



Expressions for the evaporation of sessile liquid droplets incorporating the evaporative cooling effect



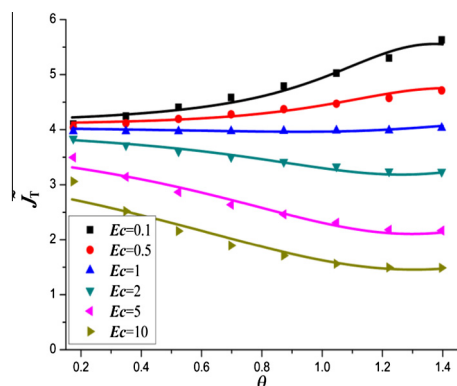
Yilin Wang^a, Liran Ma^b, Xuefeng Xu^{a,*}, Jianbin Luo^b

^a School of Technology, Beijing Forestry University, Beijing 100083, China

^b State Key Laboratory of Tribology, Tsinghua University, Beijing 100084, China

GRAPHICAL ABSTRACT

Simple, yet accurate expressions for the local interfacial evaporation flux and for the total evaporation rate of drying sessile droplets are obtained numerically by using the combined field approach. The results indicate that the variation of the total evaporation rate with the contact angle will change its trend as the evaporative cooling effect becomes significant.



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ABSTRACT

The evaporation along the surface of pinned, sessile droplets is investigated numerically by using the combined field approach. In the present model, the evaporative cooling at the droplet surface which leads to a reduction in the evaporation is taken into account. Simple, yet accurate analytical expressions for the local evaporation flux and for the total evaporation rate of sessile droplets are obtained. The theoretical analyses indicate that the reduction in the evaporation becomes more pronounced as the evaporative cooling number Ec increases. The results also reveal that the variation of total evaporation rate with contact angle will change its trend as the intensity of the evaporative cooling changes. For small values of Ec , the total evaporation rate increases with the contact angle, the same as predicted by Deegan et al. and by Hu and Larson in their isothermal models in which the evaporative cooling is neglected. Contrarily, when the evaporative cooling effect is strong enough, the total evaporation rate will decrease as the contact angle increases. The present theory is corroborated experimentally, and found in good agreement with the expressions proposed by Hu and Larson in the limiting isothermal case.

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* Corresponding author.

E-mail address: xuxuefeng@bjfu.edu.cn (X. Xu).

1. Introduction

The evaporation of sessile droplets is not only related with common, everyday phenomena, but also be of significant importance in many scientific and industrial fields. A comprehensive understanding of the droplet evaporation is urgently needed in the evaporation-based applications, such as the high-throughput automatic DNA mapping [1,2], the disease diagnosis [3], the fabrication of colloidal photonic crystals [4] and the ink-jet printing of functional materials [5–7].

Maxwell [8] firstly developed a basic model that related the droplet evaporation to the vapor diffusion in the quiescent air, and derived an analytical equation for the evaporation of spherical droplets. Picknett and Bexon [9] further investigated the evaporation of sessile droplets and pointed out that the evaporation of drops on a surface mainly includes two distinct phases: a constant contact radius phase in which the contact angle decreases while the contact line is pinned, and a constant contact angle phase where the contact radius recedes while the contact angle remains constant. The pinning of the droplet during evaporation can be attributed to the strong solid–liquid intermolecular interaction of the liquid with the substrate, which may be the result of the re-orientation, rearrangements, overturning, and desorption of the substrate surface molecules [10–14]. Since nearly the whole evaporating time often occurs at the first phase, we discuss here the constant contact radius phase in which the contact line is pinned.

By supposing the evaporation is governed by the vapor diffusion in the surrounding air, the droplet evaporation can be regarded as a quasi-steady state since the vapor concentration above the droplet surface adjusts rapidly compared to the time required to evaporation [15]. This means that the vapor concentration in the atmosphere is applicable to the Laplace's equation. By further assuming that the vapor concentration just above the liquid–air surface is saturated and equals to a constant (referred to as the “isothermal model” in the following), an analytic solution for the evaporation flux distribution along the droplet surface was obtained by Picknett and Bexon [9], Deegan et al. [16,17] and Popov [18]. The results indicate that, when the contact angle is not larger than 90°, the evaporation flux along the droplet surface increases from the top center of the droplet to the contact line, and being singular at the droplet edge.

