Derivation of effective macroscopic Stokes-Cahn-Hilliard equations for periodic immiscible flows in porous media

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Abstract

Using thermodynamic and variational principles we examine a basic phase field model for a the mixture of two incompressible fluids in strongly perforated domains. With the help of the multiple scale method with drift and our recently introduced splitting strategy for Ginzburg-Landau/Cahn-Hilliard-type equations, we rigorously derive an effective macroscopic phase field formulation under the assumption of periodic flow and a sufficiently large Péclet number. As for the classical convection-diffusion problem, we obtain systematically diffusion-dispersion relations (including Taylor-Aris-dispersion). To ensure the applicability of the equations we derive in practical settings, we also provide a convenient computational framework to macroscopically track interfaces in porous media. In view of the well-known versatility of phase field models, our study proposes a promising model for many engineering and scientific applications such as multiphase flows in porous media, microfluidics, and fuel cells.

1 Introduction

Consider the total energy density

$$e(\mathbf{x}(\mathbf{X},t),t) for an interface between two phases, := \frac{1}{2}|\mathbf{x}_t(\mathbf{X},t)|^2 - \frac{\lambda}{2}|\nabla_{\mathbf{x}}\phi(\mathbf{x}(\mathbf{X},t),t))|^2 - \frac{\lambda}{2}F(\phi(\mathbf{x}(\mathbf{X},t),t))\,, \tag{1.1}$$

where ϕ is a conserved order-parameter that evolves between different liquid phases represented as the minima of a homogeneous free energy F. The parameter λ represents the surface tension effect, i.e. $\lambda \propto$ (surface tension) $\times$ (capillary width(. Assume that F represents polynomials of the form

$$F(\phi) := \int_0^\phi f(s)\,ds\,, \quad \text{and} \quad f(s) := a_3 s^3 + a_2 s^2 + a_1 s\,. \tag{1.2}$$

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The variable $\mathbf{X}$ stands for the Lagrangian (initial) material coordinate and $\mathbf{x}(\mathbf{X},t)$ represents the Eulerian (reference) coordinate. The last two terms in (1.1) form the well-known density of the Cahn-Hilliard/Ginzburg-Landau phase field formulation adapted to the flow map $\mathbf{x}(\mathbf{X},t)$ defined by

$$\begin{cases} \mathbf{x}_t = \mathbf{u}(\mathbf{x}(\mathbf{X},t),t)\,, \\ \mathbf{x}(\mathbf{X},0) = \mathbf{X}\,. \end{cases} \tag{1.3}$$

The first term in (1.1) is the kinetic energy, which accounts for fluid flow of incompressible materials showing the same viscosity constantμ, i.e.,

$$\begin{cases} \mathbf{u}_t + (\mathbf{u}\cdot\nabla)\mathbf{u} - \mu\Delta\mathbf{u} + \nabla p = \mathbf{g}\,, \\ \operatorname{div}\mathbf{u} = 0\,, \end{cases} \tag{1.4}$$

where $\mathbf{g}$ is a driving force acting on the fluid. We are interested in the mixture of two incompressible and immiscible fluids of the same viscosity μ. Hence, we can employ generic free energies (1.1) showing a double-well form as is the case often in applications (e.g. [41]).

Suppose that $D\subset\mathbb{R}^d$ with $d>0$ the dimension of space denotes the domain which is initially occupied by the fluid. Then, we can define for an arbitrary length of time $T>0$ the total energy by

$$E(\mathbf{x}) := \int_0^T \int_D e(\mathbf{x}(\mathbf{X},t),t)\, d\mathbf{X}\, dt\,. \tag{1.5}$$

The energy (1.5) combines an action functional for the flow map $\mathbf{x}(\mathbf{X},t)$ and a free energy for the order parameter ϕ. This combination of mechanical and thermodynamic energies seems to go back to [13, 14, 17, 18, 20]. Subsequently, we will focus on quasi-stationary, i.e., $\mathbf{u}_t=\mathbf{0}$ and $\mathbf{g}\neq\mathbf{0}$, and low-Reynolds number flows, i.e., $(\mathbf{u}\cdot\nabla)\mathbf{u}=\mathbf{0}$. Then, classical ideas from the calculus of variations [38] and the theory of gradient flows together with the imposed wetting boundary condition $\int_{\partial\Omega} w(\mathbf{x})\, do(\mathbf{x})$ for $w(\mathbf{x})\in H^{3/2}(\partial\Omega)$ lead to the following set of equations

$$\text{(Homogeneous case)}\begin{cases} -\mu\Delta\mathbf{u} + \nabla p = \mathbf{g} & \text{in } \Omega_T\,, \\ \quad\operatorname{div}\mathbf{u} = 0 & \text{in } \Omega_T\,, \\ \quad\mathbf{u} = \mathbf{0} & \text{on } \partial\Omega_T\,, \\ \phi_t + \mathrm{Pe}(\mathbf{u}\cdot\nabla)\phi = \lambda\operatorname{div}\left(\nabla\left(f(\phi)-\Delta\phi\right)\right) & \text{in } \Omega_T\,, \\ \quad\nabla_n\phi := \mathbf{n}\cdot\nabla\phi = w(\mathbf{x}) & \text{on } \partial\Omega_T\,, \\ \quad\nabla_n\Delta\phi = 0 & \text{on } \partial\Omega_T\,, \\ \quad\phi(\mathbf{x},0) = h(\mathbf{x}) & \text{on } \Omega\,, \end{cases} \tag{1.6}$$

where $\Omega_T := \Omega\times]0,T[$, $\partial\Omega_T := \partial\Omega^1\times]0,T[$, λ represents the elastic relaxation time of the system, and the driving force $\mathbf{g}$ accounts for the elastic energy [18]

$$\mathbf{g} = -\gamma\operatorname{div}\left(\nabla\phi\otimes\nabla\phi\right)\,, \tag{1.7}$$

where γ corresponds to the surface tension [19] and hence $\gamma=\lambda$ for simplicity in [1]. The dimensionless parameter $\mathrm{Pe} := \frac{kTLU}{D}$ is the Péclet number for a reference fluid velocity $\mathrm{U} := |\mathbf{u}|$, L is the characteristic length of the porous medium, and the diffusion constant $D=kTM$ obtained from the mobility via Einstein's relation. We note that the immiscible flow equations can immediately be written for the full incompressible Navier-Stokes equations as in [18]. Our restriction to the Stokes equation is motivated here by the fact that such flows turn into Darcy's law in porous media [10].

