LINEARLY DECOUPLED ENERGY-STABLE NUMERICAL METHODS FOR MULTICOMPONENT TWO-PHASE COMPRESSIBLE FLOW*

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Abstract. In this paper, for the first time we propose two linear, decoupled, energy-stable numerical schemes for multicomponent two-phase compressible flow with a realistic equation of state (e.g., Peng–Robinson equation of state). The methods are constructed based on the scalar auxiliary variable (SAV) approaches for Helmholtz free energy and the intermediate velocities that are designed to decouple the tight relationship between velocity and molar densities. The intermediate velocities are also involved in the discrete momentum equation to ensure consistency with the mass balance equations. Moreover, we propose a componentwise SAV approach for a multicomponent fluid, which requires solving a sequence of linear, separate mass balance equations. The fully discrete schemes are also constructed based on the finite difference/volume methods with the upwind scheme on staggered grids. We prove that the semidiscrete and fully discrete schemes preserve the unconditional energy-dissipation feature. Numerical results are presented to verify the effectiveness of the proposed methods.

Keywords. multicomponent two-phase flow, diffuse interface model, energy stability, realistic equation of state

AMS subject classifications. 65N12, 76T10, 49S05

DOI. 10.1137/17M1162287

1. Introduction. It is a very important issue to simulate subsurface flow and transport processes [8, 11, 27, 37, 38, 40, 47, 48, 49], especially multicomponent two-phase compressible fluid systems with a realistic equation of state (e.g., Peng–Robinson equation of state [41]). It has a wide range of applications in chemical and reservoir engineering [20, 21, 22, 23, 24, 25, 40, 43], especially pore-scale modeling of subsurface fluid flow, including shale gas reservoirs. The classical models of incompressible two-phase flows or compositional flows have been extensively studied and employed [3, 5, 15, 27, 37], the primal state variables of which are often pressure, temperature, and chemical composition. Although the classical models have been widely used, they suffer from a few essential limitations, as pointed out in [34, 40]; for example, it is required to construct a pressure equation since there is no intrinsic pressure equation [40].

An alternative modeling framework, which uses the moles, volume, and temperature (the so-called NVT-based framework) as the primal state variables, has been intensively studied recently [20, 21, 22, 23, 24, 25, 26, 40, 43]. The NVT-based modeling framework originates from the phase-splitting calculations of multicomponent fluids at specified moles, volume, and temperature [33, 34, 39]. Very recently, in the NVT-based framework, a general multicomponent two-phase compressible flow model

*Received by the editors December 20, 2017; accepted for publication (in revised form) September 5, 2018; published electronically November 15, 2018.
http://www.siam.org/journals/sinum/56-6/M116228.html

Funding: This work was supported by funding from King Abdullah University of Science and Technology (KAUST) through grants BAS/1/1351-01, URF/1/2993-01, and REP/1/2879-01.
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was rigorously derived by Kou and Sun in [29] based on the thermodynamic laws and realistic equations of state (e.g., Peng–Robinson equation of state). This model has at least three important features that are distinguished from the classical models:

- It has thermodynamically consistent unified formulations for general average velocities and mass diffusion fluxes.
- It uses diffusive interfaces and realistic equations of state, thereby being capable of characterizing the fluid compressibility and partial miscibility between different phases.
- It uses a general thermodynamic pressure, which is a function of molar densities and temperature, and consequently it is never required to construct the pressure equation.

In addition, another formulation of the momentum conservation equation, which is convenient for numerical simulation, was derived in [29] by the relation between the pressure gradient and chemical potential gradients. In this paper, we consider how to develop and analyze efficient numerical methods for this model problem.

A key challenge in numerical simulation of diffuse interface models is to construct efficient numerical schemes preserving the discrete energy-dissipation law [6, 45]. In constructing energy-stable numerical schemes for a multicomponent two-phase compressible flow model, there are at least two main difficulties: One is the strong nonlinearity of bulk Helmholtz free energy density; the other is the tight coupling relationship between molar densities and flow velocity through the convection term in the mass balance equations and the stress force arising from chemical potential gradients in the momentum balance equation. An energy-dissipation numerical scheme was developed in [29] based on a convex-concave splitting of Helmholtz free energy density, but it leads to a nonlinear and coupled system of the mass balance equations and momentum balance equation. In this paper, we focus on the linear, decoupled, energy-stable numerical schemes.

Recently, for incompressible two-phase flows, a decoupled approach [35] was developed by introducing an intermediate velocity in the phase equation to resolve the coupling relation between the velocity and phase function, and this technique was used to construct linear, decoupled, efficient numerical methods for phase-field models of incompressible two-phase flows [6, 45]. When applying this technique to the compressible multicomponent two-phase flow model considered in this paper, we encounter two challenging problems: The first is how to construct the intermediate velocities since the stress force in the momentum balance equation is different from phase-field models; the second is how to treat the momentum balance equation using intermediate velocities. We run into the second problem because at the time-discrete level, the velocity variable in the convection term of the momentum balance equation shall be consistent with the intermediate velocities when we combine the mass balance equation of each component and the momentum balance equation to derive the variation of the kinetic energy. In this work, we will construct two intermediate velocities, both of which can uncouple the relationship between velocity and molar densities; we will also propose a discrete formulation of the momentum balance equation, which involves the intermediate velocities and is consequently consistent with the mass balance equations. One of the introduced intermediate velocities is for the first time defined in a componentwise way, and thus it is very efficient for a special multicomponent fluid.

There have been at least four approaches in the literature dealing with the bulk Helmholtz free energy density derived from Peng–Robinson equation of state for constructing energy-stable numerical schemes. The first approach is the convex splitting...
method [10, 13], which has been popularly used in phase-field models [2, 13, 45, 52]. The energy-stable numerical scheme based on the convex splitting method was also developed and analyzed for the diffuse-interface models with Peng–Robinson equation of state [14, 25, 28, 29, 30, 42, 43]. The second approach is a modified Newton’s method with a relaxation parameter that is dynamically chosen to ensure the energy decay property [23]. The third approach is a fully implicit unconditionally stable scheme [24], which uses the difference of Helmholtz free energy density to approximate the chemical potential. The fourth numerical scheme was developed in [31] based on the invariant energy quadratization (IEQ) approach that is a novel, efficient method and has been applied intensively to many phase-field models recently [53, 54, 55]. Even more recently, a scalar auxiliary variable (SAV) approach [46] was built upon the IEQ approach, leading to unconditionally stable numerical schemes, which only need to solve the linear equations with constant coefficients at each time step. In this paper, we will apply the SAV approach to treat the mass balance equations and construct linear, unconditionally stable numerical schemes. Moreover, we will develop a componentwise SAV approach for a multicomponent flow model, which uncouples the relationships between multiple components and allows us to solve each component mass balance equation separately. The schemes for gradient flows of multiple functions in [46] usually require the computation of eigenmatrix and eigenvalues to achieve the decoupled forms, but this computation cost is free for the proposed componentwise SAV approach. So the proposed scheme is efficient and easy to implement for the case of multiple components.

We note that the proposed numerical schemes for multicomponent two-phase flows are perfect combinations of the above intermediate velocity approaches and SAV approaches, which lead to a sequence of linearly decoupled equations. The proposed time-discrete schemes are proved to be unconditionally energy stable.

The proposed semidiscrete time schemes are further extended to the fully discrete schemes based on the finite difference/volume methods with the upwind scheme on staggered grids [19, 50], also known as marker-and-cell (MAC) schemes [12, 16, 17, 32, 44], and can be equivalent to special mixed finite element methods under specified quadrature rules [1, 18]. Although the finite difference/volume methods on staggered grids have been widely applied in computational fluid dynamics, there are relatively few studies on the related energy stability analysis, perhaps due to the complexity resulting from the use of different grids for different variables. The convergence of the MAC method for the Stokes and Navier–Stokes equations was rigorously proved in [12, 16, 17], and the superconvergence of the MAC scheme for the Stokes equations was studied in [32, 44]. The unconditionally energy-stable finite difference schemes were presented for the phase field crystal equation [52] and for the Cahn–Hilliard–Hele–Shaw equations [51], and afterwards an energy-stable finite difference scheme was developed in [6] for Cahn–Hilliard Navier–Stokes system. The energy-stability estimates of some fully discrete schemes for incompressible Navier–Stokes equations on staggered grids were analyzed recently in [4]. In this paper, the fully discrete schemes on staggered grids will be developed for multicomponent compressible flows, and the fully discrete energy stability will also be proved rigorously.

The rest of this paper is organized as follows. In section 2, we give a brief description of the multicomponent two-phase flow model. In section 3, we propose the semidiscrete time schemes and prove their unconditional energy stability, while the fully discrete schemes are presented and analyzed in section 4. In section 5, numerical tests are carried out to validate the proposed methods. Finally, concluding remarks are provided in section 6.
2. Mathematical model of multicomponent two-phase flow. In this section, we briefly introduce the mathematical model of multicomponent two-phase flow with Peng–Robinson equation of state, which was very recently proposed in [29].

We consider the motion of a mixture fluid composed of $M$ chemical components at a constant temperature. Let $n_\alpha$ be the molar density of component $\alpha$, and we denote the molar density vector by $n = [n_1, n_2, \ldots, n_M]^T$. The overall molar density of a mixture is denoted by $n = \sum_{\alpha=1}^M n_\alpha$. Let $M_{w,\alpha}$ denote the molar weight of component $\alpha$, and then we denote the mass density of component $\alpha$ by $\rho_\alpha = n_\alpha M_{w,\alpha}$ and denote the overall mass density by $\rho = \sum_{\alpha=1}^M \rho_\alpha$.

The mathematical model developed in [29] can employ any realistic equation of state, for instance, the van der Waals equation of state and Peng–Robinson equation of state (PR-EOS) [41]. PR-EOS has been widely applied in oil reservoir and chemical engineering due to its accuracy. In this work, we focus on the PR-EOS-based Helmholtz free energy density $f_b(n)$ of a homogeneous bulk fluid, which has the form

\begin{equation}
(2.1) \quad f_b(n) = f_b^\text{ideal}(n) + f_b^\text{repulsion}(n) + f_b^\text{attraction}(n),
\end{equation}

where $f_b^\text{ideal}$, $f_b^\text{repulsion}$, and $f_b^\text{attraction}$ are formulated in Appendix A.

Diffuse interfaces always occur between multiple phases of a realistic fluid. To characterize this feature, a local density gradient contribution is added to the free energy density of an inhomogeneous fluid, and consequently the general Helmholtz free energy density (denoted by $f$) is expressed as

\begin{equation}
(2.2) \quad f(n) = f_b(n) + \frac{1}{2} \sum_{\alpha,\beta=1}^M c_{\alpha\beta} \nabla n_\alpha \cdot \nabla n_\beta,
\end{equation}

where $c_{\alpha\beta} \geq 0$ ($1 \leq \alpha, \beta \leq M$) are the cross influence parameters depending on temperature but independent of molar densities. The formulations of $c_{\alpha\beta}$ can be found in Appendix B. We assume that the influence parameter matrix $(c_{\alpha\beta})_{\alpha,\beta=1}^M$ is symmetric, and moreover it is positive definite or positive semidefinite.

The chemical potential of component $\alpha$ is defined as

\begin{equation}
(2.3) \quad \mu_\alpha(n) = \frac{\delta f(n)}{\delta n_\alpha} = \mu_\alpha^b(n) - \sum_{\beta=1}^M \nabla \cdot (c_{\alpha\beta} \nabla n_\beta), \quad \mu_\alpha^b(n) = \frac{\partial f_b(n)}{\partial n_\alpha},
\end{equation}

where $\frac{\delta}{\delta n_\alpha}$ denotes the variational derivative.

