PHASE TRANSITION AND SEPARATION
IN COMPRESSIBLE CAHN-HILLIARD FLUIDS

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ABSTRACT. The paper provides a scheme for phase separation and transition by accounting for diffusion, dynamic equations and consistency with thermodynamics. The constituents are compressible fluids thus improving the model of a previous approach. Moreover a possible saturation effect for the concentration of a constituent is made explicit. The mass densities of the constituents are independent of temperature. The evolution of concentration is described by the standard equation for mixtures but the balance of energy and entropy of the mixture are stated as for a single constituent. However, due to the non-simple character of the mixture, an extra-energy flux is allowed to occur. Also motion and diffusion effects are considered by letting the stress in the mixture have additive viscous terms and, remarkably, the chemical potential contains a quadratic term in the stretching tensor. As a result a whole set of evolution equations is set up for the concentration, the velocity, and the temperature. Shear-induced mixing and demixing are examined. A maximum theorem is proved which implies that the concentration of the mixture has values from 0 to 1 as is required from the physical standpoint.

1. Introduction. Cahn-Hilliard fluids are meant as mixtures of immiscible fluids where the mean composition may be uniform in space or rather subregions occur with a predominant concentration of one of the two constituents. We can view the two settings as two phases of a continuum (mixture). In dealing with phase transitions, the two phases may be thought of as separated by a sharp interface between the phases. Alternatively, the transition may be described by phase-field models where the sharp interface is replaced by a narrow transition layer across which the fluids may mix. In the transition region, diffusion and motion of the
mixture occur and the mixture is described by an additional field, that of an order parameter. This is the essence of phase-field models (see [2, §4.4] and [3, 4, 5]).

Owing to the jump of mass density across a transition layer, the modelling allows for non-uniform densities and, mathematically, for a dependence of energy also on the mass density gradients. This idea traces back to van der Waals [6] and is at the basis of the Cahn-Hilliard model [7] for a purely diffusional problem.

Improvements of the model have been set up to account for phase separation where the (two) components of the mixture form separated or homogeneous domains. In this regard a realistic description has to involve the coupling of the equations of fluid dynamics with the occurrence of diffusion [8, 9, 10].

This view is at the basis of [11] where the dependence of the energy upon concentration gradients plays a central role both for balance equations and for constitutive properties.

The purpose of this paper is to provide a scheme for phase separation and shear-induced mixing [12] by accounting for diffusion, dynamic equations and consistency with thermodynamics. In this regard we mention that thermally-induced phase transitions in fluids are modelled in [13] by allowing for appropriate dependences on gradients (of temperature, mass density and phase field) and for an extra flux. In a recent paper [14], a model for phase separation in mixtures is set up within a phase-field approach where the concentration \( c \) of a constituent is the order parameter. The key point is the observation that a linear dependence of the stress tensor on the time derivative \( \dot{c} \) may be viewed as an additive term of the chemical potential that is quadratic in the stretching tensor. Two improvements of the model are in order. First, the constituents are allowed to be compressible. Though the incompressibility of the constituents is reasonable in many contexts, certainly the compressibility is a clear improvement which in turn avoids technical problems for the evolution equations due to the incompressibility constraint [14]. Second, the data on the whole mass of the constituents have to enter the model and to characterize the uniform solution. Such improvement is realized in this paper by means of an appropriate free-energy potential.

To keep the model as simple as possible we account for the evolution of concentration but look at the balance of energy and entropy as for a single constituent. However, due to the internal structure of the mixture we allow for an extra-energy flux, in addition to the heat flux, and an extra-entropy flux. We also account for motion and diffusion effects by letting the stress in the mixture have additive viscous terms. As a result we obtain a whole set of evolution equations for the concentration, the velocity, and the temperature through the balance of mass, linear momentum, and energy. Next we show that an appropriate free-energy potential allows for a thermally-induced phase transition and a nonzero stretching lowers the transition temperature. Also we examine the effect of stretching on the concentration and conditions for mixing and demixing are exhibited. Finally, without any assumption on the mobility except the non-negative valuedness, we are able to prove a maximum principle theorem which implies that the concentration of the mixture remains between 0 and 1 at any point and at any time, as is required from the physical standpoint. Appendices A and B provide the derivation of the thermodynamic restrictions and the relation to similar approaches to the Cahn-Hilliard fluids.

2. Mixture of fluid constituents. We consider a binary non-reacting mixture, occupying a possibly time-dependent region \( \Omega \subset \mathbb{R}^3 \). We allow for two possibilities,
namely that both constituents are compressible (fluids) or that only one of them is compressible.

Let \( \rho_1, \rho_2 \) be the mass densities of the two constituents. Hence
\[
\rho = \rho_1 + \rho_2
\]
is the mass density of the mixture and
\[
c = \frac{\rho_1}{\rho}
\]
is the concentration (or mass fraction) of constituent 1. The mass densities \( \rho, \rho_1, \rho_2 \) and the concentrations \( c, 1 - c \) are functions of the position \( x \in \Omega \) and time \( t \in \mathbb{R} \).

If the constituents are incompressible then \( \rho_1, \rho_2 \) are constants but \( \rho \) depends on \( x \) and \( t \) unless \( \rho_1 = \rho_2 \). The mass densities \( \rho_1, \rho_2 \) are related to \( \rho \) and \( c \) by
\[
\rho_1 = \rho c, \quad \rho_2 = \rho (1 - c).
\]
(2.1)
It is then equivalent to describe the mixture densities by \( \rho_1, \rho_2 \) or \( \rho, c \). To account for diffusion it is more convenient to regard \( \rho \) and \( c \) as independent variables.