The main objective of our study is the derivation of effective macroscopic equations describing (1.6) in the case of perforated domains $\Omega^\epsilon\subset\mathbb{R}^d$ instead of a homogeneous $\Omega\subset\mathbb{R}^d$. A useful and reasonable

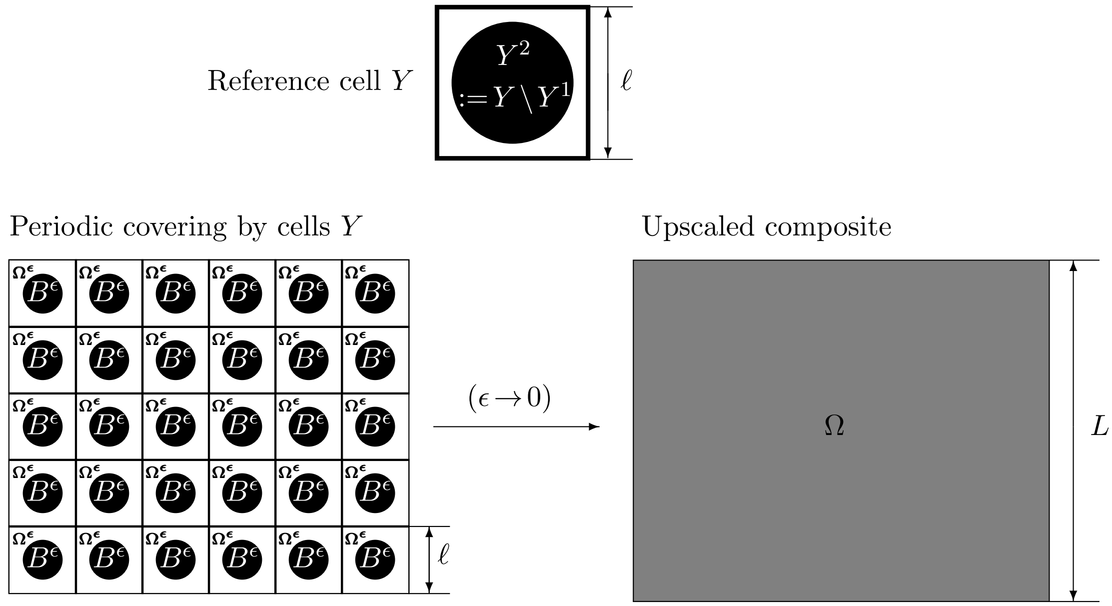

Figure 1: **Left:** Porous medium $\Omega^\epsilon := \Omega \setminus B^\epsilon$ as a periodic covering of reference cells $Y := [0, \ell]^d$. **Top:** Definition of the reference cell $Y = Y^1 \cup Y^2$ with $\ell = 1$. **Right:** The "homogenization limit" $\epsilon := \frac{\ell}{L} \to 0$ scales the perforated domain such that perforations become invisible in the macroscale.

approach is to represent a porous medium $\Omega = \Omega^\epsilon \cup B^\epsilon$ periodically with pore space Ω^ϵ and solid phase B^ϵ. The arising interface $\partial\Omega^\epsilon \cap \partial B^\epsilon$ is denoted by I^ϵ. As usual, the dimensionless variable $\epsilon > 0$ defines the heterogeneity $\epsilon = \frac{\ell}{L}$ where ℓ represents the characteristic pore size and L is the characteristic length of the porous medium, see Figure 1. The porous medium is defined by a periodic coverage of a reference cell $Y := [0, \ell_1] \times [0, \ell_2] \times \cdots \times [0, \ell_d]$ which represents a single, characteristic pore. For simplicity, we set $\ell_1 = \ell_2 = \cdots = \ell_d = 1$. The periodicity assumption allows, by passing to the limit $\epsilon \to 0$ – see Figure 1, for the derivation effective macroscopic porous media equations, rather attractive for applications. The pore and the solid phase of the medium are defined as usual by,

$$\Omega^\epsilon := \bigcup_{\mathbf{z} \in \mathbb{Z}^d} \epsilon\left(Y^1 + \mathbf{z}\right) \cap \Omega, \qquad B^\epsilon := \bigcup_{\mathbf{z} \in \mathbb{Z}^d} \epsilon\left(Y^2 + \mathbf{z}\right) \cap \Omega = \Omega \setminus \Omega^\epsilon, \tag{1.8}$$

where the subsets $Y^1, Y^2 \subset Y$ are defined such that Ω^ϵ is a connected set. More precisely, Y^1 denotes the pore phase (e.g. liquid or gas phase in wetting problems), see Figure 1.

These definitions allow us to rewrite (1.6) by the following microscopic formulation

$$\text{(Periodic porous medium)} \quad \begin{cases} -\epsilon^2 \mu \Delta \mathbf{u}_\epsilon + \nabla p_\epsilon = -\gamma \operatorname{div}\left(\nabla \phi_\epsilon \otimes \nabla \phi_\epsilon\right) & \text{in } \Omega^\epsilon_T, \\ \operatorname{div} \mathbf{u}_\epsilon = 0 & \text{in } \Omega^\epsilon_T, \\ \mathbf{u}_\epsilon = \mathbf{0} & \text{on } I^\epsilon_T, \\ \frac{\partial}{\partial t}\phi_\epsilon + \mathrm{Pe}(\mathbf{u}_\epsilon \cdot \nabla)\phi_\epsilon = \lambda \operatorname{div}\left(\nabla\left(f(\phi_\epsilon) - \Delta\phi_\epsilon\right)\right) & \text{in } \Omega^\epsilon_T, \\ \nabla_n \phi_\epsilon := \mathbf{n} \cdot \nabla \phi_\epsilon = w(\mathbf{x}) & \text{on } I^\epsilon_T, \\ \nabla_n \Delta \phi_\epsilon = 0 & \text{on } I^\epsilon_T, \\ \phi_\epsilon(\cdot, 0) = h(\cdot) & \text{on } \Omega^\epsilon, \end{cases} \tag{1.9}$$

where $I^\epsilon_T := I^\epsilon \times]0, T[$. The microscopic system (1.9) leads to a high-dimensional problem even under the assumption of periodicity since the space discretization parameter needs to be chosen to be much smaller than the characteristic size ϵ of the heterogeneities of the porous structure, e.g. left-hand side of Figure 1. This is a well-known issue and the homogenization method provides a systematic tool to reduce the intrinsic dimensional complexity by reliably averaging over the microscale represented by a single periodic reference pore Y. We note that the nonlinear problem (1.9) is characterized by a complex coupling between the micro- and the macroscale. This scale interaction leads to non-standard reference cell problems which also depend on the macroscale. As a consequence, the reference cell problems need to be computed for each macroscopic point now. **Serafim → Markus: the above sentence is confusing and needs re-writing** This high-dimensionality is an intrinsic feature of coupled non-linear partial differential equations studied in porous or highly heterogeneous structures, see [35, 36, 37]

