



## Evaporation of sessile droplets

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### ABSTRACT

Recent developments in the studies of evaporation of liquid droplets placed on a solid substrate are reviewed for the droplet size typically larger than 1 μm, so that kinetics effects of evaporation are neglected. The attention is paid to the limits of applicability of classical diffusion model of evaporation, effect of substrate, evaporation of complex fluids and applicability for its description of the theory developed for pure liquids, and hydrothermal waves accompanying evaporation.

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### 1. Introduction

Evaporation of liquid droplets is omnipresent in our everyday life and in many industrial processes. The examples are cooling and combustion, using sprays in painting, cosmetics and drug delivery etc. (see, for example, Refs. [1,2] and references therein). There are a lot of publications on the evaporation of free suspended droplets (see Refs. [2,3] for review), but the problem complicates considerably for the droplets placed on a solid substrate, because in this case the mass transfer between liquid and vapour is coupled with the heat transfer between three phases involved. Moreover, solid/liquid interaction comes into play and the wetting properties of liquid should be taken into account [1]. In the case of droplets on a solid substrate the early stage of evaporation is coupled with spreading process, whereas later evaporation can cause dewetting of solid [4]. A very comprehensive review on evaporation of droplet of pure liquids, covering more than century of research in the field, was published very recently [5].

The further complication appears for complex liquids, such as surfactant solutions and nano-fluids, as the interactions with constituents of those liquids becomes important [6]. The evaporation of sessile droplets of complex liquids is accompanied by self-assembly and self-organization processes resulting in formation of homogeneous or nano- and micro- structured patterns on solid interface [7,8]. Patterned solid substrates are widely used in industrial processes, such as printing, patterning, fabrication of MEMS (microelectromechanical systems), memory and microelectronic devices, separation of polymers, agrochemistry,

and micro- and nanofluidics. Bottom-up techniques of patterning [9], based on self-assembly and self-organization processes are expected to be cheaper and less energy consuming than most of existing top-down techniques. That is why the understanding and control of evaporation of complex liquids are of great scientific and industrial importance and attract considerable scientific attention.

In this review we discuss the most recent results of study on evaporation of liquid droplets on solid substrates published mainly during the last 3 years (some important results from the previous years are included as well) with special attention paid to the limits of applicability of classical diffusion model of evaporation, evaporation of complex fluids and hydrothermal waves accompanying evaporation.

### 2. Dynamic regimes and kinetics of simultaneous spreading and evaporation

The classical model for theoretical study of evaporation of liquid droplets on solid substrate is based on the assumptions of diffusion controlled mass transfer in gas phase and constant temperature over the whole system (isothermal conditions) [10,11]. In this case evaporation kinetics is governed by the equation:

$$\frac{dm}{dt} = \bar{m} = -2\pi DL\Delta cF(\theta), \quad (1)$$

where  $m$  is the droplet mass,  $t$  is the time,  $D$  is the vapour diffusion coefficient,  $\Delta c$  is the vapour concentration difference between the liquid surface (saturated vapour,  $c_s$ ) and ambient air far from the droplet,  $F(\theta)$  is the function of the contact angle  $\theta$  (see [4,12] for details and

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references), and  $L$  is the contact line radius. It should be stressed that although the evaporation flux has maximum near the three phase contact line (in the case  $\theta < 90^\circ$ ), the proportionality of evaporation rate to the droplet perimeter, but not its surface, is merely the consequence of the diffusion controlled evaporation [4;11;13].

The kinetic mechanisms can be rate controlling in the case of small droplets, smaller than  $1 \mu\text{m}$  [14]. However in what follows only the results for larger droplets are considered and therefore the kinetic effects are neglected.

2.1. Violation of isothermal conditions

The most important contribution made recently to the model Eq. (1) was introducing the thermal effects due to evaporative cooling [12;13;15–20], which slows down the rate of evaporation, i.e. consideration of the fact that the surface temperature  $T_{surf}$  differs from the ambient temperature  $T_\infty$ . The deviations from isothermal case ( $T_{surf} = T_\infty$ ) is considered in [12] in terms of introduced here dimensionless Sefiane–Bennacer (SB) number. The results of this study are presented in Fig. 1 showing in particular, that the evaporation slows down by an increase of the latent heat of evaporation and the substrate thickness as well as by a decrease of the substrate thermo conductivity. The effect of the substrate thermo conductivity has been confirmed by direct numerical simulations of droplet evaporation for both pinned and de-pinned contact line [15;16;17]. The theoretical predictions are in good agreement with the experimental results presented in [17;18]. It is interesting that despite much more intensive evaporation near the three phase contact line, the temperature here is the highest one over the droplet surface [19]. The reason is that the surface at the contact line has the best conditions to get energy from the substrate. The detailed consideration performed in [20] displayed that using the average surface temperature  $T_{surf} = \frac{1}{S} \int T_s dS$  ( $S$  is the surface area,  $T_s$  is the local temperature related to each surface point) enables to bring to one universal curve the numerical data on evaporation of droplets on substrates of various thermo conductivities.

