

# Theoretical analysis of the lifetime of sessile evaporating droplet with surface cooling effect

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**Abstract** - Liquid droplets are ubiquitous in daily life and industries, therefore the insight of the evolution of droplet evaporation can be of great significance on practical applications. So far the majority of studies on the evaporation of sessile drops are based on the isothermal quasi-steady cases, in which the temperature distribution is assumed uniform along the droplet interface. However, in the actual process, due to the influence of uneven evaporation flux along the air-liquid interface, there is significant variation of temperature along the air-liquid interface, leading to longer lifetime of evaporation. In this paper, by taking account into the interfacial cooling on the basis of isothermal model, the new theoretical model for sessile droplet evaporation with surface cooling effect is built up in toroidal coordinate, three evaporation modes are analysed during the evaporation lifetime, including “Constant Contact Radius”(CCR) mode, “Constant Contact Angle”(CCA) mode and “Stick-Slip”(SS) mode. The dimensionless number  $E_0$  is introduced to indicate the strength of the evaporative cooling, and it is defined based on the thermal properties of the liquid and the atmosphere. It is found that the larger the dimensionless number  $E_0$  is, the longer the lifetime of three evaporation modes is; The variation of droplet volume over time still follows “2/3 power law” in the CCA mode, as in the isothermal model without the cooling effect; In addition, the correction factor for predicting instantaneous volume of the droplet is also derived, which illustrate the difference between the isothermal model and non-isothermal models. These findings may be of great significance to explore the dynamics and heat transfer of sessile droplet evaporation.

**Keywords:** Droplet evaporation; Interfacial cooling; Theoretical analysis; Lifetime.

## 1. Introduction

Liquid droplet evaporation has wide applications in daily life and industries, such as ink-jet printing[1], DNA chip manufacturing[2], and spray-cooling technology[3], etc. Therefore, a comprehensive understanding the characteristics of the evolution of droplet evaporation is getting more and more attention from researchers. In pioneer work on the evaporation of sessile droplets, Picknet and Bexon[4] calculated the lifetimes of droplets evaporating in two extreme modes of evaporation, namely the CCR (Constant Contact Radius) mode and CCA (Constant Contact Angle) mode. Later Bourgès-Monnier and Shanahan [5] found droplets can evaporate in combined mode, “Stick-Slip”(ss) mode, in which initially the drop evaporates in CCR mode until the contact angle reaches a transition value  $\theta_{tr}$ , then the CCA mode dominates in the later stage. For simplification, the isothermal quasi-steady state assumption is usually made[6-7]. In this assumption the effect of surface cooling due to evaporative heat transfer is ignored, causing the uniform temperature and vapor concentration distribution along the droplet interface[8-9]. However, in the actual process, due to the surface cooling effect, uneven the evaporation flux along the air-liquid interface can lead to uneven temperature distribution. Dash and Garimella [10] found the lifetime of droplet evaporation is longer than that predicted by the previous theoretical predictions. Nguyen et al [11] derived an expression of evaporation flux along the air-liquid interface by considering the effect of surface cooling, but the lifetime is not provided. The aim of the present work is to derive the explicit expressions of lifetime of different evaporation modes, and hence gain further insight into the characteristics of the evolution of droplet evaporation. These findings may be of great significance to explore the dynamics and heat transfer of sessile droplet evaporation.

## 2. Theoretical Analysis

When a sessile droplet is sufficiently small, the effect of gravity is negligible compared with that of surface tension, then the droplet takes the shape of spherical cap with a contact angle, its boundary can be mapped in toroidal coordinates( $\alpha, \beta$ ), as shown in Fig.1.

