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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 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 (Schmuck *et al* 2012 *Proc. R. Soc.* A **468** 3705–24), we rigorously derive an effective macroscopic phase field formulation under the assumption of periodic flow and a sufficiently large Péclet number. As for classical convection–diffusion problems, we obtain systematically diffusion–dispersion relations (including Taylor–Aris-dispersion). Our results 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.

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1. Introduction

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 [9, 12, 17, 22, 34]. A recent attempt of a systematic extension towards non-equilibrium two-phase systems is [38], where the authors discuss the concept of local thermodynamic equilibrium of a Gibbs interface in order to relax the global thermodynamic equilibrium assumption. In [24], 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. Also of increasing interest is the mathematical and physical understanding of wetting using diffuse interface formulations [45, 46] as well as wetting in the presence of complexities such as electric fields [16, 25].

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 [2, 20]. Also, from an empirical perspective the consideration of the full multiscale problem is very challenging due to the difficulty of obtaining detailed information about the 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 [36, 50]. The method of moments [4, 10] and multiple scale expansions [7, 13, 33] 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, 10, 48] 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 [37], it is shown that the multiscale expansion strategy allows to recover the dispersion relation found in [10]. 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 [26] and a diffuse interface model, effective two-phase flow equations are presented in [31, 32]. In [5], Atkin and Craine even present constitutive theories for a binary mixture of fluids and a porous elastic solid. A combination of the homogenization method and a multiphase extension of Darcy's law as a description of multiphase flows in porous media is applied in the articles [8, 43], for instance.

An application of increasing importance for a renewable energy infrastructure are fuel cells [35]. 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 [35] 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 [40, 41].

Consider the total energy density for an interface between two phases,

$$e(\boldsymbol{x}(\boldsymbol{X},t),t) := \frac{1}{2}\left|\frac{\partial \boldsymbol{x}(\boldsymbol{X},t)}{\partial t}\right|^2 - \frac{\lambda}{2}\left|\nabla_{\boldsymbol{x}}\phi(\boldsymbol{x}(\boldsymbol{X},t),t))\right|^2 - \frac{\lambda}{2}F(\phi(\boldsymbol{x}(\boldsymbol{X},t),t)), \qquad (1)$$

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) $= \sigma\eta$. The variable $\boldsymbol{X}$ stands for the Lagrangian (initial) material coordinate and $\boldsymbol{x}(\boldsymbol{X}, t)$ represents the Eulerian (reference) coordinate. Our derivation (section 4) is valid for general free energies F and uses the method of an asymptotic multiscale expansion with drift [3]. Furthermore, we establish the well-posedness (theorem 3.5) of the upscaled/homogenized equations for polynomial free energies of the following form [49]:

Assumption (PF). *The free energy densities F in (1) are polynomials of order $2r-1$, i.e.,*

$$f(u) = \sum_{i=1}^{2r-1} a_i u^i, \qquad r \in \mathbb{N}, \quad r \geqslant 2, \tag{2}$$

with $f(u)=F'(u)$ vanishing at $u=0$,

$$F(u) = \sum_{i=2}^{2r} b_i u^i, \qquad i b_i = a_{i-1}, \quad 2 \leqslant i \leqslant 2r, \tag{3}$$

where the leading coefficient of both F and f is positive, i.e. $a_{2r-1} = 2r b_{2r} > 0$.

Remark 1.1 (Double-well potential). Free energies F satisfying the *assumption (PF)* form a general class which also includes the double-well potential for $r = 2$ with $f(u) = -\alpha u + \beta u^3$, α, $\beta > 0$, for which $(7)_4$ is called the convective Cahn–Hilliard equation. We note that the double-well is scaled by $1/4\eta^2$, i.e. $F(u) = (1/4\eta^2)(u^2-1)^2$ such that one recovers the Hele–Shaw problem in the limit $\eta \to 0$ [19, 22].

The last two terms in (1) form the well-known density of the Cahn–Hilliard/Ginzburg–Landau phase field formulation adapted to the flow map $\boldsymbol{x}(\boldsymbol{X}, t)$ defined by

$$\begin{aligned} &\frac{\partial \boldsymbol{x}}{\partial t} = \boldsymbol{u}(\boldsymbol{x}(\boldsymbol{X}, t), t), \\ &\boldsymbol{x}(\boldsymbol{X}, 0) = \boldsymbol{X}. \end{aligned} \tag{4}$$

The first term in (1) is the kinetic energy, which accounts for the fluid flow of incompressible materials, i.e.

$$\begin{aligned} &\frac{\partial \boldsymbol{u}}{\partial t} + (\boldsymbol{u}\cdot\nabla)\boldsymbol{u} - \mu\Delta\boldsymbol{u} + \nabla p = \boldsymbol{\eta}, \\ &\operatorname{div} \boldsymbol{u} = 0, \end{aligned} \tag{5}$$

where we additionally added the second order term multiplied by the viscosity μ. The variable $\boldsymbol{\eta}$ 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) showing a double-well form as is the case often in applications, e.g. [51].

Suppose that $\Omega \subset \mathbb{R}^d$, 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

$$E(\boldsymbol{x}) := \int_0^T \int_\Omega e(\boldsymbol{x}(\boldsymbol{X}, t), t)\, \mathrm{d}\boldsymbol{X}\, \mathrm{d}t. \tag{6}$$

The energy (6) combines an action functional for the flow map $\boldsymbol{x}(\boldsymbol{X}, t)$ and a free energy for the order parameter ϕ. This combination of mechanical and thermodynamic energies seems to go back to [14, 15, 21, 22, 24]. Subsequently, we will focus on quasi-stationary, i.e., $\boldsymbol{u}_t = \boldsymbol{0}$ and $\boldsymbol{\eta} \neq \boldsymbol{0}$, and low-Reynolds number flows such that we can neglect the nonlinear term $(\boldsymbol{u}\cdot\nabla)\boldsymbol{u}$. Then, classical ideas from the calculus of variations [47] and the theory of gradient flows

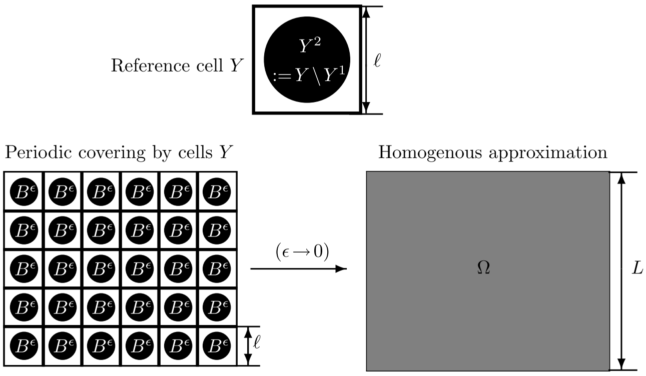

Figure 1. Left: porous medium $\Omega^\epsilon := \Omega \setminus B^\epsilon$ as a periodic covering of reference cells $Y := [0, \ell]^d$. Top: definition of the reference cell $Y = Y^1 \cup Y^2$ with $\ell = 1$. Right: the 'homogenization limit' $\epsilon := (\ell/L) \to 0$ scales the perforated domain such that perforations become invisible in the macroscale.

together with an imposed wetting boundary condition $\int_{\partial\Omega} g(\boldsymbol{x})\, \mathrm{d}o(\boldsymbol{x})$ for $g(\boldsymbol{x}) \in H^{3/2}(\partial\Omega)$ lead to the following set of equations:

$$
\text{(Homogeneous case)} \begin{cases} -\mu\Delta\boldsymbol{u} + \nabla p = \boldsymbol{\eta} & \text{in } \Omega_T, \\ \operatorname{div}\boldsymbol{u} = 0 & \text{in } \Omega_T, \\ \boldsymbol{u} = \boldsymbol{0} & \text{on } \partial\Omega_T, \\ \dfrac{\partial\phi}{\partial t} + \mathrm{Pe}(\boldsymbol{u}\cdot\nabla)\phi = \lambda\operatorname{div}(\nabla(f(\phi) - \Delta\phi)) & \text{in } \Omega_T, \\ \nabla_n\phi := \boldsymbol{n}\cdot\nabla\phi = g(\boldsymbol{x}) & \text{on } \partial\Omega_T, \\ \nabla_n\Delta\phi = 0 & \text{on } \partial\Omega_T, \\ \phi(\boldsymbol{x}, 0) = h(\boldsymbol{x}) & \text{on } \Omega, \end{cases} \tag{7}
$$

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 $\boldsymbol{\eta}$ accounts for the elastic energy [22]

$$
\boldsymbol{\eta} = -\gamma\operatorname{div}(\nabla\phi \otimes \nabla\phi), \tag{8}
$$

where γ corresponds to the surface tension [23]. As in [1], we will set $\gamma = \lambda$ for simplicity. The dimensionless parameter $\mathrm{Pe} := k\tau L\mathrm{U}/D$ is the Péclet number for a reference fluid velocity $\mathrm{U} := |\boldsymbol{u}|$, L is the characteristic length of the porous medium, and the diffusion constant $D = k\tau M$ obtained from the mobility via Einstein's relation for the temperature τ and the Bolzmann constant k. We note that the immiscible flow equations can immediately be written for the full incompressible Navier–Stokes equations as in [22]. Our restriction to the Stokes equation is motivated here by the fact that such flows turn into Darcy's law in porous media [11, 18].