Let \( M_1, M_2 \) be the whole mass of the constituents. Owing to the mass conservation,
\[
\int_{\Omega} \rho(x, t)c(x, t)dv = M_1.
\]
(2.2)
Later on we denote by \( c^* \) the constant mean value of \( c \),
\[
c^* = \frac{M_1}{M_1 + M_2}.
\]
### 2.1. Balance equations.
We now state the local form of the balance equations. Let \( \mathbf{v}_1, \mathbf{v}_2 \) be the velocities of the constituents and
\[
\mathbf{v} = c\mathbf{v}_1 + (1 - c)\mathbf{v}_2
\]
the barycentric velocity. The balance of mass for the mixture as a whole provides the classical continuity equation
\[
\dot{\rho} = -\nabla \cdot \mathbf{v}.
\]
(2.3)
Let \( \mathbf{u} \) be the diffusion velocity and \( \mathbf{j} \) the mass flux,
\[
\mathbf{u} = \mathbf{v}_1 - \mathbf{v}, \quad \mathbf{j} = \rho c \mathbf{u}.
\]
The balance of mass for constituent 1, in a non-reacting mixture, provides
\[
\rho \dot{c} = -\nabla \cdot \mathbf{j}.
\]
(2.4)
The global conservation of mass (2.2) implies that
\[
0 = \frac{d}{dt} \int_{\Omega} \rho c dv = \int_{\Omega} \rho \dot{c} dv.
\]
By (2.4) and the divergence theorem we have
\[
\int_{\partial \Omega} \mathbf{j} \cdot \mathbf{n} da = 0,
\]
(2.5)
where \( \mathbf{n} \) is the unit outward normal to the boundary \( \partial \Omega \) of \( \Omega \). For definiteness we account for (2.5) by letting
\[
\mathbf{j} \cdot \mathbf{n} = 0 \quad \text{at} \quad \partial \Omega.
\]
(2.6)
We might go on and consider the evolution equation for \( j \) [15, 16]. For simplicity, though, following a standard view in the modelling of mixtures, we regard \( j \) as an unknown function subject to thermodynamic restrictions.

The balance of linear momentum for the mixture as a whole is written in the standard form,

\[
\rho \dot{v} = \nabla \cdot T + \rho f,
\]

where \( T \) is the stress tensor and \( f \) is the body force density. The stress \( T \) is taken to be symmetric, as it follows from the balance of angular momentum.

Let \( q \) be the heat flux. The balance of energy is taken in the form

\[
\rho \left( e + \frac{1}{2} v^2 \right) = \nabla \cdot (T v - q) - \nabla \cdot w + \rho f \cdot v + \rho r,
\]

where \( e \) is the energy density and \( r \) is the heat supply (per unit mass). In view of (2.7) it follows that

\[
\rho \dot{e} = T \cdot L - \nabla \cdot q - \nabla \cdot w + \rho r,
\]

where \( L \) is the velocity gradient.

We represent the entropy flux as \( q / \theta \), where \( \theta \) is the absolute temperature. The second law is taken as the statement that the entropy density \( \eta \) satisfies the inequality

\[
\rho \dot{\eta} \geq \frac{\rho r}{\theta} - \nabla \cdot \frac{q}{\theta}
\]

for every process compatible with the balance equations (2.3), (2.4), (2.7), (2.8). Substitution of \( \rho r - \nabla \cdot q \) from (2.8) and use of the free energy \( \psi = e - \theta \eta \) provide the Clausius-Duhem (or second law) inequality in the form

\[
- \rho (\dot{\psi} + \eta \dot{\theta}) + T \cdot L - \frac{1}{\theta} q \cdot \nabla \theta - \nabla \cdot w \geq 0.
\]

As a comment we remark that a motivation for the occurrence of an extra-energy flux follows from the mixture model (see, e.g., [15]). Summing over the energy balance equations for the single constituents, we obtain (2.8) with

\[
w = \sum_{\alpha} \left( \frac{1}{2} \rho_{\alpha} u_{\alpha}^2 (1 - T_{\alpha}) \right) u_{\alpha}, \quad u_{\alpha} = v_{\alpha} - v,
\]

where \( \alpha \) runs over the constituents, whereas \( q = \sum_{\alpha} q_{\alpha} \) is the sum of the single heat fluxes.

2.2. Constitutive equations. We now establish a set of constitutive equations; the detailed derivation is given in Appendix A. By analogy with [17], we can say that if the sole macroscopic manifestation of atomistic kinematics is the order parameter, here \( c \), then it seems reasonable that interatomic forces may be characterized macroscopically by fields that perform work when \( c \) undergoes changes. We mention that, in a model of incompressible constituents [11], a flux \( t c \), \( t \) being a generalized force, is considered to account for the energy balance. Also, in a phase-field model of deformation twinning, a surface term, and hence a flux, is considered that is proportional to the variation of the order parameter, here \( c \) (see (25) of [18]). This motivates that we take the stress tensor \( T \) as

\[
T = T_0 + 2\nu \dot{D} + \sigma (\nabla \cdot v) 1 + \dot{G} \left[ 2\beta \dot{D} + \alpha (\nabla \cdot v) 1 \right],
\]

(2.11)
where $T_0$ is independent of $D$ whereas $G$ is a function of $c$ so that $\dot{G} = G \dot{c}$. Also, a superposed ring $\circ$ denotes the deviatoric part. Since $\text{tr} L = \nabla \cdot v$ then
\[
\circ L = L - \frac{1}{3} (\nabla \cdot v) 1, \quad \circ D = D - \frac{1}{3} (\nabla \cdot v) 1.
\]
Yet compatibility with thermodynamics suggests that we proceed as follows.

We assume that $j$, $T_0$, $\eta$, and $w$ are determined by the set of variables $\Gamma = (\theta, \rho, c, \nabla \theta, \nabla c, D, ...)$, through appropriate constitutive functions, the dots indicating possible higher-order gradients of $\theta, c, D$. Also, we let $\psi$ be given by a $C^2$-function of $\Gamma_0 = (\theta, \rho, c, \nabla c)$.

It follows that (see Appendix A)
\[
\begin{align*}
\eta &= -\psi_\theta, \\
T_0 &= -\rho^2 \psi_\rho 1 - \rho \nabla c \otimes \psi_\nabla c, \quad \nu, \sigma \geq 0, \quad (2.12) \\
w &= \psi_\nabla c \nabla \cdot j + \mu j, \quad 1 \frac{1}{\beta} q \cdot \nabla \theta + j \cdot \nabla \mu \leq 0, \quad (2.13) \\
\mu &= \delta_c \psi - \frac{1}{\rho} G c D, \quad (2.14)
\end{align*}
\]

where
\[
\delta_c \psi = \psi_c - \frac{1}{\rho} \nabla \cdot (\rho \psi_\nabla c), \quad D := 2 \beta \circ D \circ D + \alpha (\nabla \cdot v)^2, \quad \beta < 0, \quad \alpha < 0,
\]
the subscripts $\theta, \rho$, and $c$ denoting partial derivatives.

