On the formulations of interfacial force in the phase-field-based lattice Boltzmann method

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Abstract
Different formulations of interfacial force have been adopted in the existing phase-field-based lattice Boltzmann method for two-phase flows. Although they are identical mathematically, their numerical performances may be different due to truncation errors at discrete level. In this article, four-type formulations of interfacial force available in the literature, namely, stress tensor form (STF), chemical potential form (CPF), pressure form, and continuum surface force (CSF) form, are summarized and theoretical analyzed. A systematic study of the performances of all formulations is made by a series of benchmark problems, including stationary droplet, two merging droplets, Capillary wave, rising bubble and drop deformation in shear flow. Numerical results show that CPF is a good choice for small surface deformation problems while STF is preferred for dynamical problems, both STF and CSF presents better numerical stability in terms of Peclet number.

KEYWORDS
Cahn–Hilliard, interfacial force, lattice Boltzmann, multiphase flows, Navier–Stokes, phase field, two-phase flow

1 | INTRODUCTION

Multiphase flows are ubiquitous in both natural processes and industrial applications, such as droplet dynamics,1 lab-on-chip devices,2 surfactant behavior,3 underground water flows,4 and enhanced oil recovery.5 A number of numerical methods have been developed for simulating such flows, which can be divided into two categories, that is, interface tracking approach and interface capturing approach. In the former, interfaces are explicitly tracked, such as the marker and cell method6 and front-tracking method.7 In the latter, interfaces are implicitly tracked and an interface function that marks the location of the interface is governed by the advection (diffusion) equations, such as volume of fluid (VOF) method,8 level set (LS) method,9 and phase field method.10

Among these methods, the phase field method is an increasingly popular choice for multiphase flow simulations. The basic idea is to introduce a so-called order parameter that has distinct values in the bulk phases but varies smoothly over the interfacial region. The order parameter defined as the volume fraction or mass fraction is usually governed by the phase field equations, including the Cahn–Hilliard equation or the Allen–Cahn equation, which leads to the Navier–Stokes–Cahn–Hilliard (NSCH) or the Navier–Stokes–Allen–Cahn (NSAC) system. If the fluid density is taken as an order parameter, the two-phase flow can be described by the Navier–Stokes–Kortweg (NSK) system.11,12 In the traditional computational fluid dynamics (CFD), many discretization methods have been developed to numerically solve the
above governing equations. Recently, the lattice Boltzmann method (LBM) has grown as an alternative tool for multiphase flow simulations due to its merits.\textsuperscript{13-16} The LBM is regarded as a mesoscopic method from the kinetic theory of gases. In LBM, the fluid is represented by a discrete set of particle distribution functions which only perform propagation along fixed directions and collision processes on fixed lattice. The macroscopic quantities (mass, momentum and energy) of the flow are defined as the moments of the particle distribution functions. Although the governing equation of the distribution functions is linear compared with the traditional CFD to discretize the nonlinear equations, the macroscopic phase field equation and hydrodynamic equations can be fully recovered from LBM through the multiscale analysis.

In computational methods for multiphase flows, approximating the surface tension force accurately is critical to capture correct flow behaviors. To do this, a number of mathematical models for the interfacial force have been developed. In the framework of phase field model for multiphase flow, the interfacial force can be strictly derived based on the entropy principle of rational thermodynamics.\textsuperscript{10,17-19} The resulting interfacial force appears as a gradient of the stress tensor of the order parameter in the modified momentum equation. This type of interfacial force is called stress tensor form (STF). The stress form can be further simplified by absorbing the square of gradient terms into the pressure term. Then, the interfacial force can be expressed as the forms dependent on the gradients of the order parameter,\textsuperscript{20-24} which can be called pressure form (PF). If the chemical potential related to the order parameter is employed, the interfacial force can also be expressed as the forms dependent on the chemical potential,\textsuperscript{25-28} which is usually called chemical potential form (CPF). Mathematically, STF, PF, and CPF are equivalent. In addition, in the VOF and LS methods, the continuum surface force (CSF) model of Brackbill et al.\textsuperscript{29} that is treated as a volumetric force proportional to the normal vector and curvature of the interface and a surface Dirac function has been widely used. Inspired by the CSF model, Kim\textsuperscript{30} proposed a CSF type interfacial force for phase field method. The basic idea is to replace the LS by the order parameter and take the square of gradient of the order parameter as the surface Dirac function. An advantage of the CSF formulation is that the pressure field can be calculated explicitly. The surface Dirac function in CSF model that has an important effect on the performance of CSF can also be defined in other ways. For instance, Lee and Kim\textsuperscript{31} compared various types of surface Dirac functions in the CSF model. They argued that the absolute value of the gradient of the order parameter has the best performances in their considered numerical experiments. These formulations are named as CSF form of the interfacial force.

Although the above interfacial force formulations are mathematically equivalent, the performance of each formulation may be different in practical computations. To clarify this issue, the performances of several formulations have been compared by simulating the stationary droplet. For example, Lee and Fischer\textsuperscript{20} compared the parasitic currents predicted by PF and CPF, and the results showed that potential form yielded much smaller parasitic currents. Chao et al.\textsuperscript{32} compared the interface force distribution of PF and the CSF, and the results showed that the PF could generate wiggles over the interface region while the CSF produced smooth profiles. These comparisons are helpful for researches to carry out their calculations. However, a systematic study of the performances of all these interfacial force formulations is lacking so far. In addition, both advantages and disadvantages of different interfacial force formulations are unclear for the dynamic interface problems. In this article, we will focus on this topic. Specifically, the present work will make the following contributions in addition to previous studies. First, the STF, PF, and CSF form of the surface tension force are summarized except for the familiar CPF. Second, in addition to simulate static droplet, the simulations of the moving interface problems are considered. Third, a comprehensive comparison for the interfacial force formulations will be made.

Furthermore, it is noted that in the phase field model for multiphase flows, both the NSCH and NSAC equations can be used for multicomponent two-phase flow, and the NSK can be used for one component two phase flow. Although the interface-capturing equation in each system is different, the formulations of the interfacial force are similar. If the distributions of the order parameter and density predicted by different interface-capturing equations are same, the performance of each interfacial force formulation should be consistent in different systems. Here, we focus on the performances of interfacial force itself in the momentum equation, the NSCH system with different formulations of interfacial force is used as a demonstration. Comparisons of different interface tracking equations have been discussed in References 33,34, which is beyond the scope of this work.

The article is organized as follows. In Section 2, the governing equations of the phase field model for binary fluids are presented and the formulas of interfacial force are summarized in detail. The phase-field-based LBM is briefly introduced in Section 3. In Section 4, several benchmark problems with theoretical solutions are simulated and the results are compared. Finally, conclusions are drawn in Section 5.
2 | MATHEMATICAL FORMULATION

2.1 | Governing equations

In the NSCH system, the Cahn–Hilliard equation is expressed as\(^{10,35}\)

\[ \frac{\partial \phi}{\partial t} + \nabla \cdot (\phi \mathbf{u}) = \nabla \cdot (M \nabla \mu_\phi), \tag{1} \]

where \( \phi \) is the order parameter to identify different phases, \( M \) is the mobility, \( \mu_\phi \) is the chemical potential that is defined as

\[ \mu_\phi \equiv \frac{\delta \psi}{\delta \phi} = \frac{\partial f_0}{\partial \phi} - \kappa \nabla^2 \phi, \tag{2} \]

where \( \psi \) is the system free energy,

\[ \psi = \int_V \left[ f_0(\phi) + \frac{\kappa}{2} |\nabla \phi|^2 \right] dV, \tag{3} \]

where \( f_0 = \beta (1 - \phi^2)^2 \) is the bulk energy density, the second term in parentheses is the interface energy density, \( \beta \) and \( \kappa \) are determined by the surface tension \( \sigma \) and the interface width \( W \).

