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Reactive flows in layered porous media, I. Homogenization of free boundary problems *

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Abstract

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A model of reactive flow in a layered porous medium is considered in which the layering is represented by small-scale periodic structure. A novel form of homogenization analysis is presented, combining geometric optics and multiple scales expansions together with matched asymptotics to derive an effective free boundary problem for the motion of the reactive interface. Applications of the effective free boundary equations are given in which travelling wave solutions and the stability of shape perturbations are considered.

1. Introduction

The study of reactive flows in porous media is important in the fundamental understanding of many geochemical situations such as the diagenesis and evolution of mineral deposits, oil and gas reserves, assessing the integrity of chemical and nuclear waste repositories, and even in mineral extraction processes such as *in situ* coal gasification, enhanced oil recovery and leaching of minerals.

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The model we consider for this problem involves a solvent which is being forced into a porous medium. The solute will be dissolved upstream while sufficiently far downstream the solvent will become saturated. In between these two regions there is a reaction interface that is typically thin and across which the porosity changes rapidly from its downstream value to its upstream value as a result of the dissolution process taking place at the front. Understanding the possible formation and stability of protrusions along the front and the shape selection of morphologically more complicated reaction zones such as fingers is important in the above geochemical situations. Such protrusions can result from a relatively higher permeability at one point of the front than at neighboring points on the front. As a result of this permeability increase, the unsaturated solvent is then focused at the tip of the protrusion which in turn advances more rapidly than the other points of the front. This destabilizing mechanism is known as the reaction-infiltration instability. There is, however, a competing process which tends to inhibit the uncontrolled growth of such protrusions. As the aspect ratio of the protrusion increases, diffusion of solute from the sides of the protrusion increases the concentration of the solute in the solvent at the tip, thereby inhibiting the advancement of the protrusion (see Fig. 1). The shape stability of the reaction-infiltration instability has been studied by us [1–3] in the case of a homogeneous porous medium.

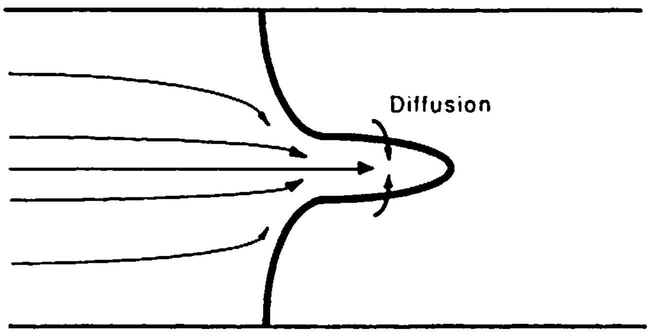

Fig. 1.

In this paper we consider the case of reactive flow through layered porous media. The inclusion of layering in the model is important as many of the world's natural gas and oil deposits are found in underground chambers whose impervious sides are made up of alternating low and higher porosity layers. These layers were formed by the oscillatory deposition caused by nucleation feedback and coupled mechano-chemical processes. Similar processes also formed under-pressured chambers, which instead of containing gas and oil are under vacuum. These under-pressured chambers are currently being investigated as potential chemical and nuclear waste repositories. An assessment of the integrity of the layered walls of such chambers depends on a fundamental understanding of their resistance to unstable breakout by the flow of highly reactive fluids. Conversely acid injection processes are also used in petroleum engineering to repair well damage to banded reservoirs. Therefore, the effect of layering on the reaction-infiltration process is of fundamental importance in these areas.

Layering occurs naturally on a wide range of length scales varying from a few grain diameters to tens or even hundreds of meters. Specific examples of layering are found in the Anadarko basin in the Simpson group of sandstones [12], the marl/limestone alterations [13] and selected shales in the Woodford formation. Reaction fronts, such as those mentioned above, occur both naturally and in the mineral extraction process. Variations in temperature or grain rate coefficients result in reaction fronts whose widths range from the grain scale to hundreds of meters. Therefore the ratio of front-width to layer thickness can have asymptotic limits ∞, 1, and 0 – all of which are relevant. In this paper we model layering by assuming that the porosity of the medium has a fine-scale periodic structure in the direction of the layering. We restrict ourselves to the distinguished limit in which the period of the layering is the same as the width of the front. A practical example of such a distinguished limit is provided by the HCl/HF acid injection process used to repair layered reservoirs damaged by wells [12]. In this case the period of the layering is typically 10 mm whereas the reaction front widths fall within the range 1–100 mm.

In this paper we provide the details of a novel application of homogenization in the context of free boundary problems. Homogenization is used to reduce the reactive flow equations with fine-scale layered structure to an effective free boundary problem. The approach uses combined multiple scale-geometric optics expansions and matched asymptotics (similar to boundary layer theory) in order to derive effective medium equations and jump conditions for the free boundary. In this context the matching process is subtle as the true solution can be expected to exhibit the same fine-scale oscillations present in the porosity function. Matching is achieved between inner and outer solutions by stitching averaged inner variables to averaged outer variables. The effective free boundary problem that results from this analysis is more tractable than the original reactive flow equations. Indeed, it is possible to use the effective free boundary problem and to asses the effect of layering on the stability of travelling wave solutions to morphological perturbations. The details of the shape stability analysis for the effective free boundary problem have been presented elsewhere [6] and will only be summarized briefly in this paper.

In Section 2 we present the mathematical model for reactive flow in a porous medium. By considering the model equations in the large solid density limit and performing an appropriate rescaling of the variables the equations are reduced to a singular perturbation problem with a small parameter ε. In terms of this small parameter ε, the width of the reaction front can be shown by rigorous estimates [16] to be $O(\varepsilon^{1/2})$. In Section 3 we provide a detailed derivation of the effective free boundary problem for the case of a horizontally layered medium in the distinguished limit in which the layer period δ equals the width of the front, i.e., $\delta = \varepsilon^{1/2}$. Other cases such as vertical layering can be treated in a completely analogous fashion. In Section 4 we demonstrate how the effective free boundary equations can be applied by providing a brief summary of the results of the shape stability analysis.

2. The mathematical model

For the purposes of the model we consider reactive flow within the infinite strip $(x, y) \in (-\infty, \infty)$. Within this strip the rate of increase in porosity, $\varphi(x, y, t)$, (equivalently the rate of dissolution of the soluble minerals) is proportional to the reaction rate:

$$\varphi_t = -k(\varphi_f - \varphi)^{2/3}(c - c_{eq}) \tag{2.1}$$

where k is the reaction rate constant, $\varphi_f(x, y, t)$ is the final porosity after complete dissolution and $c(x, y, t)$ is the concentration of solute in the solvent with its equilibrium concentration being c_{eq}. The 2/3-power indicates that we are considering surface reactions, but as we shall see in the next section, the actual form of the reaction rate in (2.1) does not affect our results since these details will be confined to an infinitesimally thin reaction zone when we take the distinguished limit. The solute concentration per rock volume, φc, satisfies a mass conservation equation:

$$(\varphi c)_t = \nabla \cdot \left[\varphi D(\varphi)\nabla c + \varphi c \varkappa(\varphi)\nabla p\right] + \rho \varphi_t \tag{2.2}$$

where D and $\varkappa$ are the porosity dependent diffusion coefficient and permeability, and ρ is the density of the minerals being dissolved. In (2.2), p is the pressure and Darcy's law for the velocity $\underline{v} = -\varkappa(\varphi)\nabla p$ has been used. Finally the conservation of water implies

$$\varphi_t + \nabla \cdot \left(\varphi \varkappa(\varphi)\nabla p\right) = 0. \tag{2.3}$$

The three equations (2.1)–(2.3) are to be solved for the three unknowns φ, c, p subject to following the imposed asymptotic conditions,

$$c \to 0, \ \varphi \to \varphi_f, \text{ and } p_x \to p_f' \quad \text{as } x \to -\infty \tag{2.4a}$$

and

$$c \to c_{eq}, \ \varphi \to \varphi_b, \text{ and } p_x \to ? \quad \text{as } x \to +\infty \tag{2.4b}$$

along with initial data. The conditions (2.4a) have the following physical interpretations: at the inlet ($x = -\infty$), the water is free of solute, the porous medium has reached its final altered state in which all the soluble minerals have been previously dissolved out, and a horizontal pressure gradient (equivalently, velocity, through Darcy's law) has been imposed. Similarly, the conditions (2.4b) imply that at the outlet far downstream ($x = +\infty$), the water is saturated with solute, the porous medium is in its original state with porosity $\varphi_b(x, y, t)$ and the horizontal pressure gradient is to be determined as part of the problem. We take zero-flux boundary conditions on the transverse boundaries of the strip.

