Electrowetting-on-liquid-dielectric (EWOLD) enables droplet manipulation with a few volts

Ken Yamamoto¹, ², +, Shimpei Takagi³, *, Yoshiyasu Ichikawa², ³, Masahiro Motosuke², ³

¹ Department of Earth and Space Science, Graduate School of Science, Osaka University, 1-1 Machikaneyama, Toyonaka, Osaka 560-0043, Japan
² Water Frontier Research Center (WaTUS), Tokyo University of Science, 6-3-1 Niijuku, Katsushika-ku, Tokyo 125-8585, Japan
³ Department of Mechanical Engineering, Tokyo University of Science, 6-3-1 Niijuku, Katsushika-ku, Tokyo 125-8585, Japan

+ These authors contributed equally.
* Corresponding author.
E-mail: yam@ess.sci.osaka-u.ac.jp

Abstract
Electrowetting has a potential to realize stand-alone point-of-care (POC) devices. Here we report a novel droplet manipulation method “electrowetting-on-liquid-dielectric (EWOLD)”. The method reduces the droplet manipulation voltage to a level that the voltage can be supplied by four AA batteries. It is achieved by removing the solid dielectric layer and replacing it with a liquid dielectric film. The mechanism of the low-voltage actuation is explained by coupling a scaling analysis and droplet velocity measurements. Consequently, we report that a 5-μL droplet can be manipulated by 5 V.
Electrowetting (EW) is a phenomenon in which an externally applied voltage changes the wettability of a solid surface. It is exploited for some tunable lenses, prisms, and optical switches [1–6], and its application was expanded in 2000s to biochemical applications called digital microfluidics [1, 7–10], as electrowetting-on-dielectric (EWOD) was developed. This is because EWOD, which inserts a dielectric layer in-between droplets and electrodes, can manipulate droplets outside microfluidic channels and in real time [11]. This feature, as well as its high connectivity to other technologies used in biochemical processes, opened a possibility of the droplet-based biochemical analysis on demand.

As our society has been progressing to specialize medical care and tests for individual (point-of-care, POC) [10], programmable nature of EWOD droplet manipulation has much advantage. POC biosensors are also expected to be a powerful tool under pandemic situations because a possible infected individual can test by oneself while avoiding a contact with others. On the other hand, there are many requirements (e.g., simplicity, cost, rapidity, portability, and sensitivity) for the POC devices and they often conflict each other. For instance, nucleic acid tests are highly sensitive but needs multiple steps, thereby realizing it with paper-based (simple and cost-effective) devices is challenging [12]. From this viewpoint, EWOD devices are suitable for automatically processing the multiple steps whereas there are rooms for improvement in such as fabrication cost [13], reliability [14], portability [10], and high-voltage requirement (typically 10² V) [10, 15, 16]. Among the abovementioned points-to-be-improved, driving voltage is one of the crucial points because access to a power source is frequently limited for the POC devices [12, 17]. Lowering the driving voltage will be also important for improving the device reliability because it is beyond the thorough understanding of the droplet manipulation mechanism.

There are two ways to lower the driving voltage of EWOD: increasing the driving force or decreasing the resistance. Employing thin and high-permittivity dielectric layer [18] is a solution for the first way and employing a hydrophobic coating [19], infusing an oil layer [6, 20], and reducing the droplet size [21] are solutions for the other. Moon et al. [18] employed a very thin (700 Å) and high-dielectric-constant (~180) layer and confirmed a transport of a droplet (460 nL) with 15 V. Yi et al. [19] achieved to lower the manipulation voltage to 65 V with a thin dielectric layer coupled with a hydrophobic coating. To further decrease the applied voltage, Bormashenko et al. [20] applied a low-drag surface similar to slippery liquid-infused porous surfaces (SLIPS) [22–24]. They installed a honeycomb-shaped polycarbonate layer on electrodes as a dielectric layer
and added silicone oil into the honeycomb structure to reduce the adhesive force between droplets and the dielectric layer. This setup (named as electrowetting-on-liquid-infused-film, EWOLF [6]) can lower the applied voltage by eliminating the so-called pinning effect in principle as the oil layer separates the droplet from contacting the substrate [25, 26]. As a result, they achieved the minimum voltage of 35 V to drive an 8-µL droplet. Lin et al. [21] used small droplets of 300 pL in an oil-filled microchannel (crossec section of 150 × 20 µm²) and achieved a driving voltage of 7.2 V.

Although these studies demonstrated that factors listed above effectively reduce the driving voltage, contributions of each factor are still unclear due to the different setup and substrate for different studies and a lack of understanding the fluid motion. Therefore, we investigate the resistance-reduction effects by comparing droplet motions with the same setups on comparable substrates (dry, oil-infused, etc.). Moreover, we propose a novel resistance-reduced EWOD concept “electrowetting-on-liquid-dielectric (EWOLD)” that removes the solid dielectric layer and replaces it with a liquid dielectric layer (Fig. 1). The resistance-reduction mechanism is explained by a physical model based on the lubrication theory.