For a plane interface at equilibrium, the equilibrium profile for the order parameter can be obtained by solving \( \mu_\phi = 0 \),

\[ \phi(r) = \tanh \left( \sqrt{\frac{2\beta}{\kappa}} r \right), \tag{4} \]

where \( r \) is the signed distance coordinate normal to the interface. \( \sqrt{\kappa/2\beta} \) has a length scale of interface thickness. As the surface tension is interpreted as energy per unit surface area, the surface tension for a flat interface with equilibrium profile can be calculated by

\[ \sigma = \int_{-\infty}^{+\infty} \left( f_0(\phi) + \frac{\kappa}{2} |\nabla \phi|^2 \right) dr \]
\[ = \kappa \int_{-\infty}^{+\infty} |\nabla \phi|^2 dx = \frac{4}{3} \sqrt{2\beta} \kappa. \tag{5} \]

In Reference 10, \( \sqrt{\kappa/2\beta} \) is defined as \( W/2 \), which leads to

\[ \beta = \frac{3}{4} \frac{\sigma}{W}, \quad \kappa = \frac{3}{8} W \sigma. \tag{6} \]

The dynamics of a fluid mixture of two incompressible viscous fluids can be described by the Navier–Stokes equations with interfacial force\(^{10,27}\)

\[ \nabla \cdot \mathbf{u} = 0, \tag{7} \]

\[ \frac{\partial (\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u} \mathbf{u}) = -\nabla P_{sf} + \nabla \cdot (\mu (\nabla \mathbf{u} + (\nabla \mathbf{u})^T)) + \mathbf{F}_g + \mathbf{F}_{sf}, \tag{8} \]

where \( \rho \) is the fluid density, \( \mathbf{u} \) is the flow velocity, \( P_{sf} \) is the generalized pressure dependent on the definition of the interfacial force, \( \mu \) is the dynamic viscosity, \( \mathbf{F}_g = (\rho - \rho_0) \mathbf{g} \) is the gravitational force with \( \mathbf{g} \) being the gravitational acceleration and \( \rho_0 \) being the background density, \( \mathbf{F}_{sf} \) is the interfacial force. The subscript \( sf(=stf, cpf, pf, csf) \) denotes different formulations of interfacial force.

The mixture density \( \rho \) and viscosity \( \mu \) can be given by

\[ \rho = \rho_1 \frac{1 + \phi}{2} + \rho_2 \frac{1 - \phi}{2}, \tag{9} \]
\[ \mu = \mu_1 \frac{1 + \phi}{2} + \mu_2 \frac{1 - \phi}{2}, \]  
(10)

where the subscripts 1 and 2 indicate fluid 1 and fluid 2.

To nondimensionalize the equations in NSCH system, the following dimensionless variables are used,

\[ u' = \frac{u}{U_c}, \quad x' = \frac{x}{L_c}, \quad t' = \frac{t}{T_c}, \quad p' = \frac{P_{sf}}{p_c}, \quad \mu' = \frac{\mu_\phi}{\mu_{\phi,c}}, \quad F'_{sf} = \frac{F_{sf}L_c^2}{\sigma}, \]  
(11)

where \( U_c, L_c, T_c (= L_c/U_c), p_c (= \rho_c U_c^2), \mu_{\phi,c} (= 4\beta) \) are, respectively, the reference velocity, length, time, pressure, and chemical potential. In this article, the density and dynamical viscosity of fluid 1 are chosen as the reference quantities, that is, \( \rho_c = \rho_1, \mu_c = \mu_1 \). With the above variables and dropping the primes, the resulting dimensionless governing equations can be written as

\[ \partial_t \phi + \nabla \cdot (\phi u) = \frac{1}{Pe} \nabla \cdot (M \nabla \mu_\phi), \]  
(12)

\[ \partial_t (\rho u) + \nabla \cdot (\rho uu) = -\nabla P_{sf} + \frac{1}{Re} \nabla \cdot (\mu (\nabla u + \nabla u^T)) + \frac{1}{We} F_{sf} + \frac{1}{Fr^2} F_g, \]  
(13)

\[ \nabla \cdot u = 0, \]  
(14)

with

\[ \mu_\phi = \phi(f^2 - 1) - \frac{Cn^2}{8} \nabla^2 \phi, \]

\[ \rho = \frac{1 + \phi}{2} + \frac{1 - \phi}{2} \rho_2, \]

\[ \mu = \frac{1 + \phi}{2} + \frac{1 - \phi}{2} \mu_2. \]  
(15)

The dimensionless groups used above are the Reynolds number \( Re \), Peclet number \( Pe \), Weber number \( We \), Froude number \( Fr \), and Cahn number \( Cn \), which are, respectively, defined by

\[ Re = \frac{\rho_c U_c L_c}{\mu_c}, \quad Pe = \frac{U_c L_c}{4M\beta}, \quad We = \frac{\rho_c L_c U_c^2}{\sigma}, \quad Fr = \frac{U_c}{\sqrt{gL_c}}, \quad Cn = W L_c. \]  
(16)

2.2 | Interfacial force formulations

Based on the energetic variational approach or the free energy inequality, the surface tension force in the momentum equation can be defined as \(^{18,36,37}\)

\[ F_{sf-1} = -\nabla \cdot \kappa (\nabla \phi \otimes \nabla \phi), \]  
(17)

where \( \nabla \phi \otimes \nabla \phi \) is the usual tensor product and denotes the induced elastic stress due to the mixing of the different fluids. In this case, the generalized pressure \( P_{sf} \) in Equation (8) includes both the hydrostatic pressure \( p_h \) due to the incompressibility and the contributions from the induced stress, \( P_{sf-1} = p_h + \kappa |\nabla \phi|^2 \). In References 38,39, the surface tension force term is defined as

\[ F_{sf-2} = \nabla \cdot \kappa (|\nabla \phi|^2 I - \nabla \phi \otimes \nabla \phi), \]  
(18)

which implies that the principle axes of the tensor are perpendicular to the tangent plane of the interface. The normal stress perpendicular to the tangent plane of the interface is zero and the two tangent normal stresses are equal. In this case, the generalized pressure in Equation (8) becomes the true pressure, namely, \( P_{sf-2} = p_h \). \(^{10,39}\)
For simplicity, we assume that the surface tension \( \sigma \) is constant. By using the following identity
\[
\kappa \nabla \cdot (\nabla \phi \otimes \nabla \phi) = \frac{\kappa}{2} \nabla |\nabla \phi|^2 + \kappa \nabla \phi \nabla \phi
\]

\[
= \nabla \left( \frac{\kappa}{2} |\nabla \phi|^2 + \kappa \phi \Delta \phi \right) - \kappa \phi \nabla \phi
\]

\[
= \nabla \left( \frac{\kappa}{2} |\nabla \phi|^2 + f_0 \right) - \mu \phi \nabla \phi
\]

\[
= \nabla \left( \frac{\kappa}{2} |\nabla \phi|^2 + f_0 - \phi \mu \phi \right) + \phi \nabla \mu \phi.
\] (19)

and absorbing the gradient terms into pressure \( p_h \), the surface tension force can be reformulated as follows

\[
F_{cpf-1} = -\phi \nabla \mu \phi, \quad F_{cpf-2} = \mu \phi \nabla \phi,
\]

\[
F_{pf-1} = -\kappa \nabla \phi \Delta \phi, \quad F_{pf-2} = \kappa \phi \nabla \Delta \phi.
\] (20)

The corresponding generalized pressure is redefined as

\[
P_{cpf-1} = p_h + f_0 - \phi \mu \phi - \frac{\kappa}{2} |\nabla \phi|^2,
\]

\[
P_{cpf-2} = p_h + f_0 - \frac{\kappa}{2} |\nabla \phi|^2,
\]

\[
P_{pf-1} = p_h - \frac{\kappa}{2} |\nabla \phi|^2,
\]

\[
P_{pf-2} = p_h + \kappa \phi \Delta \phi - \frac{\kappa}{2} |\nabla \phi|^2.
\] (21)

\( F_{cpf-1} \) and \( F_{cpf-2} \) are CPF. \( F_{pf-1} \) and \( F_{pf-2} \) are the PF. It is noted that \( F_{stf-1} \) is used in References 40,41 and \( F_{stf-2} \) is used in References 38,42-44, \( F_{cpf-1} \) is used in References 25,45,46 and \( F_{cpf-2} \) is used in References 13,26-28, \( F_{pf-1} \) is used in References 23,24 and \( F_{pf-2} \) is used in References 21,22.

