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Electrowetting Film Formation and Capillary Spreading in Dielectrics: A Theoretical Approach

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Abstract: In this study, we have shown that the film formation and capillary spreading on a typical dielectric (real surfaces) is dependent mainly on the contact angle and contact angle hysteresis with the contact angle of the quasistable spreading (for $l_c \gg R$) given as $\theta_d = \tan \theta_s - (9 \log b C_a)^{1/3}$.

Key words: Contact angle, capillary spreading, dielectric, electrowetting

INTRODUCTION

The interface between liquid metal and electrolyte has an intrinsic property that reveals important information about adsorption process. The adsorption of ions on the interface causes the surface tension across the interface to vary significantly on the application of electrical potential difference. The surface tension has been revealed in many scaling analyses to be one of the most important parameter for microscale devices and structures especially, for microfluid based devices. The magnitude of the interfacial tension has been found to be a characteristic of the particular liquid in its pure form and it is dependent on external parameters like temperature and electric field strength, even by addition of impurity (Ikata, 1991).

Wetting and film spreading are of key importance for many applications. At microscopic level, wetting or non-wetting plays an important role for oil recovery (Bertrand *et al.*, 2002; Wooding and Morel-Seytoux, 1976), the efficient deposition of pesticides on plant leaves (Bergeron *et al.*, 2000) and the cooling of industrial reactors. On a microscopic scale, wetting solutions have been proposed to resolve technological problems in microfluidics and nanoprinting, inkjet printing etc (Tabeling, 2004; Asai, 1991; Allen *et al.*, 1985). All these phenomena are governed by the surface and interfacial interactions, acting usually at small (a few nm for van derWaals or electrostatic interactions) or very small (molecular) distances.

Wetting phenomena are ubiquitous in nature and technology. The concept of electrical double layer (EDL) is vital in describing the concept of electrowetting. The interface between two phases tends to be electrically charged due to unbalanced directional forces exerted on the particles near interfacial region. Among all the interfaces, metalelectrolyte interface is of primary significance in electrochemistry because of its well-defined electrified interface as well as its frequent use in electrochemical systems. Many models have been proposed for the understanding of the electrical double layer. For instance, Bockris and Reddy (1970) proposed a simplified model based on charged capacitor called Helmholtz-Perrin model.

The skill to control the wettability of a liquid, perfectly without any form of mechanically moving parts is paramount in the actuation of fluids in microfluidic devices. This has prompted a recent resurgence in studies on electrowetting, which allows a rapid, reversible and precise means for

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manipulating small liquid volumes with relatively low power consumption (Leslie *et al.*, 2006). Also, success in generating fluid velocities in excess of several cm s^{-1} has generated momentous interest in electrowetting for other applications such as electrostatic-assist coating (Blake *et al.*, 2000) and miniaturization of optical focusing devices (Berge and Peseux, 2000).

The understanding of the underlying principle in electrowetting and electrocapillarity is the key to the miniaturizing of flow control devices because; they show active use of surface tension at microscale, which is relatively a large force as compared to gravity or structural stiffness (Kwang-Seok *et al.*, 2002). Beni *et al.* (1982) had demonstrated the importance of surface-tension-driven motion of a mercury drop in a glass capillary and he applied the principle to an optical switch. The advantage of miniaturization of flow control devices is overwhelming; for instance, increased speed, parallelization of analysis and portability.

The present research is aimed at studying theoretically, the effect of capillary spreading, consequent upon the nature of the surface on electrowetting film formation on the basic properties of microscale devices. Attempts will also be made to develop a model that will link the electrokinetic and wetting hydrodynamic theories as to predict the spreading dynamics of the impulsively electrowetting films for a dielectric material.

THEORETICAL APPROACH

In this study, we present our theoretical approach to study the electrowetting film formation and capillary spreading. We will present a general theoretical framework for the controlling macroscopic parameters of film formation on a parallel plate electrode configuration. The second part of this section presents the dynamic theory that we use to study the kinetics of the film formation.

