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Non-linear dynamics of a viscoelastic film subjected to a spatially periodic electric field

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ABSTRACT

We investigate the non-linear dynamics of the electrohydrodynamic instability of a viscoelastic polymeric film under a patterned mask. We develop a computational model and carry out 2D numerical simulations fully accounting for the flow and electric field in both phases. We perform a thorough parametric study and investigate the influence of the various rheological parameters, the applied voltage and the period of the protrusions of the mask in order to define the fabrication limits of this process in the case of patterned electrodes. Our results indicate that the effect of elasticity is destabilizing, in agreement with earlier studies in the literature based on linear stability analysis for homogeneous electric fields. However, the significance of the normal and shear polymeric stress components is found to change drastically as deformation advances, rendering inappropriate the lubrication approximation that neglects normal stresses. We also find that for low values of the *Ca* number a metastable state arises with finite interfacial deformation, the amplitude of which compares favourably with experimental observations in contrast with earlier predictions using linear theory. Though the critical voltage for this metastable state appears to be unaffected by the elasticity of the material, viscoelasticity affects the fabrication limit on the period of the protrusions of the top electrode.

1. Introduction

The interaction of an externally applied electric field with a liquid can give rise to interesting flow instabilities and pattern formation [1]. The work of Russel and co-workers [2–6] has demonstrated that the application of an electric field to an initially flat polymer–air or polymer–polymer interface results in an electrohydrodynamic (EHD) instability leading to the formation of columnar structures. These instabilities can be used in order to form well-controlled patterns at the microscale and nanoscale with many practical engineering applications.

The electrically-induced flow of thin liquid films has attracted the interest of many theoretical studies. More specifically, Schaffer et al. [3] used the lubrication approximation to determine the dependence of the fastest growing linear mode on system parameters for a polymer-air interface. Lin et al. [4] conducted experimental as well as theoretical work to study the dependence of pattern wavelength on the viscosity ratio in two-layer polymeric systems. Their modelling study predicts the wavelength to be independent of the viscosity ratio for perfect dielectric fluids. Pease and Russel [7] considered the stability of the interface between a leaky dielectric liquid and air and showed that the presence of conductivity exerts a destabilizing influence leading to patterns of smaller wavelength and much larger growth rates. Shankar and Sharma [8] also conducted a linear stability analysis using lubrication theory and their results indicate that, in contrast to the perfect dielectric case, for leaky dielectrics, increasing the viscosity ratio has a profound influence on the pattern wavelength.

More recently, Heier et al. [9] were interested in systems with heterogeneous electric fields and showed through experiments that it is possible to achieve a steady state with finite interfacial deformation when Maxwell stresses in the fluids and surface tension are balanced. They also developed a linear model and were able to derive an expression for the critical voltage beyond which the amplitude grows exponentially in qualitative agreement with their experiments. However, it should be noted that according to Heier et al. [9] linear theory severely underestimates the amplitude of the steady finite deformations in comparison with experimental observations.

The nonlinear evolution of two leaky dielectric layers in a homogeneous electric field was examined by Craster and Matar [10] showing that initially small perturbations grow under the action of the destabilizing electrical forces and eventually their amplitude saturates in the non-linear regime to give rise to spatially periodic

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patterns. Two-dimensional numerical simulations using the lubrication theory helped in elucidating the interfacial evolution, the role of the initial thickness ratio and the effect of patterned "masks" on the observed three-dimensional patterns [11–14]. Several studies have also been devoted in the investigation of the stability and dynamics of bilayers under air or another viscous liquid [15–19]. Finally, the effect of AC fields has been taken into account through linear stability analysis and non-linear simulations by Roberts and Kumar [20] and Gambhire and Thaokar [21].

As discussed above, the surface instability of a Newtonian fluid under the effect of electric field has been studied extensively by several researchers and it is now well understood. The dynamics of fluids with complex rheology, however, has received much less attention in the literature. The first attempt to take into account the polymer viscoelasticity in electrically-induced flows was made by Wu and Chou [22]. These researchers used the lubrication theory and performed a linear stability analysis of a initially static thin polymer film underneath a flat electrode using the Oldroyd-B constitutive equation for the elastic stresses. Their results have shown that the polymer elasticity destabilizes the system and when the Deborah number is large enough, a resonant phenomenon appears as a result of the interaction between the two destabilizing mechanisms (the electrostatic force and the polymer elasticity). Later on, Tomar et al. [23] used a linear constitutive equation for the stresses (Jeffreys model) and presented a linear stability analysis taking also into account the effect of inertia. Interestingly, they found that in the presence of a small amount of inertia the wavelength of the fastest growing mode (i.e. the dominant lengthscale of the instability) is independent of the rheological properties such as relaxation time and solvent viscosity whereas the growth rate is affected significantly. Their findings were confirmed recently by Espin et al. [24] using an asymptotic expansion. The latter authors also examined the viscoelastic effects under the influence of AC fields and found that the impact is largest when the relaxation time and oscillation time scale are comparable. In the case of AC fields, it is shown that the wavelength is also affected contrary to the predictions of linear theory for the case of DC fields [23]. The rheological characteristics of the fluid were also shown to play a role in the case of trilayers, indicating that its effect on the evolution of two coupled interfaces is more involved than a purely kinetic role [25]. It should be noted here that the aforementioned studies for viscoelastic fluids considered homogeneous electric fields (flat electrodes) and the linear stability analysis was performed around a quiescent base state. However, in the case of a patterned mask the field becomes heterogeneous and growth generates a time-dependent base state for which linear or weakly non-linear stability analysis is difficult necessitating the use of time-dependent simulations.