We now describe the governing equations. First, the mass balance equation for component $\alpha$ is

\begin{equation}
(2.4) \quad \frac{\partial n_\alpha}{\partial t} + \nabla \cdot (un_\alpha) + \nabla \cdot J_\alpha = 0,
\end{equation}

where $u$ is a reference velocity and $J_\alpha$ is the diffusion flux of component $\alpha$. In general, we can express the diffusion flux of component $\alpha$ as [7, 26, 29]

\begin{equation}
(2.5) \quad J_\alpha = -\sum_{\beta=1}^M \mathcal{M}_{\alpha\beta} \nabla \mu_\beta, \quad \alpha = 1, \ldots, M,
\end{equation}

where $\mathcal{M} = (\mathcal{M}_{\alpha\beta})_{\alpha,\beta=1}^M$ is the mobility tensor that shall be symmetric and at least positive semidefinite so that Onsager’s reciprocal principle [9] and the second law of thermodynamics are satisfied. Three choices of $\mathcal{M}$ are provided in [29] and shown below.
(A1) The first mobility choice is to take $\bfscrM$ as a diagonal positive definite matrix with diagonal elements
\begin{equation}
M_\alpha (n) = M_{\alpha\alpha} (n) = \frac{D_\alpha n_\alpha}{RT},
\end{equation}
where $R$ stands for the universal gas constant and $D_\alpha > 0$ is the diffusion coefficient of component $\alpha$. The diffusion flux has the form [7, 25] $J_\alpha = -\frac{D_\alpha n_\alpha}{RT} \nabla \mu_\alpha$. In this case, $u$ and $J_\alpha$ are viewed as the mean velocity and general diffusion fluxes at the constant temperature and pressure, respectively.

(A2) The second choice is to take $\bfscrM$ as a full matrix,
\begin{equation}
M_{\alpha\beta} (n) = \sum_{\beta=1}^{M} \frac{D_{\alpha\beta} n_\alpha n_\beta}{RT}, \quad M_{\alpha\beta} (n) = -\frac{D_{\alpha\beta} n_\alpha n_\beta}{RT}, \quad \beta \neq \alpha,
\end{equation}
where the mole diffusion coefficients $D_{\alpha\beta}$ satisfy $D_{\alpha\alpha} = 0$ and $D_{\alpha\beta} = D_{\beta\alpha} > 0$ for $\alpha \neq \beta$. In this case, $u$ is the molar-average velocity.

(A3) The third mobility $\bfscrM$ has the following formulation:
\begin{equation}
M_{\alpha\alpha} (n) = \sum_{\beta=1}^{M} \frac{D_{\alpha\beta} \rho_\alpha \rho_\beta}{\rho M_{w,\alpha} RT}, \quad M_{\alpha\beta} (n) = -\frac{D_{\alpha\beta} \rho_\alpha \rho_\beta}{\rho M_{w,\alpha} M_{w,\beta} RT}, \quad \beta \neq \alpha,
\end{equation}
where the mass diffusion coefficients $D_{\alpha\beta}$ satisfy $D_{\alpha\alpha} = 0$ and $D_{\alpha\beta} = D_{\beta\alpha} > 0$ for $\alpha \neq \beta$. In this case, $u$ is actually the mass-average velocity.

We now introduce the thermodynamically consistent momentum balance equation, which is expressed as [29]
\begin{equation}
\begin{align*}
\frac{\partial (\rho u)}{\partial t} + \nabla \cdot (\rho u \otimes u) + \sum_{\alpha=1}^{M} M_{w,\alpha} \nabla \cdot (J_\alpha \otimes u) &= -\sum_{\alpha=1}^{M} n_\alpha \nabla \mu_\alpha \\
&+ \nabla \cdot \left( \lambda \nabla \cdot u \right) + \nabla \cdot \eta \left( \nabla u + \nabla u^T \right),
\end{align*}
\end{equation}
where $\lambda = \xi - \frac{2}{3} \eta$, and $\xi$ and $\eta$ represent the volumetric viscosity and shear viscosity, respectively. We assume that $\eta > 0$ and $\xi > \frac{2}{3} \eta$, and thus $\lambda > 0$. If $u$ is the mass-average velocity, the term $\sum_{\alpha=1}^{M} M_{w,\alpha} J_\alpha \cdot \nabla u$ vanishes, while for the other types of $u$, it is crucial to ensure the thermodynamical consistency.

In this work, we consider numerical schemes for the model formulated by (2.4) and (2.9) coupling with the chemical potential (2.3) and the diffusion flux (2.5). For the boundary conditions, we assume that all boundary terms in (2.4) and (2.9) will vanish when integration by parts is performed; for example, we can use homogeneous Neumann boundary conditions or periodic boundary conditions.

We assume that $\Omega \subset \mathbb{R}^d (1 \leq d \leq 3)$ is an open, bounded and connected domain with the sufficiently smooth boundary $\partial \Omega$. The Helmholtz free energy and kinetic energy within $\Omega$ at a specified time are defined as
\begin{equation}
F = F_b + F_\nabla, \quad F_b = \int_{\Omega} f_b dx, \quad F_\nabla = \frac{1}{2} \sum_{\alpha,\beta=1}^{M} \int_{\Omega} c_{\alpha\beta} \nabla n_\alpha \cdot \nabla n_\beta dx, \quad E = \frac{1}{2} \int_{\Omega} \rho |u|^2 dx.
\end{equation}
It is proved in [29] that the total energy, i.e., the sum of the Helmholtz free energy and kinetic energy, is dissipated with time as
\begin{equation}
\frac{\partial (F + E)}{\partial t} \leq 0.
\end{equation}
In order to use the SAV approach [46], we define \( H(t) = \sqrt{F_b + \sum_{\alpha=1}^{M} C_{T,\alpha} N_{\alpha}^t} \), where \( N_{\alpha} = \int_{\Omega} n_{\alpha} dx \). Here, \( C_{T,\alpha} \geq 0 \) is the thermodynamical coefficient of component \( \alpha \) to ensure \( F_b + \sum_{\alpha=1}^{M} C_{T,\alpha} N_{\alpha}^t \geq 0 \), and the choice of \( C_{T,\alpha} \geq 0 \) may depend on \( T \). But independent of molar densities. Then the chemical potential of component \( \alpha (1 \leq \alpha \leq M) \) can be reformulated as

\[
\mu_{\alpha} = \frac{H(t)}{\sqrt{F_b + \sum_{\alpha=1}^{M} C_{T,\alpha} N_{\alpha}^t}} \mu_{\alpha}^b - \sum_{\beta=1}^{M} \nabla \cdot (c_{\alpha\beta} \nabla n_{\beta}) ,
\]

\[
\frac{\partial H}{\partial t} = \sum_{\alpha=1}^{M} \int_{\Omega} 2 \sqrt{F_b + \sum_{\alpha=1}^{M} C_{T,\alpha} N_{\alpha}^t} \frac{\partial n_{\alpha}}{\partial t} dx.
\]

The modified Helmholtz free energy is defined as \( \mathcal{F} = H^2 + F_b - \sum_{\alpha=1}^{M} C_{T,\alpha} N_{\alpha}^t \). In the continuous model, we have \( \mathcal{F} = F \), but at the discrete level, the modified Helmholtz free energy may be generally different from the original Helmholtz free energy.

3. Energy-stable semi-implicit time schemes. In this section, we aim to develop efficient energy-dissipated semi-implicit time marching schemes for simulating the above multicomponent flow model. The key difficulties result from the strong nonlinearity of Helmholtz free energy density and fully coupling relations between molar densities and velocity. In this work, our purpose is to uncouple this tight coupling relation between molar densities and velocity, and from this, we will develop linearly decoupled numerical schemes preserving the feature of energy dissipation.

For a given time interval \( \mathcal{I} = (0, T_f) \), where \( T_f > 0 \), we divide \( \mathcal{I} \) into \( N \) subintervals \( \mathcal{I}_k = (t_k, t_{k+1}) \), where \( t_0 = 0 \) and \( t_N = T_f \), and we denote \( \tau_k = t_{k+1} - t_k \). For any scalar \( v(t) \) or vector \( v(t) \), we denote by \( v^k \) or \( v^k \) its approximation at time \( t_k \). The traditional notation \( (\cdot, \cdot) \) and \( \| \cdot \| \) is used to represent the inner product and norm of \( L^2(\Omega) \), \((L^2(\Omega))^d \), or \((L^2(\Omega))^{d \times d} \), respectively.

3.1. Velocity-density decoupled semi-implicit scheme. We try to develop a linear semi-implicit scheme that decouples the tight relationship between molar densities and velocity. This scheme allows us to solve the mass balance equations and momentum equation separately and can be applied to the model problems with general diffusion mobility, especially cases in which the mobility is a full tensor.

We denote \( n^k = [n_1^k, n_2^k, \ldots, n_M^k]^T \) and define \( \mu_{\alpha}^{k+1} \) as

\[
\mu_{\alpha}^{k+1} = \frac{H_{k+1}^b + H^b}{2 \sqrt{F_b(n^k) + \sum_{\beta=1}^{M} C_{T,\alpha} N_{\beta}^t}} \mu_{\alpha}^b(n^k) - \sum_{\beta=1}^{M} \nabla \cdot (c_{\alpha\beta} \nabla n_{\beta}^{k+1}) ,
\]

\[
\frac{H_{k+1}^b - H^b}{\tau_k} = \sum_{\alpha=1}^{M} \int_{\Omega} 2 \sqrt{F_b(n^k) + \sum_{\beta=1}^{M} C_{T,\alpha} N_{\beta}^t} \frac{\partial n_{\alpha}}{\partial t} dx.
\]

Furthermore, we define an intermediate velocity \( u^*_k \) as

\[
u^*_k = u_k - \frac{\tau_k}{\rho_k} \sum_{\alpha=1}^{M} n_{\alpha}^k \nabla \mu_{\alpha}^{k+1} .
\]
Using the intermediate velocity \( u^K_k \), we construct the semi-implicit time scheme for the molar density balance equation (2.4) as

\[
\frac{\rho^{k+1} - \rho^k}{\tau_k} + \nabla \cdot (\rho^k u^K_k) + \nabla \cdot J^k = 0, \quad J^k = -\sum_{\beta=1}^M M_{\alpha\beta}(n^k) \nabla \mu^k_{\beta},
\]

where we denote by \( M_{\alpha\beta}(n^k) \) the mobility coefficients calculated from molar densities \( n^k \) since the mobility coefficients \( M_{\alpha\beta} \), generally depending on molar densities, can be treated explicitly.

We can see that only \( n^{k+1} \) is the unknown variable of equations (3.3). This means that the use of \( u^K_k \) eliminates the tight coupling relationship between molar densities and velocity. We can solve (3.3) to obtain molar densities \( n^{k+1} \). Once \( n^{k+1} \) is calculated, we can get \( \rho^{k+1} \), \( J^{k+1} \), and \( u^K_k \). A semi-implicit scheme for the momentum balance equation (2.9) is formulated using the definition of \( u^K_k \) as

\[
\frac{\rho^{k+1} u^{k+1} - \rho^k u^k}{\tau_k} + \nabla \cdot (\rho^k u^K_k \otimes u^{k+1}) + \sum_{\alpha=1}^M M_{\alpha} \nabla \cdot (J^k_{\alpha} \otimes u^{k+1}) = \nabla (\lambda^k \nabla \cdot u^{k+1}) + \nabla \cdot \eta^k \left( \nabla u^{k+1} + (\nabla u^{k+1})^T \right),
\]

which is a linear equation of velocity \( u^{k+1} \) only. In the convection term of the discrete momentum balance equation, the use of \( u^K_k \) instead of \( u^k \) is consistent with the mass balance equations, as shown in the proof of Theorem 3.1, and moreover it avoids using the existing approach in [35, 45], which must impose the overall mass equation into balance equations, as shown in the proof of Theorem 3.1, and moreover it avoids using the existing approach in [35, 45], which must impose the overall mass equation into the momentum equation for the sake of achieving energy dissipation for phase-field models with the large density ratios.

