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Physical formulation and numerical algorithm for simulating N immiscible incompressible fluids involving general order parameters

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ABSTRACT

We present a family of physical formulations, and a numerical algorithm, based on a class of general order parameters for simulating the motion of a mixture of N ($N \geqslant 2$) immiscible incompressible fluids with given densities, dynamic viscosities, and pairwise surface tensions. The N-phase formulations stem from a phase field model we developed in a recent work based on the conservations of mass/momentum, and the second law of thermodynamics. The introduction of general order parameters leads to an extremely strongly-coupled system of $(N-1)$ phase field equations. On the other hand, the general form enables one to compute the N-phase mixing energy density coefficients in an *explicit* fashion in terms of the pairwise surface tensions. We show that the increased complexity in the form of the phase field equations associated with general order parameters in actuality does not cause essential computational difficulties. Our numerical algorithm reformulates the $(N-1)$ strongly-coupled phase field equations for general order parameters into $2(N-1)$ Helmholtz-type equations that are completely de-coupled from one another. This leads to a computational complexity comparable to that for the simplified phase field equations associated with certain special choice of the order parameters. We demonstrate the capabilities of the method developed herein using several test problems involving multiple fluid phases and large contrasts in densities and viscosities among the multitude of fluids. In particular, by comparing simulation results with the Langmuir–de Gennes theory of floating liquid lenses we show that the method using general order parameters produces physically accurate results for multiple fluid phases.

1. Introduction

The present work focuses on the motion of a mixture of N ($N \geqslant 2$) immiscible incompressible fluids with given densities, dynamic viscosities, and pairwise surface tensions. The system is assumed to contain no solid phase (e.g. solid particles), except for possible solid-wall boundaries. The situation is a generalization of incompressible two-phase flows, which have been under intensive investigations by the community for decades. The applications and potential implications of N-phase problems are enormous, from both the practical engineering perspective and fundamental physics perspective [42,8,37].

N-phase flows have been the subject of a number of past research efforts in the literature. A summary of the existing studies is provided in the following paragraphs. Several researchers have reviewed two-phase flows comprehensively for

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different approaches and techniques [36,44,2,31,19,38,43,39,30]; see also the references therein. We will therefore restrict our attention in the following review, and also in the main work of the present paper, to flows with three or more fluid phases ($N \geqslant 3$), noting that the technique developed herein equally applies to two-phase flows. It should also be noted that our attention is limited to incompressible fluids.

Since the overall approach of the current work pertains to the phase field framework, we will first briefly mention the representative works for multiple phases based on other related approaches such as level set or volume of fluids, and then will concentrate on the existing studies with phase fields. The level set technique as proposed by [36] is extended from two to N components in several studies (see e.g. [35,47,41,49,45], among others), where different fluid components are differentiated by using N [35,47], $(N-1)$ [41,49], or $\lceil \log_2^N \rceil$ [7] level set functions. In [37] the multiple phases are characterized by a single unsigned level set function, and the ϵ ($\epsilon > 0$ is a small number) level sets are convected in the usual way by the flow, while the fluid interfaces at the new step are re-constructed from the ϵ level sets by a Voronoi tessellation. The work of [3] combines the experiments and numerical simulations based on a volume-of-fluid approach, and investigates in detail the dynamics of an air bubble crossing an interface between two fluids.

Let us now concentrate on the past studies of N-phase flows with the phase field (or diffuse interface) approach, which appears to constitute the majority of efforts in this area. The main developments are thanks to the works of Kim and collaborators [24,21,22], Boyer and collaborators [4–6], and Heida and collaborators [18]. Among these, several investigations have been devoted to the discretizations of the three-component [23,6] or N-component [26,27] Cahn–Hilliard equations or Allen–Cahn equations, where the hydrodynamic interaction is absent. In particular, energy-stable schemes for the three-component Cahn–Hilliard equations are discussed in [6], and a nonlinear multigrid method combined with a finite-difference discretization is presented in [23,26]. When the hydrodynamic interaction is present, in [24] a thermodynamically-consistent phase field model for N fluid components is derived based on the balance equations and the second law of thermodynamics. This is a generalization of the two-phase model proposed by [31]. An important feature is that, the mixture velocity in this N-phase model is the mass-averaged velocity and therefore it is not divergence free. In [4] a three-component Cahn–Hilliard system is coupled with the Navier–Stokes equation, supplemented by a capillary force, to model three-phase flows. The effects of different forms for the bulk free energy have been studied. Noting a solvability difficulty in determining the coefficients for the surface-tension forces when more than three phases are involved, Kim [21] proposed a phenomenological surface-tension force for multiple fluid components; see its applications in [22,28]. In [5] a combined Cahn–Hilliard/Navier–Stokes model has been studied for three-phase flows, in which the Cahn–Hilliard model uses a particular free-energy form due to [4] and the Navier–Stokes equation uses a special form for the inertia term due to [16]. More recently, Heida et al. [18] present a Cahn–Hilliard–Navier–Stokes type phase field model for N fluid phases in which the constitutive relations are obtained by requiring the maximization of the rate of entropy production. The mixture velocity in this model, similar to that of [24,29], is also the mass-averaged velocity and is not divergence free. Another interesting work is [34], in which an Euler type equation, i.e. barring the dissipation terms, with the surface tensions of an N-phase field has been derived based on the variational principle.

Very recently, by considering the mass conservations of the N individual fluid phases, the momentum conservation, Galilean invariance and the second law of thermodynamics, we have derived in [10] a general phase field model (isothermal) for the mixture of N ($N \geqslant 2$) immiscible incompressible fluids; see also Section 1.1 below. This model is fundamentally different from those of [24,18,29], in that the mixture velocity in our model is the *volume-averaged* velocity, which can be rigorously shown to be divergence free [10]. In contrast, the velocity in the models of [24,18,29] is the *mass-averaged* velocity, and is not divergence free. Our N-phase model can be considered as a generalization of the formulation in [1] for two-phase flows.

In order to provide an N-phase formulation suitable for numerical simulations, the general N-phase model of [10] requires the further specification of two items: (1) a set of $(N-1)$ order parameters, and (2) the form of the N-phase free energy density function. By order parameters we refer to a set of independent variables used to characterize the state of the system, and *in the current paper we use this term interchangeably with phase field variables or phase field functions*. In [10] we have employed a very special set of order parameters, which is also given in Section 2.1, because this set significantly simplifies the form of the resulting phase field equations. Employing this particular phase field formulation, we have further developed a method for computing the mixing energy density coefficients (see Section 2.1 for definition) involved in the formulation by solving a linear algebraic system based on the pairwise surface tensions among the N fluids. We have also developed an algorithm in [10] for solving the coupled system of governing equations in that formulation.

In this paper we generalize the N-phase formulation to a class of general order parameters. This gives rise to a family of N-phase physical formulations suitable for numerical simulations. Within this family, by specifying a constant non-singular matrix and a constant vector, one will single out a specific N-phase formulation. This class of physical formulations with general order parameters includes the one of [10] as a particular case.

The introduction of the class of general order parameters has two main implications:

- It enables us to derive an *explicit form* for the mixing energy density coefficients in terms of the pairwise surface tensions. Therefore, the mixing energy density coefficients with general order parameters can be *explicitly* computed. In contrast, the method of [10] requires one to solve a linear algebraic system to determine the mixing energy density coefficients.

- The resulting phase field equations have a markedly more complicated form than that employing the special set of order parameters of [10]. In particular, the $(N-1)$ phase field equations with general order parameters become extremely strongly-coupled with one another. The increased complexity raises new challenges to their numerical solutions.

We have developed an algorithm for solving the new phase field equations associated with general order parameters, and it overcomes the computational challenge caused by the increased complexity. Our algorithm reformulates the $(N-1)$ strongly-coupled phase field equations for general order parameters into $2(N-1)$ Helmholtz type equations that are completely de-coupled from one another. With this algorithm the computational complexity for the general order parameters is comparable to that of [10] for the simplified phase field equations with the special order parameters. This algorithm for the phase field equations with general order parameters, combined with an algorithm for the N-phase momentum equations, provides an efficient method for simulating N-phase flows, which has also overcome the computational issues associated with variable mixture density and viscosity.

The current work is in line with the following view about the order parameters (or phase field variables). The order parameters or phase field variables serve merely as a set of state variables chosen to characterize the system, and they can be chosen in different ways. Using a different set of order parameters leads to a different representation of the N-phase system. While the resulting phase field equations may have varying degrees of complexity with different order parameters, the various representations of the N-phase system should be equivalent to one another.

The novelties of this paper lie in three aspects: (1) the family of N-phase physical formulations associated with general order parameters, (2) the explicit form of the N-phase mixing energy density coefficients in terms of the pairwise surface tensions for general order parameters, and (3) the numerical algorithm for solving the $(N-1)$ strongly-coupled phase field equations with general order parameters. The algorithm for solving the N-phase momentum equations in the current paper, given in Appendix B, is also new in the context of N-phase flows. This algorithm is different from that of [10] for the N-phase momentum equations. The essential strategies for dealing with the variable density, variable viscosity and the pressure–velocity coupling employed here, however, stem from our previous work [14] for two-phase flows. Finally, in Appendix A of this paper we have provided a proof that the linear algebraic system about the mixing energy density coefficients derived in [10] for the special set of order parameters is uniquely solvable. The unique solvability of that system is an un-settled issue in [10].

We employ C^0-continuous spectral elements [40,20,48] for spatial discretizations in the current paper. It should however be noted that the numerical algorithm developed herein is general and can also be used with other techniques such as finite element, finite difference or finite volume.

The rest of this paper is organized as follows. In Section 1.1 we provide a summary of the general phase field model for a mixture of N immiscible incompressible fluids we derived in [10], which serves as the basis for the N-phase physical formulations with general order parameters of the current paper. In Section 2 we discuss a class of general order parameters and the N-phase formulations with the general order parameters. We also derive an explicit form for the mixing energy density coefficients in terms of the pairwise surface tensions among the N fluids. Section 3 provides an efficient algorithm for solving the $(N-1)$ strongly-coupled phase field equations with general order parameters. We further combine this algorithm, with a scheme for the N-phase momentum equations discussed in Appendix B, to form an overall method for N-phase flow simulations. In Section 4 we look into several numerical examples involving three and four fluid phases to demonstrate the accuracies and capabilities of the presented method with general order parameters. Section 5 concludes the discussions with a summary of the key points. Finally, Appendix A provides a proof for the unique solvability of the linear algebraic system about the mixing energy density coefficients derived in [10]. Appendix B summarizes a scheme for the N-phase momentum equations.

1.1. A general phase-field model for an N-fluid mixture

This subsection summarizes the general phase field model we derived in [10] based on the conservations of mass and momentum, the second law of thermodynamics, and Galilean invariance. We refer to [10] for detailed derivations of this system.

Let Ω denote the flow domain in two or three dimensions, and $\partial\Omega$ denote the boundary of Ω. Consider the mixture of N $(N \geqslant 2)$ immiscible incompressible fluids contained in Ω. Let $\tilde{\rho}_i$ $(1 \leqslant i \leqslant N)$ denote the constant densities of these N pure fluids (before mixing), and $\tilde{\mu}_i$ $(1 \leqslant i \leqslant N)$ denote their constant dynamic viscosities. We define the auxiliary parameters

$$\tilde{\gamma}_i = \frac{1}{\tilde{\rho}_i} \quad \text{for } 1 \leqslant i \leqslant N, \qquad \Gamma = \sum_{i=1}^{N} \tilde{\gamma}_i, \qquad \Gamma_\mu = \sum_{i=1}^{N} \tilde{\gamma}_i \tilde{\mu}_i. \tag{1}$$

Let ϕ_i $(1 \leqslant i \leqslant N-1)$ denote the $(N-1)$ independent order parameters (or interchangeably, phase field variables) that characterize the N-phase system, and $\vec{\phi} = (\phi_1, \ldots, \phi_{N-1})$ denote the vector of phase field variables. Let $\rho_i(\vec{\phi})$ and $c_i(\vec{\phi})$

($1 \leqslant i \leqslant N$) respectively denote the density and the volume fraction of the i-th fluid *within the mixture*. Let $\rho(\vec{\phi})$ denote the mixture density. We have the relations

$$c_i = \frac{\rho_i}{\tilde{\rho}_i} = \tilde{\gamma}_i \rho_i, \qquad \sum_{i=1}^{N} c_i = 1, \qquad \rho = \sum_{i=1}^{N} \rho_i. \tag{2}$$

Let $W(\vec{\phi}, \nabla\vec{\phi})$ denote the free energy density function of the system, which must satisfy the following condition

$$\sum_{i=1}^{N-1} \nabla\phi_i \otimes \frac{\partial W}{\partial(\nabla\phi_i)} = \sum_{i=1}^{N-1} \frac{\partial W}{\partial(\nabla\phi_i)} \otimes \nabla\phi_i, \tag{3}$$

where $\otimes$ denotes the tensor product. Then this N-phase system is described by the following equations [10]:

$$\rho\left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{u}\right) + \tilde{\mathbf{J}}\cdot\nabla\mathbf{u} = -\nabla p + \nabla\cdot\left[\mu(\vec{\phi})\mathbf{D}(\mathbf{u})\right] - \sum_{i=1}^{N-1} \nabla\cdot\left(\nabla\phi_i \otimes \frac{\partial W}{\partial(\nabla\phi_i)}\right), \tag{4a}$$

$$\nabla\cdot\mathbf{u} = 0, \tag{4b}$$

$$\sum_{j=1}^{N-1} \frac{\partial\varphi_i}{\partial\phi_j}\left(\frac{\partial\phi_j}{\partial t} + \mathbf{u}\cdot\nabla\phi_j\right) = \nabla\cdot\left[\tilde{m}_i(\vec{\phi})\nabla C_i\right], \quad 1 \leqslant i \leqslant N-1, \tag{4c}$$

where $\mathbf{u}(\mathbf{x}, t)$ is velocity, $p(\mathbf{x}, t)$ is pressure, $\mathbf{D}(\mathbf{u}) = \nabla\mathbf{u} + \nabla\mathbf{u}^T$ (superscript T denoting transpose), $\mathbf{x}$ and t are respectively the spatial and temporal coordinates. $\tilde{m}_i(\vec{\phi}) \geqslant 0$ ($1 \leqslant i \leqslant N-1$) are the mobilities corresponding to ϕ_i. $\varphi_i(\vec{\phi})$ are defined by

$$\varphi_i \equiv \rho_i(\vec{\phi}) - \rho_N(\vec{\phi}) = \varphi_i(\vec{\phi}), \quad 1 \leqslant i \leqslant N-1. \tag{5}$$

The chemical potentials $C_i(\vec{\phi}, \nabla\vec{\phi})$ ($1 \leqslant i \leqslant N-1$) are given by the following linear algebraic system

$$\sum_{j=1}^{N-1} \frac{\partial\varphi_j}{\partial\phi_i} C_j = \frac{\partial W}{\partial\phi_i} - \nabla\cdot\frac{\partial W}{\partial(\nabla\phi_i)}, \quad 1 \leqslant i \leqslant N-1, \tag{6}$$

which can be solved given $W(\vec{\phi}, \nabla\vec{\phi})$ and $\varphi_i(\vec{\phi})$. $\tilde{\mathbf{J}}(\vec{\phi}, \nabla\vec{\phi})$ is given by

$$\tilde{\mathbf{J}} = -\sum_{i=1}^{N-1}\left(1 - \frac{N}{\Gamma}\tilde{\gamma}_i\right)\tilde{m}_i(\vec{\phi})\nabla C_i. \tag{7}$$

The density of fluid k ($1 \leqslant k \leqslant N$) *within the mixture* is given by

$$\rho_k(\vec{\phi}) = \begin{cases} \frac{1}{\Gamma}[1 - \sum_{i=1}^{N-1}\tilde{\gamma}_i\varphi_i(\vec{\phi})], & k = N \\ \rho_N(\vec{\phi}) + \varphi_k(\vec{\phi}), & 1 \leqslant k \leqslant N-1. \end{cases} \tag{8}$$

The volume fractions $c_k(\vec{\phi})$ ($1 \leqslant k \leqslant N$) can then be computed from (2). The mixture density $\rho(\vec{\phi})$ and dynamic viscosity $\mu(\vec{\phi})$ are given by

$$\begin{cases} \rho(\vec{\phi}) = \sum_{i=1}^{N}\rho_i(\vec{\phi}) = \frac{N}{\Gamma} + \sum_{i=1}^{N-1}\left(1 - \frac{N}{\Gamma}\tilde{\gamma}_i\right)\varphi_i(\vec{\phi}), \\ \mu(\vec{\phi}) = \sum_{i=1}^{N}\tilde{\mu}_i c_i(\vec{\phi}) = \frac{\Gamma_\mu}{\Gamma} + \sum_{i=1}^{N-1}\left(\tilde{\mu}_i - \frac{\Gamma_\mu}{\Gamma}\right)\tilde{\gamma}_i\varphi_i(\vec{\phi}). \end{cases} \tag{9}$$

Once the functions $\varphi_i(\vec{\phi})$ ($1 \leqslant i \leqslant N-1$) and the free energy density function $W(\vec{\phi}, \nabla\vec{\phi})$ are known, essentially all the other quantities in the model can be computed.

2. Order parameters and N-phase physical formulation

2.1. N-phase formulations with general order parameters

To arrive at an N-phase physical formulation suitable for numerical simulations, the phase field model given in Section 1.1 requires the specification of: (i) the form of the free energy density function $W(\vec{\phi}, \nabla\vec{\phi})$, and (ii) the set of order parameters ϕ_i ($1 \leqslant i \leqslant N-1$) through Eq. (5).

Following [10], we assume the following form for the free energy density function of the N-phase system

$$W(\vec{\phi}, \nabla\vec{\phi}) = \sum_{i,j=1}^{N-1} \frac{\lambda_{ij}}{2} \nabla\phi_i \cdot \nabla\phi_j + \frac{\beta^2}{2\eta^2} H(\vec{\phi}), \quad H(\vec{\phi}) = \sum_{k=1}^{N} c_k^2 (1 - c_k)^2, \tag{10}$$

where β^2 is a characteristic scale for the energy, and η is a characteristic scale for the interfacial thickness. $c_k(\vec{\phi})$ $(1 \leqslant k \leqslant N)$ is the volume fraction of the fluid k in the mixture. λ_{ij} $(1 \leqslant i, j \leqslant N-1)$ are referred to as the mixing energy density coefficients, and they are assumed to be constant in the current paper. The condition (3) requires that the matrix

$$\mathbf{A} = [\lambda_{ij}]_{(N-1)\times(N-1)} \tag{11}$$

be symmetric. We further require that $\mathbf{A}$ be positive definite to ensure the positivity of the first term in the $W(\vec{\phi}, \nabla\vec{\phi})$ expression. Therefore, the matrix $\mathbf{A}$ is required to be symmetric positive definite (SPD) in the current paper.

The form of the free energy density function (10), in particular the cross terms $\nabla\phi_i \cdot \nabla\phi_j$ $(i \neq j)$ therein, give rise to a set of phase-field equations that are very different from those of the existing N-phase studies [24,4,21,5,22,28]. It is the key that enables us to determine the mixing energy density coefficients λ_{ij} uniquely, and to provide their explicit expressions, based on the pairwise surface tensions among the N fluids. This will be discussed subsequently in Section 2.2.

We now focus on the order parameters $\vec{\phi}$, and this is the departure point of the current work. Let

$$\mathbf{A}_1 = [a_{ij}]_{(N-1)\times(N-1)}, \qquad \mathbf{b}_1 = [b_i]_{(N-1)\times 1} \tag{12}$$

respectively denote a prescribed non-singular constant matrix and a prescribed constant vector. We define the $(N-1)$ order parameters ϕ_i as follows,

$$\varphi_i(\vec{\phi}) = \rho_i(\vec{\phi}) - \rho_N(\vec{\phi}) = \sum_{j=1}^{N-1} a_{ij}\phi_j + b_i, \quad 1 \leqslant i \leqslant N-1. \tag{13}$$

Note that ϕ_i $(1 \leqslant i \leqslant N-1)$ as defined above can in general be dimensional or non-dimensional variables. However, in the current paper we will require that ϕ_i be non-dimensional in the simulations. Eq. (13) defines a family of order parameters. Given a specific set of $\mathbf{A}_1$ and $\mathbf{b}_1$ in (12), Eq. (13) will define a unique set of order parameters ϕ_i $(1 \leqslant i \leqslant N-1)$. We refer to the family of order parameters defined by (13) as the general order parameters.

