Modeling of Oil/Water Interfacial Dynamics in Three-Dimensional Bistable Electrowetting Display Pixels

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ABSTRACT: Electrowetting has drawn significant interest because of the potential applications of displays, lab-on-a-chip microfluidic devices, electro-optical switches, and so forth. However, electrowetting display (EWD) is monostable, which needs extra continuous voltage supply to keep contracting the oil. This paper is concerned with the simulation of two-phase liquid flow in three-dimensional EWD pixels with two electrodes (E1 and E2) demonstrating bistability, where power is only needed to move the oil droplet between two stable states. The effects of E1 geometry, E2 geometry, and E2 pulse characteristics on the dynamics of the oil droplet motion have been analyzed. Also, predictions of the transient states in four stages of the reversible bistable operation process have been carried out by employing the finite element method, in qualitative agreement with our experimental results of the monostable EWD and the existing literature. We seek to shed more light on the fundamental two-phase liquid flow in three-dimensional pixels exhibiting bistability for low power EWD and guide optimizing the electrodes to the perfect patterns with the aid of rigorous modeling.

1. INTRODUCTION

Electrowetting has shown the potential of microscale fluid motion manipulation by changing the surface tension, which has been widely used in chemistry, bioengineering, sensors, and displays. Electrowetting displays (EWDs) reflect ambient light to provide a paper-like display with competitive advantages such as capable of video playback, low power consumption, sunlight readability, and reading comfort. Hayes and Feenstra first proposed an EWD to show the ability of video-speed response time (<10 ms), which was published in 2003; then, <2 ms switching speed has been reached by Smith in 2007. Heikenfeld used Young–Laplace transposition of brilliant pigment dispersions to realize electro-fluidic display, showing its low power consumption in 2009. EWDs also show >50% white state reflectance, flexibility, multigray scale, and full color.

Despite the great success of conventional EWD, it is monostable, which needs extra continuous voltage supply to keep contracting the oil. The oil will return to the initial relaxation off-state if the voltage is removed. In bistable electrowetting display (BEWD) pixels, droplets are confined in either of two distinct stable states that are electrically switchable, power is only needed to switch the pixel but not required to maintain the pixel state once switching is finished. BEWD is the desire for very low power dynamic display (1 mW/in.²) or zero power static display better than typical EWD (20 mW/in.²), LCD (80 mW/in.²), and OLED (170 mW/in.²). However, the two stable states of liquids are usually realized by specially designed and more complicated pixel structures, which causes difficulties in the fabrication process. Because of these challenges, research studies on BEWD lag far behind that of conventional EWD. Therefore, more effort is needed to push the BEWD technology forward.
design, it is necessary to fully understand the motion of microfluidics in a pixel during operation. For this reason, numerical simulations are used as a tool for investigations. Ku et al. are among the first who attempted the simulation of the microfluidic movement of different electrode patterns for use in EWDs. Hsieh et al. proposed a more rigorous and accurate model by coupling the electrohydrodynamic (EHD) force deduced from the Maxwell stress tensor with the laminar phase field of the oil/water dual phase. In general, the research models of microfluid flow including the boundary element method, the spine-flux method, the volume of fluid method, the level set method, and phase-field method (PFM) are widely used. PFM is the most promising and reliable among the abovementioned models as it facilitates a thermodynamic treatment of the phase interfaces, rendering it more physically consistent with the direct simulation of two-phase flow by coupling the classical macroscopic laws of Cahn–Hilliard (CH) equation and Navier–Stokes (NS) equations, where the necessary terms are exchanged between the two sets of equations. However, there are no PFM modeling published works devoted to the investigation of two-phase microfluidic in three-dimensional BEWD pixels to the best of our knowledge.

This work aims to investigate, through numerical solutions and computational modeling analysis, the tracking of the oil/water interfacial dynamics in BEWD pixels. The model is obtained by employing the COMSOL Multiphysics software package via coupling the CH equation, the NS equation, and fundamental laws of electrostatics (FLE) calculations, which contributes to the topic, on the theoretical side. The effects of electrodes geometry including nonplanar L-shaped and U-shaped E1, planar E1, and notched E2 will also be analyzed in the model. The transient states of oil/water behavior actuated shaped E1, planar E1, and notched E2 will also be analyzed in three-dimensional BEWD pixel structure with oil motion in the water on a planar hydrophobic surface showing the ON state (30 V) and the OFF state (no voltage). The presented model is based on the cross-sectional view (white dashed arrows) of the oil droplet in the water phase motion controlled by nonplanar E1 and E2 in a three-dimensional BEWD pixel.