Based on the CSF model, the surface tension force is defined as

\[
F_{csf} = \sigma \tilde{\kappa} \delta_s \mathbf{n},
\] (22)

where \( \mathbf{n} \) is the unit normal vector, \( \tilde{\kappa} = -\nabla \cdot \mathbf{n} \) is the local mean curvature, \( \delta_s \) is the surface Dirac function used to ensure the force acting on the interfacial region. To match the surface tension of the sharp interface model, the Dirac function should satisfy

\[
\int_{-\infty}^{\infty} \delta_s dr = 1.
\] (23)

There are many possible choices for \( \delta_s \). Kim\textsuperscript{30} used \( a|\nabla \phi|^2 \) as the Dirac function with \( a = 3W/8 \), leading to

\[
F_{csf-1} = -\kappa \nabla \phi |\nabla \phi| \nabla \cdot \mathbf{n}.
\] (24)

Lee and Kim\textsuperscript{31} treated \( a|\nabla \phi| \) as the Dirac function with \( a = 0.5 \),

\[
F_{csf-2} = -\frac{\sigma}{2} \nabla \phi (\nabla \cdot \mathbf{n}).
\] (25)

The derivation of \( a \) is referred to Appendix.B1. In the above interfacial force formulations, Equations (17), (18), and (20) are identical mathematically. In fact, these formulations can be rewritten as

\[
F_{stf-1} = F_{csf-1} - \left[ \frac{\kappa |\nabla \phi|^2}{2} + \frac{\kappa \nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{|\nabla \phi|} \right],
\]

\[
F_{stf-2} = F_{csf-1} - \left[ -\frac{\kappa |\nabla \phi|^2}{2} + \frac{\kappa \nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{|\nabla \phi|} \right],
\]
\[ F_{\text{cf} - 1} = F_{\text{cf} - 1} - \left[ \nabla (\phi \mu \phi) - \nabla f_0 + \frac{\kappa \nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{|\nabla \phi|} \right]. \]
\[ F_{\text{cf} - 2} = F_{\text{cf} - 1} - \left[ -\nabla f_0 + \frac{\kappa \nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{|\nabla \phi|} \right], \]
\[ F_{\text{pf} - 1} = F_{\text{cf} - 1} - \frac{\kappa \nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{|\nabla \phi|}, \]
\[ F_{\text{pf} - 2} = F_{\text{cf} - 1} - \left[ -\nabla (\kappa \phi \Delta \phi) + \frac{\kappa \nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{|\nabla \phi|} \right]. \tag{26} \]

It is clear that there are some gradient terms in \( F_{\text{sf}} \), \( F_{\text{cf}} \), and \( F_{\text{pf}} \). This is why the previous formulations cannot be used to calculate the pressure field explicitly.\textsuperscript{30}

By using Equation (4), the following relations can be built,
\[ |\nabla \phi| = \frac{2}{W}(1 - \phi^2), \]
\[ \frac{\nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{|\nabla \phi|} = \frac{1}{2} \nabla |\nabla \phi|^2. \tag{27} \]

Inserting Equation (27) into Equation (26) leads to
\[ F_{\text{sf} - 1} = F_{\text{cf} - 1} - \nabla \kappa |\nabla \phi|^2, \]
\[ F_{\text{cf} - 1} = F_{\text{cf} - 1} - \nabla (\phi \mu \phi), \]
\[ F_{\text{pf} - 1} = F_{\text{cf} - 1} - \frac{\nabla \kappa}{2} |\nabla \phi|^2, \]
\[ F_{\text{pf} - 2} = F_{\text{cf} - 1} - \nabla (-\kappa \phi \Delta \phi + \frac{\kappa}{2} |\nabla \phi|^2), \]
\[ F_{\text{sf} - 2} = F_{\text{cf} - 2} = F_{\text{cf} - 1}. \tag{28} \]

Therefore, \( F_{\text{cf} - 1} \), \( F_{\text{sf} - 2} \), and \( F_{\text{cf} - 2} \) are identical when the system is at equilibrium. The main difference between \( F_{\text{cf} - 1} \) and \( F_{\text{cf} - 2} \) is the definition of the Dirac delta function. All above formulations have been used to mimic the interfacial force in the phase-field-based LBM. In Section 5, the performance of the LBM with the above eight formulations of interfacial force will be compared.

### 3 PHASE-FIELD-BASED LBM

We adopted the multiphase LBM of He et al.\textsuperscript{21} for the hydrodynamic equations and the improved LBM of Zhang et al.\textsuperscript{48} for Cahn–Hilliard equation. The evolutions of the distribution functions \( f_i \) and \( h_i \) are, respectively, expressed as
\[ f_i(x + \Delta t, t + \delta t) - f_i(x, t) = -\frac{1}{\tau_f}[f_i(x, t) - f_i^\text{eq}(x, t)] + \delta t \left( 1 - \frac{1}{2\tau_f} \right) F_i, \tag{29} \]
\[ h_i(x + \Delta t, t + \delta t) - h_i(x, t) = -\frac{1}{\tau_h}[h_i(x, t) - h_i^\text{eq}(x, t)] + \delta t \left( 1 - \frac{1}{2\tau_h} \right) H_i, \tag{30} \]
where \( f_i(x, t) \) and \( h_i(x, t) \) are the distribution functions for the hydrodynamics and order parameter fields, respectively, \( \Delta \) is the discrete velocity in the \( i \)th direction, \( \delta t \) is the time step, \( \tau_f \) and \( \tau_h \) are the dimensionless relaxation times related to the shear viscosity and mobility, respectively, \( F_i \) and \( H_i \) are the discrete force terms. To recover the correct governing equations, the equilibrium distributions \( f_i^\text{eq} \) and \( h_i^\text{eq} \) are respectively defined as
\[ f_i^\text{eq} = \phi_i [P_{\text{sf}} + \rho \phi_i (\mathbf{u})] \tag{31} \]
\[ h_i^\text{eq} = \begin{cases} \phi + (\phi_0 - 1) \eta \mu \phi_i, & i = 0 \\ \eta \phi_i \phi_i \phi_i + \phi_i \phi_i, & i \neq 0 \end{cases} \tag{32} \]
with

\[
s_i(u) = \frac{c_i \cdot u}{c_i^2} + \frac{uu : (c_i c_i - c_i^2 I)}{2c_i^4},
\]

(33)

where \( \omega_i \) is the weighting coefficient corresponding to the discrete velocity \( c_i, c_s = c/\sqrt{3} \) is the lattice sound speed, \( c = \partial x/\partial t \) is the lattice speed with \( \partial x \) being the lattice length scale, and \( \eta \) is an adjustable parameter for the mobility. In this work, the two-dimensional nine-velocity (D2Q9) model is used in which the discrete velocity is

\[
(c_0, c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8) = c \begin{pmatrix}
0 & 1 & 0 & -1 & 0 & 1 & -1 & -1 & 1 \\
0 & 0 & 1 & 0 & -1 & 1 & 1 & -1 & -1
\end{pmatrix}
\]

(34)

and the corresponding weighting coefficients are \( \omega_0 = 4/9, \omega_{1-4} = 1/9, \) and \( \omega_{5-8} = 1/36. \)

