Phase-Field Descriptions of
Two-Phase Compressible Fluid Flow:
Interstitial Working and a Reduction
to Korteweg Theory

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1. INTRODUCTION

First formulated by Truskinovsky [14] and Lowengrub and Truskinovsky [10], the 'Navier-Stokes-Allen-Cahn' equations[1] and the 'Navier-Stokes-Cahn-Hilliard' equations describe diphasic fluid flow. They combine the conservation laws for mass, momentum, and energy with a balance law for the phases which governs the concentration of one (or the other) phase as an order parameter. By contrast, the 'Navier-Stokes-Korteweg' theory for the dynamics of a one-phase capillary fluid that Dunn and Serrin [3] gave following Korteweg [9] and Slemrod [12] uses only the three said conservation laws and has the mass density itself as its order parameter.[2]

In [6], the authors considered the derivation of the NSAC, the NSCH, and the NSK equations, and showed that in the case that the two phases are incompressible with different specific volumes, both NSAC and NSCH reduce to versions of NSK. However, as we realized only after that article was published, our assumptions in [6] tacitly correspond to neglecting the so-called microforces [8]. While the argumentation in [6] seems mathematically consistent, the microforces are a physical reality, and the purpose of the present note is to show that the reduction of NSAC and NSCH to NSK still holds when one does take them into account.

Also in NSAC and NSCH, the microforces do what Dunn and Serrin named *interstitial work*. In Section 2 we study the role that this particular effect plays in the entropy production along particle paths. Section 3 serves to prove the reduction property of NSAC and NSCH *with* interstitial work. Section 4 discusses the *objectivity* of the three models briefly. In the sequel, we use notation from [6] and the results of Section 1 of that paper without defining / stating them again.

Since the publication of [6] we also noticed that already Korteweg himself realized his tensor should be useful in connection with incompressible phases — the modern NSAC and NSCH equations would be the "autres d'une complication plus grande encore"[3] Korteweg envisaged as generalizations of what is nowadays called 'dynamic Korteweg theory'.

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[1]Cf. also the interesting paper by Blesgen [2].

[2]We will henceforth abbreviate Navier-Stokes-Allen-Cahn, Navier-Stokes-Cahn-Hilliard, and Navier-Stokes-Korteweg as NSAC,NSCH, and NSK.

[3]Cf. p. 12 of [9] and footnote 2 on p. 514 of [13].

2. Interstitial work and entropy production

2.1. The Navier-Stokes-Korteweg system

In this subsection we briefly recall entropy production in the Navier-Stokes-Korteweg system. This system, now exactly in its form derived by Dunn and Serrin in [3], reads

$$\begin{aligned}
\partial_t \rho + \nabla \cdot (\rho u) &= 0, \\
\partial_t (\rho u) + \nabla \cdot (\rho u \otimes u - \bar{\mathbf{T}}) &= 0, \\
\partial_t \bar{\mathcal{E}} + \nabla \cdot ((\bar{\mathcal{E}}\mathbf{I} - \bar{\mathbf{T}})u + \bar{w}) - \nabla \cdot (\beta \nabla \theta) &= 0,
\end{aligned} \tag{2.1}$$

with

$$\bar{\mathcal{E}} \equiv \rho(\bar{E} + \frac{1}{2}|u|^2) \quad \text{and} \quad \bar{\mathbf{T}} \equiv -p\mathbf{I} + \mathbf{K} + \mathbf{S}.$$

The fluid is specified by its Helmholtz energy

$$\bar{F}(\rho, \theta, \nabla\rho) = \check{F}(\rho, \theta, |\nabla\rho|^2/2), \tag{2.2}$$

from which its internal energy $\bar{E}$ derives via the Legendre transform

$$\bar{E}(\rho, s, \nabla\rho) \equiv \bar{F}(\rho, \theta, \nabla\rho) + \theta s,$$

with temperature θ and specific entropy $s = -\partial_\theta F$ as dual variables. The Korteweg tensor and the interstitial work flux are

