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# Determination of Diffusion Coefficient in Droplet Evaporation Experiment Using Response Surface Method

Xue Chen<sup>1</sup> · Xun Wang<sup>2</sup> · Paul G. Chen<sup>3</sup> · Qiusheng Liu<sup>4,5</sup>

## Abstract

Evaporation of a liquid droplet resting on a heated substrate is a complex free-surface advection-diffusion problem, in which the main driving force of the evaporation is the vapor concentration gradient across the droplet surface. Given the uncertainty associated with the diffusion coefficient of the vapor in the atmosphere during space evaporation experiments due to the environmental conditions, a simple and accurate determination of its value is of paramount importance for a better understanding of the evaporation process. Here we present a novel approach combining numerical simulations and experimental results to address this issue. Specifically, we construct a continuous function of output using a Kriging-based response surface method, which allows to use the numerical results as a black-box with a limited number of inputs and outputs. Relevant values of the diffusion coefficient can then be determined by solving an inverse problem which is based on accessible experimental data and the proposed response surface. In addition, on the basis of our numerical simulation results, we revisit a widely used formula for the prediction of the evaporation rate in the literature and propose a refined expression for the droplets evaporating on a heated substrate.

**Keywords** Droplet evaporation · Diffusion coefficient · Response surface · Marangoni flow · Microgravity

## Introduction

Evaporation of a liquid droplet deposited on a solid substrate has been studied intensively over the past decades due

to its technological importance in a wide variety of applications, such as micro-cooling devices, ink-jet printing and biochemical assays (Sartre et al. 2000; Park and Lee 2003; Bonn et al. 2009; Erbil 2012; Chen et al. 2018). The evaporation of a sessile droplet is a complex free-surface advection-diffusion problem where heat and mass transfer processes occur simultaneously. Though the flow field within an evaporating droplet on a heated substrate is largely determined by surface-tension-induced (Marangoni) flow, the main driving force of the evaporation is the vapor concentration gradient taken between the droplet surface and its ambient gas phase, and by definition, the rate at which the liquid droplet evaporates into the ambient gas scales linearly with the (molecular) diffusion coefficient of the vapor. Therefore, an accurate determination of the diffusion coefficient under various environmental conditions is of great importance to better understand the evaporation process.

Maxwell (1890) was the first to propose a diffusion coefficient model to describe the evaporation rate in a uniform and infinite gaseous medium. Since then, many researchers have followed the same approach to study the evaporation of a spherical drop. Morse and Pierce (1903) and Langmuir (1918) performed the experiments of suspended iodine drops and obtained a linear scaling

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of the evaporation rate with the droplet radius. Picknett and Bexon (1977) analyzed the problem of a droplet resting on a solid substrate by using an analogy between diffusive flux and electrostatic potential. The linear scaling expression included a diffusive term (diffusion coefficient, concentration distribution) and a term representing a coupling between the liquid and the solid phases (initial contact radius and a function of contact angle). However, the question of how the function is related to the contact angle and other parameters remains controversially debated (Birdi et al. 1989; Rowan et al. 1995; Hu and Larson 2002; Popov 2005); Hu and Larson (2002) put forward an intuitively simple expression through a contact-angle function, which is valid for angles between 0 and  $\pi/2$ .

In all of the above-mentioned studies, the thermal properties of the substrate, as well as the effect of evaporative cooling, have not been taken into account. Recent studies (Dunn et al. 2009; Brutin et al. 2010; Bouchenna et al. 2017; Chen et al. 2017a, b) highlighted those effects on the structure of bulk fluid flow in evaporating drops. Ristenpart et al. (2007) showed that the Marangoni flow is determined by both relative thermal conductivities and relative substrate thicknesses; however, the study was limited to the case of a non-heated substrate. Very recently, Chen et al. (2017a) obtained simple scaling laws for the evaporation rate that scales linearly with the drop radius but follows a power-law with the substrate temperature. Combining numerical simulations with response surface method, Chen et al. (2017b) further investigated the thermal effects of the substrate on the Marangoni flow and clearly identified three characteristic bulk flow structures. These recent studies called for an improved empirical relation of Hu and Larson (2002) for the prediction of the evaporation rate.