3. Free energy. In addition to the standard linear viscosity terms, $2\nu \circ D$ and $\sigma(\nabla \cdot v) 1$, the effects of the stretching tensor $D$ occur in the additive terms $2\beta \dot{G} \circ D$ and $\alpha \dot{G} (\nabla \cdot v) 1$ of the stress tensor and $-G_c D / \rho$ of the potential $\mu$. Apart from the dissipative stretching-dependent terms, the stress tensor $T$, the potential $\mu$ and the internal energy $e$ are determined by the effective free-energy function $\tilde{\psi} = \psi - G D / \rho$.

For definiteness we now give an example of free-energy function which allows an immediate description of the phase transition.

Let
\[
\psi(\theta, \rho, c, \nabla c) = \Psi(\theta, \rho) + \theta G(c) + \Theta F(c) + \frac{1}{2} \chi(\theta, \rho, c)|\nabla c|^2. \quad (3.1)
\]
By (2.14) and (2.12) we have
\[ \mu = \Theta \left( F_c + \frac{\theta}{\Theta} G_c \right) - \frac{1}{\rho} \nabla \cdot (\chi \rho \nabla c) - \frac{G_c}{\rho} D + \frac{1}{2} \chi c |\nabla c|^2, \] (3.2)
\[ T = -P \mathbf{1} - \rho \chi \nabla c \otimes \nabla c + 2 \nu \nabla \cdot (\sigma \cdot \nabla) \mathbf{1}, \] (3.3)
where
\[ P = \rho^2 \Psi + \frac{1}{2} \rho^2 \chi |\nabla c|^2. \]

Also, we find that
\[ \eta = -\Psi_{\theta} - G - \frac{1}{2} \chi_{\theta} |\nabla c|^2 \]
and hence the internal energy becomes
\[ e = \Psi - \theta \Psi_{\theta} + \Theta F + \frac{1}{2} (\chi - \theta \chi_{\theta}) |\nabla c|^2. \]

Of course
\[ e_{\theta} = -\theta \Psi_{\theta\theta} - \frac{1}{2} \chi_{\theta\theta} |\nabla c|^2 \]
is the specific heat.

For the time being, we let \( F(c) \) and \( G(c) \) be defined on the reals. Next we will show that \( c \) is in fact subject to \( c \in [0, 1] \). The function \( F \) is required to be positive as \( c \neq 0, 1 \) and to vanish at \( c = 0, 1 \) along with the derivative \( F_{c} \). The function \( G \) is assumed to be negative as \( c \neq 0, 1 \) and to vanish at \( c = 0, 1 \) along with \( G_{c} \). For a simple qualitative analysis we select the functions \( F \) and \( G \) in the form
\[ F(c) = \begin{cases} F_1 \sin^4 \pi u(c) & c \in [0, 1], \\ F_2 c^2 (1 - c)^2 & \text{otherwise,} \end{cases} \]
\[ G(c) = \begin{cases} -2F_1 \sin^2 \pi u(c) & c \in [0, 1], \\ 0 & \text{otherwise,} \end{cases} \] (3.4)
where \( F_1 \) and \( F_2 \) are positive constants (\( F_1 / F_2 = 0.3 \) in figures 1 and 2) and \( u(c) \) is a monotone function on \([0, 1]\) subject to \( u(0) = 0, u(1) = 1, u(c_*) = 1/2 \). For definiteness we let
\[ u(c) = c^\gamma \sin \pi c/2, \quad c_0^\gamma = 1/\sqrt{2}. \]
This function makes the derivative,
\[ u'(c) = \pi c^\gamma \left[ \frac{\sin \pi c/2}{c} + \frac{\pi}{2} \cos \pi c/2 \right], \]
vanish at \( c = 0_+ \) and be positive as \( c \in (0, 1] \).

It is worth remarking that the potential \( G \) favours the approach of \( c \) to equilibrium in that
\[(c - c_*)G_c > 0, \quad c \neq c_* . \]

4. Transition temperature and stretching effect. We now ask for the condition characterizing transition. As we see in a moment, the mixture has two phases, one with a single solution (uniform concentration) \( c_* \), the other with a double solution, say \( c_-, c_+ \), for \( c \). The transition is characterized as the passage between the two phases. This is ascertained by investigating the stationary points of \( \hat{\psi} \), and hence of \( \psi \), with respect to \( c \). Accordingly we examine the vanishing of \( \mu = \delta_c \hat{\psi} \).
To see the effect of motion on the transition we restrict attention to homogeneous regions and stationary conditions. We then let $\theta, \rho, c$ and $D$ be uniform, so that $\nabla \mu = 0$, and $\dot{c} = 0$. Hence $\mu$ reduces to

$$
\mu = \hat{\psi}_c = \left( \theta - \frac{1}{\rho} D \right) G_c + \Theta F_c.
$$

By (3.4) we obtain

$$
\mu = \begin{cases} 
2F_1 \pi \sin 2\pi u [\Theta \sin^2 \pi u - (\theta - D/\rho)]u', & c \in [0, 1], \\
2\Theta F_2 c (c - 1)(2c - 1), & \text{otherwise.}
\end{cases}
$$

The nonzero value of $\mu$ outside the physical interval $c \in [0, 1]$, gives relevance to the maximum principle theorem whereby $c$ is confined to $[0, 1]$.

Separation of phases is induced by changes in the convexity/concavity of $\hat{\psi}$ and such changes are related to the passage from maxima to minima or vice versa. Hence the analysis of stationary points of $\hat{\psi}$ is relevant for separation. We then require $\mu = 0$, which, by (3.4), provides

$$
\sin 2\pi u \left( \theta - \frac{1}{\rho} D - \Theta \sin^2 \pi u \right) u' = 0.
$$

The factor $\sin 2\pi u$ in (4.5) implies that the solutions $u = 0, 1/2, 1$, whence $c = 0, c_-, 1$, always occur. To understand the effect of the second factor, preliminarily let $\alpha, \beta = 0$, and hence $D = 0$, so that (4.5) becomes

$$
\sin 2\pi u (\theta - \Theta \sin^2 \pi u) = 0.
$$

If $\theta \geq \Theta$ then no additional solution occurs. Further, $\Theta F + \theta G$ has a maximum at $c = 0, 1$ and a minimum at $c = c_*$. If, instead, $\theta < \Theta$ then also the solutions

$$
u_+ - \frac{1}{\pi} \arcsin \sqrt{\theta/\Theta}, \quad u_+ = \frac{1}{\pi} \arcsin \sqrt{\theta/\Theta}
$$

occur with $u_- < 1/2 < u_+$. Because $u$ is monotone increasing then the corresponding solutions $c_-, c_+$ for $c$ satisfy $c_- < c_* < c_+$. While $c = c_*$ represents a mixture with a uniform composition (induced by the whole masses $M_1, M_2$ of the constituents), the solution $c = c_-$ denotes a (local) composition with $c < c_*$ whereas $c = c_+$ means that $c > c_*$. This allows $\Theta$ to be viewed as the transition temperature.

