Numerical Study of Microdroplet Deformation and Breakup under Electric Fields: Postprint

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Abstract

Precise manipulation of microdroplets using electric fields holds broad application prospects in chemical engineering, biomedicine, energy, and environmental fields. Accurate prediction of the deformation and breakup behavior of viscous droplets under applied electric fields is an important prerequisite for precisely controlling microdroplet behavior. In this paper, a hybrid lattice Boltzmann-finite difference numerical method based on the leaky dielectric model is developed to investigate droplet deformation and breakup behavior under steady electric fields. The results show that: the numerical method developed in this paper can accurately predict small deformations of droplets under steady electric fields, verifying the validity and accuracy of the model; for droplets deforming perpendicular to the electric field direction (oblate type), the droplet exhibits four distinct morphologies with increasing electric capillary number: elliptical, quasi-elliptical, dumbbell-shaped, and breakup; for droplets deforming along the electric field direction (prolate type), the variation of its deformation parameter with electric capillary number is significantly different from that of oblate droplets, and the morphologies exhibited by the droplet include elliptical, quasi-elliptical, periodic oscillation, and breakup. During the droplet deformation stage, both oblate and prolate droplets exhibit increased deformation with increasing electric capillary number; during the droplet breakup stage, the time required for breakup decreases with increasing electric capillary number. The research work provides a theoretical foundation for a profound understanding of the mechanisms of microdroplet deformation and breakup under electric fields.

Full Text

Preamble

Numerical Study of Microdroplet Deformation and Breakup under a Steady Electric Field

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Abstract
Manipulation of microdroplets in a precise manner by an external electric field has received much attention due to its importance in the fields of chemical engineering, biomedicine, energy and environment. In order to accurately manipulate the droplet behavior in microfluidic devices, it is necessary to study and gain an in-depth understanding of the deformation and breakup of a viscous droplet under an external electric field. In this paper, a hybrid method, in which the color lattice Boltzmann and the finite difference methods are coupled by the leaky dielectric model, is used to study the deformation and breakup of a droplet under a steady electric field. The hybrid method is first validated by simulating the droplet subject to a small deformation. We then investigate the influence of the intensity of the electric field, the ratio of the dielectric permittivity inside and outside of the droplet, as well as the ratio of the electric conductivity on the droplet behavior. It is found that the oblate droplet can exhibit four different shapes or states, i.e. ellipse, quasi-ellipse, dumbbell, and droplet breakup, which are strongly dependent on the electrical capillary number. Distinct from the oblate droplet, the prolate droplet does not exhibit the dumbbell shape but the periodic oscillation. For either prolate or oblate droplet, increasing electrical capillary number leads to an increased droplet deformation when the deformation occurs, and to a shorter time for the droplet to break up when the breakup occurs. This study provides the theoretical foundation for a deep understanding of droplet deformation and breakup in the presence of electric field.

Key words
Lattice Boltzmann method; Droplet deformation and breakup; Leaky dielectric model; Electrical capillary number

Microdroplets offer numerous advantages including small volume, low diffusion, absence of cross-contamination, rapid reaction kinetics, and high-throughput analytical potential. With continuous advances in microfluidic technology, droplet-based microfluidics has gained widespread attention and application in chemical engineering, biomedicine, energy, and environmental fields. For instance, in chemistry, droplet microfluidics is gradually replacing traditional laboratory settings in analytical chemistry, with microdroplets increasingly serving as nanoliter-scale reactors or synthesizers that precisely control molecular concentrations and reaction times to automatically and rapidly complete various complex chemical reactions and analytical functions. In these applications, precise manipulation of droplet dynamics is essential. While various methods exist for generating and controlling microdroplets, electric field manipulation represents an effective non-contact approach that has been successfully applied in industries such as atomization, inkjet printing, enhanced droplet coalescence, and demulsification. Under an external electric field, droplets suspended in viscous liquids can exhibit complex behaviors including deformation, motion, rotation, and breakup, which primarily depend on electric field intensity and fluid properties such as viscosity, interfacial tension, conductivity, and dielectric constant. Therefore, investigating droplet deformation and breakup under electric fields is both an inherent requirement for microfluidic technology development and provides theoretical guidance for industrial applications.

