COMMUN. MATH. SCI. © 2020 International Press
Vol. 18, No. 5, pp. 1441–1468

THERMODYNAMICALLY CONSISTENT HYDRODYNAMIC MODELS OF MULTI-COMPONENT COMPRESSIBLE FLUID FLOWS*

XUEPING ZHAO†, TIEZHENG QIAN‡, AND QI WANG§

Abstract. We present a systematic derivation of thermodynamically consistent hydrodynamic models for multi-component, compressible viscous fluid mixtures under isothermal conditions using the generalized Onsager principle and the one-fluid multi-component formulation. By maintaining momentum conservation while enforcing mass conservation at different levels, we obtain two compressible models. When the fluid components in the mixture are incompressible, we show that one compressible model reduces to the quasi-incompressible model via a Lagrange multiplier approach. Several different approaches to arriving at the quasi-incompressible model are discussed. Finally, we conduct a linear stability analysis on all the binary fluid models derived in the paper to show the differences of the models in near equilibrium dynamics.

Keywords. Compressible fluid; Quasi-incompressible fluid; Multi-component fluid mixtures; Phase field model; Linear stability.

AMS subject classifications. 35Q30; 35Q35; 35Q79;

1. Introduction

Fluid mixtures are ubiquitous in nature as well as in industrial applications. In a fluid mixture, when fluid components are compressible, the fluid mixture remains compressible. While in some fluid mixtures, when each fluid component is incompressible, the fluid mixture may not be incompressible especially when the densities are not equal. This fluid mixture was named a quasi-incompressible fluid and its hydrodynamic model has been derived under the assumption of the simple mixture and applied to various multi-phase fluid flows [18, 19, 27, 32]. The fluid mixture may be truly incompressible only when all the fluid components are incompressible and of the same specific densities. For immiscible fluid mixtures, sharp interface models and phase field models can both be used to describe fluid motions. While for miscible fluid mixtures, sharp interface models are no longer applicable. So, the phase field model becomes a primary platform to describe the fluid motion in the mixture. We use the name of phase field model in this paper to refer to the model in which order parameters in the form of concentrations (molar or mass density) or component fractions (volume or mass fraction) are used to describe the internal component or structure of the material system. It encompasses both immiscible and miscible fluid mixture models.

Phase field methods have been used successfully to formulate models for fluid mixtures in many applications like in life sciences [38, 39, 42, 46, 49] (cell biology [26, 33, 38, 48, 50, 51], biofilms [45–47], cell adhesion and motility [8, 30, 33, 34, 38], cell membrane [2, 15, 41, 43], tumor growth [42]), materials science [3, 7, 9], fluid dynamics [14, 31, 32, 40], image processing [6, 28], etc. The most widely studied phase field model for binary fluid mixtures is the one for immiscible fluid mixtures of two incompressible fluids of identical densities [1, 29]. While modeling binary fluids of two

*Received: January 25, 2019; Accepted (in revised form): March 14, 2020. Communicated by Jie Shen.

†Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Str. 38, 01187 Dresden, Germany (xzhao@pks.mpg.de).

‡Department of Mathematics, The Hong Kong University of Science & Technology, Clear Water Bay, Kowloon, Hong Kong (maqian@ust.hk).

§Department of Mathematics, University of South Carolina, Columbia, SC 29208, USA (qwang@math.sc.edu).

immiscible fluids using phase field models, one commonly uses an order parameter, known as the labeling or a phase variable (a volume fraction or a mass fraction), ϕ to distinguish between distinct material phases. For instance $\phi=1$ indicates one fluid phase while $\phi=0$ denotes the other fluid phase in the binary fluid mixture. For the immiscible binary fluid, the interfacial region is tracked by $0<\phi<1$. For miscible fluid mixtures or blends, order parameter in the phase field model is normally given in the form of component fractions or concentrations, in which phases of the fluid mixture are distinguished by the physically significant stable mixtures of the material system. For instance, in blends of polymer A and B, two stable phases can co-exist in which neither are pure polymer A or polymer B. A transport equation for the phase variable(s) along with the conservation equations of momentum, the continuity equation together with necessary constitutive equations constitute the governing system of equations for the fluid mixture.

In a compressible fluid, the total density ρ is a variable and the mass conservation is given by

$$\frac{\partial\rho}{\partial t}+\nabla\cdot(\rho\mathbf{v})=0, \tag{1.1}$$

where $\mathbf{v}$ is the mass average velocity field. In a single component incompressible fluid, the density ρ is a constant so that the mass conservation Equation (1.1) reduces to

$$\nabla\cdot\mathbf{v}=0. \tag{1.2}$$

In multi-component fluid models in forms of phase field models, continuity condition (1.2) may not be valid in light of the consistency condition with the second law of thermodynamics. In these models, the divergence-free condition has to be modified to accommodate the potential fluid compressibility or quasi-incompressibility. A systematic derivation of the phase field model for such fluid mixtures of viscous fluids was given by Lowengrub and Truskinovsky using the mass fraction as the phase variable for binary fluid mixtures [32] and by Li and Wang using the volume fractions as the phase variables for multi-component fluid mixtures [27]. The derivations were based on the thermodynamic laws, especially, the second law of thermodynamics and the simple mixture assumption [25] coupled with the additional constraints imposed by the transport equation of the components consistent with the Onsager linear response theory.

It's known that a hydrodynamic model of a single phase incompressible fluid can be derived from the corresponding compressible model by imposing the incompressibility constraint. The resulting model is called a constrained theory in continuum mechanics. In nature and industrial applications, there are many material systems comprising of multi-component compressible or incompressible components. For instance, in modeling tissues, there is the issue of cell proliferation which makes the volume of the material system and mass grow; in tertiary oil recovery, the mixture of CO_2 and n-decane are two important compressible fluid components in the gas-oil mixture. In fact, there are many more material systems in real world applications in this category, where the material components are compressible.

In this paper, we present a systematic derivation of thermodynamically consistent compressible phase field models for multi-component fluid mixtures through a variational approach in light of the generalized Onsager principle [44]. The generalized Onsager principle consists of the Onsager linear response theory and positive entropy production rule [35, 36]. Thermodynamical consistency indicates the models satisfy the thermodynamical laws (including the first and second laws of thermodynamics). Historically, there have been several theoretical frameworks for one to derive thermodynamical and hydrodynamical models for time-dependent dynamics. The Onsager principle is the one we adopt in this paper. The Onsager maximum entropy production principle based on the Onsager-Matchlup action potential is another approach to deriving models for purely dissipative systems [13, 35]. Equivalently, the second law of thermodynamics formulated in the form of Clausius-Duhem relation is yet another classical approach to deriving transient dynamical models [22]. The Hamilton least action principle is a classical one for formulating models for Hamiltonian or conservative systems. The Hamilton-Rayleigh principle is an incarnation of the Onsager maximum entropy principle [4, 12, 16, 17, 20, 21]. There are also more elaborate GENERIC and Poisson bracket formalism for non-equilibrium theories [5, 10, 11, 23]. These formulations share the commonality in that the non-equilibrium models have a unified mathematical structure consisting of a reversible (hyperbolic) and irreversible (parabolic, dissipative) component in the model governing the evolutionary process. Some of these equations represent conservation laws for the material system such as mass, momentum and energy conservation while others serve as constitutive equations pertinent to the material properties of the material system. The different methods may differ however in how they handle the boundary conditions as well as if one uses the dissipation functional or the mobility (or the friction coefficient) to derive the constitutive equations.

There are two general approaches to describing multiphasic materials in general. One uses multi-fluid formulation to describe the density and velocity for each phase explicitly [4, 12, 16, 17, 20, 21]. Another one uses an average velocity, normally the mass average velocity, together with chemical potentials to describe kinematics for each phase [5, 10]. In the latter approach, the average velocity is a measurable hydrodynamic quantity in fluids. For this reason, we choose this approach to formulate our phase field model for multiphasic fluid flows. Since we consider isothermal fluid systems in this paper, we will use the word "multi-phase" and "multi-component" fluid interchangeably.

We formulate the hydrodynamic phase field model for compressible fluid of N-fluid components ($N > 1$) using the one fluid multi-component formulation in this paper [5]. As it is alluded to earlier, hydrodynamic models that obey necessary conservation laws do not necessarily satisfy the second law of thermodynamics, if the constitutive equations are not derived in a thermodynamically consistent way. The second law or equivalently the Onsager positive entropy production principle is thus an additional condition that any well-posed model should satisfy. It does not yield an additional governing equation for the model. Instead, it does impose an additional constraint on the model and dictates how entropy is produced during the transient dynamical process as the system approaches the steady state.

In this paper, we first derive two classes of hydrodynamic phase field models for compressible fluid mixtures using the Onsager principle. After we obtain the "general" compressible models for multi-component fluid mixtures, we hierarchically impose additional "conservation" and/or "incompressibility" conditions to the material system to arrive at constrained, quasi-incompressible theories to show the hierarchical relationship between the compressible model and the constrained models for multi-component fluid mixtures. Through this systematic approach, we demonstrate how one can derive the constrained models via the generalized Onsager principle in a variational formulation involving Lagrange multipliers, extending the method applied to single phase materials to multi-component material mixtures in the context of one fluid multi-component framework. In the more general compressible model, we enforce the global mass conservation so that the model can be used to describe material systems undergoing mass conversion among different components. We then study near equilibrium dynamics of the general models and their various limits through a linear stability analysis. Note that we derive the models for viscous fluid components in this paper. However, this approach can be readily extended to complex fluids to account for viscoelastic effects due to mesoscopic structures in the fluid systems [44].

The paper is organized as follows. In §2, we formulate two classes of hydrodynamic phase field models for fluid mixtures of compressible fluid components and a quasi-incompressible model for the fluid mixture of two incompressible fluids with different mass conservation constraints. In §3, we generalize the derivation to compressible fluid mixtures of N fluid components. The non-dimensionalization of the models is carried out in §4. In §5, we discuss near-equilibrium dynamics of the models using a linear stability analysis. We give concluding remarks in §6.

2. Hydrodynamic phase field models for binary fluid flows

We present a systematic derivation of thermodynamically consistent hydrodynamic phase field models for binary compressible fluid flows with respect to various conditions on mass conservation and incompressibility following the generalized Onsager principle [44].

2.1. Compressible model with the global mass conservation law. We first consider a mixture of two compressible viscous fluids with density and velocity pairs $(\rho_1, \mathbf{v}_1)$ and $(\rho_2, \mathbf{v}_2)$, respectively. We define the total mass of the fluid mixture as $\rho = \rho_1 + \rho_2$ and the mass average velocity as $\mathbf{v} = \frac{1}{\rho}(\rho_1 \mathbf{v}_1 + \rho_2 \mathbf{v}_2)$. We allow the mass of fluid components to change via conversion, generation, or annihilation at specified rates. In this general framework, the mass balance equation for each fluid component is given respectively by

$$\frac{\partial \rho_i}{\partial t} + \nabla \cdot (\rho_i \mathbf{v}_i) = r_i, \quad i = 1,2, \tag{2.1}$$

where r_i is the mass conversion/generation/annihilation rate for the ith component. The corresponding momentum conservation equations are given by

$$\frac{\partial (\rho_i \mathbf{v}_i)}{\partial t} + \nabla \cdot (\rho_i \mathbf{v}_i \mathbf{v}_i) = \nabla \cdot \sigma_i + \mathbf{F}_{i,e} + r_i \mathbf{v}_i, \quad i = 1,2, \tag{2.2}$$

where $(\rho_i \mathbf{v}_i \mathbf{v}_i)_{\alpha\beta} = \rho_i \mathbf{v}_{i,\alpha} \mathbf{v}_{i,\beta}, \alpha, \beta = 1,2,3$, σ_i is the viscous stress of the ith fluid component, $(\nabla \cdot \sigma_i)_\alpha = \frac{\partial \sigma_{i,\alpha\beta}}{\partial \beta}, \alpha = 1,2,3$, $\mathbf{F}_{i,e}$ the extra force density of the ith fluid component including the friction force between different fluid components and some elastic forces, and $r_i \mathbf{v}_i$ the rate of momentum change due to mass conversion/generation/annihilation in the ith fluid component.

We rewrite the mass conservation equations using the average velocity as follows

$$\frac{\partial \rho_i}{\partial t} + \nabla \cdot (\rho_i \mathbf{v}) = j_i, \quad i = 1,2, \tag{2.3}$$

where $j_i = \nabla \cdot (\rho_i (\mathbf{v} - \mathbf{v}_i)) + r_i$ is the excessive production rate of the ith fluid component.

If we add mass balance Equations (2.3) and linear momentum Equations (2.2) of all the components, respectively, we obtain the total mass balance equation and total linear momentum balance equation as follows

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = \sum_{i=1}^{2} j_i = \sum_{i=1}^{2} r_i,$$

$$\frac{\partial(\rho\mathbf{v})}{\partial t}+\nabla\cdot(\rho\mathbf{v}\mathbf{v})=\nabla\cdot\sigma^s+\mathbf{F}_e, \tag{2.4}$$

where $\mathbf{F}_e=\sum_{i=1}^{i=2}(\mathbf{F}_{i,e}+r_i\mathbf{v}_i)$ and $\sigma^s=\sum_{i=1}^{i=2}(\sigma_i-\rho_i(\mathbf{v}_i-\mathbf{v})(\mathbf{v}_i-\mathbf{v}))$ is the stress tensor. The angular momentum balance implies the symmetry of σ^s. All j_i, i=1, 2, σ^s and $\mathbf{F}_e$ will be determined later through constitutive relations.

We assume the free energy of the system is given by

$$F=\int_V f(\rho_1,\rho_2,\nabla\rho_1,\nabla\rho_2)d\mathbf{x}, \tag{2.5}$$

where f is the free energy density function and V is the domain which the fluid mixture occupies. The total energy of the system is given by

$$E_{total}=\int_V[\frac{1}{2}\rho||\mathbf{v}||^2+f]d\mathbf{x}. \tag{2.6}$$

We next calculate the total energy dissipation rate as follows.

$$\begin{aligned}\frac{dE_{total}}{dt}=&\int_V[-\sigma^s:\mathbf{D}+(\mathbf{F}_e+\rho_1\nabla\mu_1+\rho_2\nabla\mu_2-\frac{1}{2}(j_1+j_2)\mathbf{v})\cdot\mathbf{v}+\mu_1(j_1)+\mu_2(j_2)]d\mathbf{x}\\&+\int_{\partial V}[(\sigma^s-\frac{1}{2}\|\mathbf{v}\|^2-\mu_1\rho_1-\mu_2\rho_2)\cdot\mathbf{v}+\frac{\partial f}{\partial(\nabla\rho_1)}\frac{\partial\rho_1}{\partial t}+\frac{\partial f}{\partial(\nabla\rho_2)}\frac{\partial\rho_2}{\partial t}]\cdot\mathbf{n}dS.\end{aligned} \tag{2.7}$$

where $\mu_1=\frac{\partial f}{\partial\rho_1}-\nabla\cdot\frac{\partial f}{\partial\nabla\rho_1}$, $\mu_2=\frac{\partial f}{\partial\rho_2}-\nabla\cdot\frac{\partial f}{\partial\nabla\rho_2}$ are the chemical potentials with respect to ρ_1 and ρ_2, respectively, $\mathbf{D}=\frac{1}{2}(\nabla\mathbf{v}+\nabla\mathbf{v}^T)$ is the rate of strain tensor. It contains the energy rate of change in time in the bulk, which is known as the energy dissipation rate and the rate of change due to boundary fluxes. We focus on the bulk energy dissipation rate firstly.