$$\mathbf{K} = \left[\rho\nabla \cdot \left(\partial_{\nabla\rho}(\rho\bar{F})\right)\mathbf{I} - \nabla\rho \otimes \partial_{\nabla\rho}(\rho\bar{F}) \right], \quad \bar{w} = \kappa\rho(\nabla \cdot u)\nabla\rho. \tag{2.3}$$

with κ given by

$$\rho\partial_{\nabla\rho}\bar{F} = \kappa\nabla\rho. \tag{2.4}$$

Viscous stress

$$\mathbf{S} = 2\eta(Du)^{s0} + \zeta\nabla \cdot u\,\mathbf{I}, \quad (Du)^{s0} \equiv \frac{1}{2}(Du + (Du)^\top) - \frac{1}{3}\nabla \cdot u\mathbf{I}$$

and heat flux, $-\beta\nabla\theta$, are quantified by means of the coefficients η, ζ, β of shear viscosity, bulk viscosity, and heat conductivity. The reasoning in [3][4] yields the entropy balance

$$\partial_t(\rho s) + \nabla \cdot (\rho s u) = \nabla \cdot \Sigma + \sigma \tag{2.5}$$

with

$$\Sigma \equiv \frac{\beta}{\theta}\nabla\theta \tag{2.6}$$

and

$$\sigma \equiv \frac{1}{\theta}\left(2\eta(Du)^{s0} : (Du)^{s0} + \zeta(\nabla \cdot u)^2\right) + \frac{\beta}{\theta^2}|\nabla\theta|^2. \tag{2.7}$$

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[4]which can technically be expressed as a simple computation very similar to one in Sec. 2.3 of [6]

2.2. THE NAVIER-STOKES-PHASE-FIELD MODELS

Before specializing to Navier-Stokes-Allen-Cahn and Navier-Stokes-Cahn-Hilliard, we show some properties which both systems share. We start from their common form

$$
\begin{aligned}
\partial_t \rho + \nabla \cdot (\rho u) &= 0, \\
\partial_t (\rho u) + \nabla \cdot (\rho u \otimes u - \mathbf{T}) &= 0, \\
\partial_t \mathcal{E} + \nabla \cdot ((\mathcal{E}\mathbf{I} - \mathbf{T})u + w) - \nabla \cdot (\beta \nabla \theta) &= 0, \\
\partial_t (\rho \chi) + \nabla \cdot (\rho \chi u) - \rho j &= 0
\end{aligned}
\tag{2.8}
$$

in which

$$
\mathcal{E} \equiv \rho(E + \tfrac{1}{2}|u|^2), \quad \mathbf{T} \equiv -p\mathbf{I} + \mathbf{C} + \mathbf{S}, \quad w
$$

are the total energy, the total Cauchy stress, and the interstitial work flux. We specify the fluid by its Gibbs energy G, i.e., the internal energy is

$$
E(\tau, s, \chi, \nabla\chi) \equiv G(p, \theta, \chi, \nabla\chi) - p\tau + \theta S,
$$

with temperature θ and specific entropy $s = -\partial_\theta G$ as well as pressure p and specific volume $\tau = 1/\rho = \partial_p G$ as pairs of conjugate variables, and the Ericksen tensor is

$$
\mathbf{C} = -\rho \nabla\chi \otimes \frac{\partial G}{\partial \nabla\chi}.
$$

We will also use the generalized chemical potential

$$
\mu \equiv \partial_\chi G - \frac{1}{\rho} \nabla \cdot \left(\rho \partial_{\nabla\chi} G \right),
$$

Lemma 2.1. *For solutions to* (2.8), *the entropy production along particle paths satisfies*

$$
\rho\theta\dot{s} = \mathbf{S} : Du + \nabla \cdot (\beta\nabla\theta) - \nabla \cdot w - \rho(G_{\nabla\chi} \cdot \nabla j + G_\chi j).
\tag{2.9}
$$

Proof. This is shown in exactly the same way as equation (2.7) in [6]. ☐

Lemma 2.2. *With*

$$
w = -\rho j \partial_{\nabla\chi} G,
\tag{2.10}
$$

equation (2.9) *reads*

$$
\rho\theta\dot{s} = \mathbf{S} : Du + \nabla \cdot (\beta\nabla\theta) - \rho j \mu.
\tag{2.11}
$$

Proof. One computes

$$
-\nabla \cdot w - \rho(G_{\nabla\chi} \cdot \nabla j + G_\chi j) = j(-\rho G_\chi + \nabla \cdot (\rho G_{\nabla\chi})).
$$

☐

The next result distinguishes between NSAC and NSCH.

