



Contribution of convective transport to evaporation of sessile droplets: Empirical model



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ABSTRACT

Despite the fact that sessile droplets evaporation dynamics has been studied for more than half a century, the scientific community struggles with the creation of an accurate quantitative description of the rate of evaporation. The classically used description considers evaporation as a quasi-steady process controlled by the diffusion of vapour into the air and the whole system is assumed to be isothermal at the ambient temperature. However, when two types of fluids (alcohols and alkanes) are let to evaporate on heated substrates, their evaporation rates tend to be underestimated by this model, mostly due to convection. This experimental study aims to understand how atmospheric convective transport in the vapour phase influences evaporation in order to develop an empirical model that accurately describes the evaporation rate. The Rayleigh number is used to analyse the contribution of natural convection and an empirical model is developed combining diffusive and convective transport for each type of fluid. The influence of the molecular chain length (and the increasing number of carbon atoms) is also being discussed.

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1. Introduction

Sessile droplets are commonly encountered in daily life, including coffee spills [1], rain on a waterproof raincoat [2], and water falling onto a cooking plate [3]. Thanks to their ubiquity, research about sessile droplets has been conducted for centuries, and the phenomena that affect droplets, such as evaporation, diffusion and fluid mechanics, were studied even earlier. Their intensive use in various scientific and industrial applications (combustion, medicine, chemistry or again environmental processes) raised the need to predict the evaporation time which arose and became one of the main goals of droplets research field. Estimating the evaporation rate can lead to the time of evaporation and the quantity of vapour produced by a droplet. This knowledge is highly valuable for several applications especially in the field of combustion in which fuel is sprayed into a combustion chamber in the form of spherical droplets [4]. Moreover, knowing the evaporation time can allow to easily describe the droplets interface motion as the evaporation occurs [5].

The evaporation flux rate is strongly conditioned by the molecular exchange at the droplet-vapour interface and the diffusion

of the vapour through the air: if molecules stay around the droplets and only move by diffusion, the air around the droplet will become saturated earlier than if molecules are carried away by an air flow. The concentration gradient between the apex *i.e.* the top of the droplet, and the air far from the droplet is the mechanism that drives and limits the evaporation. The hydrodynamic approach considers the evaporation rate being directly proportional to the rate of vapour diffusion since the air at the interface is assumed to be saturated with vapour. Maxwell was the first to describe the evaporation by the mean of a diffusion coefficient [6] and later, evaporation of spherical droplets has been studied with the same approach [7]. More recently, researches in the last decade dealt with the evaporation kinetics [2,8–11] and resulted in models predicting the evaporation rate of sessile droplets [12,13].

These classical descriptions consider evaporation as a quasi-steady process controlled by the diffusion of vapour into the air, and the whole system is being assumed to be isothermal at the ambient temperature. However, in the previous study [14], we have experimentally evidenced the contribution of atmospheric convective transport to the evaporation of a sessile droplet. In the case of diffusive evaporation (at ambient temperature, or on heated substrates under microgravity conditions), diffusive models accurately describe the evaporation rate. However, when convection appears in the vapour phase (substrate temperature above ambient

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Nomenclature

c_x	Vapour concentration, kg/m ³
C_x	Empirical coefficients
D	Diffusion coefficients, m ² /s
L_c	Capillary length, m
M_w	Molar mass, g/mol
N	Carbon number
P_{sat}	Saturated pressure, Pa
R	Initial droplet radius, m
\hat{R}	Ideal gas constant, J/molK
T_{sat}	Saturated temperature, K
α	Thermal diffusivity, m ² /s
γ	surface tension, N/m
θ	Initial contact angle, °
ν	Kinematic viscosity, m ² /s
ρ	Density, kg/m ³

temperature under normal gravity conditions), all these models under-predict the evaporation rate.

Evaporating droplets under microgravity conditions enabled us to study the purely diffusive evaporation with intensive substrate heating, which is impossible under normal gravity conditions. As the buoyancy forces are strongly weakened, the convection process stops and the vapour behaviour is changed from convection rolls that renew the air around the droplet and enhanced the evaporation rate to only diffusion.

Vapour distribution around a sessile droplet have been very recently studied using experimental [15,16] and numerical [17,18] approaches in the case of purely diffusive evaporation. Kelly–Zion et al. [15] have recently shown by infrared spectroscopy and computed tomography that the vapour emitted by sessile drops at room temperature behaves differently as compared to the commonly accepted diffusion limited model. In this present article, we propose to somehow extend this study of vapour diffusion for droplets for a larger range of temperature differences and use microgravity environment to study pure diffusion vapour diffusion.