We define the elastic force as

$$\mathbf{F}_e=-\rho_1\nabla\mu_1-\rho_2\nabla\mu_2+\frac{1}{2}(j_1+j_2)\mathbf{v}. \tag{2.8}$$

This indicates that the elastic force density $\mathbf{F}_e$ does not contribute to the energy dissipation. Using the Onsager principle, we propose

$$\sigma^s=2\eta\mathbf{D}+\nu tr(\mathbf{D})\mathbf{I}, \tag{2.9}$$

$$\begin{pmatrix}j_1\\j_2\end{pmatrix}=-\mathcal{M}\begin{pmatrix}\mu_1\\\mu_2\end{pmatrix}, \tag{2.10}$$

where η,ν are mass-average shear and volumetric viscosities, respectively, and $\mathcal{M}$ is an operator. The bulk energy dissipation rate reduces to

$$\frac{dE_{total}}{dt}=-\int_V[2\eta\mathbf{D}:\mathbf{D}+\nu tr(\mathbf{D})^2+(\mu_1,\mu_2)\cdot\mathcal{M}\cdot(\mu_1,\mu_2)]d\mathbf{x}. \tag{2.11}$$

It is non-positive definite provided $\mathcal{M}$ is non-negative definite and η,ν are non-negative. The constitutive relation gives a general compressible model for binary fluid flows.

The energy dissipation rate related to the boundary conditions is equally important. It defines how the material system interacts with the surrounding environment. For example, the following boundary conditions make the term vanish:

$$\mathbf{v}=0,\quad \mathbf{n}\cdot\frac{\partial f}{\partial\nabla\rho_1}=\mathbf{n}\cdot\frac{\partial f}{\partial\nabla\rho_2}=0\quad\text{ or }\quad\rho_1=\rho_1(\mathbf{x}),\rho_2=\rho_2(\mathbf{x}). \tag{2.12}$$

Another set of boundary conditions makes it dissipative:

$$\mathbf{v}=0, \quad \frac{\partial \rho_1}{\partial t}=-\lambda_1 \mathbf{n}\cdot\frac{\partial f}{\partial \nabla \rho_1}, \quad \frac{\partial \rho_2}{\partial t}=-\lambda_2 \mathbf{n}\cdot\frac{\partial f}{\partial \nabla \rho_2}, \tag{2.13}$$

where $\lambda_{1,2}>0$. We can consider more general boundary conditions using the Onsager principle at the boundary, which we will not pursue in this study.

In practice, the interesting scenarios are the following two:

(1) $\int_V \sum_{i=1}^2 r_i=0$; so, $\int_V \sum_{i=1}^2 j_i=0$.

(2) $r_i=0, i=1,2$; so, $\sum_{i=1}^2 j_i=0$.

The first condition yields the compressible model of global mass conservation law while the second one gives the compressible model of local mass conservation law. For the first case, a special choice of the mobility operator is the following

$$j_1=\nabla\cdot M_{11}\nabla\mu_1+\nabla\cdot M_{12}\nabla\mu_2,$$

$$j_2=\nabla\cdot M_{21}\nabla\mu_1+\nabla\cdot M_{22}\nabla\mu_2, \tag{2.14}$$

where $M_{ij}, i,j=1,2$ are mobility coefficients. If we set the boundary conditions as in (2.12), the surface terms vanish in the energy dissipation functional so that the energy dissipation rate reduces to

$$\frac{dE_{total}}{dt}=-\int_V [2\eta\mathbf{D}:\mathbf{D}+\nu tr(\mathbf{D})^2+(\nabla\mu_1,\nabla\mu_2)\cdot\mathbf{M}\cdot(\nabla\mu_1,\nabla\mu_2)]d\mathbf{x}, \tag{2.15}$$

where $\mathbf{M}=(M_{ij})$. It is non-positive definite provided $\eta,\nu\geq 0$ and $\mathbf{M}$ is non-negative definite.

We summarize the governing system of equations in the hydrodynamic model for binary compressible fluids with a global mass conservation law as follows:

$$\begin{cases} \frac{\partial\rho_1}{\partial t}+\nabla\cdot(\rho_1\mathbf{v})=j_1=\nabla\cdot M_{11}\cdot\nabla\mu_1+\nabla\cdot M_{12}\cdot\nabla\mu_2, \\ [2mm]\frac{\partial\rho_2}{\partial t}+\nabla\cdot(\rho_2\mathbf{v})=j_2=\nabla\cdot M_{21}\cdot\nabla\mu_1+\nabla\cdot M_{22}\cdot\nabla\mu_2, \\ [2mm]\frac{\partial(\rho\mathbf{v})}{\partial t}+\nabla\cdot(\rho\mathbf{v}\mathbf{v})-\frac{1}{2}(j_1+j_2)\mathbf{v}=2\nabla\cdot(\eta\mathbf{D})+\nabla(\nu\nabla\cdot\mathbf{v})-\rho_1\nabla\mu_1-\rho_2\nabla\mu_2. \end{cases} \tag{2.16}$$

We denote the shear viscosities of the fluid component 1 and 2 as η_1,η_2, and the volumetric viscosities of the two components as ν_1,ν_2, respectively. There are several options of defining average viscosity coefficients in the binary model.

(1) Viscosity coefficients are interpolated using mass fractions and given by

$$\eta=\frac{\rho_1}{\rho}\eta_1+\frac{\rho_2}{\rho}\eta_2, \qquad \nu=\frac{\rho_1}{\rho}\nu_1+\frac{\rho_2}{\rho}\nu_2. \tag{2.17}$$

(2) Viscosity coefficients are interpolated through volume fractions ϕ and $(1-\phi)$ in quasi-incompressible models (presented later) and given by

$$\eta=\phi\eta_1+(1-\phi)\eta_2, \qquad \nu=\phi\nu_1+(1-\phi)\nu_2, \tag{2.18}$$

where ϕ is the volume fraction of fluid 1.

(3) By the Krieger-Dougherty law, the shear viscosity η exhibits a strong non-linear dependence on the local solute concentration and is given by

$$\eta(x)=\eta_0(1-x)^{-\nu}, \tag{2.19}$$

in which x is the solute concentration (ρ_1 or ρ_2 in this model), η_0 is the viscosity of the pure solvent. For example in mixtures of CO_2 and n-decane, the solvent is n-decane and solute is CO_2. The volumetric viscosity is obtained analogously.

As a customary approximation, we assume the free energy density function f is composed of the conformational entropy, and the bulk energy h as follows

$$f(\rho_1,\rho_2,\nabla\rho_1,\nabla\rho_2)=h(\rho_1,\rho_2,T)+\frac{1}{2}(\kappa_{\rho_1\rho_1}(\nabla\rho_1)^2+2\kappa_{\rho_1\rho_2}(\nabla\rho_1,\nabla\rho_2)+\kappa_{\rho_2\rho_2}(\nabla\rho_2)^2). \tag{2.20}$$

where T is the absolute temperature, $h(\rho_1,\rho_2,T)$ is the homogeneous bulk free energy density function, $\kappa_{\rho_1\rho_1},\kappa_{\rho_1\rho_2}$ and $\kappa_{\rho_2\rho_2}$ are parameters parameterizing the conformational entropy, which are all functions of T. For example, for the partially miscible binary fluid mixture of n-decane and CO_2, where n-decane is denoted as fluid 1 and CO_2 as fluid 2, the Peng-Robinson bulk free energy density is defined by the following

$$\begin{aligned}h(\rho_1,\rho_2,T)=&\frac{r_m\rho_1+\rho_2}{m_2}\varphi(T)-\frac{r_m\rho_1+\rho_2}{m_2}RTln(\frac{m_2}{r_m\rho_1+\rho_2}-b)\\&-\frac{r_m\rho_1+\rho_2}{m_2}\frac{a}{2\sqrt{2}b}ln[\frac{m_2+(r_m\rho_1+\rho_2)b(1+\sqrt{2})}{m_2+(r_m\rho_1+\rho_2)b(1-\sqrt{2})}]\\&+\frac{r_m\rho_1+\rho_2}{m_2}RT[\frac{r_m\rho_1}{r_m\rho_1+\rho_2}ln\frac{r_m\rho_1}{r_m\rho_1+\rho_2}+\frac{\rho_2}{r_m\rho_1+\rho_2}ln\frac{\rho_2}{r_m\rho_1+\rho_2}].\end{aligned} \tag{2.21}$$

where R is the ideal gas constant, $\varphi(T)=-RT(1-log(\lambda^3))$ is a temperature-dependent function, λ is the thermal wavelength of a massive particle, m_i is the molar mass of component i for $i=1,2$, respectively, $r_m=m_2/m_1$ is the ratio of the molar mass of carbon dioxide m_2 to the molar mass of n-decane m_1, $b(\rho_1,\rho_2)$ is a volume parameter and $a(\rho_1,\rho_2,T)$ is an interaction parameter. This free energy was proposed to extend that of the Van der Waals' to describe the deviation away from the ideal gas model.

Another example of the bulk free energy density for polymeric liquids is given by the Flory-Huggins-type mixing energy density

$$h(\rho_1,\rho_2,T)=\frac{k_BT}{m}[\frac{\rho_1}{N_1}ln\frac{\rho_1}{\rho}+\frac{\rho_2}{N_2}ln\frac{\rho_2}{\rho}+\chi\frac{\rho_1\rho_2}{\rho}], \tag{2.22}$$

where m is the mass of an average molecule in the mixture and $N_{1,2}$ are two polymerization indices.

Notice that $j_i,i=1,2$ in (2.16) are obtained from the constitutive equation and if $\sum_{i=1}^2 j_i\neq 0$, this model does not necessarily conserve mass locally. However, $\int_V(\rho_1+\rho_2)d\mathbf{x}$ is a constant. So, the mass of the system is conserved globally. This model describes a binary viscous compressible fluid system in which mass is conserved globally but not locally. In this model, the exact physical meaning of the velocity is lost due to the lack of local mass conservation. It is no longer a mass average velocity! Therefore, what does the momentum equation stand for becomes less clear physically. The applicability of this model needs to be scrutinized further. A more general model can be built from (2.9) by specifying a more general mobility operator $\mathcal{M}$. However, we will not pursue it in this study.

Next, we impose the local mass conservation constraint to arrive at the model that conserves mass locally.

2.2. Compressible model with local mass conservation law. If $j_1+j_2=0$, the total mass of the system is conserved locally, i.e.,

$$\frac{\partial \rho}{\partial t}+\nabla\cdot(\rho\mathbf{v})=0, \tag{2.23}$$

which imposes a constraint on the mass fluxes:

$$\sum_{i=1}^{2}\sum_{j=1}^{2}\nabla\cdot M_{ij}\cdot\nabla\mu_j=0. \tag{2.24}$$

We obtain the governing system of equations for the compressible fluid mixture as follows

$$\begin{cases} \frac{\partial \rho}{\partial t}+\nabla\cdot(\rho\mathbf{v})=0,\\ \frac{\partial \rho_i}{\partial t}+\nabla\cdot(\rho_i\mathbf{v})=\nabla\cdot M_{i1}\cdot\nabla\mu_1+\nabla\cdot M_{i2}\cdot\nabla\mu_2, \quad i=1,2,\\ \frac{\partial(\rho\mathbf{v})}{\partial t}+\nabla\cdot(\rho\mathbf{v}\mathbf{v})=2\nabla\cdot(\eta\mathbf{D})+\nabla(\nu\nabla\cdot\mathbf{v})-\sum_{i=1}^{2}\rho_i\nabla\mu_i. \end{cases} \tag{2.25}$$

We note that among the first three equations in the system, only two are independent. So, we could have used ρ, ρ_1 as the fundamental variables in the derivation of the thermodynamic model in lieu of ρ_1 and ρ_2 since $\rho=\rho_1+\rho_2$. With these variables, we reformulate the free energy density function

$$\begin{aligned} f(\rho_1,\rho_2,\nabla\rho_1,\nabla\rho_2)&=f(\rho_1,\rho-\rho_1,\nabla\rho_1,\nabla(\rho-\rho_1))=\tilde{f}(\rho_1,\rho,\nabla\rho_1,\nabla\rho)\\ &=\tilde{h}(\rho_1,\rho,T)+\frac{1}{2}(\tilde{\kappa}_{\rho_1\rho_1}(\nabla\rho_1)^2+2\tilde{\kappa}_{\rho\rho_1}(\nabla\rho,\nabla\rho_1)+\tilde{\kappa}_{\rho\rho}(\nabla\rho)^2), \end{aligned} \tag{2.26}$$

where $\tilde{\kappa}_{\rho_1\rho_1}=\kappa_{\rho_1\rho_1}+\kappa_{\rho_2\rho_2}-2\kappa_{\rho_1\rho_2}, \tilde{\kappa}_{\rho\rho_1}=\kappa_{\rho_1\rho_2}-\kappa_{\rho_2\rho_2}$, and $\tilde{\kappa}_{\rho\rho}=\kappa_{\rho_2\rho_2}$, where $\kappa_{\rho_1\rho_1},\kappa_{\rho_1\rho_2},\kappa_{\rho_2\rho_2}$ are the coefficients of the gradient terms in free energy (2.20). The corresponding chemical potentials are given by

$$\tilde{\mu}_1=\frac{\delta\tilde{f}}{\delta\rho_1}=\frac{\delta f}{\delta\rho_1}+\frac{\delta f}{\delta\rho_2}\frac{\partial\rho_2}{\partial\rho_1}=\mu_1-\mu_2, \qquad \tilde{\mu}=\frac{\delta\tilde{f}}{\delta\rho}=\frac{\delta f}{\delta\rho_2}\frac{\partial\rho_2}{\partial\rho}=\mu_2. \tag{2.27}$$

From these, we have,

$$\mu_1=\tilde{\mu}_1+\tilde{\mu}, \qquad \mu_2=\tilde{\mu}. \tag{2.28}$$

System (2.25) reduces to

$$\begin{cases} \frac{\partial \rho}{\partial t}+\nabla\cdot(\rho\mathbf{v})=0,\\ \frac{\partial \rho_1}{\partial t}+\nabla\cdot(\rho_1\mathbf{v})=\nabla\cdot[M_{11}\cdot\nabla\tilde{\mu}_1+(M_{11}+M_{12})\cdot\nabla\tilde{\mu}],\\ \frac{\partial(\rho\mathbf{v})}{\partial t}+\nabla\cdot(\rho\mathbf{v}\mathbf{v})=2\nabla\cdot(\eta\mathbf{D})+\nabla(\nu\nabla\cdot\mathbf{v})-\rho_1\nabla\tilde{\mu}_1-\rho\nabla\tilde{\mu}. \end{cases} \tag{2.29}$$

If we assign

$$M_{12}=M_{21}=-M_{11}, \quad M_{22}=M_{11}, \tag{2.30}$$

system (2.25) reduces further to a special model

$$\begin{cases} \frac{\partial \rho}{\partial t}+\nabla\cdot(\rho\mathbf{v})=0,\\ \frac{\partial \rho_1}{\partial t}+\nabla\cdot(\rho_1\mathbf{v})=\nabla\cdot M_{11}\cdot\nabla\tilde{\mu}_1,\\ \frac{\partial(\rho\mathbf{v})}{\partial t}+\nabla\cdot(\rho\mathbf{v}\mathbf{v})=2\nabla\cdot(\eta\mathbf{D})+\nabla(\nu\nabla\cdot\mathbf{v})-\rho_1\nabla\tilde{\mu}_1-\rho\nabla\tilde{\mu}. \end{cases} \tag{2.31}$$

This is a special model for compressible binary fluid mixtures among infinitely many choices in the mobility matrix. Apparently, model (2.29) is more general.