The force terms \( F_i \) and \( H_i \) are given by

\[
F_i = (c_i - u) \cdot \left[ \Gamma_i(u)(F_{sf} + F_g) + s_i(u)\nabla c_i^2 \rho \right]
\]

(35)

\[
H_i = \bar{\omega}_i \frac{3\tau_2}{\tau_0 \tau_1 \delta t} \nabla \cdot (u \phi) + \omega_i \frac{c_i \cdot \partial_i(\phi u)}{c_i^2}.
\]

(36)

where \( \Gamma(u) = \omega_i + s_i(u) \), \( \tau_2 = (\tau_h^2 - \tau_h + \frac{1}{6})\delta t^2 \), \( \tau_1 = (\tau_h - 0.5)\delta t \) and \( \bar{\omega}_i \) is a new weight coefficient and satisfies \( \sum_i \bar{\omega}_i = \sum_i \omega_i c_i = 0 \). \( \sum_i \bar{\omega}_i c_i = c_i^2 I \). In particular, if \( \tau_g = 0.5 + \sqrt{3}/6 \), the above improved LBM for CH can be simplified to the one of Liang et al. \(^{13} \)

However, the relationship between the Peclet number and Cahn number should be \( \text{Pe} \sim \text{Cn}^{-1} \) to achieve the sharp-interface limit with continuous mesh refinement. \(^{49} \) Then, the relaxation time may have a value except the optimum one in some situations.

The macroscopic quantities are calculated by

\[
P_{sf} = \sum_i f_i + \frac{\delta t}{2} c_i^2 u \cdot \nabla \rho,
\]

\[
u = \frac{1}{c_i^2 \rho} \left[ \sum_i c_i f_i + \frac{\delta t}{2} c_i^2 (F_{sf} + F_g) \right]
\]

\[
\phi = \sum_i h_i.
\]

(37)

Through the Chapman–Enskog expansion, the macroscopic governing equations recovered from the above LBM are

\[
\frac{1}{c_i^2 \rho} \partial_t P_{sf} + \nabla \cdot u = 0,
\]

(38)

\[
\partial_t \left( \rho \nabla \left( \rho uu \right) \right) = -\nabla P_{sf} + \nabla \cdot \nabla (\nabla \rho + \nabla u^T) + F_{sf} + F_g,
\]

(39)

\[
\partial_t \phi + \nabla \cdot (\phi u) = \nabla \cdot M \nabla \mu \phi,
\]

(40)

where the viscosity \( \mu \) and the mobility \( M \) are defined as \( \mu = \rho c_i^2 (\tau_f - 0.5)\delta t \) and \( M = c_i^2 \eta (\tau_h - 0.5)\delta t \), respectively.

### 4 | BOUNDARY CONDITIONS

Boundary treatment is one of the most important tasks in numerical methods. In LBM, the classical boundary condition to model no-slip walls is the bounce-back method, which can be realized by both the full-way bounce-back and the half-way bounce-back. \(^{50} \) As the half-way bounce-back can be implemented without solid nodes and is more accurate for unsteady flows, we will carry out the half-way bounce-back in simulations. As shown in Figure 1, following Ladd’s
half-way bounce-back scheme, the unknown distribution function is determined by\textsuperscript{14,51}

\[
\begin{align*}
f_+^c(x_f, t + \delta t) &= f^-_c(x_f, t) - 2\omega_i\rho(x_w, t) c_i \cdot \mathbf{u}_w, \\
h_+^c(x_f, t + \delta t) &= h^-_c(x_f, t) - 2\omega_i\phi(x_w, t) c_i \cdot \mathbf{u}_w,
\end{align*}
\tag{41}
\]

where \(f_+\) and \(h_+\) are the distribution functions with the velocity \(c_+ = -c_i\), the superscript “+” denotes the postcollision value of the corresponding distribution function and \(\mathbf{u}_w\) is the prescribed wall velocity. The calculation of the unknown distribution function \(h_+\) involves the order parameter \(\phi\) at the wall surface. Generally, it can be obtained by employing the wetting boundary conditions. In this work, the order parameter at the wall is calculated by enforcing \(\mathbf{n}_w \cdot \nabla \phi = 0\), which implies neutral wetting boundary condition. Then the density \(\rho(x_w, t)\) at the wall can be obtained based on Equation (9).

To maintain the mass conservation, the following boundary conditions is employed,

\[
\mathbf{n}_w \cdot \nabla \mu_\phi = 0, \tag{42}
\]

where \(\mathbf{n}_w\) is the unit outward normal defined at the solid boundary.

The gradient terms in each formulation of interfacial force can be calculated with different schemes.\textsuperscript{52,53} In the present work, the widely used second-order isotropic central scheme is employed,\textsuperscript{54}

\[
\begin{align*}
\nabla \Psi &= \frac{1}{c_2^2 \delta t} \sum_{i=1}^{8} \omega_i \mathbf{c}_i \Psi(x + \mathbf{c}_i \delta t), \tag{43}
\\
\nabla^2 \Psi &= \frac{2}{c_2^2 \delta t} \sum_{i=1}^{8} \omega_i \left[ \Psi(x + \mathbf{c}_i \delta t) - \Psi(x) \right]. \tag{44}
\end{align*}
\]

where \(\Psi\) denotes arbitrary quantity. For the node located at wall boundary, a second-order one-side finite difference is employed. As shown in Figure 1, we take the derivative of \(\phi\) with respect to \(x\) as an example.

\[
\left. \frac{\partial \phi}{\partial x} \right|_w = \frac{9\phi(x_f) - \phi(x_g) - 8\phi_w}{3}. \tag{45}
\]

To easy perform Equations (43) and (44), the ghost point \(x_s\) is assigned the value of \(\phi(x_s) = 2\phi_w - \phi(x_f)\).

## 5 NUMERICAL RESULTS AND DISCUSSION

In this section, the performance of each interfacial force formulation is validated by a series of benchmark tests, including stationary droplet, two merging droplets, capillary wave, rising bubble, and the deformation droplet in a shear flow. The first three tests focus on the performances of the force formulations in the absence of external force, while the last two tests consider problems under external forces. For each test, the results obtained by the LBM with different interfacial force formulations are compared with the theoretical solutions or the available reference solutions in the literature. In
Table 1 Comparison of numerical surface tension based on Laplace law
\((\sigma = 0.357)\)

| \(F_{sf} \) | 60 \times 60 | 120 \times 120 | 240 \times 240 |
|--------------|-------------|-------------|-------------|
| \(\sigma_{num} \) | \(\sigma_{exact} \) | \(\sigma_{num} \) | \(\sigma_{exact} \) |
| \(F_{sf-1} \) | 0.3401 | 0.3404 | 0.3404 | 0.3410 | 0.480 |
| \(F_{sf-2} \) | 0.3401 | 0.3404 | 0.3404 | 0.3410 | 0.484 |
| \(F_{pf-1} \) | 0.3505 | 1.833 | 0.3451 | 1.625 | 0.3519 | 1.418 |
| \(F_{pf-2} \) | 0.3507 | 1.765 | 0.3512 | 1.619 | 0.3520 | 1.404 |
| \(F_{pf-1} \) | 0.3512 | 1.629 | 0.3517 | 1.475 | 0.3524 | 1.277 |
| \(F_{pf-2} \) | 0.3289 | 7.877 | 0.3290 | 7.846 | 0.3296 | 7.688 |
| \(F_{csf-1} \) | 0.3404 | 4.657 | 0.3404 | 4.637 | 0.3410 | 4.475 |
| \(F_{csf-2} \) | 0.3599 | 0.825 | 0.3558 | 0.326 | 0.3566 | 0.108 |

Equation (36), the time derivative is calculated by explicit Euler scheme, and \(x_0 = \omega_0 - 1, \omega_i = \omega_i \) for \(i > 0 \). The Peclet number is set to be 1.0/Cn and the interface width is set to be four grids unless otherwise stated.