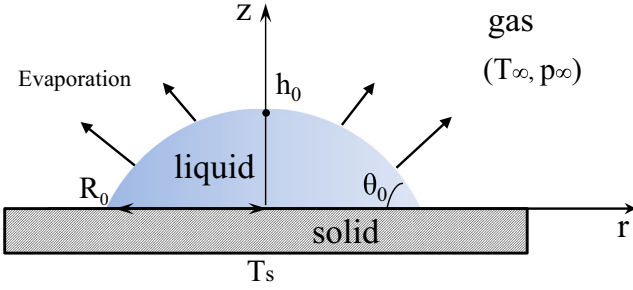
Besides, it is not feasible in space evaporation experiments to insert the instruments allowing a precise measurement of the diffusion coefficient. Given the uncertainty associated with the diffusion coefficient in a specific experiment in space, the question then arises as to what values should be used. The failure of some values used in the numerical and theoretical predictions, when compared with the experimental data, indicates that the diffusion coefficient is largely affected by the environmental conditions (Erbil and Dogan 2000; Carle et al. 2016). Thus, predicting reliably the diffusion coefficient is primarily of importance for a thorough understanding of the evaporation process. The present work is a first attempt to address this issue. To this end, we construct a continuous function of output using response surface method based on a Kriging-based response surface (Krige 1951; Matheron 1963), which allows to use numerical results as a black-box with a small amount of input and output data. This approach has been effectively used to deal with model parameter uncertainties in fluid

dynamics and acoustic problems (Echard et al. 2011; Gayton et al. 2003; Wang et al. 2017). With the Kriging-based response surface, output result for any input model data can be easily estimated without further numerical computation. Relevant values of the diffusion coefficient can then be estimated by solving an inverse problem which is based on the experimental data and the proposed response surface. This response surface approach helps to interpret the experimental results and has also been applied in recent drop evaporation experiment under microgravity conditions onboard the Chinese scientific satellite SJ-10 (Hu et al. 2014).

The rest of the paper is organized as follows. The problem formulation and the methodology are described in “[Problem Formulation](#)”. Numerical results are presented and discussed in “[Results and Discussion](#)”, followed by conclusions drawn from this work.

## Problem Formulation

We consider a millimeter-sized drop of an initial contact angle  $\theta_0$  and a constant contact radius  $R_0$  resting on a heated substrate in still air, as shown in Fig. 1. The liquid is an Newtonian, incompressible fluid of constant density  $\rho$ , dynamical viscosity  $\mu$  and thermal conductivity  $k$ . The surface tension  $\sigma$  is given by  $\sigma = \sigma_0 - \gamma(T - T_0)$ , where  $\sigma_0$  and  $\gamma$  are the surface tension evaluated at the reference temperature  $T_0$  and its negative surface-tension coefficient, respectively. The bottom of the substrate is maintained at a temperature  $T_s$ , and the gas far from drop is kept at a constant ambient temperature  $T_\infty$ . It is assumed that the axisymmetric sessile droplet maintains a spherical-cap shape and forms a constant contact line. It should be noted that, in practice, any chemical interactions, roughness and defects of the substrate, and the extend of wetting can cause contact angle hysteresis. We consider a pure liquid droplet on an ideal (smooth, perfectly flat and completely homogeneous) solid, a unique value of equilibrium contact angle is then determined by Young’s equation. In other works, the contact angle hysteresis is neglected in our simulations. The assumption of constant-radius model is consistent with the evaporation process that we observed in experiments. In our ground and space experiments, we used a solid substrate and injected sufficient liquid volume to make sure that the contact line is well anchored to the substrate with its radii being at the edge of the substrate. Under such conditions, the evaporation process observed follows essentially the constant-radius mode during most of the drop’s lifetime (90%). Therefore, it can be reasonably assumed that the contact radius  $R$  remains uncharged in our numerical simulations. Notice also that the lifetime of an evaporating drop is obtained by extrapolation to vanishing



**Fig. 1** A sessile droplet evaporating on a heated substrate in still air in a cylindrical coordinate system with radial coordinate  $r$  and axial coordinate  $z$

volume of the drop when compared to the lifetime observed in experiments.

## Governing Equations

The physical problem is governed by the incompressible Navier-Stokes equations and the energy equation for the velocity vector  $\mathbf{u}$ , hydrodynamic pressure  $p$  and temperature  $T$ , in the liquid phase :

$$\rho(\partial_t \mathbf{u} + \mathbf{u} \cdot \nabla \mathbf{u}) = \nabla \cdot [-p\mathbf{I} + \mu(\nabla \mathbf{u} + (\nabla \mathbf{u})^T)], \quad (1)$$

$$\nabla \cdot \mathbf{u} = 0, \quad (2)$$

$$\partial_t T + \mathbf{u} \cdot \nabla T = \alpha \nabla^2 T, \quad (3)$$

where  $\mathbf{I}$  is identity tensor. Note that  $\alpha = k/\rho c_p$  denotes the thermal diffusivity,  $c_p$  is the specific heat capacity.

