



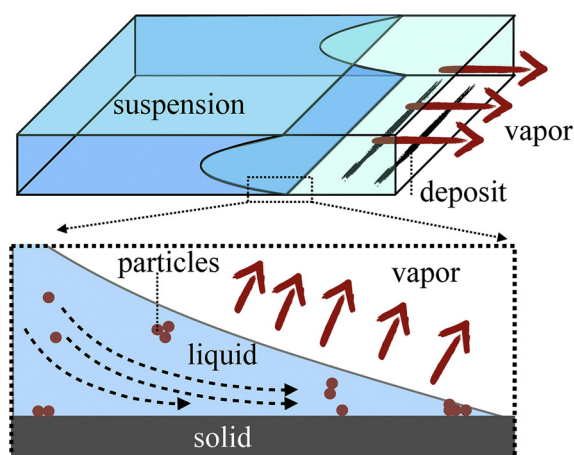
A theoretical analysis of the deposition of colloidal particles from a volatile liquid meniscus in a rectangular chamber

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GRAPHICAL ABSTRACT



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ABSTRACT

Traditionally, experiments of pattern deposition are conducted using volatile suspensions or solutions in sessile drops. However, several recent studies on the pattern deposition of colloidal particles, bio-molecular films, and membranes employed volatile carrier liquids in micro-chambers or micro-channels with a rectangular perimeter. We show that the local rate of evaporation from a concave meniscus of liquid in a chamber increases in the vicinity of the contact line like $X^{-1/2}$ for a vanishingly small three phase contact angle, where the contact line is positioned at $X = 0$. This result is a reminiscent of the local rate of evaporation of liquid from a convex sessile drop. Moreover, an increase in the magnitude of the contact angle reduces the singularity in the rate of evaporation near the contact line. We employ our findings to study the deposition of colloidal particles from a volatile liquid meniscus in the presence of particle coagulation and particle adsorption to the substrate of the chamber, while demonstrating the convective accumulation of particles near the pinned contact line.

1. Introduction

The evaporation of a volatile liquid containing colloidal dispersions (e.g., polymers, proteins, viruses, bacteria, DNA, microspheres,

nanoparticles) is a widely used mechanism to deposit particles or molecules onto a substrate. In particular, the drying process is a well-known simple technique to yield self-assembled 1D or 2D structures with controlled spatial dimensions [1]. The spatial dimensions may

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reduce down to widths of microns and thicknesses of nanometers. Hence, the evaporation of suspensions in confined geometries – the most simple being a sessile drop – has been attracting a great deal of attention for several decades [2–10].

The deposition of colloidal particles from a freely evaporating body of a volatile liquid on a solid substrate is often stochastic and lacks regularity. The judicious use of solid confinement, however, imparts exquisite control over the rate of evaporation and flow in the liquid, which in turn, allows for crafting complex deposit patterns with unprecedented regularity. Examples are capillary tubes [11], “curve-on-flat” geometry [12], wedge-on substrate geometry [13], two parallel circular plates [14,15], and two crossed-cylinders placed at right angle, one relative to the other [16]. Furthermore, the confinement of the volatile liquid within a rectangular micro-chamber supports a well defined and linear three phase contact line, which has been employed for the pattern deposition of colloidal particles, bio-molecules, and membranes [17–19]. The linear geometry of the contact line allows for a uniform and unidirectional forcing of the volatile liquid meniscus, constraining the state of deposition of the suspended particles. Examples include constraining the meniscus by displacing the lower or upper side of the channel and thus imposing shear in the liquid [20,21], and by generating surface acoustic waves (SAWs) in the substrate, thus altering the geometry of the liquid meniscus by induced mass transport [22]. A variation in the channel geometry is obtained by positioning two non-parallel plates; the upper plate is stationary and placed at a fixed angle relative to the lower moving plate [23].

Controlled deposition of particles from a volatile carrier liquid requires insight into the local rate of evaporation of the liquid from the meniscus. The evaporation generates flow in the liquid, which convects the suspended particles toward the three phase contact line. Theoretical studies on the evaporation and flow in a meniscus of liquid usually rely on the three main geometries of the free surface of the liquid near the contact line, which are discussed in detail below.

One geometry is a uniform thin micro-scale film of liquid which is governed by both capillary and viscous stresses. The treatment of the evaporation and flow problems in this geometry is often done by asymptotic matching of the micro-scale part of the meniscus near the contact line and the macro-scale part of the meniscus. More specifically, by matching the contact line region to the macro-scale part of the meniscus, Morris [24] determined the heat flow of the wetting liquid in a channel. On a different note, under the assumption of a small contact angle, by using an asymptotic matching, Rednikov and Collinet [25], extracted the evaporation flux from a steady meniscus. Park et al. [26] solved numerically the heat and vapor flow problems in the vicinity of the contact line of a liquid in a micro-chamber, where the small thickness (and slope) of the meniscus allowed them to apply the lubrication approximation. Moreover, Doumenc and Guerrier [27] solved a model problem comprising the shape of a volatile meniscus and the corresponding vapor flux from a liquid body a top of moving substrate.

