 W vertical ultrasonic vibration. The left axis shows the contact angle and the right axis shows the normalized base radius.

Figure 5 shows the time variation of the base radius and contact angle of a sessile drop subjected to horizontal vibration. It is observed that the drop, with the same size, vanishes faster and therefore the evaporation rate is higher than the two previous cases. A tentative theory explanting the reason behind this for thin liquid films has been discussed in our previous work [38]. In brief, horizontal vibration results in a more effective air circulation above the liquid-air interface than that created by vertical vibration. Another observation is a more profound drop oscillation induced due to horizontal vibration at large contact angles compared to the case of vertical vibration. It is noted that the fluctuations in

Figure 5 after depinning of the contact line at contact angle of $\theta^* \approx 36^\circ$ and t = 266 s are not due to vibration-induced drop oscillation, instead these fluctuations are due to a rapid change in the evaporation modes of stick-slip phenomenon.

According to our measured data, two parameters may be defined as follows: (1) the frequency of drop oscillation and its relationship with the frequency of the imposed vibration; (2) drop stiffness or damping factor. Other topics which are currently under investigation include the effect of vibration on the Gibbs free energy, potential energy barrier, depinning force, and the evaporation rate of a variety of entirely volatile, as well as solution drops.

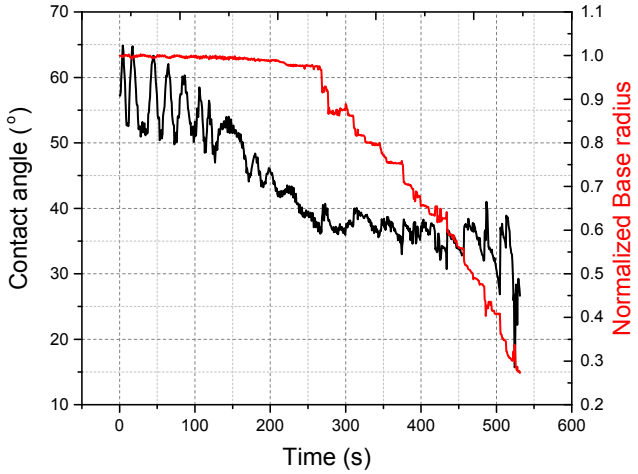

Figure 5. Evaporation behaviour of DMF drop on Teflon substrate subjected to 50 W horizontal vibration. The left axis shows the contact angle and the right axis shows the normalized base radius.

CONCLUSIONS

In this paper, droplet dynamics and evaporation behavior of DMF sessile drops placed on Teflon substrates was studied. In some cases, the drop was excited by ultrasonic vibration by putting the substrates on a vibrating surface. It was observed that the imposed vibration results in significant oscillations in droplet height and contact angle, which damp with time as the drop

evaporates. Also, it was evidenced that the imposed ultrasonic vibration increases the evaporation rate, significantly, where the horizontal vibration has a more significant effect than vertical vibration. Further research is in progress to define a stiffness coefficient for droplet oscillation, and develop explanatory theories on how vertical and horizontal vibrations affect the droplet dynamics and evaporation.

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