Lemma 2.3. *Assume* (2.10).
(i) With

$$j = j_{AC} = -\frac{\mu}{\epsilon}, \tag{2.12}$$

the entropy production on solutions of (2.8) *is given by* (2.5) *with*

$$\Sigma = \frac{\beta}{\theta}\nabla\theta \tag{2.13}$$

and

$$\sigma = \frac{1}{\theta}\mathbf{S} : Du + \frac{\beta}{\theta^2}|\nabla\theta|^2 + \frac{\epsilon}{\theta}\rho j^2. \tag{2.14}$$

(ii) With

$$j = j_{CH} = \frac{1}{\rho}\nabla\cdot\mathcal{J}, \quad \mathcal{J} = \gamma\nabla\left(\frac{\mu}{\theta}\right), \tag{2.15}$$

the entropy production on solutions of (2.8) *is given by* (2.5) *with*

$$\Sigma = \frac{\beta}{\theta}\nabla\theta - \gamma\frac{\mu}{\theta}\nabla\left(\frac{\mu}{\theta}\right) \tag{2.16}$$

and

$$\sigma = \frac{1}{\theta}\mathbf{S} : Du + \frac{\beta}{\theta^2}|\nabla\theta|^2 + \gamma\left|\nabla\left(\frac{\mu}{\theta}\right)\right|^2. \tag{2.17}$$

Proof. Immediate.   □

Equations (2.8) with interstitial work flux w as in (2.10) and $j = j_{AC}, j = j_{CH}$ as in (2.12),(2.15) are the NSAC and NSCH equations proposed in [14, 10, 2].[5] We view the considerations of this subsection certainly not as a new derivation of NSAC and NSCH[6], but as perhaps elucidating the role that interstitial work plays in connection with the second law of thermodynamics also in these two phase-field models.

3. Reduction to Korteweg models

Assume now that a fluid consists of two incompressible phases of different temperature-independent specific volumes. I.e. (cf. [6]), its Gibbs energy has the form

$$G(p, \theta, \chi, \nabla\chi) = T(\chi)p + W(\theta, \chi, \nabla\chi) \quad \text{with} \quad W(\theta, \chi, \nabla\chi) = \check{W}(\theta, \chi, |\nabla\chi|^2), \tag{3.1}$$

where

$$T(\chi) = \chi\tau_1 + (1 - \chi)\tau_2 \tag{3.2}$$

with constants $\tau_1, \tau_2 > 0$ satisfying

$$\tau_* \equiv \tau_1 - \tau_2 \neq 0. \tag{3.3}$$

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[5]Though not to *every* detail in the NSCH case, as Lowengrub and Truskinovsky seem to either use a law from isothermal diffusion or leave constitutive laws in particular for the heat and diffusion fluxes widely open; see equations (3.24b), (3.30) in [10]. The latter is very meaningful, cf. [11].

[6]Rather must we point out that Remarks 2.1, 2.2, 2.3 in [6] are no longer in order as soon as one does take the microforces into account.

3.1.   REDUCTION OF THE NAVIER-STOKES-ALLEN-CAHN SYSTEM

Theorem 3.1. *In the case of two molecularly immiscible incompressible phases of different, temperature-independent specific volumes, (3.1), (3.2), (3.3), the Navier-Stokes-Allen-Cahn equations (2.8) with $j = j_{AC}$ from (2.12) and w from (2.10) can be written as the Navier-Stokes-Korteweg system*