Early theoretical and experimental investigations of immiscible droplet deformation under electric fields began with the work of Taylor and Melcher and Taylor. Taylor' s theoretical study of droplet deformation was based on several fundamental assumptions: the droplet is electrically neutral, the electric field is quasi-static, both the droplet and carrier fluid are leaky dielectric fluids, the droplet undergoes only small deformation, and there is no charge mobility on the droplet surface. This framework is known as the leaky dielectric model. Under these assumptions, the droplet itself carries no net charge but allows charge accumulation on its surface. Since charge accumulation occurs much faster than fluid motion, the system reaches a pseudo-steady charge distribution, satisfying the quasi-static electric field assumption. Hua et al. employed both leaky dielectric and perfect dielectric models using a front-tracking method to study the deformation of charged droplets in uniform electric fields, confirming that the theory can effectively predict the deformation of droplets with small deformation, finite conductivity, and moderate differences in dielectric constants under steady electric fields. However, experimental studies by Torza et al. revealed significant deviations between Taylor' s model predictions and experimental results when droplet deformation became large. Subsequently, Ajayi extended Taylor' s linear theory by proposing a second-order theoretical model, though this approach still failed to fundamentally resolve the discrepancy between theoretical predictions and experimental observations.

Under electric field influence, droplets not only deform but may also rupture. Induced charges accumulate at the droplet surface while coupling with the internal and external viscous flows, and interfacial tension between the fluids presents significant challenges for theoretical analysis of electrohydrodynamic problems. With rapid improvements in computational speed, numerical modeling and simulation have become another important tool for studying droplet dynamics under electric fields. Sherwood investigated droplet breakup in electromagnetic fields using a boundary integral method to solve the Laplace equation governing the electric field and the Stokes equations governing the flow field. Based on the leaky dielectric model, Baygents employed a similar approach to study the interaction of two droplets in a uniform electric field. Feng and Scott also used the leaky dielectric model with a Galerkin finite element method to establish relationships between droplet deformation and electric field intensity under Stokes flow conditions and finite Reynolds numbers. Lac and Homsy conducted three-dimensional numerical studies of droplet deformation under steady electric fields using the boundary integral method, while Fernández et al. analyzed droplet deformation in microchannels under electric forces, and Tomar et al. simulated the deformation behavior of conductive droplets under electric fields using the Volume-of-Fluid method.

In recent years, the lattice Boltzmann method (LBM) has evolved into an effective numerical tool for simulating complex fluid flows. LBM possesses inherent parallel characteristics, simple boundary treatment, and straightforward implementation. Furthermore, its kinetic theory nature enables convenient description of interactions between different phases with automatic interface tracking, making LBM particularly suitable for simulating microdroplet dynamics. This paper employs the LBM color model to solve two-phase flow and the finite difference method to solve the electric field equations, with the two approaches coupled through the leaky dielectric model to numerically investigate the deformation and breakup behavior of incompressible viscous droplets under steady electric fields, revealing the influence of electric field intensity, dielectric constant ratio, and conductivity ratio on droplet behavior.

1.1 Research Object

A two-dimensional circular droplet with initial radius R and electrical neutrality is placed in a steady electric field formed by two parallel-plate capacitors. The droplet is positioned at the center between the plates and suspended in a carrier fluid. Based on symmetry considerations, the computational domain above the droplet' s horizontal axis is used in this study, as illustrated in [Figure 1: see original paper]. The upper plate maintains a constant electric potential Φ, while the lower boundary of the computational domain serves as the horizontal symmetry axis with zero potential, establishing a vertically downward steady electric field with intensity E.

The physical properties are denoted by subscripts "i" for the droplet and "o" for the carrier fluid: densities ρ_i and ρ_o, dielectric permittivities ε_i and ε_o, electrical conductivities σ_i and σ_o, and dynamic viscosities μ_i and μ_o. These define the following dimensionless parameters: density ratio $M = \rho_i/\rho_o$, dielectric constant ratio $Q = \varepsilon_i/\varepsilon_o$, conductivity ratio $R = \sigma_i/\sigma_o$, and viscosity ratio $\lambda = \mu_i/\mu_o$. For simplicity, this study assumes identical density and viscosity for the droplet and carrier fluid ($M = 1$, $\lambda = 1$) and neglects gravitational effects. The interfacial tension coefficient between the droplet and external carrier fluid is γ.

