Electrowetting on Dielectrics (EWOD): Reducing Voltage Requirements for Microfluidics

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INTRODUCTION

Surface tension is an inherently dominant force in the microscale. Electrocapillarity, the modification of surface tension by applying an electric field, has received renewed attention because of its potential usefulness in microfluidics. Pollack et al. demonstrated a microactuator for rapid manipulation of discrete microdroplets by direct electric control of the surface tension. [1] Berge and Peseux demonstrated the use of electrocapillarity in order to change the contact angle of a transparent drop, creating a lens of variable focal length. [2] Prins et al. showed the control of fluid motion by electrocapillary pressure in three-dimensional structures containing thousands of microchannels. [3]

Electrowetting was first demonstrated by Berge. [4] The general experiment scheme for electrowetting on dielectrics (EWOD) is shown in Figure 1.



Figure 1. Open EWOD Experiment. A) amp meter; B) power supply; C) digital camera.

A potential is applied between a conducting liquid on an insulating layer and a counter electrode positioned below the insulating layer. Charge accumulates at the solid-liquid interface, leading to a change in contact angle from hydrophobic to more hydrophilic, shown in Figure 2.



Figure 2. Change in Contact Angle with Applied Potential.

Large changes in contact angle were observed for droplets on a dielectric-covered conductive substrate. Using the setup shown in Figure 1 with the counter electrode coated with dielectric (Teflon-AF1600 on Parylene), Walter and Fokkink applied a potential (0-200V) to acheive fast and reversible switching of the contact angle (from 110 to 60°) of aqueous droplets containing 10^{-4} M KNO₃. [5] They and others have found that at higher applied potentials, the contact angle does not decrease further with increasing potential (contact angle

saturation) contrary to theoretical predictions. The origin of contact angle saturation at high applied potential is not understood.

Walter and Fokkink [5] applied 200 V to obtain maximum contact angle change with their dielectric-coated (Parylene and Teflon AF). Verheijen and Prins [6] used 250 V for their EWOD experiments with Parylene and Teflon-coated counter electrode. Equation (1) shows the relationship between contact angle and applied voltage.

$$\cos\theta_{V} = \cos\theta_{o} + \frac{1}{2} \frac{\varepsilon\varepsilon_{o}}{\gamma_{IV}t} V^{2}$$
(1)

where θ_V is the contact angle when a potential is applied, θ_o is the contact angle without applied potential, ϵ is the dielectric constant of the insulating layer, ϵ_o is the dielectric constrant of air, γ_{LV} is the surface tension at the liquid-vapor interface, t is the thickness of the insulating layer, and V is the applied voltage. As the equation shows that the smaller the thickness t or the larger the dielectric constant ϵ , the larger the contact angle change at a given applied voltage. Or a smaller potential is needed in order to obtain the desired contact angle change. The lowest voltage for EWOD reported to-date is 30 V. [1] To reduce these voltage requirements, our strategy has been to use a thinner dielectric film with a higher dielectric constant.

In this paper we describe the conditions to achieve maximum contact angle changes for EWOD, including the thickness of the dielectric layer, the dielectric film contiguity and smoothness, and the dielectric constant. We show that by decreasing the thickness and roughness of the dielectric layer and by increasing its dielectric constant, the threshold voltage for EWOD can be reduced to ~6 V.

EXPERIMENTAL

Hexadecanethiol (C16SH) on gold. A hexadecanethiolate (C16S⁻) self-assembled monolayer (SAM) was prepared on a vacuum evaporated Au substrate (2000 Å, with 100 Å thick Cr underlayer on Si wafer) by immersing the Au substrate in 2 mM C16SH in ethanol inside a nitrogen-purged glove box for 12 h. The monolayer was then rinsed thoroughly with water. The thickness of the monolayer is about 30 Å, as measured by an ellipsometer.

Octadecyltrichlorosilane (OTS) on silicone dioxide. The substrate (1000 Å or 7000 Å SiO_2) was transferred to a 5 mM solution of octadecyltrichlorosilane (OTS) in toluene (distilled over CaH_2) inside an argon-filled glove bag. After 15 h, the substrate was removed from the solution, rinsed with chloroform (2 x), sonicated in chloroform, washed with SuperQ-filtered water (Super-Q Water System, Millipore Corp., Bedford, MA), and dried under a stream of argon.

Parylene-C (Parylene). A Parylene thin film (1.16 $\mu\text{m})$ was deposited on a silicon substrate using an SCS parylene evaporation deposition system.

Teflon. A 6% Teflon AF solution (DuPont, Wilmington, Delaware) was spin-coated on the silicon substrate at 1600 rpm for 50 s, giving 1.2 μ m-thick Teflon film. The film thickness was controlled by varying the concentration of Teflon solution by diluting with Fluorinert FC 75 (3M Company, St. Paul, MN). Twelve different thicknesses of Teflon, from 1.65 μ m to 130 Å, were obtained by spin-coating various solutions at 1000 rpm for 30 s.

Open EWOD Experiment. The contact angle change as a function of applied voltage was examined through a digital camera, as shown in Figure 1. For each substrate, 10 μ l droplets of Super-Q water or 0.1 M aqueous NaClO₄ solution were deposited on the dielectric by pipet. By magnifying the droplet image, small contact angle changes could be detected. 1-mm diameter platinum wires were used as the working and counter electrodes. An amp meter was used to determine the breakdown voltage, as described below.

RESULTS AND DISCUSSION

EWOD Experiment. A typical EWOD result under ambient conditions is shown in Figure 2. The contact angle changes from $> 90^{\circ}$ to $< 90^{\circ}$ upon applying a potential to the droplet. The contact angle was measured as a function of applied potential, and the reversibility of wetting was also checked. The threshold voltage is the voltage at which the contact angle changes visibly. The breakdown voltage is

defined as the voltage at which current flows and the contact angle change is irreversible.

