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Journal of Colloid and Interface Science 311 (2007) 595–608
JOURNAL OF Colloid and Interface Science
www.elsevier.com/locate/jcis

Electric field induced instabilities in thin confined bilayers

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Received 21 September 2006; accepted 28 February 2007

Available online 7 March 2007

Abstract

A long wave nonlinear theory and simulations on the electric field induced instability of a thin (<1000 nm thick) viscous bilayer resting on a solid substrate are presented. The instabilities in these systems are initiated by one of the two basic short time modes of deformation at the twin interfaces—in-phase *bending* or out of phase *squeezing*. Linear stability analysis (LSA) is carried out to identify the conditions for these modes. It is shown that these modes can be switched and the relative amplitudes of deformation at the interfaces can be profoundly altered by varying the thicknesses, viscosities, interfacial tensions and dielectric constants of the films. Nonlinear simulations are presented to support the results obtained from the LSA. In addition, simulations show a number of interesting interfacial morphologies including: (a) embedded upper layer in the array of lower layer columns, (b) columns of the upper layer grown towards the substrate and sheathed by the lower layer liquid, (c) lower layer columns sheathed by the upper layer liquid leading to concentric core–shell columns, (d) droplets of upper liquid on the largely undisturbed lower layer, (e) symmetry breaking traveling waves at the interfaces, and (f) evolution of two different wavelengths at the two interfaces of a bilayer. The effects of viscous and the capillary resistances on the evolution of instability and morphology are also discussed.

Keywords: Thin film; Bilayer; Electric field instability; Interfacial dynamics

1. Introduction

Theoretical understanding of the stability, dynamics, dewetting, and morphology of ultra thin (<100-nm) films has attracted much attention [1–13] because of their presence in various products and processes ranging from coatings, adhesives, flotation, and biological membranes to a host of areas in nanotechnology. In addition to their technological content, thin liquid films are model mesoscale systems for the study of several fundamental scientific issues such as intermolecular forces, self-organization, confinement and finite-size effects, mesoscale dewetting, multilayer adsorption, and phase transitions. Thus, much theoretical and experimental effort has been devoted to understand the instability and dewetting of a thin single layer by various intermolecular forces, most notably the long-range van der Waals force [1–13].

Apart from the van der Waals forces which cannot be modulated, the application of an external electric field is emerging to be another very promising and versatile method for generation and control of micro-patterns using thin (100–1000 nm) polymer melt films. The electric field induced instabilities at the macroscopic fluid interfaces have been studied since the early works by Taylor and McEwan [14] and Melcher and Smith [15]. These works and a review by Saville [16] summarize the electrohydrodynamic instabilities at the interfaces, where the long and the short wavelength disturbances are stabilized by the gravity and the capillary forces, respectively, and the disturbances of the intermediate wave numbers grow to show instability above a critical strength of electric field.

The experimental works from the groups of Russell and Steiner et al. [17–21] have shown that the application of electric field across initially flat interfaces of ultra thin films leads to the formation of regular and hierarchical columnar structures [20]. Lin et al. [22,23] have extended these observations further and shown that when the electric field is relatively weak, the polymers dewet each other under the influence of van der Waals forces, whereas a strong electrical field across the interfaces leads to the formation of columns between the electrodes. These works also show a variety of interfacial morphologies resulting

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0021-9797/$ – see front matter
doi:10.1016/j.jcis.2007.02.089

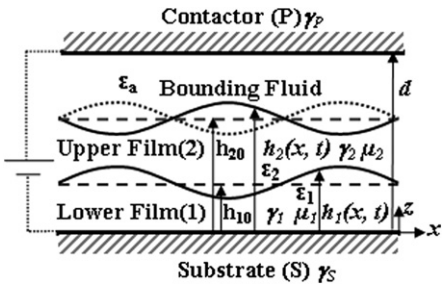

Fig. 1. A schematic diagram of a confined viscous bilayer under the influence of electric field. The electric filed is applied through the electrodes, (i) the solid substrate on which the bilayer rests, and (ii) the contactor that confines it at the top. The mean and the local thickness of the lower layer are h_{10} and $h_1(x,t)$, respectively, and the same for the composite layers are h_{20} and $h_2(x,t)$, respectively. γ_1, γ_2 and γ_S are the surface energies of the lower layer, the upper layer and the solid substrate, respectively. μ_2 and μ_1 are the upper and lower layer viscosity, respectively, and d is the distance between the electrodes. ε_1, ε_2 and ε_a are the dielectric constants of the upper layer, lower layer and bounding fluid like air, respectively.

from a single (liquid–liquid [22]) and multiple (liquid–liquid and liquid–air [23]) deformable interface(s) under the influence of electric fields. The multiple deformable interfaces can also lead to phase inverted columnar structures when subjected to an electric field, as shown in a recent experimental work [24].