In most geological examples of interest the (transverse) size of the reaction zone is several orders of magnitude greater than its thickness and the details inside this zone are not of interest *per se* except in the way in which the cumulative effect governs the evolution of the reaction zone on this larger scale. To this end we scale the above equations in terms of the parameter [1]

$$\varepsilon = c_{eq}/\rho \tag{2.5}$$

(typical values of ε range from 10^{-3} to 10^{-10}) and examine the limit $\varepsilon \to 0$ (the so-called large solid density limit) in the next section. One expects the thickness of the resulting reaction front separating the two values of φ to be very thin (indeed, it is $O(\varepsilon^{1/2})$) and the front itself to move very slowly. Thus we introduce a slow dimensionless time $\tilde{t}$ by

$$\tilde{t} = \varepsilon(kc_{eq})t \tag{2.6}$$

and space variables by

$$\tilde{\underline{r}} = (kc_{eq})^{1/2}\underline{r} \tag{2.7}$$

along with the scaled concentration

$$\gamma = c/c_{eq} \tag{2.8}$$

Equations (2.1)–(2.4) can then be written as (dropping the tildes, and writing $d(\varphi) = \varphi D(\varphi)$, $\lambda(\varphi) = \varphi \varkappa(\varphi)$)

$$\varepsilon(\varphi\gamma)_t = \nabla \cdot \left[d\nabla\gamma + \lambda\gamma\nabla p\right] + \varphi_t, \tag{2.9}$$

$$\varepsilon\varphi_t = -(\varphi_f - \varphi)^{2/3}(\gamma - 1), \tag{2.10}$$

$$\varepsilon\varphi_t = \nabla \cdot [\lambda\nabla p], \tag{2.11}$$

$$\gamma \to 0,\ \varphi \to \varphi_f,\ \text{and } p_x \to p_f' \quad \text{as } x \to -\infty, \tag{2.12a}$$

$$\gamma \to 1,\ \varphi \to \varphi_b,\ \text{and } p_x \to ? \quad \text{as } x \to +\infty. \tag{2.12b}$$

3. Derivation of effective equations and interface conditions

The model presented in Section 2 is general in that no assumptions (such as layering) were made about the porous medium itself. We build layering into the model by assuming that the initial porosity ϕ_b, in addition to its macroscopic dependence on x and y, has a fine-scale periodic structure in one or both of the independent variables. In order to account for the effect of this fine-scale layering on the movement of the reaction front, we develop a novel homogenization approach that ultimately yields an effective free boundary problem. In order to illustrate this analysis we consider the case of an initially vertically layered medium, which after interaction with the solvent, has a homogeneous distribution of porosity, i.e., $\phi_b = \phi_b(y/\varepsilon^{1/2})$ and ϕ_f = constant. This case is sufficiently rich to illustrate the methodology while retaining simplicity for the purposes of presentation. The effective equations in a more general setting follow analogously and are merely stated.

The reaction front is characterized by a narrow region having a width $O(\varepsilon^{1/2})$ across which the dependent variables change rapidly. Our strategy is to assume inner and outer multiple scale perturbation expansions of the unknowns. The outer expansion involves the macroscopic variables x, y and t as well as the microscopic variable y/δ to represent the vertical periodicity on the length scale $\delta = \varepsilon^{1/2}$. The inner expansion involves the microscope variables $S(x, y, t, \delta)/\delta$ and y/δ representing the normal coordinate to the unknown free boundary and medium periodicity, respectively, and the macroscopic variables $T(x, y, t, \delta)$ and t representing the tangential coordinate to the unknown free boundary and the time t, respectively. Redundancy in these representations are removed by exploiting the periodicity assumption. We then derive a consistent set of effective equations and interface conditions by matching averaged (i.e., macroscopically varying) inner variables to averaged outer variables.

3.1. Outer expansions

We expand each of the independent variables ϕ, γ and p in a multiple scale perturbation expansion of the form:

$$\phi_\delta = \tilde{\phi}(x, y, y/\delta, t) = \tilde{\phi}_0(x, y, y/\delta, t) + \delta\tilde{\phi}_1(x, y, y/\delta, t) + \dots . \tag{3.1.1}$$

Here we have used $(\tilde{\cdot})$ to distinguish an outer variable from an inner variable. However, within this section dealing only with outer expansions we shall drop this notation.

Scaled equations

Let $\eta = y/\delta$ then $\partial_y \phi = (\partial_y + \delta^{-1}\partial_\eta)\tilde{\phi}$ and (dropping the $\tilde{\ }$) (2.9)–(2.11) becomes:

$$\delta^2(\phi\gamma)_t = (d\gamma_x + \lambda\gamma p_x)_x + \left(\partial_y + \delta^{-1}\partial_\eta\right)\left[d\left(\gamma_y + \delta^{-1}\gamma_\eta\right) + \lambda\gamma\left(p_y + \delta^{-1}p_\eta\right)\right] + \phi_t, \tag{3.1.2a}$$

$$\delta^2\phi_t = -(\phi_f - \phi)^{2/3}(\gamma - 1), \tag{3.1.2b}$$

$$(\lambda p_x)_x + \left(\partial y + \delta^{-1}\partial_\eta\right)\left[\lambda\left(p_y + \delta^{-1}p_\eta\right)\right] = -(\phi_f - \phi)^{2/3}(\gamma - 1). \tag{3.1.2c}$$

Perturbation equations order by order

Substituting the expansions for ϕ, γ and p of the form (3.1.1) into (3.1.2) we obtain:
$O(\delta^{-2}) >$

$$\left[d(\phi_0)\gamma_{0,\eta} + \lambda(\phi_0)\gamma_0 p_{0,\eta}\right]_\eta = 0, \tag{3.1.3a}$$

$$\left[\lambda(\phi_0)p_{0,\eta}\right]_\eta = 0. \tag{3.1.3b}$$

From (3.1.3b) it follows that $p_{0,\eta} = g(x,y,t)/\lambda(\phi_0)$ and since p_0 is periodic in η:

$$0 = \langle p_{0,\eta}\rangle = g(x,y,t)\langle\lambda^{-1}(\phi_0)\rangle.$$

Now assuming that $\phi_0 > 0$ it follows that $g \equiv 0$ from which it follows that the zeroth order pressure function varies on the macroscopic scale only:

$$p_0 = p_0(x,y,t). \tag{3.1.3c}$$

Similarly (3.1.3a) implies that

$$\gamma_0 = \gamma_0(x,y,t). \tag{3.1.3d}$$

Exploiting the zeroth order macroscopic dependences (3.1.3c, d) the next order equations become:
$O(\delta^{-1}) >$

$$\left[d(\phi_0)(\gamma_{0,y} + \gamma_{1,\eta}) + \lambda(\phi_0)\gamma_0(p_{0,y} + p_{1,\eta})\right]_\eta = 0, \tag{3.1.4a}$$

$$\left[\lambda(\phi_0)(p_{0,y} + p_{1,\eta})\right]_\eta = 0. \tag{3.1.4b}$$

Integrating (3.1.4b), dividing by $\lambda(\phi_0)$ and averaging we obtain

$$\lambda(\phi_0)(p_{0,y} + p_{1,\eta}) \equiv F(x,y,t) = \langle\lambda(\phi_0)^{-1}\rangle^{-1}p_{0,y}. \tag{3.1.4c}$$

Averaging once again we obtain

$$\langle\lambda(\phi_0)p_{1,\eta}\rangle = p_{0,y}\left(\langle\lambda(\phi_0)^{-1}\rangle^{-1} - \langle\lambda(\phi_0)\rangle\right). \tag{3.1.4d}$$

Integrating (3.1.4a) and using (3.1.4c) we obtain

$$d(\phi_0)(\gamma_{0,y} + \gamma_{1,\eta}) + \gamma_0 F(x,y,t) = G(x,y,t). \tag{3.1.4e}$$

Dividing by $d(\phi_0)$ and averaging we obtain

$$\langle d(\phi_0)^{-1}\rangle^{-1}\gamma_{0,y} + \gamma_0 F(x,y,t) = G(x,y,t). \tag{3.1.4f}$$

Combining these last two equations we obtain

$$d(\phi_0)\gamma_{1,\eta} = \left(\langle d(\phi_0)^{-1}\rangle^{-1} - d(\phi_0)\right)\gamma_{0,y}. \tag{3.1.4g}$$

Averaging (3.1.4g) we obtain

$$\langle d(\phi_0)\gamma_{1,\eta}\rangle = \gamma_{0,y}\left(\langle d(\phi_0)^{-1}\rangle^{-1} - \langle d(\phi_0)\rangle\right). \tag{3.1.4h}$$