The main objective of our study is the derivation of effective macroscopic equations describing (7) 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 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 $\epsilon = \ell/L$ 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 $Y := [0, \ell_1] \times [0, \ell_2] \times \ldots \times [0, \ell_d]$, $\ell_i \in \mathbb{R}$, $i = 1, \ldots, d$, which represents a single, characteristic pore. The periodicity assumption allows, by passing to the limit $\epsilon \to 0$ (see figure 1) for the derivation effective macroscopic porous media equations. The pore and the solid phase of the medium are defined as usual by,

$$\Omega^\epsilon := \bigcup_{z\in\mathbb{Z}^d} \epsilon\big(Y^1 + z\big) \cap \Omega, \qquad B^\epsilon := \bigcup_{z\in\mathbb{Z}^d} \epsilon\big(Y^2 + z\big) \cap \Omega = \Omega \setminus \Omega^\epsilon, \tag{9}$$

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 (7) by the following microscopic formulation:

$$\text{(Porous case)}\ \begin{cases} -\epsilon^2\mu\Delta \boldsymbol{u}_\epsilon + \nabla p_\epsilon = -\gamma \operatorname{div}(\nabla\phi_\epsilon \otimes \nabla\phi_\epsilon) & \text{in } \Omega^\epsilon_T, \\ \operatorname{div} \boldsymbol{u}_\epsilon = 0 & \text{in } \Omega^\epsilon_T, \\ \boldsymbol{u}_\epsilon = \boldsymbol{0} & \text{on } I^\epsilon_T, \\ \dfrac{\partial}{\partial t}\phi_\epsilon + \mathrm{Pe}(\boldsymbol{u}_\epsilon\cdot\nabla)\phi_\epsilon = \lambda \operatorname{div}(\nabla(f(\phi_\epsilon) - \Delta\phi_\epsilon)) & \text{in } \Omega^\epsilon_T, \\ \nabla_n\phi_\epsilon := \boldsymbol{n}\cdot\nabla\phi_\epsilon = g_\epsilon(\boldsymbol{x}) := g(\boldsymbol{x}/\epsilon) & \text{on } I^\epsilon_T, \\ \nabla_n\Delta\phi_\epsilon = 0 & \text{on } I^\epsilon_T, \\ \phi_\epsilon(\cdot,0) = \psi(\cdot) & \text{on } \Omega^\epsilon, \end{cases} \tag{10}$$

where $I^\epsilon_T := I^\epsilon \times]0, T[$. $g_\epsilon(\boldsymbol{x}) = g(\boldsymbol{x}/\epsilon)$ is now a periodic wetting boundary condition accounting for the wetting properties of the pore walls. Even under the assumption of periodicity, the microscopic system (10) leads to a high-dimensional problem, 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. The homogenization method provides a systematic tool for reducing the intrinsic dimensional complexity by reliably averaging over the microscale represented by a single periodic reference pore Y. We note that the nonlinear nature of problem (10) exploits a scale separation with respect to the upscaled chemical potential, see definition 3.1, for the derivation of the effective macroscopic interfacial evolution in strongly heterogeneous environments. Such a scale separation turns out to be the key for the upscaling/homogenization of nonlinear problems, see [39–41].

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 (10) further by restricting (10) to periodic fluid flow. By taking the stationary version of equation $(10)_4$ on to a single periodic reference pore Y and by denoting the according stationary solution by $\Phi(\cdot)$, we can formulate the following periodic flow problem:

$$\text{(Periodic flow)}\ \begin{cases} -\mu\Delta_y \boldsymbol{u} + \nabla_y p = \boldsymbol{\eta} & \text{in } Y^1, \\ \operatorname{div}_y \boldsymbol{u} = 0 & \text{in } Y^1, \\ \boldsymbol{u} = \boldsymbol{0} & \text{on } \partial Y^2, \\ \boldsymbol{u} \text{ is Y}^1\text{-periodic}, & \\ \mathrm{Pe}(\boldsymbol{u}\cdot\nabla_y)\Phi = \lambda \operatorname{div}_y\big(\nabla_y(f(\Phi) - \Delta_y\Phi)\big) & \text{in} Y^1, \\ \nabla_n\Phi := (\boldsymbol{n}\cdot\nabla_y)\Phi = g(\boldsymbol{y}) & \text{on } \partial Y^2, \\ \nabla_n\Delta_y\Phi = 0 & \text{on } \partial Y^2, \\ \psi \text{ is Y}^1\text{-periodic}. & \end{cases} \tag{11}$$

We remark that in certain occasions it might be suitable to further reduce problem (11). For instance, in general the reference cell is only filled by one fluid phase, i.e. $\nabla_y\Phi = 0$ almost everywhere in Y^1, and hence one only needs to solve for the periodic Stokes problem $(11)_1$–$(11)_4$ by replacing the self-induced driving force (8) with the constant driving force $\boldsymbol{\eta} := \boldsymbol{e}_1$ where $\boldsymbol{e}_1$ denotes the canonical basis vector in the $\boldsymbol{x}_1$-direction of the Euclidean space. The periodic fluid velocity defined by (11) for such an $\boldsymbol{\eta}$ can be considered as the spatially periodic velocity of a moving frame [3]. Motivated by [3, 28, 37], we study the case of large Péclet number and consider the following distinguished case:

Assumption (LP). *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 (LP). If one introduces the microscopic Péclet number $\mathrm{Pe}_{\mathrm{mic}} := k\tau\ell \mathrm{U}/D$, then it follows immediately that $\mathrm{Pe} = \mathrm{Pe}_{\mathrm{mic}}/\epsilon$. Since we introduced a periodic flow problem on the characteristic length scale $\ell > 0$ of the pores by problem (11), it is obvious that we have to apply the microscopic Péclet number in a corresponding microscopic formulation, see (12). Hence, the periodic fluid velocity $\boldsymbol{u}(\boldsymbol{x}/\epsilon) := \boldsymbol{u}(\boldsymbol{y})$ enters the microscopic phase field problem as follows:

$$
\text{(Microscopic problem)} \begin{cases} \dfrac{\partial}{\partial t}\phi_\epsilon + \dfrac{\mathrm{Pe}_{\mathrm{mic}}}{\epsilon}(\boldsymbol{u}(\boldsymbol{x}/\epsilon)\cdot\nabla)\phi_\epsilon & \\ \quad = \lambda \mathrm{div}(\nabla(f(\phi_\epsilon) - \Delta\phi_\epsilon)) & \text{in } \Omega_T^\epsilon, \\ \nabla_n\phi_\epsilon := \boldsymbol{n}\cdot\nabla\phi_\epsilon = g(\boldsymbol{x}/\epsilon) & \text{on } I_T^\epsilon, \\ \nabla_n\Delta\phi_\epsilon = 0 & \text{on } I_T^\epsilon, \\ \phi_\epsilon(\cdot, 0) = \psi(\cdot) & \text{on } \Omega^\epsilon. \end{cases} \tag{12}
$$

We note that with our subsequently applied upscaling strategy, we do not account for boundary layers in the vicinity of rigid boundaries. Such boundary layers become increasingly important in the case of large Péclet numbers. Moreover, we make use of the splitting strategy introduced in [42] and here extended to fluid flow, i.e.

$$
\text{(Splitting)} \begin{cases} \dfrac{\partial}{\partial t}(-\Delta_\epsilon)^{-1}w_\epsilon + \dfrac{\mathrm{Pe}_{\mathrm{mic}}}{\epsilon}(\boldsymbol{u}(\boldsymbol{x}/\epsilon)\cdot\nabla)(-\Delta_\epsilon)^{-1}w_\epsilon & \\ \quad = \lambda\Big(\mathrm{div}\Big(\hat{\mathrm{M}}\nabla w_\epsilon\Big) + \mathrm{div}\Big(\hat{\mathrm{M}}\nabla f(\phi_\epsilon)\Big)\Big) & \text{in } \Omega_T^\epsilon, \\ -\nabla_n\Delta\phi_\epsilon = \nabla_n w_\epsilon = 0 & \text{on } I_T^\epsilon, \\ -\Delta_\epsilon\phi_\epsilon = w_\epsilon & \\ \nabla_n\phi_\epsilon = g(\boldsymbol{x}/\epsilon) = g_\epsilon(\boldsymbol{x}) & \text{on } I_T^\epsilon, \\ \phi_\epsilon(\cdot, 0) = \psi(\cdot) & \text{on } \Omega^\epsilon, \end{cases} \tag{13}
$$

where we will properly define $\Delta_\epsilon = \mathcal{A}_\epsilon$ in section 4.

The main result of our study is the systematic derivation of upscaled immiscible flow equations which effectively account for the pore geometry starting from the microscopic system (11)–(12), i.e.

$$
\text{(Upscaled equation)} \begin{cases} p\dfrac{\partial\phi_0}{\partial t} - \mathrm{div}\Big(\hat{\mathrm{C}}\nabla\phi_0\Big) = \lambda\mathrm{div}\Big(\hat{\mathrm{M}}_\phi\nabla f(\phi_0)\Big) \\ \quad -\dfrac{\lambda}{p}\mathrm{div}\Big(\hat{\mathrm{M}}_w\nabla\Big(\mathrm{div}\Big(\hat{\mathrm{D}}\nabla\phi_0\Big) - \tilde{g}_0\Big)\Big), \end{cases} \tag{14}
$$

where $\hat{\mathrm{C}}$ takes the fluid convection into account. This tensor account for the so-called diffusion–dispersion relations (e.g. Taylor–Aris-dispersion [4, 10, 48]). The result (14) makes use of the recently proposed splitting strategy for the homogenization of fourth-order problems in [42] and an asymptotic multiscale expansion with drift (i.e. moving frame) introduced in [3, 27].

