A PRACTICALLY UNCONDITIONALLY GRADIENT STABLE SCHEME FOR THE N-COMPONENT CAHN–HILLIARD SYSTEM

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ABSTRACT

We present a practically unconditionally gradient stable conservative nonlinear numerical scheme for the N-component Cahn–Hilliard system modeling the phase separation of an N-component mixture. The scheme is based on a nonlinear splitting method and is solved by an efficient and accurate nonlinear multigrid method. And the scheme allows us to convert the N-component Cahn–Hilliard system into a system of $N-1$ binary Cahn–Hilliard equations and significantly reduces the required computer memory and CPU time. We observe that our numerical solutions are consistent with the linear stability analysis results. We also demonstrate the efficiency of the proposed scheme with various numerical experiments.

INTRODUCTION

The Cahn–Hilliard (CH) equation is the prototypical continuum model of phase separation in a binary alloy. It was originally derived by Cahn and Hilliard [1] to describe spinodal decomposition and has been widely adopted to model many other physical phenomena such as contact angle and wetting problem [2], mixing [3], pinchoff of liquid-liquid jets [4], Rayleigh–Taylor instability [5], and solid tumor growth [6]. Most of the technologically important alloys are multi-component systems exhibiting multiple phases in their microstructures. Moreover, one or more of these phases are formed as a result of phase transformations induced during processing. Since performance of these multi-component alloys depends crucially on the morphology of the phases, a fundamental understanding of the kinetics of phase transformations is important for controlling the microstructures of these multi-phase alloys [7].

One of the main difficulties in solving the CH system numerically is that the system is fourth-order in space which makes difference stencils very large and introduces a severe time step restriction for stability, i.e., $\Delta t \sim (\Delta x)^4$ for explicit methods. And there is a nonlinearity at the lower order spatial derivatives which can also contribute to numerical stability. In numerous papers there are stability restrictions on the time step which cause huge computational costs and make the calculation very inefficient. Therefore we need a scheme that allows the use of a sufficiently large time step without the technical limitations. But, though such an algorithm allows the use of a sufficiently large time step, it seems to be less attractive, because we need to invert an $(N-1) \times (N-1)$ matrix to obtain the solution of N-component CH system (except for the explicit method) and the matrix inversion becomes more and more complicated for increasing N. This problem was shown in previous papers. The authors in [8] developed

a nonlinear multigrid method to efficiently solve the discrete N-component CH system at the implicit time level, but an iteration step for the nonlinear multigrid method consists of a $2(N-1) \times 2(N-1)$ matrix inversion.

In this paper, we present a practically unconditionally gradient stable conservative numerical method for solving the CH system representing a model for phase separation in an N-component mixture. This method allows us to solve the N-component CH system in a decoupled way and significantly reduces the CPU time and memory requirements.

GOVERNING EQUATIONS

We consider the evolution of the N-component CH system on a domain $\Omega \subset \mathbb{R}^d$, $d = 1, 2, 3$. Let $\mathbf{c} = (c_1, \ldots, c_N)$ be the phase variables (i.e., the mole fractions of different components). Clearly the total mole fractions must sum to 1, i.e., $c_1 + \cdots + c_N = 1$, so that, admissible states will belong to the Gibbs N-simplex

$$G := \left\{ \mathbf{c} \in \mathbb{R}^N \;\middle|\; \sum_{i=1}^{N} c_i = 1,\ 0 \le c_i \le 1 \right\}. \tag{1}$$

Without loss of generality, we postulate that the free energy can be written as follows:

$$\mathcal{F}(\mathbf{c}) = \int_\Omega \left(F(\mathbf{c}) + \frac{\epsilon^2}{2} \sum_{i=1}^{N} |\nabla c_i|^2 \right) d\mathbf{x},$$

where $F(\mathbf{c}) = 0.25 \sum_{i=1}^{N} c_i^2 (1 - c_i)^2$ and $\epsilon > 0$ is the gradient energy coefficient. The natural boundary condition for the N-component CH system is the zero Neumann boundary condition:

$$\nabla c_i \cdot \mathbf{n} = 0 \quad \text{on } \partial\Omega,$$

where $\mathbf{n}$ is the unit normal vector to $\partial\Omega$. The time evolution of $\mathbf{c}$ is governed by the gradient of the energy with respect to the $\dot{H}^{-1}$ inner product under the additional constraint (1). This constraint has to hold everywhere at any time. In order to ensure this last constraint, we use a variable Lagrangian multiplier $\beta(\mathbf{c})$ [9]. The time dependence of c_i is given by the following CH equation:

