Effective macroscopic Stokes-Cahn-Hilliard equations for periodic immiscible flows in porous media

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Abstract. Using thermodynamic and variational principles we study a basic phase field model for the mixture of two incompressible fluids in strongly perforated domains. We rigorously derive an effective macroscopic phase field equation under the assumption of periodic flow and a sufficiently large Péclet number with the help of the multiple scale method with drift and our recently introduced splitting strategy for Ginzburg-Landau/Cahn-Hilliard-type equations [19]. As for the classical convection-diffusion problem, we obtain systematically diffusion-dispersion relations (including Taylor-Aris-dispersion). 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.

Keywords: homogenization, diffusion-dispersion relations, porous structures, Stokes-Cahn-Hilliard equations

1 Introduction and results

We describe an arbitrary interface between two fluids by the total energy density,

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

where ϕ is a conserved order-parameter that evolves between different fluid 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). We allow for free energies F which represent polynomials of the form

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

We introduce the Lagrangian coordinate $\mathbf{X}$ for the (initial) material configuration and we denote by $\mathbf{x}(\mathbf{X},t)$ the Eulerian (reference) coordinate. The last two terms in (1) represent the well-known Cahn-Hilliard/Ginzburg-Landau free energy density adapted to the flow map $\mathbf{x}(\mathbf{X},t)$ defined by

$$\begin{cases} \frac{\partial \mathbf{x}}{\partial t} = \mathbf{u}(\mathbf{x}(\mathbf{X},t),t)\,, \\ \mathbf{x}(\mathbf{X},0) = \mathbf{X}\,. \end{cases} \tag{3}$$

By the kinetic energy, i.e., the first term in (1), we can account for fluid flow of incompressible materials with the viscosity μ, i.e.,

$$\begin{cases} \frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u}\cdot\nabla)\mathbf{u} - \mu\Delta\mathbf{u} + \nabla p = \mathbf{g}\,, \\ \operatorname{div}\mathbf{u} = 0\,, \end{cases} \tag{4}$$

where $\mathbf{g}$ is a driving force acting on the fluid. We consider mixtures of two incompressible and immiscible fluids of the same viscosity μ which is satisfied in many practical situations.

Suppose that the fluid initially occupies a domain $\Omega \subset \mathbb{R}^d$, with $d > 0$ the dimension of space. For an arbitrary length of time $T > 0$ we then define the total energy by

$$E(\mathbf{x}) := \int_0^T \int_\Omega e(\mathbf{x}(\mathbf{X},t),t)\,d\mathbf{X}\,dt\,. \tag{5}$$

Equation (5) combines an action functional for the flow map $\mathbf{x}(\mathbf{X},t)$ and a free energy for the order parameter ϕ and hence combines mechanical and thermodynamic energies [7,6,9,10,12]. We will focus our studies on quasi-stationary, i.e., $\mathbf{u}_t = \mathbf{0}$ and $\mathbf{g} \neq \mathbf{0}$, and low-Reynolds number flows, i.e., $(\mathbf{u}\cdot\nabla)\mathbf{u} = \mathbf{0}$. By using calculus of variations [20] and the theory of gradient flows together with the imposed boundary condition $\int_{\partial\Omega} w(\mathbf{x})\,do(\mathbf{x})$ for $w(\mathbf{x}) \in H^{3/2}(\partial\Omega)$, where do is the surface maesure, we derive the following set of equations

$$\text{(Homogeneous case)} \begin{cases} -\mu\Delta\mathbf{u} + \nabla p = \mathbf{g} & \text{in } \Omega_T\,, \\ \phi_t + \mathrm{Pe}(\mathbf{u}\cdot\nabla)\phi = \lambda \operatorname{div}\left(\nabla\left(f(\phi) - \Delta\phi\right)\right) & \text{in } \Omega_T\,, \\ \quad \nabla_n\phi := \mathbf{n}\cdot\nabla\phi = w(\mathbf{x}) & \text{on } \partial\Omega_T\,, \\ \quad \nabla_n\Delta\phi = 0 & \text{on } \partial\Omega_T\,, \\ \quad \phi(\mathbf{x},0) = h(\mathbf{x}) & \text{on } \Omega\,, \end{cases} \tag{6}$$