We now prove that the above linearly decoupled scheme satisfies the discrete energy dissipation law. To do this, we define the discrete kinetic energy and the modified Helmholtz free energy as

\[
E^k = \frac{1}{2} \int_{\Omega} \rho^k |u^k|^2 dx,
\]

\[
F^k = |H^k|^2 + F_b^k - \sum_{\alpha=1}^M C_{\alpha} n^\alpha_{\alpha}, \quad F_b^k = \frac{1}{2} \int_{\Omega} \sum_{\alpha, \beta=1}^M c_{\alpha\beta} \nabla n^k_{\alpha} \cdot \nabla n^k_{\beta} dx.
\]

**Theorem 3.1.** The modified total (free) energy, i.e., the sum of the modified Helmholtz free energy and kinetic energy, determined by (3.3) and (3.4) associated with (3.1) and (3.2), is dissipated with time steps, i.e.,

\[
E^{k+1} + F^{k+1} \leq E^k + F^k.
\]

**Proof.** We first estimate \( |H^{k+1}|^2 - |H^k|^2 \) using (3.1b) as

\[
|H^{k+1}|^2 - |H^k|^2 = \sum_{\alpha=1}^M \left( \frac{(H^{k+1} + H^k) \mu^k_{\alpha}}{2 \sqrt{F_b(n^k) + C_{T\alpha} n^\alpha_{\alpha}}} (n^{k+1}_{\alpha} - n^k_{\alpha}) \right).
\]
Since the influence parameter matrix \((c_{\alpha \beta})_{i,j=1}^M\) is symmetric and is positive definite or positive semidefinite, we have

\[
F^{k+1} - F^k = \sum_{\alpha,\beta=1}^M \left( \nabla \left( n^{k+1}_\alpha - n^k_\alpha \right), c_{\alpha \beta} \nabla n^{k+1}_\beta \right) - \frac{1}{2} \sum_{\alpha,\beta=1}^M \left( c_{\alpha \beta} \nabla \left( n^{k+1}_\alpha - n^k_\alpha \right), \nabla \left( n^{k+1}_\beta - n^k_\beta \right) \right)
\leq \sum_{\alpha,\beta=1}^M \left( \nabla \left( n^{k+1}_\alpha - n^k_\alpha \right), c_{\alpha \beta} \nabla n^{k+1}_\beta \right)
\leq - \sum_{\alpha,\beta=1}^M \left( n^{k+1}_\alpha - n^k_\alpha, \nabla \cdot (c_{\alpha \beta} \nabla n^{k+1}_\beta) \right).
\]

The inequalities (3.7) and (3.8) yield

\[
F^{k+1} - F^k = |H^{k+1}|^2 - |H^k|^2 + F^{k+1}_\nabla - F^k_\nabla \leq \sum_{\alpha=1}^M \left( \mu^{k+1}_\alpha, n^{k+1}_\alpha - n^k_\alpha \right).
\]

Substituting (3.3) into (3.9), we derive

\[
\frac{F^{k+1} - F^k}{\tau_k} \leq - \sum_{\alpha=1}^M \left( \nabla \cdot (n^k_\alpha u^k_\alpha) + \nabla \cdot J^{k+1}_{\alpha}, \mu^{k+1}_\alpha \right)
\leq - \sum_{\alpha=1}^M \left( \nabla \cdot (n^k_\alpha u^k_\alpha), \mu^{k+1}_\alpha \right) - \sum_{\alpha,\beta=1}^M \left( M_{\alpha \beta} (n^k \nabla \mu^{k+1}_\alpha \nabla \mu^{k+1}_\beta) \right).
\]

We now turn to consider the difference between \(E^{k+1}\) and \(E^k\). We introduce the intermediate kinetic energy as

\[
E^k_* = \frac{1}{2} \left( \rho^k u^k_*, u^k_* \right).
\]

For \(a, b, c, d \in \mathbb{R}\) and \(c \geq 0\), we have

\[
ab^2 - cd^2 = 2b(ab - cd) - c(b - d)^2 = 2b(ab - cd) - b^2(a - c) \leq 2b(ab - cd) - b^2(a - c).
\]

The difference between \(E^{k+1}\) and \(E^k\) is estimated using (3.11) as

\[
E^{k+1} - E_*^k \leq \left( \rho^{k+1} u^{k+1} - \rho^k u^k, u^{k+1} \right) - \frac{1}{2} \left( \rho^{k+1} - \rho^k, |u^{k+1}|^2 \right).
\]

On the other hand, we have the overall mass balance equation as

\[
\frac{\rho^{k+1} - \rho^k}{\tau_k} + \nabla \cdot (\rho^k u^k) + \sum_{\alpha=1}^M M_{\alpha w} \nabla \cdot J^{k+1}_{\alpha} = 0.
\]
Substituting (3.13) and (3.4) into (3.12) yields

\[
\frac{E^{k+1} - E^k}{\tau_k} \leq - \left( \nabla \cdot (\rho^k u^k \otimes u^{k+1}) + \sum_{\alpha=1}^{M} M_{w,\alpha} \nabla \cdot (J^{k+1}_\alpha \otimes u^{k+1}) , u^{k+1} \right) \\
+ \left( \nabla \cdot (\lambda^k \nabla \cdot u^{k+1}) + \nabla \cdot \eta^k \left( \nabla u^{k+1} + (\nabla u^{k+1})^T \right) , u^{k+1} \right) \\
+ \frac{1}{2} \left( \nabla \cdot (\rho^k u^k) + \sum_{\alpha=1}^{M} M_{w,\alpha} \nabla \cdot J^{k+1}_\alpha , |u^{k+1}|^2 \right) \\
\leq - \left( (\rho^k u^k + \sum_{\alpha=1}^{M} M_{w,\alpha} J^{k+1}_\alpha) \nabla u^{k+1} , u^{k+1} \right) \\
- \left\| \sqrt{\lambda^k} \nabla \cdot u^{k+1} \right\|^2 - \frac{1}{2} \left\| \sqrt{\eta^k} \left( \nabla u^{k+1} + (\nabla u^{k+1})^T \right) \right\|^2 \\
- \frac{1}{2} \left( \nabla \cdot (\rho^k u^k) + \sum_{\alpha=1}^{M} M_{w,\alpha} \nabla \cdot J^{k+1}_\alpha , |u^{k+1}|^2 \right) \\
\leq - \left\| \sqrt{\lambda^k} \nabla \cdot u^{k+1} \right\|^2 - \frac{1}{2} \left\| \sqrt{\eta^k} \left( \nabla u^{k+1} + (\nabla u^{k+1})^T \right) \right\|^2.
\]

(3.14)

We apply the definition of \( u^*_k \) to derive

\[
E^*_k - E^k = (\rho^k (u^*_k - u^k) , u^k) - \frac{1}{2} (\rho^k , |u^*_k - u^k|^2) \leq (\rho^k (u^*_k - u^k) , u^*_k)
\]

(3.15)

\[
\leq - \tau_k \sum_{\alpha=1}^{M} n^k_\alpha \nabla \mu^{k+1}_\alpha , u^*_k \leq \tau_k \sum_{\alpha=1}^{M} \left( \nabla \cdot (n^k_\alpha u^*_\alpha) , \mu^{k+1}_\alpha \right).
\]

Combining (3.10), (3.14), and (3.15) yields

\[
\frac{E^{k+1} - E^k + \mathcal{E}^{k+1} - \mathcal{E}^k}{\tau_k} \leq - \sum_{\alpha,\beta=1}^{M} \left( M_{\alpha\beta}(n^k) \nabla \mu^{k+1}_\alpha , \nabla \mu^{k+1}_\beta \right) - \left\| \sqrt{\lambda^k} \nabla \cdot u^{k+1} \right\|^2 \\
- \frac{1}{2} \left\| \sqrt{\eta^k} \left( \nabla u^{k+1} + (\nabla u^{k+1})^T \right) \right\|^2 \leq 0,
\]

(3.16)

which yields the energy dissipation (3.6).

\[\square\]

3.2. Componentwise, decoupled semi-implicit scheme. For the case that diffusion fluxes have a diagonal mobility tensor, we can design a componentwise, decoupled semi-implicit scheme, which not only uncouples the tight relationship between molar densities and velocity, but also solves the mass balance equations in a componentwise way.

We still use \( n^k = [n^k_1, n^k_2, \ldots, n^k_M]^T \) to denote the molar density vector at the integer time step \( k \). Furthermore, we introduce the molar density vector at the fractional time step \( (k + \frac{\alpha}{M}) \) and denote it by \( n^{k+\frac{\alpha}{M}} = [n^{k+1}_1, n^{k+1}_2, \ldots, n^{k+1}_M]^T \), where \( 0 \leq \alpha \leq M \); in particular, \( n^{k+\frac{\alpha}{M}} = n^k \) for \( \alpha = 0 \) and \( n^{k+\frac{\alpha}{M}} = n^{k+1} \) for \( \alpha = M \). The
discrete chemical potential \( \mu_{\alpha}^{k+\frac{\tau}{\rho}} \) (1 ≤ \( \alpha \) ≤ \( M \)) of component \( \alpha \) is defined as
\[
\mu_{\alpha}^{k+\frac{\tau}{\rho}} = \frac{H^{k+\frac{\tau}{\rho}} + H^{k+\frac{\tau}{\rho}-1}}{2\sqrt{F_b(n^k) + \sum_{\beta = 1}^{M} C_{T,\beta} N_{\beta}^t}} \mu_{\alpha}^{k} \left(n^{k+\frac{\tau}{\rho}}\right)
\]
(3.17a)
\[
- \frac{\tau}{\rho} \sum_{\beta = 1}^{M} \nabla \cdot \left(c_{\alpha\beta} \nabla n_{\beta}^{k+1}\right) - \sum_{\beta = \alpha + 1}^{M} \nabla \cdot \left(c_{\alpha\beta} \nabla n_{\beta}^{k}\right),
\]
(3.20a)
\[
\frac{H^{k+\frac{\tau}{\rho}} - H^{k+\frac{\tau}{\rho}-1}}{\tau_k} = \int_{\Omega} \frac{\mu_{\alpha}^{k} \left(n^{k+\frac{\tau}{\rho}}\right)}{2\sqrt{F_b(n^k) + \sum_{\beta = 1}^{M} C_{T,\beta} N_{\beta}^t}} n_{\alpha}^{k+1} - n_{\alpha}^{k} \, dx.
\]
(3.17b)

A componentwise intermediate velocity \( u_{\star}^{k+\frac{\tau}{\rho}} \) is defined as
\[
u_{\star}^{k+\frac{\tau}{\rho}} = \nu_{\star}^{k+\frac{\tau}{\rho}-1} - \frac{\tau_k}{\rho} n_{\alpha}^{k} \nabla \mu_{\alpha}^{k+\frac{\tau}{\rho}}, \quad 1 \leq \alpha \leq M,
\]
(3.18)
where \( u_{\star}^{k+0} = u_{\star}^{k} = u^{k} \). Let \( \rho_{\alpha} = M_{w,\alpha} n_{\alpha} \) be the mass density of component \( \alpha \), and then we introduce a mean intermediate velocity \( u_{\star}^{k} \) as
\[
u_{\star}^{k} = \sum_{\alpha = 1}^{M} \frac{\rho_{\alpha}^{k}}{\rho_{\star}^{k}} u_{\star}^{k+\frac{\tau}{\rho}}.
\]
(3.19)