Since charges exist only at the droplet surface, electric forces act solely on the droplet surface, characterized by the electric capillary number Ca_E representing the relative magnitude of electric field strength to interfacial tension:

$$Ca_E = \frac{\varepsilon_o E^2 R}{\gamma}$$

It is worth noting that the computational domain size depends on the droplet deformation. The domain is configured to minimize computational cost while maintaining a reasonable distance between the droplet ends and the domain boundaries. Unless otherwise specified, the dimensions of the symmetric computational domain are given as···

1.2 Research Methods

This study employs the lattice Boltzmann color model to solve two-phase flow. In this model, different colors distinguish different fluids, with interfacial interactions implemented through a color gradient. Interfacial tension is modeled using the continuum surface force model by Brackbill et al. and incorporated via Guo et al.'s volume force scheme. However, this treatment does not guarantee immiscibility. To promote phase separation between the droplet and carrier fluid and maintain a sharp interface, the recoloring algorithm proposed by Latva-Kokko and Rothman is adopted, which preserves interface isotropy while effectively reducing spurious velocities at the phase interface. For enhanced numerical stability, the multiple-relaxation-time (MRT) model is used for the collision term. In the computational domain shown in [Figure 1: see original paper], the upper boundary implements a half-way bounce-back no-slip condition, the lower boundary uses a symmetry condition, and the left and right boundaries are periodic.

Based on the electrohydrodynamics research by Melcher and Taylor and Saville et al., since the dynamic current at the droplet surface is small and magnetic effects are negligible, the electric field strength $\mathbf{E}$ is irrotational ($\nabla \times \mathbf{E} = 0$) and can be expressed as the gradient of a potential: $\mathbf{E} = -\nabla\Phi$. The fluid system is described by the leaky dielectric model, which assumes the droplet has finite conductivity. Since charge accumulation at the interface occurs much faster than fluid motion timescales, the charge conservation equation can be quasi-statically simplified as $\nabla \cdot \mathbf{J} = 0$, where $\mathbf{J} = \sigma\mathbf{E}$ is the current density. Substituting $\mathbf{E} = -\nabla\Phi$ into the charge conservation equation yields:

$$\nabla \cdot (\sigma\nabla\Phi) = 0$$

For incompressible fluids, the electric stress $\mathbf{F}_e$ acting at the phase interface can be expressed as:

$$\mathbf{F}_e = \nabla \cdot \mathbf{M} = \nabla \cdot \left(\varepsilon\mathbf{E}\mathbf{E} - \frac{1}{2}\varepsilon E^2\mathbf{I}\right) = q\mathbf{E} - \frac{1}{2}E^2\nabla\varepsilon$$

where $\mathbf{M}$ is the Maxwell stress tensor and q is the volume charge density at the interface. In the implementation, equation (6) is first discretized and solved using the finite difference method to obtain the electric potential, then the electric field strength is obtained through equation (4), and finally the electric stress acting at the phase interface is calculated through equation (7) and coupled with the lattice Boltzmann color model.

2 Model Validation

The results demonstrate that the numerical model exhibits good numerical accuracy and stability in simulating small droplet deformation under steady electric fields.

This section validates the developed model by simulating the influence of electric capillary number Ca_E on droplet deformation and comparing with the theoretical solution proposed by Feng et al. Without loss of generality, three typical (R, Q) values are considered: $(1.75, 3.5)$ with $f_d = -4.69 < 0$, $(4.75, 3.5)$ with $f_d = 4.31 > 0$, and $(3.25, 3.5)$ with $f_d = 17.81 > 0$. For each (R, Q) pair, the variation of droplet deformation parameter D with electric capillary number Ca_E (where $0 \leq Ca_E \leq 0.5$) is investigated.

[Figure 2: see original paper] shows the deformation parameter D as a function of Ca_E for different (R, Q) combinations. The solid lines represent the theoretical solution from Feng et al., while the symbols denote the simulation results. [Figure 3: see original paper] displays the steady droplet shapes, streamlines (left half), and velocity vector distributions (right half) under four parameter conditions: (a) $R = 1.75$, $Q = 3.5$, $Ca_E = 0.5$; (b) $R = 1.75$, $Q = 3.5$, $Ca_E = 1.0$; (c) $R = 4.75$, $Q = 3.5$, $Ca_E = 0.5$; (d) $R = 4.75$, $Q = 3.5$, $Ca_E = 1.0$, where the blue lines represent the droplet interfaces.