Considering the nonuniform evaporation flux along the droplet surface, Deegan et al. [16,17] indicated that the radically outwards flow inside the droplet should be driven, which can carry the dispersed solids to the vicinity of contact line, and form a ring-shaped stain after the droplet dries out. Moreover, the nonuniform evaporation flux and the droplet geometry will also produce a temperature gradient along the surface. Hu and Larson [19,20] found that such a temperature gradient may generate a convective Marangoni flow inside the droplet and hence alter the deposition pattern, resulting in a center deposit or a uniform deposit. Ristenpart et al. [21] and Xu et al. [22] further discussed the influence of the thermal conductivity and the thickness of the underlying substrate on the direction of the Marangoni flow. Up to now, there is a tremendous amount of research indicating that the deposition pattern is closely related to the distribution of the evaporation flux along the droplet surface [23,24]. Thus, to predict and to control the deposition of drying droplets, the evaporation of sessile droplets has attracted extensive attention in the recent years [25–42].

Hu and Larson [15] computed the vapor concentration in the surrounding atmosphere by using a finite element method and obtained an accurate empirical expression for the evaporation flux of sessile droplets. In their model, the droplet surface is considered to be isothermal and therefore the evaporation is only determined by the vapor diffusion in the air. However, the temperature variation at the liquid–air surface can affect significantly the

droplet evaporation. Due to the temperature drop at the droplet surface, the vapor concentration just above the surface will not always be a constant, and a large discrepancy in evaluating the total evaporation rate of sessile droplet may occur [43–46].

Considering the vapor saturation concentration along the sessile droplet surface to be a function of the liquid temperature there instead of a simply constant, Dunn et al. [47–49], Sefiane et al. [29] and Saada et al. [50] generalized the basic isothermal model to include the thermal effect resulting from the evaporative cooling and thus coupled the problem for the vapor concentration in the surrounding air and that for the temperature in the liquid and the underlying substrate. Sefiane et al. [51,52] further introduced a dimensionless number **SB** which can be used to evaluate the threshold for transition from the isothermal case to a nonisothermal one. Unifying the coupled physics fields in the liquid evaporation into one single field and making the interaction unnecessary, Xu and Ma [53] presented the combined field approach and derived a dimensionless number **Ec** to identify the intensity of the evaporative cooling. Wang et al. [54] further investigated the combined effects of the evaporative cooling and the underlying substrate on the droplet evaporation, and showed how the evaporative cooling affects the influences of substrate properties on the droplet evaporation.

Because of the pronounced effect of the evaporative cooling on the evaporation, it is evidenced that it should be included in the analytic expressions for the droplet evaporation. Empirical expressions for the evaporation flux and for the total evaporation rate incorporating the evaporative cooling effect would be more useful in evaluating the Marangoni effect and in computing the flow field of drying droplets. In the following sections of the present paper, a mathematical model that includes the evaporative cooling at the droplet interface is first numerically solved. Then, simple, yet accurate analytical expressions for the evaporation flux and for the evaporation rate of drying droplets are obtained. The theoretical prediction is corroborated experimentally, and found in good agreement with the theory presented by Hu and Larson [15]. The present work may contribute to the body of knowledge concerning the droplet evaporation.

2. Mathematic model

Here, we focus our attention on the effect of the evaporative cooling at the droplet surface and neglect the influence of the underlying substrate. We consider a small, pinned, and slowly evaporating sessile droplet with contact angle of θ , contact line radius of R , and thermal conductivity of K_l on a flat isothermal substrate with the room temperature T_0 . The vapor saturation concentration along the droplet surface is assumed to be a linear function of the local liquid temperature, given by $c_{sat}(T) = c_0 + b(T - T_0)$, where $b = \frac{dc_{sat}}{dT} |_{T=T_0}$, and $c_0 = c_{sat}(T_0)$ is the saturation concentration at T_0 (see Fig. 1).

By using the combined field approach introduced by Xu and Ma [53], the equations for the vapor concentration c in the surrounding air and the temperature T in the droplet can be written in a dimensionless form as:

$$\tilde{\nabla}^2 \tilde{T}_1 = 0 \text{ for } 0 \leq \tilde{z} \leq \tilde{h}(\tilde{r}), \tilde{r} \leq 1 \quad (1)$$

$$\tilde{\nabla}^2 \tilde{T}_2 = 0 \text{ for } \tilde{z} \geq \tilde{h}(\tilde{r}), \tilde{r} \leq 1; \tilde{r} > 1 \quad (2)$$

$$\tilde{T}_1 = \tilde{T}_2, \frac{\partial \tilde{T}_1}{\partial \tilde{n}} = \mathbf{Ec} \frac{\partial \tilde{T}_2}{\partial \tilde{n}} \text{ for } \tilde{z} = \tilde{h}(\tilde{r}), \tilde{r} \leq 1 \quad (3)$$

$$\tilde{T}_1 = 0 \text{ for } \tilde{z} = 0, \tilde{r} \leq 1 \quad (4)$$

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