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Reversible electrowetting on silanized silicon nitride

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ABSTRACT

Electrowetting is a phenomenon that has received increasing attention in recent years. It offers much promise for applications in lab-on-a-chip devices, liquid lenses, and optical displays. The contact angle can be controlled electrically facilitating the transport of liquid droplets by electrocapillary forces. This paper focuses on the requirements of the dielectric layers in electrowetting on dielectric; firstly, to provide the required dielectric qualities, that is, sufficient dielectric strength and high capacitance and, secondly, to satisfy the surface wetting characteristics, that is, sufficiently high hydrophobicity and low contact angle hysteresis. We present experimental work that, firstly, compares the dielectric characteristics of several insulating layers and secondly, proposes silanization as a way to form hydrophobic layers for electrowetting. The relation between electrowetting and dielectric breakdown was tested by means of measuring the contact angle of a water droplet placed between two electrodes on an insulated surface. The leakage current and resistance between these electrodes were measured as a function of applied voltage for the following dielectric coatings: silicon dioxide, silicon nitride and Parylene C. We compare the contact angle response of electrowetting on octadecyltrichlorosilane (OTS) monolayers and plasma-enhanced chemical vapour deposition Teflon-like fluoropolymer layers. The contact angle hysteresis of the OTS monolayers is clearly lower and this results in reversible electrowetting, that is, the hydrophobicity is sufficient to avoid contact line pinning and dewetting occurs on removal of an applied potential.

1. Introduction

Electrowetting is an electrokinetic means of manipulating a droplet by electrical modification of its surface tension. It has excited much interest because of its potential for application in lab-on-a-chip devices, liquid lenses, microfluidic systems and optical displays [1–3]. An electrokinetic force pulls the droplet along the gradient of the surface energy [4]. In this paper, we will concentrate on the electrical and surface wetting properties of the insulating layers used in electrowetting on dielectric.

Lippmann first described how an applied voltage leads to a change in the contact angle [4]. In electrowetting on dielectric (EWOD) the variation of the contact angle θ on application of a voltage U can be predicted by

$$\cos\,\theta(U) = \cos\,\theta(0) + \frac{\varepsilon_0\varepsilon}{2d\gamma}U^2 \qquad (1)$$

where $\theta(0)$ is the contact angle of the liquid in absence of applied voltage, ε_0 is the permittivity of free space, ε is the dielectric constant of the insulating layer, d is the thickness of the insulating layer and γ is the liquid surface tension. Contact angle saturation is a phenomenon that limits the validity of Eq. (1) [5].

In this paper, we measure the contact angle, leakage current and resistance of an electrowetting insulation layer with regard to applied voltage. We examine a number of dielectric coatings electrically in order to assess their viability for use in an electrowetting device. Moon et al. [6] were the first to focus on the dielectric layers used for EWOD with their main experimental setup being based on thermally oxidized silicon wafers. The relation between contact angle saturation and electrowetting was proposed by Papathanasiou and Boudouvis [7] and subsequently it was shown experimentally that contact angle saturation and dielectric breakdown occurred at the same applied voltages for very thin silicon dioxide layers but that dielectric breakdown can be improved with a stacked oxide–nitride-oxide layer [8]. Unfortunately, thin silicon dioxide films suffer from significant problems with regard to dielectric breakdown and electrolysis when they are in contact with aqueous electrolyte [9–12]. Dash et al. [9] suggested that ion mobility in silicon dioxide films leads to an increased leakage current [13,14]. This paper will focus on the performance of silicon nitride and Parylene C as insulating layers in electrowetting.

In addition, we compare hydrophobization of silicon nitride by deposition of a plasma-enhanced chemical vapour deposition (PECVD) Teflon-like fluoropolymer layer with hydrophobization by

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means of a self-assembled monolayer of hydrophobic silanes. It is a particular goal of our work, to find an alternative hydrophobic layer to Teflon® AF. Most implementations of electrowetting have made use of Teflon® AF, an amorphous fluoropolymer that is applied to the electrodes by spin-coating or dip-coating [1,6,8–10,15,16]. Teflon® AF is without argument an excellent material choice for use in electrowetting. Nevertheless, its use has some significant practical disadvantages: it is relatively expensive and, perhaps more significantly, its use is limited by licensing agreements with DuPont for both research and commercial purposes. Abdelgawad and Wheeler [17] have opened the debate towards the development of less expensive electrowetting devices and presented the use of Saran™ wrap (a consumer-grade polyethylene film) as an insulating layer and Rain-x® water repellent as a hydrophobic layer. Bayiati et al. [18,19] have claimed that PECVD deposited Teflon-like fluoropolymer films offer similar hydrophobicity to that of commercially available spin-coated Teflon layers and also display low protein adsorption with regard to biotechnological applications. Here, we examine the use of Teflon-like fluoropolymer film and a self-assembled layer of silanes as hydrophobic layers in electrowetting with regard to contact angle hysteresis.