Obviously, the systematic and reliable derivation of practical, convenient, and low-dimensional approximations is the key to feasible numerics of problems posed in porous media and provides a basis for computationally efficient schemes. To this end, we relax the full microscopic formulation (1.9) further by restricting (1.9) to periodic fluid flow. By taking the stationary version of equation $(1.9)_4$ on to a single periodic reference pore Y, we can formulate the following periodic flow problem

$$
\text{(Periodic flow)}\quad\begin{cases}
-\mu\Delta_{\mathbf{y}}\mathbf{u}+\nabla_{\mathbf{y}}p=-\gamma\mathrm{div}_{\mathbf{y}}\left(\nabla_{\mathbf{y}}\psi\otimes\nabla_{\mathbf{y}}\psi\right) & \text{in } Y^1,\\
\quad\mathrm{div}_{\mathbf{y}}\,\mathbf{u}=0 & \text{in } Y^1,\\
\quad\mathbf{u}=\mathbf{0} & \text{on } \partial Y^2,\\
\quad\mathbf{u}\text{ is } Y^1\text{-periodic},\\
\mathrm{Pe}(\mathbf{u}\cdot\nabla_{\mathbf{y}})\psi=\lambda\mathrm{div}_{\mathbf{y}}\left(\nabla_{\mathbf{y}}(f(\psi)-\Delta_{\mathbf{y}}\psi)\right) & \text{in } Y^1,\\
\quad\nabla_n\psi:=(\mathbf{n}\cdot\nabla_{\mathbf{y}})\psi=w & \text{on } \partial Y^2,\\
\quad\nabla_n\Delta_{\mathbf{y}}\psi=0 & \text{on } \partial Y^2,\\
\quad\psi\text{ is } Y^1\text{-periodic}.
\end{cases}\tag{1.10}
$$

We remark that it might be possible to further reduce problem (1.10) in case that the reference cell is only filled by one fluid phase, i.e., $\nabla_{\mathbf{y}}\psi=0$. This scenario requires to only solve for the periodic Stokes problem $(1.10)_1$–$(1.10)_4$ under zero driving force, i.e., $\mathbf{g}=\mathbf{0}$. We further note that physically, the periodic fluid velocity defined by (1.10) can be considered as the spatially periodic velocity of a moving frame. Motivated by [23, 33], we study the case of large Péclet number and consider the following distinguished limit.

Assumption (DP): *The Péclet number scales with respect to the characteristic pore size $\epsilon>0$ as follows:* $\mathrm{Pe}\sim\frac{1}{\epsilon}$.

Let us first discuss Assumption (DP). If one introduces the microscopic Péclet number $\mathrm{Pe}_{mic}:-=\frac{kT\ell\mathrm{U}}{D}$, then it follows immediately that $\mathrm{Pe}=\frac{\mathrm{Pe}_{mic}}{\epsilon}$. Since we introduced a periodic flow problem on the characteristic length scale $\ell>0$ of the pores by problem (1.10), it is obvious that we have to apply the microscopic Péclet number in a corresponding microscopic formulation, see (1.11) below. Hence, the periodic fluid velocity $\mathbf{u}(\mathbf{x}/\epsilon):=\mathbf{u}(\mathbf{y})$ enters the microscopic phase field problem as follows

$$
\text{(Microscopic problem)}\quad\begin{cases}
\frac{\partial}{\partial t}\phi_\epsilon+\frac{\mathrm{Pe}_{mic}}{\epsilon}(\mathbf{u}(\mathbf{x}/\epsilon)\cdot\nabla)\phi_\epsilon=\lambda\mathrm{div}\left(\nabla\left(f(\phi_\epsilon)-\Delta\phi_\epsilon\right)\right) & \text{in } \Omega^\epsilon_T,\\
\nabla_n\phi_\epsilon:=\mathbf{n}\cdot\nabla\phi_\epsilon=w(\mathbf{x}/\epsilon) & \text{on } I^\epsilon_T,\\
\nabla_n\Delta\phi_\epsilon=0 & \text{on } I^\epsilon_T,\\
\phi_\epsilon(\cdot,0)=\psi(\cdot) & \text{on } \Omega^\epsilon.
\end{cases}\tag{1.11}
$$

The main result of our study is the systematic derivation of upscaled immiscible flow equations which effectively account for pore geometry starting from the microscopic system (1.10)–(1.11), i.e.,

$$
\text{(Effective macro equation)}\quad\begin{cases}
p\frac{\partial\phi_0}{\partial t}=\mathrm{div}\Big(\Big[pf'(\phi_0)\hat{\mathrm{M}}-\Big(2\frac{f(\phi_0)}{\phi_0}-f'(\phi_0)\Big)\hat{\mathrm{M}}_v-\hat{\Theta}(\mathbf{x},t)\\
\qquad-\hat{\mathrm{C}}(\mathbf{x},t)\Big]\nabla\phi_0\Big)-f'(\phi_0)\mathrm{div}\Big((\hat{\mathrm{M}}_v+\hat{\mathrm{K}})\nabla\phi_0\Big)\\
\qquad+\frac{\lambda^2}{p}\mathrm{div}\Big(\hat{\mathrm{M}}_w\nabla\Big(\mathrm{div}\Big(\hat{\mathrm{D}}\nabla\phi_0\Big)-\tilde{w}_0\Big)\Big),
\end{cases}\tag{1.12}
$$

where $\hat{\Theta}$ and $\hat{\mathrm{C}}$ take the fluid convection into account. These two tensors account for the so-called diffusion-dispersion relations (e.g. Taylor-Aris-dispersion [4, 9, 39]). The result (1.12) makes use of the recently proposed splitting strategy for homogenization of fourth order problems in [37] and an asymptotic multiscale expansion with drift introduced in [3, 22].

The manuscript is organized as follows. We begin with physical motivation in Section 1.1. In Section 2, we present our main results and the corresponding proofs follow in the subsequent Section 3. Concluding remarks and open questions are offered in Section 4.

1.1 Physical motivation: Immiscible flows, diffusion-dispersion, and fuel cells

Fluid mixtures are ubiquitous in many scientific and engineering applications. The dynamics of phase interfaces between fluids plays a central role in rheology and hydrodynamics [8, 11, 15, 18, 30]. A recent attempt of a systematic extension towards non-equilibrium two-phase systems is [34], where the authors discuss the concept of local thermodynamic equilibrium of a Gibbs interface in order to relax the global thermodynamic equilibrium assumption. In [20], it is shown that the Cahn-Hilliard or diffuse interface formulation of a quasi-compressible binary fluid mixture allows for topological changes of the interface.

The study of flows in porous media is a delicate multiscale problem. This is evident, for instance, by the fact, that the full problem without any approximations is not computationally tractable with the present computational power. Also, from an empirical perspective the consideration of the full multiscale problem is very challenging due to the difficulty of obtaining detailed information about pore geometry. These empirical and computational restrictions strongly call for systematic and reliable approximations which capture the essential physics and elementary dynamic characteristics of the full problem in an averaged sense. A very common and intuitive strategy is volume averaging [32, 40]. The method of moments [4, 9] and multiple scale expansions [7, 29, 12] have been used in this context. The latter method is more systematic and reliable since it allows for a rigorous mathematical verification. The volume averaging strategy still lacks a consistent and generally accepted treatment of nonlinear terms. Therefore, the multiscale expansion strategy is used as a basis for the theoretical developments in the present study.