In the gas phase, vapor and heat transports in the atmosphere are solely by diffusion:

$$\partial_t c = D_0 \nabla^2 c, \quad (4)$$

$$\partial_t T = \alpha \nabla^2 T, \quad (5)$$

where  $c$  stands for the molar concentration of the liquid vapor and  $D_0$  is the vapor-in-air diffusion coefficient. The convection in the gas phase is neglected in this work although we do account for Marangoni flow along the droplet surface. The reason for that is twofold from the point view of fluid dynamics: a very small ratio of mass density between the gas and the liquid (i.e., about 1/800) and negligible buoyancy effects under microgravity conditions.

## Boundary and Initial Conditions

The dynamic boundary condition at the droplet surface is the balance of the normal and tangential stresses:

$$\mathbf{n} \cdot (\mathbf{T} \cdot \mathbf{n}) = -(2\sigma K + p_\infty), \quad (6)$$

$$\boldsymbol{\tau} \cdot (\mathbf{T} \cdot \mathbf{n}) = \gamma \nabla_\Gamma T \cdot \boldsymbol{\tau} \quad (7)$$

where  $\mathbf{n}$  and  $\boldsymbol{\tau}$  are, respectively, the unit normal and tangential vector to the liquid-gas interface (denoted by  $\Gamma$ ),  $\mathbf{T} = -p\mathbf{I} + \mu(\nabla \mathbf{u} + (\nabla \mathbf{u})^T)$  stands for the full stress tensor of the fluid,  $K$  is the mean curvature of the liquid-gas interface, and  $\nabla_\Gamma$  denotes the gradient tangent to the interface.

The heat flux across the interface experiences discontinuity due to the latent heat of vaporization  $H_{\text{vap}}$ :

$$k_g(\nabla T)_g \cdot \mathbf{n} - k_l(\nabla T)_l \cdot \mathbf{n} = j_m H_{\text{vap}}, \quad (8)$$

where  $H_{\text{vap}}$  denotes the latent heat of evaporation. The subscripts  $l$  and  $g$  represent the liquid phase and the gas phase, respectively,  $j_m$  is the local evaporation flux, which can be calculated by Fick's law:  $j_m = -D_0 \nabla c \cdot \mathbf{n}$ .

For the concentration of vapor  $c_{\text{sat}}$  at the drop surface, it assumes to be saturated, defined by

$$c_{\text{sat}}(T) = \frac{p_{\text{sat}}(T)}{\bar{R}T}, \quad \ln\left(\frac{p_{\text{sat}}}{p_{\text{ref}}}\right) = -\frac{H_{\text{vap}}}{\bar{R}}\left(\frac{1}{T} - \frac{1}{T_{\text{ref}}}\right), \quad (9)$$

where  $p_{\text{sat}}$  and  $p_{\text{ref}}$  are the saturated vapor pressure at temperature  $T$  and  $T_{\text{ref}}$ , respectively,  $\bar{R} = 8.31451 \text{ J/(mol K)}$  is the universal gas constant.

The condition of mass conservation across the interface leads to:

$$\rho_l(\mathbf{u} \cdot \mathbf{n} - u_\Gamma) = j_m, \quad (10)$$

where  $u_\Gamma$  is the normal velocity of the interface. Given the total mass evaporation flux  $J_m = \int_\Gamma j_m d\Gamma$ , the normal velocity can be geometrically determined:

$$u_\Gamma = -\frac{J_m}{\pi \rho_l R_0^2} \left( \sqrt{1 - \left(\frac{r \sin \theta}{R_0}\right)^2} - \cos \theta \right) \frac{1 + \cos \theta}{1 - \cos \theta}. \quad (11)$$

By assuming a spherical-cap shape of the droplet with the contact angle  $\theta_0$  and the contact radius  $R_0$ , the initial height  $h_0$  can be deduced by  $h_0 = R_0 \tan(\theta_0/2)$ . At initial time  $t = 0$ , the bulk flow inside the drop is at rest (i.e.  $\mathbf{u} = 0$ ), whereas the pressure within the drop is set equal to the ambient pressure augmented by the Laplace pressure ( $\Delta p = 2\sigma_0/R_s$  with  $R_s = (h_0^2 + R_0^2)/(2h_0)$ ). The initial temperature in the whole domain is set at the ambient temperature  $T_\infty (= 20^\circ \text{C})$ . The droplet evaporates into a non-saturated surrounding air with a relative humidity  $H$ . Then, the initial concentration in the vapor equals to  $c_\infty = H p_\infty / \bar{R} T_\infty$ .