To confirm the concept of EWOLD, we applied DC voltage to a droplet (10 µL) deposited on a silicone-oil-infused ITO electrode (Fig. 1b). To ensure that the liquid layer stably exists in between the droplet and the electrodes, we carefully chose a set of materials for the components to contain a relationship of $n_e > n_l > n_d$, where $n$ denotes the refractive index and subscripts e, l, and d denote the electrode, the liquid layer, and the droplet, respectively. Under this condition, a stable film with a thickness of ~100 nm is formed owing to the van der Waals force [27]. We chose gold or ITO (indium tin oxide) for the electrodes (ITO for the bottom-view observation), silicone oil for the liquid layer, and pure water for the droplet. Bottom-view observations confirmed that a thin silicone oil layer is formed in between the droplet and the electrode. Moreover, we confirmed through an interferometric observation that the droplet edge expands by applying the voltage as low as 0.1 V (see Supplementary Information for detail experimental setup and observation). From these results we concluded that the concept is valid. However, we also found that the film thickness is variable and sometimes the electric breakdown occurs for > 1V voltage probably because of the too thin dielectric layer.

A solution to avoid the electric breakdown is adding sparse micropillars which support the droplet against gravity and maintain a thick dielectric layer (Fig. 1c and Supplementary Fig. S1). We fabricated the micropillars (14 µm × 14 µm × 0.75 µm) by the standard soft lithography method and coated them by solution
of Glaco Mirror Coat (Soft 99). (see Supplementary Information for more detailed fabrication method). Note that we performed two different coating methods to prepare fully-coated pillars and top-surface-coated pillars. Respective pillars were obtained by coating the substrate after and before developing the pillars. Although the former method implies that the electrode surface is also coated, comparative study confirmed that devices fabricated by the respective methods show equivalent performance on droplet motions described in the following part. It is probably because the coating thickness (~50 nm) is negligible in terms of the lubrication (liquid layer thickness equals pillar height of 0.75 μm) and because the insulating characteristics is comparable to silicone oil. However, as we discuss later, the performance changes if a non-negligible solid dielectric layer exists.

In the following part we estimate the driving force and the resistance force of a moving droplet. In general, a driving force exerted on a moving droplet is described by the surface tension and a difference in the receding and advancing contact angles \( \theta_r - \theta_a \) as [28]

\[
F_{\text{drive}} = \gamma_w L k (\cos \theta_a - \cos \theta_t)
\]  

(1)

where \( \gamma_w, L, \) and \( k \) denote water surface tension, contact width of the droplet, and a numerical factor determined by the droplet shape, respectively. On the other hand, under the EWOD condition, the dielectrically induced change in the contact angle is expressed by Eq. (2) [1, 11, 29]

\[
\cos \theta_E = \cos \theta_0 + \frac{\varepsilon_r \varepsilon_0}{2d \gamma_w} E^2
\]  

(2)

where \( \theta_0 \) and \( \theta_E \) are contact angles before and after applying voltage, \( \varepsilon_r, \varepsilon_0, d, \) and \( E \) denote relative permittivity of the dielectric layer, the permittivity of vacuum, thickness of the dielectric layer, and the applied voltage, respectively. With assumptions that \( \theta_E \sim \theta_a \) and the change in \( \theta_t \) is negligible at the initial stage of migration (i.e., \( \theta_t \sim \theta_0 \)), we can derive a correlation of \( F_{\text{drive}} \) (force to drive a droplet) and \( E \) by combining Eqs. (1) and (2):
where $R$ denotes droplet radius and $Lk \sim 2R$ is assumed.

We performed lateral-view observations to validate the above model. A conventional EWOD substrate, which has a thin solid dielectric layer (1-μm thick CYTOP®), was prepared for the validation and a 10-μL water droplet was manipulated with a minimum applied voltage that can drive the droplet (Supplementary Fig. S5). We measured the contact angles to obtain $F_{\text{drive}}$ from Eq. (1) under two different conditions: the EWOD substrate with and without silicone oil (20 mm$^2$ s$^{-1}$) infusion. With these conditions (and an assumption that $d$ equals the pillar height) we obtain two different sets of $F_{\text{drive}}$ and minimum applied voltages and we would obtain the following relationship if Eq. (3) is valid.