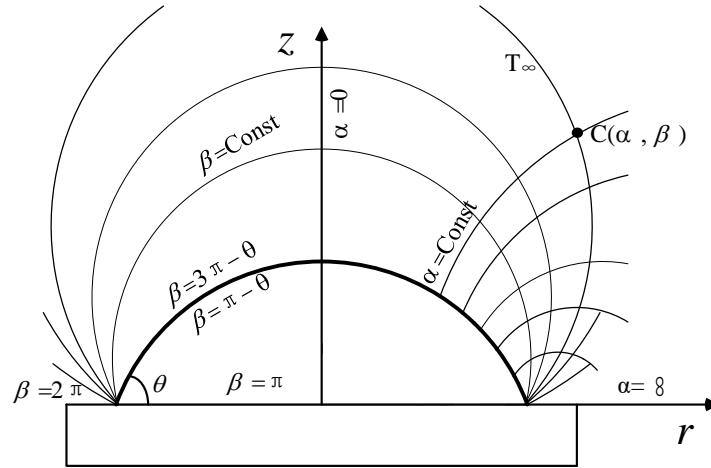


Fig. 1: Schematic diagram of a sessile evaporating droplet in toroidal coordinate.

The relationship between the toroidal coordinate( $\alpha, \beta$ ) and the cylindrical coordinate ( $r, z$ ) is shown below.

$$r / \sinh \alpha = z / \sin \beta = R_0 (\cosh \alpha - \cos \beta)^{-1} \quad (1)$$

where  $R_0$  is the base radius of droplet.

Considering the effect of evaporative cooling due to evaporative heat transfer at the liquid-vapor interface, Nguyen et al[11] has derived an expression of evaporation flux along the interface and temperature field within the droplet, as shown below.

$$j(\alpha) = \frac{D(c_s - c_\infty)}{R} \sqrt{2} (\cosh \alpha + \cos \theta)^{1.5} \int_0^\infty E_c(\tau) P_{i\tau-0.5}(\cosh \alpha) \left[ \frac{\cosh[(\theta - \pi)\tau] \sin \theta}{2(\cosh \alpha + \cos \theta)} - \tau \sinh[(\theta - \pi)\tau] \right] d\tau \quad (2)$$

$$\hat{T}(\alpha, \beta) = \frac{T(\alpha, \beta) - T_{sub}}{T_{sub} - T_\infty} = (2 \cosh \alpha + 2 \cos \theta)^{0.5} \int_0^\infty E_T(\tau) P_{i\tau-0.5}(\cosh \alpha) \sinh \theta \tau d\tau \quad (3)$$

$E_c(\tau)$  and  $E_T(\tau)$  are functions of the integration dummy, respectively.

$$E_c(\tau) = \frac{\cosh \theta \tau (\tau F \coth \theta \tau - dF / 3d\theta)}{\cosh \pi \tau \cosh[(\theta - \pi)\tau] [(\tau F \coth \theta \tau - dF / 3d\theta) - E_0(\tau F \tanh[(\theta - \pi)\tau] - dF / 3d\theta)]} \quad (4)$$

$$E_T(\tau) = \frac{E_0 \cosh \theta \tau (\tau F \tanh[(\theta - \pi)\tau] - dF / 3d\theta)}{\cosh \pi \tau \sinh \theta \tau [(\tau F \coth \theta \tau - dF / 3d\theta) - E_0(\tau F \tanh[(\theta - \pi)\tau] - dF / 3d\theta)]} \quad (5)$$

where

$$F = \sin \theta \tau / (\sinh \pi \tau \sin \theta) \quad (6)$$

$$dF / d\theta = (\tau \cosh \theta \tau \sin \theta - \sinh \theta \tau \cos \theta) / (\sinh \pi \tau \sin^2 \theta) \quad (7)$$

Here  $\tau$ ,  $P_{i\tau-0.5}$  are integration dummy and Legendre functions of the first kind respectively;  $c_s$  and  $c_\infty$  are the saturated vapor concentration at the substrate temperature and at the infinity respectively;  $E_0$  is the evaporative cooling number, as defined in Eq.(8). The larger the magnitude is, the stronger the cooling effect is.

$$E_0 = bLD / k \quad (8)$$

where  $b$  is the thermal gradient of vapor saturation concentration over temperature,  $L$  is liquid latent heat of vaporization,  $D$  is coefficient of vapor diffusion in air,  $k$  is liquid thermal conductivity. The case of  $E_0=0$  represents the idealized model without surface cooling effect.