We construct the semi-implicit time scheme for the molar density balance equation (2.4) of component \( \alpha \) as
\[
u_{\alpha}^{k+1} - \nu_{\alpha}^{k} = \frac{\tau_k}{\rho_{\star}^{k}} n_{\alpha}^{k} \nabla \mu_{\alpha}^{k+\frac{\tau}{\rho}} + \nabla \cdot \left(\nu_{\alpha}^{k} u_{\star}^{k+\frac{\tau}{\rho}}\right) + \nabla \cdot \left(J_{\alpha}^{k+\frac{\tau}{\rho}} \otimes u^{k+1}\right) = 0,
\]
(3.20a)
\[
u_{\alpha}^{k+1} - \nu_{\alpha}^{k} = \frac{\tau_k}{\rho_{\star}^{k}} n_{\alpha}^{k} \nabla \mu_{\alpha}^{k+\frac{\tau}{\rho}} + \nabla \cdot \left(\nu_{\alpha}^{k} u_{\star}^{k+\frac{\tau}{\rho}}\right) + \sum_{\alpha = 1}^{M} M_{w,\alpha} \nabla \cdot \left(J_{\alpha}^{k+\frac{\tau}{\rho}} \otimes u^{k+1}\right)
\]
(3.20b)
which is a linear equation of \( n_{\alpha}^{k+1} \) only and can be solved sequentially from \( \alpha = 1 \) to \( M \). The semi-implicit time scheme for the momentum balance equation is
\[
u_{\alpha}^{k+1} - \nu_{\alpha}^{k} = \frac{\tau_k}{\rho_{\star}^{k}} n_{\alpha}^{k} \nabla \mu_{\alpha}^{k+\frac{\tau}{\rho}} + \nabla \cdot \left(\nu_{\alpha}^{k} u_{\star}^{k+\frac{\tau}{\rho}}\otimes u^{k+1}\right) + \sum_{\alpha = 1}^{M} M_{w,\alpha} \nabla \cdot \left(J_{\alpha}^{k+\frac{\tau}{\rho}} \otimes u^{k+1}\right)
\]
(3.21)
Summing (3.18) from \( \alpha = 1 \) to \( M \) yields
\[
u_{\star}^{k+1} = u^{k} - \frac{\tau_k}{\rho_{\star}^{k}} \sum_{\alpha = 1}^{M} n_{\alpha}^{k} \nabla \mu_{\alpha}^{k+\frac{\tau}{\rho}}.
\]
(3.22)
Consequently, (3.21) can be reformulated as
\[
u_{\alpha}^{k+1} - \nu_{\alpha}^{k} = \frac{\tau_k}{\rho_{\star}^{k}} n_{\alpha}^{k} \nabla \mu_{\alpha}^{k+\frac{\tau}{\rho}} + \nabla \cdot \left(\nu_{\alpha}^{k} u_{\star}^{k+\frac{\tau}{\rho}}\otimes u^{k+1}\right) + \sum_{\alpha = 1}^{M} M_{w,\alpha} \nabla \cdot \left(J_{\alpha}^{k+\frac{\tau}{\rho}} \otimes u^{k+1}\right)
\]
(3.23)
which is still a linear equation of velocity \( \mathbf{u}^{k+1} \) and easy to solve. In the convection term of (3.23), we use the mean intermediate velocity \( \bar{u}^k \), instead of \( u^k \) or \( u^{k+\frac{2}{3}} \), to match the mass balance equations.

It is apparent that the above componentwise approach can be directly applied for the IEQ-based componentwise schemes and for the Cahn–Hilliard-type models studied in [24]. We now prove that the componentwise, decoupled scheme satisfies the discrete energy dissipation law.

**Theorem 3.2.** Let \( E^k \) and \( \mathcal{F}^k \) still be defined as in (3.5). The sum of the modified Helmholtz free energy and kinetic energy determined by (3.20) and (3.23) associated with (3.17), (3.18), and (3.19) is dissipated with time steps, i.e.,

\[
E^{k+1} + \mathcal{F}^{k+1} \leq E^k + \mathcal{F}^k.
\]

**Proof.** Using (3.17b), we derive that

\[
|H^{k+\frac{2}{3}}|^2 - |H^{k+\frac{2}{3}}|^2 = \frac{(H^{k+\frac{2}{3}} + H^{k+\frac{2}{3}}) \left( \mu^b (n^{k+\frac{2}{3}}), n^{k+1}_\alpha - n^k_\alpha \right)}{2\sqrt{F_b(n^k) + \sum_{\beta=1}^M C_{\alpha\beta} N^\beta}}
\]

The gradient Helmholtz free energy at the time step \((k + \frac{2}{3})\) can be expressed as

\[
F^{k+\frac{2}{3}} = \frac{1}{2} \int_{\Omega} \sum_{\alpha=1}^\alpha c_{\alpha\beta} \nabla n^{k+1}_\beta \cdot \nabla n^{k+1}_\alpha \, dx + \frac{1}{2} \int_{\Omega} \sum_{\beta=1}^\beta c_{\beta\alpha} \nabla n^{k+1}_\beta \cdot \nabla n^{k}_\alpha \, dx
\]

Taking into account \( c_{\alpha\beta} = c_{\beta\alpha} \) and \( c_{\alpha\beta} \geq 0 \), we derive

\[
F^{k+\frac{2}{3}} - F^{k+\frac{2}{3}} = \int_{\Omega} \sum_{\beta=1}^{\alpha-1} c_{\alpha\beta} \nabla \left( n^{k+1}_\alpha - n^{k}_\alpha \right) \cdot \nabla n^{k+1}_\alpha \, dx + \int_{\Omega} \sum_{\beta=1}^\beta c_{\beta\alpha} \nabla \left( n^{k+1}_\alpha - n^{k}_\alpha \right) \cdot \nabla n^{k+1}_\alpha \, dx
\]

By the definition of \( \mu^b_k \) given in (3.17), we obtain from (3.25) and (3.27) that

\[
|H^{k+\frac{2}{3}}|^2 - |H^{k+\frac{2}{3}}|^2 + F^{k+\frac{2}{3}} - F^{k+\frac{2}{3}} \leq \left( \mu^{k+\frac{2}{3}}, n^{k+1}_\alpha - n^{k}_\alpha \right)
\]

\[
\leq -\tau_k \left( \mu^{k+\frac{2}{3}}, \nabla \cdot \left( n^{k}_\alpha \nabla \mu^{k+\frac{2}{3}} \right) - \nabla \cdot \left( \mathcal{M}_\alpha (n^k) \nabla \mu^{k+\frac{2}{3}} \right) \right)
\]

\[
\leq \tau_k \left( \mu^{k+\frac{2}{3}}, \nabla \mu^{k+\frac{2}{3}} \right) - \tau_k \left\| \mathcal{M}_\alpha (n^k) \nabla \mu^{k+\frac{2}{3}} \right\|^2
\]

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where the second equality is obtained by using (3.20). Summing up (3.28) from $\alpha = 1$ to $M$ yields
\[
\mathcal{F}^{k+1} - \mathcal{F}^k = |H^{k+1}|^2 - |H^k|^2 + F_{\mathcal{V}}^{k+1} - F_{\mathcal{V}}^k \\
= \sum_{\alpha=1}^{M} \left( |H^{k+\alpha}|^2 - |H^{k+\alpha-1}|^2 + F_{\mathcal{V}}^{k+\alpha} - F_{\mathcal{V}}^{k+\alpha-1} \right) \\
\leq \tau_k \sum_{\alpha=1}^{M} \left( \rho^k \nabla \cdot \bfitn_{\alpha} n_{\alpha}^k \nabla \mu_{\alpha}^{k+\alpha}, \nabla \mu_{\alpha}^{k+\alpha} \right) - \tau_k \sum_{\alpha=1}^{M} \left\| \sqrt{M_{\alpha}(n^k)} \nabla \mu_{\alpha}^{k+\alpha} \right\|^2.
\] (3.29)

We define the intermediate kinetic energy as
\[
E_{k+\alpha} = \frac{1}{2} \left( \rho^k \nabla \cdot (\rho^k \nabla J_{\alpha}^{k+\alpha}), \nabla \mu_{\alpha}^{k+\alpha} \right).
\]
Using the definition (3.18) of intermediate velocities, we derive
\[
E_{k+\alpha} - E_{k+\alpha-1} \leq \left( \rho^k \nabla \cdot (\rho^k \nabla J_{\alpha}^{k+\alpha}), \nabla \mu_{\alpha}^{k+\alpha} \right) \\
\leq -\tau_k \left( n_{\alpha}^k \nabla \mu_{\alpha}^{k+\alpha}, \nabla \mu_{\alpha}^{k+\alpha} \right).
\] (3.30)

The sum of (3.20) multiplied by $M_{w,\alpha}$ leads to the overall mass balance equation
\[
\frac{\rho^{k+1} - \rho^k}{\tau_k} = -\nabla \cdot (\rho^k \nabla J_{\alpha}^{k+\alpha}) - \sum_{\alpha=1}^{M} M_{w,\alpha} \nabla \cdot J_{\alpha}^{k+\alpha},
\] (3.31)

where the first term on the right-hand side is obtained using (3.19). Similarly to (3.12) and (3.14), we estimate the difference between $E^{k+1}$ and $E_k$ as
\[
\frac{E^{k+1} - E^{k+1}}{\tau_k} \leq \frac{1}{\tau_k} \left( \rho^{k+1} u^{k+1} - \rho^k u_{k+1}, u^{k+1} \right) - \frac{1}{2\tau_k} \left( \rho^{k+1} - \rho^k, |u^{k+1}|^2 \right) \\
\leq - \left( \nabla \cdot (\rho^k \nabla J_{\alpha}^{k+\alpha} \otimes u^{k+1}) + \sum_{\alpha=1}^{M} M_{w,\alpha} \nabla \cdot (J_{\alpha}^{k+\alpha} \otimes u^{k+1}), u^{k+1} \right) \\
+ \left( \nabla \cdot (\rho^k \nabla u^{k+1}) + \sum_{\alpha=1}^{M} M_{w,\alpha} \nabla \cdot (J_{\alpha}^{k+\alpha} \otimes (\nabla u^{k+1})^T), u^{k+1} \right) \\
+ \frac{1}{2} \left( \nabla \cdot (\rho^k \nabla u^{k+1}) + \sum_{\alpha=1}^{M} M_{w,\alpha} \nabla \cdot (J_{\alpha}^{k+\alpha} \otimes |u^{k+1}|^2) \right) \\
\leq - \left\| \sqrt{\lambda^k} \nabla \cdot u^{k+1} \right\|^2 - \frac{1}{2} \left\| \sqrt{\eta^k} \left( \nabla u^{k+1} + (\nabla u^{k+1})^T \right) \right\|^2,
\] (3.32)

where we also used (3.23) and (3.31). Combining (3.30) and (3.32) yields
\[
E^{k+1} - E_k = E^{k+1} - E_{k+1} + \sum_{\alpha=1}^{M} \left( E_{k+\alpha} - E_{k+\alpha-1} \right) \\
\leq -\tau_k \sum_{\alpha=1}^{M} \left( n_{\alpha}^k \nabla \mu_{\alpha}^{k+\alpha}, \nabla \mu_{\alpha}^{k+\alpha} \right).
\] (3.33)

Finally, the energy dissipation (3.24) is derived from (3.29) and (3.33). \[\Box\]
4. Fully discrete schemes and unconditional energy stability. In this section, we consider the spatial discretization of the semidiscrete time schemes developed in section 3. The boundary conditions are taken as \( \mathbf{u} = 0 \), \( \mathbf{J}_\alpha \cdot \gamma_{\partial \Omega} = 0 \) and \( \nabla n_\alpha \cdot \gamma_{\partial \Omega} = 0 \) (\( 1 \leq \alpha \leq M \)) on the boundary \( \partial \Omega \) of \( \Omega \), where \( \gamma_{\partial \Omega} \) is the normal unit outward vector to \( \partial \Omega \). The fully discrete schemes and their theoretical analysis can be easily extended to periodic boundary conditions. For simplification of presentation, we consider the two-dimensional case only, but the extension to the three-dimensional case is straightforward.

We take a rectangular domain to be aligned with the coordinate axes, where the horizontal axis is the \( x \)-direction and the vertical axis is the \( y \)-direction; more precisely, \( \Omega = [0, L_x] \times [0, L_y] \), where \( L_x > 0 \) and \( L_y > 0 \). The velocity is expressed as \( \mathbf{u} = [u, v]^T \), where \( u \) is the \( x \)-direction velocity component and \( v \) is the \( y \)-direction velocity component. For simplicity, we use a uniform division of \( \Omega \) as \( 0 = x_0 < x_1 < \cdots < x_{N_x} = L_x \) and \( 0 = y_0 < y_1 < \cdots < y_{N_y} = L_y \), where \( N_x \) and \( N_y \) are positive integers.

We denote the mesh size \( h = x_{i+1} - x_i = y_{j+1} - y_j \), where \( x_0 < i < N_x \) and \( 0 < j < N_y \) are integers. We also introduce \( x_{i+\frac{1}{2}} = x_i + \frac{h}{2} \) and \( y_{j+\frac{1}{2}} = y_j + \frac{h}{2} \).

