

Prediction of the contact angle of colloids and microfluids during electrowetting

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A new generalised model of the electrowetting of microfluids has been proposed. Instead of the conventional Young–Lippmann formula, dynamic minimising of the total energy of system which is superposition of capillary free energy and electric energy has been utilised to calculate the contact angle as indicator of wetting behaviour. The change of interfacial energy of liquid/solid interface and change in relative permittivity of solution in the presence of colloid particles have also been considered in the developed model. The model predicts that contact angle saturation decreases in the presence of colloid particles both in DC and AC conditions. The model also predicts that the reduction of contact angle by applying potential is higher in the case of TiO_2 /water microfluidic system which is in agreement with experimental data reported in literature.

1. Introduction: Electrowetting is a phenomenon in which contact angle of liquid could be controlled by applying electrical potential between the liquid and a solid substrate [1]. Electrowetting is consequence of the change in thermodynamic of system by applying potential [2]. This process has gained much attention in recent years for application in modern technologies due to its capability for controlling the liquid motion in very low volume without the usage of any mechanical tools [3]. For instance, this process could be utilised in liquid lenses and lab-on-a-chip systems. The electrowetting has also reversibly been applied as energy harvesting system in moving tools for example in special shoes to charge battery of small electrical devices (e.g. cell phones) [4]. Several models have been developed for modelling of electrowetting, among them Young–Lippmann model [5] is the most famous one for prediction of contact angle changes by applying the potential. Despite these models, still lots of research have been conducted to extend the accuracy of contact angle prediction [6, 7].

Electrowetting-on-dielectric (EWOD) substrate introduced as main system to have reversible electrowetting without degradation of conducting liquid. In this system, a droplet of liquid separated from the conductive substrate electrode by a thin dielectric layer in order to prevent electrochemical reactions or other Faradic charge exchange at the electrode. Usually this dielectric layer is then coated with an even thinner hydrophobic (i.e. Teflon) layer in order to have high contact angle at start and increase the range of contact angle changes by potential.

Young–Lippmann model (1) is indeed modification of Young equation to include the change of solid/liquid interfacial energy upon applying electrical potential. This relation has some limitations which restricts its application, for instance it fails to predict contact angle saturation. One of the other limitations is that frequency dependence of contact angle is not considered in this relation [7, 8]

$$\cos \theta(U) = \cos \theta_0 + \frac{1}{2} \frac{\varepsilon_d \varepsilon_0}{\gamma_{la} d} U^2 \quad (1)$$

Moreover, the salt concentration and conductivity of liquid are not considered in this model while they also affecting the electrowetting. In order to overcome these limitations of the Young–Lippmann equation, several efforts have been performed. Peykov *et al.* [7] tried to include contact angle saturation in their developed model. They assumed that interface energy of solid/liquid is altered by applying the potential and at certain potential this energy

become zero which results in the contact angle saturation. Small deviation from prediction by their model occurred at high potential which is attributed to the kinetics of system. Chamakos *et al.* [6] tried to solve the nonlinear free-boundary problem of droplet shape using numerical method by the finite-element method. The main advantage of this approach is that structured dielectric surfaces can also be modelled. In the other word, surface roughness can be considered in modelling, but they do not report contact angle saturation in their developed model. One of the most successful models for electrowetting was developed by Klarman *et al.* [8] which could predict contact angle saturation as well as frequency dependence of the electrowetting. This model is also able to take into account the salt concentration in liquid. In addition, their model can be further developed to be applied in finite-element method for considering the surface roughness.

All of the mentioned models deal with liquid solution without the presence of colloid particles. In this paragraph, importance and attempts for modelling of electrowetting of colloid will be presented. One of the applications of the electrowetting which recently reported is to suppress coffee ring problem [1, 9]. In addition, electrowetting of microfluidic system will able us to design and fabricate future micro-switches [4]. In both of these applications, suspension of nanoparticles is utilised and therefore for such application knowledge and modelling of the electrowetting of suspension is worthwhile. Group of Shanahan [2] revisited Young–Lippmann equation for applying in microfluidic and colloid systems but as mentioned the model of Young–Lippmann has itself several limitations and therefore could not properly address the trait of real system. Moreover, they assumed that just energy of interfaces of the solid substrate and liquid differ from the pure liquid case in this configuration while presence of the particle in liquid could also modify the dielectric properties of the liquid. In addition, presence of the particle in liquid could also modify the impedance of the liquid in AC system and these are neither investigated nor modelled in previous report. In some cases, it was just reported that at certain potentials the shape of droplet could oscillate due to resonance in system but these observation limited to the liquids [10, 11]. It should be noted that for suppression of the coffee ring usually AC potential is applied and therefore it is needed to take into account the effect of frequency. In this Letter, modelling of the electrowetting of the nanosuspension by thermodynamical approach similar to the work of Klarman *et al.* [8] with more detailed theoretical consideration (especially due to presence of particles) has been performed. The effects of the presence of the particles on different properties of