With the set of general order parameters given by (13), and the free energy density function given by (10), Eqs. (4a)–(4c) are reduced to the following system,

$$\rho(\vec{\phi})\left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{u}\right) + \tilde{\mathbf{J}}(\vec{\phi}, \nabla\vec{\phi})\cdot\nabla\mathbf{u} = -\nabla p + \nabla\cdot\left[\mu(\vec{\phi})\mathbf{D}(\mathbf{u})\right] - \sum_{i,j=1}^{N-1} \nabla\cdot(\lambda_{ij}\nabla\phi_i\nabla\phi_j) + \mathbf{f} \tag{14a}$$

$$\nabla\cdot\mathbf{u} = 0 \tag{14b}$$

$$\sum_{j=1}^{N-1} d_{ij}\left(\frac{\partial\phi_j}{\partial t} + \mathbf{u}\cdot\nabla\phi_j\right) = \nabla^2\left[-\sum_{j=1}^{N-1}\lambda_{ij}\nabla^2\phi_j + h_i(\vec{\phi})\right] + g_i(\mathbf{x}, t), \quad 1 \leqslant i \leqslant N-1, \tag{14c}$$

where we have taken into account an external body force $\mathbf{f}(\mathbf{x}, t)$ in the momentum equation (14a), and included a prescribed source term g_i in the phase field equations (14c). g_i $(1 \leqslant i \leqslant N-1)$ are for the purpose of numerical testing only, and will be set to $g_i = 0$ in actual simulations. The constants d_{ij} $(1 \leqslant i, j \leqslant N-1)$ are defined by

$$\begin{cases} \mathbf{A}_2 = [d_{ij}]_{(N-1)\times(N-1)} = \mathbf{A}_1^T\mathbf{M}^{-1}\mathbf{A}_1, \\ \mathbf{M} = \operatorname{diag}\left(m_1\left(\dfrac{\tilde{\rho}_1 + \tilde{\rho}_N}{2}\right)^2, m_2\left(\dfrac{\tilde{\rho}_2 + \tilde{\rho}_N}{2}\right)^2, \ldots, m_{N-1}\left(\dfrac{\tilde{\rho}_{N-1} + \tilde{\rho}_N}{2}\right)^2\right), \end{cases} \tag{15}$$

where $m_i > 0$ $(1 \leqslant i \leqslant N-1)$ are the interfacial mobility coefficients associated with ϕ_i and are assumed to be positive constants. Note that the matrix $\mathbf{A}_2$ is symmetric positive definite (SPD) based on the assumptions about the non-singularity of $\mathbf{A}_1$ and the positivity of m_i.

The function $h_i(\vec{\phi})$ in (14c) is given by

$$h_i(\vec{\phi}) = \frac{\beta^2}{2\eta^2}\frac{\partial H}{\partial\phi_i}, \quad 1 \leqslant i \leqslant N-1, \tag{16}$$

where $H(\vec{\phi})$ is defined in (10). The volume fractions $c_k(\vec{\phi})$ $(1 \leqslant k \leqslant N)$ with the general order parameters are therefore given by (see (2) and (8)),

$$c_k(\vec{\phi}) = \tilde{\gamma}_k\rho_k(\vec{\phi}), \qquad \rho_k(\vec{\phi}) = \begin{cases} \frac{1}{\Gamma} - \sum_{i=1}^{N-1}\frac{\tilde{\gamma}_i}{\Gamma}\left(\sum_{j=1}^{N-1} a_{ij}\phi_j + b_i\right), & \text{if } k = N, \\ \rho_N(\vec{\phi}) + \left(\sum_{j=1}^{N-1} a_{kj}\phi_j + b_k\right), & \text{if } 1 \leqslant k \leqslant N-1. \end{cases} \tag{17}$$

The mixture density $\rho(\vec{\phi})$ is given by

$$\rho(\vec{\phi}) = \sum_{i=1}^{N} \rho_i(\vec{\phi}) = \frac{N}{\Gamma} + \sum_{i=1}^{N-1} \left(1 - \frac{N}{\Gamma}\tilde{\gamma}_i\right)\left(\sum_{j=1}^{N-1} a_{ij}\phi_j + b_i\right). \tag{18}$$

The mixture dynamic viscosity $\mu(\vec{\phi})$ is given by

$$\mu(\vec{\phi}) = \sum_{k=1}^{N} \tilde{\mu}_k c_k(\vec{\phi}) = \frac{\Gamma_\mu}{\Gamma} + \sum_{i=1}^{N-1} \left(\tilde{\mu}_i - \frac{\Gamma_\mu}{\Gamma}\right)\tilde{\gamma}_i\left(\sum_{j=1}^{N-1} a_{ij}\phi_j + b_i\right). \tag{19}$$

The term $\tilde{\mathbf{J}}(\vec{\phi}, \nabla\vec{\phi})$ is given by

$$\tilde{\mathbf{J}}(\vec{\phi}, \nabla\vec{\phi}) = -\sum_{i=1}^{N-1} \left(1 - \frac{N}{\Gamma}\tilde{\gamma}_i\right)\left(\frac{\tilde{\rho}_i + \tilde{\rho}_N}{2}\right)^2 m_i \sum_{j=1}^{N-1} R_{ij} \nabla\left[-\sum_{k=1}^{N-1} \lambda_{jk}\nabla^2\phi_k + h_j(\vec{\phi})\right], \tag{20}$$

where the constants R_{ij} $(1 \leqslant i, j \leqslant N-1)$ are defined by

$$\left(\mathbf{A}_1^T\right)^{-1} = [R_{ij}]_{(N-1)\times(N-1)}. \tag{21}$$

The N-phase physical formulation given by (14a)–(14c), which is associated with the general order parameters defined by (13), is thermodynamically consistent (when $g_i = 0$), in the sense that this formulation stems from the phase field model given by (4a)–(4c), which in turn is derived based on conservations of mass/momentum and the second law of thermodynamics [10]. The $(N-1)$ phase field equations (14c) with $g_i = 0$ reflects the mass conservations for the individual fluid phases. Eq. (14a) reflects the momentum conservation of the mixture. The velocity $\mathbf{u}$ is the volume-averaged mixture velocity and can be shown to be divergence free [10]. This is reflected by Eq. (14b). We refer the reader to the appendix of [10] for detailed derivations of the general N-phase model based on the conservations of mass/momentum, Galilean invariance, and the second law of thermodynamics.

By using Eqs. (14c) with $g_i = 0$, one can show that the $\rho(\vec{\phi})$ and $\tilde{\mathbf{J}}(\vec{\phi}, \nabla\vec{\phi})$ given by (18) and (20) satisfy the relation

$$\frac{\partial \rho}{\partial t} + \mathbf{u}\cdot\nabla\rho = -\nabla\cdot\tilde{\mathbf{J}}. \tag{22}$$

Using the above relation, one can further show that the formulation given by (14a)–(14c) admits the following energy law, assuming that $g_i = 0$ in (14c) and that all surface fluxes vanish on the domain boundary,

$$\frac{\partial}{\partial t}\int_\Omega \left[\frac{1}{2}\rho(\vec{\phi})|\mathbf{u}|^2 + W(\vec{\phi}, \nabla\vec{\phi})\right] = -\int_\Omega \frac{\mu(\vec{\phi})}{2}\left\|\mathbf{D}(\mathbf{u})\right\|^2 - \sum_{i=1}^{N-1} m_i \left(\frac{\tilde{\rho}_i + \tilde{\rho}_N}{2}\right)^2 \int_\Omega |\nabla C_i|^2 + \int_\Omega \mathbf{f}\cdot\mathbf{u}, \tag{23}$$

where

$$C_i = \sum_{j=1}^{N-1} R_{ij}\left[-\sum_{k=1}^{N-1} \lambda_{jk}\nabla^2\phi_k + h_j(\vec{\phi})\right], \quad 1 \leqslant i \leqslant N-1, \tag{24}$$

are the chemical potentials.

It is instructive to compare the current N-phase formulation given by Eqs. (14a)–(14c) with that of [10]. The formulation of [10] is based on a special set of order parameters for simplifying the form of the phase field equations, specifically as follows:

$$\varphi_i(\vec{\phi}) = \rho_i(\vec{\phi}) - \rho_N(\vec{\phi}) = \frac{1}{2}(\tilde{\rho}_i - \tilde{\rho}_N) + \frac{1}{2}(\tilde{\rho}_i + \tilde{\rho}_N)\phi_i, \quad 1 \leqslant i \leqslant N-1. \tag{25}$$

This set is a special case of (13), corresponding to

$$a_{ij} = \frac{1}{2}(\tilde{\rho}_i + \tilde{\rho}_N)\delta_{ij}, \qquad b_i = \frac{1}{2}(\tilde{\rho}_i - \tilde{\rho}_N), \quad 1 \leqslant i, j \leqslant N-1, \tag{26}$$

where δ_{ij} is the Kronecker delta. Because the matrix $\mathbf{A}_2$ defined in (15) for this case is diagonal, the form of the phase field equations (14c) becomes significantly simplified.

In contrast, the order parameters defined by (13) are in a general form. They give rise to the $\mathbf{A}_2$ matrix (d_{ij} terms) in (14c). This causes the $(N-1)$ phase field equations (14c) to couple with one another in an extremely strong fashion, which presents new challenges to the design of numerical algorithms for solving these equations. Despite the increased complexity in the form of the phase field equations, the general order parameters provide a crucial advantage. The general form (13) encompasses a certain order-parameter set, with which the determination of the mixing energy density coefficients λ_{ij} based on the pairwise surface tensions will be dramatically simplified. This enables us, for *any* set of order parameters defined by (13), to express λ_{ij} in *explicit forms* in terms of the pairwise surface tensions (see Section 2.2). This eliminates the need for solving a linear algebraic system for λ_{ij}, as with the method discussed in [10]. In addition, we will show in Section 3.1 that, the increased complexity in the form of the phase field equations (14c) actually does not cause essential computational difficulties. We will present a numerical algorithm for (14c) that involves a computational complexity comparable to that for the simplified phase field equations in [10] corresponding to the special set of order parameters defined by (25).

We next briefly mention several specific sets of order parameters as illustrations of the general form given by (13):

- Volume fractions as order parameters. Let
$$\phi_i = c_i, \quad \phi_i \in [0,1], \; 1 \leqslant i \leqslant N-1 \tag{27}$$
be the order parameters, where c_i is the volume fraction of fluid i within the mixture. Then $c_N = 1 - \sum_{i=1}^{N-1}\phi_i$. Consequently, $\varphi_i = \rho_i - \rho_N = \tilde{\rho}_i c_i - \tilde{\rho}_N c_N = \sum_{j=1}^{N-1}(\tilde{\rho}_i\delta_{ij} + \tilde{\rho}_N)\phi_j - \tilde{\rho}_N$. The matrices $\mathbf{A}_1$ and $\mathbf{b}_1$ in (12) are therefore given by
$$a_{ij} = \tilde{\rho}_i\delta_{ij} + \tilde{\rho}_N, \qquad b_i = -\tilde{\rho}_N, \quad 1 \leqslant i,j \leqslant N-1. \tag{28}$$
- Re-scaled volume fraction differences as order parameters. Define the order parameters ϕ_i $(1 \leqslant i \leqslant N-1)$ by
$$2\phi_i - 1 = c_i - c_N, \quad \phi_i \in [0,1]. \tag{29}$$
By noting $\sum_{i=1}^{N} c_i = 1$, one can obtain $c_N = 1 - \frac{2}{N}\sum_{j=1}^{N-1}\phi_j$, $c_i = 2\phi_i - \frac{2}{N}\sum_{j=1}^{N-1}\phi_j$ $(1 \leqslant i \leqslant N-1)$. Consequently, $\mathbf{A}_1$ and $\mathbf{b}_1$ are given by
$$a_{ij} = 2\tilde{\rho}_i\delta_{ij} + \frac{2}{N}(\tilde{\rho}_N - \tilde{\rho}_i), \qquad b_i = -\tilde{\rho}_N, \quad 1 \leqslant i,j \leqslant N-1. \tag{30}$$
- Densities as order parameters. Let
$$\phi_i = \rho_i, \quad \phi_i \in [0, \tilde{\rho}_i], \; 1 \leqslant i \leqslant N-1 \tag{31}$$
be the order parameters. This is in fact a simple re-scaling to the case with volume fractions as order parameters, due to Eq. (2). Then $\mathbf{A}_1$ and $\mathbf{b}_1$ are given by
$$a_{ij} = \delta_{ij} + \frac{\tilde{\rho}_N}{\tilde{\rho}_j}, \qquad b_i = -\tilde{\rho}_N, \quad 1 \leqslant i,j \leqslant N-1. \tag{32}$$
- Re-scaled density differences as order parameters. Define the order parameters ϕ_i $(1 \leqslant i \leqslant N-1)$ by
$$\rho_i - \rho_{i+1} = -\tilde{\rho}_{i+1} + (\tilde{\rho}_i + \tilde{\rho}_{i+1})\phi_i, \quad \phi_i \in [0,1], \; 1 \leqslant i \leqslant N-1. \tag{33}$$
Then $\varphi_i = \rho_i - \rho_N = -\sum_{j=i}^{N-1}\tilde{\rho}_{j+1} + \sum_{j=i}^{N-1}(\tilde{\rho}_j + \tilde{\rho}_{j+1})\phi_j$ for $1 \leqslant i \leqslant N-1$. $\mathbf{A}_1$ and $\mathbf{b}_1$ are therefore given by
$$a_{ij} = \begin{cases} \tilde{\rho}_j + \tilde{\rho}_{j+1}, & i \leqslant j, \\ 0, & i > j, \end{cases} \qquad b_i = -\sum_{j=i}^{N-1}\tilde{\rho}_{j+1}, \quad 1 \leqslant i,j \leqslant N-1. \tag{34}$$
- Another set of order parameters. Define the order parameters ϕ_i by
$$\phi_i = \sum_{j=i}^{N-1}\rho_j - (N-i)\rho_N, \quad 1 \leqslant i \leqslant N-1. \tag{35}$$
Then $\varphi_i = \rho_i - \rho_N = \phi_i - \phi_{i+1}$ $(1 \leqslant i \leqslant N-2)$, and $\varphi_{N-1} = \rho_{N-1} - \rho_N = \phi_{N-1}$. So $\mathbf{A}_1$ and $\mathbf{b}_1$ are given by
$$a_{ij} = \delta_{ij} - \delta_{i+1,j}, \qquad b_i = 0, \quad 1 \leqslant i,j \leqslant N-1. \tag{36}$$

2.2. Mixing energy density coefficients λ_{ij} for general order parameters

The physical formulation (14a)–(14c) involves, noting the symmetry of matrix $\mathbf{A}$ in (11), $\frac{1}{2}N(N-1)$ independent mixing energy density coefficients λ_{ij}, which need to be determined based on other known physical parameters. In this section we derive *explicit expressions* of λ_{ij} in terms of the $\frac{1}{2}N(N-1)$ pairwise surface tensions among the N fluid components for the general order parameters defined in (13).

The result of this section, incidentally, also provides the explicit formulas of λ_{ij} for the set of special order parameters employed in [10]. Note that in [10], using the special set of order parameters (25), we obtained a system of $\frac{1}{2}N(N-1)$ linear algebraic equations about λ_{ij}, and then numerically solved that linear system to obtain λ_{ij}. Although numerical experiments in [10] indicate that the linear algebraic system for λ_{ij} always has a unique solution, the unique solvability of that system is an un-settled issue for general N $(N \geqslant 4)$ fluid phases.

In the following, we first obtain an explicit expression of λ_{ij} for the formulation with volume fractions as the order parameters. Then we generalize the result to formulations with the general order parameters defined by (13).

2.2.1. Volume fractions as order parameters

Let us first focus on the formulation with volume fractions as the order parameters. See Eq. (27) for the definition, and the coefficients a_{ij} and b_i for this formulation are given by (28). The formulation with volume fractions as the order parameters plays a special role when computing λ_{ij} among the general order parameters. To distinguish this formulation from those with the other order parameters, we use Λ_{ij} $(1 \leqslant i, j \leqslant N-1)$ to specifically denote the mixing energy density coefficients λ_{ij} for this formulation, and use

$$\boldsymbol{\Lambda} = [\Lambda_{ij}]_{(N-1)\times(N-1)} \tag{37}$$

to denote the matrix of mixing energy density coefficients of this formulation, in contrast with (11). In addition, we use $\boldsymbol{\Lambda}_1$ to specifically denote the $\mathbf{A}_1$ matrix for this formulation, that is,

$$\boldsymbol{\Lambda}_1 = [a_{ij}]_{(N-1)\times(N-1)}, \tag{38}$$

where a_{ij} are given by Eq. (28). One can verify that $\boldsymbol{\Lambda}_1$ is symmetric positive definite.

To determine Λ_{ij} for the N-phase system, we employ an idea similar to that of [10], namely, by imposing the following consistency requirement on the N-phase formulation. We recognize that in the N-phase system, if only a pair of two fluids is present (for any fluid pair) while all the other fluids are absent, then the N-phase system is equivalent to a two-phase system consisting of these two fluids. Accordingly, for such a situation, the N-phase formulation should reduce to the two-phase formulation for the equivalent two-phase system. In particular, the free-energy density function for the N-phase system should reduce to that of the equivalent two-phase system.

Note that for a two-phase system $(N = 2)$ the relation between the mixing energy density coefficient and the surface tension is well-known. Let

$$\phi_1 = c_1 - c_2 \tag{39}$$

denote the sole order parameter of the two-phase system, where c_1 and c_2 are the volume fractions of the two fluids. Then $c_1 = \frac{1}{2}(1+\phi_1)$ and $c_2 = \frac{1}{2}(1-\phi_1)$. The free energy density function (10) is then reduced to

$$W(\phi_1, \nabla\phi_1) = \frac{\lambda_{11}}{2}\nabla\phi_1 \cdot \nabla\phi_1 + \frac{\beta^2}{16\eta^2}\left(1-\phi_1^2\right)^2. \tag{40}$$

In a one-dimensional setting, by requiring that at equilibrium the integral of the above free-energy density across the interface should equal the surface tension, one can obtain the relation (see [46,10] for details)

$$\lambda_{11} = \frac{9}{2}\frac{\eta^2}{\beta^2}\sigma_{12}^2, \tag{41}$$

where σ_{12} denotes the surface tension between fluids 1 and 2 of the two-phase system. Therefore, for a two-phase system with the free energy density function given by (40), the mixing energy density coefficient is given by (41).

To determine Λ_{ij} with N fluid phases, let us assume that fluids k and l $(1 \leqslant k < l \leqslant N)$ are the only two fluids that are present in the N-phase system, that is,

$$\rho_i \equiv 0, \qquad c_i \equiv 0, \quad \text{if } i \neq k \text{ and } i \neq l, \text{ for } 1 \leqslant i \leqslant N. \tag{42}$$

Equivalently, this N-phase system can be considered as a two-phase system consisting of fluids k and l. Therefore, this system has only one independent order parameter. Noting the form of Eq. (39), we use

$$\phi_a = c_k - c_l \tag{43}$$

to denote the sole independent order parameter of this N-phase system. Then

$$c_k = \frac{1}{2}(1+\phi_a), \qquad c_l = \frac{1}{2}(1-\phi_a), \tag{44}$$

by noting $\sum_{i=1}^{N} c_i = 1$ and the condition (42).