The manuscript is organized as follows. We present our main results in section 3 and the corresponding proofs follow in the subsequent section 4. Concluding remarks and open questions are offered in section 6.

2. Preliminaries and notation

We recall basic results required for our subsequent analysis which depends also on certain notational conventions. We consider connected macroscopic domains Ω with Lipschitz continuous boundaries $\partial\Omega$. Under the usual conventions for Sobolev spaces, we say that $u \in W^{k,p}(\Omega)$ if and only if

$$\|u\|_{k,p}{}^p := \sum_{|\alpha|\leqslant k} \|D^\alpha u\|_{L^p}{}^p < \infty \tag{15}$$

for a multi-index α such that $D^\alpha := \frac{\partial^{|\alpha|}}{\partial^{\alpha_1}x_1 \ldots \partial^{\alpha_d}x_d}$ and $p < \infty$. Herewith, we can identify corresponding Hilbert spaces ($p = 2$) by $H^k(\Omega) := W^{k,2}(\Omega)$. We introduce the following (energy) space of functions:

$$H^2_E(\Omega) := \left\{ v \in H^2(\Omega) \,\middle|\, \nabla_n v = 0 \quad \text{on} \quad \partial\Omega \right\}, \tag{16}$$

which naturally appears in the context of weak solutions for the phase field equations $(7)_4$. In order to account for the periodic reference cells appearing due to asymptotic multiscale-expansions/homogenization, we define $\overline{H}^1_{\text{per}}(Y)$ as the closure of $C^\infty_{\text{per}}(Y)$ in the H^1-norm where $C^\infty_{\text{per}}(Y)$ is the subset of Y-periodic functions of $C^\infty(\mathbb{R}^d)$. As we need uniqueness of solutions, we will work with the following space of functions:

$$H^1_{\text{per}}(Y) := \left\{ u \in \overline{H}^1_{\text{per}}(Y) \,\middle|\, \mathcal{M}_Y(u) = 0 \right\}, \tag{17}$$

where $\mathcal{M}_Y(u) := (1/|Y|) \int_Y u \, \mathrm{d}\boldsymbol{y}$. In order to establish the existence and uniqueness of weak solutions of the upscaled convective phase field equations, we need the following Aubin–Lions compactness result (e.g. [44]), i.e.

Theorem 2.1 (Aubin–Lions). *Let X_0, X, X_1 be Banach spaces with $X_0 \subset X \subset X_1$ and assume that $X_0 \hookrightarrow X$ is compact and $X \hookrightarrow X_1$ is continuous. Let $1 < p < \infty$, $1 < q < \infty$ and let X_0 and X_1 be reflexive. Then, for $W := \{u \in L^p(0, T; X_0) \,|\, \partial_t u \in L^q(0, T; X_1)\}$ the inclusion $W \hookrightarrow L^p(0, T; X)$ is compact.*

3. Results: effective immiscible flow equations in porous media

The presentation of our main result depends on the following:

Definition 3.1 (Scale separation). *We say that the the macroscopic chemical potential is scale separated if and only if the upscaled chemical potential*

$$\mu_0 := f(\phi) - \Delta\phi, \tag{18}$$

satisfies $(\partial\mu_0/\partial x_l) = 0$ for each $1 \leqslant l \leqslant d$ on the level of the reference cell Y but not in the macroscopic domain Ω.

Remark 3.2. We note that the scale separation in definition 3.1 follows intuitively from the key requirement in homogenization theory that one can identify a slow (macroscopic) variable and at least one fast (microscopic) variable. Hence, the above scale separation means that the macroscopic variable does not vary over the dimension of the microscale defined by a characteristic reference cell.

The scale separation (3.1) emerges as a key requirement for the homogenization of nonlinearly coupled partial differential equations in order to guarantee the mathematical well-posedness of the corresponding cell problems which define effective transport coefficients in homogenized, nonlinear (and coupled) problems [39–41].

We note that the upscaling requires to identify effective macroscopic boundary conditions on the macroscopic domain Ω. Such a condition will be denoted by $\tilde{h}_0$ below. In fact, $\tilde{h}_0$ can be computed as $\tilde{g}_0$. We summarize our main result in the following

Theorem 3.3 (Effective convective Cahn–Hilliard equation). *We assume that the assumption (LP) holds and that the macroscopic chemical potential μ_0 satisfies the scale separation property in the sense of definition 3.1 and let $\psi(\boldsymbol{x}) \in H^2(\Omega)$.*

Then, the microscopic equations (11)–(12) for immiscible flow in porous media admit the following effective macroscopic form after averaging over the microscale, i.e.

$$\begin{cases} p\dfrac{\partial \phi_0}{\partial t} - \mathrm{div}\Big(\hat{\mathrm{C}}\nabla\phi_0\Big) = \lambda \mathrm{div}\Big(\hat{\mathrm{M}}_\phi \nabla f(\phi_0)\Big) & \\ -\dfrac{\lambda}{p}\mathrm{div}\Big(\hat{\mathrm{M}}_w\nabla\Big(\mathrm{div}\Big(\hat{\mathrm{D}}\nabla\phi_0\Big) - \tilde{g}_0\Big)\Big) & \text{in } \Omega_T, \\ \nabla_n\phi_0 := \boldsymbol{n}\cdot\nabla\phi_0 = \tilde{h}_0(\boldsymbol{x}) & \text{on } \partial\Omega\times]0,T[, \\ \nabla_n\Delta\phi_0 = 0 & \text{on } \partial\Omega\times]0,T[, \\ \phi_0(\cdot,0) = \psi(\cdot) & \text{in } \Omega, \end{cases} \tag{19}$$

where the tensors $\hat{\mathrm{C}} := \{\mathrm{c}_{ik}\}_{1\leqslant i,k\leqslant d}$, $\hat{\mathrm{D}} := \{\mathrm{d}_{ik}\}_{1\leqslant i,k\leqslant d}$, $\hat{\mathrm{M}}_\phi = \{\mathrm{m}^\phi_{ik}\}_{1\leqslant i,k\leqslant d}$, and $\hat{\mathrm{M}}_w = \{\mathrm{m}^w_{ik}\}_{1\leqslant i,k\leqslant d}$ are defined by

$$\begin{aligned} \mathrm{c}_{ik} &:= \frac{\mathrm{Pe_{mic}}}{|Y|}\int_{Y^1}(\mathrm{u}^i - \mathrm{v}^i)\delta_{ik}\xi^k_\phi\,\mathrm{d}\boldsymbol{y}, \\ \mathrm{d}_{ik} &:= \frac{1}{|Y|}\sum_{j=1}^d\int_{Y^1}\left(\delta_{ik} - \delta_{ij}\frac{\partial\xi^k_\phi}{\partial y_j}\right)\mathrm{d}\boldsymbol{y}, \\ \mathrm{m}^\phi_{ik} &:= \frac{1}{|Y|}\sum_{j=1}^d\int_{Y^1}\left(\mathrm{m}_{ik} - \mathrm{m}_{ij}\frac{\partial\xi^k_\phi}{\partial y_j}\right)\mathrm{d}\boldsymbol{y}, \\ \mathrm{m}^w_{ik} &:= \frac{1}{|Y|}\sum_{j=1}^d\int_{Y^1}\left(\mathrm{m}_{ik} - \mathrm{m}_{ij}\frac{\partial\xi^k_w}{\partial y_j}\right)\mathrm{d}\boldsymbol{y}. \end{aligned} \tag{20}$$