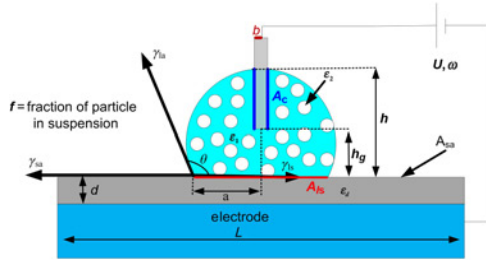


Fig. 1 Schematic diagram of electrowetting setup utilised for modelling in the current work. A droplet with shape of spherical cap and volume V and height h is placed on electrode. Droplet of suspension contains solvent with dielectric constant of ϵ_1 and particles with dielectric constant of ϵ_2 and particle volume fraction was f . The electrode is covered with a dielectric coating with thickness of d and relative permittivity of ϵ_d . A metal electrode with radius of b is inserted into the droplet and used as counter electrode. The gap between the metal wire and dielectric layer is h_g . The area of the dielectric covered by droplet is A_{ls} and that of the counter electrode is A_c . The applied electric potential is U with frequency of ω and the contact angle with the substrate is θ

liquid are also taken into account. TiO_2 nanosuspension as one of the most studied nanocolloid has been selected as a model system in this Letter since TiO_2 suspension has diverse applications in different fields such as photocatalysis, sensor, catalysis and semiconductor technologies [12]. In many of these applications, electrowetting may introduce new elaborate processing routes of TiO_2 suspension or thin film deposition techniques and therefore studying this nanosuspension is worthwhile.

2. Modelling: We assumed that the droplet in EWOD system is spherical cap as reported in [13] (Fig. 1). Fixed volume constraint is applied for modelling as the volume of droplet is constant (there is no considerable evaporation during the electrowetting). In view of the report of Klarman *et al.* [8], the total free energy of EWOD system can be separated into two distinct parts: capillary free energy and electric energy. If the total energy has been plotted against contact angle, the minimum of the plot indicates the equilibrium contact angle (see Fig. 2) at certain potential and by changing the potential the equilibrium contact angle is also modified. This method is the thermodynamical approaches for prediction of contact angle and if one considers dynamic outcomes of the effective parameters, all of the physical observation during electrowetting could be addressed. This is the main concept followed in current work for prediction of the equilibrium contact angle (equilibrium contact angle is in fact analogous of experimentally observed contact angle). In [2], it is mentioned that *contact angle is consequence of the*

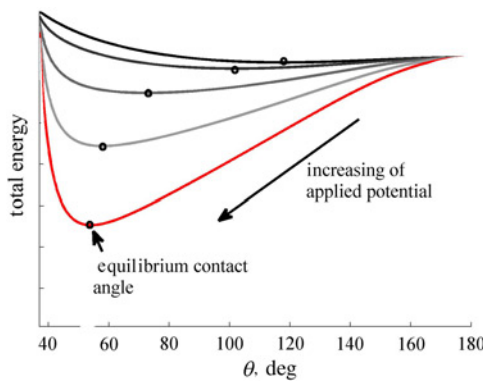


Fig. 2 Superposition of the capillary and electrical energy (total energy) at different applied voltages. The minimum of the total energy occurred in equilibrium contact angle which decreased with increasing the potential

thermodynamic of system rather than properties of a material which advocate this concept.