Again exploiting the fact that γ_0 and p_0 are macroscopic quantities the next order equations are: $O(\delta^0) >$

$$\begin{aligned}&\left(d(\phi_0)\gamma_{0,x} + \lambda(\phi_0)\gamma_0 p_{0,x}\right)_x + \left[d(\phi_0)(\gamma_{0,y} + \gamma_{1,\eta}) + \lambda(\phi_0)\gamma_0(p_{0,y} + p_{1,\eta})\right]_y \\ &\quad + \big[d(\phi_0)\gamma_{1,x} + \lambda(\phi_0)\gamma_0 p_{1,y} + \lambda(\phi_0)\gamma_1 p_{0,y} \\ &\quad + d'(\phi_0)\phi_1\gamma_{0,y} + \lambda(\phi_0)\phi_1\gamma_0 p_{0,y} \\ &\quad + d'(\phi_0)\phi_1\gamma_{1,\eta} + d(\phi_0)\gamma_{2,\eta} + \left(\lambda'(\phi_1)\phi_1\gamma_0\right. \\ &\quad \left.+ \lambda(\phi_0)\gamma_1\right)p_{1,\eta} + \lambda(\phi_0)\gamma_0 p_{2,\eta}\big]_\eta + \phi_{0,t} = 0,\end{aligned} \tag{3.1.5a}$$

$$-(\phi_f - \phi_0)^{2/3}(\gamma_0 - 1) = 0, \tag{3.1.5b}$$

$$\begin{aligned}&\left(\lambda(\phi_0)p_{0,x}\right)_x + \left(\lambda(\phi_0)p_{0,y}\right)_y + \left(\lambda(\phi_0)p_{1,\eta}\right)_y + \left(\lambda'(\phi_0)\phi_1 p_{0,y} + \lambda(\phi_0)p_{1,y}\right)_\eta \\ &\quad + \left(\lambda'(\phi_0)\phi_1 p_{1,\eta} + \lambda(\phi_0)p_{2,\eta}\right)_\eta = 0.\end{aligned} \tag{3.1.5c}$$

Let $S_0(x,y,t) = 0$ denote the limit free boundary surface, then the region upstream from the front is characterized by $S_0(x,y,t) < 0$, while the region downstream from the front is characterized by $S_0(x,y,t) > 0$. We note that (3.1.5b) implies that either $\phi_0 = \phi_f$ or $\gamma_0 = 1$ depending on whether we are upstream or downstream from the front, respectively.

Effective equations downstream from the front $S_0(x,y,t) > 0$

Using $\gamma_0 = 1$ in (3.1.4g) we see that $\gamma_{1,\eta} = 0$ so that γ_1 is a macroscopic variable:

$$\gamma_1 = \gamma_1(x,y,t). \tag{3.1.6a}$$

Averaging over η in (3.1.5c) and using (3.1.4d) we obtain the effective pressure equation:

$$\left(\langle\lambda(\phi_0)\rangle p_{0,x}\right)_x + \left(\langle\lambda(\phi_0)^{-1}\rangle^{-1}p_{0,y}\right)_y = 0. \tag{3.1.6b}$$

Now applying the downstream boundary condition (2.1.2b) for ϕ we have $\phi_0 = \phi_b$. Thus in the limit $\delta = \varepsilon^{1/2} \to 0$ we have

$$\phi_\delta \overset{w}{\to} \langle\phi_b\rangle, \quad p_\delta \to p_0(x,y,t), \quad \gamma_\delta \to 1,$$

where $p_0(x,y,t)$ satisfies (3.1.6b) with $\phi_0 = \phi_b$.

Effective equations upstream from the front $S_0(x,y,t) < 0$

In the upstream region we have $\phi_0 = \phi_f$. Averaging (3.1.5a) over η and using the fact that $\phi_f = \phi_f(\eta)$ we obtain:

$$\big(\langle d(\phi_f)\rangle\gamma_{0,x} + \langle\lambda(\phi_f)\rangle\gamma_0 p_{0,x}\big)_x + \big(\langle d(\phi_f)\rangle\gamma_{0,y} + \langle\lambda(\phi_f)\rangle\gamma_0 p_{0,y}\big)_y$$
$$+ \big(\langle d(\phi_0)\gamma_{1,\eta}\rangle + \langle\lambda(\phi_f)p_{1,\eta}\rangle\gamma_0\big)_y = 0.$$

Making use of (3.1.4d, h) we obtain the effective concentration equation:

$$\begin{aligned}&\big(\langle d(\phi_f)\rangle\gamma_{0,x} + \langle\lambda(\phi_f)\rangle\gamma_0 p_{0,x}\big)_x\\&\quad + \big(\langle d(\phi_f)^{-1}\rangle^{-1}\gamma_{0,y} + \langle\lambda(\phi_f)^{-1}\rangle^{-1}\gamma_0 p_{0,y}\big)_y = 0.\end{aligned} \tag{3.1.6c}$$

For the upstream effective pressure equation the same averaging procedure as we used for the downstream case applied to (3.1.5c) yields (3.1.6a) but with $\phi_0 = \phi_f$. Thus in the upstream region $S_0 < 0$ the solution in the limit $\delta = \varepsilon^{1/2} \to 0$ becomes:

$$\phi_\delta \overset{w}{\to} \langle\phi_f\rangle, \quad p_\delta \to p_0(x,y,t), \quad \gamma_\delta \to \gamma_0(x,y,t).$$

3.2. Inner expansions

We expand each of the independent variables ϕ, γ and p in interior multiple scale perturbation expansions of the form:

$$\phi_\delta = \phi_0\big(S(x,y,t,\delta)/\delta, T(x,y,t,\delta), y/\delta, t\big) + \delta\phi_1\big(S/\delta, T, y/\delta, t\big) + \dots, \tag{3.2.1a}$$

where we assume the following expansions for the normal and tangential coordinates:

$$\begin{aligned}S &= S(x,y,t;\delta) = S_0(x,y,t) + \delta S_1(x,y,y/\delta,t) + \dots,\\T &= T(x,y,t;\delta) = T_0(x,y,t) + \delta T_1(x,y,y/\delta,t) + \dots .\end{aligned} \tag{3.2.1b}$$

Note that the width of the front and the periodicity are assumed to be of the same order. Now defining $\xi_1 = S/\delta$, $\xi_2 = T$ and $\eta = y/\delta$ we have the following multiple scale operators:

$$\begin{aligned}\partial_x &\to \delta^{-1}S_x\partial_{\xi_1} + T_x\partial_{\xi_2},\\\partial_y &\to \delta^{-1}S_y\partial_{\xi_1} + T_y\partial_{\xi_2} + \delta^{-1}\partial_\eta,\\\partial_t &\to \delta^{-1}S_t\partial_{\xi_1} + T_t\partial_{\xi_2} + \partial_t.\end{aligned} \tag{3.2.1c}$$

Orthogonality condition

In order that the chosen coordinate variables S and T are orthogonal we impose the following orthogonality constraint:

$$\begin{aligned} 0 = S_x T_x + S_y T_y = S_{0,x} T_{0,x} &+ (S_{0,y} + S_{1,\eta})(T_{0,y} + T_{1,\eta}) \\ &+ \delta\big[S_{0,x} T_{1,x} + S_{1,x} T_{0,x} + (S_{0,y} + S_{1,\eta})(T_{1,y} + T_{2,\eta}) \\ &+ (S_{1,y} + S_{2,\eta})(T_{0,y} + T_{1,\eta})\big] + \mathrm{O}(\delta^2). \end{aligned} \tag{3.2.2}$$

Scaled equations

In terms of the rescaled variables (2.9)–(2.11) become:

$$\begin{aligned} &(\delta^{-1} S_t \partial_{\xi_1} + T_t \partial_{\xi_2} + \partial_t)\big[\delta^2 \phi\gamma - \phi\big] \\ &\quad = (\delta^{-1} S_x \partial_{\xi_1} + T_x \partial_{\xi_2})\big[d(\delta^{-1} S_x \partial_{\xi_1} + T_x \partial_{\xi_2})\gamma \\ &\quad + \lambda\gamma(\delta^{-1} S_x \partial_{\xi_1} + T_x \partial_{\xi_2})p\big] \\ &\quad + (\delta^{-1} S_y \partial_{\xi_1} + T_y \partial_{\xi_2} + \delta^{-1}\partial_\eta)\big[d(\delta^{-1} S_y \partial_{\xi_1} + T_y \partial_{\xi_2} + \delta^{-1}\partial_\eta)\gamma \\ &\quad + \lambda\gamma(\delta^{-1} S_y \partial_{\xi_1} + T_y \partial_{\xi_2} + \delta^{-1}\partial_\eta)p\big], \end{aligned} \tag{3.2.3a}$$

$$\delta^2(\delta^{-1} S_t \partial_{\xi_1} + T_t \partial_{\xi_2} + \partial_t)\phi = -(\phi_t - \phi)^{2/3}(\gamma - 1), \tag{3.2.3b}$$

$$\begin{aligned} &(\delta^{-1} S_x \partial_{\xi_1} + T_x \partial_{\xi_2})\big[\lambda(\delta^{-1} S_x \partial_{\xi_1} + T_x \partial_{\xi_2})p\big] \\ &\quad + (\delta^{-1} S_y \partial_{\xi_1} + T_y \partial_{\xi_2} + \delta^{-1}\partial_\eta)\big[\lambda(\delta^{-1} S_y \partial_{\xi_1} + T_y \partial_{\xi_2} + \delta^{-1}\partial_\eta)\big]p. \end{aligned} \tag{3.2.3c}$$