Let now $\alpha, \beta < 0$ and hence $D < 0$ and (4.5) becomes

$$
\sin 2\pi u (\hat{\theta} - \Theta \sin^2 \pi u) u' = 0,
$$

where

\begin{itemize}
  \item $\hat{\theta} = \theta - \frac{1}{\rho} D - \Theta$ for $c \in [0, 1]$,
  \item $\hat{\theta} = 2\Theta F_2 c (c - 1)(2c - 1)$ for $c \not\in [0, 1]$.
\end{itemize}
where
\[ \tilde{\theta} = \theta + \frac{1}{\rho} |D|. \]
As a consequence, the transition occurs at
\[ \tilde{\theta} = \Theta \]
namely at
\[ \theta = \Theta - \frac{1}{\rho} |D|. \]
This means that if the mixture undergoes a motion, with a non-zero \( D \), then the transition occurs at a lower temperature, the decrease being just \(|D|/\rho\).

Two remarks are in order. The function \( F \) may be selected as a fourth-order function, \( F(c) = 16c^2(c - 1)^2 \) while \( G \) is a smooth piecewise quadratic function. Examples in this sense are given in [11] and [14] (see also [1]). To our mind the functions (3.4) are more convenient in that they allow the second derivative of \( F \), at \( c = 0 \) and \( c = 1 \), to vanish thus avoiding spurious separation of components at \( c = 0 \), \( 1 \) and \( \theta > \Theta \). As the second remark, the decrease of the transition temperature may be viewed as a stretching-induced mixing [12].

It is of interest to observe that at the transition, \( \mu = 0 \), \( \tilde{\theta} = \Theta \), the free energy \( \psi \) has a point of inflection at \( c = c^* \), as is shown by Fig. 2. This is proved by evaluating the second-order derivative. Since
\[ \psi_{cc} = 4F_1\pi^2(u')^2[\Theta \sin^2 \pi u - \tilde{\theta}] + 2F_1\pi^2(u')^2 \sin^2 2\pi u \Theta + 2\pi^2 \sin 2\pi u [\Theta \sin^2 \pi u - \tilde{\theta}] \]
and each term on the right-hand side vanishes, then \( \psi_{cc} = 0 \) at \( c = c^* \).

5. Evolution equations. Substitution of \( T, w \) and \( e \) into (2.3), (2.4), (2.7), (2.8) provide the differential equations for \( \rho, c, v, \theta \). To this end we evaluate the energy density
\[ e = \psi + \theta \eta \psi = \psi - \theta \psi_{\theta}. \]
For formal simplicity we let
\[ \chi = \theta \chi_0, \]
\( \chi_0 \) being a constant. Hence, by (3.1), we obtain
\[ e = \Psi(\theta, \rho) - \theta \Psi_{\theta}(\theta, \rho) + \Theta F(c), \]
(5.1)
\[ \rho \mu = \rho \Theta F_c + (\rho \theta - D)G_c - \nabla \cdot (\chi_0 \rho \theta \nabla c). \]
(5.2)
As a consequence,
\[ \rho \dot{e} = -\rho \theta \Psi_{\theta\theta} \dot{\theta} - \rho^2 (\Psi_{\rho} - \theta \Psi_{\theta \rho}) \nabla \cdot v - \Theta F_c \nabla \cdot j. \]
Also, by (2.11),
\[ T \cdot L = -\rho^2 \Psi_{\rho} \nabla \cdot v - \rho \theta \chi_0 \nabla c \cdot D \nabla c + \dot{D} \]
where
\[ \dot{D} = 2\nu \dot{D} + \sigma (\nabla \cdot v)^2. \]
Hence, substitution of \( w \) from (2.13) makes the balance of energy (2.8) in the form
\[ -\rho \theta \Psi_{\theta\theta} \dot{\theta} = -\rho^2 \theta \Psi_{\theta\rho} \nabla \cdot v + \Theta F_c \nabla \cdot j - \rho \theta \chi_0 \nabla c \cdot D \nabla c + \dot{D} \]
\[ - \nabla \cdot (\rho j) - \nabla \cdot q + \rho r - \nabla \cdot [\theta \chi_0 (\nabla \cdot j) \nabla c]. \]
As a consequence, upon substitution of (8.8), the balance equations (2.4), (2.7), (2.8) result in
\[ \rho \dot{c} = \nabla \cdot (\kappa \nabla \mu), \] (5.3)
\[ \rho \dot{\Omega} = -\nabla P - \nabla \cdot (\rho \theta \chi_0 \nabla c \otimes \nabla c) + \nabla \cdot [2 \nu \dot{\Theta} D] + \nabla [\sigma \nabla \cdot \mathbf{v}] + \rho f, \] (5.4)
\[ -\rho \theta \Psi \theta \theta \dot{\theta} = -\rho \theta \Psi_0 \dot{\theta} = -\rho^2 \theta \Psi_0 \nabla \cdot \mathbf{v} - \Theta F \nabla \cdot (\kappa \nabla \mu) - \rho \theta \chi_0 \nabla c \cdot \mathbf{D} \nabla c + \Delta \] (5.5)
The system (2.3) and (5.3)-(5.5) in the unknowns \( \theta, \rho, c, \mathbf{v} \) is equipped with the boundary conditions
\[ \nabla \mu \cdot \mathbf{n} = 0, \quad \nabla c \cdot \mathbf{n} = 0, \quad \mathbf{v} = 0, \quad \text{at } \partial \Omega. \] (5.6)
Equations (2.3) and (5.3)-(5.5) may be named a Cahn-Hilliard-Navier-Stokes system. A simpler case, subject to the incompressibility constraint \( \nabla \cdot \mathbf{v} = 0 \), is investigated in [20] in connection with instability of flows.

6. Stretching-induced mixing and phase separation. The diffusion equation (2.4) indicates that the evolution of the concentration is governed by second-order spatial derivatives, the flux vector \( \mathbf{j} \) being related to gradients of appropriate fields. Here we look at the mixing effect of the stretching and argue as follows.