Klarman *et al.* [8] assumed that capillary free energy is just function of contact angle, while electric energy is affected by both of contact angle and potential changes. However in suspensions, the interface energy of solid/liquid is also function of the applied potential as reported by Orejon *et al.* [2]. Here, in order to generalise the calculation, it is assumed that γ_{ls} and consequently capillary free energy is also function of the potential while Klarman *et al.* [8] assumes that capillary energy is just function of contact angle. In higher concentration of particles, γ_{la} can also be affected by the presence of particles which can be implemented in current approach. Moreover, effect of presence of the particle in suspension should be considered and therefore the total free energy of system, capillary free energy and electric energy turn into function of the contact angle, particle concentration and applied electrical potential. We also know that frequency and salt concentration change the double layer behaviour and therefore they alter the electrical energy and subsequently the total energy

$$F_{\text{tot}}(\theta, U, f, \omega, C_{\text{salt}}) = F_{\text{cap}}(\theta, U, f) + F_{\text{el}}(\theta, U, f, \omega, C_{\text{salt}}) \quad (2)$$

The capillary free energy can be calculated by (3) considering the Young equation [13]. In order to take into account the changes of solid–liquid interface energy in the presence of the colloid particles, we use relation which is suggested by Shanahan group [2]. They assume that in low concentration other interface energies are constant while in higher concentration these energies are also function of concentration and presence of the particle near the interface could modify the surface tension. Interestingly, all these changes could be considered in our calculation as we started from basic thermodynamic and trying to calculate dynamic energy

$$F_{\text{cap}}(\theta, U, f) = A_{sa}\gamma_{sa} + A_{sl}\gamma_{sl} + A_{la}\gamma_{la} - V\Delta P \quad (3)$$

where γ_{ab} denotes the interface energy between medium a and medium b . A_{ab} is the areas of interface between media a and b which can be calculated considering the geometry of system. Subscripts utilised for showing the interface of different media are: subscript a denoting air, s showing solid and l representing liquid. Considering the geometry of system, a relation between contact angle and radius of droplet (5) could be obtained for conserving the volume of droplet (Appendix). Volume of spherical cap could be calculated by (4) [14]

$$V = \frac{\pi h}{6} (3a^2 + h^2) \quad (4)$$

$$R = \left[\frac{3V}{(1 - \cos \theta)^2 (2 + \cos \theta)} \right]^{1/3} \quad (5)$$

The area of interface between different medium as well as area of counter electrode (A_c) could also be obtained by following relations

$$A_{sa} = l^2 - A_{sl} \quad (6)$$

$$A_{sl} = \pi a^2 \quad (7)$$

$$A_c = 2b(h - h_g) \quad (8)$$

The typical length of system or electrode surface could also be estimated by relation (9) considering the Appendix

$$l \geq 2R(\theta_{\min}) = \left[\frac{3V}{\pi(1 - \cos \theta_{\min})^2 (2 + \cos \theta_{\min})} \right]^{1/3} \quad (9)$$

For calculating the electrical energy, the following relation can be

utilised based on [8]

$$F_{el}(\theta, U, f, \omega, C_{salt}) = -\frac{C_{tot}(\theta, f, C_{salt})}{2\sqrt{1 + \omega^2 \tau_b^2}} U^2 \quad (10)$$

In the above equation, C_{tot} is total capacitance which in fact results from two series capacitance: capacitance resulted from dielectric layer (C_1) and capacitance formed by double layer (C_2) on surface of counter electrode. Capacitance resulted from dielectric layer is very similar to flat capacitor and can be estimated by following equation

$$C_1 = C_{dl} = \frac{\epsilon_0 \epsilon_d A_{ls}}{d} \quad (11)$$

Capacitance formed by double layer on counter electrode could also be obtained by using the Debye theory and Debye length [7]

$$C_2 = \frac{\epsilon_0 \epsilon_1 A_c}{\lambda_D} \quad (13)$$

Debye length can be obtained by the following equation [7, 8]

$$\lambda_D = \left[(\epsilon_0 \epsilon_1 k_b T) / (2e^2 c_{salt}) \right]^{1/2} \quad (14)$$