The boundary conditions at a solid boundary are given by (2.12) except that the last one is replaced by $\mathbf{n}\cdot\frac{\partial f}{\partial \nabla\rho}=0$ equivalently when ρ is used as a fundamental variable. The energy dissipation rate of the special model reduces to

$$\frac{dE_{total}}{dt}=-\int_V [2\eta\mathbf{D}:\mathbf{D}+\nu tr(\mathbf{D})^2+\tilde{\mu}_1 M_{11}\tilde{\mu}_1]d\mathbf{x}\leq 0, \tag{2.32}$$

provided $\eta,\nu\geq 0$ and $M_{11}>0$. This is a compressible binary fluid model that respects mass and momentum conservation. For the more general model (2.29), the energy dissipation property is warranted so long as the mobility matrix $\mathbf{M}$ is non-negative definite. So, this class of models is thermodynamically consistent.

We next show how this (special) model reduces to another class of compressible models when the two fluid components are incompressible, known as the quasi-incompressible model [27,32]. For the more general compressible model with a local mass conservation law, an analogous result can be obtained.

2.3. Quasi-incompressible model. When the fluid mixture consists of two incompressible viscous fluid components, where the specific densities $\hat{\rho}_1$ and $\hat{\rho}_2$ are constants, we denote the volume fraction of fluid component 1 as ϕ and the other by $1-\phi$. Then, the densities of the two fluids in the mixture are given as follows

$$\rho_1=\phi\hat{\rho}_1,\quad \rho_2=(1-\phi)\hat{\rho}_2. \tag{2.33}$$

The total density of the fluid mixture is given by

$$\rho=\phi\hat{\rho}_1+(1-\phi)\hat{\rho}_2. \tag{2.34}$$

If we use ρ_1 as a fundamental physical variable, ρ is represented by ρ_1 as follows,

$$\rho=\rho_1+(1-\frac{\rho_1}{\hat{\rho}_1})\hat{\rho}_2=\hat{\rho}_2+(1-\frac{\hat{\rho}_2}{\hat{\rho}_1})\rho_1. \tag{2.35}$$

This means that the two variables ρ and ρ_1 are related linearly in this fluid mixture system. This mixture is known as the simple mixture [25]. We view this as a special case of the fully compressible model subject to a constraint given by (2.35). To accommodate the constraint, we augment the free energy density by $\pi(\hat{\rho}_2+(1-\frac{\hat{\rho}_2}{\hat{\rho}_1})\rho_1-\rho)$, where π is a Lagrange multiplier. We denote the modified free energy density function as $\hat{f}$,

$$\hat{f}=\tilde{f}(\rho_1,\nabla\rho_1,\rho,\nabla\rho)+\pi[\hat{\rho}_2+(1-\frac{\hat{\rho}_2}{\hat{\rho}_1})\rho_1-\rho]. \tag{2.36}$$

The corresponding chemical potentials and their relations to the chemical potentials in the compressible model are given as follows

$$\hat{\mu}_1=\frac{\delta\hat{f}}{\delta\rho_1}=\tilde{\mu}_1+\pi(1-\frac{\hat{\rho}_2}{\hat{\rho}_1}),\quad \hat{\mu}=\frac{\delta\hat{f}}{\delta\rho}=\tilde{\mu}-\pi,\quad \tilde{\mu}_1=\frac{\delta\tilde{f}}{\delta\rho_1}|_\rho,$$

$$\tilde{\mu}=\frac{\delta\tilde{f}}{\delta\rho}|_{\rho_1},\quad \mu_\phi=\frac{\delta\hat{f}}{\delta\rho_1}|_\rho\frac{\delta\rho_1}{\delta\phi}+\frac{\delta\hat{f}}{\delta\rho}|_{\rho_1}\frac{\delta\rho}{\delta\phi}=\hat{\rho}_1\hat{\mu}_1+(\hat{\rho}_1-\hat{\rho}_2)\hat{\mu}. \tag{2.37}$$

From the mass conservation of the mixture system (2.31)-1, we have

$$(\hat{\rho}_1-\hat{\rho}_2)[\frac{\partial\phi}{\partial t}+\nabla\cdot(\phi\mathbf{v})]+\hat{\rho}_2\nabla\cdot\mathbf{v}=0. \tag{2.38}$$

The transport equation of ρ_1 is rewritten in terms of the volume fraction as follows

$$\frac{\partial\phi}{\partial t}+\nabla\cdot(\phi\mathbf{v})=\frac{1}{\hat{\rho}_1}(\nabla\cdot M_{11}\cdot\nabla)(\hat{\mu}_1). \tag{2.39}$$

The linear momentum conservation equation is rewritten into

$$\rho(\frac{\partial\mathbf{v}}{\partial t}+\mathbf{v}\cdot\nabla\mathbf{v})=\nabla\cdot(2\eta\mathbf{D})+\nabla(\nu\nabla\cdot\mathbf{v})-\nabla\Pi-\phi\nabla\mu_\phi, \tag{2.40}$$

where η, ν are volume-averaged viscosity coefficients and the hydrostatic pressure is defined by

$$\Pi=\hat{\rho}_2(\tilde{\mu}-\pi). \tag{2.41}$$

With this definition, the transport Equation (2.39) for ϕ is written into

$$\frac{\partial\phi}{\partial t}+\nabla\cdot(\phi\mathbf{v})=\frac{1}{\hat{\rho}_1^2}(\nabla\cdot M_{11}\cdot\nabla)(\mu_\phi+\Pi(1-\frac{\hat{\rho}_1}{\hat{\rho}_2})). \tag{2.42}$$

Combining mass conservation law (2.38) and transport Equation (2.42) for ϕ, we obtain

$$\nabla\cdot\mathbf{v}=(1-\frac{\hat{\rho}_1}{\hat{\rho}_2})\frac{1}{\hat{\rho}_1^2}(\nabla\cdot M_{11}\cdot\nabla)(\mu_\phi+\Pi(1-\frac{\hat{\rho}_1}{\hat{\rho}_2})). \tag{2.43}$$

We summarize the governing equations of the quasi-incompressible model as follows

$$\begin{cases}\nabla\cdot\mathbf{v}=(1-\frac{\hat{\rho}_1}{\hat{\rho}_2})\frac{1}{\hat{\rho}_1^2}(\nabla\cdot M_{11}\cdot\nabla)(\mu_\phi+\Pi(1-\frac{\hat{\rho}_1}{\hat{\rho}_2})),\\ \frac{\partial\phi}{\partial t}+\nabla\cdot(\phi\mathbf{v})=\frac{1}{\hat{\rho}_1^2}(\nabla\cdot M_{11}\cdot\nabla)(\mu_\phi+\Pi(1-\frac{\hat{\rho}_1}{\hat{\rho}_2})),\\ \rho[\frac{\partial\mathbf{v}}{\partial t}+\mathbf{v}\cdot\nabla\mathbf{v}]=\nabla\cdot(2\eta\mathbf{D})+\nabla(\nu\nabla\cdot\mathbf{v})-\nabla\Pi-\phi\nabla\mu_\phi.\end{cases} \tag{2.44}$$

The free energy density reduces to

$$\tilde{f}(\rho_1,\rho,\nabla\rho_1,\nabla\rho)=\hat{h}(\phi)+\frac{1}{2}\hat{\kappa}_{\phi\phi}\|\nabla\phi\|^2, \tag{2.45}$$

where $\hat{h}(\phi)=\tilde{h}(\hat{\rho}_1\phi,(\hat{\rho}_1-\hat{\rho}_2)\phi+\hat{\rho}_2,T)$, $\hat{\kappa}_{\phi\phi}=\tilde{\kappa}_{\rho_1\rho_1}\hat{\rho}_1{}^2+2\tilde{\kappa}_{\rho_1\rho}\hat{\rho}_1(\hat{\rho}_1-\hat{\rho}_2)+\tilde{\kappa}_{\rho\rho}(\hat{\rho}_1-\hat{\rho}_2)^2$. This is the equation system for quasi-incompressible binary fluids obtained in [27]. The upshot of the derivation shows that we can obtain the constrained theory from the unconstrained theory by augmenting the free energy with the algebraic constraint via a Lagrange multiplier. When the boundary conditions annihilate the energy dissipation through the boundary, the energy dissipation rate of the binary quasi-incompressible fluid flow (2.44) is given by

$$\frac{dE_{total}}{dt}=-\int_V[2\eta\mathbf{D}:\mathbf{D}+\nu tr(\mathbf{D})^2+\nabla\hat{\mu}_1\cdot M_{11}\cdot\nabla\hat{\mu}_1]d\mathbf{x}\le 0, \tag{2.46}$$

provided $\eta,\nu\geq 0$, $M_{11}>0$, where $\hat{\mu}_1=\frac{1}{\hat{\rho}_1}(\mu_\phi+\Pi(1-\frac{\hat{\rho}_1}{\hat{\rho}_2}))$. When, in addition $\hat{\rho}_1=\hat{\rho}_2=\rho$, the system reduces to an incompressible model

$$\begin{cases}\nabla\cdot\mathbf{v}=0,\\ \frac{\partial\phi}{\partial t}+\nabla\cdot(\phi\mathbf{v})=\frac{1}{\rho^2}(\nabla\cdot M_{11}\cdot\nabla)\mu_\phi,\\ \rho[\frac{\partial\mathbf{v}}{\partial t}+\mathbf{v}\cdot\nabla\mathbf{v}]=\nabla\cdot(2\eta\mathbf{D})+\nabla(\nu\nabla\cdot\mathbf{v})-\nabla\Pi-\phi\nabla\mu_\phi.\end{cases}\tag{2.47}$$

This is the incompressible model derived by Halperin et al. [24].

These derivations can be readily extended to account for multi-component fluid systems.

3. Hydrodynamic phase field models for N-component multiphase compressible fluid flows

When fluid mixtures are composed of N fluid components, we use $\rho_i, i=1,2,\cdots,N$ to denote the mass density of the ith component and assume the free energy of the fluid mixture is given by

$$F=\int_\Omega f(\rho_1,\nabla\rho_1,\cdots,\rho_N,\nabla\rho_N)d\mathbf{x},\tag{3.1}$$

where f is the free energy density. The derivation of the hydrodynamic phase field models follows the procedures alluded to in the previous section. We present the results next.

3.1. Compressible model with the global mass conservation law. We choose $\rho_1,\cdots,\rho_N$ as the primitive variables. Following the procedure outlined in the previous section, we obtain the governing system of equations for the N-component multi-phase viscous fluid mixture as follows

$$\begin{cases}\frac{\partial\rho_i}{\partial t}+\nabla\cdot(\rho_i\mathbf{v})=j_i=\sum_{j=1}^N\nabla\cdot M_{ij}\cdot\nabla\mu_j,\quad i=1,2,\cdots,N,\\ \frac{\partial(\rho\mathbf{v})}{\partial t}+\nabla\cdot(\rho\mathbf{v}\mathbf{v})-\frac{1}{2}(\sum_{i=1}^N j_i)\mathbf{v}=2\nabla\cdot(\eta\mathbf{D})+\nabla(\nu\nabla\cdot\mathbf{v})-\sum_{i=1}^N\rho_i\nabla\mu_i,\end{cases}\tag{3.2}$$

where M_{ij}, i, j = 1, ..., N, are the mobility coefficients, and $\eta=\sum_{i=1}^N\eta_i\frac{\rho_i}{\rho},\nu=\sum_{i=1}^N\nu_i\frac{\rho_i}{\rho}$ are mass-average viscosities, respectively. The bulk energy dissipation rate is given by

$$\begin{aligned}\frac{dE_{total}}{dt}=&-\int_V[2\eta\mathbf{D}:\mathbf{D}+\nu tr(\mathbf{D})^2\\ &+(\nabla\mu_1,\nabla\mu_2,\cdots,\nabla\mu_N)\cdot\mathbf{M}\cdot(\nabla\mu_1,\nabla\mu_2,\cdots,\nabla\mu_N)]d\mathbf{x}\leq 0,\end{aligned}\tag{3.3}$$

provided $\eta,\nu\geq 0$, $\mathbf{M}=(M_{ij})_{ij=1}^N$ is a symmetric non-negative definite mobility coefficient matrix. The boundary conditions are chosen to annihilate the surface energy dissipation rate.

3.2. Compressible model with the local mass conservation law. If $\sum_{i=1}^N j_i=0$, the total mass of the system is conserved locally, i.e.,

$$\frac{\partial\rho}{\partial t}+\nabla\cdot(\rho\mathbf{v})=0.\tag{3.4}$$

We obtain the governing system of equations as follows

$$\begin{cases} \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0, \quad \text{or} \quad \sum_{i=1}^N \sum_{j=1}^N \nabla \cdot M_{ij} \cdot \nabla \mu_j = 0, \\ \frac{\partial \rho_i}{\partial t} + \nabla \cdot (\rho_i \mathbf{v}) = \sum_{j=1}^N \nabla \cdot M_{ij} \cdot \nabla \mu_j, \quad i = 1, 2, \cdots, N, \\ \frac{\partial (\rho \mathbf{v})}{\partial t} + \nabla \cdot (\rho \mathbf{v} \mathbf{v}) = 2 \nabla \cdot (\eta \mathbf{D}) + \nabla (\nu \nabla \cdot \mathbf{v}) - \sum_{i=1}^N \rho_i \nabla \mu_i, \end{cases} \tag{3.5}$$

where η, ν are mass-averaged shear and volumetric viscosities, $\mathbf{v}$ is the mass average velocity and $\mathbf{M} = (M_{ij})_{i,j=1}^N$ is the symmetric mobility coefficient matrix. In this case, the bulk energy dissipation rate is given by

$$\begin{aligned} \frac{dE_{total}}{dt} = -\int_V & [2\eta \mathbf{D} : \mathbf{D} + \nu tr(\mathbf{D})^2 \\ & + (\nabla \mu_1, \nabla \mu_2, \cdots, \nabla \mu_N) \cdot \mathbf{M} \cdot (\nabla \mu_1, \nabla \mu_2, \cdots, \nabla \mu_N)] d\mathbf{x} \leq 0, \end{aligned} \tag{3.6}$$

provided $\eta, \nu \geq 0$ and $\mathbf{M}$ is a symmetric non-negative definite mobility coefficient matrix subject to the constraint $\sum_{i=1}^N \sum_{j=1}^N \nabla \cdot M_{ij} \cdot \nabla \mu_j = 0$.

