3D Simulation of Electrically-induced Nano-Patterning on Polymer Films

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***Abstract*—A computational electrohydrodynamic (EHD) model is presented for the modeling of 3D electrically-induced fluid motion and free surface morphology evolution during EHD patterning of nanostructures. The model entails a finite difference solution of the electric field equation with a leaky model to account for polymer behavior, the Navier-Stokes equations for electrically-driven flows and the phase field equation for the free surface deformation. These equations are fully coupled and represent a very large complex numerical system. Once discretized, the intensive computation is alleviated with the use of parallel computing algorithms. Computed results are presented that illustrate the transient development of 3D nanostructures as they form under an electric field. The model is a useful tool to explore optimal conditions for scalable manufacturing of large scale nanostructures using the EHD patterning processes.**

***Index Terms*— Nanopatterning, Electrostatic self-assembly, Nanofluidics, Numerical simulation, Polymer films.**

I. Introduction

Patterned micro/nano structures have a wide range of applications in micro/nano optical and electronic devices [1,2] and other microelectromechanical systems [3]. Some of these structures may be made using the manufacturing processes assisted by the external fields such as the electric field [4] and the magnetic field [5]. One of such techniques that has attracted much attention is the electrohydrodynamic (EHD) patterning process, a schematic representation of which is illustrated in Fig. 1a. The process uses a pair of electrodes, one of which (often the upper one) is patterned. A liquid polymer/air (polymer/polymer) film is sandwiched in between the electrodes and structures are patterned in the film in accordance with the structured electrode, when an external electric field is applied. The final nanostructures are formed with thermally- or photo-induced curing of the patterned polymer. Fig. 1 (b) shows an example of the top electrode (i.e. template), patterned with periodic cubic protrusions. Templates with other patterned electrodes have also been used [4]. An advantage of this EHD process is perhaps the cost effectiveness and its potential for scalable nanomanufacturing of large scale nanostructures. Of crucial importance to the process development and nanostructure quality control are the electrically-induced liquid film morphology evolution and the associated fluid motion in the liquid layer.

This paper presents a full 3D transient numerical model capable of describing the electrically-induced flow and free surface phenomena in the EHD process. The numerical model is developed based on the electrostatic approximation with a leaky dielectric behavior is prescribed for the liquid-air system, which is then coupled with the Navier-Stokes equations for the fluid motion and with a phase field description of free surface deformation. The phase field modeling of free surface evolution is a result of a coarse-grain average of atomic statistical behavior of the liquid-air interface and provides an almost ideal approach to describe the nanoscaled evolution of structures associated the EHD nano-patterning processes. Numerically, this physics-based phase field model of free surfaces belongs to the category of fixed grid method for moving boundary problems. The coupled electrohydrodynamic and phase field equations are solved using the finite difference technique enhanced with parallel computing algorithms. Mathematical formulations, numerical aspects and computed results obtained for the EHD nanopatterning process are given.

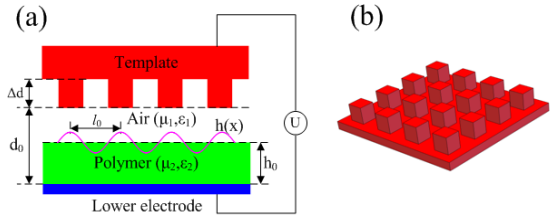

Fig. 1. (a) Configuration for the EHD patterning and (b) the morphology of the template.

II. Mathematical Formulation

Referring to Fig. 1, the mathematical equations that govern the electric field and force distribution, the fluid motion and the free surface evolution are given below.

A. Phase field equations

In phase field the free energy density is assumed to be a function of phase parameter C, the form of energy f : [0, 1] → **R** is taken as

$$f(C)=\frac{1}{2}\xi\gamma\alpha|\nabla C|^2+\xi^{-1}\gamma\alpha\bullet\frac{1}{4}C^2(1-C)^2, \quad \in\Omega_T := \Omega\times(0,T) \qquad (1)$$

where Ω is a bounded domain in $\mathbf{R}^3$, with a boundary ∂Ω, T the computing time section, and C is the phase parameter with the property that values of C=1 and C=0 correspond to two distinctive phases. γ is the coefficient of the surface tension, ξ a measure of interface thickness. $\alpha=6\sqrt{2}$ is a constant.

The Cahn-Hilliard equation with convection is employed to describe the evolution of the phase field

$$\frac{\partial C}{\partial t}+\vec{u}\bullet\nabla C-\nabla\bullet(M\nabla\phi)=0, \qquad \in\Omega_T \qquad (2)$$

where $\vec{u}$ represents the fluid velocity, M is the phase field mobility and is considered as a constant in this study, Φ is the chemical potential which is defined $\phi=\delta f/\delta C$.

B. Electric Field Equations

For poorly conducting polymers, a leaky dielectric model gives a better description of electric behavior. By the leaky model, free and bound charges both exist in the system, and the conservation of charges can be expressed as,

$$\frac{\partial \rho^e}{\partial t} + \vec{u} \bullet \nabla \rho^e = -\nabla \bullet \left(\sigma(C) \vec{E} \right) \qquad \in \Omega_T \qquad (3)$$

where $\sigma(C)$ denotes the electrical conductivity, $\vec{E} = -\nabla V$ the electric strength, V the electric potential, and ρ^e the free charge density. This equation can be further simplified for case under consideration. As the time scale of charge relaxation $t_\sigma = \varepsilon_0 \varepsilon_r / \sigma$ is much less than that of the flow $t_c = L_c/U_c$ (L_c, the length scale, U_c the characteristic velocity), Eq. (3) becomes

$$\nabla \bullet \left(\sigma(C) \nabla V \right) = 0 \qquad \in \Omega_T \qquad (4)$$

With Eq. (4), the free charge is calculated by Poisson equation, namely

$$\rho^e = -\nabla \bullet \left(\varepsilon_0 \varepsilon_r (C) \nabla V \right) \qquad \in \Omega_T \qquad (5)$$

where ε_0 is the permittivity of vacuum, and $\varepsilon_r(C)$ is the dielectric constant.