By (2.4) and (8.8) we have
\[ \rho \dot{c} = \nabla \cdot (\kappa \nabla \mu). \]
By (3.2), for simplicity we disregard the dependence of \( \mu \) on \( \nabla c \) and let
\[ \mu = \hat{\mu}(\theta, \rho, c) - \frac{1}{\rho} G c D. \]
For ease of interpretation we restrict attention to a region where \( \rho \) and \( \theta \) are uniform. Also we linearize with respect to \( \kappa \) to have
\[ \dot{c} = \frac{\kappa}{\rho} [(\hat{\mu}_c - \frac{1}{\rho} G_{cc} D) \Delta c + (\hat{\mu}_{cc} - \frac{1}{\rho} G_{cc} D) |\nabla c|^2 - \frac{1}{\rho} (G_c \Delta D + 2 G_{cc} \nabla c \cdot \nabla D)], \]
where \( \hat{\mu} = \hat{\psi}_c \).

In a fluid at rest we find the classical diffusion equation with
\[ \frac{\kappa \hat{\mu}_c}{\rho} \]
as the diffusion parameter. A non-zero stretching induces a change of the diffusion parameter
\[ \hat{\mu}_c \rightarrow \hat{\mu}_c - \frac{1}{\rho} G_{cc} D. \]
Now by the model of §6,
\[ \hat{\mu}_c - \frac{1}{\rho} G_{cc} D = \left( \theta - \frac{1}{\rho} D \right) G_{cc} + \Theta F c. \]
The stretching effect makes \( \theta - D/\rho \) to be the effective temperature and hence even the transition temperature is changed by \( -D/\rho \).

The occurrence of second-order spatial derivatives, here of \( D \), in the evolution equation is common to various approaches. For instance, [12] and refs therein add a term \(-(1/\rho) \nabla \cdot \sigma \) to \( \nabla \mu \), \( \sigma \) being a (viscous) stress tensor. We are unaware of the proof of compatibility of such approach with thermodynamics in the sense of §2. However, in such a case the stretching effect is modelled by \( \nabla \cdot (\nabla \cdot \sigma) \) and...
hence again second-order derivatives of $D$ occur in the generalized Cahn-Hilliard equation.

7. **Maximum principle theorem.** Physically, the concentration $c$ is meaningful in $[0, 1]$. It is then natural to ask whether the evolution equations so obtained, (2.3) and (5.3) to (5.5), imply that $c \in [0, 1]$. For simplicity we let $\chi$ be a constant. In addition, $\Omega$ is a time-dependent region and $\rho$ is allowed to be a function of $x \in \Omega$ and $t \in \mathbb{R}^+$. We then look at the evolution equation (5.3) for $c \in \mathbb{R}$ and let

$$\mu = \theta G_c + \Theta F_c - \frac{1}{\rho} \nabla \cdot (\chi \rho \nabla c) - \frac{1}{\rho} G_c D,$$

(7.1)

where $F$ and $G$ are given by (3.4). Now, $F > 0$ as $c \neq 0, 1$ and $F(0) = F(1) = 0$. Also,

$$G_c(c) = \begin{cases} -2\pi F_1 u'(c) \sin 2\pi u(c), & \text{if } c \in [0, 1], \\ 0, & \text{otherwise}. \end{cases}
\quad (7.2)$$

Accordingly, $G_c$ is continuous as $c \in \mathbb{R}$ and $G_c = 0$ as $c \leq 0$ and $c \geq 1$.

No assumption is placed on the mobility $\kappa$, except the non-negative valuedness; the dependence on $\theta$ is understood and not written and hence we write $\kappa(\rho, c)$ for the mobility $\kappa$. Further we suppose

$$\kappa(\rho, c) \nabla \mu \cdot \nabla \mu + \chi \rho \nabla c \cdot D \nabla c \geq 0.$$

(7.3)

In words, (7.3) means that $D$, if negative definite, is not too large relative to the first term. As a consequence, $\kappa$ is required to be positive definite.

We now look at (5.4) and consider the initial-value problem

$$\rho \dot{c} = \nabla \cdot [\kappa(\rho, c) \nabla \mu(\rho, c)], \quad c(x, 0) = c_0(x), \quad x \in \Omega.
\quad (7.4)$$

**Theorem.** Subject to (7.3), if $c_0(x) \in [0, 1]$ for each $x \in \Omega$ then the solution $c(x, t)$ to (7.4) takes values in $[0, 1]$ for each $x \in \Omega$ and each $t \in \mathbb{R}^+$.

**Proof.** For technical convenience let

$$c_- = \begin{cases} 0, & c \geq 0, \\ c, & c < 0. \end{cases}
\quad (7.5)$$

Hence

$$c_-(x, 0) = 0, \quad \nabla c_-(x, 0) = 0, \quad F(c_-(x, 0)) = 0, \quad \forall x \in \Omega.
\quad (7.5)$$

By (7.2) we have

$$G_c(c_-) = 0, \quad c \in \mathbb{R},
\quad (7.6)$$

and hence

$$\mu(\rho, c_-) = \Theta F_c(c_-) - \frac{\chi}{\rho} \nabla \cdot (\rho \nabla c_-), \quad c \in \mathbb{R}.
\quad (7.6)$$

Multiply the differential equation in (7.4) by $\mu(c_-)$. Integration on $\Omega$ and use of the divergence theorem along with the boundary condition (5.6) gives

$$\int_\Omega \rho \dot{c} \mu(\rho, c_-) dv = - \int_\Omega \kappa(\rho, c) \nabla \mu(\rho, c) \cdot \nabla \mu(\rho, c_-) dv.
\quad (7.7)$$

