

Dip- and die-coating of hydrophilic squares on flat, hydrophobic substrates



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ABSTRACT

We studied the *selective* deposition of a Newtonian liquid on the hydrophilic domains of chemically patterned substrates. We present experiments and numerical simulations of the dip-coating and self-metered die-coating of compact, geometrically convex patterns such as squares and diamonds. The coating of such patterns is intrinsically an instationary process. Nevertheless, the maximum film thickness entrained on diamonds that are either much smaller or larger than the lengthscale governing the coating process follows the theoretical predictions derived for steady-state coating of either narrow hydrophilic stripes or chemically homogeneous surfaces. The transition between these two regimes is determined by the ratio of the pattern dimensions and the characteristic length of the reservoir meniscus, i.e. the capillary length for dip-coating and the die-gap for die-coating processes. While the film thickness entrained on diamonds scales with the coating speed as a powerlaw, the entrained thickness on squares with sides oriented parallel and perpendicular to the coating direction appears to saturate for small coating speeds. This influence of the azimuthal pattern orientation is related to the capillary break-up of the coating meniscus. Residual satellite droplets observed underneath the hydrophilic patterns after coating are a consequence either of the break-up or the subsequent retraction of the coating meniscus.

1. Introduction

Solution processing of organic electronic devices holds great promise as an inexpensive and large-area compatible manufacturing technique. Roll-to-roll coating is an attractive fabrication process, because it is continuous and its throughput can be increased by extending the substrate width. Moreover, the waste of functional material is minimal compared to, for example, spin-coating. For the purpose of delineating individual devices, chemical patterns can be defined on the substrate. These typically consist of isolated hydrophilic regions, where the deposition of functional material is desired, on an otherwise hydrophobic substrate (Biebuyck and Whitesides, 1994; Qin et al., 1999; Braun and Meyer, 1999; Bechinger et al., 2000; Darhuber et al., 2000; Hartmann et al., 2001; Oh et al., 2003; Ko et al., 2004; Fan and Stebe, 2005; Darhuber and Troian, 2005; Choi and Park, 2006; Benor et al., 2007; Makaram et al., 2007; Woodson and Liu, 2007; Chowdhury et al., 2007; Zeira et al., 2008; Bardecker et al., 2008; Lin et al., 2008; Li et al., 2008; Zeira et al., 2009; Lee et al., 2009; Lin et al., 2009; Park et al., 2009; Mastrangeli et al., 2009; Meyer et al., 2009; Na et al., 2009; Cai, 2009; Lee et al., 2010; Miller et al., 2010; Reynolds et al., 2010; Svarnas et al., 2010; Zhu et al., 2010; Furuta et al., 2011; Kim et al., 2011; Mastrangeli et al., 2011; Brasjen et al., 2011; Ke and

Tang, 2013; Nakajima et al., 2013; Wang and McCarthy, 2014; Wang et al., 2014; Watanabe et al., 2014; Kim et al., 2014; Hou et al., 2014; Wu et al., 2015; Hou et al., 2015; Kobaku et al., 2015; Chang et al., 2016; Bao et al., 2016).

Two complementary coating strategies are (1) applying the liquid directly and exclusively to the hydrophilic regions (Darhuber et al., 2000; Brasjen et al., 2011) and (2) coating the entire substrate with a uniform liquid film and subsequently relying on redistribution of the solution from the hydrophobic to the hydrophilic regions (Bower et al., 2007; Kargupta and Sharma, 2003; Brasjen and Darhuber, 2011). The transition from process (1) to (2) occurs at a critical coating speed U_{crit} , which depends primarily on the contact angle of the hydrophobic part of the substrate (Blake and Ruschak, 1979; Petrov and Sedev, 1985; Petrov, 1986; Cox, 1986; Chesters and van der Zanden, 1993; Petrov and Petrov, 1995; Shikhmurzaev, 1996; Golestanian and Raphaël, 2003; Eggers, 2004; Blake, 2006; Snoeijer et al., 2007; Maleki et al., 2007).

Landau and Levich derived a relation for the film thickness h_{∞} entrained during dip-coating of chemically homogeneous and completely wetting substrates in the limit of small capillary numbers (Landau and Levich, 1942; Derjaguin, 1943)

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Nomenclature

α	Wall inclination angle, deg.
β	Exponent
b	Pattern base length, m
Ca	Capillary number
d_g	Die-gap, m
g	Gravitational acceleration, m/s ²
γ	Surface tension, N/m
h	Local film thickness, m
h_∞	Film thickness, m
h_{\max}	Maximum film thickness, m
h^*	Disjoining pressure length scale, m
h_{res}	Reservoir film thickness, m
K_D	Dip-coating constant
K_R	Die-coating constant
ℓ_c	Capillary length, m
L_x, L_y	Computational domain dimensions, m

m	Disjoining pressure exponent
μ	Dynamic viscosity, Pa s
n	Disjoining pressure exponent
p	Augmented pressure, Pa
p_{amb}	Ambient pressure, Pa
p_{res}	Reservoir pressure, Pa
φ	Azimuthal pattern orientation, deg.
Π_{SE}	Disjoining pressure, Pa
Q_y	Vertical liquid flux, m ² /s
ρ	Mass density, kg/m ³
t	Time, s
θ	Contact angle, deg.
θ_i	Hydrophilic contact angle, deg.
θ_o	Hydrophobic contact angle, deg.
U	Coating speed, m/s
U_{crit}	Critical coating speed, m/s
w	Line width, m
x, y	Coordinates, m

$$h_\infty = 0.946 \ell_c Ca^{2/3}. \quad (1)$$

Here, $\ell_c \equiv \sqrt{\gamma/(\rho g)}$ is the so-called capillary length, which determines the characteristic radius of curvature of the reservoir meniscus. The parameters γ and ρ represent the surface tension and density of the liquid, respectively, and g is the gravitational acceleration. The capillary number $Ca \equiv \mu U/\gamma$ is a non-dimensionalized coating speed U , with μ denoting the dynamic viscosity.