where $\operatorname{div}\mathbf{u} = 0$ in Ω_T, $\mathbf{u} = \mathbf{0}$ on $\partial\Omega_T$, $\Omega_T := \Omega\times]0,T[$, $\partial\Omega_T := \partial\Omega^1\times]0,T[$, λ represents the elastic relaxation time of the system, and the driving force $\mathbf{g}$ accounts for the elastic energy [10]

$$\mathbf{g} = -\gamma \operatorname{div}\left(\nabla\phi \otimes \nabla\phi\right)\,, \tag{7}$$

where γ corresponds to the surface tension [11] and hence we set $\gamma = \lambda$ for simplicity as in [1]. We denote by $\mathrm{Pe} := \frac{k\tau L U}{D}$ the dimensionless Péclet number for a reference fluid velocity $\mathrm{U} := |\mathbf{u}|$, L is the characteristic length of the porous medium, and via Einstein's relation we obtain the diffusion constant $D = k\tau M$ from the mobility M for the Boltzmann constant k and temperature τ. Our restriction to the Stokes equation (in difference to [10]) is motivated here by the fact that such flows turn into Darcy's law in porous media, e.g. [5,8]. The main

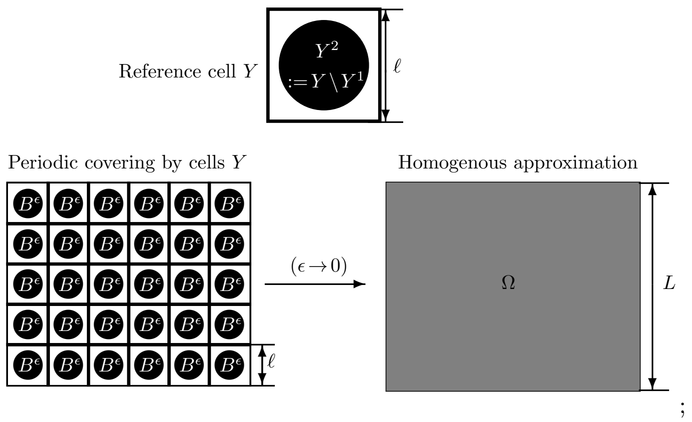

Fig. 1. Left: Porous medium $\Omega^\epsilon := \Omega \setminus B^\epsilon$ as a periodic covering of reference cells $Y := [0,\ell]^d$. **Top:** Reference cell $Y = Y^1 \cup Y^2$. **Right:** "Homogenization limit".

objective of our study is to derive effective macroscopic equations describing (6) in the case of perforated domains $\Omega^\epsilon \subset \mathbb{R}^d$ instead of a homogeneous $\Omega \subset \mathbb{R}^d$. A well-accepted approach is to represent a porous medium $\Omega = \Omega^\epsilon \cup B^\epsilon$ periodically with pore space Ω^ϵ and solid phase B^ϵ. We define $I^\epsilon := \partial\Omega^\epsilon \cap \partial B^\epsilon$ where $\epsilon > 0$ defines the heterogeneity $\epsilon = \frac{\ell}{L}$ for a characteristic pore size ℓ and the characteristic length of the porous medium L, see Figure 1. Then, we define the porous medium by a periodic covering with 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 $\epsilon \to 0$, for the derivation effective macroscopic porous media equations, as depicted in Figure 1. The pore and the solid phase of the medium are defined as usual by, $\Omega^\epsilon := \bigcup_{\mathbf{z}\in\mathbb{Z}^d} \epsilon\left(Y^1 + \mathbf{z}\right) \cap \Omega$, and $B^\epsilon := \bigcup_{\mathbf{z}\in\mathbb{Z}^d} \epsilon\left(Y^2 + \mathbf{z}\right) \cap \Omega = \Omega \setminus \Omega^\epsilon$, 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.