Look at the right-hand side of (7.7). By (7.6), $\nabla \mu(c_-)$ vanishes where $c \geq 0$ and hence

$$\int_\Omega \kappa(\rho, c) \nabla \mu(\rho, c) \cdot \nabla \mu(\rho, c_-) dv = \int_\Omega \kappa(\rho, c_-) \nabla \mu(\rho, c_-) \cdot \nabla \mu(\rho, c_-) dv \geq 0.
\quad (7.8)$$
We now consider the left-hand side of (7.7) and apply (7.6) to obtain
\[\int_{\Omega} \rho \dot{c} \mu(\rho, c_{-}) \, dv = \Theta \int_{\Omega} \rho \dot{c} F_{c}(c_{-}) \, dv - \chi_{0} \int_{\Omega} \dot{c} \nabla \cdot (\rho \nabla c_{-}) \, dv. \tag{7.9}\]
Examine the two integrals in the right hand side of (7.9). Since \(\nabla c_{-} \cdot \mathbf{n} = 0\) at \(\partial \Omega\), the divergence theorem gives
\[- \int_{\Omega} \dot{c} \nabla \cdot (\rho \nabla c_{-}) \, dv = \int_{\Omega} \rho \dot{c} \nabla \cdot \nabla c_{-} \, dv.\]
Because
\[\nabla [\dot{c} - (c_{-})] \cdot \nabla c_{-} = 0,\]
we can replace \(\dot{c}\) with \((c_{-})\) to obtain
\[\int_{\Omega} \rho \dot{c} \nabla \cdot \nabla c_{-} \, dv = \int_{\Omega} \rho (c_{-}) \cdot \nabla c_{-} \, dv.\]
By means of the identity
\[\nabla \dot{\phi} = (\nabla \phi) \dot{\gamma} + (\nabla \phi) L,\]
for any differentiable function \(\phi\), on \(\Omega \times \mathbb{R}\), we have
\[- \chi_{0} \int_{\Omega} \dot{c} \nabla \cdot (\rho \nabla c_{-}) \, dv = \frac{1}{2} \chi \frac{d}{dt} \int_{\Omega} \rho |\nabla c_{-}|^{2} \, dv + \chi \int_{\Omega} \rho \nabla c_{-} \cdot D \nabla c_{-} \, dv. \tag{7.10}\]
Also, since \(F_{c}(0) = 0\) then
\[\theta \nabla c_{-} = 0\]
so that we can replace \(\dot{c} F_{c}(c_{-})\) with \((c_{-}) F_{c}(c_{-})\) to obtain
\[\int_{\Omega} \rho \dot{c} F_{c}(c_{-}) \, dv = \int_{\Omega} \rho (c_{-}) F_{c}(c_{-}) \, dv = \int_{\Omega} \rho (c_{-}) \, dv = \int_{\Omega} \rho \dot{c} F_{c}(c_{-}) \, dv. \tag{7.10}\]
As a consequence, (7.7) to (7.10) yield
\[\frac{d}{dt} \int_{\Omega} \rho \theta F(c_{-}) + \frac{\chi}{2} |\nabla c_{-}|^{2} \, dv = \frac{d}{dt} \int_{\Omega} \rho \dot{c} F(c_{-}) \, dv = \int_{\Omega} \rho \dot{c} \nabla \mu(\rho, c_{-}) \cdot \nabla \mu(\rho, c_{-}) + \chi \rho \nabla c_{-} \cdot D \nabla c_{-} \, dv. \tag{7.11}\]
In view of (7.3) we obtain
\[\frac{d}{dt} \int_{\Omega} \rho \theta F(c_{-}) + \frac{\chi}{2} |\nabla c_{-}|^{2} \, dv \leq 0.\]
Integration on \(t \in [0, T]\) and account of the initial conditions (7.5) yield
\[\int_{\Omega} \rho \theta F(c_{-}) + \frac{\chi}{2} |\nabla c_{-}|^{2} \, dv \bigg|_{t=T} \leq 0, \quad T \in \mathbb{R}^{+}. \tag{7.12}\]
This implies that
\[F(c_{-}(x, T)) = 0, \quad \nabla c_{-}(x, T) = 0, \quad x \in \Omega, \quad T \in \mathbb{R}^{+}.\]
Since \(c_{-} \leq 0\) and \(F\) is non-negative and vanishes only at 0, 1 then it follows that
\[c_{-}(x, T) = 0, \quad c(x, T) \geq 0, \quad x \in \Omega, \quad T \in \mathbb{R}^{+}.\]
This means that \(c\) cannot be negative.

We now show that
\[c(x, T) \leq 1, \quad x \in \Omega, \quad T \in \mathbb{R}^{+}.\]
To this end we let
\[ c^+ = \begin{cases} 
1, & c \leq 1, \\
c, & c > 1.
\end{cases} \]
Hence
\[ c^+(x,0) = 1, \quad \nabla c^+(x,0) = 0, \quad F(c^+(x,0)) = 0, \quad x \in \Omega. \]
Moreover,
\[ G_c(c^+) = 0, \quad \mu(c^+) = \Theta F_c(c^+) - \chi_0 \Delta c^+, \]
as \( c \in \mathbb{R} \). By repeating step by step the procedure adopted for \( c_- \) we eventually find that
\[ c^+(x,T) = 1, \quad c(x,T) \leq 1, \quad x \in \Omega, \quad T \in \mathbb{R}^+. \]
In conclusion,
\[ c(x,T) \in [0,1] \quad x \in \Omega, \quad T \in \mathbb{R}^+ \]
and the theorem is proved.

It is worth mentioning that a maximum principle theorem (MPT) for the Cahn-Hilliard equation is proved in [21] for a simpler model. Indeed, in [21] the mobility \( \kappa \) is degenerate in that it is taken to vanish at \( c = 0,1 \). By an appropriate function \( G \) we have given a proof which is free from any assumption on \( \kappa \). Incidentally, if \( \kappa \) is degenerate then (7.3) does not hold. Moreover, here we allow also for the dependence on the mass density \( \rho \), the temperature \( \theta \) and the stretching tensor \( D \). Analogous generalizations hold relative to the MPT in [22, 23] for both phase and temperature in first-order phase transitions. Also, quite a similar proof is given in [14], for a quasi-incompressible mixture, where \( \rho \) is a known function of \( c \).

8. Conclusions. This paper provides a thermodynamic approach for the modelling of Cahn-Hilliard fluids. The phase-field scheme is applied by regarding the transition region as a possibly-narrow, but not sharp, layer where diffusion and motion of the mixture occur. The transition is induced by temperature.

To keep the model as simple as possible we account for the evolution of concentration but look at the balance of energy and entropy as for a single constituent. However, due to the internal structure of the mixture we allow for an extra-energy flux \( w \) in addition to the heat flux \( q \). Also we account for motion and diffusion effects by letting the stress in the mixture have additive viscous terms. As a result we obtain a whole set of evolution equations for the concentration, the velocity, and the temperature through the balance of mass, linear momentum, and energy. The thermodynamic analysis shows that \( w \) is eventually given by relations which are consistent with previous results on the subject.

The main features to emerge from this paper are summarized as follows. The constituents are allowed to be compressible fluids. Above the transition temperature, \( \Theta \), the uniform concentration of the mixture prevails at a value \( c^* \) that is determined by the overall masses of the constituents. Below \( \Theta \), separation occurs and the concentrations \( c_- \) (near 0) and \( c^+ \) are favoured.