Analogously, we choose $\rho_1, \cdots, \rho_{N-1}, \rho$ as the primitive variables, where $\rho = \sum_{i=1}^N \rho_i$. Then, we represent $\rho_N = \rho - \sum_{i=1}^{N-1} \rho_i$. The free energy density is written as

$$\begin{aligned} f(\rho_1, \nabla \rho_1, \cdots, \rho_N, \nabla \rho_N) &= f(\rho_1, \nabla \rho_1, \cdots, \rho - \sum_{i=1}^{N-1} \rho_i, \nabla (\rho - \sum_{i=1}^{N-1} \rho_i)) \\ &= \tilde{f}(\rho_1, \nabla \rho_1, \cdots, \rho_{N-1}, \nabla \rho), \end{aligned} \tag{3.7}$$

The corresponding chemical potentials are given by

$$\begin{aligned} \tilde{\mu}_i &= \frac{\delta \tilde{f}}{\delta \rho_i} = \frac{\delta f}{\delta \rho_i} + \frac{\delta f}{\delta \rho_N} \frac{\delta \rho_N}{\delta \rho_i} = \mu_i - \mu_N, \quad i = 1, \cdots, N-1, \\ \tilde{\mu} &= \frac{\delta \tilde{f}}{\delta \rho} = \frac{\delta f}{\delta \rho_N} \frac{\delta \rho_N}{\delta \rho} = \mu_N. \\ \mu_i &= \tilde{\mu}_i + \tilde{\mu}, \mu_N = \tilde{\mu}. \end{aligned} \tag{3.8}$$

The transport equations of the densities are given by

$$\frac{\partial \rho_i}{\partial t} + \nabla \cdot (\rho_i \mathbf{v}) = \sum_{j=1}^{N-1} \nabla \cdot M_{ij} \cdot \nabla \tilde{\mu}_j + (\sum_{j=1}^N \nabla \cdot M_{ij}) \tilde{\mu}, \quad i = 1, 2, \cdots, N-1. \tag{3.9}$$

The mass conservation equation implies

$$\sum_{i=1}^N \sum_{j=1}^{N-1} \nabla \cdot M_{ij} \cdot \nabla \tilde{\mu}_j + (\sum_{i,j=1}^N \nabla \cdot M_{ij} \cdot \nabla) \tilde{\mu} = 0. \tag{3.10}$$

The mobility coefficients must satisfy the above constraint. If we assign

$$M_{iN} = -\sum_{j=1}^{N-1} M_{ij} = M_{Ni}, \quad M_{NN} = -\sum_{i=1}^{N-1} M_{iN} = \sum_{i=1}^{N-1} \sum_{j=1}^{N-1} M_{ij}, \tag{3.11}$$

the constraint is satisfied and system (3.2) reduces to a special model

$$\begin{cases} \frac{\partial \rho}{\partial t} + \nabla\cdot(\rho \mathbf{v}) = 0, \\ \frac{\partial \rho_i}{\partial t} + \nabla\cdot(\rho_i \mathbf{v}) = \sum_{j=1}^{N-1} \nabla\cdot M_{ij}\cdot\nabla\tilde{\mu}_j, \quad i=1,2,\cdots,N-1, \\ \frac{\partial(\rho\mathbf{v})}{\partial t} + \nabla\cdot(\rho\mathbf{v}\mathbf{v}) = 2\nabla\cdot(\eta\mathbf{D}) + \nabla(\nu\nabla\cdot\mathbf{v}) - \sum_{i=1}^{N-1}\rho_i\nabla\tilde{\mu}_i - \rho\nabla\tilde{\mu}. \end{cases} \tag{3.12}$$

This is a special model for compressible fluid mixtures of N-components. The bulk energy dissipation rate is given by

$$\frac{dE_{total}}{dt} = -\int_V [2\eta\mathbf{D}:\mathbf{D} + \nu tr(\mathbf{D})^2 + (\nabla\tilde{\mu}_1, \nabla\tilde{\mu}_2, \cdots, \nabla\tilde{\mu}_{N-1})\cdot\mathbf{M}\cdot(\nabla\tilde{\mu}_1, \nabla\tilde{\mu}_2, \cdots, \nabla\tilde{\mu}_{N-1})]d\mathbf{x} \le 0, \tag{3.13}$$

provided $\eta, \nu \ge 0$ and $\mathbf{M} = (M_{ij})_{i,j=1}^{N-1}$ is a symmetric non-negative definite mobility coefficient matrix.

3.3. Quasi-incompressible model. When each of the fluid component is incompressible in the viscous fluid mixture, we denote the volume fraction of the ith component as ϕ_i and specific density as $\hat{\rho}_i$ for $i = 1, \cdots, N$, respectively. Then, $\sum_{i=1}^N \phi_i = 1$ and the total mass density in the mixture is given by

$$\rho = \sum_{i=1}^{N} \phi_i\hat{\rho}_i = \sum_{i=1}^{N-1}\phi_i\hat{\rho}_i + (1 - \sum_{i=1}^{N-1}\phi_i)\hat{\rho_N} = \sum_{i=1}^{N-1}\rho_i + (1 - \sum_{i=1}^{N-1}\frac{\rho_i}{\hat{\rho}_i})\hat{\rho_N}. \tag{3.14}$$

We assume the volume fraction of the Nth component is nonzero. Then, the free energy density is a functional of the first $N-1$ volume fractions $(\phi_1, \cdots, \phi_{N-1})$. If we augment the free energy by $\pi(\sum_{i=1}^{N-1}\rho_i + (1 - \sum_{i=1}^{N-1}\frac{\rho_i}{\hat{\rho}_i})\hat{\rho_N} - \rho)$, where π is a Lagrange multiplier, then, the modified free energy density function is given by

$$\hat{f} = \tilde{f}(\rho_1, \nabla\rho_1, ..., \rho_{N-1}, \nabla\rho_{N-1}, \rho, \nabla\rho) + \pi[\sum_{i=1}^{N-1}\rho_i + (1 - \sum_{i=1}^{N-1}\frac{\rho_i}{\hat{\rho}_i})\hat{\rho_N} - \rho]. \tag{3.15}$$

Following the procedure alluded to in the previous section, we derive the following governing system of equations of the quasi-incompressible fluid from the special compressible model as follows

$$\begin{cases} \nabla\cdot\mathbf{v} = \sum_{i=1}^{N-1}\sum_{j=1}^{N-1}(1 - \frac{\hat{\rho}_j}{\hat{\rho}_N})\frac{1}{\hat{\rho}_i\hat{\rho}_j}(\nabla\cdot M_{ij}\cdot\nabla)(\mu_{\phi_j} + \Pi(1 - \frac{\hat{\rho}_j}{\hat{\rho}_N})), \\ \frac{\partial\phi_i}{\partial t} + \nabla\cdot(\phi_i\mathbf{v}) = \sum_{j=1}^{N-1}\frac{1}{\hat{\rho}_i\hat{\rho}_j}(\nabla\cdot M_{ij}\cdot\nabla)(\mu_{\phi_j} + \Pi(1 - \frac{\hat{\rho}_j}{\hat{\rho}_N})), i = 1, 2, \cdots, N-1, \\ \rho[\frac{\partial\mathbf{v}}{\partial t} + \mathbf{v}\cdot\nabla\mathbf{v}] = \nabla\cdot(2\eta\mathbf{D}) + \nabla(\nu\nabla\cdot\mathbf{v}) - \nabla\Pi - \sum_{i=1}^{N-1}\phi_i\nabla\mu_{\phi_i}, \end{cases} \tag{3.16}$$

where

$$\tilde{\mu}_i = \frac{\delta\tilde{f}}{\delta\rho_i}, \quad \tilde{\mu} = \frac{\delta\tilde{f}}{\delta\rho}, \tag{3.17}$$

$$\hat{\mu}_i = \tilde{\mu}_i + \pi(1 - \frac{\hat{\rho}_N}{\hat{\rho}_i}), \quad i = 1, \cdots, N-1, \quad \hat{\mu} = \tilde{\mu} - \pi, \tag{3.18}$$

$$\mu_{\phi_i} = \frac{\delta\hat{f}}{\delta\phi_i} = \frac{\delta\hat{f}}{\delta\rho_i}|_\rho\frac{\delta\rho_i}{\delta\phi_i} + \frac{\delta\hat{f}}{\delta\rho}|_{\rho_i}\frac{\delta\rho}{\delta\phi_i} = \hat{\rho}_i\hat{\mu}_i + (\hat{\rho}_i - \hat{\rho}_N)\hat{\mu}, \tag{3.19}$$

$$\Pi = -\hat{\rho}_N \pi + \hat{\rho}_N \mu, \tag{3.20}$$

and Π serves as the hydrostatic pressure.

The bulk energy dissipation rate is

$$\frac{dE_{total}}{dt} = -\int_V [2\eta \mathbf{D}:\mathbf{D} + \nu tr(\mathbf{D})^2 \\ + (\nabla\hat{\mu}_1, \nabla\hat{\mu}_2, \cdots, \nabla\hat{\mu}_{N-1}) \cdot \mathbf{M} \cdot (\nabla\hat{\mu}_1, \nabla\hat{\mu}_2, \cdots, \nabla\hat{\mu}_{N-1})] d\mathbf{x} \leq 0, \tag{3.21}$$

provided $\eta, \nu \geq 0$, $\mathbf{M} = (M)_{i,j=1}^{N-1}$ is a symmetric non-negative definite matrix, where $\hat{\mu}_i = \frac{1}{\hat{\rho}_i}[\mu_{\phi_j} + \Pi(1 - \frac{\hat{\rho}_j}{\hat{\rho}_N})]$. A more general model can be derived from the general compressible model by enforcing the incompressibility constraint. But, we will not present it here.

For a fluid mixture with $\hat{\rho}_i = \rho$, the system reduces to an incompressible model

$$\begin{cases} \nabla \cdot \mathbf{v} = 0, \\ \frac{\partial \phi_i}{\partial t} + \nabla \cdot (\phi_i \mathbf{v}) = \sum_{j=1}^{N-1} \frac{1}{\rho^2} (\nabla \cdot M_{ij} \cdot \nabla) \mu_{\phi_j}, \quad i = 1, 2, \cdots, N-1, \\ \rho[\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v}] = \nabla \cdot (2\eta \mathbf{D}) + \nabla(\nu \nabla \cdot \mathbf{v}) - \nabla \Pi - \sum_{i=1}^{N-1} \phi_i \nabla \mu_{\phi_i}. \end{cases} \tag{3.22}$$

3.4. Alternative derivation of the quasi-incompressible model. For phase field models of N components where $N \geq 2$, there exists a second way to derive the quasi-incompressible phase field model. We begin with a fully compressible model of $N+1$ components, each of which is of density $\rho_i, i = 1, \cdots, N$ and ρ. We assume the free energy density depends on $(\rho_1, \cdots, \rho_N, \rho)$. The second approach to derive the quasi-incompressible model is to augment the free energy by $\pi(\sum_{i=1}^N \rho_i - \rho) + B(\sum_{i=1}^N \frac{\rho_i}{\hat{\rho}_i} - 1)$, where π and B are two Lagrange multipliers. We define the modified free energy density function by

$$\hat{f} = f(\rho_1, \nabla\rho_1, \cdots, \rho_N, \nabla\rho_N) + \pi(\sum_{i=1}^N \rho_i - \rho) + B(\sum_{i=1}^N \frac{\rho_i}{\hat{\rho}_i} - 1). \tag{3.23}$$

The chemical potentials are given by

$$\hat{\mu}_i = \frac{\delta \hat{f}}{\delta \rho_i} = \frac{\delta f}{\delta \rho_i} + \frac{1}{\hat{\rho}_i} B + \pi = \mu_i + \frac{1}{\hat{\rho}_i} B + \pi, \quad i = 1, \cdots, N, \\ \hat{\mu} = \frac{\delta \hat{f}}{\delta \rho} = -\pi. \tag{3.24}$$

The governing system of equations with $N+1$ components subject to the two constraints is given by

$$\begin{cases} \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0, \\ \frac{\partial \rho_i}{\partial t} + \nabla \cdot (\rho_i \mathbf{v}) = \sum_{j=1}^N \nabla \cdot M_{ij} \cdot \nabla \hat{\mu}_j, \quad i = 1, 2, \cdots, N, \\ \frac{\partial (\rho \mathbf{v})}{\partial t} + \nabla \cdot (\rho \mathbf{v}\mathbf{v}) = 2\nabla \cdot (\eta \mathbf{D}) + \nabla(\nu \nabla \cdot \mathbf{v}) - \sum_{i=1}^N \rho_i \nabla \hat{\mu}_i - \rho \nabla \hat{\mu}, \end{cases} \tag{3.25}$$

where $\mathbf{M}$ is the symmetric mobility matrix, which satisfies $\sum_{i,j=1}^N \nabla \cdot M_{ij} \cdot \nabla \hat{\mu}_j = 0$. This is a more general quasi-incompressible model.

In fact, if we assign $M_{Ni}=M_{iN}=-\sum_{j=1}^{N-1}M_{ij}$ and apply the constraints $\sum_{i=1}^{N}\rho_i=\rho$, $\sum_{i=1}^{N}\phi_i=1$ and $\rho_i=\phi_i\hat{\rho}_i$, we obtain the chemical potential with respect to ϕ_i, i=1,2, $\cdots$, N-1, in the quasi-incompressible limit,

$$\mu_{\phi_i}=\frac{\delta\hat{f}}{\delta\phi_i}=\frac{\delta\hat{f}}{\delta\rho_i}\frac{\partial\rho_i}{\partial\phi_i}+\frac{\delta\hat{f}}{\delta\rho}\frac{\partial\rho}{\partial\phi_i}+\frac{\delta\hat{f}}{\delta\rho_N}\frac{\partial\rho_N}{\partial\phi_i}=\hat{\mu_i}\hat{\rho_i}+\hat{\mu}(\hat{\rho_i}-\hat{\rho_N})-\hat{\rho_N}\hat{\mu_N},i=1,\cdots,N. \tag{3.26}$$

If we define

$$\Pi=\hat{\rho_N}(\hat{\mu}+\hat{\mu}_N)=\hat{\rho_N}\mu_N+B, \tag{3.27}$$

(3.4) reduces to

$$\begin{cases}\nabla\cdot\mathbf{v}=\sum_{i=1}^{N-1}\sum_{j=1}^{N-1}(1-\frac{\hat{\rho}_j}{\hat{\rho}_N})\frac{1}{\hat{\rho}_i\hat{\rho}_j}(\nabla\cdot M_{ij}\cdot\nabla)(\mu_{\phi_j}+\Pi(1-\frac{\hat{\rho}_j}{\hat{\rho}_N})),\\ \frac{\partial\phi_i}{\partial t}+\nabla\cdot(\phi_i\mathbf{v})=\sum_{j=1}^{N-1}\frac{1}{\hat{\rho}_i\hat{\rho}_j}(\nabla\cdot M_{ij}\cdot\nabla)(\mu_{\phi_j}+\Pi(1-\frac{\hat{\rho}_j}{\hat{\rho}_N})),i=1,2,\cdots,N-1,\\ \rho[\frac{\partial\mathbf{v}}{\partial t}+\mathbf{v}\cdot\nabla\mathbf{v}]=\nabla\cdot(2\eta\mathbf{D})+\nabla(\nu\nabla\cdot\mathbf{v})-\nabla\Pi-\sum_{i=1}^{N-1}\phi_i\nabla\mu_{\phi_i},\end{cases} \tag{3.28}$$

which is exactly the quasi-incompressible model given in (3.3).

4. Non-dimensionalization

Next, we non-dimensionalize the binary model equations.