5.1 Stationary droplet

We first make a comparison among different interfacial force formulations by simulating a stationary droplet. Theoretically, the exact solution is zero velocity for all time. Initially, a circle droplet with radius \(R \) is placed at the center of the domain \(L \times L \). The order parameter is set to be

\[
\phi(x, y) = \tanh \left(2 \frac{R - \sqrt{(x - x_c)^2 + (y - y_c)^2}}{W}\right),
\]

(46)

where \((x_c, y_c)\) is the center coordinate of the droplet. Periodic boundary conditions are applied to all the boundaries. The initial velocity and pressure fields are set to be zero. The physical parameters are set to be \(L = 1 \text{ m}, R = 0.25 \text{ m}, \rho_1 = 4 \text{ kg/m}^3, \rho_2 = 1 \text{ kg/m}^3, v_1 = v_2 = 0.25 \text{ m}^2/\text{s}, \) and \(\sigma = 0.357 \text{ N/m} \). Three uniform grids of \(60 \times 60, 120 \times 120, 240 \times 240 \) are used. The characteristic velocity is \(U_c = \sigma / \mu_1 \).

We first examine the shape of the droplet at equilibrium. The interface profiles of the droplet obtained by all interfacial force formulations are similar and agree well with the initial interface profiles, and the results are not shown here. It is also found that the deviation between the numerical results given by all formulations and the analytical ones becomes small as the value of mobility decreases, which is also consistent with the results in Reference 13. Since the definition of characteristic velocity is artificial to some extent, the relationship of \(\text{Pe} \sim 1/Cn \) may be unable to produce the closest results to the exact one.

From the Laplace law, the numerical surface tension can be calculated by \(\sigma_{num} = R_{out} \times (p_{h, in} - p_{h, out}) \). The relative error, \(\text{Err} = |\sigma_{num} - \sigma_{exact}| / \sigma_{exact} \times 100\%\), is listed in Table 1. It can be seen that the error decreases as the grid resolution increases. For all meshes, \(F_{csf-2} \) gives the smallest error while \(F_{pf-2} \) gives the largest one.

The pressure field \(p_h \) on the \(240 \times 240 \) mesh is presented in Figure 2. It can be seen that the pressure inside the droplet is higher than that in the surrounding fluid. However, \(F_{sf-2}, F_{csf-1} \) and \(F_{csf-2} \) give smooth pressure field across the interface while the others give obvious oscillation near the interface.

The spurious velocity for each formulation is also examined. The magnitudes of spurious velocities denoted by \(Ca = \mu_1 |u_{max}| / \sigma \) are presented in Table 2. It can be seen that both \(F_{pf-1} \) and \(F_{pf-2} \) give small spurious velocities while the others give larger ones.

Finally, the absolute values of interfacial force across the drop center with different formulations are compared. The results are shown in Figure 3. Theoretically, the interfacial force should be zero everywhere except in the vicinity of the interface. However, it is found that the absolute values of \(F_{pf-1} \) have nonzero ones in the whole domain. This may cause earlier motion of the interface due to surface tension force effect. Since the interface width is fixed, the range of nonzero interfacial force decreases with increasing grid resolution. It is also observed that the interfacial force profiles of \(F_{sf-2}, F_{pf-2}, F_{csf-1}, F_{csf-2} \) are almost symmetric while the profiles of the others are obviously asymmetrical. This is mainly
due to the force imbalance property at the discrete level. In addition, the asymmetry of the order parameter profiles due to the spontaneous shrinkage property partly contributes to the asymmetry of interfacial tension distribution. According to the law of action and reaction, we expect the force on the droplet to be equal to the one on the surrounding fluid. As such, $F_{\text{stf}}-2$, $F_{\text{cpf}}-2$, $F_{\text{csf}}-1$, and $F_{\text{csf}}-2$ are more competitive.

### 5.2 Droplets merging

To test the performance of the LBM with different interfacial force formulations, the merging of two droplets is simulated in this section. Initially, two circular droplets (density $\rho_d$ and viscosity $\nu_d$) are placed in another fluid (density $\rho_s$ and viscosity $\nu_s$) in a rectangle domain of $L_x \times L_y$. When the initial gap $d$ between two droplets is smaller than $2W$, merging will occur due to the surface tension effect. The order parameter is initialized to be

$$\phi(x, y) = 1 + \tanh \left( \frac{2R_1 - \sqrt{(x-x_1)^2 + (y-y_1)^2}}{W} \right) + \tanh \left( \frac{2R_2 - \sqrt{(x-x_2)^2 + (y-y_2)^2}}{W} \right).$$  \hspace{1cm} (47)
The interfacial forces profiles along the midline of the drop for (A) $F_{stf-1}$, (B) $F_{stf-2}$, (C) $F_{cpf-1}$, (D) $F_{cpf-2}$, (E) $F_{pf-1}$, (F) $F_{pf-2}$, (G) $F_{csf-1}$, and (H) $F_{csf-2}$ [Colour figure can be viewed at wileyonlinelibrary.com]

where \((x_1, y_1) = (L_x/2 - R_1 - d/2, L_y/2)\) and \((x_2, y_2) = (L_x/2 + R_2 + d/2, L_y/2)\) are the centers of the two droplets, respectively. The initial velocity and pressure fields are zero in the whole domain. In simulations, the computational domain of \(L_x \times L_y = 1.2m \times 1m\) is discretized by a uniform mesh \(240 \times 200\). The initial radius of the two droplets is \(R_1 = R_2 = 0.125m\) and the initial gap is \(d = 1.5W\) and \(W = 0.02m\). The densities of the two phases are \(\rho_d = 5kg/m^3\), \(\rho_s = 1kg/m^3\) and the viscosities are \(\mu_d = \mu_s = 0.01m^2/s\). The surface tension coefficient is \(\sigma = 0.1N/m\), and the characteristic velocity is given by \(U_c = \sqrt{\sigma \rho_2 / R_2}\). The Peclet number is set as \(Pe = 0.1/Cn\). Periodic boundary conditions are implemented at all boundaries. With these parameters, merging will take place. Figure 4 shows the interfacial shapes of the droplets at \(t = 2T\) and \(30T\) with \(T = \sqrt{\rho_1 R^3 / \sigma}\). The interfacial shapes at \(t = 30T\) are compared with analytical results. From Figure 4, it is observed that the two droplets gradually merge, oscillate and finally form a larger stationary droplet. Especially, the final interface shapes predicted by all formulations are in good agreement with the analytical solutions. However, the interface positions predicted by $F_{cpf-1}$, $F_{pf-2}$, $F_{csf-2}$ at \(t = 2T\) are different from those of the other formulations. The droplets predicted by $F_{cpf-1}$, $F_{pf-2}$, and $F_{csf-2}$ have started to merge while the droplets with the other interfacial force formulations remain at a distinct distance. As no external forces are presented in the system, the mass center of the droplets should not change during coalescence. Figure 5 shows the time development of the position of the mass center of the droplets. All interfacial forces present similar accuracy. It’s worth pointing out that the computations with $F_{cpf-1}$, $F_{pf-2}$, $F_{pf-2}$, and $F_{csf-2}$ become unstable when \(Pe = 1/Cn\). This is because the adjustable parameter \(\eta\) in the mobility is very small to satisfy the condition of \(Pe = 1/Cn\). In this situation, the LBM for the phase field equation is prone to numerical instability. The magnitude of velocity predicted by the momentum equation with different interfacial force formulation may vary slightly, which could bring about the numerical instability. Nevertheless, both $F_{cpf}$ and $F_{csf}$ present a better numerical stability for this problem. However, since the CH equation is highly nonlinear and only the second-order finite difference scheme for the gradient operators is considered, this improvement in numerical stability is limited. It has been demonstrated that the stability of phase-field LB models can be greatly improved by employing the conservative AC equation, 55, 56 highly isotropic central difference, 52 or mixed finite difference scheme in the force term. 53, 57