Upon linearization, we examine the various effects on \( \dot{c} \) by the contention value \( c \) and the shear tensor \( D \). If \( \psi_{cc} > 0 \) then \( c \) shows a diffusion effect. For the same value of \( c \), the effect changes according as \( \theta < \Theta \) or \( \theta > \Theta \). Also, a maximum of \( \mathcal{D} \) favours the approach of \( c \) to the equilibrium uniform value \( c^* \) whereas a minimum of \( \mathcal{D} \cdot D \) favours the phase-separation or demixing. In addition, around the minimum of \( G \), \( G_{cc} > 0 \), the shear favours diffusion through \( G_{cc} |\nabla D|^2 \). Finally, a maximum
principle theorem is proved which shows that necessarily the concentration \( c \) belongs to \([0, 1]\), which is expected on physical grounds.

**Appendix A: Thermodynamic restrictions.** Restrictions placed upon the constitutive functions by the second law of thermodynamics, as expressed by (2.10), are derived. We assume that \( j, T, \eta, \) and \( w \) are continuous functions of \( \Gamma = (\theta, \rho, c, \nabla \theta, \nabla c, D, ...) \) the dots denoting possible higher-order gradients of \( \theta, \rho, D \).

The free energy \( \psi \) is assumed to be a \( C^2 \) function of \( \Gamma_0 = (\theta, \rho, c, \nabla c) \).

Time differentiation of \( \psi(\theta, \rho, c, \nabla c) \) and substitution in (2.10) gives

\[
-\rho(\psi_\theta + \eta) \dot{\theta} + \psi_\rho \dot{\rho} + \psi_c \dot{c} + \psi_{\nabla c} \cdot \nabla \dot{c} + T \cdot L - \frac{1}{\theta} \mathbf{q} \cdot \nabla \theta - \nabla \cdot \mathbf{w} \geq 0.
\]

The inequality holds for any value of \( \dot{\theta} \) only if

\[
\eta = -\psi_\theta.
\]

Hence, by means of the identity

\[
\dot{\nabla} c = \nabla \dot{c} - L^T \nabla c
\]

and the balance equation (2.3) we can write

\[
[T + \rho^2 \psi_\rho \mathbf{1} + \rho \nabla c \otimes \psi_{\nabla c}] \cdot L - \rho \psi_c \dot{c} - \rho \psi_{\nabla c} \cdot \nabla \dot{c} - \frac{1}{\theta} \mathbf{q} \cdot \nabla \theta - \nabla \cdot \mathbf{w} \geq 0. \tag{8.1}
\]

We now determine a pair of constitutive functions \( T(\Gamma), w(\Gamma) \) for which inequality (8.1) holds.

Letting

\[
\delta_c \psi := \psi_c - \frac{1}{\rho} \nabla \cdot (\rho \psi_{\nabla c}),
\]

we can write (8.1) in the form

\[
[T + \rho^2 \psi_\rho \mathbf{1} + \rho \nabla c \otimes \psi_{\nabla c}] \cdot L - \rho \delta_c \psi \dot{c} - \nabla \cdot (\mathbf{w} - \psi_{\nabla c} \dot{c}) - \frac{1}{\theta} \mathbf{q} \cdot \nabla \theta \geq 0. \tag{8.2}
\]

To obtain restrictions placed by (8.2) we need to specify the dependence of \( T \) on \( \Gamma \).

If

\[
T = T_0 + 2 \nu \mathbf{D} + \sigma(\nabla \cdot \mathbf{v}) \mathbf{1} + \hat{G}[2 \beta \mathbf{\bar{D}} + \alpha(\nabla \cdot \mathbf{v}) \mathbf{1}],
\]

where \( T_0 \) is independent of \( \mathbf{D} \) and \( G \) is a function of \( c \), we can view \( 2 \nu \mathbf{\bar{D}} + \sigma(\nabla \cdot \mathbf{v}) \mathbf{1} \) as the viscous stress while \( \hat{G}[2 \beta \mathbf{\bar{D}} + \alpha(\nabla \cdot \mathbf{v}) \mathbf{1}] \) is a stress proportional to \( \dot{c} \). The shear-induced mixing proves effective by letting \( \beta < 0, \alpha < 0 \). Moreover, let

\[
T_0 = -p \mathbf{1} + \mathbf{\hat{D}}
\]

so that

\[
T \cdot L = -p \nabla \cdot \mathbf{v} + \mathbf{\hat{D}} \cdot \mathbf{\hat{D}} + 2 \nu \mathbf{\bar{D}} \cdot \mathbf{\bar{D}} + \sigma(\nabla \cdot \mathbf{v})^2 + G_c \mathbf{D} \dot{c}
\]

where

\[
\mathbf{D} := 2 \beta \mathbf{\bar{D}} + \mathbf{\bar{D}} + \alpha(\nabla \cdot \mathbf{v})^2.
\]

This indicates that we regard \( G_c \mathbf{D} \dot{c} \) as a contribution to the chemical potential. Let