We will distinguish two cases: (1) $l = N$, and (2) $l < N$. In the first case $1 \leqslant k < l = N$, the free energy density function (10) becomes

$$\begin{aligned} W &= \sum_{i,j=1}^{N-1}\frac{\Lambda_{ij}}{2}\nabla\phi_i \cdot \nabla\phi_j + \frac{\beta^2}{2\eta^2}\sum_{i=1}^{N} c_i^2(1-c_i)^2 \\ &= \frac{\Lambda_{kk}}{2}\nabla\phi_k \cdot \nabla\phi_k + \frac{\beta^2}{2\eta^2}\left[c_k^2(1-c_k)^2 + c_l^2(1-c_l)^2\right] \\ &= \frac{\Lambda_{kk}}{8}\nabla\phi_a \cdot \nabla\phi_a + \frac{\beta^2}{16\eta^2}\left(1-\phi_a^2\right)^2, \end{aligned} \tag{45}$$

where we have used Eqs. (27), (42) and (44). Comparing the above equation with the two-phase free energy density function (40) and using Eq. (41), we have

$$\Lambda_{kk} = \frac{18\eta^2}{\beta^2}\sigma_{kN}^2, \quad 1 \leqslant k < l = N, \tag{46}$$

where σ_{ij} $(i \neq j)$ denotes the surface tension associated with the interface formed between fluid i and fluid j.

For the second case $1 \leqslant k < l < N$, the N-phase free energy density function (10) is transformed into

$$\begin{aligned} W &= \sum_{i,j=1}^{N-1} \frac{\Lambda_{ij}}{2}\nabla\phi_i \cdot \nabla\phi_j + \frac{\beta^2}{2\eta^2}\sum_{i=1}^{N} c_i^2(1-c_i)^2 \\ &= \frac{\Lambda_{kk}}{2}\nabla\phi_k \cdot \nabla\phi_k + \frac{\Lambda_{ll}}{2}\nabla\phi_l \cdot \nabla\phi_l + \Lambda_{kl}\nabla\phi_k \cdot \nabla\phi_l + \frac{\beta^2}{2\eta^2}\left[c_k^2(1-c_k)^2 + c_l^2(1-c_l)^2\right] \\ &= \frac{1}{8}(\Lambda_{kk} + \Lambda_{ll} - 2\Lambda_{kl})\nabla\phi_a \cdot \nabla\phi_a + \frac{\beta^2}{16\eta^2}\left(1-\phi_a^2\right)^2 \end{aligned} \tag{47}$$

where we have used the symmetry of the matrix Λ and Eqs. (27), (42), and (44). Compare the above equation with Eq. (40) and use Eq. (41), and one can get

$$\frac{1}{4}(\Lambda_{kk} + \Lambda_{ll} - 2\Lambda_{kl}) = \frac{9}{2}\frac{\eta^2}{\beta^2}\sigma_{kl}^2, \quad 1 \leqslant k < l \leqslant N-1. \tag{48}$$

By using the expression (46) we have

$$\Lambda_{kl} = \Lambda_{lk} = \frac{9\eta^2}{\beta^2}\left(\sigma_{kN}^2 + \sigma_{lN}^2 - \sigma_{kl}^2\right), \quad 1 \leqslant k < l \leqslant N-1. \tag{49}$$

Therefore, with the volume fractions as the order parameters, the N-phase mixing energy density coefficients Λ_{kl} $(1 \leqslant k, l \leqslant N-1)$ are given by the explicit expressions, (46) and (49), in terms of the pairwise surface tensions σ_{ij} $(1 \leqslant i < j \leqslant N)$ among the N fluids.

2.2.2. General order parameters

Let us now consider physical formulations with the general order parameters defined by (13). Based on Eq. (17) we have the following relation

$$\nabla c_k = \sum_{i=1}^{N-1}\tilde{\gamma}_k\frac{\partial\rho_k}{\partial\phi_i}\nabla\phi_i = \sum_{i=1}^{N-1} y_{ki}\nabla\phi_i, \quad 1 \leqslant k \leqslant N. \tag{50}$$

The coefficients y_{ki} in the above equation are given by

$$y_{ki} = \sum_{j=1}^{N-1} e_{kj}a_{ji}, \quad 1 \leqslant k \leqslant N,\ 1 \leqslant i \leqslant N-1, \tag{51}$$

where

$$e_{ki} = \tilde{\gamma}_k\delta_{ki} - \frac{\tilde{\gamma}_k\tilde{\gamma}_i}{\Gamma}, \quad 1 \leqslant k \leqslant N,\ 1 \leqslant i \leqslant N-1. \tag{52}$$

Let

$$\mathbf{Y} = [y_{ij}]_{(N-1)\times(N-1)}, \qquad \mathbf{Z} = [e_{ij}]_{(N-1)\times(N-1)} \tag{53}$$

respectively denote the *square* matrices formed by y_{ij} $(1 \leqslant i, j \leqslant N-1)$ and e_{ij} $(1 \leqslant i, j \leqslant N-1)$. Then the following matrix form represents a subset of the equations in (51),

$$\mathbf{Y} = \mathbf{Z}\mathbf{A}_1, \tag{54}$$

where $\mathbf{A}_1$ is defined in (12). It is straightforward to verify that

$$\mathbf{Z} = \boldsymbol{\Lambda}_1^{-1}, \tag{55}$$

where $\boldsymbol{\Lambda}_1$ is defined in (38). The matrix $\mathbf{Y}$ is therefore non-singular.

In order to determine the mixing energy density coefficients λ_{ij}, we recognize the following point about the order parameters. The physical formulations employing different sets of order parameters are merely different representations of the N-phase system, and the different representations should be equivalent. In particular, the N-phase free energy density function can be represented in terms of any set of independent order parameters, and these representations should be equivalent. This is an embodiment of the representation invariance principle [32].

In light of the above point, we can re-write the free energy density function of the N-phase system (10) as

$$W = \sum_{i,j=1}^{N-1} \frac{\lambda_{ij}}{2} \nabla\phi_i \cdot \nabla\phi_j + \frac{\beta^2}{2\eta^2} \sum_{k=1}^{N} c_k^2(\vec{\phi})\left[1 - c_k(\vec{\phi})\right]^2$$
$$= \sum_{i,j=1}^{N-1} \frac{\Lambda_{ij}}{2} \nabla c_i \cdot \nabla c_j + \frac{\beta^2}{2\eta^2} \sum_{k=1}^{N} c_k^2(1 - c_k)^2. \tag{56}$$

In the above equation we have expressed the N-phase free energy density function in terms of the order parameters ϕ_i ($1 \leqslant i \leqslant N-1$), as well as in terms of the volume fractions c_i ($1 \leqslant i \leqslant N-1$), and we have used the results from Section 2.2.1. Using the relation in (50), we can obtain λ_{ij} from (56),

$$\lambda_{ij} = \sum_{k,l=1}^{N-1} y_{ki} y_{lj} \Lambda_{kl}, \quad 1 \leqslant i, j \leqslant N-1, \tag{57}$$

where Λ_{kl} ($1 \leqslant k, l \leqslant N-1$) are given by (46) and (49). Equivalently, the matrix form is

$$\mathbf{A} = \mathbf{Y}^T \boldsymbol{\Lambda} \mathbf{Y}, \tag{58}$$

where $\mathbf{A}$ and $\boldsymbol{\Lambda}$ are defined in (11) and (37) respectively. Eq. (57) or (58) provides the explicit forms for the mixing energy density coefficients λ_{ij} for general order parameters defined by (13).

Based on Eq. (58) we have the following observations:

- If the matrix $\boldsymbol{\Lambda}$ is SPD, then the matrix $\mathbf{A}$ for any set of order parameters defined by (13) is SPD. More generally, if the matrix $\mathbf{A}$ is SPD with one set of order parameters defined by (13), then it is SPD with all sets of order parameters defined by (13).
- The positive definiteness of the matrix $\mathbf{A}$ for general order parameters is only affected by the pairwise surface tensions σ_{ij} among the N fluids. The fluid densities $\tilde{\rho}_i$ affect the values of $\mathbf{A}$, but have no effect on its positive definiteness. This is because the dependency on σ_{ij} is through $\boldsymbol{\Lambda}$ and the dependency on $\tilde{\rho}_i$ is through $\mathbf{Y}$.
- Given a set of arbitrary positive values for the pairwise surface tensions $\sigma_{ij} > 0$ ($1 \leqslant i < j \leqslant N$), the matrix $\boldsymbol{\Lambda}$ is always symmetric, but may not be positive definite. What conditions on σ_{ij} will ensure the SPD of the matrix $\boldsymbol{\Lambda}$ is currently an open question. Numerical experiments in [10] suggest that, if the pairwise surface-tension values are such that total wetting occurs among some three-tuple of fluids among these N fluids, then the matrix $\mathbf{A}$ will have a negative eigenvalue and therefore will not be positive definite.
- The mixing energy density coefficients λ_{ij} for the N-phase formulation employed in [10] are given by the following explicit expression

  $$\mathbf{A} = [\lambda_{ij}]_{(N-1)\times(N-1)} = \mathbf{L}\mathbf{Z}\boldsymbol{\Lambda}\mathbf{Z}\mathbf{L}, \quad \mathbf{L} = \mathrm{diag}\left(\frac{\tilde{\rho}_1 + \tilde{\rho}_N}{2}, \frac{\tilde{\rho}_2 + \tilde{\rho}_N}{2}, \ldots, \frac{\tilde{\rho}_{N-1} + \tilde{\rho}_N}{2}\right). \tag{59}$$

  Note that in [10] λ_{ij} are obtained by solving a linear algebraic system. In Appendix A, we provide a proof that the λ_{ij} computed based on (59) indeed are the solution to the system of linear algebraic equations about λ_{ij} derived in [10].

Remarks In the N-phase formulation the dynamic variables are the velocity $\mathbf{u}$, the pressure p, and the $(N-1)$ phase field variables ϕ_i. Eqs. (14a)–(14c) are the governing equations to be solved for these variables. Once a specific set of order parameters is chosen, i.e. by specifying a_{ij} ($1 \leqslant i, j \leqslant N-1$) and b_i ($1 \leqslant i \leqslant N-1$), the coefficients d_{ij} ($1 \leqslant i, j \leqslant N-1$) in Eq. (14c) can be computed based on Eq. (15) and the $(N-1)$ mobility coefficients m_i that also need to be specified. The mixing energy density coefficients λ_{ij} ($1 \leqslant i, j \leqslant N-1$) in (14c) can be computed from Eq. (58) based on the pairwise surface tensions among the N fluids.

On the other hand, the dependence relations of the densities $\rho_i(\vec{\phi})$ ($1 \leqslant i \leqslant N$) and $\rho(\vec{\phi})$, the volume fractions $c_i(\vec{\phi})$ ($1 \leqslant i \leqslant N$), and the dynamic viscosity $\mu(\vec{\phi})$ in terms of $\vec{\phi}$ are known a priori as part of the model. Once the phase field variables ϕ_i ($1 \leqslant i \leqslant N-1$) are computed, one can use the second part of Eq. (17) to compute the density of fluid i within the mixture, $\rho_i(\vec{\phi})$ ($1 \leqslant i \leqslant N$), and then use the first part of this equation to compute the volume fraction of fluid i, $c_i(\vec{\phi})$ ($1 \leqslant i \leqslant N$). The mixture density $\rho(\vec{\phi})$ and the mixture dynamic viscosity $\mu(\vec{\phi})$ can be computed from Eqs. (18) and (19), respectively. The functions $\varphi_i(\vec{\phi})$ ($1 \leqslant i \leqslant N-1$) can be computed from Eq. (13), which is the definition for the order parameters in the model.

For the parameter β in the free energy density function (10), we will use the following expression (see [10])

$$\beta = \sqrt{3\sqrt{2}\sigma_{\min}\eta}, \tag{60}$$

Table 1
Field variables and parameters of different types in the N-phase physical formulation.

Input parameters/data	Variables to be solved	To be computed/post-processed	
		Variables/Parameters	Equation
a_{ij} $(1 \leqslant i, j \leqslant N-1)$	$\mathbf{u}$	d_{ij} $(1 \leqslant i, j \leqslant N-1)$	(15)
b_i $(1 \leqslant i \leqslant N-1)$	p	R_{ij} $(1 \leqslant i, j \leqslant N-1)$	(21)
$\tilde{\rho}_i$ $(1 \leqslant i \leqslant N)$	ϕ_i $(1 \leqslant i \leqslant N-1)$	λ_{ij} $(1 \leqslant i, j \leqslant N-1)$	(58)
$\tilde{\mu}_i$ $(1 \leqslant i \leqslant N)$		$\varphi_i(\vec{\phi})$ $(1 \leqslant i \leqslant N-1)$	(13)
σ_{ij} $(1 \leqslant i < j \leqslant N)$		$\rho_i(\vec{\phi})$ $(1 \leqslant i \leqslant N)$	(17)
η		$c_i(\vec{\phi})$ $(1 \leqslant i \leqslant N)$	(17)
m_i $(1 \leqslant i \leqslant N-1)$		$\rho(\vec{\phi})$	(18)
$\mathbf{f}$ (external body force)		$\mu(\vec{\phi})$	(19)
		β	(60)

where $\sigma_{\min} = \min\{\sigma_{ij}\}_{1 \leqslant i < j \leqslant N}$ denotes the minimum of the $\frac{1}{2}N(N-1)$ pairwise surface tensions among the N fluids. With this choice of β, the parameter η corresponds to the characteristic interfacial thickness of the interface associated with the minimum pairwise surface tension $\sigma_{\min}$.

One can observe that the N-phase physical formulation involves quite a few physical variables and parameters, which play different roles and need to be dealt with differently in numerical simulations. We have classified these variables into three categories: (i) input parameters or data, (ii) independent dynamic variables that need to be solved, and (iii) parameters and field variables that can be computed or post-processed based on the dynamic variables. Table 1 is a summary of the main parameters and variables in each category. For the variables and parameters that can be post-processed we have also listed the equations to be used for the computations.

The governing equations (14a)–(14c) need to be supplemented by appropriate boundary conditions and initial conditions for the velocity and phase field functions. In the current paper we consider the Dirichlet boundary condition for the velocity,

$$\mathbf{u}|_{\partial\Omega} = \mathbf{w}(\mathbf{x}, t), \tag{61}$$

where $\mathbf{w}$ is the boundary velocity, and the following boundary conditions for the phase field functions,

$$\mathbf{n} \cdot \nabla\left(\nabla^2 \phi_i\right)|_{\partial\Omega} = 0, \quad 1 \leqslant i \leqslant N-1, \tag{62a}$$

$$\mathbf{n} \cdot \nabla \phi_i|_{\partial\Omega} = 0, \quad 1 \leqslant i \leqslant N-1. \tag{62b}$$

The boundary conditions (62a) and (62b) correspond to a wall with neutral wettability, that is, if any fluid interface intersects the boundary wall, the contact angle formed between the interface and the wall shall be 90°.

3. Numerical algorithm

The governing equations (14a)–(14c), the boundary conditions (61)–(62b), and the appropriate initial conditions for the velocity $\mathbf{u}$ and the phase field variables ϕ_i $(1 \leqslant i \leqslant N-1)$ together constitute the system that need to be solved in numerical simulations.

In this section we present a numerical algorithm for solving this system. The primary challenge here lies in the system of $(N-1)$ phase field equations (14c). This system is extremely strongly-coupled. In particular, in addition to the couplings on the right side due to the terms $\lambda_{ij}\nabla^2\phi_j$ and $h_i(\vec{\phi})$, the inertia terms $\frac{\partial \phi_i}{\partial t}$ on the left hand are also coupled with one another due to the $\mathbf{A}_2$ matrix.

We will first concentrate on the numerical treatment of the coupled system of $(N-1)$ phase field equations (14c). Our algorithm will, after discretization, reduce this strongly-coupled system of fourth-order equations into $(N-1)$ *de-coupled individual* fourth-order equations, each of which can then be further reduced into two de-coupled Helmholtz-type equations using a technique originally developed for two-phase field equations.

Then we will discuss a time-marching scheme for the overall system, and provide the overall solution procedure. In particular, we present an algorithm in Appendix B for the N-phase momentum equations, (14a) and (14b). Our algorithm overcomes the numerical difficulties associated with the variable density and variable dynamic viscosity of the N-phase mixture, by using a strategy initially developed in [14] for two-phase flows.

3.1. Algorithm for coupled phase-field equations with general order parameters

Let us focus on how to numerically solve the system of $(N-1)$ coupled phase-field equations, (14c), together with the boundary conditions, (62a) and (62b). Let n denote the time step index, and $(\cdot)^n$ denote the variable $(\cdot)$ at time step n. We assume that $\mathbf{u}^n$ and ϕ_i^n $(1 \leqslant i \leqslant N-1)$ are known.

We discretize the coupled phase-field equations and the boundary conditions in time as follows,

$$\sum_{j=1}^{N-1} d_{ij}\left(\frac{\gamma_0\phi_j^{n+1}-\hat{\phi}_j}{\Delta t}+\mathbf{u}^{*,n+1}\cdot\nabla\phi_j^{*,n+1}\right)$$
$$=\nabla^2\left[-\sum_{j=1}^{N-1}\lambda_{ij}\nabla^2\phi_j^{n+1}+\frac{1}{\eta^2}\sum_{j=1}^{N-1}S_{ij}\left(\phi_j^{n+1}-\phi_j^{*,n+1}\right)+h_i\left(\vec{\phi}^{*,n+1}\right)\right]+g_i^{n+1},\quad 1\leqslant i\leqslant N-1, \tag{63a}$$

$$\mathbf{n}\cdot\nabla\left(\nabla^2\phi_i^{n+1}\right)\big|_{\partial\Omega}=0,\quad 1\leqslant i\leqslant N-1, \tag{63b}$$

$$\mathbf{n}\cdot\nabla\phi_i^{n+1}\big|_{\partial\Omega}=0,\quad 1\leqslant i\leqslant N-1. \tag{63c}$$

In the above equations, Δt is the time step size, $\mathbf{n}$ is an outward-pointing unit vector normal to $\partial\Omega$, and S_{ij} $(1\leqslant i,j\leqslant N-1)$ are $(N-1)^2$ chosen constants to be determined below. Let J ($J=1$ or 2) denote the order of temporal accuracy, and χ denote a generic variable. Then $\chi^{*,n+1}$ represents a Jth order explicit approximation of χ^{n+1} given by

$$\chi^{*,n+1}=\begin{cases}\chi^n, & J=1\\ 2\chi^n-\chi^{n-1}, & J=2.\end{cases} \tag{64}$$

$\frac{1}{\Delta t}(\gamma_0\chi^{n+1}-\hat{\chi})$ represents an approximation of $\frac{\partial\chi}{\partial t}|^{n+1}$ by a Jth order backward differentiation formula, and $\hat{\chi}$ and γ_0 are given by

$$\hat{\chi}=\begin{cases}\chi^n, & J=1\\ 2\chi^n-\frac{1}{2}\chi^{n-1}, & J=2,\end{cases}\qquad \gamma_0=\begin{cases}1, & J=1\\ \frac{3}{2}, & J=2.\end{cases} \tag{65}$$

$\vec{\phi}^{*,n+1}$ denotes the vector of $\phi_i^{*,n+1}$ $(1\leqslant i\leqslant N-1)$.

Eq. (63a) represents a set of $(N-1)$ fourth-order equations about ϕ_i^{n+1} $(1\leqslant i\leqslant N-1)$ that are strongly coupled with one another. The $(N-1)^2$ extra terms $\sum_{j=1}^{N-1}S_{ij}(\phi_j^{n+1}-\phi_j^{*,n+1})$ in the discrete form (63a) are critical to the current algorithm. After we transform (63a) into $(N-1)$ de-coupled individual fourth-order equations, these extra terms make it possible to re-formulate each individual fourth-order equation into two de-coupled Helmholtz-type (2nd order) equations, which can be discretized in space using C^0 spectral elements or finite elements in a straightforward fashion.