$$\frac{\partial c_i}{\partial t} = M \Delta \mu_i, \tag{2}$$

$$\mu_i = f(c_i) - \epsilon^2 \Delta c_i + \beta(\mathbf{c}), \quad \text{for } i = 1, \ldots, N, \tag{3}$$

where M is a mobility, $f(c) = c(c - 0.5)(c - 1)$, and $\beta(\mathbf{c}) = -\frac{1}{N} \sum_{i=1}^{N} f(c_i)$. We take $M \equiv 1$ for convenience. The mass conserving boundary condition for the N-component CH system is

$$\nabla \mu_i \cdot \mathbf{n} = 0 \quad \text{on } \partial\Omega.$$

NUMERICAL SOLUTION

The numerical solution of the N-component CH system uses a second-order accurate spatial discretization and a nonlinear splitting time stepping method. Note that we only need to solve equations with $c_1, c_2, \ldots, c_{N-1}$ since $c_N = 1 - c_1 - c_2 - \cdots - c_{N-1}$ for the N-component CH system. Let $\mathbf{c} = (c_1, c_2, \ldots, c_{N-1})$ and $\boldsymbol{\mu} = (\mu_1, \mu_2, \ldots, \mu_{N-1})$.

Let $\Omega = (a,b) \times (c,d)$ be the computational domain in 2D, N_x and N_y be positive even integers, $h = (b-a)/N_x$ be the uniform mesh size, and $\Omega_h = \{(x_i, y_j) : x_i = (i-0.5)h,\ \ y_j = (j-0.5)h,\ \ 1 \le i \le N_x,\ \ 1 \le j \le N_y\}$ be the set of cell-centers. Let $\mathbf{c}_{ij}$ and $\boldsymbol{\mu}_{ij}$ be approximations of $\mathbf{c}(x_i, y_j)$ and $\boldsymbol{\mu}(x_i, y_j)$. We define $\mathbf{f}(\mathbf{c})$ and $\mathbf{1}$ to $\mathbf{f}(\mathbf{c}) = (f(c_1), f(c_2), \ldots, f(c_{N-1}))$ and $\mathbf{1} = (1, 1, \ldots, 1) \in \mathbb{R}^{N-1}$. We discretize Eqs. (2) and (3) in time by a nonlinear splitting algorithm:

$$\frac{\mathbf{c}_{ij}^{n+1} - \mathbf{c}_{ij}^{n}}{\Delta t} = \Delta_h \boldsymbol{\nu}_{ij}^{n+1} + \Delta_h \left(\beta(\mathbf{c}_{ij}^{n})\mathbf{1} - \frac{1}{4}\mathbf{c}_{ij}^{n} \right), \tag{4}$$

$$\boldsymbol{\nu}_{ij}^{n+1} = \boldsymbol{\varphi}(\mathbf{c}_{ij}^{n+1}) - \epsilon^2 \Delta_h \mathbf{c}_{ij}^{n+1},$$

where the nonlinear function $\boldsymbol{\varphi}(\mathbf{c}) = (\varphi_1(\mathbf{c}), \varphi_2(\mathbf{c}), \ldots, \varphi_{N-1}(\mathbf{c})) = \mathbf{f}(\mathbf{c}) + \mathbf{c}/4$.

In Eq. (4), the variable Lagrangian multiplier $\beta(\mathbf{c})$ is determined by the solutions at time level n. By treating $\beta(\mathbf{c})$ explicitly, there is no relation between the solutions at time level $n+1$. Thus the N-component CH system can be solved in a decoupled way, i.e.,

$$\frac{{c_k}_{,ij}^{n+1} - {c_k}_{,ij}^{n}}{\Delta t} = \Delta_h {\nu_k}_{,ij}^{n+1} + \Delta_h \left(\beta(\mathbf{c}_{ij}^{n}) - \frac{1}{4}{c_k}_{,ij}^{n} \right),$$

$$ {\nu_k}_{,ij}^{n+1} = \varphi({c_k}_{,ij}^{n+1}) - \epsilon^2 \Delta_h {c_k}_{,ij}^{n+1},$$

for $k = 1, 2, \ldots, N-1$. This means that we only solve the binary CH equation $N-1$ times to solve the N-component CH system. The above discrete system is solved by a nonlinear multigrid method [10].