For SAMs of both C16SH on gold and OTS on SiO₂, wetting was not observed until current was detected by the amp meter. At this point, the contact angle change was irreversible because of degradation of the monolayer by desorption. Dielectric thickness and EWOD test results for samples are shown in Figure 3 and Table 1. Figure 3 shows that both the threshold and breakdown voltages increase as the film thickness is increased. We achieved ~6V of threshold voltage for EWOD on the 133 Å Teflon layer. Wetting was reversible for thick Teflon films. However, contact angle saturation was observed at high voltage, and the saturation angle was about 60°.



Figure 3. Teflon Thickness vs. Voltage.

Table 1. Teflon Layer Thickness and EWOD Tes	st Data
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Concent- ration (wt.%)	Thickness (Å)	Threshold voltage (V)	Breakdown voltage (V)
6.0	16500	46	>300
5.5	13500	45	280
5.0	12000	43.5	250
4.5	10000	41	250
4.0	7000	38	225
3.0	5500	30	120
2.0	2500	23	100
1.5	1000	18	80
1.0	800	15	75
0.6	500	15	23
0.2	200	9.5	12
0.1	130	5.9	12

Surface roughness and heterogeneity are known to affect the contact angle and contact angle hysterisis. Silicone oil was used to smooth the Teflon surface for EWOD testing. Silicone oil treatment enhances reversibility of EWOD, and it slightly lowers both threshold and breakdown voltages. For the thinner Teflon films, a dependence on the voltage polarity on EWOD results was noticed. Reversible EWOD was only observed when a positive potential was applied to the droplet. Negative applied potential causes electrolysis.

Parylene has a higher dielectric constant (2.7) than that of Teflon (1.9~2.1). It was expected from equation (1) that a higher dielectric constant material would require a lower threshold voltage for EWOD. The 45 V required by Parylene is only slightly lower than the 46 V needed for EWOD on 1.65 μ m Teflon. This result suggests that a much higher dielectric constant is required to lower the EWOD-activating voltage significantly. Parylene has been reported not to break down at 250 V applied. [6] We note also that Parylene is a good insulator, but it is not as hydrophobic as Teflon, which dimishes the effect of electrowetting.

EWOD Theory. Equation (1), used to describe the electrowetting phenomenon on EWOD, was derived by combining the Young equation with the assumption that the change in surface tension at the solid/liquid interface is the energy stored in a capacitor across the dielectric. [4] Other researchers have interpreted equation (1) as a combination of the Young and Lippmann equations. The Berge's assumption and the Lippman equation are applicable only to a simple parallel-capacitor (Helmholtz) model. The EWOD system, however, is asymmetric, having *two* interfaces at which the charge distributions are *not* identical: the electrode-dielectric interface and the liquid-dielectric interface. A correct model with predictive value must describe the potential-dependent charge distribution at both interfaces, and be able to explain experimental observations such as contact angle saturation.

We have begun refining the existing model to more accurately describe the EWOD phenomena. The problem of contact angle saturation, which has been found in the literature [5,6] and also in our experiments, cannot be predicted by the model. Several explanations for the saturation phenomena were published. [6-8] More subtle factors that nevertheless must be considered in EWOD device applications and incorporated (analytically or empirically) into the model include the potential-dependence of the surface tensions at the liquid-vapor and the solid-vapor interfaces. The model must describe the potential-dependent charge distribution at both solution-dielectic and dielectric-counter electrode interfaces, in order to be able to explain experimental observations such as contact angle saturation.

CONCLUSION

"Open EWOD" experiments, in which changes in droplet contact angle on a flat dielectric surface, were performed. Dielectric films consisting of self-assembled monolayers, Parylene, and Teflon of various thicknesses were evaluated. EWOD was achieved with only a 6 V applied potnetial for a 130 Å thin Teflon film, a 5-fold lowering of the threshold voltage compared with the most recent literature report. This was accomplished by decreasing the thickness of the dielectric layer, improving dielectric film contiguity and smoothness, and decreasing droplet size. It is expected that an additional 5-fold reduction in voltage, as well as improved reversibility, can be obtained by further optimizing these parameters and increasing the dielectric constant of the insulating layer. However, the problem of contact angle saturation requires further study because the existing theoretical model does not predict saturation.

Treatment of Teflon surfaces with silicone oil improves the reversibility of the EWOD effect, but may introduce contamination. We are currently working on the fabrication of smooth thin dielectric films with a higher dielectric constant to avoid silicone oil treatment. Also, a refined theoretical model will be a guide for us to avoid the problem of contact angle saturation and to obtain reversible wetting.

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REFERENCES

- Pollack, M. G.; Fair, R. B.; Shenderov, A. D. Appl. Phys. Lett. 2000, 77, 1725.
- 2. Berge, B.; Peseux, J. *Eur. Phys. J. E* 2000, *3*, 397.
- 3. Prins, M. W. J. ; Welters, W. J. J. ; Weekamp, J. W. Science **2001**, *291*, 277.
- 4. Berge, B. C. R. Acad. Sci. Ser. II 1993, 317, 157.
- 5. Welter, W. J. J.; Fokkink, L. G. J. Langmuir 1998, 14, 1535.
- 6. Verheijen, H. J. J.; Prins, M. W. J. Langmuir 1999, 15, 6616.
- 7. Vallet, M.; Vallet, M.; Berge, B. Eur. Phys. J. B 1999, 11, 583.
- 8. Vallet, M.; Berge, B. Polymer 1996, 37, 2465.