Perturbation equations order by order

Substituting the expansions (3.2.1) into (3.2.3) and gathering terms we have:
$\mathrm{O}(\delta^{-2}) >$

$$\begin{aligned} &S_{0,x}\partial_{\xi_1}\big[d(\phi_0) S_{0,x}\gamma_{0,\xi_1} + \lambda(\phi_0)\gamma_0 S_{0,x} p_{0,\xi_1}\big] \\ &\quad + (S_{0,y}\partial_{\xi_1} + \partial_\eta)\big[d(\phi_0)(S_{0,y}\gamma_{0,\xi_1} + \gamma_{0,\eta}) + \lambda(\phi_0)\gamma_0(S_{0,y} p_{0,\xi_1} + p_{0,\eta})\big] = 0, \end{aligned} \tag{3.2.4a}$$

$$S_{0,x}\partial_{\xi_1}\big[\lambda(\phi_0) S_{0,x} p_{0,\xi_1}\big] + (S_{0,y}\partial_{\xi_1} + \partial_\eta)\big[\lambda(\phi_0)(S_{0,y} p_{0,\xi_1} + p_{0,\eta})\big] = 0. \tag{3.2.4b}$$

Now (3.2.4b) is an equation for p_0 on the region $(\xi_1, \eta) \in (-\infty, \infty) \times T^1$. The limiting values $\xi_1 \to \pm\infty$ correspond to the matching region where the inner expansions are matched to the outer expansions, i.e.,

$$\lim_{\xi_1 \to \pm\infty} p_0(\xi_1, \xi_2, \eta, t) = \widetilde{p}_0(x_0, y_0, t) \tag{3.2.5}$$

where (x_0, y_0) satisfies

$$\begin{cases} 0 = S(x_0, y_0, t, \delta), \\ \xi_2 = T(x_0, y_0, t, \delta). \end{cases}$$

From (3.2.5) we conclude that since $p_0(\xi_1, \xi_2, \eta, t)$ has to match to the macroscopic quantity $\tilde{p}_0$ it follows that

$$\lim_{\xi_1 \to \infty} p_{0,\eta} = 0$$

and similarly with $\xi_1 \to -\infty$. We multiply (3.2.4b) by p_0, integrate over the region $(-\infty, \infty) \times T^1$ and integrate by parts to obtain

$$\int_{-\infty}^{\infty} \int_{T^1} \lambda(\phi_0) \left[S_{0,x}^2 p_{0,\xi_1}^2 + (S_{0,y} p_{0,\xi_1} + p_{0,\eta})^2 \right] \mathrm{d}\xi \, \mathrm{d}\eta = 0.$$

Now since $\lambda(\psi_0) > 0$ it follows that

$$S_{0,x} p_{0,\xi_1} = 0 \quad \text{and} \quad (S_{0,y} p_{0,\xi_1} + p_{0,\eta}) = 0. \tag{3.2.6}$$

We now assume that $|\nabla_{\xi_1 \eta} S_0| \neq 0$ and there are two possibilities:

Case 1: $S_{0,x} \neq 0$

From (3.2.6) it follows that $p_{0,\xi_1} = 0 = p_{0,\eta}$. Thus p_0 is constant over the region $(-\infty, \infty) \times T^1$ and since p_0 matches to $\tilde{p}_0$ either side of S_0 it follows that $\tilde{p}_0$ is continuous across S_0, i.e., $\tilde{p}_0(0+) = \tilde{p}_0(0-)$.

Case 2: $S_{0,y} \neq 0$

$S_{0,y} p_{0,\xi} + p_{0,\eta} = 0$ is a first order partial differential equation for p_0 whose solution is $p_0 = \psi(\xi_1 - S_{0,y}\eta)$ for some $\psi \in C^1(\mathbf{R})$. Because p_0 is periodic in η it follows that $\psi(r)$ must be a periodic function. However, since p_0 must match to a macroscopic quantity $\tilde{p}_0$ as $\xi_1 \to \infty$ the only possibility is that $p_0 = \psi =$ constant. Thus $\tilde{p}_0$ is also continuous across S_0.

Since $p_{0,\xi_1} = 0 = p_{0,\eta}$, (3.2.4a) is reduced to

$$S_{0,x} \partial_1 \left(d(\phi_0) S_{0,x} \gamma_{0,\xi_1} \right) + (S_y \partial_{\xi_1} + \partial_\eta) \left[d(\phi_0)(S_{0,y} \gamma_{0,\xi_1} + \gamma_{0,\eta}) \right] = 0.$$

This equation is the same as (3.2.4b) but with $\lambda(\phi_0)$ replaced by $d(\phi_0) > 0$. Therefore the above argument implies that $\gamma_{0,\xi_1} = 0 = \gamma_{0,\eta}$ and that $\tilde{\gamma}_0(0+) = \tilde{\gamma}_0(0-)$. Moreover, since $\gamma_0(+\infty, \xi_2, \eta, t) = \tilde{\gamma}_0 = 1$ it follows that $\gamma_0 = \gamma_0(\xi_2, \eta, t) \equiv 1$.

$O(\delta^{-1}) >$

Collecting the $O(\delta^{-1})$ terms in (A 2.3c) yields:

$$\begin{aligned} & S_{0,x} \partial_{\xi_1} \left[\lambda(\phi_0)(T_{0,x} p_{0,\xi_2} + S_{0,x} p_{1,\xi_1}) \right] \\ & \quad + \left[(S_{0,y} + S_{1,\eta}) \partial_{\xi_1} + \partial_\eta \right] \left[\lambda(\phi_0) \left\{ (T_{0,y} + T_{1,\eta}) p_{0,\xi_2} \right. \right. \\ & \quad \left. \left. + (S_{0,y} + S_{1,\eta}) p_{1,\xi_1} + p_{1,\eta} \right\} \right] = 0. \end{aligned} \tag{3.2.7}$$

Rearranging terms in (3.2.7) and using the fact that S_0, S_1, T_0 and T_1 do not depend on ξ_1 we obtain:

$$
\begin{aligned}
&[S_{0,x}^2 + (S_{0,y} + S_{1,\eta})^2]\big(\lambda(\phi_0)p_{1,\xi_1}\big)_{\xi_1} + [S_{0,x}T_{0,x} + (S_{0,y} + S_{1,\eta})(T_{0,y} + T_{1,\eta})] \\
&\quad \times \big(\lambda(\phi_0)p_{0,\xi_2}\big)_{\xi_1} + (S_{0,y} + S_{1,\eta})\big(\lambda(\phi_0)p_{1,\eta}\big)_{\xi_1} + \big[\lambda(\phi_0)(T_{0,y} + T_{1,\eta})p_{0,\xi_2} \\
&\quad + \lambda(\phi_0)(S_{0,y} + S_{1,\eta})p_{1,\xi_1} + \lambda(\phi_0)p_{1,\eta}\big]_\eta = 0.
\end{aligned}
$$

By means of the zeroth order in the orthogonality condition (3.2.2) equation (3.2.7) is further reduced to

$$
\begin{aligned}
&[S_{0,x}^2 + (S_{0,y} + S_{1,\eta})^2]\big(\lambda(\phi_0)p_{1,\xi_1}\big)_{\xi_1} + (S_{0,y} + S_{1,\eta})\big(\lambda(\phi_0)p_{1,\eta}\big)_{\xi_1} \\
&\quad + \big[\lambda(\phi_0)\{(T_{0,y} + T_{1,\eta})p_{0,\xi_2} + (S_{0,y} + S_{1,\eta})p_{1,\xi_1} + p_{1,\eta}\}\big]_\eta = 0.
\end{aligned}
\tag{3.2.8}
$$

Collecting $O(\delta^{-1})$ terms in (3.2.3a) and using $\gamma_{0,\xi_1} = 0 = \gamma_{0,\eta}$ we obtain

$$
\begin{aligned}
&S_{0,x}\partial_{\xi_1}\big[d(\phi_0)(T_{0,x}\gamma_{0,\xi_2} + S_{0,x}\gamma_{1,\xi_1}) + \lambda(\phi_0)\gamma_0(T_{0,x}p_{0,\xi_2} + S_{0,x}p_{1,\xi_1})\big] \\
&\quad + \big\{(S_{0,y} + S_{1,\eta})\partial_{\xi_1} + \partial_\eta\big\}\big[d(\phi_0)\big((T_{0,y} + T_{1,\eta})\gamma_{0,\xi_2} + (S_{0,y} + S_{1,\eta})\gamma_{1,\xi_1} + \gamma_{1,\eta}\big) \\
&\quad + \lambda(\phi_0)\gamma_0\big((T_{0,y} + T_{1,\eta})p_{0,\xi_2} + (S_{0,y} + S_{1,\eta})p_{1,\xi_1} + p_{1,\eta}\big)\big] + S_{0,t}\phi_{0,\xi_1} = 0.
\end{aligned}
$$