The celebrated works in [4, 9, 39] initiated an increasing interest in the understanding of hydrodynamic dispersion on the spreading of tracer particles transported by flow, with numerous applications, from transport of contaminants in rivers to chromatogaphy. In [33], it is shown that the multiscale expansion strategy allows to recover the dispersion relation found in [9]. The study of multiphase flows in porous media is considerably more complex; see e.g. the comprehensive review in [2] which still serves as a basis for several studies in the field. The central idea for many approaches to multiphase flows is to extend Darcy's law to multiple phases. With the help of Marle's averaging method [21] and a diffuse interface model, effective two-phase flow equations are presented in [27, 28]. In [5], Atkin and Craine even present constitutive theories for a binary mixture of fluids and a porous elastic solid.

An application of increasing importance for a renewable energy infrastructure are fuel cells [31]. This article combines the complex multiphase interactions with the help of the Cahn-Hilliard phase field method and a total free energy characterizing the fuel cell. An upscaling of the full thermodynamic model proposed in [31] is obviously very involved due to complex interactions over different scales. In this context, an upscaled macroscopic description of a simplified (i.e., no fluid flow and periodic catalyst layer) is derived in [36].

2 Results: Effective immiscible flow equations in porous media

For the presentation of our main result, we firt need to introduce the following:

Assumption (LTE): (Local thermodynamic equilibrium) *We assume that the reference cells satisfy the following local thermodynamic equilibrium condition, i.e.,*

$$\frac{\delta E(\phi)}{\delta\phi} = \mu(\phi) = f(\phi) - \lambda^2\Delta\phi = \text{const.}, \qquad \textit{for each } \frac{\mathbf{x}}{\epsilon} = \mathbf{y} \textit{ part of the same cell } Y\,, \tag{2.13}$$

where μ stands for the chemical potential which is only allowed to vary over the different reference cells.

The assumption (LTE) emerges as a key requirement for the homogenization of nonlinearly coupled partial differential equations. This equilibrium requirement is well-known and generally accepted in physical sciences if there exists a separation of macroscopic (size of the porous medium) and microscopic (characteristic pore size) length scales. This kind of equilibrium assumptions are widely applied to a variety of physical situations such as diffusion [24], macroscale thermodynamics in porous media [6], ionic transport in porous media based on dilute solution theory [35], and fuel cells [36], and density-functional theory [42, 16]. Local equilibrium assumptions (2.13) emerge as key requirements for the mathematical well-posedness of the corresponding cell problems which define effective transport coefficients in homogenized, nonlinear (and coupled) problems.

As in [37], the homogeneous free energy characterizing the equilibrium limiting values of two different phases needs to fulfill the following

Assumption (F): *Assume that the homogeneous free energy F satisfies for real parameters $\alpha_2 > \alpha_1 > 0$, which define F as a double-well potential by $F(s) = (s-\alpha_1)^2(s-\alpha_2)^2$, such that*

$$25(\alpha_1+\alpha_2)^2 - 20(\alpha_1^2+\alpha_2^2+3\alpha_1\alpha_2) > (\alpha_1+\alpha_2)^2/4\,. \tag{2.14}$$

Define also the variable $\tilde{w}_0$ and the effective fluid velocity v below!

Theorem 2.1 (Effective convective Cahn-Hilliard equation) *We assume that the* Assumptions (DP), (LTE), *and* (F) *hold and let $\psi(\mathbf{x}) \in H^2_E(\Omega)$.*

Then, the microscopic equations (1.10)–(1.11) for immiscible flows in porous media admit the following effective macroscopic form after averaging over the microscale, i.e.,

$$\begin{cases} p\frac{\partial\phi_0}{\partial t} = \mathrm{div}\Big(\Big[pf'(\phi_0)\hat{\mathrm{M}} - \Big(2\frac{f(\phi_0)}{\phi_0} - f'(\phi_0)\Big)\hat{\mathrm{M}}_v - \hat{\Theta}(\mathbf{x},t) \\ \qquad -\hat{\mathrm{C}}(\mathbf{x},t)\Big]\nabla\phi_0\Big) - f'(\phi_0)\mathrm{div}\Big((\hat{\mathrm{M}}_v+\hat{\mathrm{K}})\nabla\phi_0\Big) \\ \qquad +\frac{\lambda^2}{p}\mathrm{div}\Big(\hat{\mathrm{M}}_w\nabla\Big(\mathrm{div}\Big(\hat{\mathrm{D}}\nabla\phi_0\Big) - \tilde{w}_0\Big)\Big) & \textit{in } \Omega_T\,, \\ \nabla_n\phi_0 := \mathbf{n}\cdot\nabla\phi_0 = w(\mathbf{x}/\epsilon) & \textit{on } \{\partial\Omega\setminus I\}_T\,, \\ \nabla_n\Delta\phi_0 = 0 & \textit{on } \{\partial\Omega\setminus I\}_T\,, \\ \phi_0(\cdot,0) = \psi(\cdot) & \textit{in } \Omega\,, \end{cases} \tag{2.15}$$

where the tensors $\hat{\mathrm{K}} := \{\kappa_{kl}\}_{1\le k,l\le d}$, $\hat{\Theta} := \{\theta_{kl}\}_{1\le k,l\le d}$, $\hat{\mathrm{C}} := \{\mathrm{c}_{ik}\}_{1\le i,k\le d}$, $\hat{\mathrm{D}} := \{\mathrm{d}_{ik}\}_{1\le i,k\le d}$, $\hat{\mathrm{M}}_v = \{\mathrm{m}^v_{ik}\}_{1\le i,k\le d}$, *and* $\hat{\mathrm{M}}_w = \{\mathrm{m}^w_{ik}(\mathbf{x},t)\}_{1\le i,k\le d}$ *are defined by*

$$\begin{cases} \kappa_{kl} := \frac{1}{|Y|}\sum_{i,j=1}^d \int_{Y^1} \frac{\partial}{\partial y_i}\Big(\mathrm{m}_{ij}\frac{\partial\zeta^{kl}(\mathbf{y})}{\partial y_j}\Big)\,d\mathbf{y}\,, \\ \theta_{kl} := \frac{\mathrm{Pe}_{mic}}{|Y|}\int_{Y^1}(\mathbf{u}\cdot\nabla_{\mathbf{y}})\zeta^{kl}(\mathbf{y})\,d\mathbf{y}\,, \\ \mathrm{c}_{ik} := \frac{\mathrm{Pe}_{mic}}{|Y|}\int_{Y^1}(\mathrm{u}^i - \mathrm{v}^i)\delta_{ik}\xi^k_v(\mathbf{y})\,d\mathbf{y}\,, \\ \mathrm{d}_{ik} := \frac{1}{|Y|}\sum_{j=1}^d\int_{Y^1}\Big(\delta_{ik} - \delta_{ij}\frac{\partial\xi^k_v}{\partial y_j}\Big)\,d\mathbf{y}\,, \\ \mathrm{m}^v_{ik} := \frac{1}{|Y|}\sum_{j=1}^d\int_{Y^1}\Big(\mathrm{m}_{ik} - \mathrm{m}_{ij}\frac{\partial\xi^k_v}{\partial y_j}\Big)\,d\mathbf{y}\,, \\ \mathrm{m}^w_{ik}(\mathbf{x},t) := \frac{1}{|Y|}\sum_{j=1}^d\int_{Y^1}\Big(\mathrm{m}_{ik} - \mathrm{m}_{ij}\frac{\partial\xi^k_w(\mathbf{x},\mathbf{y},t)}{\partial y_j}\Big)\,d\mathbf{y}\,. \end{cases} \tag{2.16}$$