The effective fluid velocity $\boldsymbol{v}$ is defined by $\mathrm{v}^j := (\mathrm{Pe_{mic}}/|Y^1|)\int_{Y^1}\mathrm{u}^j(\boldsymbol{y})\,\mathrm{d}\boldsymbol{y}$ where $\boldsymbol{u}$ solves the periodic reference cell problem (11). The effective wetting boundary condition on the pore walls becomes $\tilde{g}_0 := -(\gamma/C_h)(1/|Y|)\int_{\partial Y^1}(a_1\chi_{\partial Y^1_w}(\boldsymbol{y}) + a_2\chi_{\partial Y^2_w}(\boldsymbol{y}))\,\mathrm{d}\boldsymbol{y}$ and on the boundary $\partial\Omega$ of the macroscopic domain Ω we impose $\tilde{h}_0 := -(\gamma/C_h)(1/|Y|)\int_\Gamma(a_\Gamma(\boldsymbol{y}))\,\mathrm{d}\boldsymbol{y}$. The corrector functions $\xi^k_\phi \in H^1_{\mathrm{per}}(Y^1)$ and $\xi^k_w \in L^2(\Omega; H^1_{\mathrm{per}}(Y^1))$ for $1\leqslant k,l\leqslant d$ solve in the distributional sense the following reference cell problems:

$$\xi^k_\phi : \begin{cases} -\sum_{i,j=1}^d \dfrac{\partial}{\partial y_i}\left(\delta_{ik} - \delta_{ij}\dfrac{\partial\xi^k_\phi}{\partial y_j}\right) = 0 & \text{in } Y^1, \\ \sum_{i,j=1}^d \mathrm{n}_i\left(\delta_{ij}\frac{\partial\xi^k_\phi}{\partial y_j} - \delta_{ik}\right) = 0 & \text{on } \partial Y^1, \\ \xi^k_\phi(\boldsymbol{y}) \text{ is } Y\text{-periodic and } \mathcal{M}_{Y^1}(\xi^k_\phi) = 0, & \end{cases} \tag{21}$$

$$\xi_w^k : \begin{cases} -\sum_{i,j=1}^d \frac{\partial}{\partial y_i}\left(\delta_{ik} - \delta_{ij}\frac{\partial \xi_w^k}{\partial y_j}\right) \\ = \lambda\left(\sum_{i,j=1}^d \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ik} - \mathrm{m}_{ij}\frac{\partial \xi_\phi^k}{\partial y_j}\right)\right. & \text{in } Y^1, \\ \sum_{i,j=1}^d \mathrm{n}_i\left(\left(\delta_{ij}\frac{\partial \xi_w^k}{\partial y_j} - \delta_{ik}\right)\right. \\ -\lambda\sum_{i,j=1}^d \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ik} - \mathrm{m}_{ij}\frac{\partial \xi_\phi^k}{\partial y_j}\right) = 0 & \text{on } \partial Y^1, \\ \xi_w^k(\boldsymbol{y}) \text{ is } Y\text{-periodic and } \mathcal{M}_{Y^1}(\xi_w^k) = 0. \end{cases} \tag{22}$$

Remark 3.4 (Isotropic mobility). The cell problem (22) is equal to problem (21) if we consider the case of isotropic mobility tensors, i.e. $\hat{\mathrm{M}} := m\hat{\mathrm{I}}$. In this special case, we immediately have $\xi_\phi^k = \xi_w^k$ and hence the porous media correction tensors satisfy $m\hat{\mathrm{D}} = \hat{\mathrm{M}}_\phi = \hat{\mathrm{M}}_w$.

The next theorem guarantees the well-posedness of (23) in the sense of week solutions. For convenience, we achieve existence of weak solutions for polynomial free energies in the sense of assumption (PF) [49].

Theorem 3.5 (Existence and Uniqueness). *Let $\psi \in L^2(\Omega)$, $T^* > 0$, and assume that the admissible free energy densities F in (1) satisfy assumption (PF). Then, there exists a unique solution $\phi_0 \in L^\infty(]0, T^*[; L^2(\Omega)) \cap L^2([0, T^*[; H_E^2(\Omega))$ to the following upscaled/homogenized problem:*

$$\begin{cases} p\frac{\partial \phi_0}{\partial t} + \frac{\lambda}{p}\mathrm{div}\left(\hat{\mathrm{M}}_w \nabla\left\{\mathrm{div}\left(\hat{\mathrm{D}}\nabla\phi_0\right) - \tilde{g}_0\right\}\right) \\ \quad = \mathrm{div}\left(\left[\lambda f'(\phi_0)\hat{\mathrm{M}}_\phi + \hat{\mathrm{C}}\right]\nabla\phi_0\right) & \text{in } \Omega_T, \\ \nabla_n \phi_0 = 0 & \text{on } \partial\Omega \times]0, T^*[, \\ \nabla_n \Delta\phi_0 = 0 & \text{on } \partial\Omega \times]0, T^*[, \\ \phi_0(\cdot, 0) = \psi(\cdot) & \text{in } \Omega. \end{cases} \tag{23}$$

We prove theorem 3.5 by adapting arguments from [49, section 2, p 151] to the homogenized setting including the diffusion–dispersion tensor which accounts for periodic flow.

3.1. Numerical computations

To exemplify the results presented above we perform a numerical study of the effective macroscopic Cahn–Hilliard equation (19). We consider a two-dimensional (2D) porous medium consisting of a series of periodic reference cells the geometrical shape of which is a non-straight channel of constant cross-section (see figure 2) with periodic boundary conditions at the inlet and outlet. The Cartesian coordinates in the microscopic problem are named as x and y which correspond to the y_1 and y_2 variables used in the definition of the domain Y^1, respectively. We define the geometry in such a way that the porosity of the medium is $p = 0.46$. The macroscopic domain Ω is compound of 35 reference cells in the perpendicular direction y of the flow and 50 in the x direction. We fix a constant driving force $U = 1$ at the inlet of the system by fixing the gradient of the chemical potential [51]. For simplicity we take the macroscopic mobility $M = 1$ which gives rise to a microscopic Péclet number $\mathrm{Pe}_{\mathrm{mic}} = 0.04$.

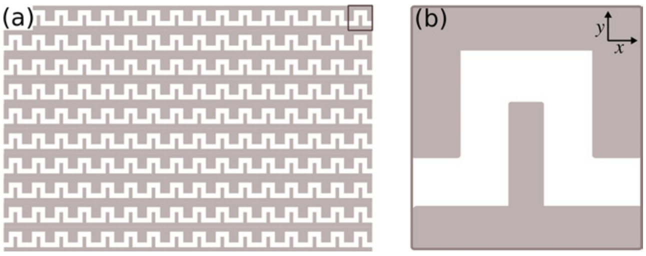

Figure 2. (*a*) Example of the 2D porous medium considered for the numerical computations. The reference cell consists of a non-straight channel of constant cross-section which is periodic in the x direction as depicted in (*b*). Grey area corresponds to the solid phase of the medium.

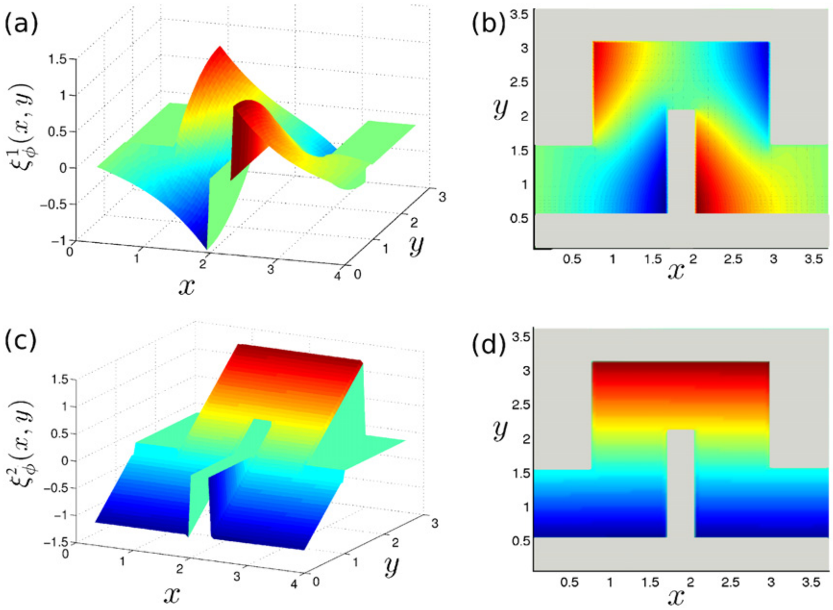

Figure 3. Corrector functions $\xi_\phi^1(x, y)$ (*a*), (*b*) and $\xi_\phi^2(x, y)$ (*c*), (*d*) for the particular reference cell defined in figure 2(*b*). Panels (*a*) and (*c*) show the three-dimensional plots, and panels (*b*) and (*d*) show the corresponding 2D projection onto the plane (x, y).

We first compute the components of the different tensors $\hat{\mathrm{C}}$, $\hat{\mathrm{D}}$, $\hat{\mathrm{M}}_\phi$, and $\hat{\mathrm{M}}_w$ for which we need to solve the reference cell problems (21) and (22). We consider the case of isotropic mobility with $m = 1$ and hence we have $\xi_\phi^k = \xi_w^k$ and $\hat{\mathrm{D}} = \hat{\mathrm{M}}_\phi = \hat{\mathrm{M}}_w$. In this case, the reference cell problem is reduced to:

$$\xi_\phi^k : \quad \begin{cases} \left(\dfrac{\partial^2}{\partial x^2} + \dfrac{\partial^2}{\partial y^2}\right)\xi_\phi^k = 0 & \text{in } Y^1, \\ \left(\mathrm{n}_1 \dfrac{\partial}{\partial x} + \mathrm{n}_2 \dfrac{\partial}{\partial y}\right)\xi_\phi^k = \mathrm{n}_k & \text{on } \partial Y^1, \end{cases} \tag{24}$$

which corresponds to the Laplace equation with special boundary conditions. The above equation is solved by using a finite differences numerical scheme and the resulting corrector functions $\xi_\phi^1(x, y)$ and $\xi_\phi^2(x, y)$ are plotted in figure 3.