On the application of external electric field, the molecules of a dielectric material at the point of adsorption site is polarized leading to increased electrostatic attraction between the molecules with accompanying reduction in the free energy of the system and the magnitude of the induced electric field lowering the energy which is related to the change in surface tension of the liquid and dependent on the polarization factor and the field strength. Hence, the energy U_a according to the Tabor (1979) becomes

$$U_a = U_{a0} + \frac{1}{2}p \cdot E_1 \quad (1)$$

where p is the dipole moment of the molecule, U_{a0} is the heat of adsorption in zero field and E_1 is the local electric field strength.

Basic Formulation

The governing equation for the spreading of a high permittivity polar dielectric classical liquid drop with volume V , constant density ρ_1 and viscosity μ_1 lying on a horizontal rigid and impermeable solid substrate, under electrocapillary action with electrode configuration of separation R_e placed horizontally on the solid substrate where the electric field is predominantly orthogonal to the three-phase contact line is that which obeys the Maxwellian force F due to dc or ac electric field expressed as

$$F = \rho_f E - \frac{\epsilon_0}{2} \nabla \left[\epsilon - \rho \left(\frac{d\epsilon}{d\rho} \right)_T \right] E \cdot E_n \quad (2)$$

where ϵ is the dielectric constant, T the temperature and ρ the density of the body.

Thermodynamics

A liquid drop on a solid substrate has basically; three different phases present (Fig. 1). Hence, three surface tensions are needed for full description of the molecular behaviour on the surface: solid-liquid, liquid-gas and solid-gas.

For instance, the shape of the film formed by a hydrophobic surface is determined by the liquid volume and the equilibrium contact angle between the liquid and the surface; where the energy of the system reaches a local minimum. Assuming the droplet volume to be of order of μ_l , the gravitational effect thus has a minimal effect and the droplet shape thus approximates that of a sphere. Since the contact equilibrium depends on the surface condition (Altti, 2003) and assuming an ideal surface: rigid, planar, homogeneous and smooth; the contact angle thus approaches Young's angle which can be approximated from the force balance between the interfacial tensions at the three-phase contact line formed by solid, liquid and vapour (Fig. 1). The force balance leads to the well known Young equation.

$$\gamma_{lv} \cos \theta_0 = \gamma_{sv} - \gamma_{sl} \tag{3}$$

where θ_0 is the Young's contact angle and γ_{lv} , γ_{sv} and γ_{sl} are the inter-surface tensions at the liquid-vapour (air), solid-vapour and solid-liquid interfaces, respectively. Here all the surface tensions are defined when the three phases: solid, liquid and vapor are in equilibrium with each other. If the three tensions are known, the wetting state of the fluid follows directly. If the substrate-vapor tension is low, a droplet with a finite contact angle minimizes the free energy of the system; we speak of partial wetting. On the other hand, if the tensions are such that the sum of the substrate-liquid and the liquid-gas surface tension equals the solid-gas interfacial tension, the contact angle will be zero.

A cursory look at Eq. 3 shows that the contact angle is a measure of the wettability of the surface (interface), thus, surfaces with contact angle less than 90° is said to be hydrophilic while surfaces with contact angle greater than 90° is said to be hydrophobic.

However in real surfaces, the contact angle is not exactly the Young value but varies within a certain range around it reaching minimum and maximum values called receding and advancing contact angles respectively; with their difference termed contact angle hysteresis. This parameter is related to real surfaces, attributed to roughness and inhomogeneity of the interface (Altti, 2003). The importance of the contact angle can be seen from Eq. 4 as it gives the force of a moving liquid on a surface:

$$F_s = \gamma_{lv} w (\cos \theta_r - \cos \theta_a) \tag{4}$$

where w is the width of the droplet bottom normal to the direction of motion of the advancing angle, θ_a and receding angle, θ_r given as

