Electromechanical Analysis of AC Electrowetting and Dielectric Breakdown of an Insulating Layer

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Abstract

The electrowetting shows some distinctive features when AC electric field is applied instead of DC field. They include the delay of dielectric breakdown of the insulating layer, the delay of contact-angle saturation, and the generation of tiny droplets near the three-phase contact line (TCL). In order to find out the causes of such phenomena, we numerically analyze the AC electric field around a droplet placed on an insulator-covered electrode. The time-averaged effective electrical wetting tension, which is a function of AC frequency, is computed by integrating the Maxwell stress. The computed wetting tension is compared with the experimental result converted from the separately obtained contact-angle data. There is a good agreement between the two results. Interestingly, the numerical results show that the electric-field strength is decreased remarkably in the insulating layer near the TCL as the AC frequency is increased. This decrease may explain the delay of the dielectric breakdown of the insulating layer in the AC case, which is known to be closely related with the contact-angle saturation phenomenon.
1 Introduction

When an electric field is applied to a droplet sitting on an insulator-covered electrode, the contact angle of the droplet changes due to the electrical force acting on the edge region of the droplet (Kang et al. 2003; Kang 2002). This phenomenon is called the electrowetting, which is also frequently called the EWOD (electrowetting on dielectrics). Electrowetting is a versatile tool to handle tiny liquid droplets and menisci, and there are many potential applications including the liquid lens (Berge and Peseux 2000; Kuiper and Hendriks 2004), electronic paper (Hayes and Feenstra 2003), and microfluidic devices (Cho et al. 2003; Mugele et al. 2006). The electrical wetting tension which induces the change of contact angle originates from the force acting on the electrical charge induced at the droplet surface (Kang 2002). The charge density is proportional to the intensity of local electric field, so that the effective electrical wetting tension is proportional to square of applied voltage and acts always outward from the droplet. Subsequently, the electrowetting phenomenon persists even when an AC signal is applied.

Interestingly, however, if an AC signal is applied instead of DC signal, it shows several distinctive features as follows. First, the occurrence of the contact-angle saturation, which is still one of the most puzzling phenomena in electrowetting, is delayed in the AC case. That is, the contact-angle saturation occurs at a smaller contact angle and at a greater root-mean-square (RMS) voltage (Quilliet and Berge 2001; Blake et al. 2000; Quinn et al. 2005). Second, the edge instability takes place, i.e., tiny satellite droplets are disintegrated from the mother droplet (Quilliet and Berge 2001; Vallet et al. 1996; Vallet et al. 1999). The edge instability is observed only in the AC case, and especially for a very low electrolyte concentration (Quilliet and Berge 2001; Mugele and Baret 2005). Third, the AC case has a less contact-angle hysteresis; and that is one of the reasons why the AC field is preferred in some applications (Berge and Peseux 2000). Fourth, ion-adsorption at the liquid-substrate interface may be decreased (Quilliet and Berge 2001; Kumar et al. 2006). In addition, two of present authors recently reported the two new interesting features of AC electrowetting. One is that the surface of a droplet oscillates in considerable amplitude even up to a
few kilo-Hertz; the other is that two distinctive hydrodynamic flows are generated inside a droplet in AC case (Ko et al. 2007). Those phenomena observed in the AC electrowetting are yet to be fully explained.

The general electromechanical characteristics of electrowetting can be, however, understood roughly by representing the droplet system by an equivalent circuit such as shown in Fig. 1. The droplet can be simply represented by a combination of a resistor $R$ and a capacitor $C$, without considering the Faradaic current and electrical double layer. The thin insulating layer which prevents the passage of the Ohmic current is represented by a capacitor $C_i$. The Ohmic current is directly proportional to voltage drop across the droplet $V_w$; and the displacement current is proportional to the rate of change of voltage $\partial V_w /\partial t$. At very low frequency the Ohmic current dominates over the displacement current, and the droplet behaves like an Ohmic conductor (Fig. 1b). At very high frequency, $\partial V_w /\partial t$ is so great that the displacement current dominates; and the droplet acts more like a perfect dielectric (Fig. 1c). The crossover frequency between the two currents is estimated $\sigma[\varepsilon_w + \varepsilon_d (R / d)]^{-1}$, in which $\sigma$ and $\varepsilon_w$ represents respectively the electrical permittivity and conductivity of water, $\varepsilon_d$ is the electrical permittivity of insulating layer, $R$ is the characteristic size of the droplet, $d$ is the thickness of insulating layer (Jones 2002; Jones et al. 2003; Jones et al. 2004). In the frequency range of $\sigma[\varepsilon_w + \varepsilon_d (R / d)]^{-1}$, the contact-angle change may show a significant frequency dependence as reported by previous investigators (Mugele and Baret 2005; Kumar et al. 2006; Jones 2002; Jones et al. 2003; Jones et al. 2004).