NUMERICAL EXPERIMENTS

The stability of the proposed scheme

We investigate the stability of five different schemes for the quaternary CH system: the explicit Euler's (EE), semi-implicit Euler's (SIE), implicit Euler's (IE), Crank-Nicolson (CN), and nonlinear splitting (NS). For dissipative dynamics such as the CH system, a discrete time stepping algorithm is defined to be gradient stable if the free energy is nonincreasing, $\mathcal{F}(\mathbf{c}^{n+1}) \le \mathcal{F}(\mathbf{c}^{n})$, for each n. Define Δt_{max} as the largest possible time step which allows stable numerical computation. In other words, if the time step is larger than Δt_{max}, then the algorithm is not gradient stable. To measure Δt_{max} for each scheme, we perform a number of simulations for a sample initial problem on a set of increasingly finer grids. The initial conditions are

$$c_1(x, y, 0) = 0.25 + 0.1\text{rand}(\,),$$

$$c_2(x, y, 0) = 0.25 + 0.1\text{rand}(\,),$$

$$c_3(x, y, 0) = 0.25 + 0.1\text{rand}(\,)$$

on a domain $\Omega = (0,1) \times (0,1)$. Here, rand( ) is a random number between -1 and 1. The numerical solutions are computed on the uniform grids, $h = 1/2^n$ for $n = 5$, 6, 7, and 8. For each case, $\epsilon = 0.64h$ is used. The values of Δt_{max} with different schemes are listed in Table 1. From the results, we observe that EE, SIE, IE, and CN schemes are not gradient stable when we use the time step larger than Δt_{max}. However, our proposed scheme (NS) is gradient stable for time steps of any size, i.e., the scheme is practically unconditionally gradient stable.

Table 1 The values of the maximum time step guaranteeing the stability of each scheme.

Case	32×32	64×64	128×128	256×256
EE	7.6×10^{-5}	1.9×10^{-5}	4.7×10^{-6}	1.1×10^{-6}
SIE	2.9×10^{-2}	7.9×10^{-3}	1.3×10^{-3}	4.3×10^{-4}
IE	3.2×10^{-2}	7.8×10^{-3}	1.7×10^{-3}	4.4×10^{-4}
CN	2.0×10^{-2}	5.0×10^{-3}	1.1×10^{-3}	2.8×10^{-4}
NS	∞	∞	∞	∞

The efficiency of the proposed scheme

As mentioned in the previous section, we can solve the N-component CH system in a decoupled way by using our scheme. In order to show the efficiency of the proposed scheme, we consider phase separation of $N = 3, 4, \ldots, 10$ components in the unit square domain $\Omega = (0, 1) \times (0, 1)$. For each number of components, the initial condition is a randomly chosen superposition of circles. We choose $h = 1/128$, $\Delta t = 10h$, and $\epsilon = 0.0047$ and perform 4000 time steps. The evolution of the interface is shown in Fig. 1. Rows 1 and 2 correspond to $t = 10\Delta t$ and $4000\Delta t$, respectively. Table 2 provides the average CPU time (in seconds) during 4000 time steps for each number of components. The average CPU time versus number of components is shown in Fig. 2. The results suggest that the convergence rate of average CPU time is linear with respect to number of components.