We performed numerical simulations of transient fluid flow together with heat and mass transfer using COMSOL Multiphysics. Numerical procedure and model validation can be found in Chen et al. (2017a, b). Simulation results

are subsequently used to construct a response surface as described below.

## Response Surface

Response surface methodology is to model the behavior of a physical phenomenon versus the influential parameter variation using a certain number of numerical simulation results. The response surface, which is a function of concerned input parameters, is constructed using Kriging method in this paper. The reliability of the Kriging-based response surface is evaluated via the leave-one-out (LOO) cross-validation strategy (Geisser 1975; Saltelli et al. 2010). Using uncertainty quantification analysis can help us numerically predict the change of concerned variables and estimate the unknown input. Here, we simply outline the methodology of the response surface method; more details about its principle and numerical procedure can be found in Chen et al. (2017b). In short, this analysis includes the following three steps:

- First, we select some discrete points of inputs (diffusion coefficient and substrate temperature) and calculate the corresponding evaporation rates via numerical simulation.
- Second, based on the numerical simulation results, a function of evaporation rate with respect to the diffusion coefficient and the substrate temperature is constructed using the Kriging-based response surface.
- Finally, based on this response surface, with a measured output (e.g., the evaporation rate) and the substrate temperature, the diffusion coefficient can be estimated by solving the inverse of the response surface function.

## Results and Discussion

Numerical results are presented for an ethanol droplet evaporating on a heated substrate covered with a thin film Teflon–PTFE. Simulation conditions correspond to our space experiments, in which the substrate temperature, as well as the environmental temperature is measured using a thermocouple. In order to maintain the substrate at a constant temperature  $T_s$ , we inserted a thin film heater and a thermocouple fitted with a proportional-integral-derivative (PID) controller to make sure that the temperature is uniformly distributed on the substrate. It is difficult, however, to measure precisely the environmental humidity; for an ethanol droplet evaporating in still air, the initial humidity is assumed to be 0, meaning that there is no ethanol vapor at the beginning of the experiment. We first present the determination of diffusion coefficient based on response surface method, and then propose an improved formula for the rate of evaporation.

## Determining the Diffusion Coefficient Using Response Surface Method

The diffusion coefficient can be predicted by an indirect method with the known numerical results and the Kriging-based response surface. We first explain the numerical procedure to obtain the unknown parameter  $D_0$ , and then compare the best estimated values with those obtained by a commonly used formulation.

It is realized that the diffusion coefficient is very difficult to measure in experiments, so we need to find a better method to determine its value. In the previous literature (Gatapova et al. 2014; Carle et al. 2016), the diffusion coefficient  $D_0$  mainly depends on temperature  $T$  and pressure  $p_\infty$ :

$$D_0(T, p_\infty) = \frac{(D_0 \cdot p)_{\text{ref}}}{p_\infty} \left( \frac{T}{T_{\text{ref}}} \right)^{3/2}, \quad (12)$$

where  $(D_0 \cdot p)_{\text{ref}} = 1.337 \text{ Pa m}^2/\text{s}$  and  $T_{\text{ref}} = 298.13 \text{ K}$  for ethanol vapor in air. Equation 12 reveals that the diffusion coefficient is proportional to  $T^{3/2}$  and inversely proportional to the atmosphere pressure  $p_\infty$ . Strictly speaking, the interfacial temperature  $T_{\text{int}}$  should be used in Eq. 12. However, the interfacial temperature of an evaporating droplet exhibits a non-monotonic spatial distribution along the droplet surface due to several thermal effects at play, namely Marangoni effect and evaporative cooling on the droplet surface, heat conduction across the substrate and the liquid. In particular, the temperature drop at the liquid interface due to substantial evaporative cooling of a rapid evaporating droplet was found to increase with increasing evaporation rates. Instead of using the interfacial temperature which can only be obtained through a numerical simulation, we use a simplifying approach in two different ways. First, the substrate temperature  $T_s$  is used in Eq. 12, which represents a limiting case for the diffusion coefficient (i.e., its upper bound), and secondly, we use an effective temperature  $T_{\text{eff}} = [\sqrt{T_s T_\infty}(\sqrt{T_s} + \sqrt{T_\infty})/2]^{2/3}$  (Semenov et al. 2017) to compute the diffusion coefficient. This effective temperature which lies between the ambient temperature  $T_\infty$  and the substrate temperature  $T_s$  may be regarded to some extent as a “mean” interfacial temperature. We note that Gatapova et al. (2014) presented an iterative procedure by which the temperature dependence of the diffusion coefficient is taken into account to compute the evaporation rate of a sessile water drop on a heated substrate. While this is an interesting numerical procedure, our aim here is to infer the diffusion coefficient involved in a droplet evaporation experiment using response surface method.