2. MODEL FORMULATION

A novel three-dimensional BEWD pixel structure model with two nonplanar electrodes is proposed based on the monostable EWD pixel with one planar electrode, as shown in Figure 1. Figure 1a show the micrographs of pixels array and the unit pixel structure of the monostable EWD fabricated by our team, where the motion of an oil droplet in the water phase on a planar hydrophobic surface is controlled by the voltage on one planar electrode. The model is based on the cross-sectional view of the three-dimensional BEWD pixel structure with quarter-circle electrode 1 (E1) at the bottom of the pixel reservoir and notched electrode 2 (E2) on the top surface of the pixel step unable to be confined in a single plane. The nonplanar E1 and E2 control the oil droplet in the water phase to move between the pixel reservoir and the pixel step, showing the potential of bistability, as shown in Figure 1b.

2.1. Governing Equations. 2.1.1. Phase-Field Method.

We adopt the PFM to track the dynamic moving interface, which has been demonstrated by numerous numerical studies to predict the effectiveness of droplet movement on a solid surface. The moving interface between oil and water is set as a tiny nonzero-thickness transition region. Thus, the physical properties at the interface of the immiscible fluids could be described by functions within this region with the help of a continuous phase-field variable \( \phi \), which varies from \(-1\) for water to 1 for oil. The volume fractions of the individual fluids of water and oil are

\[
V_{\text{water}} = \frac{1 - \phi}{2}, \quad V_{\text{oil}} = \frac{1 + \phi}{2}
\]

(1)

The multiphysics coupling feature defines \( \rho \) as the density (kg/m\(^3\)) of fluids, \( \mu \) as the viscosity (Pa·s) of fluids, and \( \varepsilon \) as the dielectric constant of fluids to vary smoothly over the interface by letting

\[
\rho = \rho_{\text{water}} V_{\text{water}} + \rho_{\text{oil}} V_{\text{oil}}
\]

(2)

\[
\mu = \mu_{\text{water}} V_{\text{water}} + \mu_{\text{oil}} V_{\text{oil}}
\]

(3)

\[
\varepsilon = \varepsilon_{\text{water}} V_{\text{water}} + \varepsilon_{\text{oil}} V_{\text{oil}}
\]

(4)

In the PFM interface, the two-phase flow dynamics is governed by a CH equation, which tracks a diffuse interface separating the immiscible phases. The diffuse interface is defined as the region where the dimensionless phase field variable goes from \(-1\) to 1. When solved in COMSOL Multiphysics, the CH equation is split up into two equations

\[
\frac{\partial \phi}{\partial t} + u \cdot \nabla \phi = -\nabla \cdot (M V G)
\]

(5)

\[
G = \lambda \left( -\nabla^2 \phi + \phi (\phi^2 - 1) / h_{\text{PP}}^2 \right)
\]

(6)

In eq 5, \( u \) is the fluid velocity (m/s). The mobility \( M \) (m\(^3\)/kg) can be expressed as \( M = \chi \rho_{\text{PP}} \), where \( \chi \) is the characteristic mobility, which controls the mobility of the interface. \( G \) is the chemical potential, a partial differential of the total free energy concerning \( \phi \), as shown in eq 6, where \( \lambda \) is the mixing energy density parameter (N). Also, \( \lambda \) and \( h_{\text{PP}} \) are related to the oil/water interfacial tension \( \gamma_{ow} \) through the relation

\[
\gamma_{ow} = 2 \sqrt{2} \lambda / 3 h_{\text{PP}}
\]

(7)

\( h_{\text{PP}} \) is a capillary width that scales with the thickness of the diffuse interface in PFM. The value of \( h_{\text{PP}} \) should be
determined empirically,\textsuperscript{30} and we adopt \( h_{\text{FF}} = h_{\text{oil}} \) in the present simulation, where \( h_{\text{oil}} \) is the oil film thickness.

\subsection*{2.1.2. Mass and Momentum Transport.} The NS equations describe the transport of mass and momentum for fluids of constant density in the PFM interface. To account for the dynamic movement of the immiscible two-phase flow, it is crucial to include surface tension in the model. The NS equations are then\textsuperscript{31}

\[
\rho \left( \frac{\partial u}{\partial t} + u \cdot \nabla u \right) = -\nabla p + \nabla \cdot (\mu \nabla u + \nabla u^T) + F_s + \rho g + F_E \tag{8}
\]

\[
\nabla \cdot u = 0 \tag{9}
\]