Integrating the evaporation flux in Eq.(2) over the liquid-gas interface in toroidal coordinates, the expression of evaporation rate can be obtained

$$\begin{aligned} \frac{dM}{dt} &= \rho_L \frac{dV}{dt} = - \int_0^\infty J(\alpha) * 2\pi R^2 \frac{\sinh \alpha}{(\cosh \alpha + \cos \theta)^2} d\alpha \\ &= -D(c_s - c_\infty) 2\sqrt{2}\pi R \times \\ &\int_0^\infty d\tau \int_0^\infty (\cosh \alpha + \cos \theta)^{-0.5} \sinh \alpha [E_c P_{-0.5+i\tau}(\cosh \alpha) \left[ \frac{\cosh[(\theta - \pi)\tau] \sin \theta}{2(\cosh \alpha + \cos \theta)} - \tau \sinh[(\theta - \pi)\tau] \right]] d\alpha \end{aligned} \quad (9)$$

The double integral term is a function of  $\theta$  and  $E_0$ , namely

$$\frac{dM}{dt} = \rho_L \frac{dV}{dt} = -D(c_s - c_\infty) 2\sqrt{2}\pi R \varphi(\theta, E_0) \quad (10)$$

$$\begin{aligned} \varphi(\theta, E_0) &= \int_0^\infty d\tau \int_0^\infty (\cosh \alpha + \cos \theta)^{-0.5} \sinh \alpha \\ &[E_c P_{-0.5+i\tau}(\cosh \alpha) \left[ \frac{\cosh[(\theta - \pi)\tau] \sin \theta}{2(\cosh \alpha + \cos \theta)} - \tau \sinh[(\theta - \pi)\tau] \right]] d\alpha \end{aligned} \quad (11)$$

The volume of the droplet is

$$V = \frac{\pi R^3}{3g(\theta)} \quad (12)$$

where

$$g(\theta) = \frac{\sin^3 \theta}{(1 - \cos \theta)^2 (2 + \cos \theta)} \quad (13)$$

The transient variation of droplet volume  $V$  over time  $t$  can be given as

$$V^{2/3} = V_0^{2/3} - \frac{2\pi D(c_s - c_\infty)}{3\rho_L} \left(\frac{3}{\pi}\right)^{1/3} (g(\theta))^{1/3} 2\sqrt{2}\varphi(\theta, E_0)t \quad (14)$$

where  $V_0$  is the initial volume of droplet.

For the isothermal model of droplet evaporation, the transient variation of droplet volume over time is

$$V^{2/3} = V_0^{2/3} - \frac{2\pi D(c_s - c_\infty)}{3\rho_L} \left(\frac{3}{\pi}\right)^{1/3} (g(\theta))^{1/3} f(\theta)t \quad (15)$$

where

$$f(\theta) = \frac{\sin \theta}{1 + \cos \theta} + 4 \int_0^\infty \frac{1 + \cos 2\theta\tau}{\sinh 2\pi\tau} \tanh[(\pi - \theta)\tau] d\tau \quad (16)$$

So the correction factor  $\kappa$  can be defined to illustrate the difference between the isothermal model and “Brand-new” model.

$$K = \frac{2\sqrt{2}\varphi(\theta, E_0)}{f(\theta)} \quad (17)$$

In CCA mode, the lifetime of droplet can be obtained

$$t_{CCA} = \frac{\rho_L R_0^2}{D(c_s - c_\infty) 4\sqrt{2}\varphi(\theta_0, E_0)g(\theta_0)} \quad (18)$$

In CCR mode, the lifetime of droplet also can be derived, as follow

$$t_{CCR} = \frac{\rho_L R_0^2}{2\sqrt{2}D(c_s - c_\infty)} \int_0^{\theta_0} \frac{1}{\varphi(\theta, E_0)(1 + \cos \theta)^2} d\theta \quad (19)$$