Gathering terms, using the orthogonality condition (3.2.2), and the fact that $\gamma_{0,\xi_1} = 0 = \gamma_{0,\eta}$ we obtain

$$
\begin{aligned}
&[S_{0,x}^2 + (S_{0,y} + S_{1,\eta})^2]\big(d(\phi_0)\gamma_{1,\xi_1}\big)_{\xi_1} + (S_{0,y} + S_{1,\eta})\big(d(\phi_0)\gamma_{1,\eta}\big)_{\xi_1} + S_{0,t}\phi_{0,\xi_1} \\
&\quad + \big[D(\phi_0)\{(T_{0,y} + T_{1,\eta})\gamma_{0,\xi_2} + (S_{0,y} + S_{1,\eta})\gamma_{1,\xi_1} + \gamma_{1,\eta}\}\big]_\eta = 0.
\end{aligned}
$$

Using the fact that $\gamma_0 = \gamma_0(\xi_2, \gamma, t) \equiv 1$ the previous equation becomes

$$
\begin{aligned}
&[S_{0,x}^2 + (S_{0,y} + S_{1,\eta})^2]\big(d(\phi_0)\gamma_{1,\xi_1}\big)_{\xi_1} + (S_{0,y} + S_{1,\eta})\big(d(\phi_0)\gamma_{1,\eta}\big)_{\xi_1} + S_{0,t}\phi_{0,\xi_1} \\
&\quad + \big[d(\phi_0)\{(S_{0,y} + S_{1,\eta})\gamma_{1,\xi_1} + \gamma_{1,\eta}\}\big]_\eta = 0.
\end{aligned}
\tag{3.2.9}
$$

3.3. Asymptotic matching

Having derived equations governing the outer and inner expansions we now present the procedure for matching the inner and outer solutions across the interface. This procedure results in jump conditions for the normal derivatives of $\widetilde{\gamma}_0$ and $\widetilde{p}_0$ and the eikonal equation for $S_0(x, y, t)$.

It should be noted that some matching has already been performed on $\widetilde{\gamma}_0$ and $\widetilde{p}_0$ themselves. The reason for not delaying all the matching to this section is that we wished to exploit the simplifications that result from the continuity of $\widetilde{\gamma}_0$ and $\widetilde{p}_0$ as soon as possible in the interests of brevity.

Local inversion of the transformation: $(\xi_1, \xi_2) \to (x, y)$

In the matching process we need to express the outer variables (x, y) in terms of the inner variables (ξ_1, ξ_2). From (3.2.1b) we have:

$$\begin{aligned} \delta\xi_1 &= S(x, y, t; \delta) = S_0(x, y, t) + \delta S_1(x, y, \eta, t) + \dots, \\ \xi_2 &= T(x, y, t; \delta) = T_0(x, y, t) + \delta T_1(x, y, \eta, t) + \dots . \end{aligned} \tag{3.3.1}$$

We want to match (ξ_1, ξ_2) to (x, y) while keeping η fixed. To achieve this consider the nominal point (x_0, y_0) defined to be the solution of the system:

$$\begin{aligned} 0 &= S(x_0, y_0, t; \delta), \\ \xi_2 &= T(x_0, y_0, t; \delta). \end{aligned} \tag{3.3.2}$$

The solution to (3.3.2) yields the parametric representation $x_0 = x_0(\xi_2)$, $y_0 = y_0(\xi_2)$. We now linearize the transformations (3.3.1) about (x_0, y_0) and invert. Let

$$x = x_0 + \delta x_1 + \dots, \qquad y = y_0 + \delta y_1 + \dots, \tag{3.3.3}$$

substitute these into (3.3.1) and expand. The $O(\delta)$ terms yield a linear system having the following solution

$$x_1 = T_{0,y}\xi_1/J, \qquad y_1 = -T_{0,x}\xi_1/J, \tag{3.3.4}$$

where $J(x_0, y_0, t) = S_{0,x}T_{0,y} - S_{0,y}T_{0,x}$ is assumed to be nonzero.

Matching inner and outer expansions for γ

Combining the properties of $\tilde{\gamma}_0$ and γ_0 established in Section 3.1 and 3.2, respectively, we obtain:

Outer expansion:

$S_0 > 0$:

$$\tilde{\gamma} = 1,$$

$S_0 < 0$:

$$\tilde{\gamma} = \tilde{\gamma}_0(x, y, t) + \delta\tilde{\gamma}_1(x, y, \eta, t) + \dots . \tag{3.3.5a}$$

Inner expansion:

$$\gamma = 1 + \delta\gamma_1(\xi_1, \xi_2, \eta, t) + \dots ; \tag{3.3.5b}$$

matching in the region $S_0 > 0$ is straightforward. To match in the region $S_0 < 0$ we substitute the expansions (3.3.3) for x and y into (3.3.5a), expand and eliminate x_1 and y_1 using (3.3.4) to obtain:

$$\begin{aligned} \tilde{\gamma} = \tilde{\gamma}_0(x_0, y_0, t) &+ \delta\big[\tilde{\gamma}_{0,x}(x_0, y_0, t)T_{0,y}\xi_1/J \\ &- \tilde{\gamma}_{0,y}(x_0, y_0, t)T_{0,x}\xi_1/J + \tilde{\gamma}_1(x_0, y_0, \eta, t)\big] + O(\delta^2). \end{aligned} \tag{3.3.5c}$$

Since $x_0 = x_0(\xi_2)$ and $y_0 = y_0(\xi_2)$, the outer expansion $\widetilde{\gamma}$ has effectively been expressed in terms of the inner variables ξ_1 and ξ_2. From (3.3.5b, c) we have the following matching conditions:
$O(\delta^0) >$

$$1 = \widetilde{\gamma}_0(x_0, y_0, t),$$

$O(\delta^1) >$

$$\gamma_1(\xi_1, \xi_2, \eta, t) = \widetilde{\gamma}_{0,x}(x_0, y_0, t)T_{0,y}\xi_1/J - \widetilde{\gamma}_{0,y}(x_0, y_0, t)T_{0,x}\xi_1/J + \widetilde{\gamma}_1(x_0, y_0, \eta, t) \quad \text{as } \xi_1 \to -\infty. \tag{3.3.5d}$$

We are now in a position to make use of the governing equation (3.2.9) for γ_1 to derive conditions for $\widetilde{\gamma}_0$. We start by determining the limiting values of the derivatives of γ_1 that appear in (3.2.9) by means of the matching condition (3.3.5d). We notice that since $x_0 = x_0(\xi_2)$, and $y_0 = y_0(\xi_2)$ it follows that γ_1 is linear in ξ_1 so that

$$\gamma_{1,\xi_1} = (\widetilde{\gamma}_{0,x}T_{0,y} - \widetilde{\gamma}_{0,y}T_{0,x})/J := F_1(\xi_2, t) \quad \text{as } \xi_1 \to -\infty. \tag{3.3.5e}$$

From (3.3.5d) it also follows that

$$\gamma_{1,\eta} = \widetilde{\gamma}_{1,\eta} \quad \text{as } \xi_1 \to -\infty.$$

Now since $\phi_0 \overset{\xi_1 \to \infty}{\to} \phi_f$ (which is independent of ξ_1) it follows that

$$\lim_{\xi_1 \to -\infty} \phi_{0,\xi_1} = 0.$$

We therefore conclude that

$$\lim_{\xi_1 \to -\infty} \left(d(\phi_0)\gamma_{1,\xi_1}\right)_{\xi_1} = 0 \quad \text{and} \quad \lim_{\xi_1 \to -\infty} \left(d(\phi_0)\gamma_{1,\eta}\right)_{\xi_1} = 0.$$

Combining all these limiting conditions we see that in the limit $\xi_1 \to -\infty$ (3.2.9) becomes

$$d(\phi_f)\left\{(S_{0,y} + S_{1,\eta})F_1(\xi_2, t) + \gamma_{1,\eta}\right\} = C \quad \text{(independent of } \eta).$$

Dividing by $d(\phi_f)$ and averaging over η we obtain

$$C = \langle d(\phi_f)^{-1}\rangle^{-1} S_{0,y}F_1(\xi_2, t)$$

which along with (3.3.5d) implies that

$$d(\phi_f)(S_{0,y} + S_{1,\eta})F_1(\xi_2, t) + d(\phi_f)\widetilde{\gamma}_{1,\eta} = \langle d(\phi_f)^{-1}\rangle^{-1} S_{0,y}F_1(\xi_2, t). \tag{3.3.6}$$

From (3.1.4g), $\widetilde{\gamma}_{1,\eta}$ is a known function of η so that (3.3.6) is an equation which can be used to determine the η dependence of S_1 and is known as the cell problem for S_1.