Jones and his co-workers successfully introduced the lumped-parameter method based on an equivalent circuit such as shown in Fig. 1 to analyze the frequency dependence of height-of-rise in a slit-like vertical channel for the AC case (Jones 2002; Jones et al. 2003; Jones et al. 2004). For a sessile droplet, Mugele and his co-workers analyzed the frequency dependence of contact angle of a droplet (Mugele and Baret 2005; Kumar et al. 2006). Their analyses were based on the lumped-parameter method, and partially considered the change of equivalent resistances and capacitances resulting from the change of contact angle (Kumar et al. 2006). The lumped-parameter method is
very useful for qualitative (or sometimes quantitative) understanding of such frequency dependence of contact angle and height-of-rise. Basically, however, the use of the lumped-parameter method is rather restricted to the case in which the equivalent capacitances and resistances of a system are independent or nearly independent of frequency, such as in the slit-like channel of Jones and co-workers. The calculation of the equivalent capacitance and resistance of a droplet system requires a numerical analysis for the electric field (Kumar et al. 2006).

We pursue reliable explanations for those intriguing phenomena mentioned above, and it includes an accurate prediction of such frequency dependence of contact angle. For this, the electric field around the droplet should be known precisely, especially at the edge region of the droplet where the hint to uncover the mystery is supposedly present. In this work, we analyzed the AC electric field around a droplet numerically and validated the results experimentally. Several groups reported the numerical results for the DC case in electrowetting problems (Zeng and Korsmeyer 2004). As far as we know, however, the full numerical analysis on the AC electrowetting has not been performed. The effective electrical wetting tension is computed to investigate the frequency response of the contact-angle change. The numerical results show a good agreement with the experimental data. Based on the numerical results on the electric-field strength, we attempt to explain the cause of delay of the contact-angle saturation, in conjunction with the delay of the dielectric breakdown of insulating layer.

2 Formulations

We analyze the electric field around a sessile droplet placed on an insulator-covered electrode (see Fig. 2). The shape of the droplet is assumed to be perfectly axisymmetric. The domain is composed of three regions, i.e., the droplet, the surrounding air, and the dielectric layer. The effect of electrical double layer is not considered. The material properties are assumed to be constant and uniform in each region. We introduce an (r,z) cylindrical coordinate system, which has its origin at the center of
the base circle of the droplet. The unit vectors in r and z directions are denoted by \( \mathbf{e}_r \) and \( \mathbf{e}_z \), respectively. In Fig. 2, \( \mathbf{n} \) represents the outward unit normal vector.

Under the assumption of quasi-electrostatic field, the electric field is separable from the magnetic field and the Maxwell equation can be simplified as follows (Haus and Melcher 1989):

\[
\nabla \times \mathbf{E} = 0 , \\
\n\nabla \cdot \mathbf{E} = \rho_f .
\]

Here, \( \mathbf{E} \) represents the electric field, \( \varepsilon = \varepsilon_r \varepsilon_0 \) is the electrical permittivity, \( \varepsilon_0 \) is the electrical permittivity of free space, \( \varepsilon_r \) is the dielectric constant, and \( \rho_f \) is the free-charge density. The conservation of free charge is written as

\[
\frac{\partial \rho_f}{\partial t} + \nabla \cdot \mathbf{J} = 0 .
\]

We assume that all the materials satisfy the Ohmic conduction law, so that the current density \( \mathbf{J} \) is related with the electric field as

\[
\mathbf{J} = \sigma \mathbf{E} ,
\]

where \( \sigma \) is the electrical conductivity. Substituting \( \rho_f \) and \( \mathbf{J} \) in Eqs. (2) and (4), respectively, to Eq. (3), we obtain the following equation:

\[
\nabla \cdot \left( \varepsilon \frac{\partial \mathbf{E}}{\partial t} + \sigma \mathbf{E} \right) = 0 .
\]

The first and second terms in the parentheses corresponds respectively to the displacement current and the Ohmic current.