For a feasible and reliable upscaling, we restrict ourselves to periodic flows defined on a single reference cell Y, i.e.,

$$\text{(Periodic flow)} \quad \begin{cases} -\mu\Delta_{\mathbf{y}}\mathbf{u} + \nabla_{\mathbf{y}}p = -\gamma\mathrm{div}_{\mathbf{y}}\left(\nabla_{\mathbf{y}}\psi \otimes \nabla_{\mathbf{y}}\psi\right) & \text{in } Y^1\,, \\ \quad \mathbf{u} \text{ is } Y^1\text{-periodic}\,, & \\ \mathrm{Pe}(\mathbf{u}\cdot\nabla_{\mathbf{y}})\psi = \lambda\mathrm{div}_{\mathbf{y}}\left(\nabla_{\mathbf{y}}(f(\psi) - \Delta_{\mathbf{y}}\psi)\right) & \text{in } Y^1\,, \\ \quad \nabla_n\psi := (\mathbf{n}\cdot\nabla_{\mathbf{y}})\psi = w & \text{on } \partial Y^2\,, \\ \quad \nabla_n\Delta_{\mathbf{y}}\psi = 0 & \text{on } \partial Y^2\,, \end{cases} \tag{8}$$

where ψ is Y^1-periodic $\mathrm{div}_{\mathbf{y}}\,\mathbf{u} = 0$ in Y^1 and $\mathbf{u} = \mathbf{0}$ on ∂Y^2.

Motivated by [14,15], we study the case of large Péclet number and consider the following distinguished limit.

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 $\mathrm{Pe}_{mic} : - = \frac{k\tau\ell\mathrm{U}}{D}$, 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 (8), it is obvious that we have to apply the microscopic Péclet number in a corresponding microscopic formulation,

$$\text{(Microscopic problem)} \quad \begin{cases} \frac{\partial}{\partial t}\phi_\epsilon + \frac{\mathrm{Pe}_{mic}}{\epsilon}(\mathbf{u}(\mathbf{x}/\epsilon)\cdot\nabla)\phi_\epsilon & \\ \qquad = \lambda\mathrm{div}\left(\nabla\left(f(\phi_\epsilon) - \Delta\phi_\epsilon\right)\right) & \text{in } \Omega^\epsilon_T\,, \\ \nabla_n\phi_\epsilon := \mathbf{n}\cdot\nabla\phi_\epsilon = w(\mathbf{x}/\epsilon) & \text{on } I^\epsilon_T\,, \\ \nabla_n\Delta\phi_\epsilon = 0 & \text{on } I^\epsilon_T\,, \end{cases} \tag{9}$$

for the initial condition $\phi_\epsilon(\cdot,0) = \psi(\cdot)$ on Ω^ϵ and the definition $I^\epsilon_T := I^\epsilon\times]0,T[$. The microscopic system (10) 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.

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. We further note that physically, the periodic fluid velocity defined by (8) can be considered as the spatially periodic velocity of a moving frame. Hence, the periodic fluid velocity $\mathbf{u}(\mathbf{x}/\epsilon) := \mathbf{u}(\mathbf{y})$ enters the microscopic phase field problem as follows

$$\text{(Microscopic problem)} \quad \begin{cases} \frac{\partial}{\partial t}\phi_\epsilon + \frac{\mathrm{Pe}_{mic}}{\epsilon}(\mathbf{u}(\mathbf{x}/\epsilon)\cdot\nabla)\phi_\epsilon & \\ \qquad = \lambda\mathrm{div}\left(\nabla\left(f(\phi_\epsilon) - \Delta\phi_\epsilon\right)\right) & \text{in } \Omega^\epsilon_T\,, \\ \nabla_n\phi_\epsilon := \mathbf{n}\cdot\nabla\phi_\epsilon = w(\mathbf{x}/\epsilon) & \text{on } I^\epsilon_T\,, \\ \nabla_n\Delta\phi_\epsilon = 0 & \text{on } I^\epsilon_T\,, \\ \phi_\epsilon(\cdot,0) = \psi(\cdot) & \text{on } \Omega^\epsilon\,. \end{cases} \tag{10}$$