The effective fluid velocity $\mathbf{v}$ *is defined by* $\mathbf{v}:=\frac{\mathrm{Pe}_{\mathrm{loc}}}{|Y^1|}\int_{Y^1}\mathrm{u}^j(y)\,dy$ *where* $\mathbf{u}$ *solves the periodic reference cell problem (1.10). The corrector functions* $\zeta^{kl},\xi_v^k\in H^1_{per}(Y^1)$ *and* $\xi_w^k\in L^2(\Omega;H^1_{per}(Y^1))$ *for* $1\leq k,l\leq d$ *solve in the distributional sense the following reference cell problems*

$$\zeta^{kl}:\quad\begin{cases}-\sum_{i,j,k,l=1}^d\frac{\partial}{\partial y_i}\left(\delta_{ij}\left(\frac{\partial}{\partial y_j}\zeta^{kl}(\mathbf{y})-2\delta_{kj}\xi_v^l(\mathbf{y})\right)\right)=0 & \text{in } Y^1\,,\\ \nabla_n\phi_2=g_\epsilon & \text{on } \partial Y_w^1\cap\partial Y_w^2\,,\\ \phi_2 \text{ is } Y\text{-periodic}, & \end{cases}\tag{2.17}$$

$$\xi_w^k:\quad\begin{cases}-\sum_{i,j,k=1}^d\frac{\partial}{\partial y_i}\left(\delta_{ik}-\delta_{ij}\frac{\partial\xi_w^k}{\partial y_j}\right)\\ \qquad=\lambda^2\sum_{k,i,j=1}^d\frac{\partial}{\partial y_i}\left(\mathrm{m}_{ik}-\frac{f(\phi_0)}{f'(\phi_0)\phi_0}\mathrm{m}_{ij}\frac{\partial\xi_v^k}{\partial y_j}\right) & \text{in } Y^1\,,\\ \sum_{i,j,k=1}^d\mathrm{n}_i\left(\delta_{ij}\frac{\partial\xi_w^k}{\partial y_j}-\delta_{ik}\right)\\ \qquad-\lambda^2\sum_{k,i,j=1}^d\frac{\partial}{\partial y_i}\left(\mathrm{m}_{ik}-\frac{f(\phi_0)}{f'(\phi_0)\phi_0}\mathrm{m}_{ij}\frac{\partial\xi_v^k}{\partial y_j}\right)\Big)=0 & \text{on } \partial Y^1\,,\\ \xi_w^k(\mathbf{y}) \text{ is } Y\text{-periodic and } \mathcal{M}_{Y^1}(\xi_w^k)=0, & \end{cases}\tag{2.18}$$

$$\xi_v^k:\quad\begin{cases}-\sum_{i,j=1}^d\frac{\partial}{\partial y_i}\left(\delta_{ik}-\delta_{ij}\frac{\partial\xi_v^k}{\partial y_j}\right)=0 & \text{in } Y^1\,,\\ \sum_{i,j=1}^d\mathrm{n}_i\left(\delta_{ij}\frac{\partial\xi_w^k}{\partial y_j}-\delta_{ik}\right)=0 & \text{on } \partial Y^1\,,\\ \xi_v^k(\mathbf{y}) \text{ is } Y\text{-periodic and } \mathcal{M}_{Y^1}(\xi_v^k)=0. & \end{cases}\tag{2.19}$$

3 Proof of Theorem 2.1

As in [37], we introduce the multiscale operators

$$\begin{aligned}\mathcal{A}_0&=-\sum_{i,j=1}^d\frac{\partial}{\partial y_i}\left(\delta_{ij}\frac{\partial}{\partial y_j}\right), & \mathcal{B}_0&=-\sum_{i,j=1}^d\frac{\partial}{\partial y_i}\left(\mathrm{m}_{ij}\frac{\partial}{\partial y_j}\right),\\ \mathcal{A}_1&=-\sum_{i,j=1}^d\left[\frac{\partial}{\partial x_i}\left(\delta_{ij}\frac{\partial}{\partial y_j}\right)\right. & \mathcal{B}_1&=-\sum_{i,j=1}^d\left[\frac{\partial}{\partial x_i}\left(\mathrm{m}_{ij}\frac{\partial}{\partial y_j}\right)\right.\\ &\qquad\left.+\frac{\partial}{\partial y_i}\left(\delta_{ij}\frac{\partial}{\partial x_j}\right)\right], & &\qquad\left.+\frac{\partial}{\partial y_i}\left(\mathrm{m}_{ij}\frac{\partial}{\partial x_j}\right)\right],\\ \mathcal{A}_2&=-\sum_{i,j=1}^d\frac{\partial}{\partial x_j}\left(\delta_{ij}\frac{\partial}{\partial x_j}\right), & \mathcal{B}_2&=-\sum_{i,j=1}^d\frac{\partial}{\partial x_j}\left(\mathrm{m}_{ij}\frac{\partial}{\partial x_j}\right),\end{aligned}\tag{3.20}$$

which make use of the micro-scale $\frac{\mathbf{x}}{\epsilon}=:\mathbf{y}\in Y$ such that $\mathcal{A}_\epsilon:=\epsilon^{-2}\mathcal{A}_0+\epsilon^{-1}\mathcal{A}_1+\mathcal{A}_2$, and $\mathcal{B}_\epsilon:=\epsilon^{-2}\mathcal{B}_0+\epsilon^{-1}\mathcal{B}_1+\mathcal{B}_2$. Herewith, the Laplace operators Δ and $\mathrm{div}\left(\hat{\mathrm{M}}\nabla\right)$ become $\Delta u^\epsilon(\mathbf{x})=\mathcal{A}_\epsilon u(\mathbf{x},\mathbf{y})$ and $\mathrm{div}\left(\hat{\mathrm{M}}\nabla\right)u^\epsilon(\mathbf{x})=\mathcal{B}_\epsilon u(\mathbf{x},\mathbf{y})$, respectively, where $u^\epsilon(\mathbf{x},t):=u(\mathbf{x}-\frac{\mathbf{v}}{\epsilon}t,\mathbf{y},t)$. Due to the drift, we additionally have

$$\frac{\partial}{\partial t}u_\epsilon=\left(\frac{\partial}{\partial t}-\frac{\mathbf{v}}{\epsilon}\right)u_\epsilon\,,\tag{3.21}$$

where we find below by the Fredholm alternative (or a solvability constraint) that $\mathbf{v}:=\frac{\mathrm{Pe}_{\mathrm{loc}}}{|Y^1|}\int_{Y^1}\mathrm{u}^j(y)\,dy$. As in [37] we apply the method of formal asymptotic multiscale expansions together with the splitting strategy introduced therein. As a consequence, we obtain the following sequence of problems by comparing terms of the same order in ϵ, with the first three problems being,