\[
E_{\text{oil}} \approx \left( \frac{F_{\text{oil}}}{F_{\text{dry}}} \right)^{1/2} E_{\text{dry}}
\]

where $E$ and $F$ denote the minimum applied voltage and the driving force and subscripts oil and dry denote the conditions with and without oil, respectively. Consequently, the droplet started moving when the difference in the receding and advancing contact angles becomes $\theta_r - \theta_a = 23.5^\circ$ at $E_{\text{dry}} = 75$ V in the case without oil, whereas it was $1.4^\circ$ at $E_{\text{oil}} = 17$ V in the case with oil, showing a similar tendency (low hysteresis resulting from the effective pinning elimination) to the case on slippery liquid-like surfaces [30]. Taking $k = 1$, we obtained $F_{\text{dry}} = 35 \mu$N and $F_{\text{oil}} = 2.1 \mu$N. Substitution of these values into Eq. (4) predicts $E_{\text{oil}} \sim 18$ V, which is excellently close to the measured value of 17 V. Therefore, we concluded that Eq. (3) is valid.

Subsequently, we estimate the drag force that should be in balance with $F_{\text{drive}}$. Under the lubricated condition, the drag is considered to cause in the thin liquid film and/or in oil meniscus [31]. In the thin oil film of thickness $h$, the viscous dissipation is proportional to oil viscosity $\eta_o$ and velocity profile $U_i / h$, where $U_i$ is the velocity at the oil–droplet interface. Taking into account the effective area \( \sim R^2 \) and the finding that velocity inside the droplet is almost the same in the $z$-direction as the droplet velocity $U$, the friction force in the film $F_{\text{film}}$ is described as

\[
F_{\text{film}} \approx \frac{R \epsilon \eta_o}{d} U^2
\]
In the oil meniscus, dynamic characteristics should be considered to estimate the viscous dissipation. The resulting friction force in the meniscus $F_{\text{meniscus}}$ is described as [31]

$$F_{\text{meniscus}} \approx \gamma_o^{1/3} \phi R (\beta \eta_0 U)^{2/3} \quad (6)$$

where $\phi$ and $\beta$ denote the solid fraction and a numerical factor (detail of the model is given in Supplementary Information).

We now correlate the above friction forces and the driving force to derive relationships between the droplet velocity and applied voltage. If the dissipation in the liquid film is dominant, the driving force $F_{\text{drive}}$ [Eq. (3)] should be equal to the friction force $F_{\text{film}}$ derived by Eq. (5). Therefore, the droplet velocity $U$ is given by

$$U \approx \frac{h \varepsilon \varepsilon_0}{d \eta \gamma_0} E^2 \quad (7)$$

On the other hand, if the dissipation in the meniscus is dominant, $U$ is given from Eqs. (3) and (6) as

$$U \approx \frac{1}{\beta \eta_0 \gamma_0^{1/2}} \left( \frac{\varepsilon \varepsilon_0}{d \phi} \right)^{3/2} E^3 \quad (8)$$

These relationships suggest that the droplet velocity is proportional to the square or cubic of the applied voltage depending on the dominant region of the dissipation. Moreover, both Eqs. (7) and (8) predict that the velocity increases as $d$ decreases. By assuming that the permittivity of the solid dielectric layer (of thickness $d_{\text{solid}}$) and silicone oil are the same, we can consider $d = d_{\text{solid}} + h$, and maximum velocity is obtained at the limit of $d_{\text{solid}} \to 0$, e.g., EWOLD state. Note that the correlations are also valid for droplets surrounded by oil if the oil film stably exists. Therefore, it is implied that the resistance can be decreased further when the oil film is formed and the droplet motion obeys Eq. (7).
To understand the droplet migration mechanism, we measured the velocity of a 10-μL droplet manipulated in the EWOLD state with various applied voltage $E$ and oil viscosity $\eta_o$ under the condition of $\phi = 25.0\%$ and $d = h = 0.75 \text{ μm}$. Note that the oil film thickness $h$ was controlled by the height of the micropillars. Figure 2 shows the relationship between the droplet velocity $U$ (maximum velocity of the center of the mass) and $E$. The result shows that $U$ is proportional to $E^2$, which indicates the dissipation in the oil film is dominant. Equation (7) also predicts that $U$ is proportional to $\eta_o^{-1}$. However, although the decreasing trend of $U$ with the increase in $\eta_o$ is observed, the data does not collapse on a single curve of $U\eta_o \sim E^2$ for small $\eta_o$ (Fig. 3a). It may be because of a small viscosity ratio of oil and water $\eta_o / \eta_w$. Figure 3b shows that extra driving force is required for small $\eta_o / \eta_w$. In that case, there are two factors that could induce additional energy loss and change the $U\eta_o \sim E^2$ relationship: change of a balance of dissipation between in the oil film and droplet and/or a relative increase in the friction at the pillar top.