Re-write (63a) as follows,

$$\sum_{j=1}^{N-1}\lambda_{ij}\nabla^2\left(\nabla^2\phi_j^{n+1}\right)-\frac{1}{\eta^2}\sum_{j=1}^{N-1}S_{ij}\nabla^2\phi_j^{n+1}+\frac{\gamma_0}{\Delta t}\sum_{j=1}^{N-1}d_{ij}\phi_j^{n+1}=Q_i=Q_i^{(1)}+\nabla^2 Q_i^{(2)},\quad 1\leqslant i\leqslant N-1, \tag{66}$$

where

$$\begin{cases}Q_i^{(1)}=g_i^{n+1}-\displaystyle\sum_{j=1}^{N-1}d_{ij}\left(-\frac{1}{\Delta t}\hat{\phi}_j+\mathbf{u}^{*,n+1}\cdot\nabla\phi_j^{*,n+1}\right)\\ Q_i^{(2)}=h_i\left(\vec{\phi}^{*,n+1}\right)-\dfrac{1}{\eta^2}\displaystyle\sum_{j=1}^{N-1}S_{ij}\phi_j^{*,n+1}.\end{cases} \tag{67}$$

We introduce the following vectors and matrix

$$\begin{cases}\boldsymbol{\Phi}=\begin{bmatrix}\vdots\\ \phi_i^{n+1}\\ \vdots\end{bmatrix}_{(N-1)\times 1},\quad \mathbf{Q}=\begin{bmatrix}\vdots\\ Q_i\\ \vdots\end{bmatrix}_{(N-1)\times 1},\quad \mathbf{Q}^{(1)}=\begin{bmatrix}\vdots\\ Q_i^{(1)}\\ \vdots\end{bmatrix}_{(N-1)\times 1},\quad \mathbf{Q}^{(2)}=\begin{bmatrix}\vdots\\ Q_i^{(2)}\\ \vdots\end{bmatrix}_{(N-1)\times 1},\\ \mathbf{S}=[S_{ij}]_{(N-1)\times(N-1)}.\end{cases} \tag{68}$$

Then the equations in (66) is equivalent to the following matrix form

$$\mathbf{A}\nabla^2\left(\nabla^2\boldsymbol{\Phi}\right)-\frac{1}{\eta^2}\mathbf{S}\nabla^2\boldsymbol{\Phi}+\frac{\gamma_0}{\Delta t}\mathbf{A}_2\boldsymbol{\Phi}=\mathbf{Q}, \tag{69}$$

where the matrices $\mathbf{A}$ and $\mathbf{A}_2$ are given by (11) and (15), and note that both matrices are symmetric positive definite.

Because it is SPD, the matrix $\mathbf{A}_2$ can be diagonalized as follows,

$$\mathbf{T}^T\mathbf{A}_2\mathbf{T}=\mathbf{E}=\operatorname{diag}(\hat{a}_1,\hat{a}_2,\ldots,\hat{a}_{N-1}),\quad \mathbf{A}_2=\mathbf{T}\mathbf{E}\mathbf{T}^T, \tag{70}$$

where $\hat{a}_i>0$ $(1\leqslant i\leqslant N-1)$ are the eigenvalues of $\mathbf{A}_2$, $\mathbf{E}$ is the diagonal matrix of $\hat{a}_i$, $\mathbf{T}$ is the orthogonal matrix formed by the eigenvectors of $\mathbf{A}_2$, and note that $\mathbf{T}^{-1}=\mathbf{T}^T$.

Using the expression of $\mathbf{A}_2$ in (70), we can transform (69) into

$$\mathbf{B}_1\nabla^2\left(\nabla^2\boldsymbol{\Phi}_1\right) - \frac{1}{\eta^2}\mathbf{S}_1\nabla^2\boldsymbol{\Phi}_1 + \frac{\gamma_0}{\Delta t}\mathbf{E}\boldsymbol{\Phi}_1 = \mathbf{T}^T\mathbf{Q}, \tag{71}$$

where

$$\mathbf{B}_1 = \mathbf{T}^T\mathbf{A}\mathbf{T}, \qquad \mathbf{S}_1 = \mathbf{T}^T\mathbf{S}\mathbf{T}, \qquad \boldsymbol{\Phi}_1 = \mathbf{T}^T\boldsymbol{\Phi}. \tag{72}$$

Let

$$\mathbf{D} = \sqrt{\frac{\gamma_0}{\Delta t}}\mathbf{E}^{\frac{1}{2}}, \qquad \frac{\gamma_0}{\Delta t}\mathbf{E} = \mathbf{D}^2, \tag{73}$$

where the exponential in $\mathbf{E}^{\frac{1}{2}}$ is understood to be element-wise operations applied to the diagonal elements, and note that $\mathbf{D}$ is a diagonal matrix. Therefore, we can transform (71) into

$$\mathbf{B}_2\nabla^2\left(\nabla^2\boldsymbol{\Phi}_2\right) - \frac{1}{\eta^2}\mathbf{S}_2\nabla^2\boldsymbol{\Phi}_2 + \boldsymbol{\Phi}_2 = \mathbf{D}^{-1}\mathbf{T}^T\mathbf{Q}, \tag{74}$$

where

$$\mathbf{B}_2 = \mathbf{D}^{-1}\mathbf{B}_1\mathbf{D}^{-1}, \qquad \mathbf{S}_2 = \mathbf{D}^{-1}\mathbf{S}_1\mathbf{D}^{-1}, \qquad \boldsymbol{\Phi}_2 = \mathbf{D}\boldsymbol{\Phi}_1. \tag{75}$$

It is straightforward to verify that $\mathbf{B}_2$ is SPD because $\mathbf{A}$ is SPD.

Because $\mathbf{B}_2$ is SPD, it can be diagonalized as follows,

$$\mathbf{P}^T\mathbf{B}_2\mathbf{P} = \mathbf{K} = \mathrm{diag}(\hat{\lambda}_1, \hat{\lambda}_2, \ldots, \hat{\lambda}_{N-1}), \quad \mathbf{B}_2 = \mathbf{P}\mathbf{K}\mathbf{P}^T, \tag{76}$$

where $\hat{\lambda}_i > 0$ $(1 \leqslant i \leqslant N-1)$ are the eigenvalues of $\mathbf{B}_2$, $\mathbf{K}$ is the diagonal matrix of $\hat{\lambda}_i$, $\mathbf{P}$ is the orthogonal matrix formed by the eigenvectors of $\mathbf{B}_2$, and note that $\mathbf{P}^{-1} = \mathbf{P}^T$.

Now we choose S_{ij} $(1 \leqslant i, j \leqslant N-1)$ in (66) such that

$$\mathbf{P}^T\mathbf{S}_2\mathbf{P} = \hat{\mathbf{S}} = \mathrm{diag}(\hat{s}_1, \hat{s}_2, \ldots, \hat{s}_{N-1}), \quad \mathbf{S}_2 = \mathbf{P}\hat{\mathbf{S}}\mathbf{P}^T, \tag{77}$$

where $\hat{s}_i$ $(1 \leqslant i \leqslant N-1)$ are $(N-1)$ chosen constants to be determined below.

Using the expression for $\mathbf{B}_2$ in (76) and the expression for $\mathbf{S}_2$ in (77), we can transform (74) into

$$\begin{aligned}\mathbf{K}\nabla^2\left(\nabla^2\mathbf{X}\right) - \frac{1}{\eta^2}\hat{\mathbf{S}}\nabla^2\mathbf{X} + \mathbf{X} &= \mathbf{P}^T\mathbf{D}^{-1}\mathbf{T}^T\mathbf{Q} \\ &= \left(\mathbf{P}^T\mathbf{D}^{-1}\mathbf{T}^T\right)\mathbf{Q}^{(1)} + \left(\mathbf{P}^T\mathbf{D}^{-1}\mathbf{T}^T\right)\nabla^2\mathbf{Q}^{(2)}\end{aligned} \tag{78}$$

where

$$\mathbf{X} = \mathbf{P}^T\boldsymbol{\Phi}_2, \quad \boldsymbol{\Phi}_2 = \mathbf{P}\mathbf{X}. \tag{79}$$

Let

$$\begin{cases} \mathbf{X} = \begin{bmatrix} \vdots \\ \xi_i^{n+1} \\ \vdots \end{bmatrix}_{(N-1)\times 1}, \quad \mathbf{P}^T\mathbf{D}^{-1}\mathbf{T}^T\mathbf{Q} = \begin{bmatrix} \vdots \\ q_i \\ \vdots \end{bmatrix}_{(N-1)\times 1}, \quad \mathbf{P}^T\mathbf{D}^{-1}\mathbf{T}^T\mathbf{Q}^{(1)} = \begin{bmatrix} \vdots \\ q_i^{(1)} \\ \vdots \end{bmatrix}_{(N-1)\times 1}, \\ \mathbf{P}^T\mathbf{D}^{-1}\mathbf{T}^T\mathbf{Q}^{(2)} = \begin{bmatrix} \vdots \\ q_i^{(2)} \\ \vdots \end{bmatrix}_{(N-1)\times 1}. \end{cases} \tag{80}$$

Then (78) can be written into $(N-1)$ de-coupled individual equations in terms of the components,

$$\nabla^2\left(\nabla^2\xi_i^{n+1}\right) - \frac{\hat{s}_i}{\hat{\lambda}_i\eta^2}\nabla^2\xi_i^{n+1} + \frac{1}{\hat{\lambda}_i}\xi_i^{n+1} = \frac{1}{\hat{\lambda}_i}q_i = \frac{1}{\hat{\lambda}_i}q_i^{(1)} + \frac{1}{\hat{\lambda}_i}\nabla^2 q_i^{(2)}, \quad 1 \leqslant i \leqslant N-1. \tag{81}$$

We have now transformed the system of strongly-coupled fourth-order equations (66) into $(N-1)$ de-coupled fourth-order scalar equations (81).

Each equation in (81) has the same form as that of [10], and therefore can be dealt with in a similar manner. They each can be re-formulated into two de-coupled Helmholtz-type equations using a technique originated from two-phase flows [46,14,9]. We provide below only the final re-formulated equations; see [14,10] for the process of reformulations.

The final reformulated forms for (81) are,

$$\nabla^2\psi_i^{n+1} - \frac{1}{\hat{\lambda}_i}\left(\alpha_i + \frac{\hat{s}_i}{\eta^2}\right)\psi_i^{n+1} = \frac{1}{\hat{\lambda}_i}q_i, \quad 1 \leqslant i \leqslant N-1, \tag{82a}$$

$$\nabla^2\xi_i^{n+1} + \frac{\alpha_i}{\hat{\lambda}_i}\xi_i^{n+1} = \psi_i^{n+1}, \quad 1 \leqslant i \leqslant N-1, \tag{82b}$$

where ψ_i^{n+1} $(1 \leqslant i \leqslant N-1)$ are auxiliary variables defined by (82b), α_i $(1 \leqslant i \leqslant N-1)$ are constants given by

$$\alpha_i = \frac{\hat{s}_i}{2\eta^2}\left(-1 - \sqrt{1 - 4\hat{\lambda}_i\frac{\eta^4}{\hat{s}_i^2}}\right), \quad \text{or} \quad \alpha_i = \frac{\hat{s}_i}{2\eta^2}\left(-1 + \sqrt{1 - 4\hat{\lambda}_i\frac{\eta^4}{\hat{s}_i^2}}\right), \quad 1 \leqslant i \leqslant N-1, \tag{83}$$

and $\hat{s}_i$ $(1 \leqslant i \leqslant N-1)$ are $(N-1)$ chosen constants that must satisfy the condition,

$$\hat{s}_i \geqslant 2\eta^2\sqrt{\hat{\lambda}_i}, \quad 1 \leqslant i \leqslant N-1. \tag{84}$$

Note that the two equations (82a) and (82b) are apparently de-coupled. One can first solve (82a) for ψ_i^{n+1}, and then solve (82b) for ξ_i^{n+1}.

With the above formulations, in order to compute ϕ_i^{n+1} from the coupled system (63a), we only need to solve $2(N-1)$ *de-coupled individual* Helmholtz-type equations given by (82a) and (82b).

Let us now consider the boundary conditions (63b) and (63c). They can be written in matrix form,

$$\mathbf{n}\cdot\nabla\left(\nabla^2\boldsymbol{\Phi}\right)\big|_{\partial\Omega} = \left(\mathbf{T}\mathbf{D}^{-1}\mathbf{P}\right)\mathbf{n}\cdot\nabla\left(\nabla^2\mathbf{X}\right)\big|_{\partial\Omega} = 0, \tag{85}$$

$$\mathbf{n}\cdot\nabla\boldsymbol{\Phi}|_{\partial\Omega} = \left(\mathbf{T}\mathbf{D}^{-1}\mathbf{P}\right)\mathbf{n}\cdot\nabla\mathbf{X}|_{\partial\Omega} = 0, \tag{86}$$

where we have used the relations in (72), (75) and (79). It follows from the above equations that

$$\mathbf{n}\cdot\nabla\left(\nabla^2\xi_i^{n+1}\right)\big|_{\partial\Omega} = 0, \quad 1 \leqslant i \leqslant N-1, \tag{87}$$

$$\mathbf{n}\cdot\nabla\xi_i^{n+1}\big|_{\partial\Omega} = 0, \quad 1 \leqslant i \leqslant N-1. \tag{88}$$

By using Eqs. (82b) and (88), we can transform (87) into

$$\mathbf{n}\cdot\nabla\psi_i^{n+1}\big|_{\partial\Omega} = 0, \quad 1 \leqslant i \leqslant N-1. \tag{89}$$

In order to facilitate the implementation with C^0 spectral elements (or finite elements), we next derive the weak forms for Eqs. (82a) and (82b), incorporating the boundary conditions (88) and (89). Let $\varpi \in H^1(\Omega)$ denote the test function. Taking the L^2 inner product between Eq. (82a) and ϖ and integrating by part, we get the weak form about ψ_i^{n+1},

$$\int_\Omega \nabla\psi_i^{n+1}\cdot\nabla\varpi + \frac{1}{\hat{\lambda}_i}\left(\alpha_i + \frac{\hat{s}_i}{\eta^2}\right)\int_\Omega \psi_i^{n+1}\varpi = -\frac{1}{\hat{\lambda}_i}\int_\Omega q_i^{(1)}\varpi + \frac{1}{\hat{\lambda}_i}\int_\Omega \nabla q_i^{(2)}\cdot\nabla\varpi, \quad \forall\varpi \in H^1(\Omega), \tag{90}$$

where we have used the boundary conditions (63c) and (89). Taking the L^2 inner product between Eq. (82b) and ϖ and integrating by part, we get the weak form about ξ_i^{n+1},

$$\int_\Omega \nabla\xi_i^{n+1}\cdot\nabla\varpi - \frac{\alpha_i}{\hat{\lambda}_i}\int_\Omega \xi_i^{n+1}\varpi = -\int_\Omega \psi_i^{n+1}\varpi, \quad \forall\varpi \in H^1(\Omega), \tag{91}$$

where we have used the boundary condition (88). The two weak forms, (90) and (91), can be discretized with C^0 spectral elements or finite elements in a straightforward fashion.

Overall, employing general order parameters for N-phase formulations involves several operations during pre-processing:

1. Choose a specific set of order parameters, by specifying the matrix $\mathbf{A}_1$ and the vector $\mathbf{b}_1$ defined in (12). Compute $\mathbf{A}_2$ from Eq. (15). Compute $(\mathbf{A}_1^{-1})^T$ in Eq. (21).
2. Compute λ_{ij} $(1 \leqslant i, j \leqslant N-1)$ from Eqs. (57), (49) and (46) based on the pairwise surface tensions σ_{kl} $(1 \leqslant k < l \leqslant N)$ among the N fluids. Form matrix $\mathbf{A}$ according to (11).
3. Solve the eigenvalue problem about matrix $\mathbf{A}_2$. Form matrices $\mathbf{E}$ and $\mathbf{T}$ in (70). Compute matrix $\mathbf{D}$ in (73).
4. Compute matrix $\mathbf{B}_2$ in (75). Solve the eigenvalue problem about $\mathbf{B}_2$. Form matrices $\mathbf{K}$ and $\mathbf{P}$ in (76).
5. Choose $(N-1)$ constants $\hat{s}_i$ $(1 \leqslant i \leqslant N-1)$ that satisfy the conditions (84). Form the diagonal matrix $\hat{\mathbf{S}}$ in (77). Compute matrix $\mathbf{S}$ based on

$$\mathbf{S} = [S_{ij}]_{(N-1)\times(N-1)} = (\mathbf{TDP})\hat{\mathbf{S}}(\mathbf{TDP})^T. \tag{92}$$

During each time step, given (ϕ_i^n, $\mathbf{u}^n$), we compute (ϕ_i^{n+1}, $\nabla^2\phi_i^{n+1}$) with the following procedure. We refer to this procedure as **Phase-Marching** hereafter in this paper. It is comprised of several steps:

Phase-Marching procedure:

1. Compute $Q_i^{(1)}$ and $Q_i^{(2)}$ ($1 \leqslant i \leqslant N-1$) based on (67). Form vectors $\mathbf{Q}^{(1)}$ and $\mathbf{Q}^{(2)}$ in (68).
2. Compute vectors $\mathbf{P}^T\mathbf{D}^{-1}\mathbf{T}^T\mathbf{Q}^{(1)}$ and $\mathbf{P}^T\mathbf{D}^{-1}\mathbf{T}^T\mathbf{Q}^{(2)}$ in (80). Then $q_i^{(1)}$ and $q_i^{(2)}$ are known.
3. Solve Eqs. (90) for ψ_i^{n+1} ($1 \leqslant i \leqslant N-1$).
4. Solve Eqs. (91) for ξ_i^{n+1} ($1 \leqslant i \leqslant N-1$). Form vector $\mathbf{X}$ in (80).
5. Compute $\boldsymbol{\Phi}$ based on

$$\boldsymbol{\Phi} = \mathbf{T}\mathbf{D}^{-1}\mathbf{P}\mathbf{X}. \tag{93}$$

   This provides ϕ_i^{n+1} ($1 \leqslant i \leqslant N-1$).
6. Compute $\nabla^2\phi_i^{n+1}$ ($1 \leqslant i \leqslant N-1$) based on

$$\begin{bmatrix} \vdots \\ \nabla^2\phi_i^{n+1} \\ \vdots \end{bmatrix}_{(N-1)\times 1} = \nabla^2\boldsymbol{\Phi} = \mathbf{T}\mathbf{D}^{-1}\mathbf{P}\nabla^2\mathbf{X} = \mathbf{T}\mathbf{D}^{-1}\mathbf{P}\begin{bmatrix} \vdots \\ \psi_i^{n+1} - \frac{\alpha_i}{\hat{\lambda}_i}\xi_i^{n+1} \\ \vdots \end{bmatrix}_{(N-1)\times 1}. \tag{94}$$

7. Compute

$$\tilde{\mathbf{J}}^{n+1} = \tilde{\mathbf{J}}(\vec{\phi}^{n+1}, \nabla\vec{\phi}^{n+1}) \tag{95}$$

   based on Eq. (20), where $\nabla^2\phi_i^{n+1}$ are obtained from the previous step.
8. Compute

$$\rho^{n+1} = \rho(\vec{\phi}^{n+1}), \qquad \mu^{n+1} = \mu(\vec{\phi}^{n+1}), \tag{96}$$

   based on Eqs. (18) and (19). When the maximum density ratio among the N fluids is large (typically beyond about 10^2), we further clamp the values of ρ^{n+1} and μ^{n+1} as follows,

$$\rho^{n+1} = \begin{cases} \rho^{n+1}, & \text{if } \rho^{n+1} \in [\tilde{\rho}_{\min}, \tilde{\rho}_{\max}] \\ \tilde{\rho}_{\max}, & \text{if } \rho^{n+1} > \tilde{\rho}_{\max} \\ \tilde{\rho}_{\min}, & \text{if } \rho^{n+1} < \tilde{\rho}_{\min}, \end{cases} \qquad \mu^{n+1} = \begin{cases} \mu^{n+1}, & \text{if } \mu^{n+1} \in [\tilde{\mu}_{\min}, \tilde{\mu}_{\max}] \\ \tilde{\mu}_{\max}, & \text{if } \mu^{n+1} > \tilde{\mu}_{\max} \\ \tilde{\mu}_{\min}, & \text{if } \mu^{n+1} < \tilde{\mu}_{\min}, \end{cases} \tag{97}$$

   where $\tilde{\rho}_{\max} = \max\{\tilde{\rho}_i\}_{1\leqslant i\leqslant N}$, $\tilde{\rho}_{\min} = \min\{\tilde{\rho}_i\}_{1\leqslant i\leqslant N}$, $\tilde{\mu}_{\max} = \max\{\tilde{\mu}_i\}_{1\leqslant i\leqslant N}$, and $\tilde{\mu}_{\min} = \min\{\tilde{\mu}_i\}_{1\leqslant i\leqslant N}$.

