A NUMERICAL METHOD FOR THE QUATERNARY CAHN-HILLIARD SYSTEM

Junseok Kim [1]

1) *Department of Mathematics, Dongguk University, Seoul 100-715, KOREA*

Corresponding Author : Junseok Kim, cfdkim@dongguk.edu

ABSTRACT

We consider a second-order conservative nonlinear multigrid method for the quaternary Cahn-Hilliard system of a model for phase separation in a quaternary mixture. First, the standard finite difference approximation in spatial discretization and the Crank-Nicholson semi-implicit scheme in temporal one are used. Then, the resulting discretized equations are solved by an efficient nonlinear multigrid method. We perform standard test problems to demonstrate the accuracy, flexibility, and robustness of this method.

INTRODUCTION

The purpose of this work is to consider a conservative nonlinear multigrid method of the quaternary Cahn-Hilliard (CH) system for four component mixture, occupying a domain $\Omega \subset \mathbb{R}^d$ $(d = 1, 2, 3)$. When a homogeneous system composed of four components, at high temperature, is rapidly cooled to a uniform temperature below a critical temperature, where it is unstable with respect to concentration fluctuations, spinodal decomposition takes place: the system separates into spatial regions rich in one component and poor in the other components and evolves into an equilibrium state with lower overall free energy [1].

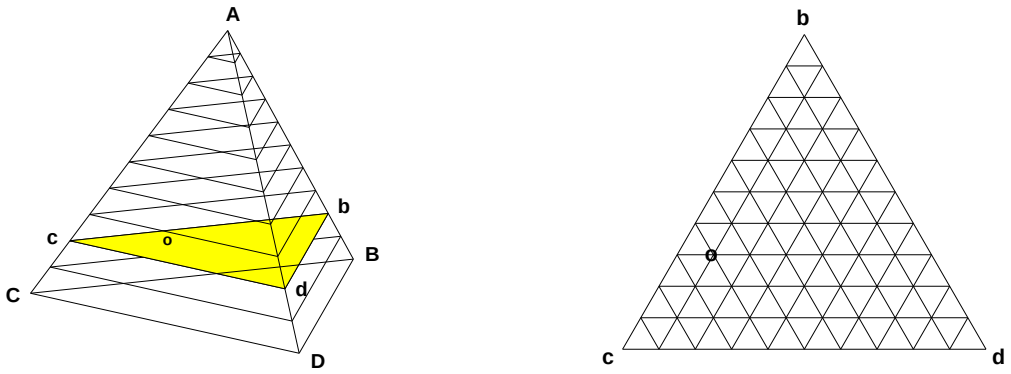
GOVERNING EQUATIONS

The composition of a quaternary mixture (A, B, C, and D) can be mapped onto an equilateral tetrahedron (the Gibbs simplex) whose corners represent 100% concentration of A, B, C or D as shown in Fig. 1. Mixtures with components lying on planes parallel to the triangle, ΔBCD contain the same percentage of A, those with planes parallel to the triangle, ΔCDA have the same percentage of B concentration, and analogously for the C and the D concentrations. In Fig. 1, the mixture at the position marked '$\circ$' contains 20% A, $0.8 \times 30\%$ B, $0.8 \times 60\%$C and $0.8 \times 10\%$ D. Clearly the total percentage must sum to 100%, or expressed as mole fraction

$$c_1 + c_2 + c_3 + c_4 = 1, \tag{1}$$

so that, admissible states will belong to the Gibbs simplex

$$S := \left\{ (c_1, c_2, c_3, c_4) \in \mathbb{R}^4 \left| \sum_{i=1}^{4} c_i = 1,\ 0 \le c_i \le 1 \right. \right\}. \tag{2}$$



(a) (b)

Figure 1. Gibbs tetrahedron.