At the interface of each region, the following jump condition should be satisfied:

\[
\| \mathbf{e} \mathbf{E} \| \cdot \mathbf{n} = q ,
\]

\[
\| \sigma \mathbf{E} \| \cdot \mathbf{n} = - \frac{\partial q}{\partial t} .
\]
where \( \| g \| \) denotes the jump, “outside-inside” of \( g \) across the interface, and \( q \) is the surface charge density. Equations (6) and (7) are just the Gauss law and the current conservation condition applied at the interface, respectively. Those two conditions in Eqs. (6) and (7) can be reduced to the following single relation by inserting \( q \) in Eq. (6) into Eq. (7):

\[
\nabla \cdot (\varepsilon \frac{\partial \mathbf{E}}{\partial t} + \sigma \mathbf{E}) \cdot \mathbf{n} = 0. \tag{8}
\]

As a matter of fact, the integral form of Eq. (5) becomes

\[
\int \left( \varepsilon \frac{\partial \mathbf{E}}{\partial t} + \sigma \mathbf{E} \right) \cdot \mathbf{n} dA = 0, \tag{9}
\]

and Eq. (8) can be directly derived from this integral condition.

Since \( \nabla \times \mathbf{E} = 0 \), there exists an electric potential \( \phi \) which satisfies \( \mathbf{E} = -\nabla \phi \). When we apply a periodic signal to the system, the potential \( \phi \) can be represented by \( \phi = \text{Re}\{\phi e^{j\omega t}\} \). Here, \( \phi = \phi_r + j\phi_i \) is the time-independent complex electric potential, or the phasor of the electric potential, \( \omega = 2\pi f \) is the angular frequency, and \( j = \sqrt{-1} \). Accordingly, the electric field is represented by \( \mathbf{E} = \text{Re}\{\tilde{\mathbf{E}} e^{j\omega t}\} \), and is related with electric potential as

\[
\mathbf{E} = -\nabla \phi = \text{Re}\{-\nabla \phi e^{j\omega t}\} = -\nabla \phi_r + \nabla \phi_i \sin \omega t. \tag{10}
\]

The complex electric field \( \tilde{\mathbf{E}} \) is related with the complex electric potential as \( \tilde{\mathbf{E}} = -\nabla \phi \).

Substituting this complex electric field to Eq. (5), we can obtain the following equation for the complex potential:

\[
\nabla \cdot (\tilde{\sigma} \nabla \phi) = 0, \tag{11}
\]

where \( \tilde{\sigma} = \sigma + j\omega \varepsilon \) is called the complex conductivity. Equation (9) is just an alternative from of Eq. (5) and can be also represented as \( \nabla \cdot \mathbf{J} = 0 \), where \( \mathbf{J} = (\sigma + j\omega \varepsilon)\tilde{\mathbf{E}} \) represents the complex current density. All the material properties are assumed to be homogeneous in each region, and therefore, the above equation is further simplified as

\[
\nabla^2 \phi = \nabla^2 (\phi_r + j\phi_i) = 0 \tag{12}
\]

in each region. Similarly, the interface condition in Eq. (8) becomes, for a sinusoidal excitation,
3 Numerical analysis

Equation (10) is solved numerically by applying the current continuity condition of Eq. (11) at
interfaces. We used a commercial package of COMSOL Multiphysics® ver. 3.3 which is based on
the finite element method. Figure 3 shows the computational domain. The electric field is assumed
axially symmetric so that only half of the domain is considered in the analysis. The hydrophobic
layer existing atop a dielectric layer is neglected, since its thickness (about 100 nm) is much thinner
than that of the insulating layer (d = 5 µm). The radius of the needle electrode is 40 µm, and its tip
position is fixed at (0, 640 µm).

