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# Electrohydrodynamic effects in the leveling of coatings

Aruna Ramkrishnan, Satish Kumar*

*Department of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, MN 55455, USA*

## HIGHLIGHTS

- Effect of electric charge on leveling of thin-film coatings is modeled.
- Both homogeneous and heterogeneous charge distributions are considered.
- Simple heuristics are proposed for determining when leveling occurs.

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## ABSTRACT

Electrostatic charges that accumulate on substrates and at liquid–air interfaces in various coating processes can drive liquid flows that lead to defects. To better understand this phenomenon, we model the leveling of thin liquid films subject to electrohydrodynamic forces. We consider cases of homogeneous and heterogeneous substrate charge distributions and contamination of the film surface by free charge. The liquid is assumed to be Newtonian, both perfect dielectric and leaky dielectric materials are considered, and lubrication theory is employed. Linear stability analysis and nonlinear simulations reveal different leveling criteria for small- and large-amplitude perturbations to the film surface. Heterogeneous charge distributions on the substrate are found to lead to steady curved interface shapes. Using asymptotic methods, we develop analytical expressions to predict these shapes, and consequently, the magnitude of coating defects. We also employ transient nonlinear simulations track the leveling of disturbances created by contamination of the film surface by free charge. The results of our study enable us to propose simple heuristics for determining the conditions under which coatings subject to electrohydrodynamic forces will level.



## 1. Introduction

Coating is the process of laying out a uniform thin liquid film onto a substrate. In many coating applications, generating a smooth defect-free surface is crucial for improved product performance. Properties like high gloss, mechanical stability, and electrochemical performance are dependent on coating uniformity (Desjumaux et al., 1998; Glatter and Bousfield, 1997; Orchard, 1963; Phair et al., 2009; Schwartz et al., 1996; Xiang and Bousfield, 2000). To get a smooth coating, interfacial defects should level before the liquid coating dries (Iyer and Bousfield, 1996; Tsai et al., 2010). Detailed studies of leveling of disturbances in coatings have been carried out and these take into account the effects of surface tension, gravity, viscosity, elasticity, drying, non-Newtonian rheology, Marangoni flows, and other phenomena (Bousfield, 1991; Iyer and Bousfield, 1996; Keunings and Bousfield, 1987; Kheshgi and Scriven, 1988; Orchard, 1963; Tsai et al., 2010; Wulf et al., 2000).

A significant source of defects in liquid coatings is electrostatic charge, which can build up on dielectric webs during web handling processes involving friction between the web and rollers, separation of surfaces, stretching of webs, and modification of surface properties by exposure to ionized gas. Electrostatic potential differences on the order of tens to hundreds of volts can be created by static charge on non-conductive webs which may take days or weeks to dissipate (Gutoff and Cohen, 1995). Static charges can be damaging to liquid coatings because they pose a spark ignition hazard, attract dust particles, and create defects or rupture coatings through electrohydrodynamic interaction with the liquid (Jendrejack et al., 2010, 2011a,b). A second source of defects is sparking in coating processes, which may cause free charges to jump onto the liquid surface and travel in the coating, creating localized disturbances which may or may not level.

It would be valuable to have models that predict the leveling behavior of liquid coatings in the presence of electrostatic charges in order to provide guidelines to design robust coatings. However, to the best of our knowledge, such models have not been developed. Most related studies focus on how electric fields can be used to create surface patterns in thin liquid films.

* Corresponding author. Tel.: +1 612 625 2558; fax: +1 612 626 7246.
*E-mail address:* kumar030@umn.edu (S. Kumar).

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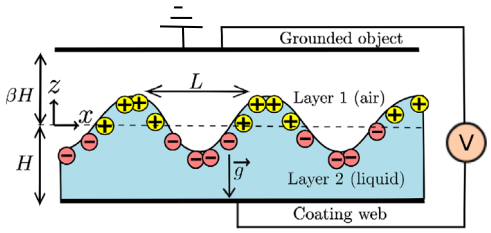


**Fig. 1.** Schematic of problem geometry. The interface is initially located at the dashed line.

The mechanism of electrostatically driven instability was first proposed by Herminghaus (1999) in order to accurately explain experimental observations by David et al. (1998), who reported dynamical instability with strong mode selection in dielectric liquid films confined between conductive surfaces. Seminal experiments performed by Chou and Zhuang (1999) and Zhuang et al. (1999) showed that thermally induced charges can create an interfacial instability in thin polymer films. Later, external electric fields were also shown to produce a similar effect (Schäffer et al., 2000, 2001). A host of experiments and theoretical modeling followed which examined the ways in which mask patterning, liquid properties, multilayer films, AC fields and other parameters can be manipulated to control the final microstructure (Arun et al., 2009; Atta et al., 2011; Bandyopadhyay and Sharma, 2007; Bandyopadhyay et al., 2009; Craster and Matar, 2005, 2009; Dickey et al., 2006; Gambhire and Thaokar, 2010, 2011; Leach et al., 2005; Mondal et al., 2013; Morariu et al., 2003; Ozen et al., 2006; Pattader et al., 2011; Pease and Russel, 2002, 2003, 2004, 2006; Reddy et al., 2010; Roberts and Kumar, 2009, 2010; Shankar and Sharma, 2004; Srivastava et al., 2010; Tian et al., 2011; Wu and Russel, 2009; Xi et al., 2012; Zhao et al., 2011).