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

$$\mathcal{F} = \int_\Omega \left[F(c_1, c_2, c_3, c_4) + \frac{\epsilon^2}{4} \sum_{i=1}^{4} |\nabla c_i|^2 \right] d\mathbf{x}, \tag{3}$$

where $F(c_1, c_2, c_3, c_4) = \frac{1}{8} \sum_{i=1}^{4} c_i^2 (1-c_i)^2$ and Ω is an open, bounded subset of $\mathbf{R}^n (n = 1, 2, 3)$ occupied by the system. Notice that the coefficients $\frac{1}{8}$ and $\frac{1}{4}$ are natural in view of the binary case. If there is no fluid components of the third and the fourth, the free energy, Eq. (3), reduces to the binary case [2],

$$\mathcal{F} = \int_\Omega \left(\frac{1}{4} c_1^2 (1 - c_1)^2 + \frac{\epsilon^2}{2} |\nabla c_1|^2 \right) d\mathbf{x}, \tag{4}$$

where we used the relation, $c_2 = 1 - c_1$. Let $\mathbf{c} = (c_1, c_2, c_3)$ be the phase variable (i.e. concentration). The time evolution of $\mathbf{c}$ is governed by the gradient of the energy with respect to the H^{-1} inner product under the additional constraint (1), which has to hold everywhere at any time. In order to ensure this last constraint, we use a Lagrangian multiplier $\beta(\mathbf{c})$. The system of equations is written for $i = 1, 2, 3, 4$

$$\frac{\partial c_i}{\partial t} = M \Delta \mu_i, \tag{5}$$

$$\mu_i = \frac{\delta \mathcal{F}}{\delta c_i} + \beta(\mathbf{c}) = \frac{\partial F(\mathbf{c})}{\partial c_i} - \epsilon^2 \Delta c_i + \beta(\mathbf{c}). \tag{6}$$

To calculate $\beta(\mathbf{c})$, we write the equation satisfied by $S = c_1 + c_2 + c_3 + c_4$, and we want $S \equiv 1$ to be a solution to this equation.

$$\frac{\partial S}{\partial t} = M \Delta \left(\sum_{i=1}^{4} \frac{\delta \mathcal{F}}{\delta c_i} + 4\beta(\mathbf{c}) \right). \tag{7}$$

Therefore, $\beta(\mathbf{c}) = -\frac{1}{4} \sum_{i=1}^{4} \frac{\delta \mathcal{F}}{\delta c_i} = -\frac{1}{4} \sum_{i=1}^{4} \frac{\partial F}{\partial c_i}$. The natural and mass conserving boundary conditions for the quaternary CH system are the zero Neumann boundary ones:

$$\nabla c_i \cdot \mathbf{n} = \nabla \mu_i \cdot \mathbf{n} = 0 \text{ on } \partial\Omega, \tag{8}$$

where $\mathbf{n}$ is the unit normal vector to $\partial\Omega$.

NUMERICAL SOLUTION

We discretize Eqs. (5) and (6) in time by the Crank-Nicholson algorithm:

$$\frac{\mathbf{c}_{ij}^{n+1} - \mathbf{c}_{ij}^{n}}{\Delta t} = M\Delta_d \boldsymbol{\mu}_{ij}^{n+\frac{1}{2}}, \tag{9}$$

$$\boldsymbol{\mu}_{ij}^{n+\frac{1}{2}} = \frac{1}{2}(\mathbf{f}(\mathbf{c}_{ij}^{n+1}) + \mathbf{f}(\mathbf{c}_{ij}^{n})) - \frac{1}{2}\Delta_d(\mathbf{c}_{ij}^{n+1} + \mathbf{c}_{ij}^{n}). \tag{10}$$

In this section, we develop a nonlinear Full Approximation Storage (FAS) multigrid method to solve the nonlinear discrete system at the implicit time level. The nonlinearity, $\mathbf{f}(\mathbf{c})$, is treated using one step of Newton's iteration and a pointwise Gauss-Seidel relaxation scheme is used as the smoother in the multigrid method. Let us rewrite equations (9)-(10) as follows.