$$\mathcal{O}(\epsilon^{-2}):\quad\begin{cases}\mathcal{B}_0w_0=\mathcal{B}_0f(\phi_0)-\mathrm{Pe}_{mic}(\mathbf{u}\cdot\nabla)_{\mathbf{y}})v_0 & \text{in } Y^1\,,\\ \quad\text{no flux b.c.}, & \\ \quad w_0 \text{ is } Y^1\text{-periodic}, & \\ \mathcal{A}_0v_0=0 & \text{in } Y^1\,,\\ \quad\nabla_nv_0=0 & \text{on } \partial Y_w^1\cap\partial Y_w^2\,,\\ \quad\phi_0 \text{ is } Y^1\text{-periodic}, & \end{cases}\tag{3.22}$$

$$
\mathcal{O}(\epsilon^{-1}):\quad \begin{cases}
\mathcal{B}_0 w_1 = -\mathcal{B}_1 w_0 - \mathcal{B}_0\left[f(\phi_0)\frac{\phi_1}{\phi_0}\right] - \mathcal{B}_1 f(\phi_0) & \\
\qquad -\mathrm{Pe}_{mic}(\mathbf{u}\cdot\nabla_{\mathbf{y}})v_1 - \mathrm{Pe}_{mic}((\mathbf{u}-\mathbf{v})\cdot\nabla)v_0 & \text{in } Y^1,\\
\quad \text{no flux b.c.}, & \\
\quad w_1 \text{ is } Y^1\text{-periodic}, & \\
\mathcal{A}_0 v_1 = -\mathcal{A}_1 v_0 & \text{in } Y^1,\\
\quad \nabla_n v_1 = 0 & \text{on } \partial Y^1_w \cap \partial Y^2_w,\\
\quad \phi_1 \text{ is } Y^1\text{-periodic}, &
\end{cases} \tag{3.23}
$$

$$
\mathcal{O}(\epsilon^{0}):\quad \begin{cases}
\mathcal{B}_0 w_2 = -\lambda^2(\mathcal{B}_2 w_0 + \mathcal{B}_1 w_1) & \\
\qquad -\mathcal{B}_0\left[\frac{1}{2}f''(\phi_0)\phi_1^2 + f'(\phi_0)\phi_2\right] & \\
\qquad -\mathcal{B}_1\left[f(\phi_0)\frac{\phi_1}{\phi_0}\right] - \mathcal{B}_2 f(\phi_0) & \\
\qquad -\mathrm{Pe}_{mic}(\mathbf{u}\cdot\nabla_{\mathbf{y}})\phi_2 - \mathrm{Pe}_{mic}((\mathbf{u}-\mathbf{v})\cdot\nabla)v_1 - \partial_t(-\Delta)^{-1}w_0 & \text{in } Y^1,\\
\quad \text{no flux b.c.}, & \\
\quad w_2 \text{ is } Y^1\text{-periodic}, & \\
\mathcal{A}_0 v_2 = -\mathcal{A}_2 v_0 - \mathcal{A}_1 v_1 + w_0 & \text{in } Y^1,\\
\quad \nabla_n v_2 = g_\epsilon & \text{on } \partial Y^1_w \cap \partial Y^2_w,\\
\quad \phi_2 \text{ is } Y^1\text{-periodic}, &
\end{cases} \tag{3.24}
$$

As usual, the first problem (3.22) induces that the leading order terms $\phi_0 = v_0 + \overline{\phi}_0$ and w_0 are independent of the micro-scale $\mathbf{y}$. The second problem (3.23) reads in explicit form for v_1 as follows,

$$
\xi_\phi:\quad \begin{cases}
-\sum_{i,j=1}^d \frac{\partial}{\partial y_i}\left(\delta_{ik} - \delta_{ij}\frac{\partial \xi_v^k}{\partial y_j}\right) = & \\
\qquad = -\mathrm{div}\left(\mathbf{e}_k - \nabla_y \xi_v^k\right) = 0 & \text{in } Y^1,\\
\mathbf{n}\cdot\left(\nabla\xi_v^k + \mathbf{e}_k\right) = 0 & \text{on } \partial Y^1_w \cap \partial Y^2_w,\\
\xi_v^k(\mathbf{y}) \text{ is } Y\text{-periodic and } \mathcal{M}_{Y^1}(\xi_v^k) = 0, &
\end{cases} \tag{3.25}
$$

which represents the reference cell problem for v_0 after identifying $v_1 = -\sum_{k=1}^d \xi_v^k(\mathbf{y})\frac{\partial v_0}{\partial x_k}$.

The cell problem for w_1 is substantially more involved since it depends on the fluid velocity $\mathbf{u}$ and the the corrector ξ_v^k from (3.25). Specifically,

$$
\begin{aligned}
\sum_{k,i,j=1}^d \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ij}\frac{\partial \xi_w^k}{\partial y_j}\right)\frac{\partial w_0}{\partial x_k} &= \sum_{i,j=1}^d \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ij}\frac{\partial w_0}{\partial x_j}\right)\\
&\quad - \frac{f(\phi_0)}{\phi_0}\sum_{k,i,j=1}^d \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ij}\frac{\partial \xi_v^k}{\partial y_j}\right)\frac{\partial \phi_0}{\partial x_k} + \sum_{k,i,j=1}^d \frac{\partial}{\partial y_i}\left(f'(\phi_0)\mathrm{m}_{ij}\frac{\partial \phi_0}{\partial x_i}\right)\\
&\quad + \mathrm{Pe}_{mic}\sum_{k,i,j=1}^d \left(\mathrm{u}_0^i\frac{\partial}{\partial y_i}\right)\xi_v^k\frac{\partial \phi_0}{\partial x_k} - \mathrm{Pe}_{mic}\sum_{i=1}^d\left(\mathrm{u}^i - \mathrm{v}^i\right)\frac{\partial \phi_0}{\partial x_i},
\end{aligned} \tag{3.26}
$$

where the last line can be simplified under the Assumption (LTE), i.e., $f'(\phi)\frac{\partial \phi}{\partial x_k} = f'(\phi)\frac{\partial v}{\partial x_k} = \lambda^2\frac{\partial w}{\partial x_k}$ for $1 \le k \le d$, to

$$
\lambda^2\left(\frac{\mathrm{Pe}_{mic}}{f'(\phi_0)}\sum_{k,i=1}^d\left(\frac{\partial \xi_v^k}{\partial y_i} - 1\right)\mathrm{u}^i + \mathrm{v}^i\right)\frac{\partial w_0}{\partial x_k}. \tag{3.27}
$$