3 Results and Discussion

3.1 Oblate Droplet Deformation and Breakup

As shown in [Figure 2: see original paper], for any given (R, Q) pair, the degree of droplet deformation (oblate or prolate) increases with increasing electric capillary number Ca_E. When $Ca_E < 0.3$, the simulation results agree well with the theoretical solution. However, when $Ca_E > 0.3$, for $R = 1.75$ or $R = 4.75$, the droplet deformation becomes large and no longer satisfies the small deformation assumption employed in the theoretical derivation, causing the simulation results to deviate from the theoretical solution, with the deviation increasing as Ca_E increases. Hu et al. reached similar conclusions in their numerical simulations of three-dimensional droplet deformation. [Figure 3: see original paper] presents the steady-state droplet deformation, streamlines, and velocity vectors under four parameter conditions, revealing that the circulation directions inside oblate and prolate droplets are opposite.

When the indicator function $f_d < 0$, the droplet undergoes oblate deformation, flattening perpendicular to the electric field direction. As the electric capillary number Ca_E increases further, droplet breakup may occur. [Figure 4: see original paper] shows the variation of droplet deformation parameter D with Ca_E for $R = 0.1$, $Q = 2.0$. The negative values of D indicate deformation perpendicular to the electric field intensity, with the magnitude increasing as Ca_E increases. Based on the capillary number, four distinct droplet morphologies are observed: elliptical, quasi-elliptical, dumbbell-shaped, and broken. When $Ca_E \leq 0.04$, the steady-state droplet is elliptical, with simulation results matching Feng et al.' s theoretical solution well. For $0.04 < Ca_E < 0.14$, the steady-state droplet becomes quasi-elliptical, and simulations begin to deviate from the theoretical solution (as the large deformation violates the small deformation assumption). When $0.14 \leq Ca_E < 0.17$, the droplet' s major axis continues to increase while the minor axis decreases, forming a horizontally oriented dumbbell shape. Notably, around $Ca_E = 0.14$, the deformation parameter D exhibits a sudden change where the electric stress at the droplet surface can no longer be balanced by the viscous stress generated by internal circulation. The droplet stretches into a dumbbell shape with enlarged ends and a flattened middle until a new equilibrium is reached between electric forces and flow shear stresses. [Figure 5: see original paper] shows the steady droplet shapes, streamlines (left), and velocity vectors (right) for $Ca_E = 0.139$ and $Ca_E = 0.168$, where the droplet aspect ratio exceeds 7. The horizontal length increases with electric field strength, and the deformation parameter D approaches -1.

When the electric capillary number increases to approximately $Ca_E = 0.17$, the droplet breaks up under the steady electric field. As shown in [Figure 6: see original paper], under electric forces, the droplet's middle section contracts while the entire droplet stretches horizontally into a dumbbell shape. During breakup, the droplet does not fracture at the vertical central axis but rather at the necks of the left and right dumbbell ends. The breakup produces two symmetric large droplets and one small central droplet. The small middle droplet, dominated by interfacial tension, retracts into an elliptical shape, while the two large side droplets stretch into asymmetric dumbbell shapes under the electric field.

3.2 Prolate Droplet Deformation and Breakup

When the indicator function $f_d > 0$, the droplet undergoes prolate deformation, stretching along the electric field direction. This section investigates prolate droplet deformation and breakup under various electric capillary numbers for $R = 10$, $Q = 0.2$.

Simulations reveal four distinct morphologies as Ca_E increases from zero: elliptical, quasi-elliptical, droplet oscillation, and breakup. As shown in [Figure 7: see original paper], at small Ca_E ($Ca_E \leq 0.2$), droplet deformation is minor, stretching into an elliptical shape along the electric field direction, consistent with theoretical predictions. As Ca_E increases, deformation grows and gradually deviates from elliptical (quasi-elliptical droplet). [Figure 8: see original paper] shows that for $0.6 \leq Ca_E \leq 0.88$, the deformation parameter D exhibits damped oscillations, eventually reaching equilibrium after some time, with larger Ca_E producing higher peak values of D and longer times to reach steady state. As Ca_E continues to increase ($Ca_E > 0.88$), prolate droplets exhibit behavior distinctly different from oblate droplets: they undergo periodic oscillations under steady electric fields, with oscillation frequency and amplitude remaining constant over time. This phenomenon has not been reported in previous studies. When periodic oscillations occur, increasing Ca_E raises the average value of D, slightly increases oscillation frequency, and decreases amplitude.