The current continuity condition of Eq. (11) is applied at droplet-air (S1), droplet-substrate
(S2), air-substrate (S3) interfaces (see Fig. 3). Far from the droplet, a hemispherical surface is
chosen (S4), whose radius is about 6a, where a is the base radius of the droplet. The volume of
droplet is fixed at 5 µl. The base radius is 1.1 mm when contact angle is 117°, which are consistent
with the experimental condition. On the hemispherical surface (S4), the insulation condition of
\[ \mathbf{n} \cdot \mathbf{\tilde{J}} = 0 \]
is applied. The constant-potential conditions are applied at the needle (S5) and substrate
(S6) electrodes. At the center of the droplet (S7), the axial-symmetry condition was applied.

The value of conductivity of air and insulating layer has little effect on the electric field
since the conductivity is so small that the displacement current \( \varepsilon \partial \mathbf{E} / \partial t \) dominates over the Ohmic
current \( \sigma \mathbf{E} \) in this region. Therefore, we let the conductivity of air and insulating layer zero. The
dielectric constants of water, air, and insulating layer are chosen as \( \varepsilon_{w} = 78.5 \), \( \varepsilon_{ra} = 1 \), and
\( \varepsilon_{rd} = 2.7 \). The conductivity of water is measured to be \( \sigma_{w} = 4.8 \times 10^{-4} \) S/m.

For a direct comparison of numerical results with the experimental data, the dimensions of
the droplet and the dielectric layer are chosen to be the same as those of the experimental condition.
Moreover, we extracted the shapes of the droplet from experimentally obtained images, and then the simulation is carried out for those geometries to consider the change of drop shape. Total number of mesh points is about 40,000 ~ 50,000 and about 60% of mesh points was concentrated near the TCL to minimize the error caused by the sharp-edge effect.

The change of contact angle in electrowetting is induced by the effective electrical wetting tension \( w_{el} \) which is the integration of electrical stress (i.e., the Maxwell stress) acting on the droplet surface (Kang et al. 2003; Kang 2002). Both the electric field and the effective electrical wetting tension vary sinusoidally with time. The instantaneous effective electrical wetting tension is

\[
w_{el} = \frac{1}{2\pi a} \int_{S} \|T \cdot n\| \cdot e_r \, ds ,
\]

where \( e_r \) is the unit vector in the positive \( r \)-direction, and as mentioned, \( \| T \cdot n \| \) denotes the jump of \( T \cdot n \) across the drop surface (air side – water side). The Maxwell stress tensor \( T \) is defined as

\[
T = \varepsilon \varepsilon_0 E E - \frac{1}{2} \varepsilon (E \cdot E) I ,
\]

where \( I \) is the second-order isotropic tensor. The time-averaged effective electrical wetting tension is obtained by integrating the time-averaged Maxwell stress which becomes (see Appendix A):

\[
\langle (T \cdot n) \cdot e_i \rangle = \frac{1}{2} \text{Re} \left\{ \varepsilon E^* (\vec{E}_r n_r + \vec{E}_z n_z) - \frac{1}{2} \varepsilon (\vec{E}_r E^*_r + \vec{E}_z E^*_z) n_r \right\} ,
\]

where \( \vec{E} = \vec{E}_r e_r + \vec{E}_z e_z \), \( n = n_r e_r + n_z e_z \), and the superscript \('\) represents the complex conjugate. Consequently, using Eqs. (12) and (14), time-averaged effective electrical wetting tension \( (W_{el} = \langle w_{el} \rangle) \) is obtained. The root-mean-square (RMS) potential \( \phi_{rms} \) and the RMS electric-field strength \( E_{rms} \) is determined as follows (see Appendix A):

\[
\phi_{rms} = \sqrt{\langle \phi^2 \rangle} = \sqrt{\phi_r^2 + \phi_z^2} / \sqrt{2} ,
\]

\[
E_{rms} = \sqrt{\langle E \cdot E \rangle} = \sqrt{E_{r,rms}^2 + E_{z,rms}^2} .
\]

Considering the balance of forces at the TCL, the change of contact angle \( (\theta) \) is related with the effective electrical wetting tension as \( \cos \theta = \cos \theta_r + W_{el} / \gamma \), where \( \theta_r \) is the Young’s contact
angle and $\gamma$ is the interfacial tension at droplet-air interface. In DC case, this equation becomes the Lippmann-Young equation since $W_{el} = \varepsilon V^2 / 2d$ (Kang et al. 2003; Kang 2002). In turn, the effective electrical wetting tension can be obtained by using the experimentally-measured contact angle ($\theta_e$) as follows:

$$\frac{W_{el}}{\gamma} = \cos \theta_e - \cos \theta_f.$$ (15)