There is also the added difficulty that the ambient pressure might not be measured or the measurements are not reliable, such that the estimation of the diffusion coefficient

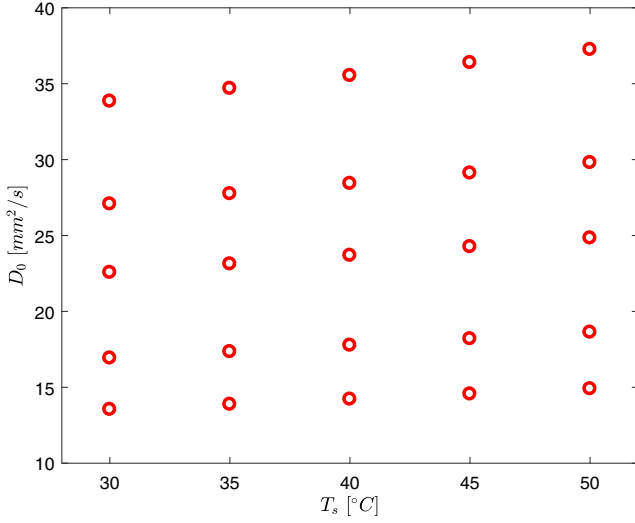


Fig. 2 Sample points in  $(D_0, T_s)$  for constructing response surface

is imprecise. Consequently, the numerical simulations are inconsistent with the experimental results due to the improper input value. For this reason, the response surface method should be applied, in order to give a new way to obtain the unknown value of diffusion coefficient.

To construct response surface, a grid with 25 samples in  $(D_0, T_s)$  is used, the values of the samples are shown by the circles in Fig. 2. The corresponding value of  $p_\infty$ , which is used in the numerical simulation, follows the relation of the parameters in Eq. 12. The required number of grid samples depends on the dimension of input parameters, the physical range of each input parameter and the complexity of response surface. In the considered numerical example, the grid with 25 samples is proven to be reasonable such that the Kriging response surface converges; we have tested cases with additional samples but they do not significantly improve the reliability of response surface. The lifetime of the evaporation  $t_{\text{vap}}$  can be achieved, giving an average rate of evaporation  $|\dot{m}| = \rho V_0 / t_{\text{vap}}$ . Here,  $V_0$  represents the initial volume of the droplet. Then, a 3D response surface of  $|\dot{m}|$ , denoted as

$$|\dot{m}| = f(D_0, T_s), \quad (13)$$

is constructed based on the Kriging method with the numerical simulations. The constructed response surface which represents the evaporation rate variations with different substrate temperatures  $T_s$  and diffusion coefficient  $D_0$ , is shown in Fig. 3. The Kriging-based response surface is validated by the LOO method: the maximum cross-validation is  $\max_s CV_s(|\dot{m}|) = 3\%$ , which means the response surface is reliable.

Note that for a given substrate temperature  $T_s$ , where Eq. 13 can be denoted by

$$|\dot{m}| = f_{T_s}(D_0), \quad (14)$$

if  $f_{T_s}$  is a monotone function, the diffusion coefficient can be uniquely determined by

$$\hat{D}_0 = f_{T_s}^{-1}(|\dot{m}|). \quad (15)$$

Therefore, by solving an inverse problem, i.e., Eq. 15 using the experimental results of substrate temperature and evaporation rate, we can infer the diffusion coefficient.