$$\partial_t \rho + \nabla \cdot (\rho u) = 0,$$
$$\partial_t (\rho u) + \nabla \cdot (\rho u \otimes u) - \nabla \cdot (-\bar{p}\mathbf{I} + \mathbf{K} + \mathbf{S}^\epsilon) = 0, \tag{3.4}$$
$$\partial_t \bar{\mathcal{E}} + \nabla \cdot (\bar{\mathcal{E}} u - (-\bar{p}\mathbf{I} + \mathbf{K} + \mathbf{S}^\epsilon)u + \bar{w}) - \nabla \cdot (\beta \nabla \theta) = 0,$$

with $\bar{\mathcal{E}}, \bar{p}, \mathbf{K}, \bar{w}$ derived as in 2.3 from the Helmholtz energy

$$\bar{F}(\theta, \rho, \nabla \rho) = W(\theta, \bar{\chi}(\rho), [\bar{\chi}'(\rho)]^2 |\nabla \rho|^2) \quad with \quad \bar{\chi}(\rho) := \frac{1/\rho - \tau_2}{\tau_*}$$

and with the modified viscous stress

$$\mathbf{S}^\epsilon = 2\eta(Du)^{s0} + (\zeta + \zeta^\epsilon)\nabla \cdot u \, \mathbf{I} \quad where \quad \zeta^\epsilon \equiv \frac{\epsilon}{\rho \tau_*^2}. \tag{3.5}$$

Proof. In this situation, the concentration χ and the total density ρ are linked through

$$\chi = \bar{\chi}(\rho), \tag{3.6}$$

and the behaviour of the concentration is described by

$$\rho j = \partial_t(\rho \chi) + \nabla \cdot (\rho \chi u) = \rho \dot{\chi} = \rho \bar{\chi}'(\rho)\dot{\rho} = \frac{1}{\tau_*}\nabla \cdot u. \tag{3.7}$$

Very similarly to [6], we have

$$\partial_{\nabla \chi} G = \partial_{\nabla \chi} W = \frac{1}{\bar{\chi}'(\rho)}\partial_{\nabla \rho}\bar{F}, \quad \nabla \chi = \bar{\chi}'(\rho)\nabla \rho. \tag{3.8}$$

Using (2.10) and (3.7), this implies

$$w = -\rho \dot{\chi} \, \partial_{\nabla \chi} G = \rho^2(\nabla \cdot u) \, \bar{\chi}'(\rho) \, \partial_{\nabla \chi} G = \rho^2(\nabla \cdot u) \, \partial_{\nabla \rho}\bar{F} = \kappa \rho (\nabla \cdot u) \, \nabla \rho = \bar{w}. \tag{3.9}$$

Furthermore,

$$\partial_\chi G = \tau_* p + \partial_\chi W \tag{3.10}$$

and

$$\frac{1}{\tau_*}\partial_\chi W = -\rho^2 \partial_\rho \bar{F} + \rho^2 \frac{\bar{\chi}''(\rho)}{\bar{\chi}'(\rho)}\nabla \rho \cdot \partial_{\nabla \rho}\bar{F}, \tag{3.11}$$

imply that

$$\rho \mu = -\left(\nabla \cdot \left(\rho \partial_{\nabla \chi} G\right) - \rho \partial_\chi G\right) = -\left(\nabla \cdot \left([\bar{\chi}'(\rho)]^{-1}\partial_{\nabla \rho}\left(\rho \bar{F}\right)\right) - \rho \tau^* p - \rho \partial_\chi W\right). \tag{3.12}$$

This allows to represent the pressure as

$$p = \frac{\mu}{\tau_*} \nabla\cdot u - \rho\bar\chi'(\rho)\nabla\cdot\left([\bar\chi'(\rho)]^{-1}\partial_{\nabla\rho}(\rho\bar F)\right) - \frac{1}{\tau_*}\partial_\chi W. \qquad (3.13)$$

Combining (3.13) and (3.7) with the Allen-Cahn relation (2.12) gives

$$p = -\frac{\epsilon}{\tau_*^2\rho} \nabla\cdot u - \rho\bar\chi'(\rho)\nabla\cdot\left([\bar\chi'(\rho)]^{-1}\partial_{\nabla\rho}(\rho\bar F)\right) - \frac{1}{\tau_*}\partial_\chi W. \qquad (3.14)$$