4.1. Compressible model with the global mass conservation law.

In model (2.16), selecting characteristic time scale t_0, characteristic length scale l_0, and characteristic density scale ρ_0, we nondimensionalize the variables and parameters as follows

$$\tilde{t}=\frac{t}{t_0},\quad \tilde{x}=\frac{x}{l_0},\quad \tilde{\rho}_1=\frac{\rho_1}{\rho_0},\quad \tilde{\rho}_2=\frac{\rho_2}{\rho_0},\quad \tilde{\mathbf{v}}=\frac{\mathbf{v}t_0}{l_0},\quad \tilde{M_{ij}}=\frac{M_{ij}}{t_0\rho_0},\quad i,j=1,2, \tag{4.1}$$
$$\frac{1}{Re_s}=\tilde{\eta}=\frac{t_0}{\rho_0 l_0^2}\eta,\quad \frac{1}{Re_v}=\tilde{\nu}=\frac{t_0}{\rho_0 l_0^2}\nu,\quad \tilde{\mu}_1=\frac{t_0^2}{l_0^2}\mu_1,\quad \tilde{\mu}_2=\frac{t_0^2}{l_0^2}\mu_2,\quad J_i=\frac{j_i t_0}{\rho_0},i=1,2,$$

where Re_s, Re_v are the Reynolds number corresponding to the shear and volumetric stresses. The scaling of chemical potentials μ_1, μ results from the non-dimensionalization of the total energy. We summarize the governing equation with non-dimensional variables and parameters as follows, dropping the ˜ for simplicity,

$$\begin{cases}\frac{\partial\rho_1}{\partial t}+\nabla\cdot(\rho_1\mathbf{v})=J_1=\nabla\cdot M_{11}\cdot\nabla\mu_1+\nabla\cdot M_{12}\cdot\nabla\mu_2,\\ \frac{\partial\rho_2}{\partial t}+\nabla\cdot(\rho_2\mathbf{v})=J_2=\nabla\cdot M_{12}\cdot\nabla\mu_1+\nabla\cdot M_{22}\cdot\nabla\mu_2,\\ \frac{\partial(\rho\mathbf{v})}{\partial t}+\nabla\cdot(\rho\mathbf{v}\mathbf{v})-\frac{1}{2}(J_1+J_2)\mathbf{v}=2\nabla\cdot(\frac{1}{Re_s}\mathbf{D})+\nabla(\frac{1}{Re_v}\nabla\cdot\mathbf{v})-\rho_1\nabla\mu_1-\rho_2\nabla\mu_2.\end{cases} \tag{4.2}$$

4.2. Compressible model with the local mass conservation law.

Analogously, in model (2.31), we nondimensionalize the variables and parameters as above and in particular

$$\tilde{M}_{11}=\frac{M_{11}}{t_0\rho_0}. \tag{4.3}$$

We summarize the governing equation with non-dimensional variables and parameters as follows, dropping the ˜ for simplicity,

$$\begin{cases} \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0, \\ \frac{\partial \rho_1}{\partial t} + \nabla \cdot (\rho_1 \mathbf{v}) = \nabla \cdot M_{11} \cdot \nabla \tilde{\mu}_1, \\ \frac{\partial (\rho \mathbf{v})}{\partial t} + \nabla \cdot (\rho \mathbf{v}\mathbf{v}) = 2\nabla \cdot (\frac{1}{Re_s}\mathbf{D}) + \nabla(\frac{1}{Re_v}\nabla \cdot \mathbf{v}) - \rho_1 \nabla \tilde{\mu}_1 - \rho \nabla \tilde{\mu}. \end{cases} \tag{4.4}$$

4.3. Quasi-incompressible model. In model (2.44), in addition to the above, we nondimensionalize two new ones as follows:

$$\tilde{\mu}_\phi = \frac{t_0^2}{\rho_0 l_0^2}\mu_\phi, \quad \tilde{\Pi} = \Pi \frac{t_0^2}{\rho_0 l_0^2}. \tag{4.5}$$

Dropping the ˜ on the non-dimensionalized variables and parameters, the governing equation system of the quasi-incompressible fluid flows is written as follows,

$$\begin{cases} \nabla \cdot \mathbf{v} = (1 - \frac{\hat{\rho}_1}{\hat{\rho}_2})\frac{1}{\hat{\rho}_1^2}(\nabla \cdot M_{11} \cdot \nabla)(\mu_\phi + \Pi(1 - \frac{\hat{\rho}_1}{\hat{\rho}_2})), \\ \frac{\partial \phi}{\partial t} + \nabla \cdot (\phi \mathbf{v}) = \frac{1}{\hat{\rho}_1^2}(\nabla \cdot M_{11} \cdot \nabla)(\mu_\phi + \Pi(1 - \frac{\hat{\rho}_1}{\hat{\rho}_2})), \\ \rho[\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v}] = 2\nabla \cdot (\frac{1}{Re_s}\mathbf{D}) + \nabla(\frac{1}{Re_v}\nabla \cdot \mathbf{v}) - \nabla \Pi - \phi \nabla \mu_\phi. \end{cases} \tag{4.6}$$

5. Near equilibrium dynamics

We investigate near equilibrium dynamics of the models derived previously by conducting a linear stability analysis of the models from each class about a constant equilibrium state. Through analyzing dispersion relations of the selected models, we identify the intrinsic relation among compressible, quasi-incompressible and incompressible models, focusing on the consequence of the hierarchical model reduction. We show the result on models of binary fluid mixtures in this study.

5.1. Linear stability analysis of the compressible model with the global mass conservation law. This compressible model admits constant solution:

$$\mathbf{v} = \mathbf{0}, \quad \rho_1 = \rho_1^0, \quad \rho_2 = \rho_2^0, \tag{5.1}$$

where ρ_1^0, ρ_2^0 are constants. We perturb the solution with the normal mode as follows:

$$\mathbf{v} = \epsilon e^{\alpha t + i\mathbf{k}\cdot\mathbf{x}}\mathbf{v}^c, \quad \rho_1 = \rho_1^0 + \epsilon e^{\alpha t + i\mathbf{k}\cdot\mathbf{x}}{\rho_1}^c, \quad \rho_2 = \rho_2^0 + \epsilon e^{\alpha t + i\mathbf{k}\cdot\mathbf{x}}{\rho_2}^c. \tag{5.2}$$

where ϵ is a small parameter, representing the magnitude of the perturbation, and $\mathbf{v}^c, \rho_1^c, \rho_2^c$ are constants, α is the growth rate, $\mathbf{k}$ is the wave number of the perturbation. Without loss of generality, we limit our study to one-dimensional perturbations in $\mathbf{k}$ in 2D space. Substituting these perturbations into the equations in (4.1) and truncating the equations at order $O(\epsilon)$, we obtain the linearized equations. The dispersion equation of the linearized equation system is given by the algebraic equation of α:

$$\begin{aligned} &(\frac{1}{Re_s}k^2 + \alpha\rho^0)\{\alpha^3\rho_0 + \alpha^2 k^2[\frac{1}{Re} + \rho^0 M_{11}(h_{\rho_1\rho_1} + \kappa_{\rho_1\rho_1}k^2) + \rho^0 M_{22}(h_{\rho_2\rho_2} + \kappa_{\rho_2\rho_2}k^2)] \\ &+ \alpha^2 k^2[2\rho^0 M_{12}(h_{\rho_1\rho_2} + \kappa_{\rho_1\rho_2}k^2)] + \alpha[\mathbf{p}^T \cdot \mathbf{C} \cdot \mathbf{p} + \mathbf{p}^T \cdot \mathbf{K} \cdot \mathbf{p}k^2]k^2 \\ &+ \alpha\frac{1}{Re}[M_{11}(h_{\rho_1\rho_1} + \kappa_{\rho_1\rho_1}k^2) + M_{22}(h_{\rho_2\rho_2} + \kappa_{\rho_2\rho_2}k^2) + 2M_{12}(h_{\rho_1\rho_2} + \kappa_{\rho_1\rho_2}k^2)]k^4 \end{aligned}$$

$$+\alpha\rho^0|\mathbf{M}|[(h_{\rho_1\rho_1}+\kappa_{\rho_1\rho_1}k^2)(h_{\rho_2\rho_2}+\kappa_{\rho_2\rho_2}k^2)-(h_{\rho_1\rho_2}+\kappa_{\rho_1\rho_2}k^2)^2]k^4$$
$$+k^4(\frac{1}{Re}|\mathbf{M}|k^2+M_{22}(\rho_1^0)^2+M_{11}(\rho_2^0)^2-2M_{12}\rho_1^0\rho_2^0).$$
$$[(h_{\rho_1\rho_1}+\kappa_{\rho_1\rho_1}k^2)(h_{\rho_2\rho_2}+\kappa_{\rho_2\rho_2}k^2)-(h_{\rho_1\rho_2}+\kappa_{\rho_1\rho_2}k^2)^2]\}=0. \tag{5.3}$$

where $\mathbf{p}=(\rho_1^0,\rho_2^0)^T$ and $\frac{1}{Re}=2\frac{1}{Re_s}+\frac{1}{Re_v}$. $|\mathbf{M}|$ is the determinant of the mobility coefficient matrix $\mathbf{M}=(M_{i,j})$, $\mathbf{K}$ is the coefficient matrix of the conformational entropy

$$\mathbf{K}=\begin{pmatrix}\kappa_{\rho_1\rho_1} & \kappa_{\rho_1\rho_2}\\ \kappa_{\rho_1\rho_2} & \kappa_{\rho_2\rho_2}\end{pmatrix}, \tag{5.4}$$

$\mathbf{C}$ is the Hessian of the bulk free energy $h(\rho_1,\rho_2,T)$ in (2.20) with respect to ρ_1 and ρ_2,

$$\mathbf{C}=\begin{pmatrix}h_{\rho_1\rho_1} & h_{\rho_1\rho_2}\\ h_{\rho_1\rho_2} & h_{\rho_2\rho_2}\end{pmatrix}, \tag{5.5}$$

$h_{\rho_i\rho_j}$ represents the second-order derivative of the bulk free energy density $h(\rho_1,\rho_2,T)$ with respect to ρ_i and ρ_j, i, j = 1, 2.

One root of Equation (5.3) is given by

$$\alpha_0=-\frac{1}{\rho^0 Re_s}k^2. \tag{5.6}$$

This is the viscous mode associated to the viscous stress. The other three roots are governed by a cubic polynomial equation and their closed forms are essentially impenetrable. Instead, we present them using asymptotic formulae in long and short wave range and numerical calculations in the intermediate wave range, respectively.

The asymptotic expressions of the three growth rates at $|k|\ll 1$ are given by

$$\alpha_1=x_1k^2+y_1k^4+O(k^5),\quad \alpha_{2,3}=x_{2,3}k+y_{2,3}k^2+O(k^3), \tag{5.7}$$

where

$$x_1=-\frac{g_1|\mathbf{C}|}{\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}},\quad x_{2,3}=\pm\sqrt{-\frac{\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}}{\rho^0}},$$

$$y_1=-\frac{1}{\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}}\big[\frac{1}{Re}|\mathbf{M}||\mathbf{C}|+dg_1\big]-\frac{1}{\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}}\big[\rho^0x_1^3+x_1^2(\frac{1}{Re}+\rho^0\mathbf{M}:\mathbf{C})$$
$$+x_1(\rho^0|\mathbf{M}||\mathbf{C}|+\frac{1}{Re}\mathbf{M}:\mathbf{C}+\mathbf{p}^T\cdot\mathbf{K}\cdot\mathbf{p})\big], \tag{5.8}$$

$$y_{2,3}=-\frac{1}{2\rho^0Re}-\frac{1}{2\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}}[M_{11}(\rho_1^0h_{\rho_1\rho_1}+\rho_2^0h_{\rho_1\rho_2})^2+M_{22}(\rho_1^0h_{\rho_1\rho_2}+\rho_2^0h_{\rho_2\rho_2})^2].$$

where $d=h_{\rho_1\rho_1}\kappa_{\rho_2\rho_2}+h_{\rho_2\rho_2}\kappa_{\rho_1\rho_1}-2h_{\rho_1\rho_2}\kappa_{\rho_1\rho_2}$ and $g_1=M_{22}(\rho_1^0)^2+M_{11}(\rho_2^0)^2-2M_{12}\rho_1^0\rho_2^0\geq 0$, since $\mathbf{M}\geq 0$.

When $|k|\gg 1$, the three growth rates are given by

$$\alpha_{1,2}=x_{1,2}k^4+y_{1,2}k^2+O(k),\quad \alpha_3=x_3k^2+y_3+O(\frac{1}{k}), \tag{5.9}$$

where

$$x_{1,2}=-\frac{\mathbf{M}:\mathbf{K}}{2}\pm\frac{1}{2\rho^0}\sqrt{(\mathbf{M}:\mathbf{K}\rho^0)^2-4[\frac{1}{Re}\mathbf{M}:\mathbf{K}+\rho^0|\mathbf{M}||\mathbf{K}|]},\quad x_3=-\frac{1}{\rho^0Re},$$

$$y_{1,2}=\frac{-\frac{1}{Re}|\mathbf{M}||\mathbf{K}|-x_{1,2}^2(\frac{1}{Re}+\rho^0\mathbf{M}:\mathbf{C})-x_{1,2}(\frac{1}{Re}\mathbf{M}:\mathbf{K}+\rho^0|\mathbf{M}|d)}{3x_{1,2}^2\rho^0+2x_{1,2}\rho^0(\mathbf{M}:\mathbf{K})+\rho^0|\mathbf{M}||\mathbf{K}|}, \tag{5.10}$$

$$y_3=-\frac{1}{\rho^0|\mathbf{M}||\mathbf{K}|}\left[x_3^2(\rho^0\mathbf{M}:\mathbf{K})+x_3(\rho^0|\mathbf{M}|d+\frac{1}{Re}\mathbf{M}:\mathbf{K})+|\mathbf{M}|\frac{1}{Re}d+g_1|\mathbf{K}|\right].$$

The thermodynamic mode α_1 is related to the mobility matrix and Hessian matrix of the bulk free energy exclusively. The other two eigenvalues $\alpha_{2,3}$ are coupled with hydrodynamics.

Obviously, α_0 is negative so the viscous mode is stable. From the asymptotic expansions of α at $|k|\gg 1$, we observe that all three eigenvalues $\alpha_{1,2,3}$ are negative (5.9) since $\mathbf{K}>0,\mathbf{M}\geq 0$ and viscosity coefficients are positive. This indicates that the model does not have any short-wave instability near its steady states, which is physically meaningful.

When $|k|\ll 1$, we notice that the leading term in α_1 is determined by the combination of mobility coefficient matrix $\mathbf{M}$ and Hessian matrix $\mathbf{C}$ of the bulk free energy. We assume that $\mathbf{M}\geq 0$ and it has at least one positive eigenvalue, so $g_1>0$. We discuss the dependence of the leading order term of α_1 on $\mathbf{C}$.

	α_0	α_1	α_2	α_3
$\mathbf{C}>0$	negative	negative	negative	negative
$\mathbf{C}<0$	negative	positive	positive	negative
$\mathbf{C}$ is indefinite	negative	$\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}$ has the same sign with $\|\mathbf{C}\|$: negative; Otherwise, positive.	If $\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}>0$: negative; If $\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}<0$: positive.	negative

TABLE 5.1. *Sign of the eigenvalues when $|k|\ll 1$ in different regimes of $\mathbf{C}$. Negative sign indicates stability while positive sign indicates instability.*

(1) When $\mathbf{C}>0$, the leading term $-\frac{g_1|\mathbf{C}|}{\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}}k^2<0$, then $\alpha_1<0$. So, this mode is stable.

(2) When $\mathbf{C}<0$, the leading term $-\frac{g_1|\mathbf{C}|}{\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}}k^2>0$, then $\alpha_1>0$. This instability is due to the spinodal decomposition in the coupled Cahn-Hilliard-type equations of ρ_1 and ρ_2.

(3) When $\mathbf{C}$ is indefinite and $\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}$ has the same sign as $|\mathbf{C}|$, the property of α_1 is the same as the case where $\mathbf{C}>0$; otherwise, the property of α_1 is the same as the case of $\mathbf{C}<0$.

$\alpha_{2,3}$ represent the two coupled modes. Their signs depend on the model parameters. Since the leading term is determined by the properties of the Hessian matrix $\mathbf{C}$, we discuss their dependence on $\mathbf{C}$ below.

(1) When $\mathbf{C}>0$, $\sqrt{(-\frac{1}{\rho^0}\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p})}$ is imaginary. In this situation, the leading order growth rate in $\alpha_{2,3}$ is the quadratic term $(-\frac{1}{Re}\frac{1}{2\rho^0}-\frac{1}{2\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}}(M_{11}(\rho_1^0h_{\rho_1\rho_1}+\rho_2^0h_{\rho_1\rho_2})^2+M_{22}(\rho_1^0h_{\rho_1\rho_2}+\rho_2^0h_{\rho_2\rho_2})^2))k^2\leq 0$. So, the two modes are stable.

(2) When $\mathbf{C}<0$, the leading term is given by $\pm\sqrt{(-\frac{1}{\rho^0}\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p})}k$, indicating there

exists an unstable mode. This verifies the fact that the steady state at a concave free energy surface is unstable.