We now integrate (3.2.9) over the region $(\xi_1, \eta) \in (-\infty, \infty) \times T^1$ and use the fact that $\gamma_1(+\infty, \xi_2, \eta, t) = 0$ (which comes from matching (3.3.5a, b) in the downstream region) to obtain:

$$F_1 S_{0,x}^2 \langle d(\phi_f)\rangle + \left\langle (S_{0,y} + S_{1,\eta})\left\{d(\phi_f)(S_{0,y} + S_{1,\eta})F_1 + d(\phi_f)\widetilde{\gamma}_{1,\eta}\right\}\right\rangle + S_{0,t}\left(\langle\phi_f\rangle - \langle\phi_b\rangle\right) = 0. \tag{3.3.7}$$

We notice that the quantity { } in (3.3.7) is just the left side of (3.3.6) so that (3.3.7) is reduced to the form

$$F_1\{\langle d(\phi_f)\rangle S_{0,x}^2 + \langle d(\phi_f)^{-1}\rangle^{-1} S_{0,y}^2\} + (\langle\phi_f\rangle - \langle\phi_b\rangle)S_{0,t} = 0$$

which is the eikonal equation for S_0. Eliminating F_1 using (3.3.5e) we obtain

$$\begin{aligned}(\widetilde{\gamma}_{0,x}T_{0,y} - \widetilde{\gamma}_{0,y}T_x)\,&\frac{(\langle d(\phi_f)\rangle S_{0,x}^2 + \langle d(\phi_f)^{-1}\rangle^{-1} S_{0,y}^2)}{S_{0,x}T_{0,y} - S_{0,y}T_{0,x}}\\ &+ (\langle\phi_f\rangle - \langle\phi_b\rangle)S_{0,t} = 0.\end{aligned} \tag{3.3.8}$$

Matching inner and outer expansions for p

We follow the same procedure as in (3.3.5c) to obtain the following matching conditions for p:
$O(\delta^0) >$

$$p_0(\xi_1, \xi_2, \eta, t) = \widetilde{p}_0(x_0, y_0, t),$$

$O(\delta^1) >$

$$\begin{aligned}p_1(\xi_1, \xi_2, \eta, t) = \big(&\widetilde{p}_{0,x}^{\pm}(x_0, y_0, t)T_{0,y}/J - \widetilde{p}_{0,y}^{\pm}(x_0, y_0, t)T_{0,x}/J\big)\\ &+ \widetilde{p}_1^{\pm}(x_0, y_0, \eta, t) \quad \text{as } \xi_1 \to \pm\infty,\end{aligned}$$

where $\widetilde{p}_k^{\pm}$ represent the outer expansions in the regions $S_0 > 0$, and $S_0 < 0$, respectively. Following a similar procedure as we did with γ we obtain the following limiting conditions for p:

$$p_{1,\xi_1} = (\widetilde{p}_{0,x}^{\pm}T_{0,y} - \widetilde{p}_{0,y}^{\pm}T_{0,x})/J := F_2^{\pm}(\xi_2, t) \quad \text{as } \xi_1 \to \pm\infty \tag{3.3.9a}$$

and

$$p_{1,\eta} = \widetilde{p}_{1,\eta}^{\pm}(x_0, y_0, \eta, t) \quad \text{as } \xi_1 \to \pm\infty \tag{3.3.9b}$$

and

$$p_{0,\xi_2} = \frac{\partial}{\partial\xi_2}\widetilde{p}_0^{\pm}\big(x_0(\xi_2), y_0(\xi_2), t\big) = \big(-\widetilde{p}_{0,x}^{\pm}S_{0,y} + \widetilde{p}_{0,y}^{\pm}S_{0,x}\big)/J \quad \text{as } \xi_1 \to \pm\infty. \tag{3.3.9c}$$

We now consider (3.2.8) in the limit $\xi_1 \to \pm\infty$, make use of the limiting equations (3.3.9) and integrate with respect to η to obtain

$$(T_{0,y} + T_{1,\eta})\widetilde{p}_{0,\xi_2}^{\pm} + (S_{0,y} + S_{1,\eta})F_2^{\pm}(\xi_2, t) + \widetilde{p}_{1,\eta}^{\pm} = C^{\pm}\lambda(\phi_0)^{-1} \quad \text{as } \xi_1 \to \pm\infty$$

where $C^{\pm}$ are independent of η. Averaging over η and eliminating $C^{\pm}$ we obtain

$$\begin{aligned}&\lambda(\phi_0)\big[(T_{0,y} + T_{1,\eta})\widetilde{p}_{0,\xi_2}^{\pm} + (S_{0,y} + S_{1,\eta})F_2^{\pm}(\xi_2, t) + \widetilde{p}_{1,\eta}^{\pm}\big]\\ &\quad = \langle\lambda(\phi_0)^{-1}\rangle^{-1}\big(T_{0,y}\widetilde{p}_{0,\xi_2}^{\pm} + S_{0,y}F_2^{\pm}\big).\end{aligned} \tag{3.3.10}$$

We now integrate (3.2.8) over the region $(\xi_1, \eta) \in (-\infty, \infty) \times T^1$, use (3.3.10), and the zeroth order orthogonality condition (3.2.2) to obtain

$$\begin{aligned}
&F_2^+\left(\langle\lambda(\phi_b)\rangle S_{0,x}^2 + \langle\lambda(\phi_b)^{-1}\rangle^{-1} S_{0,y}^2\right) \\
&\quad + \widetilde{p}^+_{0,\xi_2}\left(\langle\lambda(\phi_b)\rangle S_{0,x}T_{0,x} + \langle\lambda(\phi_b)^{-1}\rangle^{-1} S_{0,y}T_{0,y}\right) \\
&= F_2^-\left(\langle\lambda(\phi_f)\rangle S_{0,x}^2 + \langle\lambda(\phi_f)^{-1}\rangle^{-1} S_{0,y}^2\right) \\
&\quad + \widetilde{p}^-_{0,\xi_2}\left(\langle\lambda(\phi_f)\rangle S_{0,x}T_{0,x} + \langle\lambda(\phi_f)^{-1}\rangle^{-1} S_{0,y}T_{0,y}\right)
\end{aligned}$$

or making use of (3.3.9a, c) we obtain

$$\begin{aligned}
&\left(\widetilde{p}^+_{0,x}T_{0,y} - \widetilde{p}^+_{0,y}T_{0,x}\right)\left(\langle\lambda(\phi_b)\rangle S_{0,x}^2 + \langle\lambda(\phi_b)^{-1}\rangle^{-1} S_{0,y}^2\right) \\
&\quad + \left(\widetilde{p}^+_{0,y}S_{0,x} - \widetilde{p}^+_{0,x}S_{0,y}\right)\left(\langle\lambda(\phi_b)\rangle S_{0,x}T_{0,x} + \langle\lambda(\phi_b)^{-1}\rangle^{-1} S_{0,y}T_{0,y}\right) \\
&= \left(\widetilde{p}^-_{0,x}T_{0,y} - \widetilde{p}^-_{0,y}T_{0,x}\right)\left(\langle\lambda(\phi_f)\rangle S_{0,x}^2 + \langle\lambda(\phi_f)^{-1}\rangle^{-1} S_{0,y}^2\right) \\
&\quad + \left(\widetilde{p}^-_{0,y}S_{0,x} - \widetilde{p}^-_{0,x}S_{0,y}\right)\left(\langle\lambda(\phi_f)\rangle S_{0,x}T_{0,x} + \langle\lambda(\phi_f)^{-1}\rangle^{-1} S_{0,y}T_{0,y}\right)
\end{aligned} \tag{3.3.11}$$

which is the jump condition for the pressure.

3.4. *The effective free boundary problem for a general layered medium*

For the sake of brevity, the asymptotic analysis required to derive the effective free boundary problem in the large solid density limit ($c_{eq}/\rho \to 0$) was presented only for the case in which the layering ahead of the front was horizontal while behind the front the medium was assumed to be homogeneous. Precisely the same procedure can be followed for the cases in which the layering is vertical and in which layering also occurs behind the front. In this subsection we present the effective free boundary problem in a general form which incorporates all of the various cases mentioned above. The general effective free boundary problem may be summarized as follows:

Upstream from the reaction front $S(x,y,t) = 0$ one has

$$\left.\begin{aligned}
&d_f\gamma_{xx} + D_f\gamma_{yy} + \lambda_f\gamma_x p_x + \Lambda_f\gamma_y p_y = 0 && (3.4.1)\\
&\lambda_f p_{xx} + \Lambda_f p_{yy} = 0 && (3.4.2)
\end{aligned}\right\} \quad S(x,y,t) < 0,$$

while downstream the concentration has reached its equilibrium concentration so that

$$\left.\begin{aligned}
&\gamma \equiv 1 && (3.4.3)\\
&\lambda_b q_{xx} + \Lambda_b q_{yy} = 0 && (3.4.4)
\end{aligned}\right\} \quad S(x,y,t) > 0.$$

At the unknown reaction interface, $S(x,y,t) = 0$, one has

$$\gamma = 1, \tag{3.4.5}$$

$$p = q, \tag{3.4.6}$$

$$(p_xT_y - p_yT_x)(\lambda_f S_x^2 + \Lambda_f S_y^2) + (p_yS_x - p_xS_y)(\lambda_f S_xT_x + \Lambda_f S_yT_y)$$
$$= (q_xT_y - q_yT_x)(\lambda_b S_x^2 + \Lambda_b S_y^2) + (q_yS_x - q_xS_y)(\lambda_b S_xT_x + \Lambda_b S_yT_y), \tag{3.4.7}$$

$$(\gamma_xT_y - \gamma_yT_x)\left(\frac{d_fS_x^2 + D_fS_y^2}{S_xT_y - S_yT_x}\right) = -S_t(\langle\varphi_f\rangle - \langle\varphi_b\rangle). \tag{3.4.8}$$