For the spatial discretization, we will apply the finite difference/volume methods with the upwind scheme for the convection terms on staggered grids [19, 50]. In Figure 4.1, we show a two-dimensional uniform staggered mesh, in which different variables are discretized on different grids that are shifted half a grid point. A primal control volume is outlined by a red solid line in Figure 4.1; at its center \( (x_{i+\frac{1}{2}}, y_{j+\frac{1}{2}}) \), molar density of component \( \alpha \) is placed, whereas \( u \) is discretized at the middle (i.e., \( (x_i, y_{j+\frac{1}{2}}) \)) of its left and right edges (marked by the blue triangle—see the electronic version of this article for color figures), and \( v \) is discretized at the middle (i.e., \( (x_{i+\frac{1}{2}}, y_j) \)) of its top and bottom edges (marked by the green triangle).

4.1. Discrete function spaces and discrete operators. We define the following discrete function spaces:

\[ \mathcal{S}_c = \{ \phi : (x_{i+\frac{1}{2}}, y_{j+\frac{1}{2}}) \mapsto \mathbb{R}, \ 0 \leq i \leq N_x - 1, \ 0 \leq j \leq N_y - 1 \}, \]

\[ \mathcal{S}_u = \{ u : (x_i, y_{j+\frac{1}{2}}) \mapsto \mathbb{R}, \ 0 \leq i \leq N_x, \ 0 \leq j \leq N_y - 1 \}, \]
Functions in these discrete function spaces are identified by the subscripts in their component forms, for example, \( \phi_{i+\frac{1}{2}, j+\frac{1}{2}} = \phi(x_{i+\frac{1}{2}}, y_{j+\frac{1}{2}}) \) for \( \phi \in \mathcal{S}_c \). Taking into account the boundary conditions, we further define the subsets of \( \mathcal{S}_u \) and \( \mathcal{S}_v \) as

\[
\mathcal{S}_u^0 = \{ u \in \mathcal{S}_u | u_{i,j+\frac{1}{2}} = u_{N_x,j+\frac{1}{2}} = 0, \ 0 \leq j \leq N_y - 1 \},
\]

\[
\mathcal{S}_v^0 = \{ v \in \mathcal{S}_v | v_{i+\frac{1}{2}, j} = v_{i+\frac{1}{2}, N_y} = 0, \ 0 \leq i \leq N_x - 1 \}.
\]

To impose conveniently the boundary conditions for the velocity, we introduce some points out of the physical domain and define two extended discrete function spaces:

\[
\mathcal{S}_u^0 = \left\{ u : (x_i, y_{j+\frac{1}{2}}) \mapsto \begin{cases} 
\mathbb{R}, & 1 \leq i \leq N_x - 1, \ 0 \leq j \leq N_y - 1 \\
0, & \begin{cases} i \in \{0, N_x\}, \ 0 \leq j \leq N_y - 1 \\
1 \leq i \leq N_x - 1, j \in \{-1, N_y\} 
\end{cases}
\end{cases} \right\},
\]

\[
\mathcal{S}_v^0 = \left\{ v : (x_{i+\frac{1}{2}}, y_j) \mapsto \begin{cases} 
\mathbb{R}, & 0 \leq i \leq N_x - 1, \ 1 \leq j \leq N_y - 1 \\
0, & \begin{cases} 0 \leq i \leq N_x - 1, \ j \in \{0, N_y\} \\
i \in \{-1, N_x\}, \ 1 \leq j \leq N_y - 1 
\end{cases}
\end{cases} \right\}.
\]

We now introduce the discrete operators. For \( \phi \in \mathcal{S}_c \), we define the difference operators

\[
\delta_x^c[\phi]_{i,j+\frac{1}{2}} = \begin{cases} 
\phi_{i+\frac{1}{2}, j+\frac{1}{2}} - \phi_{i-\frac{1}{2}, j+\frac{1}{2}}, & 1 \leq i \leq N_x - 1, \ 0 \leq j \leq N_y - 1, \\
0, & \begin{cases} i \in \{0, N_x\}, \ 0 \leq j \leq N_y - 1 \\
1 \leq i \leq N_x - 1, j \in \{-1, N_y\} 
\end{cases}
\end{cases},
\]

\[
\delta_y^c[\phi]_{i+\frac{1}{2}, j} = \begin{cases} 
\phi_{i+\frac{1}{2}, j+\frac{1}{2}} - \phi_{i+\frac{1}{2}, j-\frac{1}{2}}, & 0 \leq i \leq N_x - 1, \ 1 \leq j \leq N_y - 1, \\
0, & \begin{cases} j \in \{0, N_y\}, \ 0 \leq i \leq N_x - 1 \\
i \in \{-1, N_x\}, \ 1 \leq j \leq N_y - 1 
\end{cases}
\end{cases},
\]

which imply \( \delta_x^c[\phi] \in \mathcal{S}_u^0 \) and \( \delta_y^c[\phi] \in \mathcal{S}_v^0 \), and we also define the average operators

\[
A_x^c[\phi]_{i,j+\frac{1}{2}} = \begin{cases} 
\frac{1}{2} (\phi_{i+\frac{1}{2}, j+\frac{1}{2}} + \phi_{i-\frac{1}{2}, j+\frac{1}{2}}), & 1 \leq i \leq N_x - 1, \ 0 \leq j \leq N_y - 1, \\
\phi_{i+\frac{1}{2}, j+\frac{1}{2}}, & i = 0, \ 0 \leq j \leq N_y - 1, \\
\phi_{i-\frac{1}{2}, j+\frac{1}{2}}, & i = N_x, \ 0 \leq j \leq N_y - 1,
\end{cases}
\]

\[
A_y^c[\phi]_{i+\frac{1}{2}, j} = \begin{cases} 
\frac{1}{2} (\phi_{i+\frac{1}{2}, j+\frac{1}{2}} + \phi_{i+\frac{1}{2}, j-\frac{1}{2}}), & 0 \leq i \leq N_x - 1, \ 1 \leq j \leq N_y - 1, \\
\phi_{i+\frac{1}{2}, j+\frac{1}{2}}, & j = 0, \ 0 \leq i \leq N_x - 1, \\
\phi_{i+\frac{1}{2}, j-\frac{1}{2}}, & j = N_y, \ 0 \leq i \leq N_x - 1,
\end{cases}
\]

which show that \( A_x^c[\phi] \in \mathcal{S}_u \) and \( A_y^c[\phi] \in \mathcal{S}_v \). The upwind scheme will be applied for the convection term, so we introduce the upwind operator for \( \phi \in \mathcal{S}_c \) in the presence of the velocity components \( u \in \mathcal{S}_u \cup \mathcal{S}_u^0 \cup \mathcal{S}_v^0 \) and \( v \in \mathcal{S}_v \cup \mathcal{S}_v^0 \cup \mathcal{S}_v^0 \) as

\[
A_x^c[\phi][u]_{i,j+\frac{1}{2}} = A_x^c[\phi]_{i,j+\frac{1}{2}} - \frac{1}{2} \text{sign}(u_{i,j+\frac{1}{2}}) \delta_x^c[\phi]_{i,j+\frac{1}{2}},
\]
\[ \Lambda_x^u[u]_{i+\frac{1}{2},j+\frac{1}{2}} = \Lambda_x^u[u]_{i+\frac{1}{2},j} - \frac{1}{2} \text{sign}(U_{i+\frac{1}{2},j}) \delta_x^u[u]_{i+\frac{1}{2},j}, \]

where \( \text{sign}(\cdot) \) is a sign function defined as

\[
\text{sign}(\phi) = \begin{cases} 
1, & \phi > 0, \\
0, & \phi = 0, \\
-1, & \phi < 0.
\end{cases}
\]

For \( u \in S_u \cup \overline{S}_u \) and \( v \in S_v \cup \overline{S}_v \), we define the difference and average operators

\[
\delta_x^u[u]_{i+\frac{1}{2},j+\frac{1}{2}} = u_{i+1,j+\frac{1}{2}} - u_{i,j+\frac{1}{2}}, \quad \delta_y^v[v]_{i+\frac{1}{2},j+\frac{1}{2}} = v_{i+\frac{1}{2},j+1} - v_{i+\frac{1}{2},j},
\]

\[
A_x^u[u]_{i+\frac{1}{2},j+\frac{1}{2}} = \frac{u_{i,j+\frac{1}{2}} + u_{i+1,j+\frac{1}{2}}}{2}, \quad A_y^v[v]_{i+\frac{1}{2},j+\frac{1}{2}} = \frac{v_{i+\frac{1}{2},j} + v_{i+\frac{1}{2},j+1}}{2}.
\]

For \( u \in \overline{S}_u \) and \( v \in \overline{S}_v \), we further define

\[
\delta_x^u[u]_{i,j} = u_{i+\frac{1}{2},j} - u_{i,j}, \quad \delta_y^v[v]_{i,j} = v_{i+\frac{1}{2},j} - v_{i,j},
\]

\[
A_x^u[u]_{i,j} = \frac{u_{i,j} + u_{i+1,j}}{2}, \quad A_y^v[v]_{i,j} = \frac{v_{i,j} + v_{i+1,j}}{2}.
\]

We can see that \( \delta_x^u[u] \in S_u \), \( \delta_y^v[v] \in S_v \), \( \delta_x^u[u] \in S_{pu} \), and \( \delta_y^v[v] \in S_{pv} \). For \( u \in \overline{S}_u \) and \( v \in \overline{S}_v \), we also define the following upwind operators:

\[
\Lambda_x^u[u]U_{i+\frac{1}{2},j+\frac{1}{2}} = \Lambda_x^u[u]_{i+\frac{1}{2},j} - \frac{1}{2} \text{sign}(U_{i+\frac{1}{2},j}) \delta_x^u[u]_{i+\frac{1}{2},j}, \quad U \in S_u,
\]

\[
\Lambda_y^v[u]V_{i,j} = A_y^v[u]_{i,j} - \frac{1}{2} \text{sign}(V_{i,j}) \delta_y^v[u]_{i,j}, \quad V \in S_{pu},
\]

\[
\Lambda_y^v[v]V'_{i+\frac{1}{2},j+\frac{1}{2}} = \Lambda_y^v[v]_{i+\frac{1}{2},j} - \frac{1}{2} \text{sign}(V'_{i+\frac{1}{2},j}) \delta_y^v[v]_{i+\frac{1}{2},j}, \quad V' \in S_v,
\]

\[
\Lambda_x^v[v]U'_{i,j} = \Lambda_x^v[v]_{i,j} - \frac{1}{2} \text{sign}(U'_{i,j}) \delta_x^v[v]_{i,j}, \quad U' \in S_{pv}.
\]

For \( \psi \in S_{pu} \cup S_{pv} \), we define the difference operators

\[
\delta_x^v[\psi]_{i+\frac{1}{2},j} = \psi_{i+1,j} - \psi_{i,j}, \quad \delta_y^v[\psi]_{i,j+\frac{1}{2}} = \psi_{i,j+1} - \psi_{i,j}.
\]

We define the following discrete inner products:

\[
\langle \phi, \phi' \rangle_c = \sum_{i=0}^{N_x-1} \sum_{j=0}^{N_y-1} \phi_{i+\frac{1}{2},j+\frac{1}{2}} \phi'_{i+\frac{1}{2},j+\frac{1}{2}}, \quad \phi, \phi' \in S_c,
\]

\[
\langle \psi, \psi' \rangle_{pu} = \sum_{i=1}^{N_x-1} \sum_{j=0}^{N_y-1} \psi_{i,j} \psi'_{i,j}, \quad \psi, \psi' \in S_{pu},
\]