Once we know the corrector functions of the reference cell problem, we can compute the different elements of the tensor $\hat{\mathrm{D}}$ as defined in (20) obtaining the values $\mathrm{d}_{11} = 0.4$, and

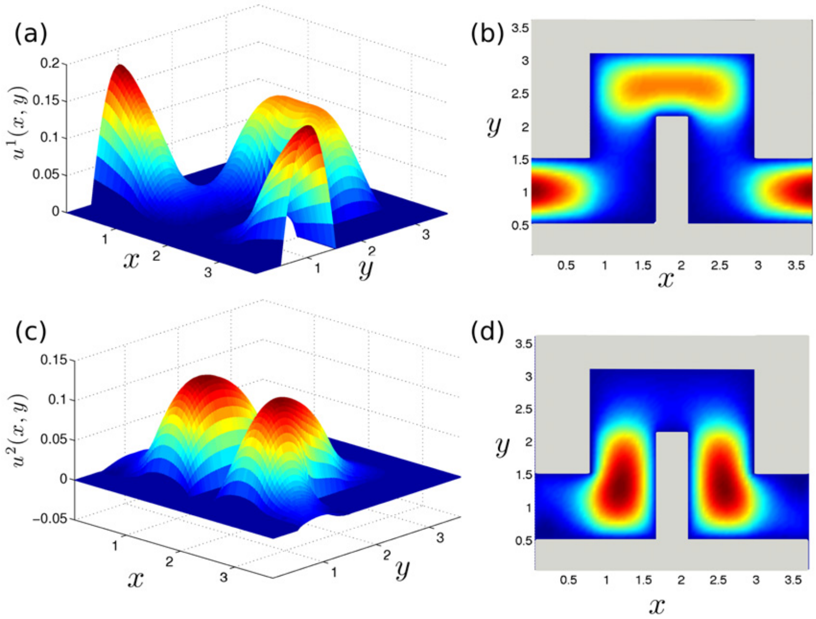

Figure 4. Two components of the velocity field $u^1(x, y)$ (*a*), (*b*) and $u^2(x, y)$ (*c*), (*d*). Panels (*a*) and (*c*) show the three-dimensional plots, and panels (*b*) and (*d*) show the corresponding 2D projection onto the plane (x, y).

$d_{12} = d_{21} = d_{22} = 0$. Note that these are similar values to those reported in [6] for a similar porous geometry. Next we solve the Stokes flow for this particular microscopic geometry by numerically integrating the periodic reference cell problem (11) to obtain the velocity $\boldsymbol{u}$ and hence the coefficients for the tensor $\hat{C}$. The results for the two velocity components $u^1(x, y)$ and $u^2(x, y)$ are presented in figure 4. By applying the formula given in (20) we obtain the coefficients $c_{11} = 0.015$ and $c_{22} = 0.023$. Note that by definition $c_{12} = c_{21} = 0$.

Finally, with all the different tensor coefficients we can numerically integrate the problem (19) in the macroscopic domain Ω, the Cartesian coordinates of which are denoted as (X, Y). We use a finite difference scheme for the spatial discretization and a fourth-order Runge–Kutta algorithm with adaptive time stepsize to march froward in time. The domain is discretized with a grid spacing $\Delta X = 0.01$ and we impose periodic boundary conditions along the transversal direction of the flow. As an initial condition, we consider a small sinusoidal shape for the interface separating the liquid from the gas phase. Also, to simulate the same condition as in the porous medium, we impose the driving force U to be fixed alternately at the inlet of the system in such a way it follows the periodicity of the microstructure. The evolution of the interface position is then found by setting $\phi_0(X, Y, t) = 0$. The results are presented in figure 5 where we observe that the profile of the interface evolves into a well defined spatial periodic shape which corresponds to the periodic porous medium that is defined at the microscopic level (had the macroscopic model ignored the microscopic details, by e.g. taking the tensors to be identity matrices, the interface would be flat at all times). For large times and after the influence of the initial disturbances dies out, the interface approaches a steady travelling front with a microstructure that reflects the porous medium structure (as expected). Our results hence show that the effective macroscopic equation is able to retain the microscopic details even though we do not resolve them numerically.

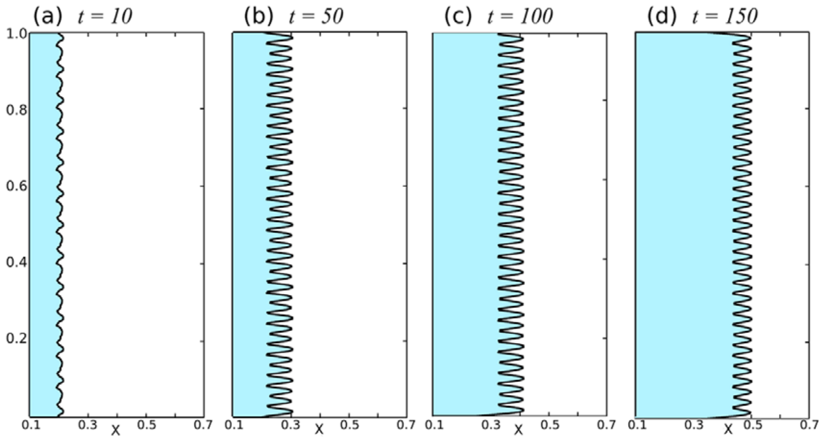

Figure 5. Numerical integrations of the effective macroscopic Cahn–Hilliard equation at different times. Blue colour represents the liquid phase and the interface position is depicted as a solid black line.

4. Formal derivation of theorem 3.3

As in [42], we introduce the differential operators

$$
\begin{aligned}
\mathcal{A}_0 &= -\textstyle\sum_{i,j=1}^{d} \dfrac{\partial}{\partial y_i}\left(\delta_{ij}\dfrac{\partial}{\partial y_j}\right), &\qquad \mathcal{B}_0 &= -\textstyle\sum_{i,j=1}^{d} \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ij}\dfrac{\partial}{\partial y_j}\right),\\
\mathcal{A}_1 &= -\textstyle\sum_{i,j=1}^{d}\Bigg[\frac{\partial}{\partial x_i}\left(\delta_{ij}\dfrac{\partial}{\partial y_j}\right) & \mathcal{B}_1 &= -\textstyle\sum_{i,j=1}^{d}\Bigg[\dfrac{\partial}{\partial x_i}\left(\mathrm{m}_{ij}\dfrac{\partial}{\partial y_j}\right)\\
&\quad+\dfrac{\partial}{\partial y_i}\left(\delta_{ij}\dfrac{\partial}{\partial x_j}\right)\Bigg], & &\quad+\dfrac{\partial}{\partial y_i}\left(\mathrm{m}_{ij}\dfrac{\partial}{\partial x_j}\right)\Bigg],\\
\mathcal{A}_2 &= -\textstyle\sum_{i,j=1}^{d} \dfrac{\partial}{\partial x_j}\left(\delta_{ij}\dfrac{\partial}{\partial x_j}\right), & \mathcal{B}_2 &= -\textstyle\sum_{i,j=1}^{d} \dfrac{\partial}{\partial x_j}\left(\mathrm{m}_{ij}\dfrac{\partial}{\partial x_j}\right),
\end{aligned}
\tag{25}
$$

which make use of the microscale $(\boldsymbol{x}/\epsilon) =: \boldsymbol{y} \in Y$ 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 $\mathrm{div}(\hat{\mathrm{M}}\nabla)$ become $\Delta u^\epsilon(\boldsymbol{x}) = \mathcal{A}_\epsilon u(\boldsymbol{x}, \boldsymbol{y})$ and $\mathrm{div}(\hat{\mathrm{M}}\nabla)u^\epsilon(\boldsymbol{x}) = \mathcal{B}_\epsilon u(\boldsymbol{x}, \boldsymbol{y})$, respectively, where $u^\epsilon(\boldsymbol{x}, t) := u(\boldsymbol{x} - \frac{\boldsymbol{v}}{\epsilon}t, \boldsymbol{y}, t)$. Due to the drift [3, 27], we additionally have

$$
\frac{\partial}{\partial t}u^\epsilon = \left(\frac{\partial}{\partial t} - \frac{\boldsymbol{v}\cdot\nabla_{\boldsymbol{x}}}{\epsilon}\right)u^\epsilon,
\tag{26}
$$

where we find below by the Fredholm alternative (or a solvability constraint) that $\boldsymbol{v} := \frac{\mathrm{Pe}_{\mathrm{loc}}}{|Y^1|}\int_{Y^1} \mathrm{u}^j(y)\,\mathrm{d}y$. As in [42] we apply the method of formal asymptotic multiscale expansions, that is,

$$
\begin{aligned}
w^\epsilon &:= w_0(\boldsymbol{x}, \boldsymbol{y}, t) + \epsilon w_1(\boldsymbol{x}, \boldsymbol{y}, t) + \epsilon^2 w_2(\boldsymbol{x}, \boldsymbol{y}, t) + \cdots,\\
\phi^\epsilon &:= \phi_0(\boldsymbol{x}, \boldsymbol{y}, t) + \epsilon\phi_1(\boldsymbol{x}, \boldsymbol{y}, t) + \epsilon^2\phi_2(\boldsymbol{x}, \boldsymbol{y}, t) + \cdots,
\end{aligned}
\tag{27}
$$

together with the splitting strategy introduced therein. In order to cope with the nonlinear form of the homogeneous free energy $f = F'$, see (2) and (3), we make use of a Taylor expansion which naturally leads to an expansion in ϵ, i.e.