$$N(\mathbf{c}^{n+1}, \boldsymbol{\mu}^{n+\frac{1}{2}}) = (\boldsymbol{\phi}^n, \boldsymbol{\psi}^n), \tag{11}$$

where the nonlinear system operator (N) is defined as

$$N(\mathbf{c}^{n+1}, \boldsymbol{\mu}^{n+\frac{1}{2}}) = \left(\frac{\mathbf{c}^{n+1}}{\Delta t} - M\Delta_d \boldsymbol{\mu}^{n+\frac{1}{2}},\ \boldsymbol{\mu}^{n+\frac{1}{2}} - \frac{1}{2}\mathbf{f}(\mathbf{c}^{n+1}) + \frac{\epsilon^2}{2}\Delta_d \mathbf{c}^{n+1}\right), \tag{12}$$

$$(\boldsymbol{\phi}^n, \boldsymbol{\psi}^n) = \left(\frac{\mathbf{c}^{n}}{\Delta t} - (\mathbf{u}\cdot\nabla_d \mathbf{c})^n,\ \frac{1}{2}\mathbf{f}(\mathbf{c}^{n}) - \frac{\epsilon^2}{2}\Delta_d \mathbf{c}^{n}\right). \tag{13}$$

Given the number ν of pre- and post- smoothing relaxation sweeps, an iteration step for the nonlinear multigrid method using the V-cycle is formally written as follows:

FAS multigrid cycle

$$\{\mathbf{c}_k^{m+1}, \boldsymbol{\mu}_k^{m+\frac{1}{2}}\} = FAScycle(k, \mathbf{c}_k^{m}, \boldsymbol{\mu}_k^{m-\frac{1}{2}}, N_k, \boldsymbol{\phi}_k^n, \boldsymbol{\psi}_k^n, \nu) \text{ on } \Omega_k \text{ grid.} \tag{14}$$

We set the initial guess, $\mathbf{c}_k^0 = \mathbf{c}_k^n$ and $\boldsymbol{\mu}_k^{-\frac{1}{2}} = \boldsymbol{\mu}_k^{n-\frac{1}{2}}$. Now, define the FAScycle.

Step 1 - Presmoothing:

$$\{\bar{\mathbf{c}}_k^m,\ \bar{\boldsymbol{\mu}}_k^{m-\frac{1}{2}}\} = SMOOTH^{\nu}(\mathbf{c}_k^m, \boldsymbol{\mu}_k^{m-\frac{1}{2}}, N_k, \boldsymbol{\phi}_k^n, \boldsymbol{\psi}_k^n) \text{ on } \Omega_k \text{ grid.} \tag{15}$$

One $SMOOTH$ relaxation operator step consists of solving the system (16)-(19) given below by 6×6 matrix inversion for each ij. First, let's discretize Eq. (9).

$$\frac{\bar{\mathbf{c}}_{ij}^m}{\Delta t} + \frac{4M}{h^2}\bar{\boldsymbol{\mu}}_{ij}^{m-\frac{1}{2}} = \boldsymbol{\phi}_{ij}^n + \frac{M}{h^2}(\boldsymbol{\mu}_{i+1,j}^{m-\frac{1}{2}} + \bar{\boldsymbol{\mu}}_{i-1,j}^{m-\frac{1}{2}} + \boldsymbol{\mu}_{i,j+1}^{m-\frac{1}{2}} + \bar{\boldsymbol{\mu}}_{i,j-1}^{m-\frac{1}{2}}). \tag{16}$$