The need for clamping the ρ^{n+1} and μ^{n+1} values in (97) when the maximum density ratio among the N fluids becomes large has been pointed out in [10]; see also the same situation in two-phase flows for large density ratios [14,9]. Practical simulations have shown that the numerical values for the phase field variables ϕ_i may slightly go out of range at certain spatial points. When the maximum density ratio among the N fluids is large, this may produce un-physical (negative) values for ρ^{n+1} and μ^{n+1} computed from (96) at certain points in the domain, and cause numerical difficulties. Therefore, when the maximum density ratio among the N fluids becomes large, we employ the operations in Eq. (97) to avoid this issue.

Let us briefly contrast the above algorithm for general order parameters with that of [10] for the set of special order parameters defined by (25). While the phase field equations (14c) for the general order parameters are considerably more complicated and more strongly coupled than those in [10], with the algorithm presented above these equations do not pose essential computational difficulties. The computational complexity per time step of the above algorithm for general order parameters is comparable to that of [10] for the simplified phase field equations.

3.2. Overall method

Let us now discuss the overall method for simulating the coupled system of governing equations, (14a)–(14c), for general order parameters.

We present in Appendix B an algorithm for the momentum equations (14a)–(14b), together with the boundary condition (61). This is a velocity correction-type scheme. Its formulation differs from that of [10] in two aspects. First, in the pressure substep (Eq. (114a) of Appendix B) all the terms in the momentum equation are approximated at the time step $(n+1)$ with the current algorithm. By contrast, in [10] while the inertia term and the pressure term are approximated at step $(n+1)$, all the rest of the terms are approximated at the step n. Second, in the velocity substep the algorithm of [10] contains a number of correction terms, while in the current algorithm there is no correction term (see Eq. (115a) of Appendix B). It is observed from numerical tests with analytic solutions that, while the current algorithm and the scheme of [10] result in comparable velocity errors, the current algorithm produces significantly smaller pressure errors. It should be noted that the current algorithm for the momentum equations results in only constant and time-independent coefficient matrices for the pressure and velocity linear algebraic systems after discretization, despite the variable nature of the mixture density and dynamic viscosity. Because the essential ideas for treating the variable density/viscosity here stem from our previous work [14] for two-phase flows and are not new, we have presented the discussions of this algorithm for the N-phase momentum equations in an appendix rather than in the main text. We refer to Appendix B for detailed discussions of this algorithm.

Let us now consider the overall time-marching scheme. Note that in Appendix B we have reformulated the momentum equation (14a) into an equivalent form (113), and introduced an auxiliary pressure P (see Eq. (112)). The following scheme solves for the field variables $(\mathbf{u}, P, \phi_i)$ from the system of governing equations consisting of (113), (14b)–(14c), together with the boundary conditions (61)–(62b). Given $(\mathbf{u}^n,\ P^n,\ \phi_i^n)$, we solve for the phase field functions, the pressure, and the velocity at time step $(n+1)$ successively in a de-coupled fashion as follows:

(phase-step) Use Eqs. (63a)–(63c);
(pressure-step) Use Eqs. (114a)–(114c) in Appendix B;
(velocity-step) Use Eqs. (115a)–(115b) in Appendix B.

It can be noted that in this scheme the phase field variables ϕ_i $(1 \leqslant i \leqslant N-1)$ are de-coupled from $(\mathbf{u}, P)$ due to the explicit treatment of the convection terms in (63a). The pressure P is de-coupled from the velocity $\mathbf{u}$ due to the velocity correction-type strategy embodied by (114a) and (115a).

We employ C^0 spectral elements [20,40,48] for spatial discretizations in the current paper. To facilitate the implementation of the above scheme, we derive the weak forms for the phase field functions, the pressure, and the velocity. These have been discussed in Section 3.1 and in Appendix B.

The final solution procedure is therefore the following. Within a time step, given $(\mathbf{u}^n,\ P^n,\ \phi_i^n)$, we compute ϕ_i^{n+1}, P^{n+1} and $\mathbf{u}^{n+1}$ successively in a de-coupled fashion:

1. Use the **Phase-Marching** procedure from Section 3.1 to compute ϕ_i^{n+1} and $\nabla^2\phi_i^{n+1}$ $(1 \leqslant i \leqslant N-1)$, $\tilde{\mathbf{J}}^{n+1}$, ρ^{n+1} and μ^{n+1};
2. Solve Eq. (119) in Appendix B for P^{n+1};
3. Solve Eq. (121) in Appendix B, together with the Dirichlet condition (115b), for $\mathbf{u}^{n+1}$.

Eqs. (90), (91), (119) and (121) are the actual equations to solve in the spectral element implementation. These equations are in weak forms, and involve no derivatives of order two or higher. All terms in these equations can be computed or evaluated directly in the discrete space using C^0 elements. The discretization of these equations with spectral elements involves only the standard procedure [20].

One can observe that our method has the following characteristics:

- It can employ arbitrary order parameters of the form (13) for N-phase flows.
- The mixing energy density coefficients λ_{ij} $(1 \leqslant i, j \leqslant N-1)$ are explicitly given in terms of the pairwise surface tensions among the N fluids.
- The computations for different flow variables are de-coupled. The computations for the $(N-1)$ phase field variables are also completely de-coupled. The computations for the different velocity components (see (121) in Appendix B) are de-coupled as well.
- The linear algebraic systems for all flow variables involve only *constant* and *time-independent* coefficient matrices after discretization. These coefficient matrices can be pre-computed during pre-processing.
- Within each time step, the method only requires the solution of individual Helmholtz-type (including Poisson) equations.

4. Representative numerical tests

In this section we use several multiphase flow problems in two dimensions to demonstrate the accuracy and capabilities of the N-phase physical formulation and numerical algorithm presented in Section 3. These problems involve large density contrasts, large viscosity contrasts, and pair-wise surface tensions. We compare our simulations with the Langmuir–de Gennes theory of floating liquid lenses for a three-phase problem to show that our method produces physically accurate results. The majority of simulation results in this section are obtained using the volume fractions c_i $(1 \leqslant i \leqslant N-1)$ as the order parameters. For several cases, the results obtained using other order parameters (e.g. $c_i - c_N$, $1 \leqslant i \leqslant N-1$) are also presented.

Let us now briefly mention the normalizations of flow variables, governing equations, and the boundary/initial conditions. One can show that, when the flow variables and physical parameters are normalized in a proper fashion, the forms of the N-phase governing equations and the boundary/initial conditions will remain un-changed upon non-dimensionalization. Specifically, the normalization constants for various flow variables and parameters are summarized in Table 2, where L and U_0 are respectively the characteristic length and velocity scales, and $\tilde{\rho}_1$ is the density of the first fluid. For example,

Table 2
Normalization of flow variables and physical/numerical parameters.

Variables/parameters	Normalization constants	Variables/parameters	Normalization constants
$\mathbf{x}$, η	L	m_i	$L/(\tilde{\rho}_1 U_0)$
$\mathbf{u}$, $\mathbf{w}$	U_0	$\tilde{\gamma}_i$, Γ	$1/\tilde{\rho}_1$
t, Δt	L/U_0	σ_{ij}	$\tilde{\rho}_1 U_0^2 L$
$\mathbf{g}_r$ (gravity)	U_0^2/L	λ_{ij}, S_{ij}	$\tilde{\rho}_1 U_0^2 L^2$
p, P, $h(\vec{\phi})$, $W(\vec{\phi}, \nabla\vec{\phi})$	$\tilde{\rho}_1 U_0^2$	d_{ij}	$\tilde{\rho}_1 U_0/L$
ϕ_i, $\vec{\phi}$, c_i	1	$\mathbf{f}$	$\tilde{\rho}_1 U_0^2/L$
β	$\sqrt{\tilde{\rho}_1} U_0 L$	g_i	$\tilde{\rho}_1 U_0^2/L^2$
$\tilde{\rho}_i$, ρ_i, ρ, a_{ij}, b_i, $\varphi_i(\vec{\phi})$, ρ_0	$\tilde{\rho}_1$	$\tilde{\mathbf{J}}$	$\tilde{\rho}_1 U_0$
$\tilde{\mu}_i$, μ	$\tilde{\rho}_1 U_0 L$	ν_0	$U_0 L$

the non-dimensional pairwise surface tensions are given by $\frac{\sigma_{ij}}{\tilde{\rho}_1 U_0^2 L}$ (inverse of Weber numbers) based on this table. In the following discussions, all variables are non-dimensional unless otherwise specified, and have been normalized according to Table 2.

4.1. Convergence rates

The goal of this section is to demonstrate the spatial and temporal convergence rates of the algorithm developed in Section 3 by using a contrived analytic solution to the N-phase governing equations.

Consider the flow domain defined by $\Omega = \{(x, y) : 0 \leqslant x \leqslant 2,\ -1 \leqslant y \leqslant 1\}$, and a four-phase fluid mixture contained in Ω. We assume the following analytic expressions for the flow and phase field variables

$$\begin{cases} u = A_u \cos \pi y \sin \bar{a} x \sin \omega t \\ v = -\dfrac{A_u \bar{a}}{\pi} \sin \pi y \cos \bar{a} x \sin \omega t \\ P = A_u \sin \pi y \sin \bar{a} x \cos \omega t \\ \phi_1 = \dfrac{1}{6}(1 + A_1 \cos \bar{a}_1 x \cos \bar{b}_1 y \sin \omega_1 t) \\ \phi_2 = \dfrac{1}{6}(1 + A_2 \cos \bar{a}_2 x \cos \bar{b}_2 y \sin \omega_2 t) \\ \phi_3 = \dfrac{1}{6}(1 + A_3 \cos \bar{a}_3 x \cos \bar{b}_3 y \sin \omega_3 t) \end{cases} \tag{98}$$

where (u, v) are the velocity components of $\mathbf{u}$, and A_u, $\bar{a}$, ω, A_i, $\bar{a}_i$, $\bar{b}_i$, ω_i $(1 \leqslant i \leqslant 3)$ are prescribed constants to be given below. It is straightforward to verify that (u, v) in (98) satisfy Eq. (14b), and that the expressions for ϕ_1, ϕ_2 and ϕ_3 satisfy the boundary conditions (62a) and (62b) with the parameter values given below. We choose the body force $\mathbf{f}(\mathbf{x}, t)$ in (113) (in Appendix B) and the source terms $g_i(\mathbf{x}, t)$ $(1 \leqslant i \leqslant N - 1)$ in (14c) such that the analytic expressions in (98) satisfy the governing equations (113) and (14c). In addition, we choose the boundary velocity $\mathbf{w}(\mathbf{x}, t)$ in (61) according to the velocity analytic expressions in (98), and choose the initial velocity and the initial phase field functions by setting $t = 0$ to the analytic expressions of (98).

We partition the domain along the x direction into two quadrilateral elements of equal size. The element order is varied systematically in the tests. We employ the algorithm developed in Section 3 to integrate the governing equations in time from $t = 0$ to $t = t_f$ (t_f to be specified later), and then compute and monitor the errors of the numerical solution at $t = t_f$ against the analytic solution from (98). The parameter values for this problem are listed in Table 3.

We have tested two sets of order parameters. The first set employs the volume fractions c_i $(1 \leqslant i \leqslant N - 1)$ as the order parameters, which is defined in Eq. (27). The other set employs the re-scaled density differences $\rho_i - \rho_{i+1}$ $(1 \leqslant i \leqslant N - 1)$ as the order parameters; see the definition in (33).

In the first group of tests we fix the time step size at $\Delta t = 0.001$ and final integration time at $t_f = 0.1$, and vary the element order systematically between 2 and 20. Fig. 1(a) shows the errors of the numerical solution in L^∞ and L^2 norms for the velocity, pressure and the phase-field variables (ϕ_1, ϕ_2, ϕ_3) at $t = t_f$ as a function of the element order, obtained with the volume fractions as the order parameters (see Eq. (27)). It is evident that the numerical errors decrease exponentially as the element order increases and is below about 12. As the element order increases beyond 12, the error curves level off owing to the saturation with the temporal truncation errors. Fig. 1(c) shows the corresponding error results obtained using the re-scaled density differences as the order parameters (see (33)). One can similarly observe an exponential spatial convergence rate.

In the second group of convergence tests we fix the element order at a large value 18 and the final integration time at $t_f = 1.0$. Then we vary the time step size systematically between $\Delta t = 0.0015625$ and $\Delta t = 0.1$. In Fig. 1(b) we plot the

Table 3
Parameter values for the convergence-rate tests.

Parameters	Values
A_u	2.0
A_1, A_2, A_3	1.0
$\bar{a}$, $\bar{a}_1$, $\bar{a}_2$, $\bar{a}_3$	π
$\bar{b}_1$, $\bar{b}_2$, $\bar{b}_3$	π
ω, ω_1	1.0
ω_2	1.2
ω_3	0.8
η	0.1
β	0.05
$\tilde{\rho}_1$	1.0
$\tilde{\rho}_2$	3.0
σ_{12}	6.236×10^{-3}
σ_{13}	7.265×10^{-3}
σ_{14}	3.727×10^{-3}
J (integration order)	2
$\tilde{\rho}_3$	2.0
$\tilde{\rho}_4$	4.0
$\tilde{\mu}_1$	0.01
$\tilde{\mu}_2$	0.02
$\tilde{\mu}_3$	0.03
$\tilde{\mu}_4$	0.04
m_1	10^{-3}
m_2	2×10^{-3}
m_3	3×10^{-3}
ρ_0	$\min(\tilde{\rho}_1, \dots, \tilde{\rho}_4)$
ν_0	$\max(\frac{\tilde{\mu}_1}{\tilde{\rho}_1}, \dots, \frac{\tilde{\mu}_4}{\tilde{\rho}_4})$
σ_{23}	8.165×10^{-3}
σ_{24}	5.270×10^{-3}
σ_{34}	6.455×10^{-3}
λ_{ij}	Computed from (58)

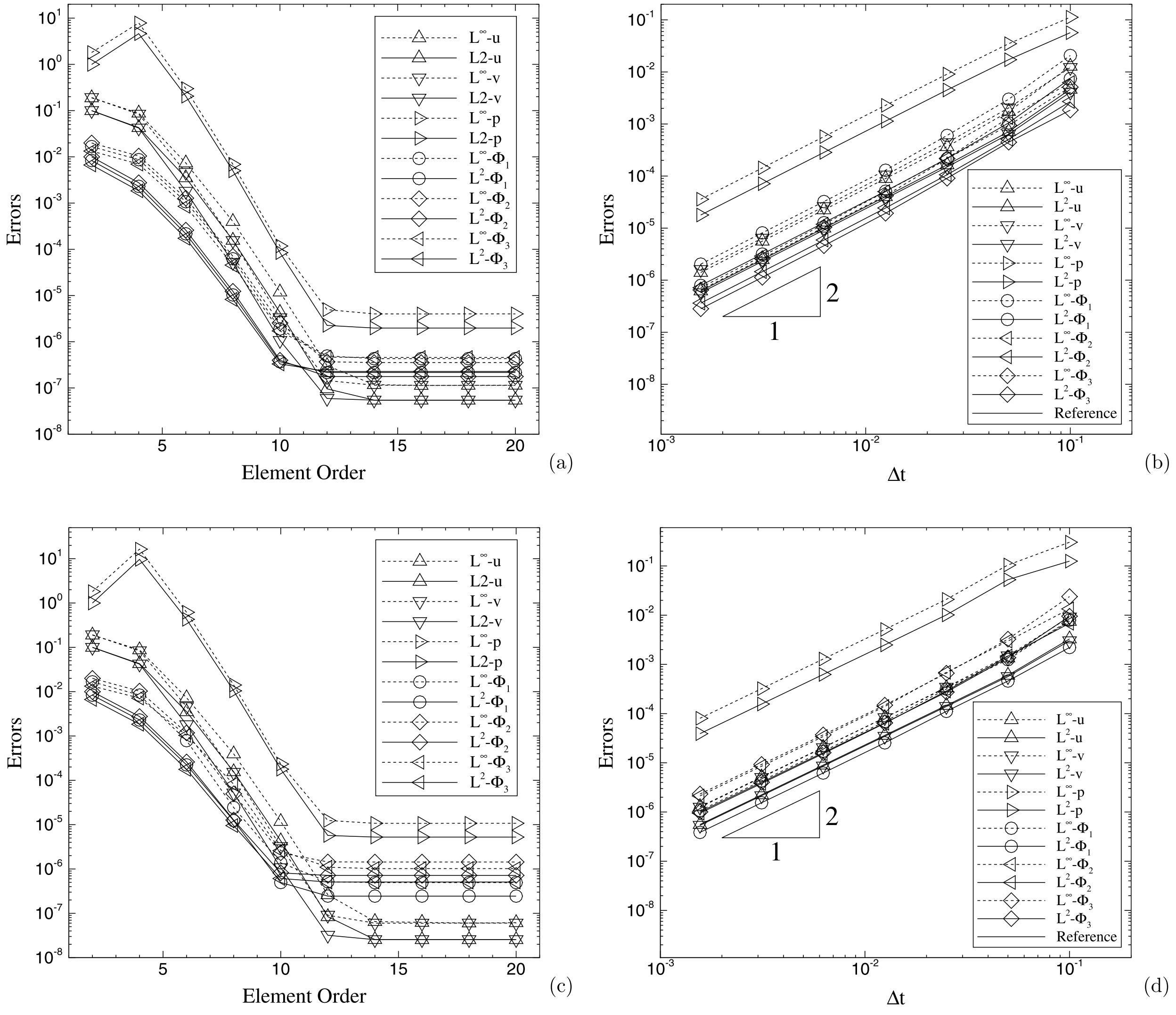

Fig. 1. Convergence rates: (a) and (c), numerical errors as a function of element order (fixed $\Delta t = 0.001$) showing spatial exponential convergence and error saturation at large element orders due to temporal truncation error. (b) and (d), numerical errors as a function of Δt (fixed element order 18) showing temporal second-order convergence rate. Results in (a) and (b) are obtained using formulation with volume fractions c_i ($1 \leqslant i \leqslant N-1$) as order parameters. Those in (c) and (d) are obtained using formulation with re-scaled density differences, $\rho_i - \rho_{i+1} = -\tilde{\rho}_{i+1} + (\tilde{\rho}_i + \tilde{\rho}_{i+1})\phi_i$ ($1 \leqslant i \leqslant N-1$), as order parameters.

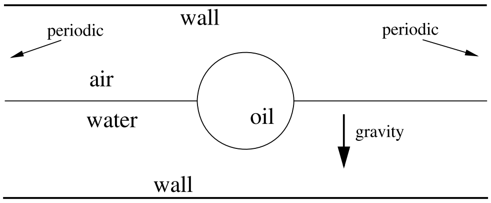

Fig. 2. Initial configuration for the three-phase problem of floating oil lens on water surface.

Table 4
Physical parameter values for the air–water–oil three phase problem.

Density [kg/m^3]:	Air	1.2041	Water	998.207	Oil	577 (or varied)
Dynamic viscosity [kg/(m s)]:	Air	1.78E−5	Water	1.002E−3	Oil	9.15E−2
Surface tension [kg/s^2]:	Air/water	0.0728	Oil/water	0.04	Air/oil	0.065 (or varied)
Gravity [m/s^2]:	9.8					

Table 5
Simulation parameter values for the air–water–oil three phase problem.

Parameters	Values
λ_{ij}	Computed based on (58)
η/L	0.0075
β	Computed based on (60)
$m_i\tilde{\rho}_1 U_0/L$, $1 \leqslant i \leqslant N-1$	$0.24(\frac{\eta}{L})^3 \frac{\tilde{\rho}_1 U_0^2 L^2}{\lambda_{\max}}$, where $\lambda_{\max} = \max\{\lambda_{ij}\}$
$\hat{s}_i$	$2\eta^2\sqrt{\hat{\lambda}_i}$
ρ_0	$\min\{\tilde{\rho}_i\}_{1\leqslant i\leqslant N}$
ν_0	$2\max\{\frac{\tilde{\mu}_i}{\tilde{\rho}_i}\}_{1\leqslant i\leqslant N}$
$U_0\Delta t/L$	2.5×10^{-6}
J (temporal order)	2

L^∞ and L^2 errors of the numerical solution at $t = t_f$ against the analytic solution for different variables as a function of Δt, obtained with the volume fractions as the order parameters. The error curves exhibit a second-order temporal convergence rate. Fig. 1(d) contains the corresponding results obtained with the re-scaled density differences as the order parameters, showing similarly a second-order temporal convergence rate.