**4 Experiments**

An experiment is carried out to validate the numerical results. The contact angle of droplet is measured with changing the frequency of the applied AC electric field. The conventional experimental setup for the electrowetting experiment is employed, as illustrated in Fig. 4. A de-ionized water droplet of 5 $\mu$l in volume is placed on a planar insulator-covered electrode by a micropipette. The conductivity of the water droplet is measured by Orion 550A of Thermo Electron Co., and has a value of $4.8 \times 10^{-4}$ S/m. A stainless steel wire of 80 $\mu$m diameter is inserted to the droplet as an electrode. A smooth stainless steel plate is used as the counter electrode. On the electrode surface, the Parylene-C layer of 5 $\mu$m thickness is deposited at an insulating layer. On top of that, the Teflon® AF1600 layer of about 100 nm thinness was spin-coated to make the surface hydrophobic. The initial contact angle and the base radius of the droplet were about 117° and 1.1 mm respectively. The capillary length for water at room temperature is about 2.7 mm, while the radius of droplet is about 1.1 mm. Thus, the distortion of droplet shape from the spherical shape due to hydrostatic pressure is neglected, and the initial shape of the droplet can be regarded as a part of a perfect sphere.

A sinusoidal AC signal is generated by a function generator (33220A, Agilent), and then it is amplified by a voltage amplifier (A800, FLC). The amplified electrical signal is applied to the droplet. The frequency is varied from 1 kHz to 128 kHz. The droplet shape is observed by using a CCD camera (INFINITY2-2M, Luminera) which has 1,280 by 1,000 resolution, and the image is
recorded in a PC. All the experiments are finished within a few minutes to minimize any troublesome effect of evaporation. The shape of the droplet is extracted by using a Matlab® program and the macroscopic contact angle is obtained by a software based on the Matlab®.

5 Results and discussions

5.1 Frequency dependence of electric field

Figure 5 shows the distributions of electric potential for $f = 1$ kHz and $f = 128$ kHz, which represent respectively here the low and high frequency limits in Fig. 1. At a low frequency, the potential distribution is not very different from that of DC case, which is represented by Fig. 5a. That is, the potential is almost uniform and there is almost no electric field inside the droplet. On the contrary, at a high frequency, as represented by Fig. 5b, the electric field becomes similar to that of a perfectly dielectric material. As a result, the droplet behaves like a conductor at a low frequency and like a dielectric at a high frequency (Jones 2002; Jones et al. 2003; Jones et al. 2004). Figure 6 shows the potential distribution especially near the TCL for the two cases. There appear fewer equipotential lines for the case of $f = 128$ kHz compared to the case of $f = 1$ kHz, which means the electric-field strength is relatively weaker. It follows that the electrical stress which is proportional to square of electric field would be smaller for $f = 128$ kHz.

The contact angle is measured with changing the input frequency from 1 kHz to 128 kHz for four different RMS voltages of 57, 93, 113, and 143 V. Figure 7 demonstrates the frequency dependence of droplet shape for 143 V. Although it is not shown here, the contact angle shows a minor change below 1 kHz. As shown in Fig. 8a, the contact angle increases significantly with the frequency (Mugele and Baret 2005). As mentioned in the introduction, however, there appear capillary waves on the surface of a droplet, and the droplet surface shows an oscillating motion up to frequency range of a few kilo Hertz. That’s why the image taken at 1 kHz in Fig. 7 is more or less blurred at the boundary of the droplet. Although, the contact angle in Fig. 8a is a value at an instant,
it may not be very different from the time-averaged value since the camera’s exposure time is long enough to fully cover one period of oscillation.

Figure 8a shows the frequency and voltage dependence of contact angle. When the frequency is below 1 kHz, there is only minor change of contact angle. However, there is significant change of contact angle between 1 kHz and 20 kHz. Above 20 kHz, the contact angle is not very different from Young’s contact angle, as shown in Fig. 7. Figure 8b shows the effective electrical wetting tension. The effective electrical wetting tension decreases with increasing the input frequency. At a low frequency, the normalized effective electrical wetting tension $W_{el}/\gamma$ is only hardly different from that of DC value of $\varepsilon V^2/2\gamma d$. For example, when $V = 143$ V and $f = 1$ kHz, the normalized effective electrical wetting tension obtained numerically is 0.489. It is not very different from the effective electrical wetting tension of 0.471 which is converted from the measured contact angle by using Eq. (15).