Six samples of  $D_0$  are assumed to be unknown and estimated by the measurements of  $T_s$  and  $|\dot{m}|$ . The estimates of  $D_0$  obtained from the response surface and the corresponding values computed using Eq. 12 (given  $T_s$  and  $p_\infty$ ) are listed in Table 1. It should be noted that once we get the experimental results of  $|\dot{m}|$  and the imposed substrate temperature  $T_s$ , the response surface method does not require ambient pressure  $p_\infty$  to determine  $D_0$ . Comparing the results in Table 1 indicates that Eq. 12 systemically underestimates diffusion coefficient. For example, if the substrate temperature is imposed at  $T_s = 40^\circ\text{C}$  and the evaporation rate is  $|\dot{m}| = 2.028 \times 10^{-4} \text{ g/s}$ , we obtain a value of  $D_0 = 30.07 \text{ mm}^2/\text{s}$  from the response surface, while Eq. 12 gives a value of  $D_0 = 28.43 \text{ mm}^2/\text{s}$  with  $T_s$  and  $27.06 \text{ mm}^2/\text{s}$  with  $T_{\text{eff}}$ .

## Revisiting the Evaporation Rate

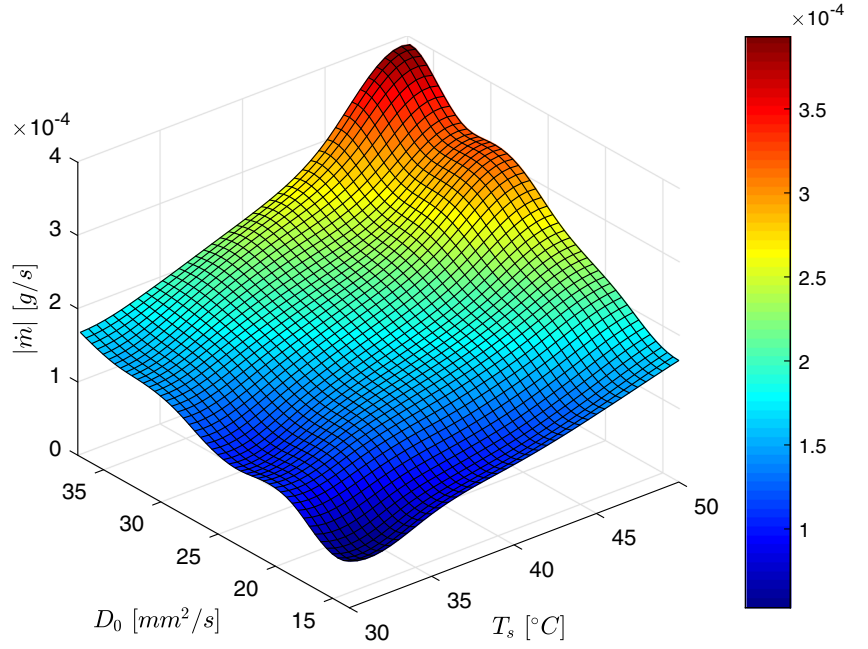
Over the past decades, many efforts have been made to achieve a simplified, empirical formula for the (average) rate of evaporation of a sessile droplet. Indeed, such an empirical formula has been found to be beneficial to drop evaporation experiments as it provides a quick guide to designing and interpreting the experiments. The simplest and thus most widely used formula was proposed by Hu and Larson (2002), based on a theoretical model of quasi-steady diffusion-driven evaporation; it consists primarily of a contact-angle function representing the wettability of substrate. The formula also includes the concentration difference between the droplet surface and the surrounding environment, defined as follows:

$$|\dot{m}| = \pi R_0 D_0 \Delta c f(\theta), \quad (16)$$

where the concentration difference  $\Delta c = c_{\text{sat}}(T_s) - H c_{\text{sat}}(T_\infty)$  and the (dimensionless) contact-angle function  $f(\theta) = 1.3 + 0.27\theta^2$  ( $\theta$  in rad).

It turns out that using Eq. 16 significantly overestimates the average rate of evaporation when compared with the space experimental results. The explanation for this follows by considering a liquid droplet where the temperature at the droplet interface is equal to the heating temperature of substrate. That means the temperature profile assumed by the model is instantly established in the whole droplet volume at the very beginning of evaporation process, which is obviously not the case, as an ambient, non-heated droplet

**Fig. 3** Response surface of  $|\dot{m}|$  as a function of  $D_0$  and  $T_s$



is placed on the substrate during the experiments, and thus it would take some time for the flow of heat to develop. In other words, Hu and Larson's (2002) model underestimates the lifetime of the droplet evaporation, leading to a higher average rate of evaporation. A quantitative analysis is provided below.