Here, \( u \) and \( \rho \) are the velocity and density of the fluids, \( p, g, F_s, F_E \) respectively denote the pressure, the gravitational acceleration, the surface tension force, and the volumetric electrodynamic force generated by an electric field. In the PFM, \( F_E \) can be calculated over the computational domain in terms of the chemical potential and phase-field variable by\textsuperscript{31}

\[
F_E = G \nabla \phi \tag{10}
\]

where \( G \) is the chemical potential defined in eq 6 of CH equations, that is, the CH and the NS equations are coupled via the surface tension force \( F_s \). Obviously, \( F_s \) approaches zero except those at the diffusive thickness of the oil/water interface. The volumetric electrodynamic force \( F_E \) can be expressed by the divergence of the Maxwell stress tensor \( T^M \):\textsuperscript{32}

\[
F_E = \nabla \cdot T^M \tag{11}
\]

The Maxwell stress tensor is given by\textsuperscript{32}

\[
T^M = \begin{bmatrix} T_{xx} & T_{xy} \\ T_{yx} & T_{yy} \end{bmatrix} \tag{12}
\]

where \( T_{ij}^M = \varepsilon E_i E_j - \varepsilon / 2 (E_i^2 + E_j^2), T_{yy}^M = \varepsilon E_y E_y \). The subscript \( i \) and \( j \) indicate the directions \( x \) and \( y \), respectively. \( E \) is the electrical field strength.

\subsection*{2.1.3. Fundamental Laws of Electrostatics.} The electrostatic force in the NS equation \( F_E \) couples the flow equations with the equations of the electrostatics, where the general form for this body force is \( \rho_E E \). The water phase behaves as a conductive liquid by assigning a high dielectric constant (80), which is compatible with the general treatment in an EW theory.\textsuperscript{33} FLE resolved in COMSOL Multiphysics commercial software are defined as\textsuperscript{32}

\[
\nabla \cdot D = \rho_E \tag{13}
\]

\[
E = -\nabla V \tag{14}
\]

Equation 13 is a differential form of Gauss’s Law, where \( D \) is the electric field displacement (\( \varepsilon_0 \varepsilon_r E \)), where \( \varepsilon_0 \) is the permittivity of the vacuum and \( \varepsilon_r \) is the relative permittivity. \( \rho_E \) is the volumetric charge density. Equation 14 is a conservative field, where \( V \) is a potential function.

\subsection*{2.2. Boundary Conditions.} In this section, we will assume the boundary conditions of the model in the present study including the electrostatic field, PFM and mass, and momentum transport boundary conditions. The modeling domain is the three-dimensional unit pixel in BEWD, as shown in Figure 2. The electrostatic field in the hydrophobic layer, the dielectric layer, hydrophilic pixel wall, oil phase, and water phase, is assumed to be governed by FLE, the zero-charge condition (\( \rho_i = 0 \)) is adopted for all of the exterior boundaries except the electrode regions. For the electrode regions, we specify the voltage \( V_{\text{com}} = 0 \) on the common electrode, \( E_{E1} \), and \( E_{E2} \) as the voltages on the electrode E1 and E2, respectively. In the boundary conditions of PFM, the solid surfaces (e.g., hydrophobic surface, pixel wall, and top substrate) are considered as wetted surfaces, and along the surfaces, we specify a wetted contact angle \( \theta_w \), which is related to \( \phi \) through

\[
n \cdot \nabla \phi = \cos(\theta_w) \nabla \phi | \nabla \phi | \tag{15}
\]

where \( n \) is the unit vector normal to the wall. With an applied voltage across the dielectric layer, the initial contact angle of a conductive liquid on the hydrophobic surface \( \theta_{FP} \) decreases to \( \theta_w \) according to Young–Lippmann equation,

\[
\cos \theta_w = \cos \theta_{FP} + \frac{CV^2}{2\varepsilon_0} \tag{16}
\]

where \( C \) is the capacitance per unit area of the hydrophobic and dielectric (\( \text{F/m}^2 \)), \( V \) is the dc voltage, and \( \varepsilon_0 \) is the interfacial surface tension between the aqueous and oil phases (\( \text{N/m} \)).

In the boundary conditions of NS equation formulation, we assume that there is no net flow in the water phase so that no pressure difference was applied on the boundary of the water phase and the pressure at the four outlets for a unit pixel is chosen as \( p = 0 \). Also, zero mass flux and no-slip boundary condition \( (u = 0) \) is used to associate with the mass and momentum equation. The detailed parameters used in the present model comprising material properties, interfacial properties, and geometric properties are listed in Table 1.