We further simulate the above system but with two droplets of unequal sizes \((R_1 = 0.125m, R_2 = 0.1m)\). Figure 6 shows the interfacial shapes of the droplets at \(t = 2T\) and \(30T\). The interface positions are different for each interfacial force formulation. In particular, the merged droplets predicted by $F_{cpf-1}$, $F_{cpf-2}$, and $F_{csf-2}$ have a distinct movement. Figure 7 shows the time development of the position of the mass center of the droplets, which shows that the positions predicted by $F_{cpf}$ \((F_{cpf-1}, F_{cpf-2})\) and $F_{csf}$ \((F_{csf-1}, F_{csf-2})\) display significant deviations from their initial positions as time increases.
FIGURE 4 Interfaces of two droplets of equal sizes at $t = 2T$(dotted line) and $30T$(dashed line): (A) $F_{stf-1}$, (B) $F_{stf-2}$, (C) $F_{cfp-1}$, (D) $F_{cfp-2}$, (E) $F_{pf-1}$, (F) $F_{pf-2}$, (G) $F_{csp-1}$, and (H) $F_{csp-2}$. Solid line represents the analytical solutions [Colour figure can be viewed at wileyonlinelibrary.com]

FIGURE 5 Time history of mass center $x_c$ for two droplets of equal size [Colour figure can be viewed at wileyonlinelibrary.com]

5.3 Capillary wave

We further test the numerical accuracy of the interfacial force formulations by a two-dimensional Capillary wave problem. Initially, a heavier fluid is placed under a lighter fluid with a small perturbation $y = 1.5H + h_0 \cos(kx)$ on the interface in a rectangle domain of $H \times 3H$, where $h_0$ is the initial perturbation amplitude and $k = 2\pi/H$ is the wave number. The evolution of the interface wave amplitude $h(t)$ is given by Prosperetti\textsuperscript{58}

$$h(t) = \frac{4(1 - 4\beta)\nu^2k^4}{8(1 - 4\beta)\nu^2k^4 + \omega_0^2} \text{erfc}(\sqrt{\nu k^2} t) + \sum_{i=1}^{4} \frac{\omega_i^2}{Z_i} \frac{\omega_0^2}{Z_i^2 - \nu k^2} e^{(z_i^2 - \nu k^2)t} \text{erfc}(z_i \sqrt{t}),$$

(48)

where $\beta = \rho_1 \rho_2 / (\rho_1 + \rho_2)^2$, $\omega_i^2 = (\sigma k^3) / (\rho_2 + \rho_1)$, $\text{erfc}(z)$ is the complementary error function of a complex variable $z$, $z_i(i = 1, \ldots, 4)$ are the four roots of the following algebraic equation...
FIGURE 6 Interfaces of two droplets of unequal sizes at $t = 2T$ (dotted line) and $30T$ (dashed line): (A) $F_{stf-1}$, (B) $F_{stf-2}$, (C) $F_{cpf-1}$, (D) $F_{cpf-2}$, (E) $F_{pf-1}$, (F) $F_{pf-2}$, (G) $F_{csf-1}$, and (H) $F_{csf-2}$. Solid line represents the analytical solutions [Colour figure can be viewed at wileyonlinelibrary.com]

FIGURE 7 Time history of mass center $x_c$ for two droplets of unequal size [Colour figure can be viewed at wileyonlinelibrary.com]

$$z^4 - 4\beta \sqrt{vk^2}z^3 + 2(1 - 6\beta)vk^2z^2 + 4(1 - 3\beta)(vk^2)^{1/2}z + (1 - 4\beta)v^2k^4 + \omega_0^2 = 0,$$  
(49)

and $Z_i$ is defined as

$$Z_i = \prod_{j \neq i} (z_j - z_i), \quad i, j = 1, \ldots, 4.$$  
(50)

In simulations, periodic boundaries are applied to the left and right sides and no-slip boundaries are imposed on the top and bottom walls. The physical parameters are set as $\bar{H} = 1\text{m}$, $\rho_1 = \rho_2 = 1\text{kg/m}^3$, $v_1 = v_2 = 0.01\text{m}^2/\text{s}$, $\sigma = 0.25\text{N/m}$. The characteristic velocity is given by $U_c = \sqrt{\sigma/L_c/\rho_1}$. Hence, the Reynolds number is $Re = 50$ and the Weber number is $We = 1$. Two uniform grids of $H = 80$ and 160 are used. Figure 8 shows the evolution of the capillary amplitude for each grid. All the numerical results agree well with the theoretical solutions in the initial stage. However, the decaying amplitudes with $F_{cpf-1}$ and $F_{pf-2}$ on $80 \times 240$ meshes reach the steady state faster than the other forms as time increases.
We found that this behavior can be improved by increasing the Peclet number. However, it is still undesirable to carefully choose a parameter in advance compared with other formulations.

We further repeated the above simulations with $\rho_1/\rho_2 = 10$. The results are shown in Figure 9. In this case, all the results give a good agreement with the theoretical solutions. For quantitative comparison, the time averaged $L_2$-norm error for the wave amplitude is measured, which is defined as

$$E_2(h) = \sqrt{\frac{\rho_0}{25} \int_0^{25} \left| h(t) - \tilde{h}_{\text{exact}}(t) \right|^2 dt}. \quad (51)$$
Table 3 presents the time averaged $L_2$-norm error of wave amplitude, from which we can observe that all the averaged errors monotonically decrease as the numerical grid increases. Among the results, it can be found that the results given by $F_{pf-1}$ and $F_{csf-2}$ are closer to the analytical ones.

### 5.4 Rising bubble

We now examine the performance of different interfacial force formulations by simulating a bubble rising in a two-dimensional domain, which was also simulated by Hysing et al.\textsuperscript{60} and Aland and Voigt.\textsuperscript{61} Although no analytical solution is available for this problem, some numerical results were presented in Reference 60. The results from group 3 on the finest grids in Reference 60 are taken as the reference solutions. The schematic of the domain is shown in Figure 10. Initially, a bubble with radius $R = 0.25\text{m}$ is placed at $(0.5\text{m}, 0.5\text{m})$ in a rectangle domain of $2\text{m} \times 1\text{m}$. For the velocities, no-slip velocity boundary conditions are applied to the top and bottom boundaries and free-slip boundary conditions are imposed on the side boundaries. The gravitational force acts in the opposite direction of the vertical direction. Two uniform grids of $120 \times 240$ and $240 \times 480$ are employed. The fluid parameters are listed in Table 4. The related nondimensional numbers are given by

\begin{align}
\text{Re} &= \frac{\rho_1 U_g L}{\mu_1}, \quad \text{Eo} = \frac{\rho_1 U_g^2 L}{\sigma}, \quad \text{Mo} = \frac{\text{Eo}^3}{\text{Re}^4} = \frac{U_g^2 \mu_1^4}{2 \rho_1 \sigma^3 R},
\end{align}

where $U_g = \sqrt{2Rg}$ and $L$ are the reference velocity and length, respectively.
For comparison, the benchmark quantities, including bubble shape at $t = 3s$, rising velocity, center of mass, and circularity are measured by

$$y_c = \frac{\int_0^\infty (1 - \phi) y dx}{\int_0^\infty (1 - \phi) dx},$$

$$v_c = \frac{\int_0^\infty (1 - \phi) dx}{\int_0^\infty (1 - \phi) dx},$$

$$C = \frac{\text{perimeter of area-equivalent circle}}{\text{perimeter of bubble}} = \frac{2\sqrt{\int_0^\infty \phi dx}}{P_b},$$

where $v$ is the velocity component in the vertical direction and $P_b$ is obtained by integration over the contour line at $\phi = 0$ in Matlab.