We define the following discrete inner products:

\[
\langle \phi, \phi' \rangle_c = \sum_{i=0}^{N_x-1} \sum_{j=0}^{N_y-1} \phi_{i+\frac{1}{2},j+\frac{1}{2}} \phi'_{i+\frac{1}{2},j+\frac{1}{2}}, \quad \phi, \phi' \in S_c,
\]

\[
\langle \psi, \psi' \rangle_{pu} = \sum_{i=1}^{N_x-1} \sum_{j=0}^{N_y-1} \psi_{i,j} \psi'_{i,j}, \quad \psi, \psi' \in S_{pu},
\]
let $H$ be the fully discrete modified bulk Helmholtz free energy. Then we introduce its variation equation as

\[
\langle \varphi, \varphi' \rangle_{pu} = \sum_{i=0}^{N_x} \sum_{j=1}^{N_y} \varphi_{i,j} \varphi_{i,j}', \quad \varphi, \varphi' \in \mathcal{S}_{pu},
\]

\[
\langle u, u' \rangle_u = \sum_{i=1}^{N_x-1} \sum_{j=0}^{N_y-1} u_{i,j+\frac{1}{2}} u_{i,j+\frac{1}{2}}', \quad u, u' \in \mathcal{S}_u \cup \mathcal{S}_u^0 \cup \mathcal{S}_u^1,
\]

\[
\langle v, v' \rangle_v = \sum_{i=0}^{N_x-1} \sum_{j=1}^{N_y} v_{i+\frac{1}{2},j} v_{i+\frac{1}{2},j}', \quad v, v' \in \mathcal{S}_v \cup \mathcal{S}_v^0 \cup \mathcal{S}_v^1.
\]

The following summation-by-parts formulas are derived by direct calculations:

\[(4.7a) \quad \langle u, \delta_x^c [\phi] \rangle_u = -\langle \delta_x^u [u], \phi \rangle_c, \quad u \in \mathcal{S}_u^0 \cup \mathcal{S}_u^1, \quad \phi \in \mathcal{S}_c,
\]

\[(4.7b) \quad \langle v, \delta_y^c [\phi] \rangle_v = -\langle \delta_y^v [v], \phi \rangle_c, \quad v \in \mathcal{S}_v^0 \cup \mathcal{S}_v^1, \quad \phi \in \mathcal{S}_c,
\]

\[(4.7c) \quad \langle \delta_y^u [\psi], u \rangle_u = -\langle \psi, \delta_y^u [u] \rangle_{pu}, \quad u \in \mathcal{S}_u^0, \quad \psi \in \mathcal{S}_{pu},
\]

\[(4.7d) \quad \langle \delta_x^u [\varphi], v \rangle_v = -\langle \varphi, \delta_x^u [v] \rangle_{pu}, \quad v \in \mathcal{S}_v^0, \quad \varphi \in \mathcal{S}_{pu}.
\]

Some similar summation-by-parts formulas have also been presented in [4, 6, 32, 51, 52].

### 4.2. Fully discrete velocity-density decoupled scheme.

Because of the limitation of article length, we consider the fully discrete forms of the velocity-density decoupled scheme proposed in subsection 3.1 only. The spatial discretization forms of the componentwise decoupled scheme proposed in subsection 3.2 can be constructed and analyzed using similar approaches and routines.

At each time step, we seek the discrete molar densities $n^{k+1}_\alpha \in \mathcal{S}_c$ and the discrete velocity components $u^{k+1} \in \mathcal{S}_u^0$ and $v^{k+1} \in \mathcal{S}_v^0$ by solving the fully discrete equations sequentially, in which $n^k \in \mathcal{S}_c$, $u^k \in \mathcal{S}_u^0$, and $v^k \in \mathcal{S}_v^0$ are provided or already computed in the previous time step.

Let $H^k_h$ be the fully discrete modified bulk Helmholtz free energy. Then we introduce its variation equation as

\[
H^{k+1}_h - H^k_h = \frac{h^2}{2 F^k_b + \sum_{\beta=1}^{M} C_{T, \beta} N^k_{\beta}} \sum_{\alpha=1}^{M} \langle \mu^b_{\alpha}, n^{k+1}_\alpha - n^k_\alpha \rangle_c,
\]

where $H^0_h = \sqrt{F^0_b + \sum_{\beta=1}^{M} C_{T, \beta} N^0_{\beta}}$ and $F^k_b = h^2 (f_b(n^k), 1)$. The fully discrete chemical potentials $\mu^{k+1}_\alpha \in \mathcal{S}_c$ can be expressed as

\[(4.9a) \quad \mu^{k+1}_\alpha = \frac{H^{k+1}_h + H^k_h}{2 F^k_b + \sum_{\beta=1}^{M} C_{T, \beta} N^k_{\beta}} \mu^b_\alpha (n^k) + \mu^{\nabla, k+1}_\alpha,
\]

\[(4.9b) \quad \mu^{\nabla, k+1}_\alpha = -\frac{1}{h^2} \sum_{\beta=1}^{M} c_{\alpha \beta} \left( \delta_x^\psi [\delta_x^c [n^{k+1}_\beta]] + \delta_y^\psi [\delta_y^c [n^{k+1}_\beta]] \right).
\]
Let $\lambda$ balance equation (3.4) and introduce $U$ from their formulations. We next consider the fully discrete scheme for the momentum (4.11b) stated: Given $n^k, \eta^k \in \mathcal{S}_c$, $u^k \in \mathcal{S}_u^0$, and $v^k \in \mathcal{S}_v^0$, find $n^{k+1} \in \mathcal{S}_c$ such that

\begin{equation}
\eta^{k+1} - \eta^k = -\frac{T_k}{h} \sum_{\alpha=1}^{M} A_\alpha^x [n^k \eta^x] \delta^x_{\alpha \beta} [\mu^k] + \frac{1}{h} \sum_{\alpha=1}^{M} A_\alpha^y [n^k \eta^y] \delta^y_{\alpha \beta} [\mu^k],
\end{equation}

where \( J^{x,k+1} \in \mathcal{S}_u \) and \( J^{y,k+1} \in \mathcal{S}_v \) are defined as

\begin{align}
J^{x,k+1} &= -\frac{1}{h} \sum_{\beta=1}^{M} M_{\alpha \beta} (A_x^\alpha [n^k]) \delta^x_{\alpha \beta}, \\
J^{y,k+1} &= -\frac{1}{h} \sum_{\beta=1}^{M} M_{\alpha \beta} (A_y^\alpha [n^k]) \delta^y_{\alpha \beta}.
\end{align}

Once $n^{k+1}$ is computed by solving (4.10), we can get $u^k, v^k, J^{x,k+1}$, and $J^{y,k+1}$ from their formulations. We next consider the fully discrete scheme for the momentum balance equation (3.4) and introduce $U^k \in \mathcal{S}_c, U^k_\nu \in \mathcal{S}_{pu}, V^k_\nu \in \mathcal{S}_c$, and $V^k_u \in \mathcal{S}_{pu}$ as

\begin{align}
U^k &= A_x^u [\eta^x] u^k + \sum_{\alpha=1}^{M} M_{w,\alpha} A_x^\alpha [J^{x,k+1}], \\
U^k_\nu &= A_y^u [\eta^x] u^k + \sum_{\alpha=1}^{M} M_{w,\alpha} A_y^\alpha [J^{x,k+1}], \\
V^k &= A_x^y [\eta^y] v^k + \sum_{\alpha=1}^{M} M_{w,\alpha} A_y^\alpha [J^{y,k+1}], \\
V^k_\nu &= A_x^y [\eta^y] v^k + \sum_{\alpha=1}^{M} M_{w,\alpha} A_y^\alpha [J^{y,k+1}].
\end{align}

Let $\lambda^k \in \mathcal{S}_c$ and $\eta^k \in \mathcal{S}_c$, and further denote by $\eta^{k}_{i,j}$ the average values of $\eta^k$ at $(x_i, y_j)$. We introduce the discrete forms $\Phi^{k+1} \in \mathcal{S}_c$ for $\lambda^k \nabla \cdot u^{k+1}$ as

\begin{equation}
\Phi^{k+1} = \frac{1}{h} \lambda^k (\delta^x_{\alpha \beta} [u^{k+1}] + \delta^y_{\alpha \beta} [v^{k+1}]).
\end{equation}

To discretize the stress tensor $\eta^k (\nabla u^{k+1} + (\nabla u^{k+1})^T)$, we introduce $\Psi^{k+1} \in \mathcal{S}_c$, $\Upsilon^{k+1} \in \mathcal{S}_{pu}$, and $\Theta^{k+1} \in \mathcal{S}_{pu} \cup \mathcal{S}_{pv}$ as

\begin{equation}
\Psi^{k+1} = \frac{2}{h} \eta^k \delta^x_{\alpha \beta} [u^{k+1}], \quad \Upsilon^{k+1} = \frac{2}{h} \eta^k \delta^y_{\alpha \beta} [v^{k+1}], \quad \Theta^{k+1} = \frac{\eta^k}{h} (\delta^x_{\alpha \beta} [u^{k+1}] + \delta^y_{\alpha \beta} [v^{k+1}]).
\end{equation}
The discrete density is computed as $\rho^k = \sum_{\alpha=1}^{M} n^k_\alpha M_{w,\alpha}$. The fully discrete scheme for the momentum balance equation (3.4) is stated: Find $u^{k+1} \in \mathcal{S}^0_u$ and $v^{k+1} \in \mathcal{S}^0_v$ such that $u_{i,j+\frac{1}{2}}^{k+1}$ ($1 \leq i \leq N_x - 1$, $0 \leq j \leq N_y - 1$) satisfy

$$\frac{A_x^c[\rho^{k+1}]u^{k+1} - A_x^c[\rho^k]u^k}{\tau_k} + \frac{1}{h} \left( \delta_x^c[U^k u^k u^k] + \delta_y^c[V^k u^k v^k] \right)$$

(4.14) $$- \frac{1}{h} \left( \delta_x^c(u_{i,j}^{k+1} + \delta_v^c[\Phi^{k+1}] + \delta_y^c[\Theta^{k+1}]) = 0,$$

and $v_{i,j}^{k+1}$ ($0 \leq i \leq N_x - 1$, $1 \leq j \leq N_y - 1$) satisfy

$$\frac{A_y^c[\rho^{k+1}]v^{k+1} - A_y^c[\rho^k]v^k}{\tau_k} + \frac{1}{h} \left( \delta_y^c[U^k v^k v^k] + \delta_x^c[V^k v^k u^k] \right)$$

(4.15) $$- \frac{1}{h} \left( \delta_y^c(u_{i,j}^{k+1} + \delta_v^c(\Phi^{k+1}) + \delta_x^c(\Theta^{k+1}) \right) = 0.$$

4.3. Fully discrete energy-dissipation law. We define the fully discrete formulations of kinetic energy and modified Helmholtz free energy as

$$E^k = \frac{h^2}{2} \left( A_x^c[\rho^k]u^k, u^k \right) + \frac{h^2}{2} \left( A_y^c[\rho^k]v^k, v^k \right),$$

$$\mathcal{F}^k = |H^k|^2 + F_{\nabla, h} - \sum_{\alpha=1}^{M} C_{T,\alpha} \mathcal{N}_\alpha^k,$$

$$F_{\nabla, h}^k = \frac{1}{2} \sum_{\alpha, \beta=1}^{M} c_{\alpha, \beta} \left( \left\langle \delta_x^c[n^k_{\alpha}], \delta_x^c[n^k_{\beta}] \right\rangle_u + \left\langle \delta_y^c[n^k_{\alpha}], \delta_y^c[n^k_{\beta}] \right\rangle_v \right).$$

We also define the discrete intermediate kinetic energy as

$$E^{k, \ast}_h = \frac{h^2}{2} \left( A_x^c[\rho^k]u^k, u^k \right) + \frac{h^2}{2} \left( A_y^c[\rho^k]v^k, v^k \right).$$

In what follows, we will prove that the above fully discrete scheme obeys a discrete energy dissipation law.