(3) When $\mathbf{C}$ is indefinite and $\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}>0$, the property of $\alpha_{2,3}$ is the same as the case where $\mathbf{C}>0$. Similarly, the property of $\alpha_{2,3}$ is the same as the case of $\mathbf{C}<0$ when $\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}<0$.

The stability property of the model with respect to $\mathbf{C}$ in the long-wave regime is summarized in Table 5.1. For the intermediate wave regime, we compute the growth rate numerically for specific free energy density functions.

5.2. Compressible model with the local mass conservation law. Notice that the compressible model with the local mass conservation law also admits the same constant solution (5.1). We repeat the same normal mode analysis analogous to the previous model and obtain the dispersion equation as follows:

$$(\alpha\rho^0+\frac{1}{Re_s}k^2)\{\alpha^3\rho^0+\alpha^2[\rho^0k^2M_{11}(\tilde{h}_{\rho_1\rho_1}+\tilde{\kappa}_{\rho_1\rho_1}k^2)+\frac{1}{Re}k^2]+\alpha[k^2M_{11}(\tilde{h}_{\rho_1\rho_1}+\tilde{\kappa}_{\rho_1\rho_1}k^2)\frac{1}{Re}k^2+\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}k^2+\mathbf{p}^T\cdot\mathbf{K}\cdot\mathbf{p}k^4]+k^4M_{11}(\rho^0)^2((\tilde{h}_{\rho_1\rho_1}+k^2\tilde{\kappa}_{\rho_1\rho_1})(\tilde{h}_{\rho\rho}+\tilde{\kappa}_{\rho\rho}k^2)-(\tilde{h}_{\rho\rho_1}+k^2\tilde{\kappa}_{\rho\rho_1})^2)\}=0. \tag{5.11}$$

Again, $\alpha_0=-\frac{1}{\rho^0}\frac{1}{Re_s}k^2$ is a root of this algebraic equation. We present the rest asymptotically.

When $|k|\ll 1$, we have

$$\alpha_1=-\frac{M_{11}(\rho^0)^2|\mathbf{C}|}{\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}}k^2+(-\frac{x_0^3\rho^0+x_0^2[\rho^0M_{11}\tilde{h}_{\rho_1\rho_1}+\frac{1}{Re}]+x_0[\mathbf{p}^T\cdot\mathbf{K}\cdot\mathbf{p}+\tilde{h}_{\rho_1\rho_1}M_{11}\frac{1}{Re}]}{\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}}-\frac{M_{11}(\rho^0)^2[\tilde{h}_{\rho_1\rho_1}\tilde{\kappa}_{\rho\rho}+\tilde{h}_{\rho\rho}\tilde{\kappa}_{\rho_1\rho_1}-2\tilde{h}_{\rho\rho_1}\tilde{\kappa}_{\rho\rho_1}]}{\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}})k^4+O(k^5), \tag{5.12}$$

$$\alpha_{2,3}=\pm\sqrt{(-\frac{1}{\rho^0}\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p})}k-(\frac{1}{Re}\frac{1}{2\rho^0}+\frac{M_{11}}{2\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}}(\rho_1^0\tilde{h}_{\rho_1\rho_1}+\rho^0\tilde{h}_{\rho\rho_1})^2)k^2+O(k^3),$$

where $x_0=-\frac{M_{11}(\rho^0)^2|\mathbf{C}|}{\mathbf{p}^T\cdot\mathbf{C}\cdot\mathbf{p}}$. When $|k|\gg 1$,

$$\alpha_1=-M_{11}\tilde{\kappa}_{\rho_1\rho_1}k^4-M_{11}\tilde{h}_{\rho_1\rho_1}k^2+O(k), \tag{5.13}$$

$$\alpha_{2,3}=\frac{-\frac{1}{Re}\pm\sqrt{(\frac{1}{Re})^2-4\tilde{\kappa}_{\rho_1\rho_1}^{-1}(\rho^0)^3|\mathbf{K}|}}{2\rho^0}k^2+\frac{-M_{11}(\rho^0)^2[\tilde{h}_{\rho_1\rho_1}\tilde{\kappa}_{\rho\rho}+\tilde{h}_{\rho\rho}\tilde{\kappa}_{\rho_1\rho_1}-2\tilde{h}_{\rho\rho_1}\tilde{\kappa}_{\rho\rho_1}]}{2x_{2,3}\rho^0M_{11}\tilde{\kappa}_{\rho_1\rho_1}+M_{11}\tilde{\kappa}_{\rho_1\rho_1}\frac{1}{Re}}-\frac{x_{2,3}^3\rho^0+x_{2,3}^2[\rho^0M_{11}\tilde{h}_{\rho_1\rho_1}+\frac{1}{Re}]+x_{2,3}[M_{11}\tilde{h}_{\rho_1\rho_1}\frac{1}{Re}+\mathbf{p}^T\cdot\mathbf{K}\cdot\mathbf{p}]}{2x_{2,3}\rho^0M_{11}\tilde{\kappa}_{\rho_1\rho_1}+M_{11}\tilde{\kappa}_{\rho_1\rho_1}\frac{1}{Re}}+O(\frac{1}{k}),$$

where $x_{2,3}=\frac{-\frac{1}{Re}\pm\sqrt{(\frac{1}{Re})^2-4\tilde{\kappa}_{\rho_1\rho_1}^{-1}(\rho^0)^3|\mathbf{K}|}}{2\rho^0}$,

$$\begin{pmatrix}\tilde{h}_{\rho\rho} & \tilde{h}_{\rho\rho_1}\\ \tilde{h}_{\rho\rho_1} & \tilde{h}_{\rho_1\rho_1}\end{pmatrix} \tag{5.14}$$

is the Hessian matrix of the bulk free energy density function h with respect to ρ and ρ_1 and evaluated at the constant steady state, and

$$\mathbf{K}=\begin{pmatrix}\tilde{\kappa}_{\rho\rho} & \tilde{\kappa}_{\rho\rho_1}\\ \tilde{\kappa}_{\rho\rho_1} & \tilde{\kappa}_{\rho_1\rho_1}\end{pmatrix} \tag{5.15}$$

is the coefficient matrix of the quadratic conformational entropy term in the free energy density function (2.26).

Like in the previous model, the first growth rate α_0 is the viscous mode associated to the viscous stress exclusively; the second growth rate α_1 is a thermodynamic mode, related to the transport equation of density ρ_1 and dictated by the mobility matrix and Hessian matrix of the bulk free energy. The other two growth rates $\alpha_{2,3}$ are coupled modes.

Obviously, α_0 is negative so the viscous mode is stable. For the other three modes, we adopt the same strategy to analyze their stability, combining asymptotic analysis in long and short waves with numerical computations in intermediate waves. From asymptotic expansions (5.13) of α at $|k|\gg 1$, we observe that all three eigenvalues $\alpha_{1,2,3}$ are negative, given that $\mathbf{K}$ and the mobility coefficients are both positive definite. This indicates that the model does not have any short-wave instability near its steady states. The properties of the three modes in the long-wave regime are identical to the cases discussed in the previous section for the more general compressible model and summarized in Table 5.1.

For the intermediate wave regime, we have to compute the growth rate using a specific free energy density function numerically. We use the Peng-Robinson bulk free energy as an example [37], given by

$$\begin{aligned}\tilde{h}(\rho_1,\rho,T)=&\frac{r_m\rho_1+(\rho-\rho_1)}{m_2}\varphi(T)-\frac{r_m\rho_1+(\rho-\rho_1)}{m_2}RTln(\frac{m_2}{r_m\rho_1+(\rho-\rho_1)}-b)\\&-\frac{r_m\rho_1+(\rho-\rho_1)}{m_2}\frac{a}{2\sqrt{2}b}ln[\frac{m_2+(r_m\rho_1+(\rho-\rho_1))b(1+\sqrt{2})}{m_2+(r_m\rho_1+(\rho-\rho_1))b(1-\sqrt{2})}]\\&+\frac{r_m\rho_1+(\rho-\rho_1)}{m_2}RT[\frac{r_m\rho_1}{r_m\rho_1+(\rho-\rho_1)}ln\frac{r_m\rho_1}{r_m\rho_1+(\rho-\rho_1)}\\&+\frac{\rho_2}{r_m\rho_1+(\rho-\rho_1)}ln\frac{(\rho-\rho_1)}{r_m\rho_1+(\rho-\rho_1)}].\end{aligned}\tag{5.16}$$

where $T=377.6K$, ρ_1 is the mass density of n-decane and ρ_2 the mass density of CO_2, $\rho=\rho_1+\rho_2$ is the total mass density. This is obtained by replacing ρ_1, ρ_2 in the free energy density given in (2.21) by $\rho_1,\rho_2=\rho-\rho_1$. This free energy density is either positive definite or indefinite in its entire physical domain. The positive definite domain and indefinite domain are shown in Figure 5.1 in (ρ_1,ρ) space. *Notice that in this example, when* $\mathbf{C}$ *is indefinite, we always have* $|\mathbf{C}|<0$*, it is impossible to have two unstable modes* α_1 *and* α_2 *exist simultaneously according to Table 5.1.* We then search the parameter space to sample all the possible instabilities associated to the compressible model with this free energy.

As an example, we choose the steady state given by $(\rho^0,\rho_1^0,\mathbf{v}_0)=(400,2,0,0)$ to show the positive growth in α_1. To show positive growth in the coupled mode α_2, we choose $(\rho^0,\rho_1^0,\mathbf{v}_0)=(1000,0.025,0,0)$. Figure 5.2 plots the three growth rates $\alpha_{1,2,3}$ with $\alpha_1>0$ at the first constant solution. The corresponding eigenvector to α_1 of the linearized system is $(0,1,0,0)$, indicating the unstable variable in the linear regime is ρ_1. The three growth rates $\alpha_{1,2,3}$ with the coupled mode $\alpha_2>0$ at the second solution are plotted in Figure 5.3. The corresponding eigenvector to α_2 is $(0,1,0,0)$ as well, indicating the instability is still associated with ρ_1. When $\mathbf{C}>0$, the corresponding constant solution is stable. We choose constant solution $(\rho^0,\rho_1^0,\mathbf{v}_0)=(400,200,0,0)$ as an example. The three growth rates $\alpha_{1,2,3}$ of negative real parts are shown in Figure 5.4. The numerical results show that the asymptotic analysis is accurate in their respective wave number ranges of applicability.

From the linear analysis above, we conclude that linear dynamics of compressible model (4.1) and (4.2) are qualitatively the same. Next, we investigate the near equilibrium dynamics of the quasi-incompressible model.

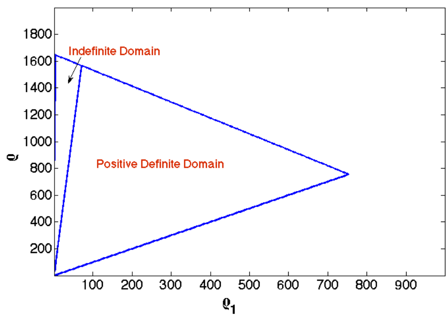
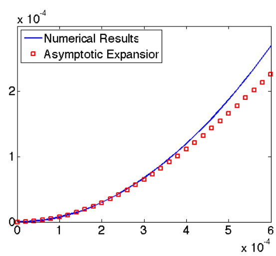
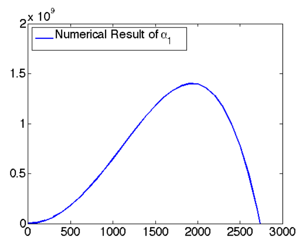


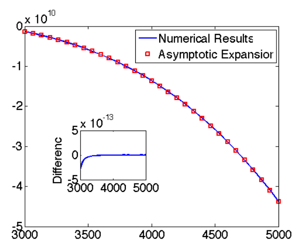
FIG. 5.1. *Domain of concavity of the Peng-Robinson free energy.*



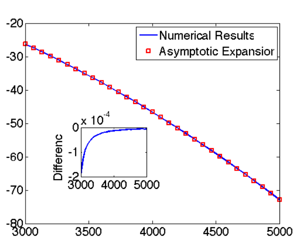
(a) α_1 when $|k| \ll 1$



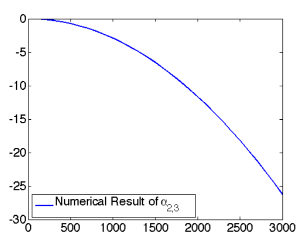
(b) α_1



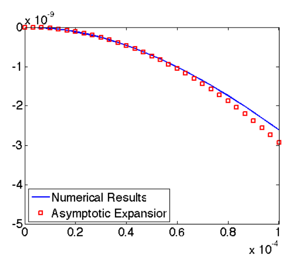
(c) α_1 when $|k| \gg 1$



(d) $\alpha_{2,3}$ when $|k| \gg 1$



(e) $\alpha_{2,3}$

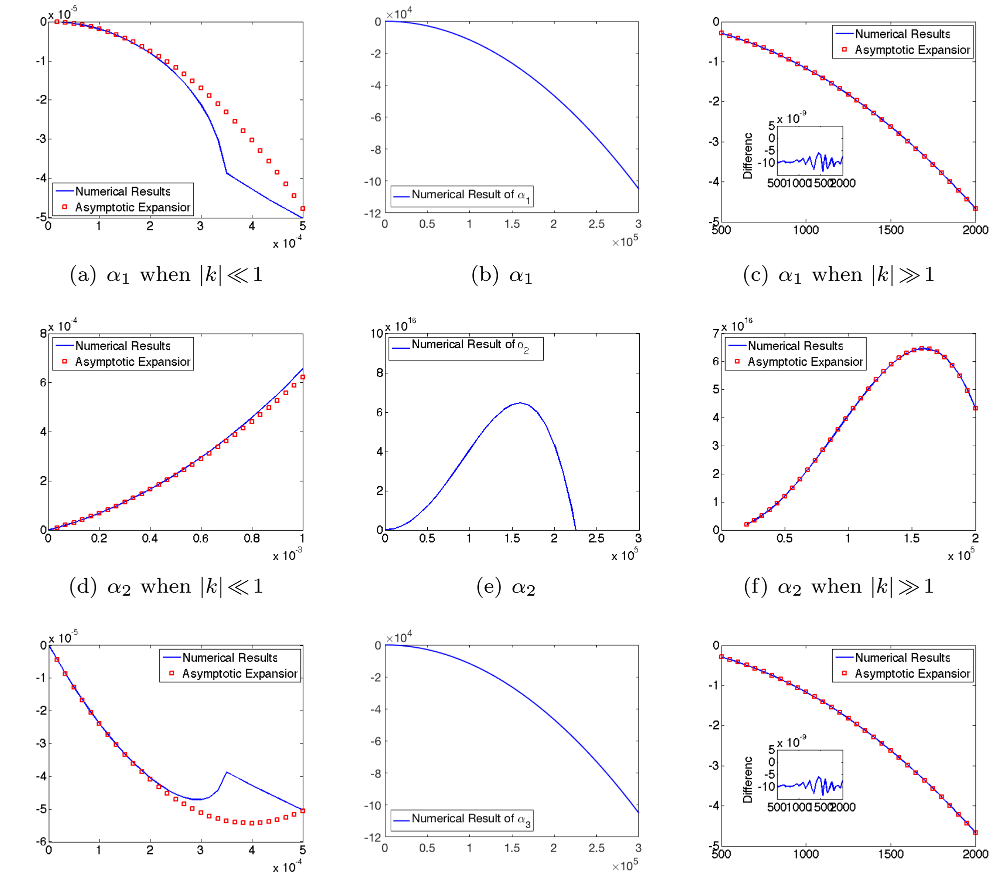


(f) $\alpha_{2,3}$ when $|k| \ll 1$

FIG. 5.2. *Numerical growth rates and the corresponding asymptotic ones as functions of the wave number when $\alpha_1 > 0$ and all others are negative in compressible model (4.2) at constant state $(\rho^0, \rho_1^0, \mathbf{v}) = (400, 2, 0, 0)$ with the Peng-Robinson free energy. The vertical axis is the growth rate and the horizontal one is the wave number. (a). α_1 in the long wave range. (b). α_1 in the intermediate wave range. (c). α_1 in the short wave range. (d). $\alpha_{2,3}$ in the short wave range. (e). $\alpha_{2,3}$ in the intermediate wave range. (f). $\alpha_{2,3}$ in the short wave range. The parameter values used are: $M_{11} = 0.0001$, $Re_s = 1$, $Re_v = 3$, $\tilde{\kappa}_{\rho\rho} = 0.000106$, $\tilde{\kappa}_{\rho_1\rho_1} = 0.0001$, $\tilde{\kappa}_{\rho\rho_1} = 0$.*