Figure 11 shows the bubble shapes predicted by various interfacial force formulations at $T = 3s$. It can be seen that all the results seem to agree well with the benchmark solutions. However, the shapes of the bubble obtained by $F_{surf-1}$, $F_{surf-2}$, and $F_{pf-1}$ are clearly lower than the reference solutions for both grids compared with those predicted by other formulations. Figure 12 shows the time histories of the center of mass. At the initial stage, all the results are in good agreement with those reported in Reference 61. However, the discrepancy between the results with $F_{surf-1}$, $F_{surf-2}$, and $F_{pf-1}$ and the reference solutions becomes larger after $T = 1.5s$. Figure 13 compares the rising velocity of the bubble. All the results are similar and lower than the reference solutions when the bubble velocity approaches its maximum value. This may be caused by the interfacial compressibility effect in the LBM. In addition, the viscous effect caused by side walls may slow down the bubble. Figure 14 shows the circularity over time for all surface tension formulations, which clearly show that the data with all interfacial force formulations on both meshes agree well with the reference values. However, the results with $F_{pf-1}$ and $F_{pf-2}$ on the coarse mesh deviate slightly from the reference solutions. The minimum circularity on the finer mesh is significantly lower than that of the reference solution except for $F_{pf-1}$.

To clearly illustrate the differences among all formulations, the maximum mass center position, the maximum rising velocity, and minimum circularity with each force formulation are calculated and compared with the reference results. The results are presented in Table 5. Overall, the values obtained by $F_{surf-1}$, $F_{surf-2}$, $F_{surf-1}$, and $F_{surf-2}$ are similar and in better agreement with the reference data.

### 5.5 Droplet deformation in shear flow

Finally, we consider a circle drop deformation in a shear flow. The schematic of the flow field is shown in Figure 15. Initially, a circle drop is located at the center of a rectangle domain of $2H \times H$. The effect of gravity is ignored. The top
The shape of the drop can be characterized by a Taylor deformation parameter defined as $F_{\text{def}}$. The evolution of the center of mass for (A) $F_{\text{def}}$, (B) $F_{\text{def}}$, (C) $F_{\text{def}}$, (D) $F_{\text{def}}$, (E) $F_{\text{def}}$, (F) $F_{\text{def}}$, (G) $F_{\text{def}}$, and (H) $F_{\text{def}}$ [Colour figure can be viewed at wileyonlinelibrary.com]

The evolution of the rising velocity for (A) $F_{\text{def}}$, (B) $F_{\text{def}}$, (C) $F_{\text{def}}$, (D) $F_{\text{def}}$, (E) $F_{\text{def}}$, (F) $F_{\text{def}}$, (G) $F_{\text{def}}$, and (H) $F_{\text{def}}$ [Colour figure can be viewed at wileyonlinelibrary.com]

and bottom walls maintain velocities $U$ and $-U$, respectively, leading to a shear rate $E = 2U/H$. The periodic boundary conditions are applied to the left and right boundaries. The same density and viscosity are specified for both the drop and surrounding fluid. In the simulation, we set $H = 8m$, $R = 1m$, $U_w = 4m/s$, $\rho_d = \rho = 1kg/m^3$. The Reynolds number $Re = E\rho d R^2/\mu L = 0.1$. The capillary number $Ca = \mu ER/\sigma$ is varied from 0.1 to 0.4 by varying $\sigma$. The uniform grid size of $200 \times 200$ is employed. The shapes of the deformed drop at steady state are shown in Figure 16. It can be seen that the shapes of the drop given by all interfacial force formulations deform into an ellipsoidal one and are elongated as $Ca$ increases. In particular, the shapes of the drop obtained by $F_{\text{def}}$ are overstretched compared with other results. The shape of the drop can be characterized by a Taylor deformation parameter defined as $D = (L - B)/(L + B)$, where $L$ and $B$ are the lengths along the major axis and the minor axis of the droplet, respectively. A theoretical solution derived on
FIGURE 14 The evolution of circularity for (A) $F_{stf}^{-1}$, (B) $F_{stf}^{-2}$, (C) $F_{cpf}^{-1}$, (D) $F_{cpf}^{-2}$, (E) $F_{pf}^{-1}$, (F) $F_{pf}^{-2}$, (G) $F_{csf}^{-1}$, and (H) $F_{csf}^{-2}$ [Colour figure can be viewed at wileyonlinelibrary.com]

TABLE 5 Benchmark quantities for rising bubble on 240 × 480 meshes

| Parameter | Reference 60 | $F_{stf}^{-1}$ | $F_{stf}^{-2}$ | $F_{cpf}^{-1}$ | $F_{cpf}^{-2}$ | $F_{pf}^{-1}$ | $F_{pf}^{-2}$ | $F_{csf}^{-1}$ | $F_{csf}^{-2}$ |
|-----------|---------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| $y_{max}$ | 1.0817        | 1.0672         | 1.0674         | 1.0539         | 1.0539         | 1.0536         | 1.0669         | 1.0675         | 1.0681         |
| $v_{max}$ | 0.2417        | 0.2278         | 0.2279         | 0.2229         | 0.2228         | 0.2223         | 0.2276         | 0.2279         | 0.2284         |
| $C_{min}$ | 0.9013        | 0.8917         | 0.8914         | 0.8983         | 0.8979         | 0.9004         | 0.8889         | 0.8933         | 0.8966         |

FIGURE 15 Drop deformation in a shear flow. $L$ is the major axis and $B$ is the minor axis

the assumptions of the Stokes flow and small deformation shows that the Taylor deformation parameter is related to the capillary number and the viscosity ratio $^{64,65}$

$$D = \frac{L - B}{L + B} = Ca\left(\frac{19\lambda + 16}{16\lambda + 16}\right),$$

(56)

where $\lambda = \mu_d/\mu_f$ is the viscosity ratio between the drop fluid and the surrounding fluid. Table 6 shows the Taylor deformation parameters with different force formulations. It can be seen that the values predicted by $F_{csf}^{-1}$ are significantly higher than the theoretical values. Overall, the values with $F_{stf}^{-1}$ and $F_{stf}^{-2}$ are close to the theoretical ones.
6 | CONCLUSIONS

In this article, we successfully implemented the phase-field-based lattice Boltzmann method with different interfacial force formulations for two phases flow. The performance of each surface tension formulation has been validated and compared. For a stationary drop, $F_{\text{csf}^-2}$ provides the most accurate prediction in terms of the surface tension coefficient. CPF can generate small spurious currents. $F_{\text{csf}^-2}$, $F_{\text{pf}^-2}$, and $F_{\text{pf}^-1}$ produce a smooth pressure field. The amplitude of the surface tension force predicted by $F_{\text{csf}^-2}$, $F_{\text{pf}^-2}$, $F_{\text{pf}^-1}$, and $F_{\text{csf}^-2}$ can become symmetric with respect to the phase interface. For the droplets merging problems, there are obvious differences for all formulations in terms of the interface shapes of the droplets during coalescence. The droplets predicted by $F_{\text{cpf}^-1}$, $F_{\text{pf}^-2}$, and $F_{\text{csf}^-2}$ is more prone to merge due to the surface tension effects. In particular, the unexpected movement of droplets with unequal sizes occurs when $F_{\text{cpf}^-1}$, $F_{\text{cpf}^-2}$, $F_{\text{pf}^-1}$, and $F_{\text{csf}^-2}$ are used. It is also found that $F_{\text{pf}^-1}$ and $F_{\text{pf}^-2}$ show better numerical stability than $F_{\text{cpf}}$ and $F_{\text{csf}}$ in terms of a large Peclet number. For the test of capillary wave, the evolution processes of the interface amplitude predicted by $F_{\text{pf}^-1}$ and $F_{\text{csf}^-2}$ are closer to the analytical solutions in all formulations. It is worth noting that $F_{\text{cpf}^-1}$ can yield good results but the Peclet number should be carefully chosen in advance. For the simulation of a rising bubble, both the stress form and CSF form give a good results in terms of the mass center of the bubble. However, $F_{\text{cpf}^-1}$, $F_{\text{pf}^-2}$, and $F_{\text{pf}^-1}$ clearly underestimate the position of the mass center at the late stage. For the rising velocity, all formulations underestimate the maximum rise velocity. In terms of the circularity, only $F_{\text{cpf}^-2}$ and $F_{\text{csf}^-2}$ give the predictions closer to
the reference solutions. For shear flow, all formulations give accurate predictions in comparison with the linear theory at $Ca = 0.1$. With the increase of capillary number, $F_{csf}^{-1}$ produces a larger deformation than the theoretical predictions. For all the considered capillary number, $F_{stf}^{-1}$ and $F_{stf}^{-2}$ can give a satisfactory prediction. In summary, it seems that no surface tension force formulation can give satisfactory results in all respects although each formulation can produce an approximate analytical solution to some extent. Different forms may be considered for different concerns. Overall, $F_{cpf}$ is good for calculating multiphase flows with small interface deformation. Both $F_{stf}$ and $F_{csf}$ are good for dynamical situations. In each category, $F_{stf}^{-2}$, $F_{stf}^{-1}$, $F_{cpf}^{-1}$, and $F_{csf}^{-2}$ are preferred.