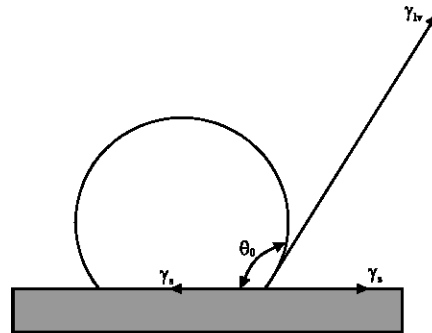


Fig. 1: Droplet actuation on a an ideal surface

$$w = 2R\sin\left(\frac{\theta_a + \theta_r}{2}\right) \quad (5)$$

Electrowetting

Electrowetting is an electrically-induced change of a material’s wettability (Kwang-Seok *et al.*, 2002) of which the contact angle is the decisive factor in controlling this electric field normally applied between droplet and electrode. The electrostatic force acting on the ions on the interface can be observed to be the reduction of the contact angle between solid and liquid as shown in Fig. 2a and b. Electrowetting is treated as charge-induced change in the interfacial energy between solid and liquid (Altti, 2003), which its overwhelming contribution in droplet actuating has lead to its proposal as the actuating driving mechanism according to Lee *et al.* (2001) and Pollack *et al.* (2000).

Basically, the term electrowetting defined as the change in the solid electrolyte contact angle due to applied potential difference between the solid and the electrolyte, is akin but distinct from electrocapillarity. It was first introduced in 1981 in an attempt to describe the effect proposed for designing a new type of display device (Beni and Hackwood, 1981) but did not see the light of the day until it was experimentally verified by Berge (1993). Berge’s analysis was merely based on Lippmann’s theory except for the replacement of the capacitance solid-liquid double layer by the insulator-separation capacitance between droplet and electrode which is reasonable enough as the insulator is far thicker than the double layer with the majority voltage drop prominent across the insulator. Attempts have also been made to relate experimental findings of electrowetting on a wide variety of dielectric or polymeric substrates by correlating the measured change in the contact angle (Tom, 2004; Decamps *et al.*, 2000) against the Lippmann equation (Leslie *et al.*, 2006) in this case, a consequence of the applied voltage V given as

$$\cos\theta_v = \cos\theta_0 + \frac{1}{2} \frac{\epsilon_0 \epsilon}{\gamma_{lv} t} V^2 \quad (6)$$

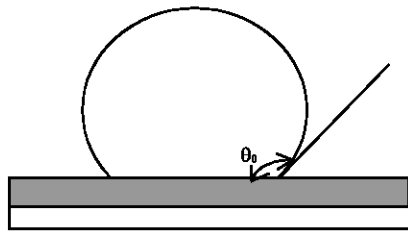


Fig. 2a: Generic electrowetting effect without any applied field

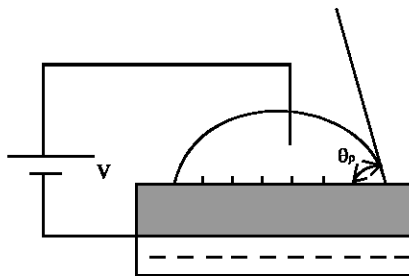


Fig. 2b: Generic electrowetting set-up with applied field

where θ_v is the contact angle when a potential is applied (apparent contact angle), θ_0 is the contact angle in the absence of an electric field (or initial contact angle), ϵ_0 the permittivity of free space, ϵ the dielectric permittivity of the insulating layer, V the applied potential, t the dielectric film thickness and γ_{lv} the liquid-vapour interfacial (surface) tension.

It must be pointed out that it has been proposed that the observed change in contact angle is due to change in line tension; hence casting overwhelming doubts and need for further elucidation of the electrowetting phenomenon.