The total capacitance caused by these two series capacitor is also calculated by (15) (presence of different dielectric layers can also simply be taken into account in this part as additional series capacitor)

$$C_{tot}^{-1} = C_1^{-1} + C_2^{-1} \quad (15)$$

Finally, the electrical energy at different potential and frequency could be calculated. It should be considered that total time for formation of double layer t_b can be obtained by the following equation

$$\tau_b = \frac{\lambda_D L}{D} \quad (16)$$

In the above equation, L is the typical length of system and D is the diffusion constant of solution. Considering the work of Shanahan group [2], the changes of energy of liquid solid interface γ_{sl} could also be estimated by the following equation

$$\gamma_{sl}(U, f) = \gamma_{sl}^0 - \gamma_{lg} A f^{1/3} U \quad (17)$$

For titanium nanoparticles, the constant (A) in (17) is about 0.0045 as reported in [2]. In addition, using the Maxwell-Garnet [15] equation, the effective dielectric constant of suspension (colloids or microfluids) could be obtained by the following equation

$$\epsilon = \epsilon_1 \times \frac{[2(1-f)\epsilon_1 + (1+2f)\epsilon_2]}{[(2+f)\epsilon_1 + (1-f)\epsilon_2]} \quad (18)$$

Moreover, it was reported that in high frequency ($\omega > 10^6$ Hz), impedance of the suspension [15] suppressed by particles presence, while in typical AC electrowetting the frequency is < 100 kHz and therefore we can omit the changes of impedance because of particle presence. The current approach could be further developed to consider this part if in certain suspension the changes are big enough to be considerable.

3. Results and discussion: In this section, results obtained from the developed model for contact angle calculation during the electrowetting will be reported. Having the value of all effective parameters like dielectric constant of suspension solvent, fraction

Table 1 List of the nomenclatures and the default values of different parameters for feeding the developed model

Parameter	Symbol	Presumed value
contact angle (initial contact angle, accessible minimum contact angle)	$\theta (\theta_0, \theta_{min})$	–
thickness of dielectric layer	d	100 nm
vacuum permittivity	ϵ_0	8.85×10^{-12} N/m ²
Boltzmann constant	K_b	1.38×10^{-23} m ² kg/(s ² K)
energy of system (subscript tot = total, cap = capillary, el = electrical)	F_{cap}, F_{el} and F_{tot}	–
Debye length	λ_D	–
capacitance of dielectric and double layer	C_1 and C_2	–
interface area (subscript in text shows two media)	A_{sl}, A_{la} and A_{sa}	–
relative permittivity of water	ϵ_1	$80 \epsilon_0$
relative permittivity of TiO ₂	ϵ_2	$86 \epsilon_0$
relative permittivity of dielectric layer	ϵ_d	$2.67 \epsilon_0$
height of the counter electrode	h_g	0.7 mm
droplet volume	V	5 μ l
diffusion constant	D	2.34 m ² /s
fraction of particles	f	0–10%
frequency of applied potential	ω	0–100 kHz
interface energy of solid/liquid	γ_{sl}	47 mN/m
interface energy of liquid/air	γ_{la}	72.8 mN/m
interface energy of solid/air	γ_{sa}	12.7 mN/m
radius of counter electrode	b	12.5 μ m
applied potential	U	0–100 V
concentration of salt (ions)	C_{salt}	50 m mol

System of distilled water and suspension of TiO₂ nanoparticles in water on substrates of teflon/miline C are selected as model for investigation.

of particle in the suspension, dielectric constant of nanoparticles, dielectric constant of insulating layer on conducting electrode, salt concentration in the solvent/conductivity of solvent, range of applied potential, frequency and geometry of electrowetting system (electrodes distance and volume of droplet), the contact angle could be predicted for different applied potential. In order to compare the result of current work with experimental data and other models, the values of different parameters were gathered from literatures [1, 2, 8, 16] and listed in Table 1. Fig. 3 shows the shape of droplet containing 1% of TiO₂ particles as well as

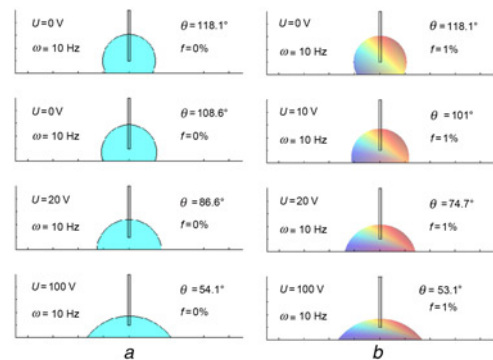


Fig. 3 Effect of different applied potential on the shape of droplet and contact angle in EWOD system
a In the absence of TiO₂ particles
b In the presence of 1% of TiO₂ particles as predicted by developed method in current work at frequency of 10 Hz ($C_{salt} = 50$ mM and all other values for calculation are listed in Table 1)