5.3. Quasi-incompressible model. The quasi-incompressible fluid flow model equation admits constant solution:

$$\mathbf{v} = \mathbf{0}, \quad \phi = \phi^0, \quad \Pi = \Pi_0, \tag{5.17}$$



(a) α_1 when $|k| \ll 1$ (b) α_1 (c) α_1 when $|k| \gg 1$

(d) α_2 when $|k| \ll 1$ (e) α_2 (f) α_2 when $|k| \gg 1$

(g) α_3 when $|k| \ll 1$ (h) α_3 (i) α_3 when $|k| \gg 1$

FIG. 5.3. *Numerical growth rates and the corresponding asymptotic ones when $\alpha_2 > 0$ while the others are negative compressible model (4.2) at constant state $(\rho^0, \rho_1^0, \mathbf{v}) = (1000, 0.025, 0, 0)$ with the Peng-Robinson free energy. The vertical axis is the growth rate and the horizontal one is the wave number. (a). (d). (g). Growth rates in the short wave range. (b). (e). (h). Growth rates in the intermediate wave range. (c). (f). (j). Growth rates in the short wave range. The parameter values used are: $M_{11} = 0.0001$, $Re_s = 1$, $Re_v = 3$, $\tilde{\kappa}_{\rho\rho} = 0.000106$, $\tilde{\kappa}_{\rho_1\rho_1} = 0.0001$, $\tilde{\kappa}_{\rho\rho_1} = 0$.*

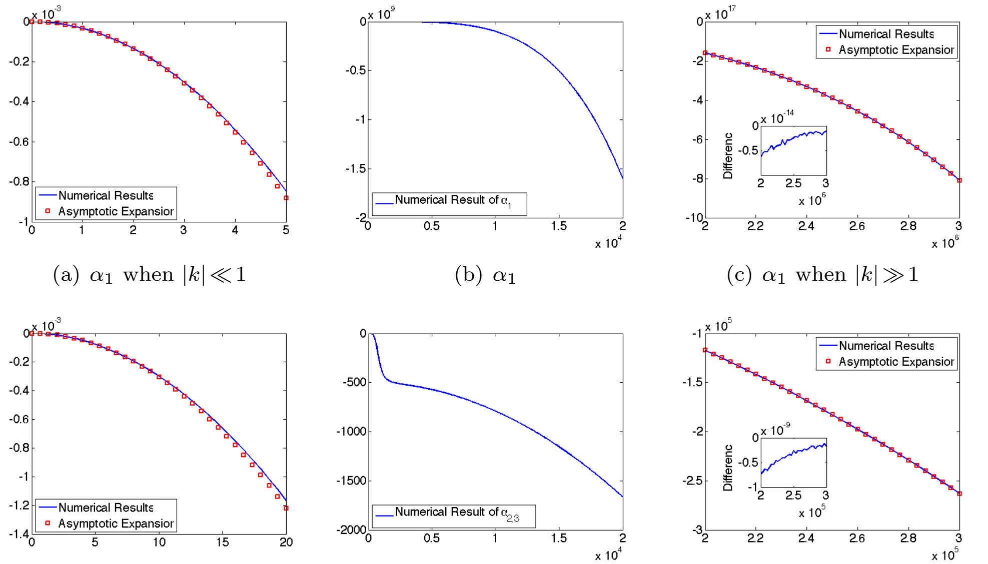
where ϕ^0, Π_0 are constants. We perturb the constant solution as follows:

$$\mathbf{v} = \epsilon e^{\alpha t + i\mathbf{k}\cdot\mathbf{x}} \mathbf{v}^c, \quad \phi = \phi^0 + \epsilon e^{\alpha t + i\mathbf{k}\cdot\mathbf{x}} \phi^c, \quad \Pi = \Pi_0 + \epsilon e^{\alpha t + i\mathbf{k}\cdot\mathbf{x}} \Pi^c, \tag{5.18}$$

where ϵ is a small perturbation, and $\mathbf{v}^c, \phi^c, \Pi^c$ are constants.

The dispersion equation of the linearized system is a factorable, third-order polynomial in α

$$\begin{aligned}
&(\frac{1}{Re_s} k^2 + \alpha \rho^0)(\alpha^2 (1 - \frac{\hat{\rho}_1}{\hat{\rho}_2})^2 \frac{1}{\hat{\rho}_1{}^2} M_{11} k^2 \rho^0 + \alpha [k^2 + \frac{1}{Re}(1 - \frac{\hat{\rho}_1}{\hat{\rho}_2})^2 \frac{1}{\hat{\rho}_1{}^2} M_{11} k^4] \\
&\quad + k^4 M_{11} (\hat{h}_{\phi\phi} + k^2 \hat{\kappa}_{\phi\phi}) \frac{1}{\hat{\rho}_1{}^2} [1 - (1 - \frac{\hat{\rho}_1}{\hat{\rho}_2}) \phi_0]^2) = 0.
\end{aligned} \tag{5.19}$$



(a) α_1 when $|k| \ll 1$ (b) α_1 (c) α_1 when $|k| \gg 1$

(d) $\alpha_{2,3}$ when $|k| \ll 1$ (e) $\alpha_{2,3}$ (f) $\alpha_{2,3}$ when $|k| \gg 1$

FIG. 5.4. *Numerical growth rates and the corresponding asymptotic ones without any unstable modes in compressible model* (4.2) *at constant state* $(\rho^0, \rho_1^0, \mathbf{v}) = (400, 200, 0, 0)$ *with the Peng-Robinson free energy. (a). and (d). Growth rates in the short wave range. (b). and (e). Growth rates in the intermediate wave range. (c). and (f). Growth rates in the short wave range. The parameter values used are:* $M_{11} = 0.0001$, $Re_s = 10^6$, $Re_v = 3 \times 10^6$, $\tilde{\kappa}_{\rho\rho} = 0.000106$, $\tilde{\kappa}_{\rho_1\rho_1} = 0.0001$, $\tilde{\kappa}_{\rho\rho_1} = 0$.

i.e.

$$\left(\frac{1}{Re_s}k^2 + \alpha\rho^0\right)\left[\left(1 - \frac{\hat{\rho}_1}{\hat{\rho}_2}\right)^2 \frac{1}{\hat{\rho}_1{}^2} M_{11} k^2\right]\left(\alpha^2\rho^0 + \alpha\left[\left[\left(1 - \frac{\hat{\rho}_1}{\hat{\rho}_2}\right)^2 \frac{1}{\hat{\rho}_1{}^2} M_{11}\right]^{-1} + \frac{1}{Re}k^2\right]\right.$$
$$\left. + k^2(\hat{h}_{\phi\phi} + k^2\hat{\kappa}_{\phi\phi})\left[\phi_0 - \frac{\hat{\rho}_2}{\hat{\rho}_2 - \hat{\rho}_1}\right]^2\right) = 0. \tag{5.20}$$

The growth rates are given explicitly by

$$\alpha_0 = -\frac{1}{Re_s}\frac{1}{\rho^0}k^2,$$
$$\alpha_1 = \frac{-2k^2(\hat{h}_{\phi\phi} + k^2\hat{\kappa}_{\phi\phi})Q^2}{\left[\left(\frac{1}{Re}k^2 + A\right) + \sqrt{\left(\frac{1}{Re}k^2 + A\right)^2 - 4\rho^0 k^2(\hat{h}_{\phi\phi} + k^2\hat{\kappa}_{\phi\phi})Q^2}\right]}, \tag{5.21}$$
$$\alpha_2 = \frac{-\left(\frac{1}{Re}k^2 + A\right) - \sqrt{\left(\frac{1}{Re}k^2 + A\right)^2 - 4\rho^0 k^2(\hat{h}_{\phi\phi} + k^2\hat{\kappa}_{\phi\phi})Q^2}}{2\rho^0},$$

where

$$Q = \phi^0 - \frac{\hat{\rho}_2}{\hat{\rho}_2 - \hat{\rho}_1}, \quad \frac{1}{Re} = 2\frac{1}{Re_s} + \frac{1}{Re_v} > 0, \quad A = \left[\left(1 - \frac{\hat{\rho}_1}{\hat{\rho}_2}\right)^2 \frac{1}{(\hat{\rho}_1)^2} M_{11}\right]^{-1} > 0. \tag{5.22}$$

The stable hydrodynamic mode is in α_0 again. The thermodynamic modes are now given by $\alpha_{1,2}$. $Re(\alpha_1)$ can be positive only when $\hat{h}_{\phi\phi} < 0$, in which $Re(\alpha_1) > 0$ when

$0 \le k \le \sqrt{-\frac{\hat{h}_{\phi\phi}}{\kappa_{\phi\phi}}}$. This instability is due to the spinodal decomposition in the coupled Cahn-Hilliard equation of ϕ. Given that the viscosity and mobility coefficients are all positive, $Re(\alpha_2) < 0$. So, the second coupled mode is a stable mode. In the long wave range ($|k| \ll 1$), $\alpha_1 \approx -\frac{M_{11}\hat{h}_{\phi\phi}}{\hat{\rho_1}^2\hat{\rho_2}^2}(\hat{\rho_1} + (\hat{\rho_1} - \hat{\rho_2})\phi^0)^2 k^2$.

When $\hat{\rho_1} = \hat{\rho_2}$, the model reduces to an incompressible model with the following two growth rates

$$\alpha_0 = -\frac{1}{Re_s}\frac{1}{\rho^0}k^2,$$

$$\alpha_1 = -\frac{1}{\hat{\rho_2}^2}M_{11}\hat{h}_{\phi\phi}k^2 - \frac{1}{\hat{\rho_1}^2}\hat{\kappa}_{\phi\phi}M_{11}k^4. \tag{5.23}$$

The thermodynamic mode decouples from the hydrodynamic mode completely in the linear regime. The possible instability only lies in the spinodal mode of the Cahn-Hilliard equation. In fact, $A, Q \to \infty$ in this limit. So, the growth rate associated with α_2 in the quasi-incompressible model is lost in the incompressible model.

5.4. Summary of linear stability results. In compressible phase field models, there are four modes in the 1D perturbation analysis: α_0 is the hydrodynamic mode dictated by the viscous stress, α_1 is the thermodynamic mode dominated by the mobility and the bulk free energy, the other two modes $\alpha_{2,3}$ are coupled, which couples dynamics of phase behavior with hydrodynamics and may be unstable depending on the composition of the fluid mixture. When the Hessian matrix of the bulk free energy $\mathbf{C} > 0$, $\sqrt{(-\frac{1}{\rho^0}\mathbf{p}^T \cdot \mathbf{C} \cdot \mathbf{p})}$ is imaginary. So $\pm\sqrt{(-\frac{1}{\rho^0}\mathbf{p}^T \cdot \mathbf{C} \cdot \mathbf{p})}k$ represents a wave that does not contribute to the amplitude change in growth rates of the linearized system. The scenario on stability of the steady state is tabulated in Table 5.1.

When the quasi-incompressible constraint is added, i.e. $\rho_1 = \hat{\rho_1}\phi, \rho_2 = \hat{\rho_2}(1-\phi)$. The positive definite matrix $\mathbf{C}$ reduces to a singular matrix

$$\mathbf{C} = h_{\phi\phi}\begin{pmatrix} \frac{1}{\hat{\rho_1}\hat{\rho_1}} & -\frac{1}{\hat{\rho_1}\hat{\rho_2}} \\ -\frac{1}{\hat{\rho_1}\hat{\rho_2}} & \frac{1}{\hat{\rho_2}\hat{\rho_2}} \end{pmatrix} \tag{5.24}$$

Obviously, $|\mathbf{C}| = 0$ and $\mathbf{p}^T \cdot \mathbf{C} \cdot \mathbf{p} = (2\phi - 1)^2$ for $\rho_1^0 = \phi\hat{\rho_1}$, $\rho_2^0 = (1-\phi)\hat{\rho_2}$. The growth rates reduce to two modes labeled as $\alpha_{1,2}$. They are not necessarily related to the $\alpha_{1,2}$ in the compressible model. When the quasi-incompressible mode reduces to the incompressible model further, the coupled hydrodynamic modes reduce to one mode in α_1.

The analysis shows that the more constraints we have on the composition of the fluid mixture, the less coupled the equations are in linearized systems. In 3D models, the total number of growth rates will increase as the number of equations increases. But, the number of unstable modes will not change. In this paper, the perturbation analysis is carried out in a 1D wave number direction in 2D space and it can be extended to the multi-dimensional wave number space as well. We will omit the details for simplicity.

6. Conclusion

We have presented a systematic derivation of hydrodynamic phase field models for multi-component fluid mixtures of compressible fluids as well as their reductions to quasi-incompressible and incompressible fluids under isothermal conditions. The governing equations in the models are composed of the mass and momentum conservation law as well as the constitutive equations, which are derived using the generalized Onsager principle to warrant an energy dissipation in time. By relaxing or enforcing local mass conservation law while keeping the total mass conserved, we obtain two classes of compressible models, one conserves the local mass while the other does not. Via a Lagrange multiplier approach, we reduce the compressible model with the local mass conservation law to a quasi-incompressible model when the constituent fluids are all incompressible and the mixture is assumed simple. The quasi-incompressible model further reduces to the incompressible model while the constituent fluid components are of the same densities.

We then study linear stability of the hydrodynamic models. The properties of linear stability are studied and differences of the models in the linear regime are identified: there exist three types of growth/decay rates among the models. The first type is dominated by the viscous property of the fluid, known as the viscous mode. The second type is the thermodynamic mode, which is dominated by the mobility and Hessian of the bulk free energy density. The third type is the coupled mode among the phase variables and hydrodynamic variables. When more constraints are imposed to reduce the models from compressible, to quasi-incompressible and then to incompressible models, the number of coupled modes reduces accordingly, indicating that these constraints weaken the coupling of the equations in the model at least in the linear regime. This study not only develops a general framework for the derivation of compressible models and their reduction to quasi-incompressible and incompressible models, but also identifies differences between compressible and incompressible models in near equilibrium dynamics.

Acknowledgements. Qi Wang's research is partially supported by NSF grants DMS-1815921, OIA-1655740, DOE grant DE-SC0020272 and a SC EPSCOR GEAR award. Tiezheng Qian's research is partially supported by Hong Kong RGC Collaborative Research Fund No. C1018-17G.

Appendix. Dispersion equations of the 2D hydrodynamic models. We list the dispersion equations in determinant forms of all hydrodynamic models derived in this study in two space dimensions in this appendix.