If we also rewrite the remaining terms under Assumption (LTE) as in [37], then we end up with the following cell problem,

$$\begin{cases} -\sum_{i,j,k=1}^{d}\frac{\partial}{\partial y_i}\left(\delta_{ik}-\delta_{ij}\frac{\partial \xi_w^k}{\partial y_j}\right) & \\ \qquad =\lambda^2\Big(\sum_{k,i,j=1}^{d}\frac{\partial}{\partial y_i}\left(\mathrm{m}_{ik}-\frac{f(\phi_0)}{f'(\phi_0)\phi_0}\mathrm{m}_{ij}\frac{\partial \xi_v^k}{\partial y_j}\right) & \\ \qquad +\frac{\mathrm{Pe}_{mic}}{f'(\phi_0)}\sum_{k,i=1}^{d}\left(\frac{\partial \xi_v^k}{\partial y_i}-1\right)\mathrm{u}^i+\mathrm{v}^i\Big) & \text{in } Y^1, \\ \sum_{i,j,k=1}^{d}\mathrm{n}_i\Big(\left(\delta_{ij}\frac{\partial \xi_w^k}{\partial y_j}-\delta_{ik}\right) & \\ \qquad -\lambda^2\sum_{k,i,j=1}^{d}\frac{\partial}{\partial y_i}\left(\mathrm{m}_{ik}-\frac{f(\phi_0)}{f'(\phi_0)\phi_0}\mathrm{m}_{ij}\frac{\partial \xi_v^k}{\partial y_j}\right) & \\ \qquad +\frac{\mathrm{Pe}_{mic}}{f'(\phi_0)}\sum_{k,i=1}^{d}\left(\frac{\partial \xi_v^k}{\partial y_i}-1\right)\mathrm{u}^i+\mathrm{v}^i\Big)=0 & \text{on } \partial Y_w^1\cap\partial Y_w^2, \\ \xi_w^k(\mathbf{y}) \text{ is } Y\text{-periodic and } \mathcal{M}_{Y^1}(\xi_w^k)=0. & \end{cases} \tag{3.28}$$

We are then left to study the last problem (3.24) arising by the asymptotic multiscale expansions. Problem $(3.24)_2$ for v_2 is classical and leads immediately to the upscaled equation

$$-\Delta_{\hat{\mathrm{D}}}v_0:=-\mathrm{div}\left(\hat{\mathrm{D}}\nabla v_0\right)=\theta_1 w_0+\tilde{g}_0\,, \tag{3.29}$$

see also [37], where the porous media correction tensor $\hat{\mathrm{D}}:=\{\mathrm{d}_{ik}\}_{1\le i,k\le d}$ is defined by

$$|Y|\,\mathrm{d}_{ik}:=\sum_{j=1}^{d}\int_{Y^1}\left(\delta_{ik}-\delta_{ij}\frac{\partial \xi_v^k}{\partial y_j}\right)d\mathbf{y}\,. \tag{3.30}$$

Next, we apply the Fredholm alternative on equation $(3.24)_1$, i.e.,

$$\begin{aligned} \int_{Y^1}\Big\{&-\lambda^2(\mathcal{B}_2 w_0+\mathcal{B}_1 w_1)-\mathcal{B}_0\left(\frac{1}{2}f''(\phi_0)\phi_1^2+f'(\phi_0)\phi_2\right)\\ &-\mathcal{B}_1\left[f(\phi_0)\frac{\phi_1}{\phi_0}\right]-\mathcal{B}_2 f(\phi_0)-\partial_t(-\Delta)^{-1}w_0\\ &-\mathrm{Pe}_{mic}(\mathbf{u}\cdot\nabla_{\mathbf{y}})\phi_2-\mathrm{Pe}_{mic}((\mathbf{u}-\mathbf{v})\cdot\nabla)v_1\Big\}\,d\mathbf{y}=0\,. \end{aligned} \tag{3.31}$$

The term multiplied by λ^2 in (3.31) can immediately be rewritten by

$$\lambda^2\int_{Y^1}-(\mathcal{B}_2 w_0+\mathcal{B}_1 w_1)\,d\mathbf{y}=-\mathrm{div}\left(\hat{\mathrm{M}}_w\nabla w_0\right), \tag{3.32}$$

where the effective tensor $\hat{\mathrm{M}}_w=\{\mathrm{m}_{ik}^w\}_{1\le i,k\le d}$ is defined by

$$\mathrm{m}_{ik}^w:=\frac{1}{|Y|}\sum_{j=1}^{d}\int_{Y^1}\left(\mathrm{m}_{ik}-\mathrm{m}_{ij}\frac{\partial \xi_w^k}{\partial y_j}\right)d\mathbf{y}\,. \tag{3.33}$$

The first two terms on the second line in (3.31) transform as in [37] to

$$\begin{aligned} &\frac{1}{|Y|}\int_{Y^1}\left(-\mathcal{B}_1\left[f(\phi_0)\frac{\phi_1}{\phi_0}\right]-\mathcal{B}_2 f(\phi_0)\right)d\mathbf{y}\\ &=\mathrm{div}\left(\left[\theta_1 f'(\phi_0)\hat{\mathrm{M}}-2\frac{f(\phi_0)}{\phi_0}\hat{\mathrm{M}}_v\right]\nabla\phi_0\right), \end{aligned} \tag{3.34}$$

where the tensor $\hat{\mathrm{M}}_v = \left\{\mathrm{m}^v_{ij}\right\}_{1\le i,k\le d}$ is defined by

$$\mathrm{m}^v_{ik} := \frac{1}{|Y|}\sum_{j=1}^d \int_{Y^1} \left(\mathrm{m}_{ij}\frac{\partial \xi^k_v}{\partial y_j}\right) d\mathbf{y}\,. \tag{3.35}$$

The last term on the first line in (3.31) reveals the (non-trivial) coupling between the micro- and the macro-scale. After integrating over Y it can be written in the following compact form,

$$\begin{aligned}\frac{1}{|Y|}\int_{Y^1} -\mathcal{B}_0\left[\frac{1}{2}f''(\phi_0)\phi_1^2 + f'(\phi_0)\phi_2\right] d\mathbf{y} = {}& \mathrm{div}\left(f'(\phi_0)\hat{\mathrm{M}}_v\nabla\phi_0\right)\\ & -f'(\phi_0)\mathrm{div}\left(\hat{\mathrm{M}}_v\nabla\phi_0\right) + f'(\phi_0)\kappa(\mathbf{x},t)\,,\end{aligned} \tag{3.36}$$

where

$$\kappa(\mathbf{x},t) := \frac{1}{|Y|}\sum_{i,j=1}^d \int_{Y^1} \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ij}\frac{\partial \phi_2(\mathbf{x},\mathbf{y},t)}{\partial y_j}\right) d\mathbf{y}\,. \tag{3.37}$$

The terms in the last line of (3.31) become

$$\begin{aligned}&-\frac{1}{|Y|}\int_{Y^1} \mathrm{Pe}_{mic}(\mathbf{u}\cdot\nabla_{\mathbf{y}})\phi_2 + \mathrm{Pe}_{mic}((\mathbf{u}-\mathbf{v})\cdot\nabla)v_1\, d\mathbf{y}\\ &= -\frac{1}{|Y|}\int_{Y^1} \mathrm{Pe}_{mic}(\mathbf{u}\cdot\nabla_{\mathbf{y}})\phi_2\, d\mathbf{y} - \frac{\mathrm{Pe}_{mic}}{|Y|}\sum_{k,i=1}^d \int_{Y^1} (\mathrm{u}^i - \mathrm{v}^i)\frac{\partial}{\partial x_i}\left(\delta_{ik}\xi^k_v(\mathbf{y})\frac{\partial}{\partial x_k}v_0\right) d\mathbf{y}\end{aligned} \tag{3.38}$$