Electrocapillary Film Spreading

The wetting of dielectrics is driven by molecular forces normally at the contact line. But for highly hydrophobic fluids with small dynamic contact angles, θ_d molecular dynamics can be studied applying macroscopic slip models using continuum descriptions according to the author Kalliadasis *et al.* (1996). A typical thin molecular precursor film, where liquid from the bulk of the drop drains as a consequence of the negative disjoining and capillary pressures have also been observed experimentally using ellipsometry for wetting fluids and also in molecular dynamic simulations (Leslie *et al.*, 2006). Though, much thinner than the macroscopic precursor film, it has been observed that in most simulations, that the thickness of this precursor film has no significant effect on the overall wetting dynamics (Kalliadasis *et al.*, 1996). In electrocapillary film formation and spreading, the key parameter is the precursor film thickness. Following Leslie *et al.* (2006), in the limit of the locally quasistable limit (here, $L/\tilde{L} \gg 1$ where L is the length scale of the macroscopic region and \tilde{L} is that for precursor film region) the approximation in the bulk section with a constant dimensionless speed Ca , with reference to a coordinate frame advancing gradually with the contact line is

$$3C_a h_x = (h^3 h_{xxx})_x \quad (7)$$

where C_a is the capillary number given as

$$C_a = \frac{U\eta}{\gamma} \quad (8)$$

usually small ($C_a \approx 10^{-5} - 10^3$) for most spreading experiments; η is the viscosity of the liquid and U is the contact line speed. Note for a case where the drop radius is smaller than the capillary length

$l_c = \sqrt{\frac{\gamma}{\rho g}}$ an equilibrium liquid-vapour phase of the drop is obtained. The viscous force has very little

or no significant effect on the interfacial shape of the droplets formed for those far from the contact line such that the shape looks like a spherical cap as if the system were in static equilibrium. Hence for sufficiently flat droplet ($h' \ll 1$), the film thickness, $h(r, t)$ is

$$h(r) = \frac{2V}{\pi R^2} \left[1 - \left(\frac{r}{R} \right)^2 \right] \quad (9)$$

However, at a certain volume, V , the major determinant of the shape of the drop is the apparent contact angle, θ_{ap} and the spherical cap with the interface makes the angle given as

$$\tan \theta_{ap} = \frac{4V}{\pi R^3} \quad (10)$$

As the drop spreads, $\theta_{sp}(t) \rightarrow 0$ to give the approximate drop radius $R(t)$ in terms of the Tanner's law (Tanner, 1979; Voinov, 1976; Ogarev *et al.*, 1974) as

$$R(t) = \left[\frac{10\gamma}{9B\eta} \left(\frac{4V}{\pi} \right)^3 t \right]^n \sim t^n \quad (11)$$

where $n \sim \frac{1}{10}$ is a constant and B is also a constant found from theoretical approximation to be $B^{1/10} \sim 1.1860$.

Taking cognizance of the asymptotic matching between the outer bulk region and the inner molecular precursor film region after redimensioning, we obtain from (7) that

$$h \sim -x(9C_a \log \eta)^{\frac{1}{3}} \quad (12)$$

From which we obtain maintaining the scaling point of view and a precursor film model, that the suitable macroscopic contact angle θ_d during the quasistable spreading of a completely or partially wetting liquid under conditions of viscous flow and small gradient (for $l_c \gg R$) is given as

$$\tan \theta_d = \tan \theta_s - (9 \log b C_a)^{\frac{1}{3}} \quad (13)$$

where $b \sim \eta = \frac{\tilde{L}}{L}$ is the precursor film thickness known as the Hamaker constant and θ_s is the static contact angle and this matching correlates with the contact angle condition for the slope that scales as $C_a^{\frac{1}{3}}$ in total agreement with Tanner's law.

CONCLUSIONS

It has been observed that the spreading dynamics above all things depends mainly on the contact angle along the contact line driven mainly by the molecular forces, which induces potential gradient in the dielectric interface. The wettability of any surface (be it hydrophobic or hydrophilic) is also noted to be contact angle dependent.

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