Electrowetting on top of self-assembled monolayers of thiols on gold electrodes has been reported by Sondag-Huethorst and Fokkink [20,21]. These monolayers were only stable within the limits of double-layer charging and at higher applied potentials they provided no insulation to the flow of current and electrolysis occurred. Both Lin et al. [11] and Saeki et al. [12] performed some experimental work on the suitability of silanes as a hydrophobic layer for electrowetting. To this end, they assembled octadecyltrichlorosilane (OTS) monolayers on silicon dioxide thin films. These setups were limited by the onset of electrolysis presumably caused by the dielectric breakdown of the silicon dioxide layer used as electrical insulation. Mugele et al. [22,23] utilized silane monolayers bound to oxide surfaces without focussing on their use. In both cases, breakdown may have been avoided by using relatively thick insulating layers: a 50-μm thick glass cover slip and a 1-μm thick thermally grown silicon dioxide layer. Silanes bind covalently to surface hydroxyl groups; and silicon dioxide is by far the most obvious surface to provide such conditions. It may be that the dearth of research into electrowetting on top of hydrophobic silanes is a result of the low resistance to electric breakdown exhibited by thin silicon dioxide films in EWOD [11,12]. In this paper, we have chosen to examine the performance of OTS because it is one of the most widely used and cheapest of the available silanes. Furthermore, we bind OTS monolayers to a silicon nitride surface and as a consequence avoid the use of a silicon dioxide insulating layer.

2. Methods

Figs. 1 and 2 show schematic drawings of the setup that was used to measure the electrical and surface wetting properties of a sessile droplet sitting on top of an electrode structure that is insulated with a thin-film dielectric coating. For each experiment, a 1-μL droplet was dispensed by means of a syringe pump in the area between two parallel electrodes. A direct current (DC) voltage was applied between these electrodes using a Keithley 6517 electrometer. The electrometer is capable of generating DC voltages of up to ±1 kV and of measuring leakage currents down to femtoamperes (fA). The electrometer measured the current flow and the resistance of the system as a function of applied voltage. The electrometer is controlled via the general purpose interface bus (GPIB), which also enables digital transfer of measurement data. The setup was placed in a metal Faraday cage in order to minimise electromagnetic disturbances. It took between 2 and 5 s to measure each of the data points in the experiments presented here.

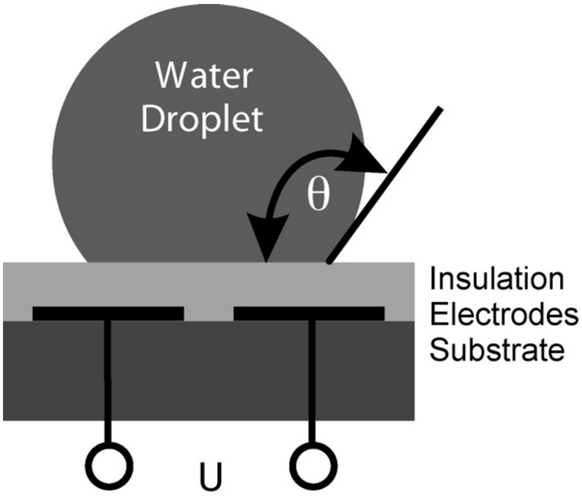

Fig. 1. Schematic drawing of the experimental setup. A sessile droplet is positioned between two parallel electrodes with various types of insulation as described in the text. The contact angle is measured by means of video microscopy. This figure is not to scale.

Parallel electrodes were used for the electric breakdown experiments. The droplet was placed between the electrodes and electrical measurements were performed between these electrodes as described in the above paragraph. The following coatings were applied: (a) a platinum electrode coated with 400 nm of Si_3N_4 and with a further 100 nm of Teflon-like fluoropolymer, (b) a copper electrode coated with 1.2 μm Parylene C and with a further 200 nm of Teflon-like fluoropolymer, and (c) a gold electrode with 800 nm of SiO_2 and 200 nm of Teflon-like fluoropolymer. Parylene C was deposited by vapour phase deposition. Parylene C and silicon nitride samples were hydrophobically coated as follows: the silicon nitride sample was coated with a further 100 nm of PECVD-coated Teflon-like fluoropolymer by GeSiM mbH (Großerkmannsdorf, Germany) and the Parylene C sample was coated by the Leibniz Institute for Plasma Science and Technology (INP) e.V. (Greifswald, Germany) with a 200 nm of PECVD-coated Teflon-like fluoropolymer layer. Although the Teflon-like fluoropolymer from INP e.V. displays a slightly higher contact angle, the layers on the samples from both suppliers showed static contact angles of greater than 90°.

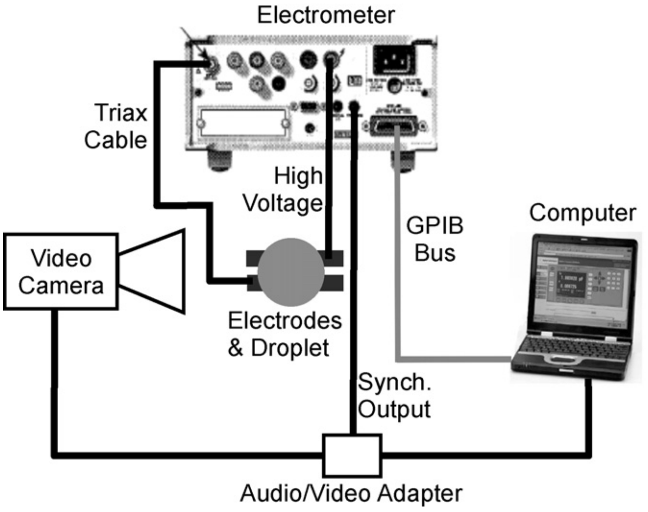

Fig. 2. A voltage is applied by means of a Keithley 6517 electrometer, which measures the current and resistance characteristics of the system as a function of this applied voltage.