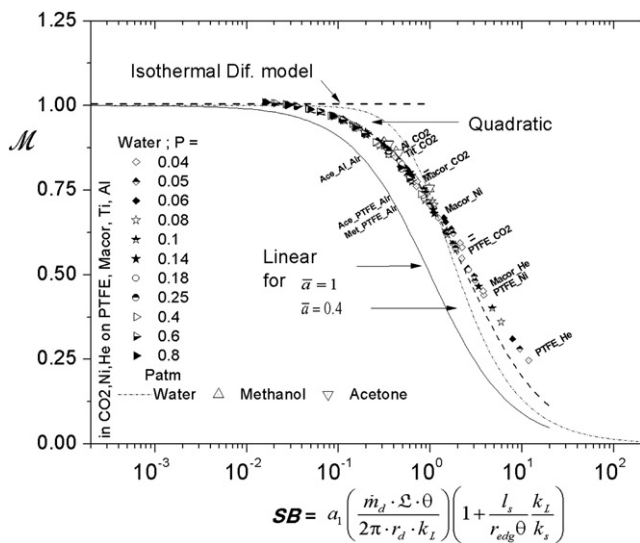


Fig. 1. Dependence of the evaporation rate of sessile droplet normalized by the evaporation rate in isothermal conditions,  $\mathcal{M}$ , on Sefiane–Bennacer number:  $a_1 = \frac{(\partial C_s / \partial T)_{T_\infty}}{(C_s)_{T_\infty}}$ ,  $m_d / \pi r_d = D C_s(T_\infty)$ ,  $\mathcal{L}$  is the latent heat of evaporation,  $k_l$  and  $k_s$ , are thermo conductivities of liquid and solid respectively, and  $r_{edg} \theta$  and  $l_s$  are the effective thicknesses of liquid and solid respectively. Adopted from [12].

2.2. Convection in the vapour phase

The model represented by Eq. (1) assumes diffusion controlled vapour transfer and neglects the effect of convection in vapour phase. Good agreement of the theoretical predictions with experimental results displayed, for example, in [16;21] for water on various substrates and in [22] for the wide range of conditions (five liquids on four coatings providing contact angles from  $17$  to  $134^\circ$ ), can be considered as a confirmation of the validity of this assumption. However, the deviation of the experimental results from the isothermal diffusion-controlled model starts to grow, if the substrate temperature deviates from the room temperature [23]. To be precise, for metallic substrates the diffusion model underestimates the evaporation rate. The possible reason of this deviation is the increasing importance on thermal buoyancy convection at higher temperatures. The results of numerical simulations presented in [24] show that the diffusion model underestimates the evaporation rate by 8.5% at the substrate temperature  $25^\circ\text{C}$  (equal to ambient temperature) and by 27.3% at the substrate temperature  $70^\circ\text{C}$ .

Several other researches have been performed during last years to outline the range of experimental conditions wherein the problem remains diffusion controlled. Two parallel sets of experiments have been performed in the terrestrial and microgravity conditions [25] displaying clearly that the deviations of the experimental results from the model is due to buoyancy driven effects (Fig. 2). The difference between the experimental results and the model predictions is rather small if the substrate temperature is close to the ambient temperature, but it increases considerably by an increase of the temperature difference. According to [26] for the substrate kept at the room temperature the deviation of experimental results from the diffusion model

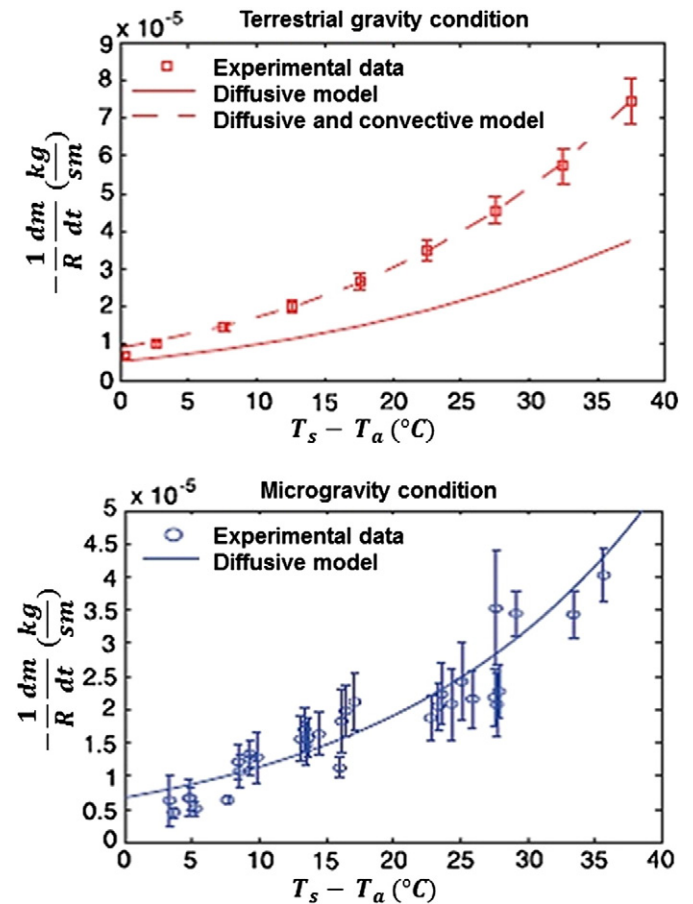


Fig. 2. Evaporation rate of an ethanol sessile droplet as a function of the temperature difference between the substrate and the ambient air for terrestrial and microgravity conditions [25].

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