Here the averaged constants d_f, D_f, λ_f, Λ_f, λ_b, Λ_b obtained via the homogenization procedure outlined above are to be understood as follows. The "dees" (d, D) and lambdas (λ, Λ) denote appropriate averages of the functions $d(\varphi) = \varphi D(\varphi)$ and $\lambda(\varphi) = \varphi\varkappa(\varphi)$, respectively. The subscripts b and f refer to the original (unaltered, downstream) region and the final (altered, upstream) region, respectively. The lower case indicates it is averaged in the x-direction in whatever manner the homogenization dictates, while the upper case is the same for the y-direction. As an example of this notation consider the case considered in the asymptotic analysis presented above, i.e., in which the porous medium ahead of the front is layered horizontally and behind the front it is homogeneous as in Fig. 2. Then φ_f is a constant so that $\varphi_f D(\varphi_f)$ and $\varphi_f\varkappa(\varphi_f)$ are also constant resulting in $d_f = D_f$ and $\lambda_f = \Lambda_f$. In the downstream region $\lambda_b = \langle\varphi_b\varkappa(\varphi_b)\rangle$ and $\Lambda_b = \langle\varphi_b^{-1}\varkappa(\varphi_b)^{-1}\rangle^{-1}$ where the braces $\langle\ \rangle$ denote averaging over one period. In this example homogenization dictates that in the x-direction the usual, arithmetic average is to be used, while in the y-direction the harmonic average is appropriate.

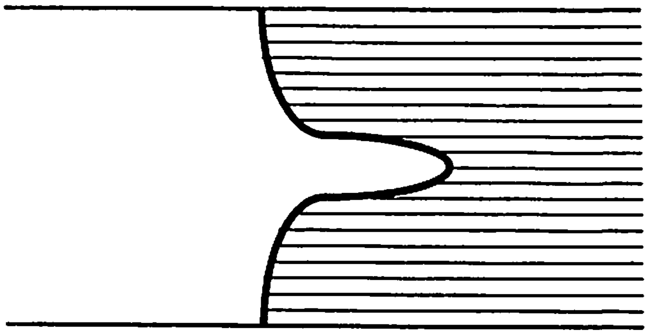

Fig. 2.

These equations are to be solved for γ in the upstream region (since $\gamma \equiv 1$ in the downstream region), p, q and S subject to the asymptotic conditions

$$\gamma = 0,\ \varphi = \varphi_f,\ p_x = p'_f \quad \text{as } x \to -\infty \tag{3.4.9}$$

and

$$\varphi = \varphi_b \quad \text{as } x \to -\infty. \tag{3.4.10}$$

Note that as $x \to +\infty$, $\gamma = 1$ automatically and q_x is to be determined as part of the solution in terms of the desiderata of the problem, especially the inlet pressure gradient p'_f. On the transverse boundaries of the aquifer, which by scaling we can take as $y = 0, \pi$ we shall impose the no-flow boundary conditions that $\gamma_y, p_y, q_y = 0$.

4. Application of effective equations: analysis of morphological instabilities in layered porous media

In this section we briefly summarize the results of the application of the effective free boundary equations to the investigation of the shape stability of the reaction interface in layered porous media. Full details of this analysis are presented in [6]. The approach follows the method used in the homogeneous case studied in references [1, 2] in which the planar solutions are computed explicitly and then a linearized stability analysis is used to examine the stability of a complete set of perturbations of the form $\cos my$. We shall obtain a formula for the spectrum of the linearized problem for each m and compare it with that derived for the homogeneous case [1, 2] in order to determine if the layering has a stabilizing or destabilizing effect.

4.1. Planar solutions

We seek planar solutions in which the normal and tangential coordinates of reaction interface are of the form

$$S(x,y,t) = x - Vt, \quad T(x,y,t) = y,$$

and the unknown functions are of the form

$$\gamma(x,y,t) = \overline{\gamma}(x - Vt), \quad p(x,y,t) = \overline{p}(x - Vt), \quad q(x,y,t) = \overline{q}(x - Vt).$$

Substituting these special functional forms into (3.4.1)–(3.4.8) and solving the resulting system of ordinary differential equations, boundary and interface conditions we obtain the following planar solution:

$$\overline{\gamma}(x - Vt) = \mathrm{e}^{\alpha(x-Vt)}, \quad \alpha = \frac{-p'_f\lambda_f}{d_f} > 0, \quad x < Vt, \tag{4.1.1}$$

$$\overline{p}(x - Vt) = p'_f(x - Vt), \tag{4.1.2}$$

$$\overline{q}(x - Vt) = \frac{p'_f}{\Gamma}(x - Vt), \quad \Gamma = \lambda_b/\lambda_f, \tag{4.1.3}$$

with the velocity of the travelling front, obtained from (3.4.8), being

$$V = -p'_f\lambda_f/\big(\langle\varphi_f\rangle - \langle\varphi_b\rangle\big) = \frac{v_f}{\langle\varphi_f\rangle - \langle\varphi_b\rangle} \tag{4.1.4}$$

where the inlet velocity $v_f = -p'_f\lambda_f$ by Darcy's law.

4.2. Linear shape instabilities

We consider small perturbations of the above planar solution of the form

$$S(x,y,t) = x - Vt + \Delta r(y,t) \tag{4.2.1}$$

where Δ, the size of the morphological disturbance, is much larger than the δ the width of the reaction front. By averaging the zeroth order orthogonality condition (3.2.2) over η we conclude that, in order to recover the planar solution as $\Delta \to 0$, the orthogonality condition up to $O(\Delta)$ is:

$$S_{0,x}T_{0,x} + s_{0,y}T_{0,y} = 0. \tag{4.2.2}$$

To fix the front at $x = 0$ and to build in the orthogonality condition (4.2.2) then we make the change of variables:

$$x' = S_0(x, y, t) = x - Vt - \Delta r(y, t), \tag{4.2.3}$$

$$y' = T_0(x, y, t) = y - \Delta x r_y(y, t). \tag{4.2.4}$$

Writing (3.4.1)–(3.4.10) in terms of the new coordinates (4.2.3), introducing perturbations $\overline{\gamma} + \Delta\gamma$, $\overline{p} + \Delta p$, $\overline{q} + \Delta q$ of the planar solutions (4.1.1)–(4.1.3), and retaining only terms up to $O(\Delta)$ we obtain the linearized versions of (3.4.1)–(3.4.10). In order to investigate the stability of these linearized equations it suffices to solve for a complete set of perturbations of the form

$$r(y, t) = e^{\sigma(m)t}\cos my, \quad \gamma(x, y, t) = \gamma_m(x)e^{\sigma(m)t}\cos my, \tag{4.2.5}$$

and similarly with p and q, where $\sigma(m)$ is the spectrum of the linearized problem. Substituting (4.2.5) into the linearized equations yields (with $' = d/dx$):

$$\left.\begin{aligned} &d_f\gamma'' + \lambda_f p_f'\gamma' - m^2 D_f\gamma - m^2 D_f\alpha e^{\alpha x} + \lambda_f\alpha e^{\alpha x}p' = 0 \quad (4.2.6)\\ &\lambda_f q'' - m^2\Lambda_f p - m^2\Lambda_f p_f' = 0 \quad (4.2.7)\end{aligned}\right\} \quad x < 0,$$

$$\lambda_b q'' - m^2\Lambda_b q - m^2\Lambda_b p_f'/\Gamma = 0, \quad x > 0, \tag{4.2.8}$$

$$\left.\begin{aligned} &\gamma = 0 \quad (4.2.9)\\ &p = q \quad (4.2.10)\\ &p' = \Gamma q' \quad (4.2.11)\\ &d_f\gamma' = \big(-\langle\varphi_f\rangle + \langle\varphi_b\rangle\big)\sigma(m) \quad (4.2.12)\end{aligned}\right\} \quad x = 0,$$

$$\gamma \to 0, \; p' \to 0 \quad \text{as } x \to -\infty, \tag{4.2.13}$$

and

$$q' \to 0 \quad \text{as } x \to +\infty. \tag{4.2.14}$$

Solving (4.2.7), (4.2.8) for p and q, and matching them at $x = 0$, we obtain

$$p = -p_f' + \frac{p_f'\left(1 - \frac{1}{\Gamma}\right)}{1 + \frac{\beta_f}{\beta_0\Gamma}} e^{|m|\beta_f x}, \quad x < 0, \tag{4.2.15}$$

where

$$\beta_f = \left(\frac{\Lambda_f}{\lambda_f}\right)^{1/2}, \quad \beta_0 = \left(\frac{\Lambda_0}{\lambda_0}\right)^{1/2}, \quad \Gamma = \frac{\lambda_0}{\lambda_f}.$$