In self-metered die-coating processes, the reservoir meniscus is confined between the moving substrate and a stationary solid object, the so-called coating die. They are separated by a distance d_g , which is commonly referred to as the die-gap. As a consequence, the radius of curvature of the meniscus becomes proportional to d_g and the entrained thickness is given by

$$h_{\max} = K_R d_g Ca^{2/3}, \quad (2)$$

where K_R is a constant depending on operating conditions and the die geometry (Ruschak, 1976).

Hydrodynamical aspects of the dip-coating of chemically patterned substrates were first studied by Darhuber et al. (2000). For the case of long, narrow hydrophilic lines of width $w \ll \ell_c$, the confinement of the liquid to the hydrophilic stripe significantly alters the entrainment dynamics, leading to (Darhuber et al., 2000)

$$h_{\max} = K_D w Ca^{1/3}. \quad (3)$$

Using an asymptotic matching technique, Davis (2005) determined the numerical value of $K_D = 0.356$. Brasjen et al. (2011) performed systematic experiments regarding the dip-coating of hydrophilic lines and tip-down oriented triangles with varying base length b . Unexpectedly, Eqs. (1) and (3) were found to provide accurate predictions of the thickness of films entrained on tip-down triangles, despite the fact that Eqs. (1) and (3) were derived for steady-state coating processes, whereas the coating of chemical patterns of finite dimensions necessarily is an *instationary* process.

In this manuscript, we consider the selective deposition of liquids on isolated, geometrically convex, hydrophilic regions on otherwise hydrophobic substrates. We systematically measured the thickness of the film entrained during dip-coating and die-coating of hydrophilic squares as a function of the baselength, coating speed and azimuthal orientation. The dip-coating experiments were complemented with numerical simulations, which reproduced the scaling behavior observed in the experiments. Moreover, they elucidated the decisive role of the break-up of the coating meniscus in determining the liquid volume entrained on the squares. The occurrence of residual satellite droplets is caused by this break-up process as well as the subsequent meniscus retraction.

2. Materials and methods

Samples were manufactured from $5 \times 5 \text{ cm}^2$ and $7.5 \times 5 \text{ cm}^2$ large D263 glass slides (Schott) using the photolithography procedure described by Brasjen et al. (2011). Substrates fabricated by this procedure consist of a hydrophobic self-assembled monomolecular layer of 1H,1H,2H,2H-perfluorooctyltrichlorosilane (PFOTS, Aldrich,

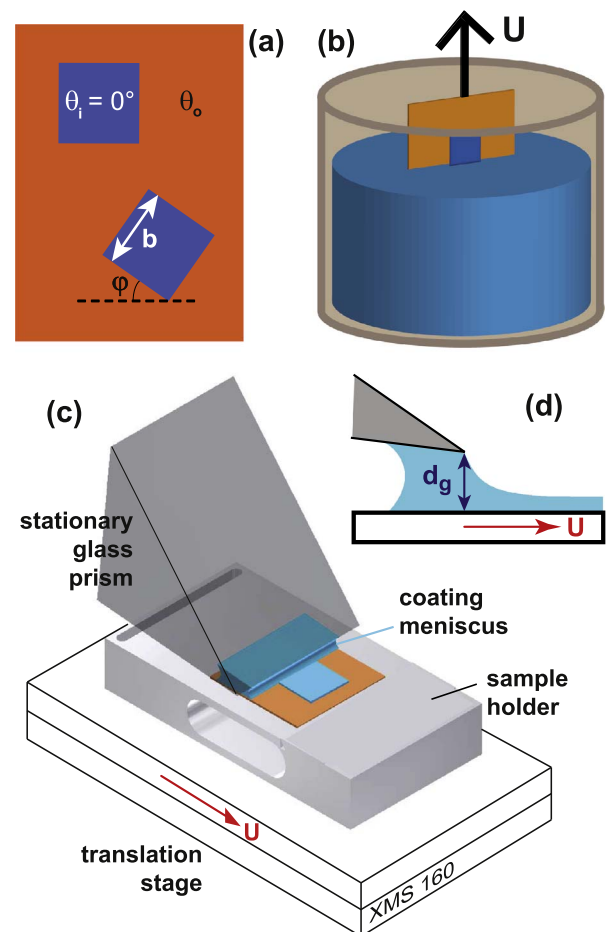


Fig. 1. (a) Schematic of a chemically patterned substrate with hydrophilic squares of different azimuthal orientation φ . (b) Sketch of the dip-coating process and (c) and (d) the self-metered die-coating setup. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

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