Finally, it is noted that a number of multiphase LBM models have been developed based on the conservative (local and nonlocal) AC equation recently. In this type of model, the interfacial force formulations are the same as those in Section 2. Compared with the CH equation, the values of the order parameter calculated by the conserved AC equation are easy to be kept within a reasonable range. In this work, the benchmark problems with small density contrasts are considered, the differences of the order parameter predicted by both CH and AC can be negligible. Therefore, the conclusions may be still valid for the NSAC system. It is also worth pointing out that the differences of the performance of the interfacial force formulations mainly come from two aspects. One is from the order parameter predicted by the interface-tracking equation, the values of which may be beyond the physical range $[-1, 1]$ and the profile of which may produce no-smooth solution near the interface region, even though the program is still numerically stable. The other is from the discretization of interfacial force that is independent of the density ratio. Therefore, for multiphase flows with large density ratios, the capabilities of different interfacial force formulations may be different due to the abnormal change of the values of the order parameter. Nevertheless, the conclusions remain instructive since the performances of the interfacial force formulations themselves in the momentum equation have been validated and compared. We hope the present comparisons can provide insights into the advantages and limitations of each formulation.

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DATA AVAILABILITY STATEMENT
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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APPENDIX A. RELATIONS AMONG DIFFERENT INTERFACIAL FORCE FORMULATIONS

This appendix presents the relations among different interfacial force formulations. In Equation (24), the curvature term can be written as

\[ \nabla \cdot n = \nabla \cdot \left( \frac{\nabla \phi}{|\nabla \phi|} \right) = \frac{1}{|\nabla \phi|} \left( \nabla^2 \phi - \frac{\nabla \phi \cdot \nabla |\nabla \phi|}{|\nabla \phi|} \right). \]  
\[ (A1) \]

Substituting the above equation into Equation (24) yields

\[ F_{csf-1} = -\kappa \nabla \phi |\nabla \phi| \nabla \cdot n \\
= -\kappa \nabla \phi \left( \nabla^2 \phi - \frac{\nabla \phi \cdot \nabla |\nabla \phi|}{|\nabla \phi|} \right) \\
= F_{pf-2} + \kappa \frac{\nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{|\nabla \phi|}, \]  
\[ (A2) \]

By using the equality \( \nabla \phi \nabla^2 \phi = \nabla (\phi \nabla^2 \phi) - \phi \nabla \nabla^2 \phi \), one can obtain the following relationship,

\[ F_{csf-1} = F_{pf-2} - \kappa \nabla (\phi \nabla^2 \phi) + \kappa \frac{\nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{|\nabla \phi|}, \]  
\[ (A3) \]
Based on Equation (2), \( F_{csf-1} \) can be rewritten as

\[
F_{csf-1} = \nabla \phi \left( \mu_\phi - \frac{\partial f_0}{\partial \phi} \right) + \kappa \frac{\nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{| \nabla \phi |},
\]

\[
= F_{csf-2} - \nabla f_0 + \kappa \frac{\nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{| \nabla \phi |},
\]

\[
= F_{csf-1} + \nabla (\phi \mu_\phi) - \nabla f_0 + \kappa \frac{\nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{| \nabla \phi |},
\]

(A4)

where we have used the equality \( \nabla (\phi \mu_\phi) = \mu_\phi \nabla \phi + \phi \nabla \mu_\phi \). By using the following equality,

\[
-\kappa \nabla \phi \Delta \phi = \frac{\kappa}{2} |\nabla \phi|^2 - \nabla \cdot \kappa (\nabla \phi \otimes \nabla \phi),
\]

(A5)

one can obtain

\[
F_{csf-1} = -\kappa \nabla \phi \nabla^2 \phi + \kappa \frac{\nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{| \nabla \phi |},
\]

\[
= F_{stf-1} + \frac{\kappa}{2} |\nabla \phi|^2 + \kappa \frac{\nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{| \nabla \phi |}.
\]

(A6)

By virtue of Equation (4), we have

\[
|\nabla \phi| = \frac{\partial \phi}{\partial r} = \frac{2}{W}(1 - \phi^2),
\]

\[
\frac{\nabla \phi (\nabla \phi \cdot \nabla |\nabla \phi|)}{| \nabla \phi |} = \frac{1}{2} |\nabla \phi|^2 = \frac{8}{W^2} \phi (\phi^2 - 1) \nabla \phi.
\]

(A7)

Equation (A6) is then derived as

\[
F_{csf-1} = -\nabla \cdot \kappa (\nabla \phi \otimes \nabla \phi) + \kappa \nabla |\nabla \phi|^2 = F_{stf-2}.
\]

(A8)

**APPENDIX B. THE DIRAC FUNCTION IN \( F_{csf-1} \) AND \( F_{csf-2} \)**

In \( F_{csf-1} \), the surface Dirac function is chosen as \( \alpha |\nabla \phi|^2 \) with \( \alpha \) being an undetermined parameter. Based on Equation (4),

\[
|\nabla \phi| = \frac{\partial \phi}{\partial r} = \frac{2}{W}(1 - \phi^2),
\]

(B1)

Inserting the above equation into Equation (23) yields,

\[
\int_{-\infty}^{\infty} \alpha |\nabla \phi|^2 dr = \int_{-\infty}^{\infty} \alpha \frac{2}{W}(1 - \phi^2) \frac{\partial \phi}{\partial r} dr
\]

\[
= \int_{-\infty}^{\infty} \alpha \frac{2}{W}(1 - \phi^2) d\phi
\]

\[
= \alpha \frac{2}{W} \int_{-\infty}^{\infty} d \left( \phi - \frac{\phi^3}{3} \right)
\]

\[
= \frac{8\alpha}{3W} = 1,
\]

(B2)

where \( \phi|_{r=\infty} = 1 \) and \( \phi|_{r=-\infty} = -1 \) are used. As a result, \( \alpha = \frac{3W}{8} \).
In $F_{csf-2}$, the surface Dirac function is chosen as $a|\nabla \phi|$. Analogously, one can have

$$
\int_{-\infty}^{\infty} a|\nabla \phi| \, dr = \int_{-\infty}^{\infty} \frac{\partial \phi}{\partial r} \, dr = \int_{-\infty}^{\infty} a \, d\phi = 2a = 1.
$$

(B3)

This leads to $a = \frac{1}{2}$. 