Combine the two equations above, the lifetime of droplet evaporates in the “Stick-Slip”(ss) mode can be obtained

$$t_{ss} = \frac{\rho_L R_0^2}{2\sqrt{2}D(c_s - c_\infty)} \left[ \int_{\theta_r}^{\theta_0} \frac{1}{\varphi(\theta, E_0)(1 + \cos \theta)^2} d\theta + \frac{1}{2\varphi(\theta_r, E_0)g(\theta_r)} \right] \quad (20)$$

### 3. Results and discussion

#### 3. 1. Validation of current model

To verify the correctness of the current analytical model, the predicted values of evaporation rates with the “Brand-new” model(solid lines) are compared with the experimental measurement (data points)[12-13], as shown in Fig.2. The

evaporative cooling numbers  $E_0$  for acetone are 1.03 at ambient temperature  $T=295\text{K}$  and ambient pressure  $P=99.8\text{kpa}$ . It is clearly seen that the predicted results agree quite well with the experimental results for three types of liquid droplets.

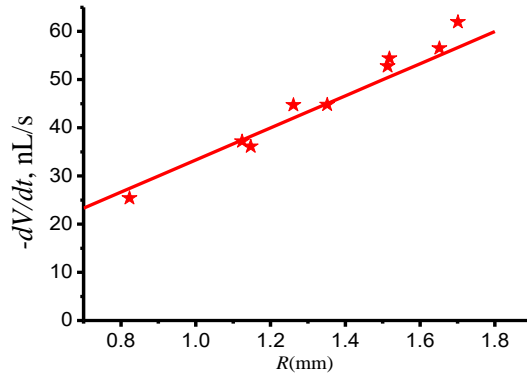


Fig. 2: Comparison of total evaporation rates over different base radii between current theoretical model and experimental data.

### a. Droplet Evaporation in CCR Mode

For the CCR mode, the contact angle gradually decreases from  $\theta_0$  to 0, the expression of droplet evaporation lifetime at this stage is shown in Eq.19. In this section, the effects of significant parameters such as contact angle and evaporative cooling number on the lifetime are provided, the values of initial contact angle are  $90^\circ$  and  $120^\circ$  respectively; the values of evaporative cooling numbers are 0 and 3 respectively. For the accuracy of comparison, the initial droplet volumes are identical under different contact angles, the dimensions are all normalized with the base radius at contact angle  $\theta_0 = 90^\circ$ . The lifetimes at different stages are normalized with the maximum value, which is the case of evaporating in CCA mode at  $\theta_0 = 120^\circ$  and  $E_0 = 3$ . As shown in Fig.3, the larger the initial contact angle is, the longer the lifetime. It is also found that with the increasing initial contact angle, the effect of the evaporative cooling number on lifetime becomes larger.

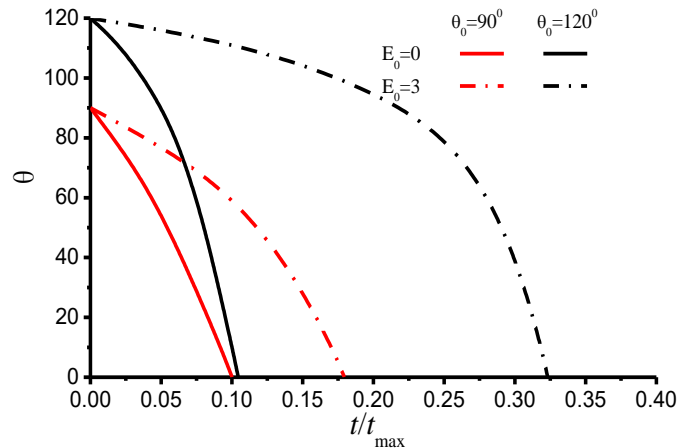


Fig. 3: Transient variations of contact angle for droplets under different initial contact angles  $\theta_0$  and evaporative cooling numbers  $E_0$  in the CCR mode ( $t_{\max}$  is the lifetime at CCA mode at  $\theta_0 = 120^\circ$  and  $E_0 = 3$ ).