The viscous resistance by the shear inside the droplet is estimated as $F_{\text{drop}} \sim \eta_w R^2 \frac{dU}{dz}$, where $\frac{dU}{dz}$ is the velocity gradient in the droplet. Because the maximum $F_{\text{drop}}$ can be estimated by taking $\frac{dU}{dz}$ from a velocity profile in the case of EWOD (because lubrication suppresses the inner flow, see Supplementary Figs. S7 and S8), the effect of the inner flow can be evaluated by comparing $F_{\text{drop}}$ for EWOD and $F_{\text{film}}$ for low oil viscosity. As a result, $F_{\text{drop}}$ for EWOD (at 75 V) is calculated as $10^{-2} \text{ μN}$ and $F_{\text{film}}$ for $\eta_o = 2 \text{ mm s}^{-1}$ is calculated as $10^{-1} \text{ μN}$ with $U = 0.1 \text{ mm s}^{-1}$, which is one order smaller than the actual migration velocity. Although it is difficult to estimate the frictional loss, the above result implies that the friction affects the migration for low $\eta_o$ and the energy loss inside the droplet is negligible.

As $U$ obeys Eq. (7), we can expect that $U$ increases with $R^{-1}$. Figure 4a shows a dependence of $U$ on $R$ for $\eta_o = 20 \text{ mm}^2 \text{ s}^{-1}$ and $E = 75 \text{ V}$. The diagram shows an increasing trend of $U$ with decreasing $R$. Moreover, Fig. 4b shows that $UR$ for different $R$ take almost a constant value. It implies that smaller droplets have higher mobility as we expected.

In contrast to Eq. (8) and instinct, Eq. (7) predicts that $U$ does not depend on neither $h$ nor $\phi$ for EWOLD (because $h/d = 1$ for EWOLD). Figure 5a and 5b show $U$ as a function of $E$ for different $h$ and $\phi$, respectively. The result shows a slight increase in $U$ with $h$, but the difference is not significant, at least in this range of $h$. On the other hand, the effect of $\phi$ is non-negligible (Fig. 5b). In these three cases, $U$ takes maximum with the middle of the fraction $\phi = 16.0\%$. This could be explained as follows: the frictional loss and the viscous
dissipation becomes smaller as $\phi$ decreases, but it turns back at a particular point, under which drag increases as the bottom of the droplet deforms, and the pillars act as bumps [32].

Finally, we performed the droplet manipulation with applied voltage as low as possible. With a condition of $\eta_o = 2 \text{ mm}^2 \text{s}^{-1}$, $\phi = 16.0\%$, $h = 0.75 \mu\text{m}$, we confirmed that a 10-$\mu$L droplet migrates with $E = 6$ V (Supplementary Movie 1). Moreover, as we can expect from Eq. (7) that smaller droplet can migrate with smaller voltage, we drove a 5-$\mu$L droplet. Consequently, we confirmed that it migrates with $E = 5$ V.

We developed a novel electrowetting-derived droplet manipulation method: electrowetting-on-liquid-dielectric (EWOLD). By removing a solid dielectric layer and replacing it with a liquid dielectric layer, deformation of a droplet with 0.1 V applied voltage was achieved. Moreover, we also performed the droplet migration. The minimum applied voltage that can manipulate the droplet was 93 % reduced in comparison with the conventional electrowetting-on-dielectric (EWOD). This reduction stems primarily from the elimination of the pinning effect and addition of the lubrication effect and secondary from the removal of the solid dielectric layer. We developed a model to predict the velocity of the droplet migration and found that the velocity increases with the square of the applied voltage and it decreases with an increase in the liquid-dielectric viscosity. Finally, manipulation of a 5-$\mu$L droplet by 5 V was performed: it opens a door for manipulations in various ways; for instance, coupling with photoelectrowetting [33–36] may lead droplet handling on a smartphone, novel adjustable lenses, and portable lab-on-a-chip devices.

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Figures

**Fig. 1.** (a) Photograph of a migrating droplet by EWOLD. Schematics of the EWOLD substrates (b) for the droplet shape deformation (thin silicone oil film exists in between the droplet and electrode) and (c) for the droplet migration.

**Fig. 2.** Relationship between the droplet velocity $U$ and the applied voltage $E$ with different oil viscosity $\eta_o$. 

![Diagram showing the relationship between droplet velocity and applied voltage](image-url)
Fig. 3. Effects of the oil viscosity in the relationship between $U$ and $E$. (a) $U \eta_o$ as a function of $E$. (b) Balance of the resistance force and the driving force $F_{\text{film}} / F_{\text{drive}}$ for various oil / water viscosity ratio $\eta_o / \eta_w$.

Fig. 4. Droplet velocity for different droplet radius $R$. (a) $U$ vs. $R$. (b) $UR$ vs. $R$.

Fig. 5. Dependence of the droplet velocity on substrate geometries. (a) Effects of the thickness of the dielectric liquid $h$. (b) Effects of the solid fraction $\varphi$. 
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