**Lemma 4.1.** The fully discrete modified Helmholtz free energy varies with time steps as

$$\frac{\mathcal{F}^{k+1} - \mathcal{F}^k}{\tau_k} \leq -h \sum_{\alpha=1}^{M} \left\langle \mu^{k+1}_\alpha, \delta_x u^k \delta_x^c[n^k_{\alpha} u^k] + \delta_y v^k \delta_y^c[n^k_{\alpha} v^k] \right\rangle_c.$$  (4.17)

**Proof.** The difference between $|H^k|^2$ and $|H^k|^2$ is deduced using (4.8) as

$$|H^{k+1}|^2 - |H^k|^2 = \frac{h^2}{2} \left( \frac{H^{k+1} + H^k}{2} \right) \sum_{\alpha=1}^{M} \left\langle \mu^k_\alpha, n^{k+1}_\alpha - n^k_\alpha \right\rangle_c.$$  (4.18)
By an approach similar to that used in (3.8), we can obtain

\[
F_{\Sigma,h}^{k+1} - F_{\Sigma,h}^k \leq \sum_{\alpha,\beta=1}^M c_{\alpha\beta} \left( \delta^c_{\alpha} [n_{\alpha}^{k+1}] - \delta^c_{\alpha} [n_{\alpha}^k], \delta^c_{\beta} [n_{\beta}^{k+1}] \right)_u
+ \sum_{\alpha,\beta=1}^M c_{\alpha\beta} \left( \delta^c_{\alpha} [n_{\alpha}^{k+1}] - \delta^c_{\alpha} [n_{\alpha}^k], \delta^c_{\beta} [n_{\beta}^{k+1}] \right)_v
\]

\[
(4.22)
\]

where \( \mu_{\alpha}^{\Sigma,k+1} \) is defined in (4.9b). From (4.18) and (4.19), we get

\[
F_{h}^{k+1} - F_{h}^k \leq h^2 \sum_{\alpha=1}^M \left( \mu_{\alpha}^{k+1}, n_{\alpha}^{k+1} - n_{\alpha}^k \right)_c
\]

\[
\leq -h \sum_{\alpha=1}^M \left( \mu_{\alpha}^{k+1}, \delta^v [u^k \Lambda^c_x [n_{\alpha}^k | u^k]] + \delta^v [v^k \Lambda^c_y [n_{\alpha}^k | v^k]] \right)_c
\]

\[
- h \sum_{\alpha=1}^M \left( \mu_{\alpha}^{k+1}, \delta^v [J_x^{k+1}] + \delta^v [J_y^{k+1}] \right)_c
\]

\[
\leq -h \sum_{\alpha=1}^M \left( \mu_{\alpha}^{k+1}, \delta^v [u^k \Lambda^c_x [n_{\alpha}^k | u^k]] + \delta^v [v^k \Lambda^c_y [n_{\alpha}^k | v^k]] \right)_c
\]

\[
- \sum_{\alpha,\beta=1}^M \left( M_{\alpha\beta} (A^c_x [n^k]) \delta^c_{\alpha} [\mu_{\alpha}^{k+1}], \delta^c_{\beta} [\mu_{\beta}^{k+1}] \right)_u
\]

\[
- \sum_{\alpha,\beta=1}^M \left( M_{\alpha\beta} (A^c_y [n^k]) \delta^c_{\alpha} [\mu_{\alpha}^{k+1}], \delta^c_{\beta} [\mu_{\beta}^{k+1}] \right)_v
\]

(4.20)

Thanks to the property of \( M \), (4.17) is derived.

Lemma 4.2. The difference between \( E_{*,h}^{k} \) and \( E_{h}^{k} \) satisfies

\[
\frac{E_{*,h}^{k+1} - E_{*,h}^{k}}{\tau_k} \leq h \sum_{\alpha=1}^M \left( \mu_{\alpha}^{k+1}, \delta^v [u^k \Lambda^c_x [n_{\alpha}^k | u^k]] + \delta^v [v^k \Lambda^c_y [n_{\alpha}^k | v^k]] \right)_c
\]

(4.21)

Proof. Using the fully discrete formulations of intermediate velocity, we derive the difference between \( E_{*,h}^{k} \) and \( E_{h}^{k} \) as

\[
\frac{E_{*,h}^{k+1} - E_{*,h}^{k}}{\tau_k} \leq \frac{h^2}{\tau_k} \left( \delta^c_{\alpha} [u^k A^c_y [\rho^k]] \right)_u + \frac{h^2}{\tau_k} \left( \delta^c_{\alpha} [v^k A^c_y [\rho^k]] \right)_v
\]

\[
\leq -h \sum_{\alpha=1}^M \left( \left( u^k A^c_x [n_{\alpha}^k | u^k], \delta^c_{\alpha} [\mu_{\alpha}^{k+1}] \right)_u + \left( v^k A^c_y [n_{\alpha}^k | v^k], \delta^c_{\alpha} [\mu_{\alpha}^{k+1}] \right)_v \right).
\]

(4.22)

Applying (4.7a) and (4.7b) to (4.22) yields (4.21).

Lemma 4.3. The kinetic energies \( E_{h}^{k+1} \) and \( E_{*,h}^{k} \) satisfy

\[
E_{h}^{k+1} - E_{*,h}^{k} \leq 0.
\]

(4.23)
Applying (3.11), we can derive that

\[
E_h^{k+1} - E_h^k \leq \frac{h^2}{\tau_k} \left( A_x^e[\rho^{k+1}]u^{k+1} - A_x^e[\rho^k]u^k, u^{k+1} \right)_u \\
- \frac{\tau_k}{2} \left( A_x^e[\rho^{k+1}] - A_x^e[\rho^k], |u^{k+1}|^2 \right)_u \\
+ \frac{\tau_k}{2} \left( A_y^e[\rho^{k+1}]v^{k+1} - A_y^e[\rho^k]v^k, v^{k+1} \right)_v \\
- \frac{\tau_k}{2} \left( A_y^e[\rho^{k+1}] - A_y^e[\rho^k], |v^{k+1}|^2 \right)_v,
\]  

(4.24)

For \( u \in \mathcal{S}_u^0 \cup \mathcal{S}_v^0 \) and \( v \in \mathcal{S}_u^0 \cup \mathcal{S}_v^0 \), we can deduce by direct calculations that

\[
A_x^e[\delta_x^i u]_{i,j,\frac{1}{2}} = \delta_x^i [A_x^e[u]]_{i,j,\frac{1}{2}}, \quad 1 \leq i \leq N_x - 1, \quad 0 \leq j \leq N_y - 1,
\]

(4.25a)

\[
A_x^e[\delta_y^j v]_{i,\frac{1}{2},j} = \delta_y^j [A_x^e[u]]_{i,\frac{1}{2},j}, \quad 1 \leq i \leq N_x - 1, \quad 0 \leq j \leq N_y - 1,
\]

(4.25b)

\[
A_y^e[\delta_x^i u]_{i,\frac{1}{2},j} = \delta_x^i [A_y^e[u]]_{i,\frac{1}{2},j}, \quad 0 \leq i \leq N_x - 1, \quad 1 \leq j \leq N_y - 1,
\]

(4.25c)

\[
A_y^e[\delta_y^j v]_{i,j,\frac{1}{2}} = \delta_y^j [A_y^e[v]]_{i,j,\frac{1}{2}}, \quad 0 \leq i \leq N_x - 1, \quad 1 \leq j \leq N_y - 1.
\]

(4.25d)

Applying (4.25) to the fully discrete component mass balance equations, we obtain

\[
A_x^e[\delta_x^i u]_{i,j,\frac{1}{2}} - A_x^e[\rho^k]_{i,j,\frac{1}{2}} + \frac{1}{h} \left( \delta_x^i [U^k]_{i,j,\frac{1}{2}} + \delta_y^j [V^k]_{i,j,\frac{1}{2}} \right) = 0,
\]

(4.26)

\[
A_y^e[\delta_y^j v]_{i,\frac{1}{2},j} - A_y^e[\rho^k]_{i,\frac{1}{2},j} + \frac{1}{h} \left( \delta_y^j [U^k]_{i,\frac{1}{2},j} + \delta_y^j [V^k]_{i,\frac{1}{2},j} \right) = 0,
\]

(4.27)

where (4.26) holds for \( 1 \leq i \leq N_x - 1 \) and \( 0 \leq j \leq N_y - 1 \), while (4.27) holds for \( 0 \leq i \leq N_x - 1 \) and \( 1 \leq j \leq N_y - 1 \). Substituting (4.14), (4.15), (4.26), and (4.27) into (4.24) yields

\[
E_h^{k+1} - E_h^k \leq h (T_1 + T_2 + T_3 + T_4 + T_5 + T_6 + T_7),
\]

(4.28)

where

\[
T_1 = \frac{1}{2} \left( \delta_x^i [U^k]_{i,j,\frac{1}{2}} [u^{k+1}]_{i} + \delta_x^i [U^k]_{i,j,\frac{1}{2}} [u^k]_{i} \right)_u,
\]

\[
T_2 = \frac{1}{2} \left( \delta_y^j [V^k]_{i,\frac{1}{2},j} [u^{k+1}]_{i} + \delta_y^j [V^k]_{i,\frac{1}{2},j} [u^k]_{i} \right)_u,
\]

\[
T_3 = \frac{1}{2} \left( \delta_x^i [U^k]_{i,j,\frac{1}{2}} [v^{k+1}]_{j} + \delta_x^i [U^k]_{i,j,\frac{1}{2}} [v^k]_{j} \right)_v,
\]

\[
T_4 = \frac{1}{2} \left( \delta_y^j [V^k]_{i,\frac{1}{2},j} [v^{k+1}]_{j} + \delta_y^j [V^k]_{i,\frac{1}{2},j} [v^k]_{j} \right)_v,
\]
\[ T_5 = \langle \delta_x^c [\Phi^{k+1}], u^{k+1} \rangle_u + \langle \delta_y^c [\Phi^{k+1}], v^{k+1} \rangle_v, \]
\[ T_6 = \langle \delta_x^c [\Psi^{k+1}], u^{k+1} \rangle_u + \langle \delta_y^c [\Upsilon^{k+1}], v^{k+1} \rangle_v, \]
\[ T_7 = \langle \delta_y^p [\Theta^{k+1}], u^{k+1} \rangle_u + \langle \delta_x^p [\Theta^{k+1}], v^{k+1} \rangle_v. \]

For the term \( T_1 \), using (4.7a) and (4.7b), we can deduce that
\[
T_1 = \langle U_u^k \Lambda_y^u [u^{k+1}], \delta_y^u [u^{k+1}] \rangle_c - \frac{1}{2} \langle U_u^k, \delta_y^u [u^{k+1}]^2 \rangle_c
= \langle U_u^k (\Lambda_x^u [u^{k+1}] + \Lambda_y^u [u^{k+1}]), \delta_x^u [u^{k+1}] \rangle_c
= -\frac{1}{2} \langle U_u^k, \delta_y^u [u^{k+1}], \delta_x^u [u^{k+1}] \rangle_c.
\]

Applying (4.7c) to \( T_2 \) yields
\[
T_2 = \langle V_u^k \Lambda_y^p [u^{k+1}], \delta_y^u [u^{k+1}] \rangle_{pu} - \frac{1}{2} \langle V_u^k, \delta_y^u [u^{k+1}]^2 \rangle_{pu}
= -\frac{1}{2} \langle V_u^k, \delta_y^u [u^{k+1}], \delta_y^u [u^{k+1}] \rangle_{pu}.
\]

This is similar to deducing that
\[
T_3 + T_4 = -\frac{1}{2} \langle V_v^k, \delta_x^u [u^{k+1}], \delta_x^u [v^{k+1}] \rangle_{pv} - \frac{1}{2} \langle V_v^k, \delta_y^u [v^{k+1}], \delta_y^u [v^{k+1}] \rangle_c.
\]

The terms \( T_5 \) and \( T_7 \) can be derived applying (4.7) as
\[
T_5 = -\langle \Phi^{k+1}, \delta_x^u [u^{k+1}] \rangle_c - \langle \Phi^{k+1}, \delta_y^u [v^{k+1}] \rangle_c
= -\frac{1}{h} \langle \lambda^k (\delta_x^u [u^{k+1}] + \delta_y^u [v^{k+1}]), \delta_x^u [u^{k+1}] + \delta_y^u [v^{k+1}] \rangle_c,
\]
\[
T_7 = -\langle \Theta^{k+1}, \delta_y^p [u^{k+1}] \rangle_{pu} - \langle \Theta^{k+1}, \delta_y^p [v^{k+1}] \rangle_{pv}
= -\frac{1}{h} \langle \eta^k \delta_y^p [u^{k+1}], \delta_y^p [u^{k+1}] \rangle_{pu}
\]
\[
\leq -\frac{1}{h} \sum_{i=1}^{N_x} \sum_{j=1}^{N_x-1} \eta_{i,j}^k \left( \delta_y^p [u^{k+1}] + \delta_y^p [v^{k+1}] \right)^2,
\]
where we have also used (4.12) and (4.13). We can see that the terms \( T_1 \) to \( T_7 \) are all less than zero, and thus we get (4.23).
Theorem 4.1. Fully discrete total (free) energies are dissipated with time steps

\[ E_h^{k+1} + \mathcal{F}_h^{k+1} \leq E_h^k + \mathcal{F}_h^k. \]  

Proof. Inequality (4.35) is immediately obtained combining (4.17), (4.21), and (4.23).