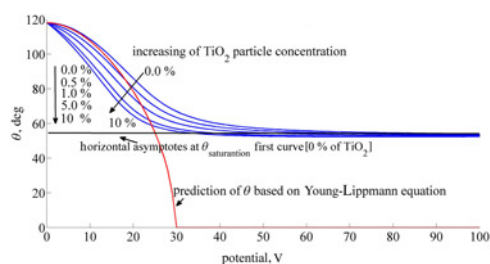


Fig. 4 Effect of different applied potential on contact angle of EWOD system in the presence of different value of TiO_2 nanoparticles (0–10%) as predicted by developed method in current work, for comparison the prediction by Young–Lippmann equation is also illustrated in figure ($C_{\text{salt}} = 50$ mM and all other values for calculation are listed in Table 1)

pure water solvent after applying different potential in EWOD system water at frequency of 10 Hz.

It could be seen that changes of the contact angle are faster for suspension of TiO_2 particles and the final value of contact angle is lower in the presence of particle. The changes of contact angle against TiO_2 particle loading in the suspension are also presented in Fig. 4 which shows the similar results. Increasing the particle concentration decreases the contact angle saturation value and also the reduction of contact angle with lower potential becomes faster. These results are in good agreement with results reported for suspensions and microfluidic system [17, 18].

Comparison of the prediction obtained by current model and experimental data reported by Shanahan group [2] is depicted in Fig. 5. It can be seen that there is good correlation between the experimental data and prediction of the current model. There is small deviation in the start point where initial contact angle predicted by model (118.1°) is higher than experimental value (about 109° – 111°). One should consider that for perfect Teflon layer the contact angle is 118.1° as predicted by current model (numerous literature report this value [8]). However, initial contact angle reported in [2] is lower than this value. Different experimental conditions like hysteresis or defect and impurity of the deposited layer may cause this deviation. In any case, the prediction of model is generally better than non-modified Young–Lippmann equation.

To get more information about behaviour of suspension during the electrowetting, the frequency-dependent changes of contact angle in suspension are also predicted by developed approaches (Fig. 6) which may be useful in forecasting the final pattern after

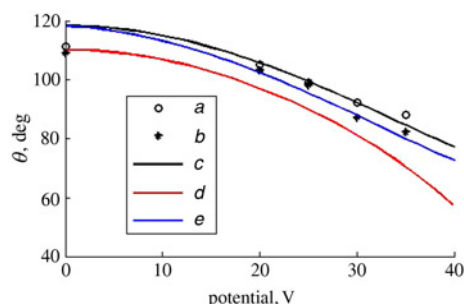


Fig. 5 Comparison of the prediction by current model with experimental data reported in literature; changes of the contact angle by applying different potential in case of (a) water solution without nanoparticles (experimental data in [2]), (b) suspension of TiO_2 nanoparticles with concentration of 0.1% (experimental data in [2]), (c) prediction of developed model in case of water solution, (d) prediction by Young–Lippmann, (e) prediction of developed model in case of TiO_2 nanoparticles suspension with concentration of 0.1% in water. The thicknesses of double dielectric layers were selected to be similar to [2], 95 nm of Ta_2O_5 layer with relative permittivity of about $100\epsilon_0$ [16] and 300 nm of Teflon layer with relative permittivity of $2.67\epsilon_0$ [8] (other values of parameters for calculation were listed in Table 1)