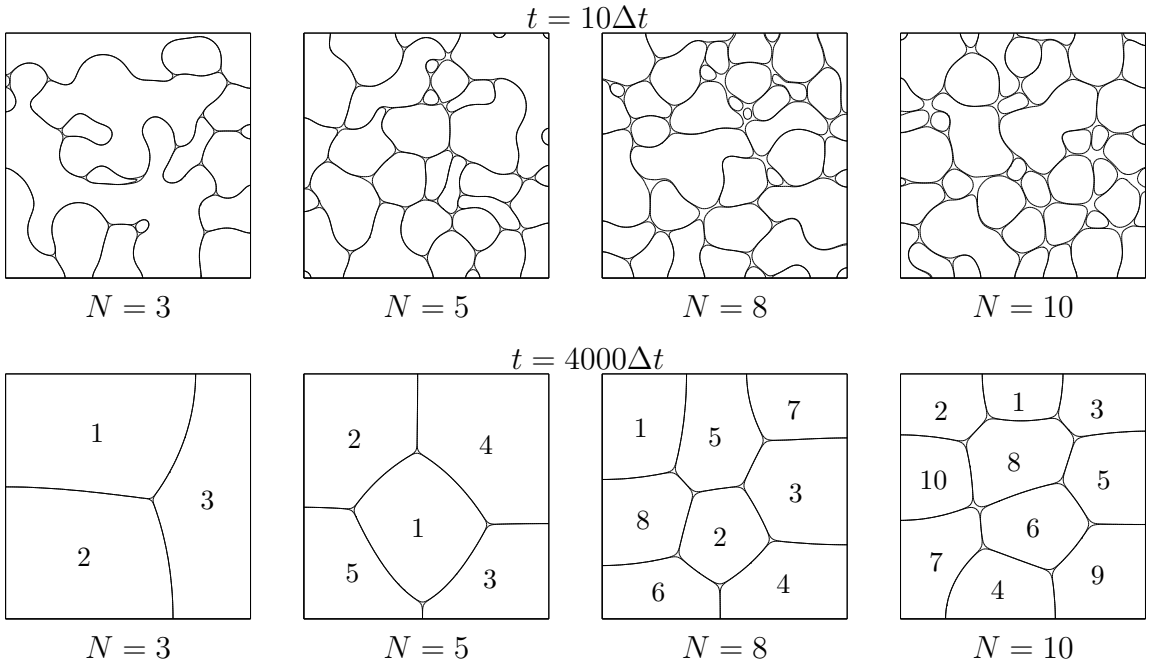

Figure 1. Phase separation of $N = 3$, 5, 8, and 10 components. Rows 1 and 2 correspond to $t = 10\Delta t$ and $4000\Delta t$, respectively. Numbers in row 2 indicate the number of components.

Table 2 Average CPU times ($sec.$) for different numbers of components.

N	3	4	5	6	7	8	9	10
Average CPU time	2.060	2.990	4.070	5.028	5.791	6.686	7.492	8.533

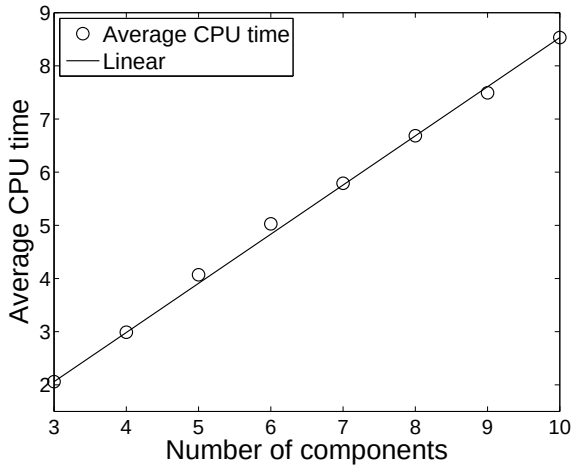

Figure 2. Average CPU time versus number of components.

The Rayleigh–Taylor instability of a five-component mixture

We consider multi-component incompressible viscous fluid flow. The fluid dynamics is described by the Navier–Stokes–Cahn–Hilliard (NSCH) equations [3–5,11–16]:

$$\rho(\mathbf{c})\left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{u}\right) = -\nabla p + \frac{1}{Re}\Delta\mathbf{u} + \frac{\rho(\mathbf{c})}{Fr^2}\mathbf{g}, \tag{5}$$

$$\nabla\cdot\mathbf{u} = 0, \tag{6}$$

$$\frac{\partial \mathbf{c}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{c} = \frac{1}{Pe}\Delta\boldsymbol{\mu}, \tag{7}$$

$$\boldsymbol{\mu} = \mathbf{f}(\mathbf{c}) - \epsilon^2\Delta\mathbf{c} + \beta(\mathbf{c}), \tag{8}$$

where $\mathbf{g} = (0, -1)$. By applying our scheme, we can solve the multi-component advective CH system (7) and (8) in a decoupled way and solving the multi-component NSCH system (5)-(8) becomes solving the binary NSCH system. For a detailed description of the numerical method used in solving the binary NSCH system, please refer to Ref. [5].

To model the Rayleigh–Taylor instability of a five-component mixture, we take an initial state as shown in Fig. 3 (a). The initial velocity is zero. $\rho(\mathbf{c}) = \sum_{i=1}^{5}\rho_i c_i$ ($c_5 = 1 - c_1 - c_2 - c_3 - c_4$ and ρ_i is the ith fluid density) and $\rho_i = 6 - i$ for $i = 1, \ldots, 5$. A mesh size 128×512 is used on a domain $\Omega = (0, 1)\times(0, 4)$ and we choose $\Delta t = 2.0\times10^{-3}$, $\epsilon = 0.0047$, $Re = 3000$, and $Pe = 0.1/\epsilon$. The results are presented in Figs. 3 (b)-(g). The area shown by black indicates the fluid 1 region, while the dark gray, gray, light gray, and white color regions stand for the fluid 2, 3, 4, and 5 domains, respectively. We observe that the gravity affects a multi-component simulation by pulling the heavy fluid to the bottom of the computational domain and our proposed method is a powerful tool to simulate the Rayleigh–Taylor instability between multi-component fluids.

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(a) (b) (c) (d) (e) (f) (g)

Figure 3. The Rayleigh–Taylor instability of a five-component mixture. Times are $t = 0$, 3, 6, 10, 30, 50, and 200 (left to right).

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