A.1. Dispersion equation of the compressible model with the global mass conservation. The dispersion equation of the linearized equation system of the compressible model with the global mass conservation is given by a 4×4 determinant as follows

$$det\begin{pmatrix} \alpha+A_{11} & A_{12} & i\rho_1^0 k & 0 \\ A_{21} & \alpha+A_{22} & i\rho_2^0 k & 0 \\ ik(\rho_1^0 D_{11}+\rho_2^0 D_{12}) & ik(\rho_2^0 D_{22}+\rho_1^0 D_{12}) & \alpha\rho^0+\frac{1}{Re}k^2 & 0 \\ 0 & 0 & 0 & \alpha\rho^0+\frac{1}{Re_s}k^2 \end{pmatrix}=0, \qquad \text{(A.1)}$$

where $A_{11}=k^2(M_{11}D_{11}+M_{12}D_{12})$, $A_{12}=k^2(M_{11}D_{12}+M_{12}D_{22})$, $A_{21}=k^2(M_{12}D_{11}+M_{22}D_{12})$, $A_{22}=k^2(M_{12}D_{12}+M_{22}D_{22})$ and $D_{11}=h_{\rho_1\rho_1}+k^2\kappa_{\rho_1\rho_1}$, $D_{22}=h_{\rho_2\rho_2}+k^2\kappa_{\rho_2\rho_2}$, $D_{12}=h_{\rho_1\rho_2}+k^2\kappa_{\rho_1\rho_2}$, $\frac{1}{Re}=2\frac{1}{Re_s}+\frac{1}{Re_v}$. The growth/decay rate in the hydrodynamic mode associated to the viscous stress is given explicitly by $\alpha=-\frac{1}{Re_s}\frac{1}{\rho^0}k^2$, which decouples from the rest of the modes. This decoupling is inherited by all its limiting models given below.

A.2. Dispersion equation of the compressible model with local mass conservation. The dispersion equation of the linearized equation system of this model is given by a 4×4 determinant as follows

$$
det\begin{pmatrix} \alpha & 0 & i\rho^0 k & 0 \\ (k^2 M_{11})D_{12} & \alpha+(k^2 M_{11})D_{22} & i\rho_1^0 k & 0 \\ ik(\rho_1^0 D_{12}+\rho^0 D_{11}) & ik(\rho_1^0 D_{22}+\rho^0 D_{12}) & \alpha\rho^0+\frac{1}{Re}k^2 & 0 \\ 0 & 0 & 0 & \alpha\rho^0+\frac{1}{Re_s}k^2 \end{pmatrix}=0, \qquad (A.2)
$$

where $D_{11}=\tilde{h}_{\rho\rho}+k^2\tilde{\kappa}_{\rho\rho}$, $D_{22}=\tilde{h}_{\rho_1\rho_1}+k^2\tilde{\kappa}_{\rho_1\rho_1}$, $D_{12}=\tilde{h}_{\rho\rho_1}+k^2\tilde{\kappa}_{\rho\rho_1}$, and $\frac{1}{Re}=2\frac{1}{Re_s}+\frac{1}{Re_v}$.

A.3. Dispersion equation of the quasi-incompressible model. The resulting dispersion equation of the linearized system of this model is given by a 4×4 determinant as follows

$$
det\begin{pmatrix} 0 & -\alpha(1-\frac{\hat{\rho}_1}{\hat{\rho}_2}) & ik-ik\phi^0(1-\frac{\hat{\rho}_1}{\hat{\rho}_2}) & 0 \\ \frac{1}{\hat{\rho}_1{}^2}M_{11}k^2(1-\frac{\hat{\rho}_1}{\hat{\rho}_2}) & \alpha+\frac{1}{\hat{\rho}_1{}^2}M_{11}k^2 D_\phi & ik\phi^0 & 0 \\ ik & ik\phi^0 D_\phi & \alpha\rho^0+\frac{1}{Re}k^2 & 0 \\ 0 & 0 & 0 & \alpha\rho^0+\frac{1}{Re_s}k^2 \end{pmatrix}=0, \qquad (A.3)
$$

where $D_\phi=\hat{h}_{\phi\phi}+\hat{\kappa}_{\phi\phi}k^2$, $\hat{h}_{\phi\phi}=\frac{\partial^2 h}{\partial\phi^2}$ is the second-order derivative of the bulk free energy density function h with respect to volume fraction ϕ at the constant solution, and $\hat{\kappa}_{\phi\phi}$ is the coefficient of the conformational entropy. If we multiply $(1-\frac{\hat{\rho}_1}{\hat{\rho}_2})$ by the second row and add it to the first row of the dispersion relation matrix, we obtain

$$
det\begin{pmatrix} \frac{1}{\hat{\rho}_1{}^2}M_{11}(1-\frac{\hat{\rho}_1}{\hat{\rho}_2})^2 k^2 & (1-\frac{\hat{\rho}_1}{\hat{\rho}_2})\frac{1}{\hat{\rho}_1{}^2}M_{11}k^2 D_\phi & ik & 0 \\ \frac{1}{\hat{\rho}_1{}^2}M_{11}(1-\frac{\hat{\rho}_1}{\hat{\rho}_2})k^2 & \alpha+\frac{1}{\hat{\rho}_1{}^2}M_{11}k^2 D_\phi & ik\phi^0 & 0 \\ ik & ik\phi^0 D_\phi & \alpha\rho^0+\frac{1}{Re}k^2 & 0 \\ 0 & 0 & 0 & \alpha\rho^0+\frac{1}{Re_s}k^2 \end{pmatrix}=0, \qquad (A.4)
$$

A.4. Dispersion equation of the incompressible model. The dispersion equation of the linearized system of the incompressible model is given by

$$
det\begin{pmatrix} 0 & 0 & ik & 0 \\ 0 & \alpha+\frac{1}{\hat{\rho}_1{}^2}M_{11}k^2 D_\phi & ik\phi^0 & 0 \\ ik & ik\phi^0 D_\phi & \alpha\rho^0+\frac{1}{Re}k^2 & 0 \\ 0 & 0 & 0 & \alpha\rho^0+\frac{1}{Re_s}k^2 \end{pmatrix}=0. \qquad (A.5)
$$

This can be obtained from that in the quasi-incompressible model by equating $\hat{\rho}_1=\hat{\rho}_2$ in (A.3).

REFERENCES

[1] H. Abels, *On a diffuse interface model for two-phase flows of viscous, incompressible fluids with matched densities*, Arch. Ration. Mech. Anal., 194(2):463–506, 2009. 1

[2] S. Aland, S. Egerer, J. Lowengrub, and A. Voigt, *Diffuse interface models of locally inextensible vesicles in a viscous fluid*, J. Comput. Phys., 277:32–47, 2014. 1

[3] S. Aland, J. Lowengrub, and A. Voigt, *Particles at fluid-fluid interfaces: a new Navier-Stokes-Cahn-Hilliard surface-phase-field model*, Phys. Rev. E, 86:046321, 2012. 1

[4] N. Auffray, F. dell'Isola, V. Eremeyev, A. Madeo, and G. Rosi, *Analytical continuum mechanics à la Hamilton-Piola least action principle for second gradient continua and capillary fluids*, Math. Mech. Solids, 20(4):375–417, 2015. 1

[5] A.N. Beris and B. Edwards, *Thermodynamics of Flowing Systems*, Oxford Science Publications, New York, 1994. 1

[6] A. Bertozzi, S. Esedoglu, and A. Gillette, *Inpainting of binary images using the Cahn-Hilliard equation*, IEEE Trans. Image Process., 16(1):285–291, 2007. 1

[7] M. Borden, C. Verhoosej, M. Scott, T. Hughes, and C. Landis, *A phase-field description of dynamic brittle fracture*, Comput. Meth. Appl. Mech. Eng., 217(220):77–95, 2012. 1

[8] B. Camley, Y. Zhao, Bo Li, H. Levine, and W. Rappel, *Crawling and turning in a minimal reaction-diffusion cell motility model: coupling cell shape and biochemistry*, Phys. Rev. E, 95:012401, 2017. 1

[9] L.Q. Chen and W. Yang, *Computer simulation of the dynamics of a quenched system with large number of non-conserved order parameters*, Phys. Rev. B, 60:15752–15756, 1994. 1

[10] H.C. Öttinger, *Beyond Equilibrium Thermodynamics*, Wiley, Hoboken, 2005. 1

[11] H.C. Öttinger and M. Grmela, *Dynamics and thermodynamics of complex fluids. II. Illustrations of a general formalism*, Phys. Rev. E, 56:6633, 1997. 1

[12] F. dell'Isola, A. Madeo, and P. Seppecher, *Boundary conditions at fluid-permeable interfaces in porous media: A variational approach*, Int. J. Solids Struct., 46(17):3150–3164, 2009. 1

[13] M. Doi, *Onsager's variational principle in soft matter*, J. Phys. Condens. Matter, 23:284118, 2011. 1

[14] Q. Du, C. Liu, R. Ryham, and X. Wang, *A phase field formulation of the Willmore problem*, Nonlinearity, 18:1249–1267, 2005. 1

[15] N. Gavish, G. Hayrapetyan, K. Promislow, and L. Yang, *Curvature driven flow of bilayer interfaces*, Phys. D, 240:675–693, 2011. 1

[16] S. Gavrilyuk and H. Gouin, *A new form of governing equations of fluids arising from Hamilton's principle*, Int. J. Eng. Sci., 37(12):1495–1520, 1999. 1

[17] S. Gavrilyuk, H. Gouin, and Y. Perepechko, *Hyperbolic models of homogeneous two-fluid mixtures*, Meccanica, 33(2):161–175, 1998. 1

[18] Y. Gong, J. Zhao, and Q. Wang, *Second order fully discrete energy stable methods on staggered grids for hydrodynamic phase field models of binary viscous fluids*, SIAM J. Sci. Comput., 40(2):B528–B553, 2018. 1

[19] Y. Gong, J. Zhao, X. Yang, and Q. Wang, *Fully discrete second-order linear schemes for hydrodynamic phase field models of binary viscous fluid flows with variable densities*, SIAM J. Sci. Comput., 40(1):B138–B167, 2018. 1

[20] H. Gouin, *Variational theory of mixtures in continuum mechanics*, Eur. J. Mech. B Fluids, 9(5):469–491, 1990. 1

[21] H. Gouin and S. Gavrilyuk, *Hamilton's principle and Rankine-Hugoniot conditions for general motions of mixtures*, Meccanica, 34(1):39–47, 1999. 1

[22] A.E Green and P.M. Naghdi, *A re-examination of the basic postulates of thermomechanics*, Proc. R. Soc. A, 432(1885):171–194, 1991. 1

[23] M. Grmela and H.C. Öttinger, *Dynamics and thermodynamics of complex fluids. I. Development of a general formalism*, Phys. Rev. E, 56:6620, 1997. 1

[24] P.C. Hohenberg and B.I. Halperin, *Theory of dynamic critical phenomena*, Rev. Modern Phys., 49(3):435–479, 1977. 2.3

[25] D.D. Joseph and Y.Y. Renardy, *Fundamentals of Two-Fluid Dynamics Part II: Lubricated Transport, Drops and Miscible Liquids*, Springer Science+Business Media, LLC, 1993. 1, 2.3

[26] M. Kapustina, D. Tsygankov, J. Zhao, T. Wessler, X. Yang, A. Chen, N. Roach, T.C. Elston, Q. Wang, K. Jacobson, and M.G. Forest, *Modeling the excess cell surface stored in a complex morphology of bleb-like protrusions*, PLOS Comput. Biol., 12(3):1–25, 2016. 1

[27] J. Li and Q. Wang, *A class of conservative phase field models for multiphase fluid flows*, J. Appl. Mech., 81(2):021004, 2014. 1, 1, 2.2, 2.3

[28] Y. Li and J. Kim, *Multiphase image segmentation using a phase-field model*, Comput. Math. Appl., 62:737–745, 2011. 1

[29] C. Liu and J. Shen, *A phase field model for the mixture of two incompressible fluids and its approximation by a Fourier-spectral method*, Phys. D, 179:211–228, 2003. 1

[30] J. Lober, F. Ziebert, and I.S. Aranson, *Modeling crawling cell movement on soft engineered substrates*, Soft Matter, 10:1365, 2014. 1

[31] J. Lowengrub, A. Ratz, and A. Voigt, *Phase field modeling of the dynamics of multicomponent vesicles spinodal decomposition coarsening budding and fission*, Phys. Rev. E, 79:031926, 2009. 1

[32] J.S. Lowengrub and L. Truskinovsky, *Quasi incompressible Cahn-Hilliard fluids and topological transitions*, Proc. R. Soc. A, 454:2617–2654, 1998. 1, 1, 2.2

[33] S. Najem and M. Grant, *Phase-field model for collective cell migration*, Phys. Rev. E, 93:052405, 2016. 1

[34] M. Nonomura, *Study on multicellular systems using a phase field model*, PLoS One, 7(4):0033501, 2012. 1
[35] L. Onsager, *Reciprocal relations in irreversible processes I*, Phys. Rev., 37:405–426, 1931. 1
[36] L. Onsager, *Reciprocal relations in irreversible processes II*, Phys. Rev., 38:2265–2279, 1931. 1
[37] D.-Y. Peng and D.B. Robinson, *A new two-constant equation of state*, Ind. Eng. Chem. Fundamen., 15(1):59–64, 1976. 5.2
[38] D. Shao, H. Levine, and W. Pappel, *Coupling actin flow, adhesion, and morphology in a computational cell motility model*, Proc. Natl. Acad. Sci. USA, 109(18):6851–6856, 2012. 1
[39] D. Shao, W. Pappel, and H. Levine, *Computational model for cell morphodynamics*, Phys. Rev. Lett., 105:108104, 2010. 1
[40] S. Torabi, J. Lowengrub, A. Voigt, and S. Wise, *A new phase-field model for strongly anisotropic systems*, Proc. R. Soc. A, 265:1337–1359, 2009. 1
[41] X. Wang and Q. Du, *Modeling and simulations of multi-component lipid membranes and open membranes via diffuse interface approaches*, J. Math. Bio., 56:347–371, 2008. 1
[42] S. Wise, J. Lowengrub, H. Frieboes, and B. Cristini, *Three dimensional multispecies nonlinear tumor growth. I. Model and numerical method*, J. Theor. Biol., 253(3):524–543, 2008. 1
[43] T. Witkowski, R. Backofen, and A. Voigt, *The influence of membrane bound proteins on phase separation and coarsening in cell membranes*, Phys. Chem. Chem. Phys., 14(42):14403–14712, 2012. 1
[44] X. Yang, J. Li, G. Forest, and Q. Wang, *Hydrodynamic theories for flows of active liquid crystals and the generalized Onsager principle*, Entropy, 18(6):202, 2016. 1, 2
[45] J. Zhao, P. Seeluangsawat, and Q. Wang, *Modeling antimicrobial tolerance and treatment of heterogeneous biofilms*, Math. Biosci., 282:1–15, 2016. 1
[46] J. Zhao, Y. Shen, M. Happasalo, Z.J. Wang, and Q. Wang, *A 3D numerical study of antimicrobial persistence in heterogeneous multi-species biofilms*, J. Theor. Biol., 392:83–98, 2016. 1
[47] J. Zhao and Q. Wang, *Three-dimensional numerical simulations of biofilm dynamics with quorum sensing in a flow cell*, Bull. Math. Biol., 79(4):884–919, 2017. 1
[48] J. Zhao and Q. Wang, *Modeling cytokinesis of eukaryotic cells driven by the actomyosin contractile ring*, Int. J. Numer. Meth. Biomed. Eng., 32(12):e02774, 2016. 1
[49] F. Ziebert and I.S. Aranson, *Effects of adhesion dynamics and substrate compliance on the shape and motility of crawling cells*, PLOS One, 8(5):e64511, 2013. 1
[50] F. Ziebert, S. Swaminathan, and I.S. Aranson, *Model for self-polarization and motility of keratocyte fragments*, J. R. Soc. Interface, 9:1084–1092, 2011. 1
[51] D. Zwicker, R. Seyboldt, C. Weber, A. Hyman, and F. Julicher, *Growth and division of active droplets provides a model for protocells*, Nature Phys., 13:408–413, 2017. 1