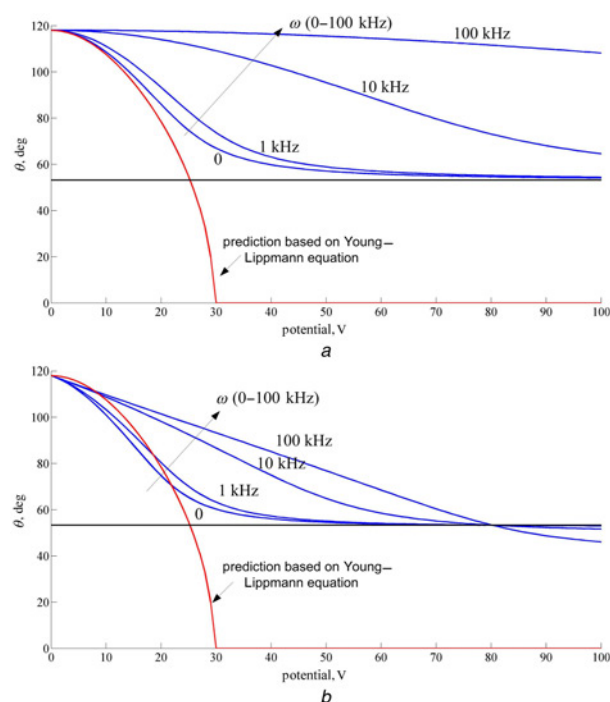


Fig. 6 Effect of AC potential with different frequencies on the contact angle of EWOD system

a In the absence of TiO_2 nanoparticles

b In the presence of TiO_2 nanoparticles (1%) as predicted by developed method in current work ($C_{\text{salt}} = 50$ mM and all other values for calculation are listed in Table 1), for comparison the prediction by Young–Lippmann equation is also illustrated in figure

electrowetting assisted droplet evaporation [9]. First observation which is also reported by different groups [8, 9, 11] is that the changes of the contact angle at higher frequency are lower. At higher frequency, charging the capacitors takes longer time and therefore the electrical energy is smaller. Consequently, the equilibrium contact angle is less affected at higher frequencies. Another observation is that in the presence of particles the contact angle saturation decreased in higher frequency while during the electrowetting of water, the saturation value is not changed by frequency (comparing Figs. 6a and b).

Concentration of ions in the solution which could be correlated with its conductivity could also affect the electrowetting mainly by altering the length of double layer and capacitance of double layer. Investigating the effect of this parameter in the solution is

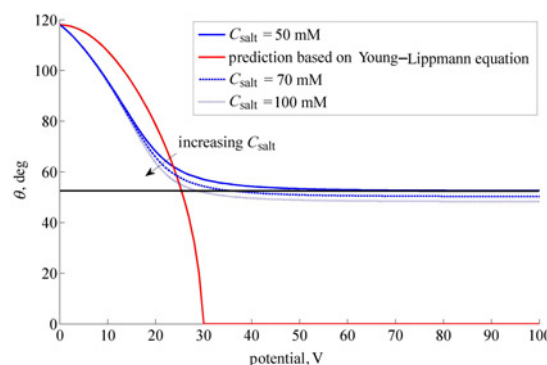


Fig. 7 Effect of potential on the contact angle of EWOD system in the presence of TiO_2 nanoparticles (1%) at frequency of 10 Hz and different salt concentration of solvent predicted by developed model, for comparison the prediction by Young–Lippmann equation is also illustrated in figure (all other values for calculation are listed in Table 1)

important because during the preparation of microfluidic system some new ions could also be formed in addition to the particles in solvent [17]. Fig. 7 shows the effect of salt concentration on the changes of contact angle upon applying the voltages. It can be seen that the contact angle saturation slightly delayed with increasing the salt concentration. This result can be verified by Dash *et al.* [17]. Dash *et al.* [17] did not observe contact angle saturation at the range of applied potential (0–60 V) for bismuth telluride microfluidic systems and they attribute this observation to the presence of COO^- anion on surface of particles and diffusion of H^+ ion to the solution (change in ionic strength of solution). The results obtained by this model verify their interpretation (delayed saturation of contact angle). It should also be considered that contact angle saturation will be occurred finally according to the prediction of our thermodynamical model. The reason that Dash *et al.* [17] did not observe contact angle saturation could be related to the fact that they just studied limited voltage range and according to current results the saturation voltage will be happened at higher potentials (e.g. >70 V).

4. Conclusion: The model developed in the current work is based on thermodynamical relation for calculating and superposition of the dynamic capillary and electrical energies. This model addresses some of the experimental observation during the electrowetting of suspension including: (i) fast reduction of contact angle upon applying voltage in EWOD system of suspensions and microfluids, (ii) delayed and lower contact angle saturation in suspensions and microfluids, (iii) frequency-dependent electrowetting of suspensions and (iv) delayed contact angle saturation with increasing the ion concentration in solution.