The main result of our study is the systematic derivation of upscaled immiscible flow equations which effectively account for pore geometries starting from the microscopic system (8)–(10) by passing to the limit $\epsilon \to 0$, i.e.,

$$\text{(Upscaled equation)} \quad \begin{cases} p\frac{\partial \phi_0}{\partial t} = \mathrm{div}\Big(\Big[pf'(\phi_0)\hat{\mathrm{M}} - \Big(2\frac{f(\phi_0)}{\phi_0} - f'(\phi_0)\Big)\hat{\mathrm{M}}_v - \hat{\Theta}(\mathbf{x},t) \\ \quad -\hat{\mathrm{C}}(\mathbf{x},t)\Big]\nabla\phi_0\Big) - f'(\phi_0)\mathrm{div}\left((\hat{\mathrm{M}}_v + \hat{\mathrm{K}})\nabla\phi_0\right) \\ \quad +\frac{\lambda^2}{p}\mathrm{div}\left(\hat{\mathrm{M}}_w\nabla\left(\mathrm{div}\left(\hat{\mathrm{D}}\nabla\phi_0\right) - \tilde{w}_0\right)\right)\,, \end{cases} \tag{11}$$

where $\hat{\Theta}(\mathbf{x},t) := \{\theta_{kl}\}_{1\leq k,l\leq d}$ and $\hat{C}(\mathbf{x},t) := \{\mathrm{c}_{ik}\}_{1\leq k,l\leq d}$ take the fluid convection into account, i.e.,

$$\theta_{kl} := \frac{\mathrm{Pe}_{mic}}{|Y|}\int_{Y^1}(\mathbf{u}\cdot\nabla_{\mathbf{y}})\zeta^{kl}(\mathbf{y})\,d\mathbf{y}\,,\quad \mathrm{c}_{ik} := \frac{\mathrm{Pe}_{mic}}{|Y|}\int_{Y^1}(\mathrm{u}^i - \mathrm{v}^i)\delta_{ik}\xi_v^k(\mathbf{y})\,d\mathbf{y}\,. \tag{12}$$

These two tensors account for the so-called diffusion-dispersion relations (e.g. Taylor-Aris-dispersion [3,4,21]). The tensors $\hat{\mathrm{M}}$, $\hat{\mathrm{M}}_v$ and $\hat{\mathrm{M}}_w$ are derived and defined in [18]. The result (11) makes use of the recently proposed splitting strategy for homogenization of fourth order problems in [19] and an asymptotic multiscale expansion with drift introduced in [2,13]. We note that the nonlinear problem (11) is characterized by a complex coupling between the micro- and the macroscale. As a consequence, the reference cell problems need to be computed for each macroscopic degree of freedom now and seems to be an intrinsic feature of upscaling nonlinearly coupled problems [16,18,19].

2 Conclusion

The main new result here is the extension of the results in the study by Schmuck *et al.* in the absence of flow [19] to include a periodic fluid flow in the case of sufficiently large Péclet number. The resulting new effective porous media approximation (11) of the microscopic Stokes-Cahn-Hilliard problem (8)–(10) reveals interesting physical characteristics such as diffusion-dispersion [4] relations by (12). The homogenization methodology serves as a systematic tool for the reliable and rigorous derivation of effective macroscopic porous media equations starting with the fundamental work on Darcy's law [5,8]. A qualitative quantification of the new equations by error estimates [17] would be very interesting.