### b. Droplet Evaporation in CCA Mode

In this section, the variations of droplet's radius over evaporation time are analysed in CCA mode, as shown in Fig.4. With the increasing initial contact angle, the lifetime becomes longer; In addition to that, for the case of larger initial contact angle, the effect of evaporative cooling number can be more significant on lifetime. When evaporative cooling number  $E_0$  increases from 0 to 3, for the case of initial contact angle  $\theta_0 = 90^\circ$ , the evaporation time is increased by less than 4 times, however, for the case of  $\theta_0 = 120^\circ$ , it is increased by 10 times.

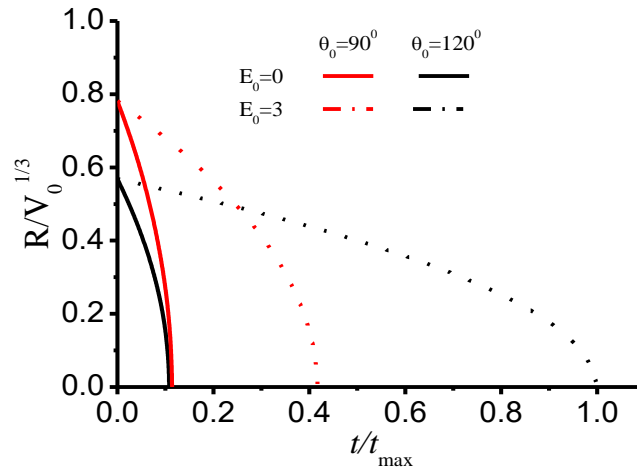


Fig. 4: Transient variations of the scaled contact radius  $R/V_0^{1/3}$  for droplets under different initial contact angles  $\theta_0$  and evaporative cooling numbers  $E_0$  in the CCA mode.

Under isothermal case without surface cooling effect, it is known that  $V^{2/3}$  decreases linearly over time in the CCA mode, as seen in Eq.(15). In non-isothermal case with the evaporative cooling effect, this rule still holds, as seen in Eq.14. Fig.5 shows the quantitative linear relationship between the scaled volume  $(V/V_0)^{2/3}$  and scaled time under different initial contact angles at evaporative cooling number  $E_0=1$ , so it is proved that in the non-isothermal model with surface cooling effect, the so-called “2/3 power law” of volume and time still holds.

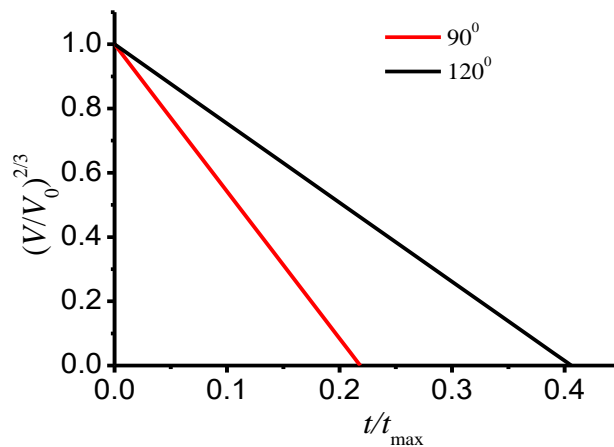


Fig. 5: Transient variations of the scaled volume  $(V/V_0)^{2/3}$  for droplets under different initial contact angles  $\theta_0$  at evaporative cooling number  $E_0 = 1$  in the CCA mode.