\subsection*{2.3. Mesh Independency Study.} Three different triangular mesh sizes, coarse mesh, medium mesh, and fine mesh, were employed in this study, which corresponds to 11.4, 1.15, and 0.12 \( \mu \text{m} \) per unit, respectively. Figure 3 illustrates the predicted time evolution of the contact radius of the oil droplet on the pixel step as compared to the experimental measurements of our monostable EWD when dc of 30 V was applied. The results show that the 1.15 \( \mu \text{m} \) mesh yields a satisfactory prediction of the measurements and further mesh refinement does not drastically improve the prediction accuracy. Hence, a mesh size of 1.15 \( \mu \text{m} \) per unit was used in the simulations of this work.
3. RESULTS AND DISCUSSION

We next proceed to evaluate the effect of various parameters like E1 geometry, E2 geometry, and E2 pulse characteristics on the dynamics of the oil droplet motion. Finally, we will demonstrate the application of the reversible operation process of BEWD in a confined three-dimensional pixel with two electrodes exhibiting bistability.

3.1. Effect of E1 Geometry. The oil droplet motion in the pixel reservoir is actuated by the localized change in contact angle as a result of the electric field at the triple contact line. Hence, it will be interesting to investigate the effect of E1 geometry on the electric field distributions (EFDs) in the pixel reservoir to find the lowest actuation threshold voltage of the oil droplet. E1 with electrodes on the left sidewall and bottom of the pixel reservoir, E1 with electrodes on both sidewalls and the bottom of the pixel reservoir, and E1 with electrode only at the bottom of the pixel reservoir are defined as L-shaped, U-shaped, and planar E1, respectively. The EFD data along the oil/water interface of the initial state from left 0 to right 50 μm position illustrated in the inset are plotted in Figure 4. The EFD of 60, 80, and 100 V threshold voltages on L-shaped, U-shaped, and planar E1, respectively, capable of moving the oil out of the pixel reservoir in the model. The data were obtained from the simulation results of COMSOL Multiphysics software package. The voltage of the L-shaped E1 decreases gradually in the widest range (44–4 V) among the three types of E1 from left to right along the whole 50 μm interface, where the pressure gradient introduced by a nonuniform contact angle is the driving force behind electrowetting-based microfluidics. Hence, L-shaped E1 is favorable for fluids’ motion and the actuation threshold voltage is 60 V. However, the decreasing range of voltage along the whole interface is small (47–38 V) in U-shaped E1, negative for oil/water motion, so the actuation threshold voltage is 80 V. Most of the voltages along the interface stay at around 60 V with a sharp decrease to 28 V at the end of the interface (48–50 μm) in the planar E1, so it requires the highest actuation threshold voltage (100 V) to move the oil out of the pixel reservoir.

The switching speed between ON and OFF states is crucial for video playback with no flicker of displays. We assumed that the same 85 V was applied on three types of E1 in the model; then, we plotted the center of the main oil droplet position in the x-axis direction as a function of time consumed to move the oil from the pixel reservoir to the pixel step, as shown in Figure 5. The modeling results show that the L-shaped E1 design can be altered to optimize the speed at which the oil droplet moves and also improve the reversibility of the fluids.

When dc 85 V was applied, no residual oil stayed on the L-shaped E1, residual oil stayed at the right corner of the pixel reservoir of U-shaped E1, and residual oil stayed at both the left and right corners of the pixel reservoir of planar E1, as
shown in the insets of Figure 5. In addition, the residual oil is also voltage-dependent as demonstrated in the model. The reason is that when the driving force reaches the threshold to overcome the interfacial tension and moves the oil, the oil droplet may easily split into different parts, resulting in the residual oil left in the pixel reservoir. However, if the voltage is increased, the driving force will be much larger than the interfacial tension and the oil droplet will move out of the reservoir completely.

3.2. Effect of E2 Geometry. To quantitatively characterize the motion of the oil droplet on the pixel step actuated by different E2 geometry, the right edge position of the main oil droplet as a function of time is plotted in Figure 5. When dc of 30 V was applied, voltage-dependent oil deformation actuated by the nonplanar E2 design on the top surface of the pixel step was more obvious than the planar E2 design at the bottom of the pixel step, easily explained by the difference of a thick dielectric pixel step ($\varepsilon_{PS} = 3.28$, $h_{step} = 12 \mu m$) on planar E2 reducing the electrostatic field that the water experiences at the interface. As a result, only notched E2 on the top surface of the pixel step succeeded in moving the oil droplet close to the pixel reservoir, in agreement with the experimental results of monostable EWD pixels of our team shown in the insets.