\[
\mu := \delta_c \psi - \frac{1}{\rho} G_c \mathbf{D} \dot{c}. \tag{8.3}
\]
In view of (2.3) and (2.4), substitution in (8.2) and some rearrangements yield
\begin{equation}
(T_0 + \rho \nabla c \otimes \psi \nabla c) \cdot \dot{\mathbf{L}} + (p + \rho^2 \psi_p + \frac{1}{3} \rho \nabla c \cdot \psi \nabla c) \nabla \cdot \mathbf{v} + \sigma (\nabla \cdot \mathbf{v})^2 \\
+ 2\nu \mathbf{D} : \dot{\mathbf{D}} - \mu \nabla \cdot \mathbf{j} - \nabla \cdot (\mathbf{w} + \psi \nabla c \dot{c}) - \frac{1}{\theta} \mathbf{q} : \nabla \theta \geq 0.
\end{equation}
\begin{equation}
(8.4)
\end{equation}
Since
\[-\mu \nabla \cdot \mathbf{j} - \nabla \cdot (\mathbf{w} + \psi \nabla c \dot{c}) = -\mathbf{j} : \nabla \mu - \nabla \cdot (\mathbf{w} + \psi \nabla c \dot{c} + \mu \mathbf{j}),
\]
it follows that inequality (8.4) holds if
\begin{equation}
T_0 = -\rho^2 \psi_p \mathbf{1} - \rho \nabla c \otimes \psi \nabla c, \quad \nu, \sigma \geq 0,
\end{equation}
\begin{equation}
w = \psi \nabla c \cdot \mathbf{j} + \mu \mathbf{j} \quad \frac{1}{\theta} \mathbf{q} : \nabla \theta + \mathbf{j} : \nabla \mu \leq 0.
\end{equation}
\begin{equation}
(8.5) \quad (8.6)
\end{equation}
In addition, since \( T \) (and hence \( T_0 \)) is symmetric then \( \psi \) is required to satisfy
\[\nabla c \otimes \psi \nabla c = \psi \nabla c \otimes \nabla c.\]
Hence it follows that \( \psi \nabla c \) is parallel to \( \nabla c \),
\begin{equation}
\psi \nabla c = \chi(\theta, \rho, c, \nabla c) \nabla c,
\end{equation}
\begin{equation}
(8.7)
\end{equation}
which occurs if \( \psi \) depends on \( \nabla c \) through \( |\nabla c| \). Unless \( \chi \) is constant, this means that the dependence on \( \nabla c \) need not be in the customary additive form
\[\psi = \psi_0(\theta, \rho, c) + \frac{1}{2} \chi |\nabla c|^2.\]
Though the inequality in (8.6) involves both \( \mathbf{q} \) and \( \mathbf{j} \), for the sake of simplicity we require both terms to be non-positive, which in turn implies that
\[\mathbf{j} = -\kappa \nabla \mu, \quad \mathbf{q} = -k \nabla \theta \]
\begin{equation}
(8.8)
\end{equation}
\( \kappa \) and \( k \) being positive-valued functions of \( \Gamma \).

Some remarks are in order about the constitutive equation for the stress tensor \( T \) and the potential \( \mu \). It is natural to regard \( \rho^2 \psi_p \) as the (dynamic) pressure \( P \) which is then a function of \( \theta, \rho, c, \nabla c \). Also, by (2.12),
\[p = -\frac{1}{3} \text{tr} T_0 = P + \frac{1}{3} \rho \nabla c \cdot \psi \nabla c.\]
This means that the spherical part \( p \) of \( T \) consists of a pressure term \( P \) and of an additional contribution \( \rho \nabla c \cdot \psi \nabla c/3 \) due to the inhomogeneity of the concentration.

The potential \( \mu \), in (2.14), consists of two different contributions. First, \( \delta_c \psi \) originates from the free-energy potential \( \psi \) and can be viewed as classical chemical potential (see, e.g., [17]) which is defined as the variational derivative of \( \psi \) relative to the order parameter. Second, the term \( -G_c \mathcal{D}/\rho \) is due to the stretching \( \mathbf{D} \). Indeed, letting
\[\hat{\psi} = \psi - \frac{1}{\rho} G \mathcal{D},\]
we can write
\[\mu = \delta_c \hat{\psi}.\]
We can then say that the diffusion flux \( \mathbf{j} \) involves the effective potential \( \mu \) and hence the effective free energy \( \hat{\psi} \), instead of the thermodynamic potential \( \delta_c \psi \) only.
Appendix B: Relation to other approaches. Based on a variational analysis, Lowengrub and Truskinovsky [11] assume that the rate of work \( A \) done on the boundary \( \partial P \) of a material domain \( P \) is given by

\[
A = \int_{\partial P} [(\mathbf{T}n) \cdot \mathbf{v} + \mathbf{t} \cdot n \mathbf{c}] \, da
\]

where

\[
t = \rho \psi c.
\]

As a consequence the balance of energy becomes

\[
\rho \dot{e} = \mathbf{T} \cdot \mathbf{L} + \nabla \cdot (\mathbf{t} \dot{c}) + \rho r.
\]

The formal absence of \( q \) in [11] is due to the identification of the energy flux with \( \mathbf{t} \dot{c} \).

Gurtin [17] (see also [19]) restricts attention to undeformable bodies. Upon identifying the order parameter \( \rho \) of [17] with \( c \), we can write the rate of working (of microforces) as

\[
A = \int_{\partial P} \mathbf{\xi} \cdot n \dot{c} \, da + \int_{P} \gamma \dot{c} \, dv,
\]

Next \( \mathbf{\xi} \) too is shown to be related to the free energy, in our notation \( \mathbf{\xi} = \rho \psi c \).

By (8.6), the present balance of energy (2.8) and the entropy inequality (2.9) become

\[
\rho \dot{\theta} \geq \frac{\rho r}{\theta} - \nabla \cdot \frac{\mathbf{q}}{\theta}.
\]

Accordingly, the power term \( \nabla \cdot (\rho \psi c \dot{c}) \) coincides with that of [11], \( \nabla \cdot (\mathbf{t} \dot{c}) \), and that of [17], \( \nabla \cdot (\mathbf{\xi} \dot{c}) \).

In [14] both constituents are regarded as incompressible, and hence the mixture is said to be quasi-incompressible. Mathematically, the incompressibility of the constituents results into a dependence of \( \rho \) on \( c \), \( \rho = \rho(c) \), and hence in the further constraint

\[
\rho c \dot{c} = -\rho \nabla \cdot \mathbf{v}.
\]

This in turn implies that

\[
\nabla \cdot \mathbf{v} = \frac{1}{\rho^2} \rho c \nabla \cdot \mathbf{j}
\]

and hence \( \nabla \cdot \mathbf{v} \) is not independent of \( \nabla \cdot \mathbf{j} \). This implies that the relation \( P = \rho^2 \psi_\rho + (\rho/3)c \nabla \cdot \mathbf{c} \) cannot be derived and, furthermore, the additive term in \( \rho \mu \) is \( -2\beta G_c \mathbf{D} \cdot \mathbf{D} \) thus ruling out a dependence of \( \mu \) on \( \nabla \cdot \mathbf{v} \). According to [11],

\[
\mathbf{T}_0 = -P \mathbf{I} - \rho \nabla c \otimes \psi c,
\]

where \( P \) is an arbitrary function of position and time, instead of (2.12).

We might have developed a dual scheme with both an extra-energy flux \( \mathbf{w} \) and an extra-entropy flux \( \mathbf{k} \) [13] to obtain

\[
\mathbf{w} = -\rho \psi c \dot{c}, \quad \mathbf{k} = -\frac{\mu}{\theta} \mathbf{j}, \quad \mathbf{j} \cdot \nabla \frac{\mu}{\theta} \leq 0,
\]

which is consistent with (3.29) of [11].
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