Using this, we get

$$\begin{aligned}
-p\mathbf{I} + \mathbf{C} + \mathbf{S} &= -p\,\mathbf{I} - \nabla\chi\otimes\partial_{\nabla\chi}(\rho W) + \mathbf{S} \\
&= \frac{1}{\tau_*}\partial_\chi W\,\mathbf{I} + \rho\bar\chi'(\rho)\nabla\cdot\left([\bar\chi'(\rho)]^{-1}\partial_{\nabla\rho}(\rho\bar F)\right)\mathbf{I} - \nabla\rho\otimes\partial_{\nabla\rho}(\rho\bar F) + \mathbf{S}^\epsilon \\
&= \frac{1}{\tau_*}\partial_\chi W\,\mathbf{I} + \rho\nabla\cdot\left(\partial_{\nabla\rho}(\rho\bar F)\right)\mathbf{I} - \rho^2\frac{\bar\chi''(\rho)}{\bar\chi'(\rho)}\nabla\rho\cdot\partial_{\nabla\rho}\bar F\mathbf{I} - \nabla\rho\otimes\partial_{\nabla\rho}(\rho\bar F) + \mathbf{S}^\epsilon,
\end{aligned}$$

where we have used (3.6) and (3.5). Replacing now the term $\partial_\chi W$ according to the identity (3.11), we indeed find

$$\begin{aligned}
-p\mathbf{I} + \mathbf{C} + \mathbf{S} &= -\bar p\mathbf{I} + \left[\rho\nabla\cdot\left(\partial_{\nabla\rho}\left(\rho\bar F\right)\right)\mathbf{I} - \nabla\rho\otimes\partial_{\nabla\rho}\left(\rho\bar F\right)\right] + \mathbf{S}^\epsilon \\
&= -\bar p\mathbf{I} + \mathbf{K} + \mathbf{S}^\epsilon
\end{aligned} \qquad (3.15)$$

with the Korteweg tensor $\mathbf{K}$ as defined in (2.3).   □

As a consistency check, we note that in the situation of Theorem 3.1, the entropy production rate (2.14) agrees with what (2.7) becomes upon replacing ζ with $\zeta + \zeta^\epsilon$:

$$\frac{\epsilon\rho}{\theta}j_{AC}^2 = \frac{\epsilon}{\rho\theta\tau_*^2}|\nabla\cdot u|^2 = \frac{\zeta^\epsilon}{\theta}|\nabla\cdot u|^2.$$

3.2.   Reduction of the Navier-Stokes-Cahn-Hilliard system

Theorem 3.2. *In the case of two molecularly immiscible incompressible phases of different, temperature-independent specific volumes, (3.1), (3.2), (3.3), the Navier-Stokes-Allen-Cahn equations (2.8) with $j = j_{CH}$ from (2.15) and w from (2.10) can be written as the Navier-Stokes-Korteweg system*

$$\begin{aligned}
\partial_t\rho + \nabla\cdot(\rho u) &= 0, \\
\partial_t(\rho u) + \nabla\cdot(\rho u\otimes u) - \nabla\cdot(-\bar p\mathbf{I} + \mathbf{K} + \mathbf{S}_\gamma) &= 0, \\
\partial_t\bar{\mathcal{E}} + \nabla\cdot(\bar{\mathcal{E}}u - (-\bar p\mathbf{I} + \mathbf{K} + \mathbf{S}_\gamma)u + \mathbf{w}) - \nabla\cdot(\beta\nabla\theta) &= 0,
\end{aligned} \qquad (3.16)$$

with $\bar{\mathcal{E}}, \bar p, \mathbf{K}, \bar w$ derived as in 2.3 from the Helmholtz energy

$$\bar F(\theta, \rho, \nabla\rho) = W(\theta, \bar\chi(\rho), [\bar\chi'(\rho)]^2|\nabla\rho|^2) \quad with \quad \bar\chi(\rho) := \frac{1/\rho - \tau_2}{\tau_*}$$