3.3. Effect of E2 Pulse Characteristics. The pulse on nonplanar E2 (notched) was divided into two parts, including part one which aimed to merge the oil film to a droplet close to the pixel reservoir, and part two of dc 60 V threshold voltage which moved the oil droplet back to the pixel reservoir. We herein investigate the pulse voltage and duration of part one. The position of the main oil droplet right edge in the x-axis direction was plotted as a function of part one voltage, as shown in Figure 7. dc 30 V should be the best voltage producing the maximum oil contraction close to the pixel reservoir. Also, we could observe from Figure 6 that the oil finished contracting in 1 ms, so we optimize the 5 ms E2 pulse to 30 V (1 ms) and 60 V (4 ms) according to the analysis.

3.4. Reversible Operation Process of BEWD. The reversible operation process of the oil droplet motion in a confined three-dimensional BEWD pixel is divided into four stages (a to d) in the model, actuated by combined square pulse voltages of nonplanar L-shaped E1 and notched E2 in Figure 8 investigated in the previous section. The detailed oil droplet motion in every stage of the present model in Figure 9 (Video S1) will be described in this section.

3.4.1. Stable ON State (Stage a). Figure 9a shows the colored oil concentrated in the pixel reservoir from 0 to 5 ms, where the larger area of pixel step reflects a white backplane in the ON state. The optimized L-shaped E1 and notched E2 demonstrated with advantages were applied in the model. The hydrophobic pixel reservoir surface provides a physical barrier to confine the oil phase contributing to the stability of the ON-
state without voltage. The absence of velocity vector distributions (yellow arrows) in Figure 9a also demonstrates that there is no fluid flow during 5 ms in stage a.

3.4.2. Oil Crosses the Pixel Step (Stage b). The oil droplet in the water phase overcomes an energy barrier of the pixel step edge with a 60 V square pulse to move the oil onto the pixel step in stage b of this model. The evolution results of the modeling demonstrate the full motion process of the oil phase changing the position in stage b, as shown in Figure 9b, where the yellow arrows represent the velocity vector indicating the trend of the fluid flow. Most part of the oil droplet is gathered on the top surface of the pixel step after stage b, resulting in a significant increase of the oil coverage area on the pixel step.

3.4.3. Stable OFF State (Stage c). Figure 9c shows the evolution of the oil phase relaxed without voltage on top of the pixel step surface from a droplet to a film. When the voltage is removed, the oil film remains stable on the top surface of the pixel step because of the interfacial tension of oil/water fluids. As a result, the colored oil covers almost the whole surface of the pixel step without voltage, so this state of the pixel is called a stable OFF state.

3.4.4. Oil Flows Back to the Reservoir (Stage d). A similar mechanism describes the reverse motion of the oil droplet from the top surface of the pixel step to the pixel reservoir when 30 V (1 ms) and 60 V (4 ms) stepwise square pulses are applied to E2, analyzed in the previous section. The design of stepwise dc voltages is also in accordance with the experimental result of Zhang et al.\textsuperscript{34} in monostable EWD pixels to avoid oil film splitting. After most of the oil moves back to the pixel reservoir of stage d in the model at 20 ms, it changes into the stable ON state of stage a again, as shown in Figure 9d.

The reversible operation process of BEWD with 100 V square pulses on planar E1 and E2 in three-dimensional pixels predicted by our model could reproduce the existing experimental results of the prior literature reported by Charipar et al. in ref 15, demonstrating bistability. Also, the modeling results of exhibiting bistability in three-dimensional BEWD pixels with nonplanar electrodes are supported by the experimental results reported by Zhang and Liang.\textsuperscript{35} The numerical solutions of the presented modeling qualitatively agree with the existing literature, which indicates the accuracy and capacity of our model in dealing with oil/water interface tracking in three-dimensional BEWD pixels.

4. CONCLUSIONS

BEWD has received much attention in the literature owing to its marked low power consumption. However, most studies were experimental. Here, rigorous modeling of oil/water interface tracking in three-dimensional BEWD pixels was obtained by employing the PFM of COMSOL Multiphysics software package, numerical solutions, and contributions to the topic, on the theoretical side. More importantly, the effects of E1 geometry, E2 geometry, and E2 pulse characteristics on the dynamics of the oil droplet motion have been analyzed. The advantages of nonplanar L-shaped E1 and notched E2 design
with stepwise square pulses were also demonstrated. Then, the application of the reversible operation process in a confined three-dimensional BEWD pixel has been realized, which reproduced the existing experimental results, exhibiting bistability in a numerical way. These results shed some light on the structure optimization of the electrodes to the perfect pattern for oil/water two-phase flow in three-dimensional BEWD pixels.

## ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.9b04352.

Slow-playing animation of a complete BEWD operation process as shown in Figure 9 exported from the COMSOL Multiphysics software package (AVI)

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### Notes

The authors declare no competing financial interest.

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