The second geometry is associated with a sessile drop, for which a vast amount of literature exists. Rowan et al. [3] proposed a vapor-phase diffusion model to estimate the evaporation rate for a spherical-cap drop atop a substrate. They derived an analytical solution for the rate of volume loss due to the evaporation under the assumption of a small contact angle. Later on, with the aid of the theory derived by Lebedev [2], Deegan et al. [1,5] and Popov [7] presented an analytical solution for vapor-phase diffusion model for a spherical drop of a general contact angle. Their closed form solution yielded an integrable singularity for the vapor flux near the contact line. By experimentally investigating the evaporation of a sessile droplet with a pinned contact line and by applying the finite element method (FEM) to compute the diffusive distribution of the vapor concentration, Hu and Larson [6] proposed a correction to Deegan's formula for the rate of evaporation. On a different note, by using Deegan's solution, Poulard et al. [28], who proposed a model which generalizes Tanner's law, suggested a power law dependence for the radius and for the contact angle of the

evaporating droplet on time. Another generalization to Deegan's model was proposed by Dunn et al. [29], which includes the effect of evaporative cooling on the saturation concentration of vapor at the free surface of the drop, and the dependence of the diffusion coefficient of the vapor on the atmospheric pressure. A completely different approach was proposed by Gelderblom et al. [30]. They studied the singular behavior of the evaporative flux in the vicinity of the contact line of a sessile droplet by analytically solving the Stokes equation for viscous flow in a wedge geometry of arbitrary contact angle.

The third geometry is the so-called “strip-like droplet” (planar case). It was studied both theoretically and experimentally by Yarin et al. [8], where the solvent evaporation rate was calculated over the whole free surface of the liquid. Yarin and coworkers showed that the vapor flux has an integrable power law singularity in the proximity of the contact line, where the power has the similar dependence on the contact angle as the one obtained earlier by Deegan et al. [1,5]. Later on, Du and Deegan [31] used Yarin's formula for the evaporative flux near the contact line in order to study pattern deposition from a volatile drop atop an inclined substrate.

The second aspect of our paper is the deposition of colloidal particles from a volatile carrier liquid. The morphology of the deposit of evaporating sessile drops was widely studied in the past decade; here we bring a short count of the main previous works in the field. Briefly, Bhardwaj et al. [32], showed that by altering the pH levels of colloidal suspensions in drops they managed to alter the morphology of the particulate deposit following the full evaporation of the volatile carrier liquid. Different pH levels rendered different surface forces of molecular origin that acted on the suspended particles, promoting or arresting particle coagulation and the adsorption of the particles to the substrate. In a different experimental study, Anyfantakis et al. [33] used charged surfactants to alter the zeta potential of the suspended particles for the same purpose. A theoretical approach was suggested e.g., by Crivoi and Duan [34,35] who, by employing a diffusion limited cluster–cluster aggregation approach, investigated the connection between the growth of particle aggregates in a volatile liquid and the morphology of the deposit. On a different note, Kaplan and Mahadevan [36], proposed a multiphase model to describe the evaporation of a drop of a colloidal suspension. They predicted the formation of deposits of the shape of a single ring, multiple rings, broad bands, and uniform layers as a function of two parameters: the initial solute concentration and the scaled inverse capillary number. In addition, Zigelman and Manor [37] proposed a theoretical model for the deposition of colloidal particles from a volatile drop, where the contributions of surface forces were directly connected to the morphology of the deposit through classic models of particle coagulation and adsorption. They further gave a comprehensive review on corresponding experimental work.

In this paper we use theory in order to study the local rate of evaporation of liquid from a volatile meniscus in a rectangular chamber and the resulting dynamics of the deposition of colloidal particles that are originally suspended in the liquid. In our analysis we account for particle coagulation and particle adsorption to the solid substrate. The chamber is assumed to possess a thickness that is small relative to its width and length, which is a common geometry in microfluidic channels and chambers. We explain the geometry of our problem and the corresponding mathematical model for the evaporation and flow problems in Section 2. We discuss the shape of the meniscus in Section 2.1, solve the evaporation problem in Section 2.2, discuss the method for calculating the quasi-steady advection velocity for a constant contact angle (for the case of slipping contact line) in Section 2.3, discuss the corresponding case of a dynamic contact angle (pinned contact line) in Section 2.4, and estimate the overall rate of evaporation, the singularity of the evaporative flux near the contact line, and the corresponding velocity of the liquid in Section 2.5. We then discuss the deposition problem in Section 3. We present the model equations in Section 3.1 and discuss analytical solutions for the problem of particle transport in Section 3.2.1, for the problem of particle transport and adsorption in

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