Figure 9 compares the numerically-computed effective electrical wetting tension and the experimentally-obtained effective electrical wetting tension. We carry out the analysis considering the change of droplet shape due to contact-angle change. The computed effective electrical wetting tension shows a fairly good agreement with the experimental data. In fact, the contact angle in the present configuration is somehow dependent on the position of needle electrode and the conductivity of solution. There can be some uncertainty in measuring the position of electrode and the conductivity of solution. Such a good agreement shown in Fig. 9 should be interpreted that the results of present numerical analysis lie within the limit of uncertainty, rather than give perfectly accurate values for the wetting tension.

Figure 10 shows the electrical force (per unit area) acting on droplet surface in the radial direction (what we call the radial stress) for several frequencies. Actually, the effective electrical wetting tension is just an integration of the radial stress (see Eq. (12)). It is evident that the radial stress is much greater at a low frequency around the TCL, i.e., $z = 0$. The stress gradually decreases with an increase of frequency. Note that the stress distribution is only hardly changed from the DC case up to $f = 2$ kHz. As mentioned earlier, the conduction current dominates over the displacement.
current at a low frequency, so that the electric-field distribution is not very different from the DC case.

### 5.2 Delay of breakdown of dielectric layer

Previous experimental results showed that the contact-angle saturation is delayed when the AC voltage is used (Quilliet and Berge 2001; Blake et al. 2000). That is, the saturation occurs at a smaller contact angle and at a higher voltage than in DC case. The fundamental physics of the phenomenon has not been fully clarified yet. There exist several theories on the cause of the contact-angle saturation. Those theories are based on dielectric breakdown of dielectric layer (Papathanasiou and Boudouvis 2005; Seyrat and Hayes 2001), charge trapping (Verheijen and Prins 1999), thermodynamic interfacial instability (Peykov et al. 2000; Quinn et al. 2005), leaky-dielectric effect (Shapiro et al. 2003), air ionization (Vallet et al. 1999), and so on. The saturation could be even caused by different mechanisms depending on circumstances (Kedzierski and Berry 2006).

Here, we computed the electrical stress near the TCL, and investigate whether the delay of contact-angle saturation in AC case can be explained in terms of delay of dielectric breakdown. Figure 11 shows the RMS electric-field strength across the dielectric layer, directly below the TCL. It has a maximum value just below the TCL due to the sharp-edge effect, and generally decreases with increasing frequency. On the other hand, the electric field near the TCL is not well-defined due to the inherent corner singularity, and therefore, the electric field has an infinitely large value at the TCL. This singular behavior in fact does not appear in the numerical analysis due to discretization of domain and the subsequent numerical-averaging process. It is shown in Appendix B that the integration of the electrical stress on a small region including the singular point has a finite value. In this work, the small (surface) region covers the adjacent node points of the TCL, which ranges 10 to 15 nm in physical dimension (Papathanasiou and Boudouvis 2005).

The dielectric breakdown is certainly associated with the electrical stress acting on the layer. Since the electrical stress is proportional to $E_{\text{rms}}^2$, the dielectric breakdown may be delayed somehow.
when we increase the frequency. For $f = 1$ kHz, for example, the maximum electric-field strength which appears near the TCL is 289.4 MV/m. This value is about 80 % of the dielectric breakdown strength of the parylene-C of 350.6 MV/m (which is obtained from the Cookson Electronics, Specialty Coating Systems). For $f = 4$ kHz, however, the maximum electric-field strength is reduced to 243.3 MV/m which is about 70 % of the dielectric strength.