4.2. Floating oil lens on water surface

In this section we look into an air–water–oil three-phase problem, where the oil forms a liquid lens floating on the water surface at equilibrium. We quantitatively compare the simulation results with the theory by Langmuir and de Gennes [25,8] to show that our method produces physically accurate results. A similar liquid-lens problem was considered in [10] using the special set of order parameters defined by (25). Here we simulate the problem with general order parameters, and investigate the effects of several sets of physical parameters on the air–water–oil configurations.

Consider the flow domain depicted in Fig. 2, $-\frac{L}{2} \leqslant x \leqslant \frac{L}{2}$ and $0 \leqslant y \leqslant \frac{4}{5}L$, where $L = 4$ cm. The top and the bottom of the domain ($y = 0$ and $\frac{4}{5}L$) are two solid walls. In the horizontal direction the domain is periodic at $x = \frac{L}{2}$ and $x = -\frac{L}{2}$. At $t = 0$, the domain is filled with air in its top half and filled with water in its bottom half, and a circular oil drop of radius $R_0 = \frac{L}{5}$ is held at rest on the water surface with its center located at $\mathbf{x}_c = (x_c, y_c) = (0, \frac{2}{5}L)$. It is assumed that the gravity is in the $-y$ direction, and that there is no initial flow. The system is then released, and evolves to equilibrium due to the interactions among the three fluid components and the effects of the gravity and the surface tensions. Our goal is investigate the effects of several physical parameters on the equilibrium configurations of the oil.

Table 4 lists the values of the physical parameters, including the densities, dynamic viscosities, and the pairwise surface tensions of the three fluids involved in this problem. We use L as the characteristic length scale, and choose a characteristic velocity scale $U_0 = \sqrt{g_{r0}L}$, where $g_{r0} = 1$ m/s^2. We assign the air, water and oil as the first, second and third fluid, respectively. All the flow variables and physical parameters are then non-dimensionalized based on the normalization constants specified in Table 2. We set $g_i = 0$ $(1 \leqslant i \leqslant N-1)$ in (14c) for the simulations.

In order to simulate the problem, we discretize the domain using 160 equal-sized quadrilateral elements, with 20 elements along the x direction and 8 along the y direction. The element order is 16 in the simulations. The algorithm developed in Section 3 has been used to solve the coupled system of governing equations. For the boundary conditions, at the top/bottom walls, we impose the condition (61) with $\mathbf{w} = 0$ for the velocity and the conditions (62a) and (62b) for the phase field functions. In the horizontal direction, all the flow variables (velocity, pressure, phase field functions) are set to be periodic.

We employ the formulation with the volume fractions as the order parameters, defined in (27), for simulations of all the cases in this section. We have also simulated several selected cases using the formulation with the re-scaled volume fraction differences as the order parameters, as defined in (29). The initial velocity is set to zero. The initial phase field functions are set to

$$\phi_i(\mathbf{x}, t=0) = c_{i0}, \quad 1 \leqslant i \leqslant N-1, \tag{99}$$

for the order parameters defined by (27), and

$$\phi_i(\mathbf{x}, t=0) = \frac{1}{2} + \frac{1}{2}(c_{i0} - c_{N0}), \quad 1 \leqslant i \leqslant N-1, \tag{100}$$

for the order parameters defined by (29). In these equations, c_{i0} $(1 \leqslant i \leqslant N)$ are the initial volume fractions given by $(N=3)$

$$\begin{aligned}
c_{10} &= \big[1 - \Theta(x, x_c - R_0) + \Theta(x, x_c + R_0)\big]\frac{1}{2}\left(1 + \tanh\frac{y - y_c}{\sqrt{2}\eta}\right) \\
&\quad + \big[\Theta(x, x_c - R_0) - \Theta(x, x_c + R_0)\big]\frac{1}{2}\left(1 + \tanh\frac{|\mathbf{x} - \mathbf{x}_c| - R_0}{\sqrt{2}\eta}\right)\Theta(y, y_c), \\
c_{20} &= \big[1 - \Theta(x, x_c - R_0) + \Theta(x, x_c + R_0)\big]\frac{1}{2}\left(1 - \tanh\frac{y - y_c}{\sqrt{2}\eta}\right) \\
&\quad + \big[\Theta(x, x_c - R_0) - \Theta(x, x_c + R_0)\big]\frac{1}{2}\left(1 + \tanh\frac{|\mathbf{x} - \mathbf{x}_c| - R_0}{\sqrt{2}\eta}\right)\big[1 - \Theta(y, y_c)\big], \\
c_{30} &= \frac{1}{2}\left(1 - \tanh\frac{|\mathbf{x} - \mathbf{x}_c| - R_0}{\sqrt{2}\eta}\right),
\end{aligned}$$

where $\Theta(x, a)$ is the unit step function, assuming unit value if $x \geqslant a$ and zero otherwise.

Table 5 summarizes the numerical parameter values in the simulations. We have used $\rho_0 = \min\{\tilde{\rho}_i\}_{1 \leqslant i \leqslant N}$ and $\nu_0 = 2\max\{\frac{\tilde{\mu}_i}{\tilde{\rho}_i}\}_{1 \leqslant i \leqslant N}$, which respectively satisfy the conditions (116) and (117). When solving the phase field equations we use $\hat{s}_i = 2\eta^2\sqrt{\hat{\lambda}_i}$, which satisfies the condition (84). We employ the same value for the mobility coefficients m_i $(1 \leqslant i \leqslant N-1)$, and choose this value according to the theoretical analysis of [33]. Magaletti et al. [33] have provided a detailed asymptotic analysis of the phase-field governing equations for two-phase flows (coupled Navier–Stokes and Cahn–Hilliard equations) at the sharp interface limit. They have shown that, as the interfacial thickness η approaches zero, in order for the diffuse-interface model to recover the sharp interface model and provide the correct surface tension at the sharp interface limit, the non-dimensional mobility number M^* must scale as the square of the Cahn number ($C_n = \frac{\eta}{L}$), i.e.

$$M^* = \frac{3}{2\sqrt{2}}\frac{\sigma}{\tilde{\rho}_1 U_0^2 L}\frac{M\tilde{\rho}_1 U_0}{L} \propto \left(\frac{\eta}{L}\right)^2, \quad \text{or equivalently,} \quad \frac{\lambda}{\tilde{\rho}_1 U_0^2 L^2}\frac{M\tilde{\rho}_1 U_0}{L} \propto \left(\frac{\eta}{L}\right)^3, \tag{101}$$

where M, σ, and λ ($\lambda = \frac{3}{2\sqrt{2}}\sigma\eta$) are respectively the mobility coefficient, the surface tension, and the mixing energy density coefficient for two fluid phases. Based on the relation (101) we therefore choose the mobility coefficients m_i by

$$\frac{\lambda_{\max}}{\tilde{\rho}_1 U_0^2 L^2}\frac{m_i\tilde{\rho}_1 U_0}{L} = \alpha_s\left(\frac{\eta}{L}\right)^3, \quad 1 \leqslant i \leqslant N-1 \tag{102}$$

where $\lambda_{\max} = \max\{\lambda_{ij}\}_{1 \leqslant i,j \leqslant N-1}$, and α_s is a proportionality coefficient. Note that the specific value for α_s is not of consequence as $\eta \to 0$ [33]. We set $\alpha_s = 0.24$ for this problem. We have performed simulations using two interfacial thickness values $\frac{\eta}{L} = 0.01$ and 0.0075, and their results are comparable. Results reported below correspond to $\frac{\eta}{L} = 0.0075$. We have used a sufficiently small time step size $\frac{U_0 \Delta t}{L} = 2.5 \times 10^{-6}$ in the simulations.

The physics of floating liquid lenses for three phases was discussed in Langmuir [25] and de Gennes et al. [8] (pages 54–56). The equilibrium oil-drop shape is determined by the interplay of the gravity and the three pair-wise surface tensions, and also influenced by the three densities. When the gravity effect dominates, the oil will form a puddle on the water surface. If the surface tension effects dominate, the oil-drop shape will consist of two circular caps in two dimensions or two spherical caps in three dimensions. One can approximately determine which effect is dominant by comparing the characteristic drop size with the three capillary lengths associated with the three types of interfaces; see [8].

We will investigate the effects on the equilibrium oil configurations of two physical parameters: the oil density and the air–oil surface tension.

Let us first consider the effect of the oil density. In this group of tests, we vary the density of the oil systematically ranging from 300 kg/m^3 to 750 kg/m^3, and fix all the other physical parameters at values given in Table 4 (air–oil surface tension fixed at 0.065 kg/s^2). In Fig. 3 we show the equilibrium oil configurations corresponding to three oil-density values $\rho_o = 300$ kg/m^3, 500 kg/m^3 and 750 kg/m^3. They are obtained using the volume fractions as the order parameters. Plotted

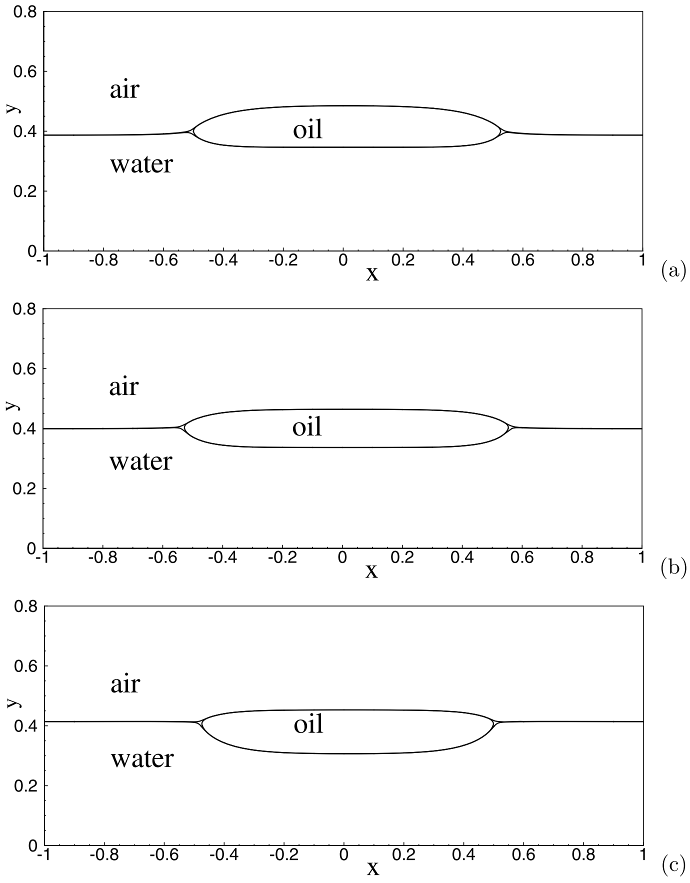

Fig. 3. Effect of oil density on equilibrium oil puddle configurations (3 fluid phases): (a) $\rho_o = 300$ kg/m^3, (b) $\rho_o = 500$ kg/m^3, (c) $\rho_o = 750$ kg/m^3. Results obtained using volume fractions c_i $(1 \leqslant i \leqslant N-1)$ as order parameters.

are the contours of volume fractions $c_i = \frac{1}{2}$ $(i = 1, 2, 3)$ for the three fluids. One can note the "star"-shaped regions around the three-phase contact lines in these figures. This is because in these regions none of the three fluids has a volume fraction larger than $\frac{1}{2}$. It is evident that the oil forms puddles floating on the water surface. Subtle differences can be noticed in their shapes, for example, in the curvature of the oil profiles not far from the three-phase contact lines. In addition, the immersion depths of the oil in the water are notably different as the oil density changes.

The simulation results obtained using different sets of order parameters are similar, as is expected. This is shown by Fig. 4. Here we compare the equilibrium oil configurations, as shown by the contour levels $c_i = \frac{1}{2}$, corresponding to the oil density $\rho_0 = 400$ kg/m^3 obtained using the volume fractions as the order parameters (Eq. (27)), see Fig. 4(a), and using the re-scaled volume fraction differences as the order parameters (Eq. (29)), see Fig. 4(b). The results are qualitatively similar.

We have computed the thickness of the oil puddles at equilibrium, defined as the distance along the vertical direction between the upper and lower puddle surfaces, corresponding to different oil densities. The symbols in Fig. 5 show the normalized oil-puddle thickness as a function of the normalized oil density obtained from the current simulations. The results from both sets of order parameters have been included in this figure, differentiated using different symbols. It is shown in [25,8] that for three fluid phases, when the gravity is dominant (i.e. oil forming puddles), the puddle thickness can be explicitly expressed in terms of the known physical parameters as follows,

$$e_c = \sqrt{\frac{2(\sigma_{ao} + \sigma_{ow} - \sigma_{ow})\rho_w}{(\rho_w - \rho_o)\rho_o g_r}}, \tag{103}$$

where e_c is the asymptotic puddle thickness when the gravity is dominant, g_r is the gravitational acceleration, ρ_w and ρ_o are respectively the water and oil densities, and σ_{ao}, σ_{aw} and σ_{ow} are respectively the air–oil, air–water, and oil–water surface tensions. For comparison, the dashed curve in Fig. 5 shows the relation between e_c and ρ_o given by (103). The results from the current simulations agree with the results based on the theory of [25,8] quite well.

We next look into the effect of the air–oil surface tension on the equilibrium configuration of this three-phase system. In this group of tests we vary the air–oil surface tension systematically between 0.055 kg/s^2 and 0.095 kg/s^2, while fixing all the other parameters at those values in Table 4 (oil density is 577 kg/m^3). Fig. 6 shows the equilibrium configurations of

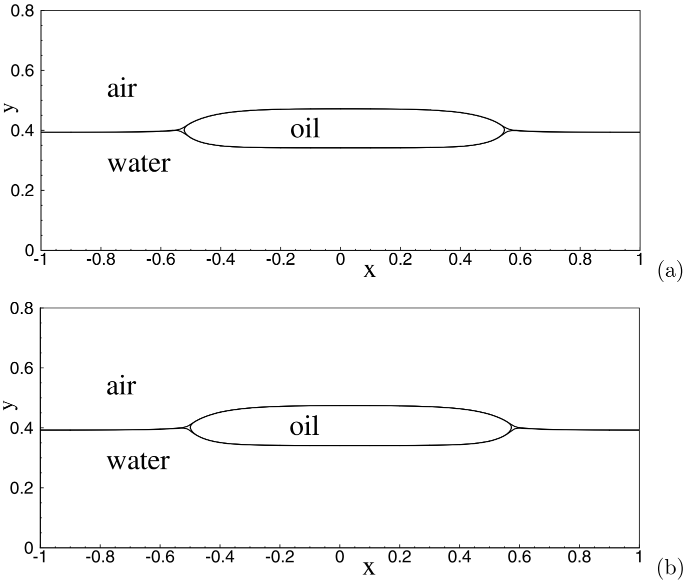

Fig. 4. Equilibrium oil-puddle configurations for oil density $\rho_o = 400$ kg/m^3 obtained using (a) volume fractions c_i ($1 \leqslant i \leqslant N-1$) as order parameters, and (b) re-scaled volume fraction differences as order parameters (Eq. (29)).

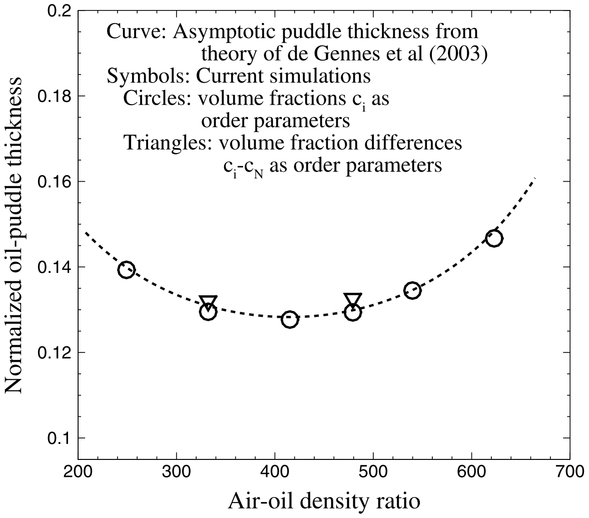

Fig. 5. Comparison of the oil-puddle thickness as a function of the oil density between current simulations and the Langmuir–de Gennes theory [8] for three fluid phases.

the system (contour lines $c_i = \frac{1}{2}$, $1 \leqslant i \leqslant 3$) corresponding to the air–oil surface tensions 0.065 kg/s^2 and 0.085 kg/s^2. They are obtained with the volume fractions as the order parameters. The oil forms a puddle, and the puddle size and thickness have a clear dependence on the value of the air–oil surface tension. A larger air–oil surface tension leads to a smaller but thicker oil puddle at equilibrium.

The quantitative relationship between the oil-puddle thickness and the air–oil surface tension is demonstrated by Fig. 7. Here we plot the puddle-thickness *squared* as a function of the air–oil surface tension. The symbols represent results from the simulations. The circles denote the results obtained using the volume fractions as the order parameters (Eq. (27)), and the triangles denote those obtained using the re-scaled volume-fraction differences (Eq. (29)) as the order parameters. For comparison, the theoretical relationship between these two quantities, see Eq. (103), due to [25,8] is also shown in this figure, by the dashed line. It can be observed that the data from current simulations are in good agreement with the theory.

To summarize, the floating liquid lens problem studied in this section involves multiple fluid phases, multiple pairwise surface tensions, gravity, large density ratios and viscosity ratios. The quantitative comparisons between current simulations and the theory of Langmuir and de Gennes [25,8] show that, the physical formulations and the numerical algorithm with general order parameters developed in the current work produce physically accurate results. The results of this section

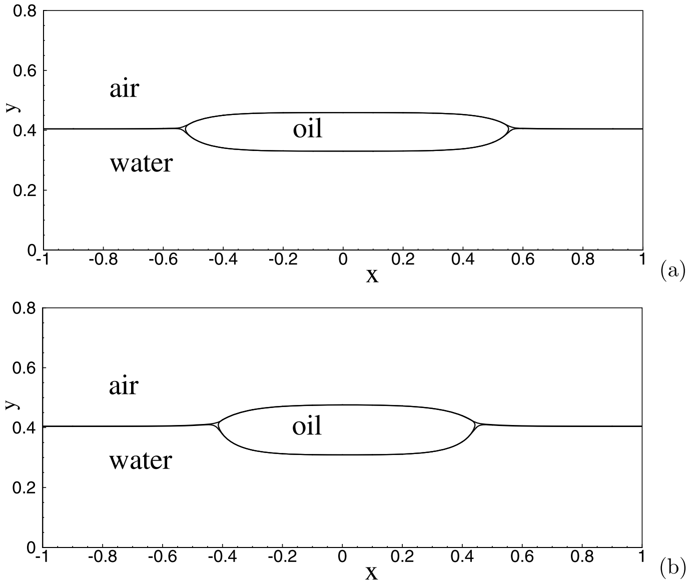

Fig. 6. Equilibrium configurations of an oil puddle on the air–water interface corresponding to air–oil surface tensions: (a) 0.065 kg/s^2, (b) 0.085 kg/s^2. All other physical parameters are fixed. Results are obtained with the volume fractions c_i $(1 \leqslant i \leqslant N-1)$ as the order parameters.

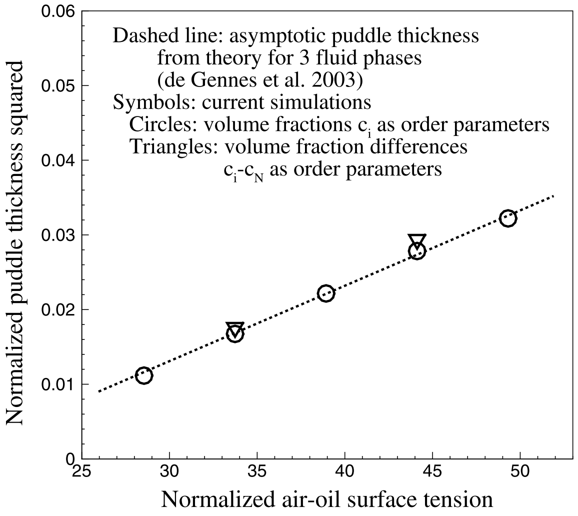

Fig. 7. Comparison between current simulations and the Langmuir–de Gennes theory [8] for air–water–oil three fluid phases: Oil-puddle thickness squared as a function of the air–oil surface tension. Circles denote results obtained with the volume fractions as order parameters. Triangles denote results obtained with the re-scaled volume-fraction differences as order parameters.

also demonstrate the significant effects of the densities and the pairwise surface tensions on the configurations of this three-phase system.