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References

1. Abels, H.: Double obstacle limit for a Navier-Stokes/Cahn-Hilliard system. in Progress in Nonlinear Differential Equations and Their Applications 43, 1–20 (Springer 2011)

2. Allaire, G., Brizzi, R., Mikelić, A., Piatnitski, A.: Two-scale expansion with drift approach to the Taylor dispersion for reactive transport through porous media. Chem. Eng. Sci. 65, 2292–2300 (2010)
3. Aris, R.: On the Dispersion of a Solute in a Fluid Flowing through a Tube. Proc. R. Soc. A 235(1200), 67–77 (1956), http://dx.doi.org/10.2307/100013
4. Brenner, H.: Dispersion Resulting from Flow through Spatially Periodic Porous Media. Phil. Trans. Roy. Soc. London, A 297(1430), 81–133 (Jul 1980), http://dx.doi.org/10.1098/rsta.1980.0205
5. Carbonell, R., Whitaker, S.: Dispersion in pulsed systems – II: Theoretical developments for passive dispersion in porous media. Chem. Engng Sci. 38(11), 1795 – 1802 (1983), http://www.sciencedirect.com/science/article/pii/0009250983850362
6. Doi, M.: The Theory of Polymer Dynamics. Oxford Science Publication (1986)
7. de Gennes, P.G., Prost, R.L.: The Physics of Liquid Crystals. Oxford University Press (1993)
8. Hornung, U.: Homogenization and Porous Media. Springer (1997)
9. Lin, F.H., Liu, C.: Nonparabolic dissipative systems, modeling the flow of liquid crystals. Comm. Pure Appl. Math. XLVIII, 501–537 (1995)
10. Liu, C., Shen, J.: A phase field model for the mixture of two incompressible fluids and its approximation by a fourier-spectral method. Phys. D 179, 211–228 (2003)
11. Liu, C., Walkington, N.J.: An Eulerian description of fluids containing visco-hyperelastic particles. Arch. Rat. Mech. Anal. 159, 229–252 (2001)
12. Lowengrub, J., Truskinovsky, L.: Quasi-incompressible Cahn-Hilliard fluids and topological transitions. Proc. R. Soc. A 454, 2617–2654 (1998)
13. Marušic-Paloka, E., Piatnitski, A.L.: Homogenization of a nonlinear convection-diffusion equation with rapidly oscillating coefficients and strong convection. J. London Math. Soc. 72(02), 391 (Oct 2005), http://jlms.oxfordjournals.org/cgi/doi/10.1112/S0024610705006824
14. Mei, C.C.: Method of homogenization applied to dispersion in porous media. Transp. Porous Media 9, 261–274 (1992), http://dx.doi.org/10.1007/BF00611970, 10.1007/BF00611970
15. Rubinstein, J., Mauri, R.: Dispersion and convection in periodic porous media. SIAM J. Appl. Math. 46(6), 1018–1023 (1986), http://www.jstor.org/stable/2101656
16. Schmuck, M.: A new upscaled Poisson-Nernst-Planck system for strongly oscillating potentials. arXiv:1209.6618v1 preprint (2012), http://arxiv.org/abs/1209.6618
17. Schmuck, M.: First error bounds for the porous media approximation of the Poisson-Nernst-Planck equations. Z. angew. Math. Mech. 92(4), 304–319 (2012)
18. Schmuck, M., Berg, P.: Homogenization of a catalyst layer model for periodically distributed pore geometries in PEM fuel cells. Appl. Math. Res. Express. AMRX in press (2012), http://amrx.oxfordjournals.org/content/early/2012/07/08/amrx.abs011.full.pdf+html
19. Schmuck, M., Pradas, M., Pavliotis, G.A., Kalliadasis, S.: Upscaled phase-field models for interfacial dynamics in strongly heterogeneous domains. Proc. R. Soc. A 468, 3705–3724 (2012)
20. Struwe, M.: Variational Methods: Applications to Nonlinear Partial Differential Equations and Hamiltonian Systems. Springer, Dordrecht (2008)
21. Taylor, G.: Dispersion of Soluble Matter in Solvent Flowing Slowly through a Tube. Proc. R. Soc. A 219(1137), 186–203 (Aug 1953), http://dx.doi.org/10.1098/rspa.1953.0139