These considerations finally lead to the following upscaled phase field equation

$$\begin{aligned}p\frac{\partial \phi_0}{\partial t} = {}& \mathrm{div}\Big(\Big[pf'(\phi_0)\hat{\mathrm{M}} - \left(2\frac{f(\phi_0)}{\phi_0} - f'(\phi_0)\right)\hat{\mathrm{M}}_v - \hat{\Theta}(\mathbf{x},t)\\ & -\hat{\mathrm{C}}(\mathbf{x},t)\Big]\nabla\phi_0\Big) - f'(\phi_0)\mathrm{div}\left((\hat{\mathrm{M}}_v + \hat{\mathrm{K}})\nabla\phi_0\right)\\ & + \frac{\lambda^2}{p}\mathrm{div}\left(\hat{\mathrm{M}}_w\nabla\left(\mathrm{div}\left(\hat{\mathrm{D}}\nabla\phi_0\right) - \tilde{w}_0\right)\right)\,,\end{aligned} \tag{3.39}$$

where the tensors $\hat{\mathrm{K}} := \{\kappa_{kl}\}_{1\le k,l\le d}$, $\hat{\Theta} := \{\theta_{kl}\}_{1\le k,l\le d}$, and $\hat{\mathrm{C}} := \{\mathrm{c}_{ik}\}_{1\le i,k\le d}$ are defined by

$$\begin{cases} \kappa_{kl}(\mathbf{x},t) := \frac{1}{|Y|}\sum_{i,j=1}^d \int_{Y^1} \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ij}\frac{\partial \zeta^{kl}(\mathbf{y})}{\partial y_j}\right) d\mathbf{y}\,,\\ \theta_{kl}(\mathbf{x},t) := \frac{\mathrm{Pe}_{mic}}{|Y|}\int_{Y^1} (\mathbf{u}\cdot\nabla_{\mathbf{y}})\zeta^{kl}(\mathbf{y})\, d\mathbf{y}\,,\\ \mathrm{c}_{ik}(\mathbf{x},t) := \frac{\mathrm{Pe}_{mic}}{|Y|}\int_{Y^1} (\mathrm{u}^i - \mathrm{v}^i)\delta_{ik}\xi^k_v(\mathbf{y})\, d\mathbf{y}\,,\end{cases} \tag{3.40}$$

and ζ^{kl} is a second order corrector, which is defined by $\phi_2(\mathbf{x},\mathbf{y},t) = \sum_{k,l=1}^d \zeta^{kl}(\mathbf{y})\frac{\partial^2\phi_0(\mathbf{x})}{\partial x_k \partial x_l}$. The second order term ϕ^2 solves

$$\begin{cases} \mathcal{A}_0\phi_2(\mathbf{x}.\mathbf{y},t) = \sum_{k,i,j=1}^d \Big\{ \frac{\partial}{\partial y_i}\left(\delta_{ij}\xi^k_v(\mathbf{y})\frac{\partial^2\phi_0(\mathbf{x},t)}{\partial x_j \partial x_k}\right) & \\ \qquad\qquad + \frac{\partial}{\partial x_i}\left(\delta_{ij}\frac{\partial}{\partial y_j}\xi^k_v(\mathbf{y})\frac{\partial \phi_0(\mathbf{x},t)}{\partial x_k}\right)\Big\} & \text{in } Y^1,\\ \nabla_n\phi_2 = g_\epsilon & \text{on } \partial Y^1_w \cap \partial Y^2_w,\\ \phi_2 \text{ is } Y^1\text{-periodic}, & \end{cases} \tag{3.41}$$

which can be further simplified to

$$\zeta: \quad \begin{cases} -\sum_{i,j,k,l=1}^d \frac{\partial}{\partial y_i}\left(\delta_{ij}\left(\frac{\partial}{\partial y_j}\zeta^{kl}(\mathbf{y}) - 2\delta_{kj}\xi^l_v(\mathbf{y})\right)\right) = 0 & \text{in } Y^1,\\ \nabla_n\phi_2 = g_\epsilon & \text{on } \partial Y^1_w \cap \partial Y^2_w,\\ \phi_2 \text{ is } Y^1\text{-periodic}. & \end{cases} \tag{3.42}$$

4 Conclusion

The main new result here is the extension of the results in the study by Schmuck *et al.* in the absence of flow [37] to include a periodic fluid flow in the case of sufficiently large Péclet number. The resulting new effective porous media approximation (2.15) of the microscopic Stokes-Cahn-Hilliard problem (1.10)–(1.11) reveals interesting physical characteristics such as diffusion-dispersion relations by $(2.16)_2$–$(2.16)_3$ for instance. The homogenization methodology serves as a well-established tool for the systematic and rigorous derivation of effective macroscopic porous media equations starting with the fundamental works on Darcy's law [10]. As a byproduct of the methodology developed here, one recovers rigorously and systematically the dispersion relations proposed in [9].

We note that the Cahn-Hilliard and related equations generally model more complex material transport [25, 31] than classical Fickian diffusion. In this context, our result of an upscaled convective Cahn-Hilliard equation hence provides diffusion-dispersion relations for generalized non-Fickian material transport, that is, not just the product of a gradient of particle concentration and a constant diffusion matrix.

Of course, it would be of interest to remove the periodicity assumption on the fluid flow imposed by (1.10). This assumption implies a quasi-steady state on the fluid velocity and seems currently to be inevitable for the homogenization machinery to work such as the assumption of local thermodynamic equilibrium on the level of the reference cells.

So far, we are restricted to mixtures of two fluid by our model. **Serafim → Markus: I changed "liquids" to "fluids" throughout. I believe that the methodology is applied to an interface between two fluids** An extension towards mixtures of $N>0$ components is studied in [26] where the authors compare a local and non-local model for incompressible fluids. In specific applications, it might be of interest to extend the here developed framework towards such multi-component mixtures.

Finally, the rigorous and systematic derivation of effective macroscopic immiscible flow equations provides a promising and convenient alternative in view of the broad applicability of the Cahn-Hilliard equations. The strength of our approach is based on its foundation on a thermodynamically motivated homogeneous free energy which is generally derived on systematic physical grounds. Moreover, even if a systematic derivation is impossible, one can often mathematically design such a free energy based on physical principles and phenomenological observations. We expect promising insights of the systematically averaged interplay of micro- and macroscale in a wide variety of applications, e.g. for the optimization of microfluidic devices where the scale effects discovered here are crucial. A dedicated numerical study might also allow to rationally analyze current applications in science and engineering and hopefully reveal potential new ones.

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