On the basis of our numerical results, we attempt to refine this formula taking into account the thermal effects such as substrate heating, evaporative cooling and Marangoni flow, which could allow us to quickly predict the evaporation rate of the space experiments in SJ-10. The key idea was to obtain a similar function which not only varies with the contact angle  $\theta$  but also with the substrate temperature  $T_s$ . To this end, we performed a series of numerical simulations for different initial contact angles  $\theta$ , diffusion coefficient  $D_0$  and substrate temperatures  $T_s$ , resulting in the average rates of evaporation  $|\dot{m}|$  as a function of  $\theta$ ,  $D_0$  and  $T_s$ .

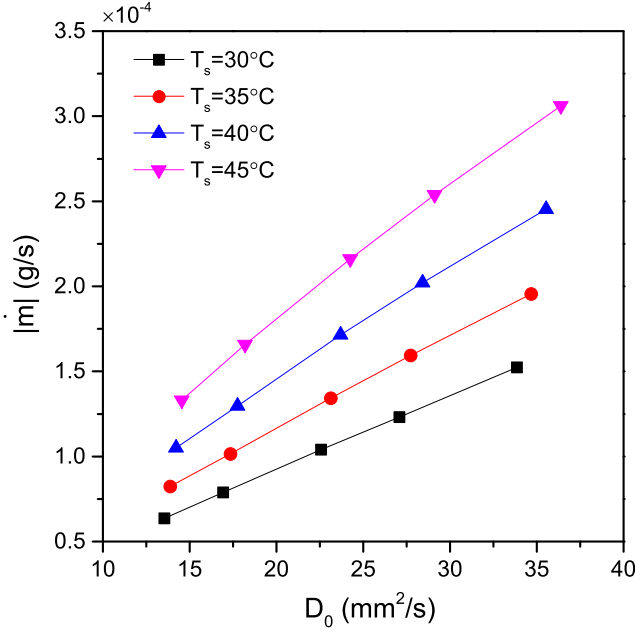
We first checked the relationship between the evaporation rate  $|\dot{m}|$  and the diffusion coefficient  $D_0$  under different heating conditions. Numerical results are shown in Fig. 4. As expected, a linear scaling, i.e.,  $|\dot{m}| \propto D_0$ , is recovered, in accordance with Eq. 16. It is further noted that the evaporation rate increases remarkably with the substrate temperature. This result is consistent with our previous findings (Chen et al. 2017a) that the evaporation rate of droplets could be well fitted by a power law with the substrate temperature, i.e.,  $|\dot{m}| \propto T_s^b$  with the power exponent  $b$  in the range of about 2.5.

Now defining the ratio  $|\dot{m}|/(\pi R_0 D_0 \Delta c) (\equiv f(\theta, T_s))$ , we plot its value as a function of the contact angle for the substrate temperatures ranging from 25 to 40 °C (Fig. 5). Indeed, the value is substantially lower than that given by the contact-angle function proposed by Hu and Larson (2002), and the ratio decreases as the

**Table 1** Diffusion coefficient  $D_0$  (mm<sup>2</sup>/s) obtained from the Kriging response surfaces, for given experimental  $T_s$  (°C) and  $|\dot{m}|$  (g/s), compared with its values from Eq. 12

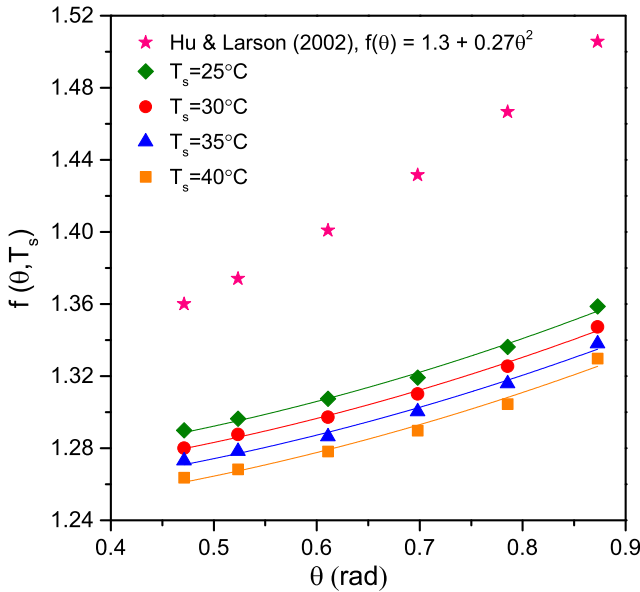
$(T_s,  \dot{m} )$	Determination of $D_0$		
	Using response surface	Using Eq. 12 with $T_s$	Using Eq. 12 with $T_{\text{eff}}$
(32, 1.497)	29.94	27.71	26.54
(32, 0.698)	16.54	13.85	13.27
(40, 2.028)	30.07	28.43	27.06
(40, 1.051)	15.13	14.21	13.53
(50, 3.195)	31.99	29.80	27.71
(50, 1.710)	15.04	14.90	13.85