**c. Droplet Evaporation in “Stick-Slip” (ss) Mode**

The “Stick-Slip” (SS) Mode consists of the CCR phase followed by CCA phase, as expressed by Eq.20. The overall lifetime of this mode is closely related to initial contact angle, transition contact angle, base radius and evaporative cooling number, based on the above analysis. In this section, the effects of transition contact angle and evaporative cooling number on the lifetime are analysed in details. Fig.6 shows the lifetimes of two stages of “Stick-Slip” mode over transition contact angles under different evaporative cooling numbers at initial contact angle  $\theta_0 = 120^\circ$ . The dash line is the first stage, namely CCR mode, and the solid line is the second stage, namely CCA mode. It can be clearly seen that the relation between the lifetimes of two stages and transition contact angle are almost linear, where the CCR mode is negatively correlated. In contrast, the CCA mode is positively correlated. It is also worth noting that for the case of fixed transition contact angle, the larger the evaporative cooling number is, the longer the lifetimes of two stages, especially for the CCR mode.

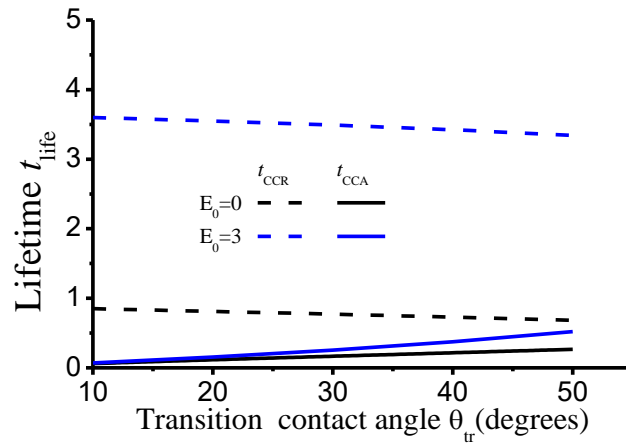


Fig. 6. The lifetimes of two stages of “Stick-Slip” mode over transition contact angles under different evaporative cooling numbers at contact angle  $\theta_0 = 120^\circ$ .

Combined the lifetimes of two stages, the overall lifetime of “Stick-Slip” mode can be obtained, as shown in Fig.7. As the transition contact angle increases, the lifetime gradually increases, especially for the high evaporative cooling number. It is also noted that for the case of fixed transition contact angle, as the evaporative cooling number increases, the lifetime dramatically increases.

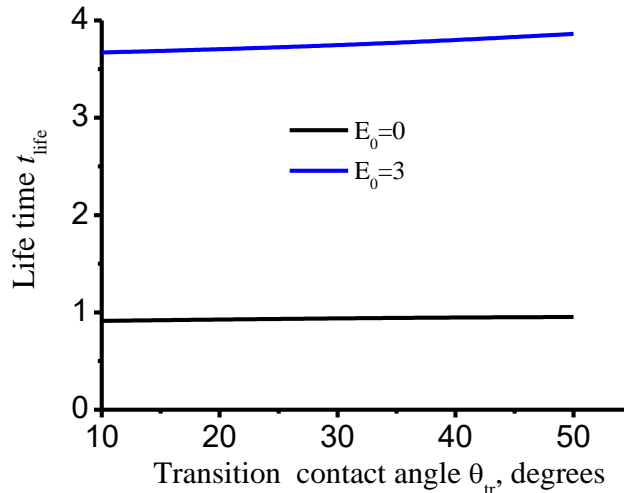


Fig. 7: Evaporating droplet lifetime of “stick-slide” mode over transition contact angles under different evaporative cooling numbers at initial contact angle  $\theta_0 = 120^\circ$ .

#### 4. Conclusion

In this study, the non-isothermal model for the sessile droplet evaporation with surface cooling effect is built up in toroidal coordinate, the temporal variation of droplet volume, contact angle and contact radius are presented under CCR, CCA and SS mode, the following conclusions are obtained.

1. The larger the dimensionless number  $E_0$  can lead to the longer lifetime of three modes of droplet evaporation.
2. The droplet volume over time still follows “ $2/3$  power law” in the CCA mode, as in the isothermal model without the surface cooling effect.
3. In the “SS” mode, the large transition contact angle can reduce the evaporation time in CCR mode, and increase the time in CCA mode, the overall lifetime will be increased
4. The correction factor for predicting instantaneous volume of the droplet is defined and derived which illustrate the difference between the isothermal model and non-isothermal model.

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