4.3. *Dynamics of a four-phase fluid mixture*

In this section we look into the dynamics of a mixture of four immiscible incompressible fluids. The goal is to demonstrate the capability of our method from Section 3 for simulating dynamical problems.

The setting of this problem is as follows. Consider the rectangular flow domain as shown in Fig. 8(a), $-\frac{L}{2} \leqslant x \leqslant \frac{L}{2}$ and $0 \leqslant y \leqslant \frac{8}{5}L$, where $L = 2$ cm. The top and bottom sides of the domain are solid walls. If any of the fluid interfaces involved in this problem intersects with the walls, the contact angle is assumed to be 90°. In the horizontal direction the domain is assumed to be periodic at $x = \pm\frac{L}{2}$. The gravity is assumed be along the $-y$ direction. At $t = 0$, the top half of the domain is filled with air, and the bottom half is filled with water. A circular drop (diameter $0.3L$) of a fluid, denoted by “F2”, is suspended in the air at rest. Simultaneously, an air bubble and a drop of another fluid, denoted by “F1”, both circular

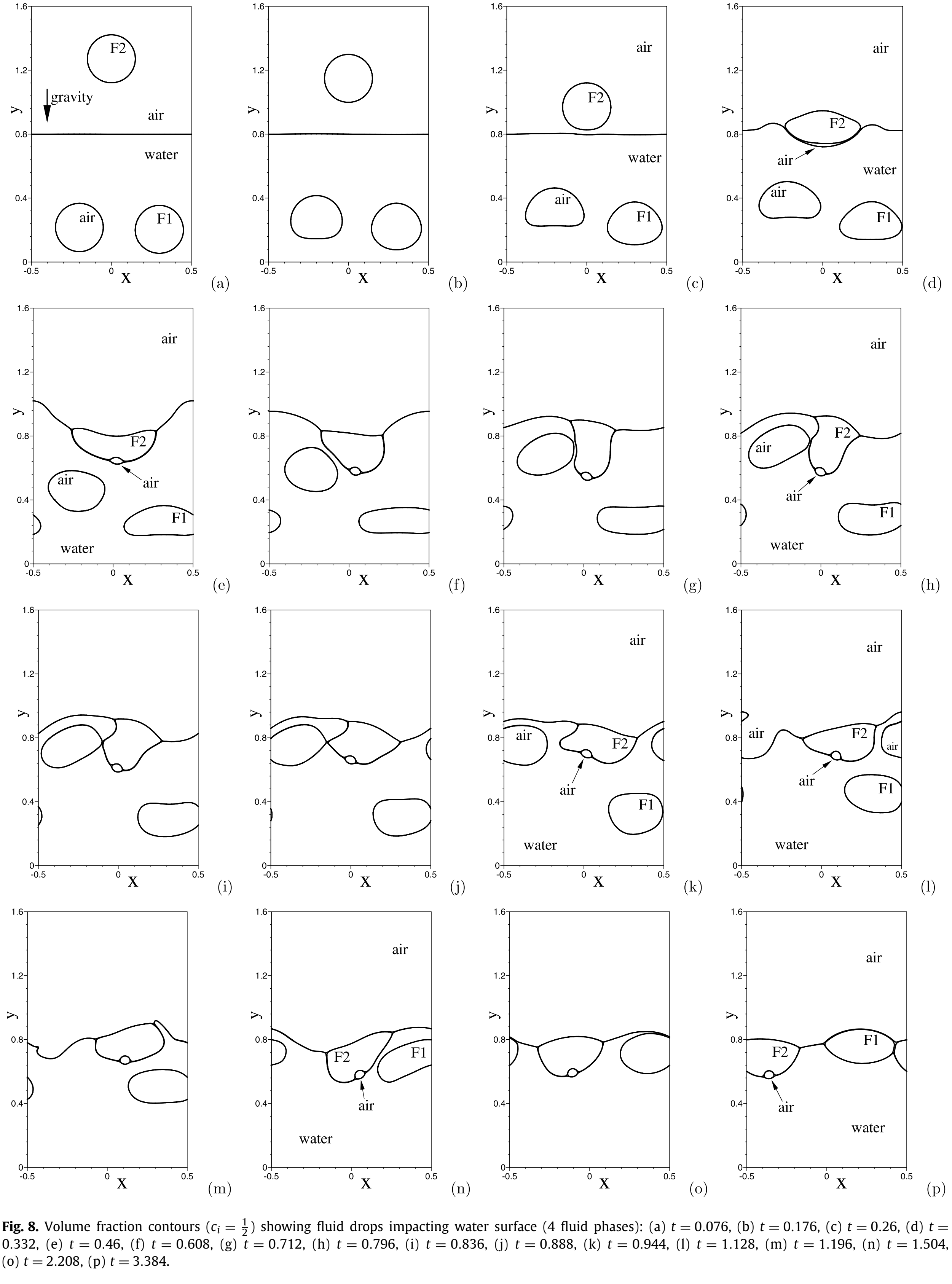

Fig. 8. Volume fraction contours ($c_i = \frac{1}{2}$) showing fluid drops impacting water surface (4 fluid phases): (a) $t = 0.076$, (b) $t = 0.176$, (c) $t = 0.26$, (d) $t = 0.332$, (e) $t = 0.46$, (f) $t = 0.608$, (g) $t = 0.712$, (h) $t = 0.796$, (i) $t = 0.836$, (j) $t = 0.888$, (k) $t = 0.944$, (l) $t = 1.128$, (m) $t = 1.196$, (n) $t = 1.504$, (o) $t = 2.208$, (p) $t = 3.384$.

Table 6
Physical parameter values for the air–water–F1–F2 four phase problem.

Density [kg/m^3]	Air	1.2041	Water	998.207	F1	400	F2	870
Dynamic viscosity [kg/(m s)]	Air	1.78E−5	Water	1.002E−3	F1	0.02	F2	0.0915
Surface tension [kg/s^2]	Air/water	0.0728	Air/F1	0.06	Air/F2	0.055		
	Water/F1	0.045	Water/F2	0.044	F1/F2	0.048		
Gravity [m/s^2]	9.8							

Table 7
Simulation parameter values for the air–water–F1–F2 four phase problem. $N = 4$ in this table.

Parameters	Values
λ_{ij}	Computed based on (58)
η/L	0.005
β	Computed based on (60)
$m_i\tilde{\rho}_1 U_0/L$, $1 \leqslant i \leqslant N-1$	$0.16(\frac{\eta}{L})^3 \frac{\tilde{\rho}_1 U_0^2 L^2}{\lambda_{\max}}$, where $\lambda_{\max} = \max\{\lambda_{ij}\}$
$\hat{s}_i$	$2\eta^2\sqrt{\hat{\lambda}_i}$
ρ_0	$\min\{\tilde{\rho}_i\}_{1\leqslant i\leqslant N}$
ν_0	$5\max\{\frac{\tilde{\mu}_i}{\tilde{\rho}_i}\}_{1\leqslant i\leqslant N}$
$U_0\Delta t/L$	2×10^{-6}
J (temporal order)	2

initially with diameter $0.3L$, are trapped in the water. The centers of the air bubble and the fluid drops have the following coordinates

$$\begin{cases} \mathbf{x}_{F1} = (x_{F1}, y_{F1}) = (0.3L, 0.2L) \quad \text{(F1 drop)} \\ \mathbf{x}_{F2} = (x_{F2}, y_{F2}) = (0, 1.3L) \quad \text{(F2 drop)} \\ \mathbf{x}_a = (x_a, y_a) = (-0.2L, 0.2L) \quad \text{(air bubble).} \end{cases} \tag{104}$$

The four types of fluids (air, water, F1, and F2) are assumed to be incompressible and all immiscible with one another, and it is assumed that there is no initial velocity. The system is then released. The liquid drops and the air bubble fall through the air or rise through the water, and then impact the water surface. The objective is to simulate the dynamics of this process.

In Table 6 we list the values of the physical parameters involved in this problem, including the densities and dynamic viscosities of the four fluids, and the six pairwise surface tensions among them. We assign the air, water, F2 and F1 fluids respectively as the first, second, third and fourth fluid. We choose L as the characteristic length scale and $U_0 = \sqrt{g_{r0}L}$ as the characteristic velocity scale, where $g_{r0} = 1$ m/s^2. The non-dimensionalization of the problem then follows in a straightforward fashion based on the constants given in Table 2. We set $g_i = 0$ $(1 \leqslant i \leqslant N-1)$ in (14c) in the simulations.

In order to simulate the problem, we discretize the domain using 1440 quadrilateral elements of equal sizes, with 30 elements in the x direction and 48 elements in the y direction. An element order of 12 (with over-integration) has been used in the simulations for all elements. The algorithm developed in Section 3, and the formulation with the volume fractions c_i $(1 \leqslant i \leqslant N-1$, where $N = 4)$ as the order parameters as defined by (27), have been employed to integrate this four-phase system in time. For the boundary conditions on the top and bottom walls, the condition (61) with $\mathbf{w} = 0$ has been imposed for the velocity, and the conditions (62a) and (62b) have been imposed for the phase field functions ϕ_i $(1 \leqslant i \leqslant 3)$. In the horizontal direction, periodic conditions have been imposed at $x = \pm\frac{L}{2}$ for all flow variables. The initial velocity is set to zero. The initial phase field functions are set according to Eq. (99) with $N = 4$, where the initial volume fractions are

$$c_{10} = \frac{1}{2}\left(1 + \tanh\frac{y - y_w}{\sqrt{2}\eta}\right)\left[1 - \frac{1}{2}\left(1 - \tanh\frac{|\mathbf{x} - \mathbf{x}_{F2}| - R_0}{\sqrt{2}\eta}\right)\right] + \frac{1}{2}\left(1 - \tanh\frac{|\mathbf{x} - \mathbf{x}_a| - R_0}{\sqrt{2}\eta}\right),$$

$$c_{20} = \frac{1}{2}\left(1 - \tanh\frac{y - y_w}{\sqrt{2}\eta}\right)\left[1 - \frac{1}{2}\left(1 - \tanh\frac{|\mathbf{x} - \mathbf{x}_a| - R_0}{\sqrt{2}\eta}\right) - \frac{1}{2}\left(1 - \tanh\frac{|\mathbf{x} - \mathbf{x}_{F1}| - R_0}{\sqrt{2}\eta}\right)\right],$$

$$c_{30} = \frac{1}{2}\left(1 - \tanh\frac{|\mathbf{x} - \mathbf{x}_{F2}| - R_0}{\sqrt{2}\eta}\right),$$

$$c_{40} = \frac{1}{2}\left(1 - \tanh\frac{|\mathbf{x} - \mathbf{x}_{F1}| - R_0}{\sqrt{2}\eta}\right),$$

where $y_w = \frac{4}{5}L$ is the initial position of the water surface in the y direction, and $R_0 = 0.15L$ is the initial radii of the air bubble and fluid drops.

Table 7 summarizes the values of all the numerical parameters involved in the algorithm and the simulations. The mobility coefficients m_i have been chosen according to Eq. (102), which stems from the asymptotic analysis of [33] as explained in Section 4.2, and we have used $\alpha_s = 0.16$ for this problem. We have conducted simulations using several values for the characteristic interfacial thickness $\frac{\eta}{L} = 0.01$, 0.0075 and 0.005, and observe that the flow features resulting from the two smallest values are essentially the same. The simulation results presented below correspond to $\frac{\eta}{L} = 0.005$.

We now look into the dynamics of this four-phase flow. In Fig. 8 we show a temporal sequence of snapshots of the fluid interfaces in the flow by plotting the contour lines of the volume fractions $c_i = \frac{1}{2}$ $(1 \leqslant i \leqslant 4)$. From Figs. 8(a)–(c), one can observe that upon release the F2 drop falls rapidly through the air due to the gravity, and is about to impact the water surface (Fig. 8(c)). Its profile maintains essentially the original shape during the falling process. Simultaneously, the air bubble and the F1 drop rise through the water due to buoyancy, albeit much more slowly compared to the falling F2 drop. They experience significant deformations in their shapes. The air bubble has the shape of a "cap", with a flat underside (Fig. 8(c)). Fig. 8(d) shows that the F2 drop impacts the water surface, and generates a ripple that spreads outward. The F2 drop traps a pocket of air at its underside. Later on, the F2 fluid forms a pool floating on the water surface and the trapped air pocket forms a small bubble (Fig. 8(e)). Figs. 8(f)–(k) show the interaction between the rising air bubble and the floating F2 pool on the water surface. Notice that the F2 fluid is mostly immersed in the water owing to the small density contrast between F2 and water (see e.g. Fig. 8(g)). As the air bubble rises, it approaches and "kisses" the F2 fluid immersed in the water (Figs. 8(g)–(j)), and then pulls apart (Fig. 8(k)). This interaction and the motion of the air bubble has caused a dramatic deformation in the profile of the F2 fluid (Figs. 8(i)–(k)). One also observes that, during this period of time, the upward motion of the F1 drop appears to have stalled (Figs. 8(f)–(j)), but the drop exhibits significant deformations in shape. Figs. 8(k)–(m) show that the air bubble touches the water surface and merges with the bulk of air above the water. As time goes on, the F1 drop rises slowly through the water and forms a floating F1 drop on the water surface (Figs. 8(n)–(p)). Eventually, the water surface becomes mostly covered by the floating F1 and F2 drops, and the F2 drop has a small air bubble trapped at its underside (Fig. 8(p)).

We further illustrate the dynamical features of this flow by looking into the velocity distributions. Fig. 9 is a temporal sequence of snapshots of the velocity fields of this four-phase flow at the same time instants as those in Fig. 8. In order to make the figures clearer, the velocity vectors have been plotted on every 15th quadrature point in each direction within each element. Figs. 9(a)–(c) indicate that the falling F2 drop induces a velocity field inside the air, forming a pair of vortices near the shoulders of the F2 drop (see e.g. Figs. 9(b)–(c)). On the other hand, the velocity inside the F2 drop is largely uniform. Prior to the impact on the water surface (Fig. 9(c)), the air is squeezed out from between the F2 drop and the water surface, resulting in a strong lateral air flow just above the water surface. Upon impact, the strong air flow produces a pair of vortices behind (i.e. above) either side of the F2 drop (Fig. 9(d)), noting the periodicity in the horizontal direction. The pair of vortices subsequently travel upward in the air and dies down gradually over time (Figs. 9(e)–(k)). Simultaneously, the rise of the air bubble through the water induces a pair of vortices behind (see e.g. Fig. 9(d)). A pair of vortices can also be recognized behind the F1 drop; see e.g. Fig. 9(f). An interaction of these two pairs of vortices in the water can be observed (Figs. 9(f)–(k)). Subsequently, the merger of the air bubble and the bulk of air generates an energetic air flow near the water surface (Figs. 9(l)–(m)). Figs. 9(n)–(p) show that the velocity field inside the water, and also in the air, dies down as the F1 drop rises to the water surface and the system approaches an equilibrium state.

5. Concluding remarks

The main contributions of the current work consist of the following three aspects:

- We have presented a family of N-phase physical formulations corresponding to a class of general order parameters. They generalize the N-phase formulation presented in [10]. This generalization has three implications: (i) The set of $(N-1)$ phase field equations becomes extremely strongly-coupled with one another. In particular, the inertia terms $\frac{\partial \phi_i}{\partial t}$ are also all coupled with one another. (ii) It makes it possible to derive an *explicit* form for the mixing energy density coefficients λ_{ij}. (iii) Numerical solution of the coupled phase field equations becomes more challenging.
- We have provided an *explicit* form for computing the mixing energy density coefficients λ_{ij} with general order parameters. This also provides an explicit solution to the linear algebraic system about λ_{ij} derived in [10] corresponding to a special set of order parameters.
- We have developed an efficient algorithm for solving the coupled system of phase field equations with general order parameters. The algorithm transforms the $(N-1)$ strongly-coupled fourth order phase field equations for general order parameters into $2(N-1)$ Helmholtz type equations that are completely de-coupled from one another. The computational complexity of the current algorithm for general order parameters is comparable to that of [10] for the special order parameters. The current work shows that, although the phase field equations for general order parameters have a considerably more complicated form, the computational work involved in their numerical solution is essentially the same as that for the simplified phase field equations corresponding to the special order parameters of [10].

In addition, in Appendix A we have provided a proof that the linear algebraic system about λ_{ij} derived in [10] corresponding to the special order parameters is uniquely solvable. The unique solvability of that system is an un-settled issue in [10].

We have presented several example problems to demonstrate the accuracy and capability of the physical formulations and the numerical algorithm developed herein. These test problems involve multiple fluid phases, large density contrasts, large viscosity contrasts, and multiple pairwise surface tensions. Several different sets of order parameters have been employed in the simulations. By comparing with the theory of Langmuir and de Gennes [25,8], we have shown that the

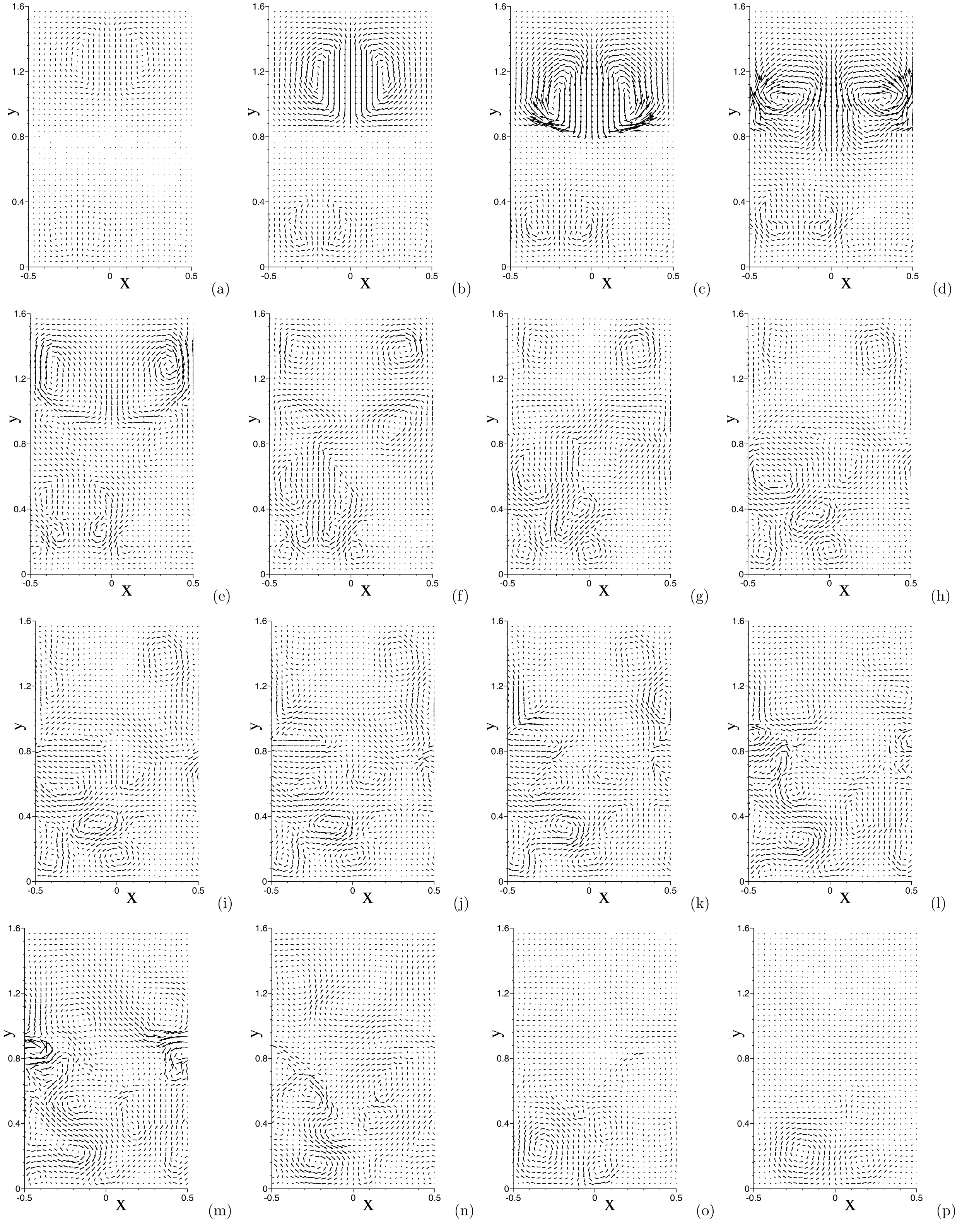

Fig. 9. Velocity fields of fluid drops impacting water surface (4 fluid phases): (a) $t = 0.076$, (b) $t = 0.176$, (c) $t = 0.26$, (d) $t = 0.332$, (e) $t = 0.46$, (f) $t = 0.608$, (g) $t = 0.712$, (h) $t = 0.796$, (i) $t = 0.836$, (j) $t = 0.888$, (k) $t = 0.944$, (l) $t = 1.128$, (m) $t = 1.196$, (n) $t = 1.504$, (o) $t = 2.208$, (p) $t = 3.384$. Velocity vectors are plotted on every 15th quadrature points in each direction on each element.

formulations and the numerical algorithm developed herein with general order parameters have produced physically accurate results for multiple fluid phases. The simulation results also demonstrate the complex dynamics induced by the interactions among multiple types of fluid interfaces.