and with the modified viscous stress

$$\mathbf{S}_\gamma = 2\eta(Du)^{s0} + \zeta\nabla\cdot u\,\mathbf{I} + \frac{\theta}{\tau_*^2}\Lambda_\gamma(\nabla\cdot u)\mathbf{I}, \qquad (3.17)$$

where Λ_γ denotes the solution operator of the elliptic problem

$$-\nabla\cdot(\gamma\nabla\phi) = \nabla\cdot u \quad on \ \Omega.$$

Proof. The only difference from the proof of Theorem 3.1 lies in the form of the representation for the pressure p. We keep using identities (3.6) through (3.13). As the Cahn-Hilliard relation (2.15) means that

$$\mu = -\frac{\theta}{\tau_*}\Lambda_\gamma(\nabla\cdot u), \tag{3.18}$$

identity (3.13) now yields

$$p = -\frac{\theta}{\tau_*^2}\Lambda_\gamma(\nabla\cdot u) - \rho\bar\chi'(\rho)\nabla\cdot\left([\bar\chi'(\rho)]^{-1}\partial_{\nabla\rho}(\rho\bar F)\right) - \frac{1}{\tau_*}\partial_\chi W \tag{3.19}$$

instead of (3.13). This leads to

$$-p\mathbf{I} + \mathbf{C} + \mathbf{S} = -\bar p\mathbf{I} + \mathbf{K} + \mathbf{S}_\gamma \tag{3.20}$$

instead of (3.15). $\square$

For a confirmation, set

$$\phi = \Lambda_\gamma(\nabla\cdot u),$$

note that for (3.16), the entropy balance (2.5) holds with

$$\Sigma \equiv \frac{\beta}{\theta}\nabla\theta - \frac{\gamma}{\tau_*^2}\phi\nabla\phi$$

and

$$\sigma \equiv \frac{1}{\theta}\left(\eta(Du)^{s0} : (Du)^{s0} + \zeta(\nabla\cdot u)^2\right) + \frac{\beta}{\theta^2}|\nabla\theta|^2. + \frac{\gamma}{\tau_*^2}|\nabla\phi|^2$$

instead of (2.6) and (2.7), and observe that in the present situation (3.18) implies that

$$\phi = -\tau_*\frac{\mu}{\theta}$$

in accordance with (2.16) and (2.17).

4. Spatiotemporal objectivity

In [6], arguing against the interstitial work that was so ingeniously discovered by Dunn and Serrin in [3], we claimed that using the corresponding term in the energy equation "would violate the fundamental requirement that the contributions of a stress tensor $\mathbf{T}$ to momentum and energy balance are related as $\nabla\cdot\mathbf{T}$ and $\nabla\cdot(\mathbf{T}u)$" (p. 10). However, instead of being fundamental, this 'requirement' is fundamentally wrong – in particular it is *not* dictated by the principle of objectivity; see below. Regarding NSK, besides by the persuasiveness of Dunn and Serrin's argumentation based on the Clausius-Duhem inequality, we should have felt alarmed by the fact that Benzoni-Gavage et al. in [1] had already identified a Hamiltonian structure for the Euler-Korteweg equations which confirms Dunn and Serrin's formulation. Regarding NSAC and NSCH, the equally solid derivations by Truskinovsky, Lowengrub and Truskinovsky, and Blesgen were other signs of warning.

Still, we noticed the inappropriateness[7] of the above 'requirement' only when we began[8] to formulate relativistic versions of NSK, NSAC, and NSCH. Also 3-tensors that reflect purely mechanical stresses must respect *spatiotemporal objectivity*. In particular, the Korteweg tensor occuring in NSK and the phase transition rate / interphase diffusion flux figuring as j in NSAC and NSCH come from relativistically covariant tensorial quantities that derive from energies which depend, objectively in the sense of Rational Mechanics, on the spatiotemporal 4-gradient of density/concentration, and interstitial work is a natural implication of this property. Related details can be found in [4, 5].

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[7]at the level of physics — it remains amazing to us that the NSAC-NSCH-NSK triple stays as consistent as shown in [6] at the mathematical level, when one suppresses the interstitial work in all three models!

[8]partly paralleling work on dissipation [7]

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