C. Fluid Flow Equations

The flow field distribution is described by the governing equations of mass conservation and momentum equations,

$$\nabla \bullet \vec{u} = 0 \qquad \in \Omega_T \qquad (6)$$

$$\nabla \bullet \Pi + \vec{f_e} + \vec{f_\gamma} = 0 \qquad \in \Omega_T \qquad (7)$$

In the EHD system, the electric force and the surface tension force on the interface should be added. The modified Navier-Stokes equation for variable density and viscosity is employed here as the governing equation. In micro/nano scale, the Reynolds number is very low (Re<<1), the flow becomes creeping flow, the unsteady term and convective term can be ignored. In the above equations, $\rho(C)$ is the density of fluid, p the pressure, μ the viscosity, $\delta_{i,j}$ identity matrix, Φ the chemical potential. Also, Π is the stress tensor, and the electric force due to polarization charge and free charge, and surface tension force is defined below,

$$\vec{f_\gamma} = \phi \nabla c, \quad \Pi = -p\delta_{i,j} + 2\mu\varepsilon_{i,j} = -p\delta_{i,j} + \mu(C)\left(\nabla \vec{u} + \nabla \vec{u}^T\right)$$

$$\vec{f_e} = \nabla \bullet \left(\varepsilon_0 \vec{E} \otimes \vec{E} - \frac{1}{2}\varepsilon_0 E^2 \delta_{i,j} \right) = \rho^e \vec{E} - \frac{1}{2}\varepsilon_0 E^2 \nabla \varepsilon(C)$$

III. Results and Discussion

The above coupled electrohydrodynamic and phase field equations are solved using the finite difference method. The 3D computing is numerical intensive and the parallel computing algorithm is employed to enhance the numerical performance. The numerical model itself is highly versatile, capable of handling any arbitrarily patterned electrodes and the associated complex liquid-air interface morphologies and fluid motion. As an example, the case shown in Fig. 1 is studied here. The characteristic length is chose as the distance of the two electrodes, which is d_0 in Fig. 1(a). The properties of the polymer-air film (polymer to air) are taken as follows: λ_ρ=1000, λ_ε=3, λ_μ=1000, λ_σ=10^4. The template is shown in Fig. 1(b), with the periodic feature of cubic protrusion. The periodic length l_0 is 200nm, and d_0=100nm, Δd=50nm, the cross section of the protrusion is 100nm*100nm. We take

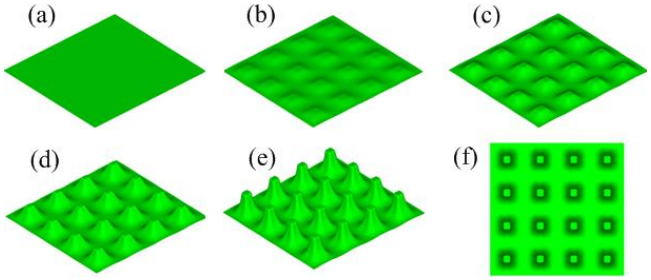

Fig. 2. Numerical simulation for the EHD Patterning, the polymer/air interface is shown here. The corresponding dimensionless time for (a) to (e) are t^*=0, t^*=8, t^*=12, t^*=16, t^*=20.

advantage of the periodic character and compute only one feature area. The computational domain is $l_0 \times (d_0 + \Delta d) \times l_0$. Special treatment is needed for the solid part on the template, the electric field is equi-potential and the flow is set to zero. The mesh is chose as 101x76x101. The featured patterning could be extended to multi-periodic after computation.

Figure 2 shows the computational results of nanostructures formed. Initially, the polymer-air interface is flat. With an external field imposed, the interface starts to deform under the effect of electric force, showing the "pine" like structures (see Figs. 2(c) and (d)). The structure evolves until it grows in height to the template, as evident in Fig. 2(e). The top surface of the nanostructure eventually becomes "flat" as it has touched the template (i.e. upper electrode). In Figs. 2(d) and (e), four ridges on one patterned structure are observed, and each of them corresponds to one corner of the template. Fig. 2(f) is the top view of Fig. 2(e), which illustrates that the structure conforms well with the template. Also as is seen in Fig. 3, the growing rate of the pattern height accelerates with time, which is attributed to the fact that the electric force is stronger as the polymer/air interface gets closer to the template. Then structure height eventually touches the upper electrode and becomes fixed by the geometry constraint.

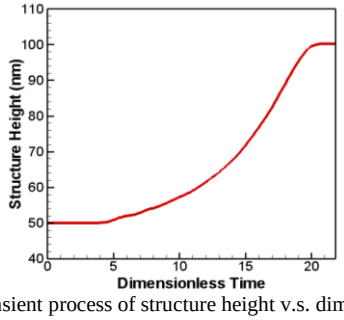

Fig. 3. The transient process of structure height v.s. dimensionless time

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