As was noted above, most of the research studies in the literature employ linear theory, which is valid only for small disturbances. One crucial issue, however, is not only to predict the band of unstable wavenumbers in the linear regime but also to determine accurately the behaviour of the system in the non-linear regime. For the latter, the majority of the research groups make use of the lubrication theory in order to interpret experimental results. Pease and Russel [26,27] argued, however, that in many cases the experiments were carried out for conditions under which the lubrication approximation is not strictly valid. They compared the predictions of a generalised model with those of lubrication theory against experimental results and found a better agreement with the former. Very recently, a detailed comparison was also presented by Gambhire and Thaokar [21] for both DC and AC fields, which indicated large deviations for the predicted wavelength. Moreover, in the case of viscoelastic fluids, the deficiencies of the lubrication approximation are expected to be enhanced due to the significant underestimation of normal stresses and to the fact that non-linear viscoelastic effects are not taken into account.

Examples of fully non-linear simulations without making use of the lubrication approximation are the works of [28–31] who studied primarily cases involving heterogeneous electric fields. Yang et al. [30], motivated by the work of Heier et al. [9], considered a sinusoidally patterned top electrode and performed non-linear simulations using a boundary/finite element method to determine the critical parameters for instability of the liquid film. Their results indicate that linear analysis can significantly over-predict the critical voltage for instability. Li et al. [31] were also interested in heterogeneous electric fields and investigated the effect of various geometric features of the patterned electrode to determine the fabrication limits of this process using a diffuse interface method.

The scope of this work is to investigate the non-linear dynamics of a viscoelastic material under the influence of an heterogeneous electric field taking fully into account the viscoelastic effects. We avoid making any assumptions, such as using lubrication approximation, in order to describe the flow dynamics as accurately as possible. We perform two-dimensional transient numerical simulations, using the finite element method combined with an elliptic grid generation scheme for the determination of the unknown position of the interface. The viscoelasticity of the polymeric film is taken into account using the affine Phan–Thien Tanner model. We perform an extensive parametric analysis to determine the effects of the various geometric and rheological parameters on the evolution of the interface and on the fabrication limits of this process. Our results indicate that the elasticity of the material does not affect the critical voltage for instability but affects the fabrication limit on the period of the top electrode protrusions. We also discuss about the validity of lubrication theory in the case of viscoelastic materials.

The remainder of the paper is organized as follows. In Section 2, we describe the system of governing equations and outline the numerical method used for its numerical solution. The results are presented and discussed in Section 3. Finally, the concluding remarks are given in Section 4.

2. Problem formulation

We consider the dynamics of two perfect dielectric fluids sandwiched between two rigid, and impermeable electrodes. The electrodes can be either flat or periodically patterned as shown in Fig. 1; *w* and *p* denote the width and the height of the protrusions, respectively, and *s* denotes the spacing between the protrusions. The bottom fluid is considered to be a polymeric viscoelastic film surrounded by a Newtonian liquid, with initial thickness, *d*. Both fluids, which are initially stationary, are taken to be incompressible with the lower (upper) fluid having a density ρ_1 (ρ_2), dielectric constants ϵ_1 (ϵ_2); these properties are assumed to be constant. The viscosity of the upper fluid is also constant and denoted by μ_2. The viscoelastic fluid has a zero-shear viscosity $\mu_1 = \mu_s + \mu_p$, where μ_s and μ_p are the viscosities of the solvent and the polymer,

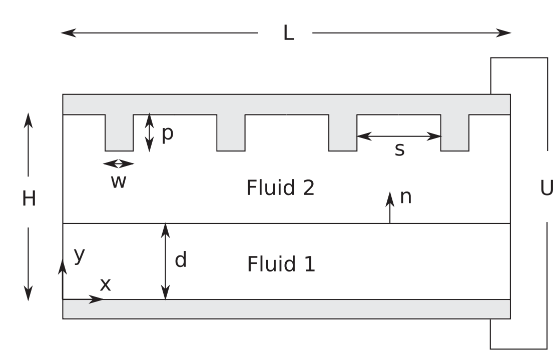

Fig. 1. Schematic of the flow geometry.