Next, let's discretize Eq. (10). Since $\mathbf{f}(\mathbf{c}_{ij}^{n+1})$ is nonlinear with respect to $\mathbf{c}_{ij}^{n+1}$, we linearize $\mathbf{f}(\mathbf{c}_{ij}^{n+1})$ at $\mathbf{c}_{ij}^{m}$, i.e.,

$$\mathbf{f}(\mathbf{c}_{ij}^{n+1}) \approx \mathbf{f}(\mathbf{c}_{ij}^{m}) + (\bar{\mathbf{c}}_{ij}^m - \mathbf{c}_{ij}^m)\frac{\partial \mathbf{f}(\mathbf{c}_{ij}^m)}{\partial \mathbf{c}}, \tag{17}$$

where

$$\frac{\partial \mathbf{f}(\mathbf{c}_{ij}^m)}{\partial \mathbf{c}} = \begin{pmatrix} \frac{\partial f_1}{\partial c_1}(\mathbf{c}_{ij}^m) & \frac{\partial f_2}{\partial c_1}(\mathbf{c}_{ij}^m) & \frac{\partial f_3}{\partial c_1}(\mathbf{c}_{ij}^m) \\ \frac{\partial f_1}{\partial c_2}(\mathbf{c}_{ij}^m) & \frac{\partial f_2}{\partial c_2}(\mathbf{c}_{ij}^m) & \frac{\partial f_3}{\partial c_2}(\mathbf{c}_{ij}^m) \\ \frac{\partial f_1}{\partial c_3}(\mathbf{c}_{ij}^m) & \frac{\partial f_2}{\partial c_3}(\mathbf{c}_{ij}^m) & \frac{\partial f_3}{\partial c_3}(\mathbf{c}_{ij}^m) \end{pmatrix}. \tag{18}$$

$$-\bar{\mathbf{c}}_{ij}^m\left(\frac{\partial \mathbf{f}(\mathbf{c}_{ij}^m)}{2\partial \mathbf{c}}+\frac{2\epsilon^2}{h^2}\right)+\bar{\boldsymbol{\mu}}_{ij}^{m-\frac{1}{2}}=\boldsymbol{\psi}_{ij}^n+\frac{1}{2}\mathbf{f}(\mathbf{c}_{ij}^m)-\mathbf{c}_{ij}^m\frac{\partial \mathbf{f}(\mathbf{c}_{ij}^m)}{2\partial \mathbf{c}}$$
$$-\frac{\epsilon^2}{2h^2}(\mathbf{c}_{i+1,j}^m+\bar{\mathbf{c}}_{i-1,j}^m+\mathbf{c}_{i,j+1}^m+\bar{\mathbf{c}}_{i,j-1}^m). \tag{19}$$

Step 2 - Compute the defect: $(\overline{\mathbf{def}}_{1\ k}^m, \overline{\mathbf{def}}_{2\ k}^m)=(\boldsymbol{\phi}_k^n,\boldsymbol{\psi}_k^n)-N_k(\bar{\mathbf{c}}_k^m,\bar{\boldsymbol{\mu}}_k^{m-\frac{1}{2}})$. *Step 3 - Restrict the defect and* $\{\bar{\mathbf{c}}_k^m,\ \bar{\boldsymbol{\mu}}_k^{m-\frac{1}{2}}\}$:

$$(\overline{\mathbf{def}}_{1\ k-1}^m, \overline{\mathbf{def}}_{2\ k-1}^m, \bar{\mathbf{c}}_{k-1}^m, \bar{\boldsymbol{\mu}}_{k-1}^{m-\frac{1}{2}})=I_k^{k-1}(\overline{\mathbf{def}}_{1\ k}^m, \overline{\mathbf{def}}_{2\ k}^m, \bar{\mathbf{c}}_k^m, \bar{\boldsymbol{\mu}}_k^{m-\frac{1}{2}}). \tag{20}$$

The restriction operator I_k^{k-1} maps k-level functions to $(k-1)$-level functions. That is, coarse grid values are obtained by averaging the four nearby fine grid values.