The theoretical understanding of electric field induced instabilities in thin films started with the linear stability analyses of a single layer [18,19]. Herminghaus [25] examined the stability of the interface between a dielectric and a conducting fluid and showed that the electric fields can also arise naturally from the contact potentials of the various interfaces and cause instabilities. Pease and Russel [26] have shown that for the systems with leaky dielectric liquid and air, the presence of conductivity leads to patterns of smaller wavelength and larger growth rates of instabilities. More recently, Shankar and Sharma [27] conducted a linear stability analysis of the interface between two leaky dielectric fluids in the presence of an electric field and showed that increasing the viscosity ratio has a profound influence on the pattern wavelength. The short and long time nonlinear morphologies at the liquid–air interfaces because of electric fields are studied in detail by Sharma et al. [28,29]. In conjunction with a linear stability analysis, Matar et al. [30] have shown the details of the nonlinear instabilities at the liquid–liquid interface involving leaky dielectrics.

However, all the previous theoretical studies [18,19,25–30] of electric field destabilization have been restricted to the deformation of a single interface, either liquid–air or liquid–liquid, whereas the electric field induced interfacial dynamics of bilayers involves the deformations of the two coupled interfaces. Fig. 1 shows a schematic of the system considered here. The presence of multiple, confined fluid interfaces, as in a free film, can lead to two distinct initial modes of surface instabilities [31,32], either in-phase *bending* or out of phase *squeezing*, as shown in Fig. 1. Theoretically, bilayer dynamics has been studied previously only under the influence of van der Waals forces. Danov et al. [33–35] presented a comprehensive analysis of the initial growth of instability of evaporating bilayers containing surfactants. The initial [36,37] and the late stages [38–41] of interfacial morphological evolution for the van der Waals bilayers have also been studied extensively. Detailed linear [42] and nonlinear analyses [43] of unstable viscoelastic bilayers have also been pursued. The electric field induced morphologies at the coupled liquid–air and liquid–liquid interfaces of viscous bilayers are of interest in view of their potential in meso-patterning applications and in understanding of a rich variety of behavior uncovered in recent experiments [20,23,24].

In this study, the stability, dynamics and morphology of confined viscous bilayers subjected to an external electric field are considered with the help of linear stability analysis (LSA) and nonlinear long wave simulations. The linear stability analysis is carried out to obtain the time and the length scale of the instability. Nonlinear simulations are presented to show the effects of electric fields and other bilayer parameters on the evolution of interfacial morphologies until the first rupture of either of the two films becomes imminent.

2. Nonlinear long-wave equations for a bilayer

In this analysis, we consider the bilayer fluids to be incompressible and Newtonian. All material properties, such as the interfacial tensions, dielectric constants, viscosities are considered to be constant. Small aspect ratios of the liquid films permit the use of lubrication approximation.

In the following section, x and z are the coordinates parallel and normal to the substrate surface (as shown in Fig. 1), respectively, and t represents time. We follow the convention where the subscript i of the variable denotes a fluid layer ($i = 1$ denotes lower liquid layer and $i = 2$ denotes upper liquid layer), s denotes solid substrate and a denotes a non-viscous fluid like air in the gap between the non-contacting electrode and the upper liquid layer. Thus, for the layer i, u_i is the x-component of velocity, w_i is the z-component of velocity, μ_i is the viscosity and P_i is the total pressure. γ_i and γ_S are the surface tensions of the ith fluid layer and the solid substrate, respectively. The subscripts t, x and z in all the equations denote differentiation with respect to that variable. Both the notations, $h_2 - h_1$ and h_3, represent upper layer thickness. The notations h_{20}, h_{10} and h_{30} represent the respective base state or mean thicknesses.

The long-wave equations of motion for the two layers, $\mu_i u_{izz} = P_{ix}$, the equations of continuity for the two layers, $u_{ix} + w_{iz} = 0$, the kinematic conditions for individual layers $h_{it} + u_i|_{\text{at}\, h_i} h_{ix} = w_i|_{\text{at}\, h_i}$ together with the equations of capillarity for the two interfaces, finally give the equations for the position of interfaces, $h_i = h_i(x,t)$. Gravity is neglected in comparison to electric field forces because films are thin ($<$1000 nm) and thus the instability arising from the density differences is not considered. The following boundary conditions are used: $\mu_2 u_{2z} = 0$ at $z = h_2$ (zero shear at liquid–air interface); $u_2 = u_1$, $w_2 = w_1$, $\mu_2 u_{2z} = \mu_1 u_{1z}$ all at $z = h_1$ (continuity of velocity and shear stress at liquid–liquid interface) and $u_1 = w_1 = 0$ at $z = 0$ (no-slip and impermeability at the solid–liquid interface). Details of the derivation can be found elsewhere [37]. The evolution equations thus obtained shown below describe the stability, dynamics and morphology