Since the effective electrical wetting tension decreases with increasing frequency, the greater voltage should be applied to keep the effective electrical wetting tension constant. We computed a necessary input voltage ($V_{req}$) necessary to keep the effective electrical wetting tension constant with increasing frequency. We also computed the RMS electric-field strength near the TCL ($E_{TCL}$) for each frequency. When the effective electrical wetting tension is constant, the droplet shape does not change. We chose the droplet shape obtained for $f = 1$ kHz. Figure 12 shows that the input voltage should be increased to maintain the effective electrical wetting tension constant with compensating the reduction due to the frequency effect. Interestingly, at a constant effective electrical wetting tension, the resultant $E_{TCL}$ decreases even though the applied voltage is increased. As shown in Fig. 12, such a tendency is maintained up to 500 V, although it is an unrealistic value in conventional electrowetting. Thus, it clearly confirms the frequency effect on the delay of the dielectric breakdown. Therefore, the high-frequency electric field induces a greater change of contact angle when the maximum electric-field strength at the TCL is the same. This may explain how in experiments we could increase the voltage than the DC case without any sign of dielectric breakdown and how we could induce greater change of contact angle in the AC case (Quilliet and Berge 2001).

6 Conclusions

We successfully implemented the AC conduction model to analyze the AC electrowetting phenomena. The numerical results show a fairly good agreement with the experimental results. It is shown numerically that the effective electrical wetting tension decreases with the increase in the
frequency, which is quantitatively consistent with the experimental data. The numerical analysis shows that, for a fixed effective electrical wetting tension, a greater input voltage should be applied at a higher frequency. It is because the effective electrical wetting tension decreases with increasing frequency. The numerical result also shows that the maximum electric-field strength is reduced even though the applied voltage is increased. The dielectric breakdown of the dielectric layer is highly likely to be one of the causes of contact-angle saturation. Therefore, the reduction of the electric-field strength with increasing frequency may give a clue to explain why the contact-angle saturation in the AC case generally occurs at a higher voltage with smaller contact angle compared to the DC case.
Appendix A Time average of the Maxwell stress and the RMS electric-field strength.

There is a following time-average theorem for the two time-harmonic complex variables of $u = \text{Re}\{\tilde{u}e^{j\omega t}\}$ and $v = \text{Re}\{\tilde{v}e^{j\omega t}\}$ (Jackson 1999):

$$\langle uv\rangle = \frac{1}{2} \text{Re}\{\tilde{u} \tilde{v}^*\} . \quad (A1)$$

Here, the bracket $\langle \rangle$ denotes the time average and the superscript $^*$ a complex conjugate. The directional vectors are time-independent, so that the time average of the electrical stress in $r$-direction in Eq. (14) can be rewritten as

$$\langle (T \cdot n) \cdot e_r \rangle = \langle (T) \cdot n \rangle \cdot e_r . \quad (A2)$$

The time average of the Maxwell stress tensor becomes from Eq. (A1),

$$\langle T \rangle = \frac{1}{2} \text{Re}\left\{-\frac{1}{2} \varepsilon (\tilde{E} \cdot \tilde{E}^*) I + \varepsilon \tilde{E} \tilde{E}^* \right\} . \quad (A3)$$

The complex electric field is decomposed as $\tilde{E} = \tilde{E}_r e_r + \tilde{E}_z e_z$ which is substituted into Eq. (A3) to obtain

$$\langle T \rangle = \frac{1}{2} \text{Re}\left\{-\frac{1}{2} \varepsilon (\tilde{E}_r \tilde{E}_r^* + \tilde{E}_z \tilde{E}_z^*) I + \varepsilon (\tilde{E}_r e_r + \tilde{E}_z e_z)(\tilde{E}_r^* e_r + \tilde{E}_z^* e_z) \right\} . \quad (A4)$$

The time-averaged Maxwell stress in Eq. (A4) is substituted to Eq. (A2) and normal vector is decomposed as $n = n_r e_r + n_z e_z$. Then the following result is obtained:

$$\langle (T \cdot n) \cdot e_r \rangle = \frac{1}{2} \text{Re}\left\{\varepsilon \tilde{E}_r^* (\tilde{E}_r n_r + \tilde{E}_z n_z) - \frac{1}{2} \varepsilon (\tilde{E}_r \tilde{E}_r^* + \tilde{E}_z \tilde{E}_z^*) n_r \right\} . \quad (A5)$$

Similarly, the RMS electric-field strength becomes

$$E_{rms} = \sqrt{\langle E \cdot E \rangle} = \sqrt{\frac{1}{2} \text{Re}\{\tilde{E} \cdot \tilde{E}^*\}} = \sqrt{E_{rms,r}^2 + E_{rms,z}^2} . \quad (A6)$$
Appendix B Average value of the Maxwell stress just below the TCL.