It is instructive to compare the general order parameters discussed here for N-phase flows with the order parameter for two-phase flows. There exists only one independent order parameter for a two-phase system, and different order parameters have been used by different researchers in the literature. Several choices of the order parameter for two phases are touched on in e.g. [1], and the resultant two-phase field equation would usually only require a simple re-scaling for a different choice of the order parameter.

On the other hand, for an N-phase system there are $(N-1)$ independent order parameters. The possibilities for choosing the $(N-1)$ order parameters are much broader, and different choices lead to varying degrees of complexity in the resulting phase field equations. The class of order parameters considered in the current work, Eq. (13), has a linear relation between $(\rho_i - \rho_N)$ and ϕ_j. One can readily imagine even broader classes of order parameters, e.g.

$$\varphi_i(\vec{\phi}) = \rho_i(\vec{\phi}) - \rho_N(\vec{\phi}) = f_i(\vec{\phi}), \quad 1 \leqslant i \leqslant N-1, \tag{105}$$

where $f_i(\vec{\phi})$ are $(N-1)$ given functions and in general can be nonlinear. This, however, will lead to even more complicated forms for the phase field equations. It should be noted that, regardless of the set of order parameters being employed, the phase field equations all represent the mass balance relations for the N individual fluid phases. We hold the view that the formulations using different order parameters are merely different representations of the N-phase system, and that the different representations should be equivalent to one another. This is an embodiment of the representation invariance principle [32]. The explicit expressions for the mixing energy density coefficients λ_{ij} derived in the current work are a direct result of this principle.

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Appendix A. Unique solvability of λ_{ij} linear algebraic system

In this appendix we prove that the linear algebraic system about λ_{ij} derived in [10] for the special set of order parameters defined by (25) has a unique solution for any $N \geqslant 2$. The unique solvability of that system for $N \geqslant 4$ is an un-settled issue in [10].

The idea of the proof is as follows. We will show that the system of equations about Λ_{ij} ((46) and (48)) in the current work, under a non-singular transform, is equivalent to the linear algebraic system about λ_{ij} in [10] for the special set of order parameters defined by (25). Since the system consisting of (46) and (48) has a unique solution for any $N \geqslant 2$, the linear system from [10] must also have a unique solution.

The following is the linear algebraic system about λ_{ij} from [10], which is based on the set of order parameters defined by (25),

$$\sum_{i,j=1}^{N-1} L_i^{kl} L_j^{kl} \lambda_{ij} = \frac{9}{2}\frac{\eta^2}{\beta^2}\sigma_{kl}^2, \quad 1 \leqslant k < l \leqslant N, \tag{106}$$

where

$$L_i^{kl} = \begin{cases} \frac{\tilde{\rho}_k}{\tilde{\rho}_k+\tilde{\rho}_N}\delta_{ik} - \frac{\tilde{\rho}_l}{\tilde{\rho}_l+\tilde{\rho}_N}\delta_{il}, & 1 \leqslant k < l \leqslant N-1,\ 1 \leqslant i \leqslant N-1, \\ \frac{\tilde{\rho}_i}{\tilde{\rho}_i+\tilde{\rho}_N}\delta_{ik} + \frac{\tilde{\rho}_N}{\tilde{\rho}_i+\tilde{\rho}_N}, & 1 \leqslant k < l = N,\ 1 \leqslant i \leqslant N-1. \end{cases} \tag{107}$$

This is a system of $\frac{1}{2}N(N-1)$ equations about $\frac{1}{2}N(N-1)$ unknowns, noting the symmetry $\lambda_{ij} = \lambda_{ji}$ $(1 \leqslant i, j \leqslant N-1)$.

We next show that the linear system consisting of (46) and (48) about Λ_{ij} can be transformed to the system (106) about λ_{ij}, under the transform (59) between $\mathbf{A}$ and $\boldsymbol{\Lambda}$. Re-write (59) as

$$[\Lambda_{ij}]_{(N-1)\times(N-1)} = \boldsymbol{\Lambda} = \mathbf{Z}^{-1}\mathbf{L}^{-1}\mathbf{A}\mathbf{L}^{-1}\mathbf{Z}^{-1} = \boldsymbol{\Lambda}_1\mathbf{L}^{-1}\mathbf{A}\mathbf{L}^{-1}\boldsymbol{\Lambda}_1 = 4\mathbf{R}^T\mathbf{A}\mathbf{R} \tag{108}$$

where we have used Eq. (55) and $\boldsymbol{\Lambda}_1$ is defined in (38), and

$$\mathbf{R} = \frac{1}{2}\mathbf{L}^{-1}\boldsymbol{\Lambda}_1 = [r_{ij}]_{(N-1)\times(N-1)}, \qquad r_{ij} = \frac{\tilde{\rho}_i}{\tilde{\rho}_i+\tilde{\rho}_N}\delta_{ij} + \frac{\tilde{\rho}_N}{\tilde{\rho}_i+\tilde{\rho}_N} = L_i^{jN}. \tag{109}$$

First consider the case $1 \leqslant k < l \leqslant N-1$. Eq. (48) becomes

$$\frac{9}{2}\frac{\eta^2}{\beta^2}\sigma_{kl}^2 = \frac{1}{4}(\Lambda_{kk} + \Lambda_{ll} - 2\Lambda_{kl})$$

$$
\begin{aligned}
&= \left(\frac{\tilde{\rho}_k}{\tilde{\rho}_k + \tilde{\rho}_N}\right)^2 \lambda_{kk} + \left(\frac{\tilde{\rho}_l}{\tilde{\rho}_l + \tilde{\rho}_N}\right)^2 \lambda_{ll} - 2\frac{\tilde{\rho}_k}{\tilde{\rho}_k + \tilde{\rho}_N}\frac{\tilde{\rho}_l}{\tilde{\rho}_l + \tilde{\rho}_N}\lambda_{kl} \\
&= \left(L_k^{kl}\right)^2 \lambda_{kk} + \left(L_l^{kl}\right)^2 \lambda_{ll} + 2L_k^{kl}L_l^{kl}\lambda_{kl} \\
&= \sum_{i,j=1}^{N-1} L_i^{kl}L_j^{kl}\lambda_{ij}, \quad 1 \leqslant k < l \leqslant N-1
\end{aligned} \tag{110}
$$

where we have used (108) and (107). Therefore Eq. (48) is transformed to (106) for the case $1 \leqslant k < l \leqslant N-1$. Next consider the case $1 \leqslant k < l = N$. Eq. (46) becomes

$$
\frac{9}{2}\frac{\eta^2}{\beta^2}\sigma_{kN}^2 = \frac{1}{4}\Lambda_{kk} = \sum_{i,j=1}^{N-1} r_{ik}r_{jk}\lambda_{ij} = \sum_{i,j=1}^{N-1} L_i^{kN}L_j^{kN}\lambda_{ij}, \quad 1 \leqslant k < l = N \tag{111}
$$

where we have used (108), (107), and (109). Therefore, Eq. (46) is transformed to (106) for the case $1 \leqslant k < l = N$.

Since the transform (59) (or equivalently (108)) is non-singular, we conclude that the linear system consisting of (46) and (48) is equivalent to the linear system given by (106). We can then conclude that the linear system (106) about λ_{ij} has a unique solution for any $N \geqslant 2$, and that its solution is given by (59), in which $\boldsymbol{\Lambda}$ is given by (37), (46) and (49).

Appendix B. Algorithm for *N*-phase momentum equations

In this appendix we present an algorithm for the N-phase momentum equations, (14a) and (14b), together with the velocity boundary condition, (61).

We assume that the phase field variables ϕ_i $(1 \leqslant i \leqslant N-1)$ are known, and our goal is to compute the velocity and pressure from (14a) and (14b). Let

$$
P = p + \sum_{i,j=1}^{N-1} \frac{\lambda_{ij}}{2}\nabla\phi_i \cdot \nabla\phi_j \tag{112}
$$

denote an auxiliary pressure, which will also be loosely referred to as pressure hereafter. We can then transform (14a) into

$$
\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{u} + \frac{1}{\rho}\tilde{\mathbf{J}}\cdot\nabla\mathbf{u} = \frac{1}{\rho}\nabla P + \frac{\mu}{\rho}\nabla^2\mathbf{u} + \frac{1}{\rho}\nabla\mu\cdot\mathbf{D}(\mathbf{u}) - \frac{1}{\rho}\sum_{i,j=1}^{N-1}\lambda_{ij}\nabla^2\phi_j\nabla\phi_i + \frac{1}{\rho}\mathbf{f}. \tag{113}
$$

The following algorithm is to solve the equations (113) and (14b), together with the boundary condition (61). Given $(\mathbf{u}^n, P^n, \phi_i^{n+1})$, we compute P^{n+1} and $\mathbf{u}^{n+1}$ successively in a de-coupled fashion as follows:

For P^{n+1}:

$$
\begin{aligned}
&\frac{\gamma_0\tilde{\mathbf{u}}^{n+1} - \hat{\mathbf{u}}}{\Delta t} + \mathbf{u}^{*,n+1}\cdot\nabla\mathbf{u}^{*,n+1} + \frac{1}{\rho^{n+1}}\tilde{\mathbf{J}}^{n+1}\cdot\nabla\mathbf{u}^{*,n+1} + \frac{1}{\rho_0}\nabla P^{n+1} \\
&\quad = \left(\frac{1}{\rho_0} - \frac{1}{\rho^{n+1}}\right)\nabla P^{*,n+1} - \frac{\mu^{n+1}}{\rho^{n+1}}\nabla\times\nabla\times\mathbf{u}^{*,n+1} + \frac{1}{\rho^{n+1}}\nabla\mu^{n+1}\cdot\mathbf{D}\left(\mathbf{u}^{*,n+1}\right) \\
&\qquad - \frac{1}{\rho^{n+1}}\sum_{i,j=1}^{N-1}\lambda_{ij}\nabla^2\phi_j^{n+1}\nabla\phi_i^{n+1} + \frac{1}{\rho^{n+1}}\mathbf{f}^{n+1},
\end{aligned} \tag{114a}
$$

$$
\nabla\cdot\tilde{\mathbf{u}}^{n+1} = 0, \tag{114b}
$$

$$
\mathbf{n}\cdot\tilde{\mathbf{u}}^{n+1}\big|_{\partial\Omega} = \mathbf{n}\cdot\mathbf{w}^{n+1}. \tag{114c}
$$

For $\mathbf{u}^{n+1}$:

$$
\frac{\gamma_0\mathbf{u}^{n+1} - \gamma_0\tilde{\mathbf{u}}^{n+1}}{\Delta t} - \nu_0\nabla^2\mathbf{u}^{n+1} = \nu_0\nabla\times\nabla\times\mathbf{u}^{*,n+1}, \tag{115a}
$$

$$
\mathbf{u}^{n+1}\big|_{\partial\Omega} = \mathbf{w}^{n+1}. \tag{115b}
$$

In the above equations all the symbols follow the notation outlined in Section 3.1. $\mathbf{u}^{*,n+1}$ and $P^{*,n+1}$ are defined by (64). $\hat{\mathbf{u}}$ and γ_0 are defined by (65). ρ^{n+1} and μ^{n+1} are given by (96), and also (97) in case of large density ratios among the N fluids. $\tilde{\mathbf{J}}^{n+1}$ is given by (95). $\mathbf{f}^{n+1}$ is the external body force evaluated at time step $(n+1)$. $\mathbf{n}$ is the outward-pointing unit vector normal to $\partial\Omega$. $\tilde{\mathbf{u}}^{n+1}$ is an auxiliary velocity that approximates $\mathbf{u}^{n+1}$. ρ_0 is a chosen constant that must satisfy the condition

$$0 < \rho_0 \leqslant \min(\tilde{\rho}_1, \tilde{\rho}_2, \ldots, \tilde{\rho}_N). \tag{116}$$

ν_0 in (115a) is a chosen positive constant that is sufficiently large. A conservative condition for ν_0 is given in [10]. But in the current paper we will generally employ a value for ν_0 satisfying

$$\nu_0 \geqslant \max\left(\frac{\tilde{\mu}_1}{\tilde{\rho}_1}, \frac{\tilde{\mu}_2}{\tilde{\rho}_2}, \cdots, \frac{\tilde{\mu}_N}{\tilde{\rho}_N},\right). \tag{117}$$

The above algorithm employs a velocity correction-type idea [17,15,13,12] to de-couple the computations for the pressure and the velocity. The differences between this algorithm and that of [10] lie in two aspects. Firstly, in the pressure substep (Eq. (114a)) all the terms in the momentum equations have been approximated at the time step $(n+1)$ in the current algorithm. In contrast, in the pressure substep of [10], while the time derivative term is approximated at time step $(n+1)$, the other terms are approximated at time step n rather than $(n+1)$. Secondly, the velocity substep of the scheme of [10] contains a number of correction terms to offset the effects caused by the less accurate approximations using data from time step n in the preceding pressure substep. On the other hand, the velocity substep of the current algorithm (Eq. (115a)) does not contain such correction terms.

It can be noted that the variable density ρ and the variable dynamic viscosity μ have been treated with a reformulation of the pressure term $\frac{1}{\rho}\nabla P$ and a reformulation of the viscous term $\frac{\mu}{\rho}\nabla^2\mathbf{u}$. These eventually lead to linear algebraic systems involving only *constant* and *time-independent* coefficient matrices for the pressure and velocity. The ideas for these reformulations stem from the original developments for two-phase flows [14,9,11].

We next derive the weak forms for the pressure and the velocity in order to facilitate the implementation using C^0 spectral elements. Let $q \in H^1(\Omega)$ denote the test function, and let

$$\begin{aligned}\mathbf{G}^{n+1} = {} & \frac{1}{\rho^{n+1}}\mathbf{f}^{n+1} - \left(\mathbf{u}^{*,n+1} + \frac{1}{\rho^{n+1}}\tilde{\mathbf{J}}^{n+1}\right)\cdot\nabla\mathbf{u}^{*,n+1} + \frac{\hat{\mathbf{u}}}{\Delta t} + \left(\frac{1}{\rho_0} - \frac{1}{\rho^{n+1}}\right)\nabla P^{*,n+1} \\ & + \frac{1}{\rho^{n+1}}\nabla\mu^{n+1}\cdot\mathbf{D}\big(\mathbf{u}^{*,n+1}\big) - \frac{1}{\rho^{n+1}}\sum_{i,j=1}^{N-1}\lambda_{ij}\nabla^2\phi_j^{n+1}\nabla\phi_i^{n+1} + \nabla\left(\frac{\mu^{n+1}}{\rho^{n+1}}\right)\times\boldsymbol{\omega}^{*,n+1},\end{aligned} \tag{118}$$

where $\boldsymbol{\omega} = \nabla\times\mathbf{u}$ is the vorticity. Take the L^2 inner product between Eq. (114a) and ∇q, and we get the weak form about P^{n+1},

$$\int_\Omega \nabla P^{n+1}\cdot\nabla q = \rho_0\int_\Omega \mathbf{G}^{n+1}\cdot\nabla q - \rho_0\int_{\partial\Omega}\frac{\mu^{n+1}}{\rho^{n+1}}\mathbf{n}\times\boldsymbol{\omega}^{*,n+1}\cdot\nabla q - \frac{\gamma_0\rho_0}{\Delta t}\int_{\partial\Omega}\mathbf{n}\cdot\mathbf{w}^{n+1}q, \quad \forall q \in H^1(\Omega) \tag{119}$$

where we have used integration by part, Eqs. (114b) and (114c), the divergence theorem, and the identity $\frac{\mu}{\rho}\nabla\times\boldsymbol{\omega}\cdot\nabla q = \nabla\cdot(\frac{\mu}{\rho}\boldsymbol{\omega}\times\nabla q) - \nabla(\frac{\mu}{\rho})\times\boldsymbol{\omega}\cdot\nabla q$.

Adding together Eqs. (114a) and (115a), we get

$$\frac{\gamma_0}{\Delta t}\mathbf{u}^{n+1} - \nu_0\nabla^2\mathbf{u}^{n+1} = \mathbf{G}^{n+1} - \nabla\left(\frac{\mu^{n+1}}{\rho^{n+1}}\right)\times\boldsymbol{\omega}^{*,n+1} - \frac{1}{\rho_0}\nabla P^{n+1} - \left(\frac{\mu^{n+1}}{\rho^{n+1}} - \nu_0\right)\nabla\times\boldsymbol{\omega}^{*,n+1} \tag{120}$$

Let $H_0^1(\Omega) = \{v \in H(\Omega) : v|_{\partial\Omega} = 0\}$, and $\varphi \in H_0^1(\Omega)$ denote the test function. Taking the L^2 inner product between Eq. (120) and φ, one can get the weak form about $\mathbf{u}^{n+1}$,

$$\begin{aligned}\int_\Omega \nabla\varphi\cdot\nabla\mathbf{u}^{n+1} + \frac{\gamma_0}{\nu_0\Delta t}\int_\Omega \varphi\mathbf{u}^{n+1} = {} & \frac{1}{\nu_0}\int_\Omega\left(\mathbf{G}^{n+1} - \frac{1}{\rho_0}\nabla P^{n+1}\right)\varphi \\ & - \frac{1}{\nu_0}\int_\Omega\left(\frac{\mu^{n+1}}{\rho^{n+1}} - \nu_0\right)\boldsymbol{\omega}^{*,n+1}\times\nabla\varphi, \quad \forall\varphi \in H_0^1(\Omega),\end{aligned} \tag{121}$$

where we have used integration by part, the divergence theorem, the identity (χ denoting a scalar function)

$$\int_\Omega \chi\nabla\times\boldsymbol{\omega}\varphi = \int_{\partial\Omega}\chi\mathbf{n}\times\boldsymbol{\omega}\varphi - \int_\Omega \nabla\chi\times\boldsymbol{\omega}\varphi + \int_\Omega \chi\boldsymbol{\omega}\times\nabla\varphi,$$

and the fact that the surface integrals of type $\int_{\partial\Omega}\chi\varphi$ vanish because $\varphi \in H_0^1(\Omega)$.

The weak forms for the pressure and the velocity, (119) and (121), can be discretized in space using C^0 spectral elements in a straightforward fashion. Note that the terms $\nabla^2\phi_i^{n+1}$ ($1 \leqslant i \leqslant N-1$) involved in the $\mathbf{G}^{n+1}$ expression (118) and in the $\tilde{\mathbf{J}}^{n+1}$ expression (see (95) and (20)) must be computed based on Eq. (94).

Therefore, solving the N-phase momentum equations amounts to the following two successive operations. First, solve Eq. (119) for pressure P^{n+1}. Then, solve Eq. (121), together with the Dirichlet condition (115b) on $\partial\Omega$, for $\mathbf{u}^{n+1}$.

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