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7 Appendix

Considering Fig. 8, few geometrical relations between the height and volume of droplet could be obtained for conservation of the volume of the droplet. The volume of the spherical cap could be obtained by (19) according to [12]

$$V = \frac{\pi h}{6}(3a^2 + h^2) = \frac{1}{3}\pi h^2(3R - h) \quad (19)$$

The radius of sphere is perpendicular on the perimeter of it

$$\varphi + \theta = \frac{\pi}{2} \quad (20)$$

Sum of the angles of the triangle is equal to π and therefore

$$\varphi + \beta + \frac{\pi}{2} = \pi \rightarrow \varphi + \beta = \frac{\pi}{2} \rightarrow \beta = \theta \quad (21)$$

Also considering the trigonometric relations we can obtain the following equation

$$R - h = R \cos \beta = R \cos \theta \rightarrow h = R(1 - \cos \theta) \quad (22)$$

similarly $a = R \sin \theta$

Combining (22) and (19) we obtain the following equations

$$\begin{aligned} V &= \frac{1}{3}\pi h^2(3R - h) = \frac{1}{3}\pi [R(1 - \cos \theta)]^2 (3R - [R(1 - \cos \theta)]) \\ &= \frac{1}{3}\pi R^3(1 - \cos \theta)^2(2 + \cos \theta) \\ R &= \left[\frac{3V}{\pi(1 - \cos \theta_{\min})^2(2 + \cos \theta_{\min})} \right]^{1/3} \end{aligned} \quad (23)$$

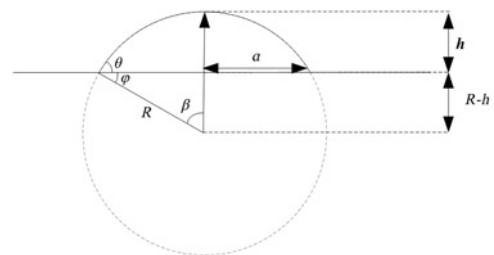


Fig. 8 Schematic illustration of droplet (spherical cap) on the substrate for deriving the geometrical relations

$$h = R(1 - \cos \theta) = \left[\frac{3V}{\pi(1 - \cos \theta_{\min})^2(2 + \cos \theta_{\min})} \right]^{1/3} (1 - \cos \theta) \quad (24)$$

$$a = R \sin \theta = \left[\frac{3V}{\pi(1 - \cos \theta_{\min})^2(2 + \cos \theta_{\min})} \right]^{1/3} \sin \theta \quad (25)$$

At possible minimum of θ , the height of droplet h is equal to distance of counter electrode and dielectric layer h_g and therefore for finding the length of necessary electrode which should have area larger than interface area of droplet and dielectric layer at $\theta_{\min}$, we can write

$$\theta \rightarrow \theta_{\min} \Rightarrow h \rightarrow h_g$$

$$\Rightarrow h_g = \left[\frac{3V}{\pi(1 - \cos \theta_{\min})^2(2 + \cos \theta_{\min})} \right]^{1/3} (1 - \cos \theta_{\min}) \quad (26)$$

$$\begin{aligned} \Rightarrow h_g &= \left[\frac{3V(1 - \cos \theta_{\min})^3}{\pi(1 - \cos \theta_{\min})^2(2 + \cos \theta_{\min})} \right]^{1/3} \\ &= \left[\frac{3V(1 - \cos \theta_{\min})}{\pi(2 + \cos \theta_{\min})} \right]^{1/3} \end{aligned} \quad (27)$$

$$\begin{aligned} h_g^3 &= \frac{3V(1 - \cos \theta_{\min})}{\pi(2 + \cos \theta_{\min})} \Rightarrow 2\pi h_g^3 + \pi h_g^3 \cos \theta_{\min} \\ &= 3V - 3V \cos \theta_{\min} \\ \Rightarrow \cos \theta_{\min} &= \frac{3V - 2h_g^3}{3V + h_g^3} \end{aligned} \quad (28)$$

$$l \geq 2R(\theta_{\min}) = \left[\frac{3V}{\pi(1 - \cos \theta_{\min})^2(2 + \cos \theta_{\min})} \right]^{1/3} \quad (29)$$