The initial conditions used in the computations are  $R_0 = 6$  mm,  $\theta_0 = 27^\circ$ ,  $T_\infty = 20$  °C, and  $H = 0$ . Using the substrate temperature  $T_s$  in Eq. 12 represents a limiting case for the diffusion coefficient (i.e., its upper bound) if one tries to use Eq. 12 to infer its value



**Fig. 4** Evaporation rate  $|\dot{m}|$  as a function of diffusion coefficient  $D_0$  for various substrate temperatures  $T_s$ . Initial conditions:  $R_0 = 6$  mm,  $\theta_0 = 27^\circ$ ,  $T_\infty = 20^\circ\text{C}$ , and  $H = 0$

substrate temperature increases, which is consistent with the explanation we described above. We may offer another plausible explanation by the fact that  $|\dot{m}| \propto D_0$ , and at the same time, the concentration difference  $\Delta c$  increases with  $T_s$  as the vapor pressure increases more rapidly than the temperature. Taken together, the ratio  $|\dot{m}|/(\pi R_0 D_0 \Delta c)$



**Fig. 5** Variation of the ratio  $f(\theta, T_s) = |\dot{m}|/(\pi R_0 D_0 \Delta c)$  as a function of initial contact angle  $\theta$  for different heating temperatures. Symbols denote the ratio for different  $T_s$ , lines are the fitted plots representing new function  $f(\theta, T_s) = 1.305 + 0.12\theta^2 - 0.00175T_s$

decreases as  $T_s$  increases, and the contact-angle function of Hu and Larson (2002) represents its upper limit, as shown in Fig. 5.

Finally, by curve fitting to the numerical results, we arrive at a function  $f(\theta, T_s) = 1.305 + 0.12\theta^2 - 0.00175T_s$  ( $\theta$  in rad and  $T_s$  in  $^\circ\text{C}$ ). At first sight, the proposed fitting function  $f(\theta, T_s) = a + b\theta^2 + cT_s$  is physically meaningless owing to the two absolutely independent parameters (the contact angle  $\theta$  and the substrate temperature  $T_s$ ), which, of course, are measured in absolutely different units. However, it should be understood that the function is dimensionless, meaning that the coefficients  $b$  and  $c$  have, respectively, the units of  $\text{rad}^{-2}$  and  $^\circ\text{C}^{-1}$ . With this new function, Eq. 16 could now be extended its application to a droplet evaporating on a heated substrate. Nevertheless, we caution that additional comparisons with other numerical and/or experimental results are needed to assess the validity of this formula under a wide range of conditions.

## Conclusions

In this paper, we have presented a numerical model to investigate the evaporation process of a pinned, millimeter-sized drop deposited on a heated solid substrate. A limited number of numerical simulations have been performed to construct a continuous function of the evaporation rate with respect to the vapor diffusion coefficient in the atmosphere and the heating temperature using the Kriging-based response surface, from which relevant values of the diffusion coefficient involved in recent drop evaporation space experiment are obtained by solving an inverse problem. Our numerical results show that the conventional method systematically leads to a lower value of the diffusion coefficient than that predicted by the present approach. This methodology relies on a combination of numerical simulations and readily accessible experimental data (namely, heating temperature and overall evaporation rate) to determine precisely the values of some key physical parameters related to the experiments, and the proposed numerical procedure is general, so that the methodology could be a useful tool for situations requiring repeated simulations. We have also revisited a widely used empirical formula for the prediction of the evaporation rates of a sessile droplet, and proposed a new, improved empirical relation for the droplets evaporating on a heated substrate. The proposed formula, inspired from Hu and Larson's (2002) work, incorporates a combined thermal effect, namely substrate heating, evaporative cooling and Marangoni flow. Overall, this study provides a new way to simply and accurately determine the diffusion coefficients involved in the drop evaporation experiments and could help better devise future experiments.

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