A stationary droplet placed in a steady electric field develops induced charges at its interface. The energy supplied by the electric field converts into kinetic energy of fluid at the droplet interface, gradually elongating the droplet. As the droplet stretches along the electric field direction, the induced charge density generated by the steady field decreases, weakening the electric force. Simultaneously, as deformation increases, interfacial tension grows. During elongation, there exists a moment when interfacial tension exceeds the electric force, and the droplet' s kinetic energy dissipates due to internal and external circulation. When kinetic energy dissipates to zero, the deformation parameter reaches its maximum (time t_1 in [Figure 9: see original paper]). Subsequently, the droplet contracts under interfacial tension; as it shrinks, interfacial tension decreases while induced surface charges and electric force increase. When kinetic energy again reaches zero, the deformation parameter D reaches its minimum, and the droplet elongates again under electric force, repeating the cycle. Thus, within a certain range of Ca_E, droplets undergo periodic oscillations under the combined action of electric force and interfacial tension.

[Figure 9: see original paper] illustrates droplet oscillatory deformation at $Ca_E = 1.00$, with [FIGURE:9(a)] showing the droplet interface shapes at five equally spaced moments within half an oscillation period (see [FIGURE:9(b)]). The droplet remains in a dumbbell shape from t_1 to t_4, much longer than the time spent in an elliptical shape (t_5).

When the electric capillary number increases sufficiently ($Ca_E^B > 2.24$), the droplet breaks up under the steady electric field. For prolate droplets, breakup occurs when the droplet is stretched along the electric field direction, the horizontal forces are balanced, and the droplet interface width on the horizontal symmetry axis approaches zero, as shown in [Figure 10: see original paper]. The droplet breaks into three sub-droplets: a small residual droplet at the central axis and two equal larger portions that stretch into elongated shapes and move along the electric field direction. Notably, the deformation parameter D at breakup is approximately 0.99 (corresponding to an aspect ratio of about 100:1), and this parameter (sub-droplet size) remains nearly unchanged with increasing Ca_E, while the time required for breakup decreases as Ca_E increases.

Based on the above discussion, for prolate droplets we can conclude that, at constant dielectric constant ratio Q and conductivity ratio R, there exist two critical electric capillary numbers: Ca_E^V, which distinguishes between steady deformation and periodic oscillation, and Ca_E^B, the minimum capillary number for droplet breakup. For the present case ($R = 10$, $Q = 0.2$), the oscillation critical capillary number Ca_E^V lies in the interval $(0.87, 0.89)$, and the breakup critical capillary number Ca_E^B lies in the interval $(2.24, 2.26)$.

4 Conclusions

This study develops a hybrid numerical method based on the lattice Boltzmann color model for two-phase flow, finite difference method for electric field calculation, and coupling through the leaky dielectric model to investigate droplet deformation and breakup under steady electric fields. Numerical investigations of droplet deformation in different stretching directions reveal that deformation (oblate or prolate) increases with electric capillary number for both cases. At small Ca_E ($Ca_E < 0.3$), the simulation results agree perfectly with Feng et al.' s theoretical solution. At larger Ca_E, deviations from the theoretical solution appear because the large deformation violates the small deformation assumption, with the deviation increasing as Ca_E increases.

The influence of electric capillary number on droplet deformation and breakup phenomena is also investigated. For oblate droplets ($f_d < 0$, $R = 0.1$, $Q = 2.0$) that stretch perpendicular to the electric field direction, four distinct morphologies emerge with increasing Ca_E: elliptical ($Ca_E \leq 0.04$), quasi-elliptical ($0.04 < Ca_E < 0.14$), dumbbell-shaped ($0.14 \leq Ca_E < 0.17$), and breakup (critical breakup capillary number $Ca_E \approx 0.17$). For prolate droplets ($f_d > 0$, $R = 10$, $Q = 0.2$) that stretch along the electric field direction, the deformation parameter D varies significantly differently from oblate droplets, exhibiting elliptical ($Ca_E \leq 0.2$), quasi-elliptical ($0.2 < Ca_E \leq 0.88$), periodic oscillation ($0.88 < Ca_E \leq 2.24$), and breakup ($Ca_E^B > 2.24$) morphologies. During the deformation stage, the deformation parameter D for both oblate and prolate droplets increases with Ca_E. During breakup, the time required for breakup decreases as Ca_E increases for both deformation modes.

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Note: Figure translations are in progress. See original paper for figures.