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Multiphase Allen–Cahn and Cahn–Hilliard models and their discretizations with the effect of pairwise surface tensions ☆

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ABSTRACT

In this paper, the mathematical properties and numerical discretizations of multiphase models that simulate the phase separation of an N-component mixture are studied. For the general choice of phase variables, the unisolvent property of the coefficient matrix involved in the N-phase models based on the pairwise surface tensions is established. Moreover, the symmetric positive-definite property of the coefficient matrix on an $(N-1)$-dimensional hyperplane — which is of fundamental importance to the well-posedness of the models — can be proved equivalent to some physical condition for pairwise surface tensions. The N-phase Allen–Cahn and N-phase Cahn–Hilliard equations can then be derived from the free-energy functional. A natural property is that the resulting dynamics of concentrations are independent of phase variables chosen. Finite element discretizations for N-phase models can be obtained as a natural extension of the existing discretizations for the two-phase model. The discrete energy law of the numerical schemes can be proved and numerically observed under some restrictions pertaining to time step size. Numerical experiments including the spinodal decomposition and the evolution of triple junctions are described in order to investigate the effect of pairwise surface tensions.

1. Introduction

Multiphase flows are frequently encountered in biomedical, chemical, and engineering applications. The dynamics of multiphase flows associate with a wide range of fundamental physical properties such as pairwise surface tensions, wetting spreading, and formating contact angles among multiple materials [1]. On the other hand, multiphase flows are challenging from the points of view of both mathematical modeling and numerical methods due to the complexity of the moving interface.

There are two main approaches to moving interface problems: the direct approach and the indirect approach. The direct approach obtains information pertaining to the interface by tracking quantities associated with it. Therefore, the direct approach relies on the parameterization method [2], the immersed boundary method [3], the volume-of-fluid method [4], and/or the front tracking method [5]. It is known that the direct approach commonly encounters difficulty handling topological changes, such as pinches, splits, and merging — all of which can be handled easily by the indirect approach. The level set method [6] and the phase field method [7] are both examples of popular indirect methods. In this paper, however, we focus on the phase field method for modeling the effect of pairwise surface tensions for N-phase flows ($N \geq 2$).

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With the phase field method, the thickness of the sharp interface between the two phases is supposed to be very small but positive. The state of the system is then represented by a set of smooth functions called phase variables or order parameters. The evolution of the system is driven by the gradient of a total free-energy, which is the sum of two terms: a bulk free-energy term, whose effect tends to separate the flows, and a capillary term, whose effect tends to mix the flows. The capillary term depends on the gradient of the order parameters, which accounts for the energy of the interfacial tensions between flows.

Drawing on the large body of research on two-phase flows [8–13], researchers have produced many theoretical and numerical studies on three-phase flows involving the effect of pairwise surface tensions [14–18]. In these models, the given pairwise surface tensions σ_{ij} are decomposed into three positive phase-specific surface-tension coefficients as

$$\sigma_{12} = \sigma_1 + \sigma_2, \quad \sigma_{13} = \sigma_1 + \sigma_3, \quad \sigma_{23} = \sigma_2 + \sigma_3,$$

whose existence is equivalent to the triangle inequality of the pairwise surface tensions. However, this decomposition encounters difficulties for cases in which $N \geq 4$, as the number of pairwise tensions would be greater than the number of phase-specific surface-tension coefficients, which leads to an overdetermined system [19,20]. In [17], a phenomenological continuum surface tension force was introduced by coupling Navier–Stokes equations through the mean curvature of the interface. Further, the generalization of this approach to an arbitrary number of phases with the purpose of avoiding the solvability issue was discussed in [19].

Generalizations of diffuse models to an arbitrary number of phases have recently been introduced and studied. In most of the existing models for multiphase flows, the phase variables are chosen specifically as concentrations of mixture c_i, whose sum is equal to 1. Examples of such models include N-phase Allen–Cahn equations [21,22] and N-phase Cahn–Hilliard equations [23–28]. A benefit of these models is that their consistency with the two-phase model can be easily proved. However, the pairwise surface tensions are not involved in the energy-density function so that the homogeneous surface tensions are implied in most of the existing models intrinsically. As the physical concentrations must belong to the $(N-1)$-dimensional Gibbs simplex [29], a variable Lagrangian multiplier should be introduced in the dynamic equations.

In order to incorporate the pairwise surface tensions into the phase field model, several generalized models have been proposed based on the generalized total free-energy functional. In [30], Elliott and Luckhaus set the total free-energy functional as

$$\mathcal{E}(\vec{c}) := \int_{\Omega} \left[\Psi(\vec{c}) + \frac{1}{2}(\mathbf{\Gamma}\nabla\vec{c}) : \nabla\vec{c} \right],$$

where $\mathbf{\Gamma}$ is the $N \times N$ symmetric-positive semi-definite matrix, i.e. a symmetric coefficient matrix is introduced in the capillary-energy term. They also gave a global existence result under constant mobility when $\mathbf{\Gamma} = \gamma \boldsymbol{I}$. Eyre [31] then studied this system and determined its equilibrium and dynamic behavior. Recently, Boyer and Minjeaud [32] proposed a generalization of the well-known two-phase Cahn–Hilliard model for the modeling of N-phase mixtures using the concentrations as the phase variables. Dong [33] established an algebraic relationship between the coefficient matrix and the pairwise surface tensions under a special choice of phase variables and gave the coupled system between the phase field and Navier–Stokes equations in the thermodynamics framework [34]. One main feature of these works is that, thanks to a relevant choice of free-energy, the model coincides exactly with the two-phase model. Dong then derived a formulation for the general phase variables in [35] by eliminating one variable in order to relax the algebraic relationship.

In this paper, we begin by applying the general phase variables without eliminating any of the variables, and we rebuild the relationship between the coefficient matrix and the pairwise surface tensions in a compact form. By drawing on a recent work on the close connection between the symmetric matrix space and simplex [36], we obtain the unisolvent property of the coefficient matrix on the tangent space of the solution manifold. Furthermore, the symmetric positive-definite (SPD) property on the tangent space proposed as an open problem in [32,33,35], is answered by two equivalent conditions from both the algebraic and the geometric point of view, see Theorem 2.3. We note that this property is fundamentally important to the well-posedness of the dynamic system. This is the first major contribution of the present study to the field.

The second principle contribution of the present study is the derivation of the N-phase Allen–Cahn and Cahn–Hilliard equations under the generalized total free-energy functional. As the gradient flow on the solution manifold, the Allen–Cahn equations make sense only under the given inner product on the tangent space in energy-variation framework. Here, we apply the inner product on the tangent space induced from the choice of generalized phase variables, so that the dynamics of the concentrations are independent of the choice of phase variables. A similar technique can be applied to N-phase Cahn–Hilliard equations to obtain the same property. When N-phase Allen–Cahn and Cahn–Hilliard equations are written in a strong formulation, the orthogonal projection to the tangent space will naturally translate into the variable Lagrangian multiplier as shown in models reported in [21,22,25–28]. This implies that our models can be viewed as a natural extension of the existing models while accounting for and including effect of pairwise surface tensions on the multiphase flows.

Based on the above properties, we propose finite element discretizations for N-phase models. The semi-implicit, fully-implicit, and modified Crank–Nicolson scheme, are considered for N-phase Allen–Cahn equations, and the semi-implicit, fully-implicit, and modified Crank–Nicolson scheme, are considered for N-phase Cahn–Hilliard equations. Each of these schemes can be viewed as a natural extension of the existing numerical schemes for two-phase flows [37,13]. The discrete energy law of the numerical schemes is also discussed.