5. Numerical tests. In this section, the proposed methods are applied to simulate multicomponent two-phase flow problems. We consider a binary mixture and a ternary mixture in a square domain \( \Omega \) with length 20 nm. We take the boundary conditions as \( \mathbf{u} = 0, J_\alpha \cdot \gamma_{\partial \Omega} = 0, \) and \( \nabla n_\alpha \cdot \gamma_{\partial \Omega} = 0 \) on \( \partial \Omega, \) where \( \gamma_{\partial \Omega} \) is the normal unit outward vector to \( \partial \Omega. \) A uniform rectangular mesh with 40 \( \times \) 40 elements is used for spatial discretization. The energy parameters are chosen as \( C_{T,\alpha} = 0. \)

5.1. A binary mixture droplet. In this example, we consider a binary mixture composed of methane (\( C_1 \)) and pentane (\( C_5 \)) at a constant temperature 310 K. At the initial time, a square shape droplet is located in the center of the domain. The initial gas molar densities of \( C_1 \) and \( C_5 \) are 7.4302 kmol/m\(^3\) and 0.6736 kmol/m\(^3\), respectively, while the initial liquid molar densities of \( C_1 \) and \( C_5 \) are 6.8663 kmol/m\(^3\) and 4.7915 kmol/m\(^3\), respectively. The initial molar density distributions for \( C_1 \) and \( C_5 \) are illustrated in Figures 5.1(a) and 5.1(d), respectively. We use the diffusion mobility formulation given by (2.7) with the coefficients \( D_{12} = D_{21} = 10^{-8} \) m\(^2\)/s. The volumetric viscosity and the shear viscosity are taken as \( \xi = \eta = 10^{-4} \) Pa-s. The time step size is taken as \( 10^{-12} \) s, and 200 time steps are simulated.

The proposed velocity-density decoupled method is applied to simulate the dynamical evolution of the square-shaped droplet. In Figure 5.2, we show the evolution profiles of the modified total energy (i.e., the sum of the modified Helmholtz free energy and kinetic energy) with time steps; we also depict the original total energy (i.e., the sum of the original Helmholtz free energy and kinetic energy) for the sake

![Binary mixture droplet: molar densities of \( C_1 \) and \( C_5 \) at different time steps.](image-url)
of comparison. It is observed from Figure 5.2(a) that although the modified total (free) energy is slightly less than the original energy, both total (free) energies are strictly dissipated with time steps, and moreover Figure 5.2(b), which is a zoom-in plot of Figure 5.2(a) in the later time steps, demonstrates that both total (free) energies continue to decrease. Consequently, the proposed method can preserve the energy-dissipation feature.

Figure 5.1 depicts the evolution process of each component molar density, and it is clearly observed that the droplet is gradually reshaping to a circle from its initial square shape due to chemical potential gradients. In Figure 5.3, we show the fluid motion driven by chemical potential gradients, including the velocity field and magnitudes of both velocity components.

5.2. A binary mixture bubble. In this example, we consider the dynamics of a binary mixture (C$_1$ and C$_5$) bubble at a constant temperature of 310 K. We take the initial gas molar densities $[n_{G_1}^{T}, n_{G_5}^{T}] = [7.4302, 10^{-15}]$ kmol/m$^3$ and the initial liquid molar densities $[n_{L_1}^{T}, n_{L_5}^{T}] = [6.8663, 4.7915]$ kmol/m$^3$ as illustrated in Figures 5.5(a) and 5.5(d), respectively. In this example, we aim to simulate a situation where one component is absent somewhere at the initial time. But such a situation leads to the singularity of the bulk free energy and chemical potential based on PR-EOS. In order to reduce this singularity, we take a very small molar density value ($10^{-15}$ kmol/m$^3$) for C$_5$ in gas to represent virtually the absence of C$_5$ inside the bubble. The diffusion mobility and viscosity are the same as those in subsection 5.1. We apply the proposed
velocity-density decoupled method to simulate this problem for 700 time steps with time step size $5 \times 10^{-13}$ s. The original and modified total energies at different time steps are depicted in Figure 5.4. Although the absence of C$_5$ in gas causes a slight destabilization of the original total energies at the beginning of the evolutorial process, the modified total energies always decrease with time steps. Molar densities at different time steps are illustrated in Figure 5.5. In the dynamical process, we can observe from Figure 5.5 that C$_5$ in liquid is evaporating into the bubble to attain an equilibrium state.

5.3. Ternary mixture. In this example, we consider a ternary mixture composed of methane (C$_1$), pentane (C$_5$), and decane (C$_{10}$) at a constant temperature 323 K. The initial gas molar densities of C$_1$, C$_5$, and C$_{10}$ are $10.516$ kmol/m$^3$, $0.77$ kmol/m$^3$, and $0.184$ kmol/m$^3$, respectively, while the initial liquid molar densities of C$_1$, C$_5$, and C$_{10}$ are $7.8412$ kmol/m$^3$, $1.9925$ kmol/m$^3$, and $1.433$ kmol/m$^3$, respec-
tively. At the initial time, there are two square-shaped droplets in the domain, as shown in Figures 5.6(a), 5.7(a), and 5.8(a). The diffusion fluxes are formulated by (2.6) with the diffusion coefficients $D_\alpha = 3 \times 10^{-8}$ m$^2$/s ($1 \leq \alpha \leq 3$). The volumetric viscosity and the shear viscosity are set as $\xi = \eta = 10^{-4}$ Pa-s. We take the time step size equal to $10^{-12}$ s and 1,000 time steps.

We employ the proposed componentwise, decoupled numerical scheme. The orig-
Fig. 5.8. Ternary mixture: $C_{10}$ molar densities at the (a) initial, (b) 30th, (c) 100th, (d) 200th, (e) 500th, and (f) 1,000th time step.

Fig. 5.9. Ternary mixture: energy dissipation with time steps.

inal and modified total energies and their zoom-in plots are shown in Figure 5.9. We still see that both of total energies are dissipated with time steps. We also note that in practical computations, the componentwise method is really effective for the mixtures composed of multiple components since it only needs to solve one more mass balance equation as a new component is added.

Figures 5.6, 5.7, and 5.8 illustrate the molar density configurations of three components. The simulation results show that, due to chemical potential gradients, two droplets are first merging with each other, and at the later time, the merged droplets are gradually reshaping into a circle.

6. Conclusions. Two decoupled energy-stable numerical schemes are developed for multicomponent two-phase compressible flows with a realistic equation of state (e.g., Peng–Robinson equation of state). In these methods, the scalar auxiliary variable (SAV) approach is applied to deal with the bulk Helmholtz free energy, and moreover we propose a componentwise SAV approach, which is extremely efficient.
and easy to implement for multicomponent flows. In order to uncouple the tight relationship between velocity and molar densities, we introduce two intermediate velocities, one of which has a componentwise form matching the componentwise SAV approach. The intermediate velocities are involved in the discrete formulation of the momentum balance equation, which establishes the consistent relationships with the mass balance equations. The proposed numerical schemes only need to solve a sequence of linear equations at each time step. Moreover, the fully discrete schemes are constructed based on the finite difference/volume methods with the upwind scheme on staggered grids. Unconditional energy-dissipation laws of the semidiscrete and fully discrete schemes are proved rigorously. Numerical results validate the effectiveness of the proposed methods.

Appendix A. Helmholtz free energy density. Let \( R \) be the universal gas constant, and let \( T \) be the absolute temperature. Three contributions of Helmholtz free energy density \( f_b(n) \) based on PR-EOS are expressed as

\[
(A.1) \quad f_{b\text{ideal}}(n) = RT \sum_{\alpha=1}^{M} n_{\alpha} (\ln n_{\alpha} - 1), \quad f_{b\text{repulsion}}(n) = -nRT \ln (1 - bn),
\]

\[
(A.2) \quad f_{b\text{attraction}}(n) = \frac{a(T)n}{2\sqrt{2b}} \ln \left( \frac{1 + (1 - \sqrt{2})bn}{1 + (1 + \sqrt{2})bn} \right),
\]

where \( n = \sum_{\alpha=1}^{M} n_{\alpha} \) is the overall molar density. We denote by \( T_{c,\alpha} \) and \( P_{c,\alpha} \) the critical temperature and critical pressure of component \( \alpha \), respectively. Let the reduced temperature be \( T_{r,\alpha} = T/T_{c,\alpha} \). The parameters \( a_{\alpha} \) and \( b_{\alpha} \) are calculated as

\[
(A.3) \quad a_{\alpha} = \frac{0.45724 R^{2} T_{c,\alpha}^{2}}{P_{c,\alpha}} \left[ 1 + m_{\alpha}(1 - \sqrt{T_{r,\alpha}}) \right], \quad b_{\alpha} = \frac{0.07780 RT_{c,\alpha}}{P_{c,\alpha}}
\]

We denote by \( \omega_{\alpha} \) the acentric factor of component \( \alpha \). The coefficients \( m_{\alpha} \) are calculated by the following relations:

\[
m_{\alpha} = 0.37464 + 1.54226\omega_{\alpha} - 0.26992\omega_{\alpha}^{2}, \quad \omega_{\alpha} \leq 0.49,
\]

\[
m_{\alpha} = 0.379642 + 1.485030\omega_{\alpha} - 0.166423\omega_{\alpha}^{2} + 0.016666\omega_{\alpha}^{3}, \quad \omega_{\alpha} > 0.49.
\]

We denote \( y_{\alpha} = n_{\alpha}/n \). Then we calculate \( a(T) \) and \( b \) as

\[
(A.4) \quad a = \sum_{\alpha=1}^{M} \sum_{\beta=1}^{M} y_{\alpha}y_{\beta} (a_{\alpha}a_{\beta})^{1/2} (1 - k_{\alpha\beta}), \quad b = \sum_{\alpha=1}^{M} y_{\alpha}b_{\alpha},
\]

where \( k_{\alpha\beta} \) stands for the binary interaction coefficients. In numerical tests, we use \( k_{C1,C1} = k_{C5,C1} = 0.041, \; k_{C1,C5} = k_{C10,C1} = 0.05 \) and treat the others as zero.

We note that, as shown in (A.4), the parameters \( a \) and \( b \) are modified geometric mean and arithmetic mean of component values, respectively, which involve the composition \( y_{\alpha} (1 \leq \alpha \leq M) \), so \( f_{b\text{repulsion}} \) and \( f_{b\text{attraction}} \) depend not only on the overall molar density \( n \), but also on \( y_{\alpha} \) (i.e., relying on each component molar density).

We list some physical parameters of three substances in Table A.1.
First, we formulate the influence parameter $c_{\alpha}$ of component $\alpha$ as \cite{36}

$$c_{\alpha} = a_{\alpha} a_{\alpha}^{2/3} \left[ \gamma_{\alpha} (1 - T_{c,\alpha}) + \phi_{\alpha} \right], \quad \gamma_{\alpha} = \frac{-10^{-16}}{1.2326 + 1.3757 \omega_{\alpha}}, \quad \phi_{\alpha} = \frac{10^{-16}}{0.9051 + 1.5410 \omega_{\alpha}}. $$

The cross influence parameter is calculated as

$$c_{\alpha \beta} = (1 - w_{\alpha \beta}) \sqrt{c_{\alpha} c_{\beta}},$$

where $w_{\alpha \beta}$ are the binary interaction coefficients satisfying the symmetry $c_{\alpha \beta} = c_{\beta \alpha}$ and $w_{\alpha \alpha} = 0$, $0 \leq w_{\alpha \beta} < 1$. In numerical tests, we take $w_{\alpha \beta} = 0.5$ for $\alpha \neq \beta$.

## References

\[ \begin{align*}
\]
LINEARLY DECOUPLED METHODS FOR TWO-PHASE FLOW


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