Models taking into account convection are developed in the combustion community to predict the evaporation time in order to obtain the best reagents concentration for combustion [19–21]. The most common, the Spalding evaporation model, takes into account convection with the calculation of mass and heat balance separately in each phase at the interface [22]. This model is based on numerous assumptions (isothermal spherical droplet, quasi-steady gas boundary, vapour/liquid phase equilibrium at the interface, air and vapour behave like a perfect gas, use of Fick's law of diffusion) that are similar to the purely diffusive model used in this study. This model, based on the Spalding mass number B_m , slightly over-predicts the evaporation rate of droplets evaporating under reduced gravity conditions *i.e.* for purely diffusive evaporation [14]. For this reason, the purely diffusive model has been chosen as a basis for our empirical model on.

New models that are able to correctly predict diffusive and convective evaporation are emerging [14,23] but their use is limited to peculiar fluids. Therefore, the aim of this article is to determine the limits of purely diffusive models and to develop, based on numerous experiments, an empirical model that accurately describes the evaporation rate of a sessile droplet on a heated substrate, regardless of the temperature or the type of fluid. The atmospheric contribution to a sessile droplet evaporation will also be investigated numerically.

2. Experiments

2.1. Fluids properties

For the purpose of this experimental study, two types of fluids (both linear carbon chains) have been chosen to be evaporated onto heated substrates: alcohols; from CH₃ OH (methanol) to C₇ H₁₅ OH (heptanol), and alkanes; from C₅ H₁₂ (pentane) to C₁₀ H₂₂ (decane). The fluids, obtained from Sigma Aldrich, are anhydrous with a purity of 99% or higher and contain only one compound except for hexane which is a mixture of isomers. The fluids have been used without any treatments prior to the experiments. All fluids properties can be found in Table 1. Properties are given at medium temperature $T_m = 40$ °C (unless otherwise indicated). The temperature dependences of properties have been taken into account in all calculations. Moreover, through this article, properties have been either measured in our laboratory or are extracted from literature sources.

2.2. Experimental set-up

The experimental set-up (Fig. 1), based on our previous work [28], enables the creation of droplets on a heated substrate (a range of temperatures between 20 °C, room temperature and 55 °C is investigated) and the monitoring of their evolution with a side-view CCD Camera which follows the evolution of the geometric parameters (radius, contact angle and height of the drop).

As this experimental set-up can be run either on Earth (1 g) or under reduced gravity conditions (microgravity 0.01 g, Moon gravity 0.16 g and Mars gravity 0.38 g), several modifications are brought to the experimental set up to obtain the most precise measurements in each conditions and are explicated in this section. The reduced gravity experiments take place aboard the Novespace's A300-ZeroG aircraft in Bordeaux (France). This aircraft is dedicated to parabolic flights and creates 22 s of reduced gravity framed by two stages of hypergravity (1.8 g).

In the laboratory, the droplets are created with a micro-dispenser controlled by computer above the substrate without the cell lid and are slowly deposited onto the substrate. As this method is technically very complex during reduced gravity experiment, the fluids are injected through a thin 0.7 mm pipe at the centre of the substrate with a motorized syringe-pump into an enclosed cell.

The droplets are then allowed to evaporate in an atmosphere composed of air at a temperature (T_a) of 20 ± 1 °C and a pressure (p_a) of 1 bar for laboratory experiments (the cell is open during all the droplet evaporation to insert the microdispenser) and 835 ± 2 mbar for microgravity experiments (cabin pressure during flight) in a closed configuration. The experimental conditions of the cell are recorded using a pressure sensor and a thermocouple of type K.

The substrate temperature and the heat-flux absorbed by the droplet are measured with a type-K thermocouple and with a heat flux-metre, respectively. The last one contains a large number of thermocouples connected in series and has a diameter of 15 mm and a thickness of 0.6 mm. Its copper body provides its high thermal conductivity and, consequently, its negligible thermal resistance to the heat flux from the heater (disk-shaped polyimide thermofoil heater regulated by a PT-100 sensor with a PID regulator at 0.1 °C) to the droplet.

The heat-flux metre, located between the heater and the substrate, is only used for reduced gravity experiments as they are performed into a closed chamber and the evaporation results in a slight increase of the chamber pressure, which pushes liquid back into droplet injection pipe. There is also a possibility of trapped

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