We introduce the control surface $\Sigma = S \cup S_1 \cup S_2 \cdots S_5$ as shown in Fig. A1. Here, $S$ denotes the surface used to obtain the average value in the numerical analysis. The mechanical equilibrium condition for the domain enclosed by $\Sigma$ is written as follows (Kang et al. 2003):

$$\int_{\Sigma} \mathbf{T} \cdot \mathbf{n} \, dS = \int_S \mathbf{T} \cdot \mathbf{n} \, dS + \int_{S_1 \cup S_2 \cdots S_5} \mathbf{T} \cdot \mathbf{n} \, dS = 0.$$ (B1)

Here, $\mathbf{n}$ the outward normal vector for each surface. By using Eq. (B1), one can calculate the force exerted on the surface $S$ from the sum of the forces acting on other surfaces, that is,

$$\int_S \mathbf{T} \cdot \mathbf{n} \, dS = -\int_{S_1 \cup S_2 \cdots S_5} \mathbf{T} \cdot \mathbf{n} \, dS.$$ (B2)

The average force acting on $S$ can be also expressed as follows:

$$\langle \mathbf{T} \cdot \mathbf{n} \rangle_S = \frac{1}{S} \int_S \mathbf{T} \cdot \mathbf{n} \, dS = -\frac{1}{S} \int_{S_1 \cup S_2 \cdots S_5} \mathbf{T} \cdot \mathbf{n} \, dS.$$ (B3)

It is quite clear the electric fields on the surfaces $S_1$, $S_3$, and $S_5$, which are nonzero contributions, have well-defined finite values. In conclusion, the average stress near the TCL is finite and then the average electric field is also finite.

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References


Fig. 1  a Equivalent circuit diagram for a droplet sitting on an insulator-covered electrode; b at very low frequency range (Ohmic currents dominates); c at very high frequency range (displacement currents dominates).

Fig. 2 A water droplet on a dielectric-coated electrode.

Fig. 3 Computational domain.

Fig. 4 Experimental set-up

Fig. 5 The root-mean-square potential distributions ($\phi_{rms}$) and the corresponding images for droplets where the applied RMS voltage ($V_{rms}$) is 143 V: a $f = 1$ kHz; b $f = 128$ kHz. In the contour plots, the contour interval ($\Delta V$) is 4.77 V.

Fig. 6 The root-mean-square potential distributions near the TCL where the applied RMS voltage ($V_{rms}$) is 143 V: a $f = 1$ kHz; b $f = 128$ kHz. In the contour plots, the contour interval ($\Delta V$) is 4.77 V.

Fig. 7. Frequency dependence of the shape of droplet at a fixed RMS voltage, $V_{rms} = 143$ V.

Fig. 8. Frequency and voltage dependence of: a contact angles and b time-averaged effective electrical wetting tensions ($W_{el}$). In the figure, $\theta$ represents the Young’s contact angle, and the effective electrical wetting tension is normalized by the droplet-air interfacial tension ($\gamma = 0.072$ N/m).

Fig. 9 Comparison of the normalized effective electrical wetting tension ($W_{el} / \gamma$) between the experimental results and the numerical results for $V_{rms} = 143$ V.

Fig. 10 Time-averaged radial force per unit area (radial stress) acting on the drop surface near the TCL, where $d = 5 \mu m$ is the thickness of the dielectric layer.

Fig. 11 RMS electric-field strength ($E_{rms}$) across the dielectric layer just below the TCL.

Fig. 12 Frequency dependence of the localized electric field at TCL ($E_{TCL}$, circles) and the necessary voltage ($V_{req}$, squares) to keep the effective electrical wetting tension constant ($W_{el} / \gamma = 0.481$). The inset is the chosen shape of droplet for calculation.

Fig. A1 Control surface for calculation of average stress just below the TCL
Fig. 8 a

Contact angle vs. Frequency (Hz)

- $\theta_\gamma$
- 57V
- 93V
- 113V
- 143V

Fig. 8 b

$W_{cl}/\gamma$ vs. Frequency (Hz)

- 57 V
- 93 V
- 113 V
- 143 V
Fig. 11

Fig. 12