However, although patterning applications require the growth of deformations in thin films, precision coating applications demand the opposite: disturbances in coatings should level out quickly to prevent the formation and locking in of defects. The goal of this work is to develop models that enable us to understand how bound and free electrostatic charges interact with liquid coatings and to predict, through a detailed study, conditions under which coatings will level even in the presence of charges at the substrate or interface.

In Section 2 of this paper, we describe our mathematical model and governing equations for the case of a general heterogeneous charge distribution at the substrate. In Section 3, we consider a homogeneous substrate charge distribution and use linear stability analysis and nonlinear simulations to study leveling of small- and large-amplitude disturbances. In Section 4, we examine the effect of substrate charge heterogeneity on film stability and use asymptotic expansions to model steady interface profiles. Finally, in Section 5, we employ transient nonlinear simulations and a scaling analysis to track the evolution of disturbances created by surface charge contaminants and follow this with conclusions in Section 6.

## 2. Mathematical model and governing equations

Our presentation closely follows earlier works on thin-film electrohydrodynamic instabilities (Roberts and Kumar, 2009; Shankar and Sharma, 2004; Craster and Matar, 2005). We employ a two-dimensional model consisting of an incompressible Newtonian liquid layer confined between two electrodes, as shown in Fig. 1. The top electrode is grounded and separated from the liquid by an air gap, and the bottom electrode has a fixed potential $V(x)$ which could be uniform or spatially varying. The ratio of the air gap to the liquid thickness $H$ is characterized by the dimensionless parameter $\beta$. In practice, the top electrode corresponds to any material close enough to the liquid to act as a ground and create a non-zero electric field in the liquid and the air gap. The $x$–$z$ coordinate axes rest on the initially flat interface between phase 1 (air) and phase 2 (liquid). In general, the position of the interface is given by $h(x,t)$. Flow in the air phase is neglected. For the liquid phase, the viscosity $\mu$ and density $\rho$ are constants. The two phases have dielectric constants $\epsilon_i$, the air phase is assumed to be non-conducting, and in the general case, the liquid has a finite conductivity $\sigma$. The interfacial tension $\gamma$ and the gravitational acceleration $g$ drive flow that tries to level out any interfacial perturbations that grow due to the destabilizing electrostatic forces.

In our analysis, we have described the electrical response of liquids using the perfect and leaky dielectric models. Leaky dielectrics have a finite, low conductivity which allows for accumulation of free charge at the liquid–air interface, represented by $q(x,t)$ (Saville, 1997; Melcher and Taylor, 1969). In the limit of zero conductivity and zero free charge, the leaky dielectric reduces to a perfect dielectric, which is a material incapable of conduction and only polarizes when acted upon by an electric field, creating bound charge at the interface. A comparison of the two models enables us to understand the role of free charge in the leveling process.

### 2.1. Governing equations

The electrostatics for both phases ($i=1,2$) are modeled by the Laplace equation for electrostatic potential $\psi_i$

$$\nabla^2\psi_i = 0, \tag{1}$$

which requires two boundary conditions on potential in each phase. Since the top substrate is grounded and the bottom substrate has a fixed potential

$$\psi_1 = 0 \quad \text{at } z=\beta H, \tag{2}$$

$$\psi_2 = V(x) \quad \text{at } z=-H. \tag{3}$$

The two other boundary conditions are provided by the continuity condition for the potential at the interface

$$\psi_1 = \psi_2 \quad \text{at } z=h(x,t), \tag{4}$$

and the interfacial jump condition for the electric field

$$\|\epsilon_i\epsilon_0\nabla\psi_i\|_i \cdot \boldsymbol{n} = q(x,t) \quad \text{at } z=h(x,t), \tag{5}$$

where $\|\ldots\|_i$ is the jump operator $\|\ldots\|_2-\|\ldots\|_1$ and $\epsilon_0$ is the permittivity of free space. We denote $\boldsymbol{n}$ as the normal vector pointing outward from the interface into the air layer.

For the hydrodynamics, the creeping-flow equations apply in the liquid

$$\nabla\cdot\boldsymbol{v} = 0, \tag{6}$$

$$\nabla\cdot\boldsymbol{T} = 0. \tag{7}$$

where $\boldsymbol{v}$ is the velocity vector and $\boldsymbol{T}=\mu[\nabla\boldsymbol{v}+(\nabla\boldsymbol{v})^T]-p\boldsymbol{\delta}+\boldsymbol{M}$ is the total stress tensor. Here, $p$ is the pressure, $\boldsymbol{\delta}$ is the identity tensor, and $\boldsymbol{M}=\epsilon\epsilon_0[\boldsymbol{E}_i\boldsymbol{E}_i-\frac{1}{2}(\boldsymbol{E}_i\boldsymbol{E}_i)]\boldsymbol{\delta}$ is the Maxwell stress tensor. The electric field $\boldsymbol{E}_i$ can be calculated from the electrostatic potential using the relation $\boldsymbol{E}_i=-\nabla\psi_i$. In the bulk of the liquid, the $\nabla\cdot\boldsymbol{M}$ contribution to the creeping-flow equations uniformly goes to zero (Saville, 1997), simplifying (7) to

$$-\nabla p+\mu\nabla^2\boldsymbol{v} = 0. \tag{8}$$

The boundary conditions employed for the fluid velocities are the no-slip and no-penetration conditions at the substrate

$$\boldsymbol{v} = 0 \quad \text{at } z=-H. \tag{9}$$