Step 4 - Compute the right-hand side:

$$(\boldsymbol{\phi}_{k-1}^n,\boldsymbol{\psi}_{k-1}^n)=(\overline{\mathbf{def}}_{1\ k-1}^m, \overline{\mathbf{def}}_{2\ k-1}^m)+N_{k-1}(\bar{\mathbf{c}}_{k-1}^m,\bar{\boldsymbol{\mu}}_{k-1}^{m-\frac{1}{2}}). \tag{21}$$

Step 5 - Compute an approximate solution $\{\hat{\mathbf{c}}_{k-1}^m,\ \hat{\boldsymbol{\mu}}_{k-1}^{m-\frac{1}{2}}\}$ *of the coarse grid equation on* Ω_{k-1}*:*

$$N_{k-1}(\mathbf{c}_{k-1}^m,\boldsymbol{\mu}_{k-1}^{m-\frac{1}{2}})=(\boldsymbol{\phi}_{k-1}^n,\boldsymbol{\psi}_{k-1}^n). \tag{22}$$

If $k=1$, we explicitly invert a 4×4 matrix to obtain the solution. If $k>1$, we solve (22) by performing a FAS k-grid cycle using $\{\bar{\mathbf{c}}_{k-1}^m,\ \bar{\boldsymbol{\mu}}_{k-1}^{m-\frac{1}{2}}\}$ as an initial approximation:

$$\{\hat{\mathbf{c}}_{k-1}^m,\hat{\boldsymbol{\mu}}_{k-1}^{m-\frac{1}{2}}\}=\text{FAScycle}(k-1,\bar{\mathbf{c}}_{k-1}^m,\bar{\boldsymbol{\mu}}_{k-1}^{m-\frac{1}{2}},N_{k-1},\boldsymbol{\phi}_{k-1}^n,\boldsymbol{\psi}_{k-1}^n,\nu). \tag{23}$$

Step 6 - Compute the coarse grid correction (CGC):

$$\hat{\mathbf{v}}_{k-1}^m=\hat{\mathbf{c}}_{k-1}^m-\bar{\mathbf{c}}_{k-1}^m,\quad \hat{\mathbf{w}}_{k-1}^{m-\frac{1}{2}}=\hat{\boldsymbol{\mu}}_{k-1}^{m-\frac{1}{2}}-\bar{\boldsymbol{\mu}}_{k-1}^{m-\frac{1}{2}}. \tag{24}$$

Step 7 - Interpolate the correction: $(\hat{\mathbf{v}}_k^m,\hat{\mathbf{w}}_k^{m-\frac{1}{2}})=I_{k-1}^k(\hat{\mathbf{v}}_{k-1}^m,\hat{\mathbf{w}}_{k-1}^{m-\frac{1}{2}})$.
Step 8 - Compute the corrected approximation on Ω_k*:*

$$\mathbf{c}_k^{m,\text{ after }CGC}=\bar{\mathbf{c}}_k^m+\hat{\mathbf{v}}_k^m,\quad \boldsymbol{\mu}_k^{m-\frac{1}{2},\text{ after }CGC}=\bar{\boldsymbol{\mu}}_k^{m-\frac{1}{2}}+\hat{\mathbf{w}}_k^{m-\frac{1}{2}} \tag{25}$$

Step 9 - Postsmoothing:

$$\{\mathbf{c}_k^{m+1},\boldsymbol{\mu}_k^{m+\frac{1}{2}}\} \tag{26}$$
$$=SMOOTH^\nu(\mathbf{c}_k^{m,\text{ after }CGC},\boldsymbol{\mu}_k^{m-\frac{1}{2},\text{ after }CGC},N_k,\boldsymbol{\phi}_k^n,\boldsymbol{\psi}_k^n)\text{ on }\Omega_k\text{ grid}. \tag{27}$$

This completes the description of a nonlinear FAScycle.

REFERENCES

1. Copetti, M.I.M., "Numerical experiments of phase separation in ternary mixtures", *Math. Comput. Simulation*, Vol. 52, 2000, pp. 41-51.
2. Kim, J.S., "A continuous surface tension force formulation for diffuse-interface models", *J. Comp. Phys.*, Vol 204, 2005, pp. 784-804.