$$
f(\phi^\epsilon) = f(\phi_0) + f'(\phi_0)(\phi^\epsilon - \phi_0) + \tfrac{1}{2}f''(\phi_0)(\phi^\epsilon - \phi_0)^2 + \mathcal{O}((\phi^\epsilon - \phi_0)^3).
\tag{28}
$$

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,

$$\mathcal{O}(\epsilon^{-2}):\quad \begin{cases} \lambda\mathcal{B}_0[w_0+f(\phi_0)]+\mathrm{Pe}_{\mathrm{mic}}(\boldsymbol{u}\cdot\nabla))_{\boldsymbol{y}}\mathcal{A}_2^{-1}w_0=0 & \text{in } Y^1,\\ \text{no flux b.c.,} & \\ w_0 \text{ is } Y^1\text{-periodic,} & \\ \mathcal{A}_0\phi_0=0 & \text{in } Y^1,\\ \nabla_n\phi_0=0 & \text{on } \partial Y_w^1\cap\partial Y_w^2,\\ \phi_0 \text{ is } Y^1\text{-periodic,} & \end{cases} \tag{29}$$

$$\mathcal{O}(\epsilon^{-1}):\quad \begin{cases} \lambda\mathcal{B}_0\big[w_1+f'(\phi_0)\phi_1\big]+\mathrm{Pe}_{\mathrm{mic}}\big(\boldsymbol{u}\cdot\nabla_{\boldsymbol{y}}\big)\mathcal{A}_2^{-1}w_1 & \\ \quad=-\lambda\mathcal{B}_1[w_0+f(\phi_0)] & \\ \quad-\mathrm{Pe}_{\mathrm{mic}}((\boldsymbol{u}-\boldsymbol{v})\cdot\nabla)\mathcal{A}_2^{-1}w_0 & \text{in } Y^1,\\ \text{no flux b.c.,} & \\ w_1 \text{ is } Y^1\text{-periodic,} & \\ \mathcal{A}_0\phi_1=-\mathcal{A}_1\phi_0 & \text{in } Y^1,\\ \nabla_n\phi_1=0 & \text{on } \partial Y_w^1\cap\partial Y_w^2,\\ \phi_1 \text{ is } Y^1\text{-periodic,} & \end{cases} \tag{30}$$

$$\mathcal{O}(\epsilon^{0}):\quad \begin{cases} \lambda\mathcal{B}_0\left[w_2+\frac{1}{2}f''(\phi_0)\phi_1^2+f'(\phi_0)\phi_2\right]+\mathrm{Pe}_{\mathrm{mic}}\big(\boldsymbol{u}\cdot\nabla_{\boldsymbol{y}}\big)\mathcal{A}_2^{-1}w_2 & \\ \quad=\lambda(\mathcal{B}_2[w_0+f(\phi_0)]+\mathcal{B}_1[w_1+f(\phi_0)\phi_1]) & \\ \quad-\mathrm{Pe}_{\mathrm{mic}}((\boldsymbol{u}-\boldsymbol{v})\cdot\nabla)\mathcal{A}_2^{-1}w_1 & \\ \quad-\partial_t\mathcal{A}_2^{-1}w_0 & \text{in} Y^1,\\ \text{no flux b.c.,} & \\ w_2 \text{ is } Y^1\text{-periodic,} & \\ \mathcal{A}_0\phi_2=-\mathcal{A}_2\phi_0-\mathcal{A}_1\phi_1+w_0 & \text{in} Y^1,\\ \nabla_n\phi_2=g(\boldsymbol{y}) & \text{on } \partial Y_w^1\cap\partial Y_w^2,\\ \phi_2 \text{ is } Y^1\text{-periodic,} & \end{cases} \tag{31}$$

As usual, the first problem (29) induces that the leading order terms ϕ_0 and w_0 are independent of the microscale $\boldsymbol{y}$. The second problem (30) reads in explicit form for ϕ_1 as follows:

$$\xi_\phi:\quad \begin{cases} -\sum_{i,j=1}^{d}\frac{\partial}{\partial y_i}\left(\delta_{ik}-\delta_{ij}\frac{\partial\xi_\phi^k}{\partial y_j}\right) & \\ \quad=-\mathrm{div}\big(\boldsymbol{e}_k-\nabla_y\xi_\phi^k\big)=0 & \text{in } Y^1,\\ \boldsymbol{n}\cdot\big(\nabla\xi_\phi^k+\boldsymbol{e}_k\big)=0 & \text{on } \partial Y_w^1\cap\partial Y_w^2,\\ \xi_\phi^k(\boldsymbol{y}) \text{ is } Y\text{-periodic and } \mathcal{M}_{Y^1}(\xi_\phi^k)=0, & \end{cases} \tag{32}$$

which represents the reference cell problem for ϕ_0 after identifying $\phi_1 = -\sum_{k=1}^{d}\xi_\phi^k(\boldsymbol{y})(\partial\phi_0/\partial x_k)$.

The cell problem for w_1 is substantially more involved since it depends on the fluid velocity $\boldsymbol{u}$ and the the corrector ξ_ϕ^k from (32). Specifically,

$$-\sum_{k,i,j=1}^{d}\frac{\partial}{\partial y_i}\left(\mathrm{m}_{ij}\left(\frac{\partial x_k}{\partial x_j}-\frac{\partial\xi_w^k}{\partial y_j}\right)\frac{\partial w_0}{\partial x_k}\right)=\sum_{k,i,j=1}^{d}\frac{\partial}{\partial y_i}\left(\mathrm{m}_{ij}\left(\frac{\partial x_k}{\partial x_j}-\frac{\partial\xi_\phi^k}{\partial y_j}\right)\frac{\partial f(\phi_0)}{\partial x_k}\right)$$
$$-\mathrm{Pe}_{\mathrm{mic}}\sum_{i=1}^{d}\big(\mathrm{u}^i-\mathrm{v}^i\big)\frac{\partial\phi_0}{\partial x_i}\qquad\text{in } Y^1, \tag{33}$$

which can be simplified under a scale seperated chemical potential in the sense of definition 3.1, i.e. $(\partial/\partial x_k)f(\phi) = f'(\phi)(\partial\phi/\partial x_k) = (\partial w/\partial x_k)$ for $1 \leqslant k \leqslant d$, to the following cell problem:

$$\begin{cases} -\sum_{i,j,k=1}^{d} \frac{\partial}{\partial y_i}\left(\delta_{ik} - \delta_{ij}\frac{\partial \xi_w^k}{\partial y_j}\right) f'(\phi_0) \\ \quad = \lambda \sum_{k,i,j=1}^{d} \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ik} - \mathrm{m}_{ij}\frac{\partial \xi_\phi^k}{\partial y_j}\right) f'(\phi_0) \\ \quad -\mathrm{Pe}_{\mathrm{mic}} \sum_{i=1}^{d} (\mathrm{u}^i - \mathrm{v}^i) & \text{in } Y^1, \\ \sum_{i,j,k=1}^{d} \mathrm{n}_i \left(\left(\delta_{ij}\frac{\partial \xi_w^k}{\partial y_j} - \delta_{ik}\right) \right. \\ \quad \left. -\lambda \sum_{k,i,j=1}^{d} \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ik} - \mathrm{m}_{ij}\frac{\partial \xi_\phi^k}{\partial y_j}\right) \right) = 0 & \text{on } \partial Y_w^1 \cap \partial Y_w^2, \\ \xi_w^k(\boldsymbol{y}) \text{ is } Y\text{-periodic and } \mathcal{M}_{Y^1}(\xi_w^k) = 0. \end{cases} \tag{34}$$

A solvability constraint (e.g. the Fredholm alternative) immediately turns (34) into the following characterization of ξ_w^k and v^i, i.e.

$$\mathrm{v}^j := \frac{\mathrm{Pe}_{\mathrm{mic}}}{|Y^1|}\int_{Y^1} \mathrm{u}^j(\boldsymbol{y})\,\mathrm{d}\boldsymbol{y}$$

$$\begin{cases} -\sum_{i,j,k=1}^{d} \frac{\partial}{\partial y_i}\left(\delta_{ik} - \delta_{ij}\frac{\partial \xi_w^k}{\partial y_j}\right) = \lambda \sum_{k,i,j=1}^{d} \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ik} - \mathrm{m}_{ij}\frac{\partial \xi_\phi^k}{\partial y_j}\right) & \text{in } Y^1 \\ \sum_{i,j,k=1}^{d} \mathrm{n}_i \left(\left(\delta_{ij}\frac{\partial \xi_w^k}{\partial y_j} - \delta_{ik}\right) - \lambda \sum_{k,i,j=1}^{d} \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ik} - \mathrm{m}_{ij}\frac{\partial \xi_\phi^k}{\partial y_j}\right)\right) = 0 & \text{on } \partial Y_w^1 \cap \partial Y_w^2, \\ \xi_w^k(\boldsymbol{y}) \text{ is } Y\text{-periodic and } \mathcal{M}_{Y^1}(\xi_w^k) = 0. \end{cases} \tag{35}$$

We are then left to study the last problem (31) arising by the asymptotic multiscale expansions. Problem $(31)_2$ for ϕ_2 is classical and leads immediately to the upscaled equation

$$-\Delta_{\hat{\mathrm{D}}}\phi_0 := -\mathrm{div}\left(\hat{\mathrm{D}}\nabla\phi_0\right) = p w_0 + \tilde{g}_0, \tag{36}$$

see also [42], where the porous media correction tensor $\hat{\mathrm{D}} := \{\mathrm{d}_{ik}\}_{1\leqslant i,k\leqslant d}$ is defined by

$$|Y|\mathrm{d}_{ik} := \sum_{j=1}^{d} \int_{Y^1} \left(\delta_{ik} - \delta_{ij}\frac{\partial \xi_\phi^k}{\partial y_j}\right) \mathrm{d}\boldsymbol{y}. \tag{37}$$

Next, we apply the Fredholm alternative on equation $(31)_1$, i.e.,

$$\int_{Y^1} \Big\{ \lambda(\mathcal{B}_2 w_0 + \mathcal{B}_1 w_1) - \lambda\mathcal{B}_1[f(\phi_0)\phi_1] - \lambda\mathcal{B}_2 f(\phi_0) - \partial_t \mathcal{A}_2^{-1} w_0 \\ -\mathrm{Pe}_{\mathrm{mic}}\left(\boldsymbol{u}\cdot\nabla_y\right)\mathcal{A}_2^{-1} w_2 - \mathrm{Pe}_{\mathrm{mic}}((\boldsymbol{u}-\boldsymbol{v})\cdot\nabla)\mathcal{A}_2^{-1} w_1 \Big\}\,\mathrm{d}\boldsymbol{y} = 0. \tag{38}$$

The term multiplied by λ in (38) can immediately be rewritten by

$$\lambda \int_{Y^1} -(\mathcal{B}_2 w_0 + \mathcal{B}_1 w_1)\,\mathrm{d}\boldsymbol{y} = -\lambda\mathrm{div}\left(\hat{\mathrm{M}}_w \nabla w_0\right), \tag{39}$$

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

$$\mathrm{m}_{ik}^w := \frac{1}{|Y|}\sum_{j=1}^{d} \int_{Y^1} \left(\mathrm{m}_{ik} - \mathrm{m}_{ij}\frac{\partial \xi_w^k}{\partial y_j}\right) \mathrm{d}\boldsymbol{y}. \tag{40}$$

The first term on the second line in (38) transform as in [42] to

$$
\begin{aligned}
&-\mathcal{B}_1\left[f'(\phi_0)\phi_1\right] \\
&\quad = -\sum_{i,j=1}^{d}\left[\frac{\partial}{\partial x_i}\left(\mathrm{m}_{ij} f'(\phi_0)\sum_{k=1}^{d}\frac{\partial \xi_\phi^k}{\partial y_j}\frac{\partial \phi_0}{\partial x_k}\right) + \frac{\partial}{\partial y_i}\left(\mathrm{m}_{ij} f'(\phi_0)\sum_{k=1}^{d}\xi_\phi^k\frac{\partial^2 \phi_0}{\partial x_k \partial x_j}\right)\right], \qquad (41)
\end{aligned}
$$

where the last term in (41) disappears after integrating by parts. The first term on the right-hand side of (41) can be rewritten with the help of the chain rule as follows:

$$
-\mathcal{B}_1\left[f'(\phi_0)\phi_1\right] = -\sum_{i,j=1}^{d}\mathrm{m}_{ij}\sum_{k=1}^{d}\frac{\partial \xi_\phi^k}{\partial y_j}\frac{\partial^2 f(\phi_0)}{\partial x_k \partial x_i}. \qquad (42)
$$

After adding to (42) the term $-\mathcal{B}_2 f(\phi_0)$, we can define a tensor $\hat{\mathrm{M}}_\phi = \{\mathrm{m}_{ij}^\phi\}_{1\leqslant i,k\leqslant d}$, i.e.,

$$
\mathrm{m}_{ik}^\phi := \frac{1}{|Y|}\sum_{j=1}^{d}\int_{Y^1}\left(\mathrm{m}_{ik} - \mathrm{m}_{ij}\frac{\partial \xi_\phi^k}{\partial y_j}\right)\mathrm{d}\boldsymbol{y}, \qquad (43)
$$

such that

$$
-\mathcal{B}_1\left[f'(\phi_0)\phi_1\right] - \mathcal{B}_2 f(\phi_0) = \mathrm{div}\left(\hat{\mathrm{M}}_\phi \nabla f(\phi_0)\right). \qquad (44)
$$

The terms in the last line of (38) become

$$
\begin{aligned}
&-\frac{1}{|Y|}\int_{Y^1}\mathrm{Pe}_{\mathrm{mic}}\left(\boldsymbol{u}\cdot\nabla_{\boldsymbol{y}}\right)\mathcal{A}_2^{-1}w_2 + \mathrm{Pe}_{\mathrm{mic}}((\boldsymbol{u}-\boldsymbol{v})\cdot\nabla)\mathcal{A}_2^{-1}w_1\,\mathrm{d}\boldsymbol{y} \\
&\qquad = -\frac{1}{|Y|}\int_{Y^1}\mathrm{Pe}_{\mathrm{mic}}\left(\boldsymbol{u}\cdot\nabla_{\boldsymbol{y}}\right)\phi_2\,\mathrm{d}\boldsymbol{y} \qquad (45) \\
&\qquad\quad + \frac{\mathrm{Pe}_{\mathrm{mic}}}{|Y|}\sum_{k,i=1}^{d}\int_{Y^1}(\mathrm{u}^i - \mathrm{v}^i)\frac{\partial}{\partial x_i}\left(\delta_{ik}\xi_\phi^k(\boldsymbol{y})\frac{\partial}{\partial x_k}\phi_0\right)\mathrm{d}\boldsymbol{y}.
\end{aligned}
$$

Using the fact that $\boldsymbol{u}$ is divergence-free and after defining the tensor $\hat{\mathrm{C}} := \{\mathrm{c}_{ik}\}_{1\leqslant i,k\leqslant d}$ by

$$
\mathrm{c}_{ik} := \frac{\mathrm{Pe}_{\mathrm{mic}}}{|Y|}\int_{Y^1}(\mathrm{u}^i - \mathrm{v}^i)\delta_{ik}\xi_\phi^k(\boldsymbol{y})\,\mathrm{d}\boldsymbol{y}, \qquad (46)
$$

we finally obtain with the previous considerations the following upscaled phase field equation:

$$
\begin{aligned}
p\frac{\partial \mathcal{A}_2^{-1}w_0}{\partial t} &= \mathrm{div}\left(\left[\lambda\hat{\mathrm{M}}_\phi f'(\phi_0) + \hat{\mathrm{C}}\right]\nabla\phi_0\right) \\
&\qquad - \frac{\lambda}{p}\mathrm{div}\left(\hat{\mathrm{M}}_w\nabla\left(\mathrm{div}\left(\hat{\mathrm{D}}\nabla\phi_0\right) - \tilde{g}_0\right)\right). \qquad (47)
\end{aligned}
$$

Using (36) finally leads to the effective macroscopic phase field equation:

$$
p\frac{\partial \phi_0}{\partial t} = \mathrm{div}\left(\left[\lambda\hat{\mathrm{M}}_\phi f'(\phi_0) + \hat{\mathrm{C}}\right]\nabla\phi_0\right) - \frac{\lambda}{p}\mathrm{div}\left(\hat{\mathrm{M}}_w\nabla\left(\mathrm{div}\left(\hat{\mathrm{D}}\nabla\phi_0\right) - \tilde{g}_0\right)\right). \qquad (48)
$$

5. Proof of theorem 3.5

The proof follows in three basic steps: In step 1, we establish *a priori* estimates that provide compactness required for step 2 where we construct a sequence of approximate solutions. In step 3, we pass to the limit in the sequence of approximate solutions which provide then existence and uniqueness of the original problem.

Step 1: (A priori estimates). (i) Basic energy estimate: as we establish the existence and uniqueness of weak solutions, we rewrite (23) in the sense of distributions, i.e. for all $\varphi \in H^2_E$ it holds that

$$\frac{p}{2}\frac{\partial}{\partial t}(\phi_0,\varphi) + \frac{\lambda}{p}\Big(\mathrm{div}\Big(\hat{\mathrm{D}}\nabla\phi_0\Big), \mathrm{div}\Big(\hat{\mathrm{M}}_w\nabla\varphi\Big)\Big) + \frac{\lambda}{p}\Big(\hat{\mathrm{M}}_w\nabla\tilde{g}_0, \nabla\varphi\Big) + \Big(\lambda f'(\phi_0)\hat{\mathrm{M}}_\phi\nabla\phi_0, \nabla\varphi\Big) + \Big(\hat{\mathrm{C}}\nabla\phi_0, \nabla\varphi\Big) = 0. \tag{49}$$

Note that for an isotropic mobility of the form $\hat{\mathrm{M}} = m\hat{\mathrm{I}}$, where $\hat{\mathrm{I}}$ is the identity tensor, the following identity holds $m\hat{\mathrm{D}} = \hat{\mathrm{M}}_w$. For simplicity, we will base our proof on this identity. The general case is then verified along the same lines using properties of symmetric positive definite tensors. Using the notation $\Delta_{\hat{\mathrm{D}}} := \mathrm{div}(\hat{\mathrm{D}}\nabla)$ and the test function $\varphi = \phi_0$ leads to the following inequality:

$$\frac{p}{2}\frac{\mathrm{d}}{\mathrm{d}t}\|\phi_0\|^2 + \frac{\lambda m}{p}\left\|\Delta_{\hat{\mathrm{D}}}\phi_0\right\|^2 + \lambda b_{2r}m_\phi\int_\Omega \phi_0^{2r-2}|\nabla\phi_0|^2\,\mathrm{d}\boldsymbol{x} \leqslant C\|\nabla\phi_0\|^2 + \frac{2\lambda m_w}{p}\|\nabla\tilde{g}_0\|\|\nabla\phi_0\| \leqslant C\|\nabla\phi_0\|^2 + \frac{\lambda}{2p}m_w\|\nabla\tilde{g}_0\|^2, \tag{50}$$

where we applied the existence of a constant $C > 0$ such that $f(s)s \geqslant pb_{2r}s^{2r} - C$, for all $s \in \mathbb{R}$ due to assumption (PF). With the equivalence of norms in $H^2(\Omega)$, i.e. $\|\phi_0\|_{H^2(\Omega)} \leqslant C(\|\Delta\phi_0\| + \|m(\phi_0)\|)$, and interpolation estimates we obtain

$$\|\nabla\phi_0\|^2 \leqslant C\|\phi_0\|\|\phi_0\|_{H^2(\Omega)} \leqslant C\|\phi_0\|(\|\Delta\phi_0\| + \alpha) \leqslant C\|\phi_0\|\Big(\left\|\Delta_{\hat{\mathrm{D}}}\phi_0\right\| + \alpha\Big) \leqslant \frac{\kappa}{2}\left\|\Delta_{\hat{\mathrm{D}}}\phi_0\right\|^2 + \frac{1}{C\kappa}\|\phi_0\|^2 + \frac{\kappa\alpha^2}{2}, \tag{51}$$

where $m(\phi_0) := \frac{1}{|\Omega|}\int_\Omega \phi_0\,\mathrm{d}\boldsymbol{x}$ with $m(\phi_0) \leqslant \alpha$. This leads to

$$\frac{p}{2}\frac{\mathrm{d}}{\mathrm{d}t}\|\phi_0\|^2 + \left(\frac{\lambda m}{p} - \frac{\kappa}{2}\right)\|\Delta\phi_0\|^2 + \lambda b_{2r}\int_\Omega \phi^{2r-2}|\nabla\phi_0|^2\,\mathrm{d}\boldsymbol{x} \leqslant C(\kappa)\|\phi_0\|^2 + C(\alpha,\kappa) + \frac{\lambda}{2p}m_w\|\nabla\tilde{g}_0\|^2, \tag{52}$$

which turns with Gronwall's inequality into the expression

$$\|\phi_0(\cdot,t)\|^2 \leqslant \|\phi_0(\cdot,0)\|^2\exp(Ct) + \int_0^t\left(C(\alpha) + \frac{\lambda}{2p}m_w\|\nabla\tilde{g}_0\|^2\right)\exp(-Cs)\,\mathrm{d}s, \tag{53}$$

for $T^* \geqslant 0$ such that we finally obtain $\phi \in L^\infty(0,T^*;L^2(\Omega)) \cap L^2(0,T^*;H^2_E(\Omega))$.

ii) Control over time derivative: Using the test function $\varphi \in H^2_E(\Omega)$ in (49) allows us to estimate the time derivative term by

$$\frac{p}{2}|(\partial_t\phi_0,\varphi)| \leqslant C\Big(\left\|\Delta_{\hat{\mathrm{D}}}\phi_0\right\| + \|\tilde{g}_0\| + \|f(\phi_0)\| + \|\phi_0\|\Big)\|\Delta\varphi\|, \tag{54}$$

such that $\|\partial_t\phi_0\|_{(H^2_E(\Omega))^*} = \sup_{\varphi\in H^2_E}\frac{|(\partial_t\phi_0,\varphi)|}{\|\varphi\|_{H^2_E}} \leqslant C$ and hence $\|\partial_t\phi_0\|_{(H^2_E(\Omega))^*}{}^2 \leqslant C(T^*)$.

Step 2: (Galerkin approximation). As $H^2_E(\Omega)$ is a separable Hilbert space, we can identify a linearly independent basis $\varphi_j \in H^2_E(\Omega)$, $j \in \mathbb{N}$, which is complete in $H^2_E(\Omega)$. This allows us to define for each $N \in \mathbb{N}$ approximate solutions $\phi_0^N = \sum_{j=1}^N \eta_j^N(t)\varphi_j$ which solve

$$\frac{p}{2}\frac{\partial}{\partial t}\left(\phi_0^N,\varphi_j\right) + \frac{\lambda m}{p}\Big(\mathrm{div}\Big(\hat{\mathrm{D}}\nabla\phi_0^N\Big), \mathrm{div}\Big(\hat{\mathrm{D}}\nabla\varphi_j\Big)\Big) + \frac{\lambda}{p}\Big(\hat{\mathrm{M}}_w\nabla\tilde{g}_0, \nabla\varphi_j\Big) + \Big(\lambda f'(\phi_0^N)\hat{\mathrm{M}}_\phi\nabla\phi_0^N, \nabla\varphi_j\Big) + \Big(\hat{\mathrm{C}}\nabla\phi_0^N, \nabla\varphi_j\Big) = 0 \qquad j = 1,\ldots,N, \tag{55}$$

for the initial condition $\phi_0^N(\cdot, 0) = \psi^N(\cdot)$. The initial value problem (55) represents a system of N ordinary differential equations (ODEs) for the coefficients $\eta_j^N(t)$. Hence, classical ODE theory immediately provides existence and uniqueness of ϕ_0^N. Moreover, we have $\phi_0^N \in C(0, T^*; H_E^2(\Omega))$ and $\partial_t \phi_0^N \in L^2(0, T^*; H_E^{-2}(\Omega))$.

Step 3: (Passing to the limit). In the same way as in *Step 1*, we can derive *a priori* estimates for the approximate solutions ϕ_0^N by using the test function ϕ_0^N instead of the basis functions φ_j in (55). Hence, by weak compactness, see theorem 2.1 in section 2, there exist subsequences (for simplicity still denoted by ϕ_0^N) such that

$$\begin{aligned} \phi_0^N &\rightharpoonup \phi_0 && \text{in} L^2(0, T^*; H_E^2(\Omega)) \text{ weakly}, \\ \phi_0^N &\overset{*}{\rightharpoonup} \phi_0 && \text{in} L^\infty(0, T^*; L^2(\Omega)) \text{ weak-star}, \\ \phi_0^N &\to \phi_0 && \text{in } L^2(0, T^*; L^2(\Omega)) \text{ strongly}, \end{aligned} \tag{56}$$

for $N \to \infty$. This allows us to pass to the limit in the initial value problem (55) such that we obtain

$$\frac{p}{2}\frac{\partial}{\partial t}(\phi_0, \varphi) + \frac{\lambda}{p}\Big(\mathrm{div}\Big(\hat{\mathrm{D}}\nabla\phi_0\Big), \mathrm{div}\Big(m\hat{\mathrm{D}}\nabla\varphi\Big)\Big) + \frac{\lambda}{p}\Big(\hat{\mathrm{M}}_w\nabla\tilde{g}_0, \nabla\varphi\Big) + \Big(\lambda f'(\phi_0)\hat{\mathrm{M}}_\phi\nabla\phi_0, \nabla\varphi\Big) + \Big(\hat{\mathrm{C}}\nabla\phi_0, \nabla\varphi\Big) = 0, \tag{57}$$

for all $\varphi \in H_E^2(\Omega)$. In the same way we can pass to the limit with respect to the initial condition ψ^N. □

6. Conclusions

The main new result here is the extension of the study by Schmuck *et al* in the absence of flow [42] to include a periodic fluid flow in the case of sufficiently large Péclet number. The resulting new effective porous media approximation (19) of the microscopic Stokes–Cahn–Hilliard problem (11)–(12) reveals interesting physical characteristics such as diffusion–dispersion relations by $(20)_2$–$(20)_3$ for instance. The homogenization methodology developed here allows for the systematic and rigorous derivation of effective macroscopic porous media equations starting with the fundamental works on Darcy's law [11, 18]. As a by-product of the methodology, one recovers rigorously and systematically the dispersion relations proposed in [10].

We note that the Cahn–Hilliard and related equations generally model more complex material transport [29, 35] 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 (11). This assumption implies a quasi-steady state on the fluid velocity and seems currently to be inevitable for the homogenization theory to work such as the assumption of a scale separated chemical potential on the level of the reference cells.

So far, we are restricted to mixtures of two fluids by our model. An extension towards mixtures of $N > 2$ components is studied in [30] 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 mathematically design such a free energy based on physical principles and phenomenological observations. As an example of the applicability of the presented upscaled equations, we have numerically solved the homogenized Cahn–Hilliard equation in a simple porous medium the unit reference cell of which consists of a periodic non-straight channel. We observed that the macroscopic solution retained the periodic properties induced at the microscopic scale. This numerical study represents a first step towards the use of the presented methodology in more complex geometries with e.g. non-periodic properties, something that we leave as a future work. In addition, a more detailed numerical study shall allow to rationally analyse current applications in science and engineering and hopefully reveal potential new ones.

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