Substituting (4.2.15) into (4.2.6) and solving for γ, we obtain

$$\gamma(x) = Ce^{\frac{\alpha+\sqrt{\alpha^2+4m^2\delta_f^2}}{2}x} - \alpha e^{\alpha x} - \alpha^2\beta_f|m|\,\frac{1-\Gamma}{\Gamma+\frac{\beta_f}{\beta_0}}\cdot e^{(\alpha+\beta_f|m|)x}\left(\alpha\beta_f|m| + m^2(\beta_f^2-\delta_f^2)\right)^{-1} \tag{4.2.16}$$

where $\delta_f = (D_f/d_f)^{1/2}$, and imposing the jump condition (4.2.9) it follows that

$$C = \alpha + \alpha^2\beta_f|m|\,\frac{1-\Gamma}{\Gamma+\frac{\beta_f}{\beta_0}}\cdot\left(\alpha\beta_f|m| + |m|^2(\beta_f^2-\delta_f^2)\right)^{-1}.$$

The jump condition (4.2.12) together with (4.2.16) yield the following expression for the spectrum $\sigma(m)$ of the linearized problem:

$$\sigma(m) = \frac{d_f}{\langle\varphi_f\rangle - \langle\varphi_0\rangle}\left[\frac{\alpha^2 - \alpha\sqrt{\alpha^2+4m^2\delta_f^2}}{2} + 2\,\frac{1-\Gamma}{\Gamma+\frac{\beta_f}{\beta_0}}\,\frac{\alpha^2\beta_f|m|}{\alpha+2\beta_f|m|+\sqrt{\alpha^2+4m^2\delta_f^2}}\right]. \tag{4.2.17}$$

We note that $\sigma(m)$ has the following properties (see Fig. 3):

$$\sigma(m)\to-\infty \text{ as } |m|\to\infty,\quad \sigma(0)=0,\quad \text{and } \sigma'(0) = \frac{1-\Gamma}{\Gamma+\frac{\beta_f}{\beta_0}}\,\alpha\beta_f > 0.$$

Solving the equation $\sigma(m_0) = 0$ we obtain after some simplification:

$$|m_0| = \frac{\beta_f\alpha}{2\delta_f}\cdot\frac{\left(\frac{\frac{\beta_f}{\beta_0}+2-\Gamma}{\Gamma+\frac{\beta_f}{\beta_0}}\right)^2 - 1}{\delta_f\cdot\left(\frac{\frac{\beta_f}{\beta_0}+2-\Gamma}{\Gamma+\frac{\beta_f}{\beta_0}}\right) + \left(\delta_f^2+\beta_f^2\left(\left(\frac{\frac{\beta_f}{\beta_0}+2-\Gamma}{\Gamma+\frac{\beta_f}{\beta_0}}\right)^2-1\right)\right)^{1/2}}. \tag{4.2.18}$$

Now by introducing the variables

$$s = \frac{\frac{\beta_f}{\beta_0}+2-\Gamma}{\Gamma+\frac{\beta_f}{\beta_0}} \geqslant 1 \quad\text{and}\quad t = \frac{\delta_f}{\beta_f} > 0$$

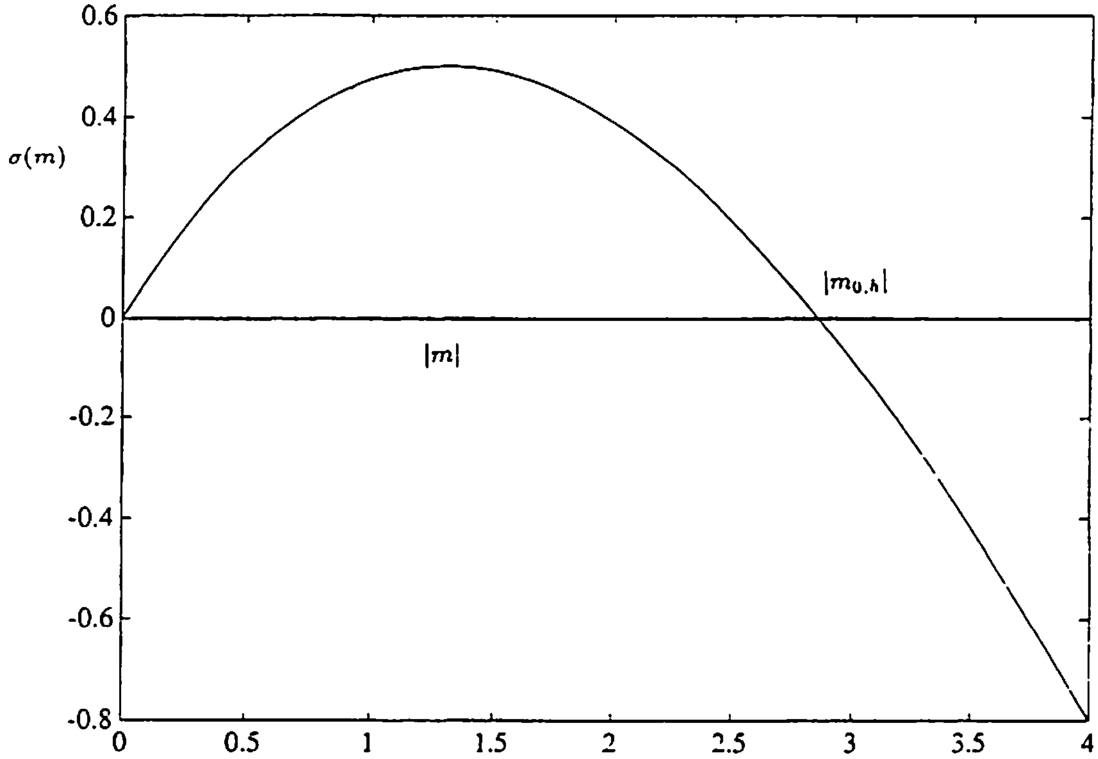

Fig. 3.

(4.2.18) can be expressed in the simplified form:

$$|m_0| = \frac{\alpha}{2\delta_f} \frac{s^2-1}{ts+\sqrt{s^2+t^2-1}} = \left(-p_f'\right) \frac{\lambda_f}{2\sqrt{D_f d_f}} \cdot \frac{s^2-1}{ts+\sqrt{s^2+t^2-1}}. \tag{4.2.19}$$

In the case that there is layering only ahead of the front, $t = 1$, and the formula reduces to

$$|m_0| = \left(-p_f'\right) \cdot \frac{\lambda_f}{2d_f} \cdot \frac{s^2-1}{2s} = \alpha \cdot \frac{(\beta_0^{-1}+1)(1-\Gamma)}{(\beta_0^{-1}+\Gamma)(\beta_0^{-1}+2-\Gamma)}. \tag{4.2.20}$$

If there is, in addition, no layering ahead of the front (i.e., the medium is homogeneous) then $\beta_0 = 1$ and formula (4.2.20) reduces the expression in [1, 2].

4.3. Specific examples of layered porous media

In this section we examine the effect of layering on the stability of planar fronts in two typical situations: layering only ahead of the front and layering ahead and behind the front with a fixed proportion of the medium dissolved out as the front passes. These results will be compared with the case of homogeneous porous media [1, 2].

a) *Layering ahead of the front*

We examine the case of horizontal layering ahead of the front as depicted in Fig. 2. The case of vertical layering ahead of the front can be treated in exactly the same manner. Behind the front the porosity is a constant, which implies $d_f = D_f =$ constant and $\lambda_f = \Lambda_f =$ constant and hence $\delta_f = \beta_f = 1$. A typical form for the permeability is $\varkappa(\varphi) = K\varphi^k$ where $K, k > 0$ so that

$$\lambda_0(\varphi) = K\varphi_0^{k+1}. \tag{4.3.1}$$

Since the layering is horizontal:

$$\beta_0^2 = \Lambda_0/\lambda_0 = \langle \varphi_0^{-(k+1)} \rangle^{-1} / \langle \varphi_0^{k+1} \rangle < \beta_{0,h}^2 = 1, \tag{4.3.2}$$

where we have used the fact that for convex functions (like x^{k+1}), the harmonic mean is less than the arithmetic mean.

Similarly, exploiting convexity we obtain the following:

$$\Gamma = \lambda_0/\lambda_f = \langle \varphi_0^{k+1} \rangle / \varphi_f^{k+1} > \langle \varphi_0 \rangle^{k+1} / \varphi_f^{k+1} = \Gamma_h. \tag{4.3.3}$$

To compare the critical value m_0 with the one for a corresponding homogeneous porous medium $m_{0,h}$, we set $\sigma(m) = 0$ in (4.2.17), which upon using $\beta_f = \delta_f = 1$ and $\alpha = -p_f' \lambda_f / d_f = \alpha_h$ yields the following equation for m_0:

$$\left(\alpha^2 + 4m^2\right)^{1/2} = \alpha + \frac{2|m|(1-\Gamma)}{(1/\beta_0 + 1)} < \alpha + |m|(1-\Gamma) < \alpha + |m|(1-\Gamma_h) \tag{4.3.4}$$

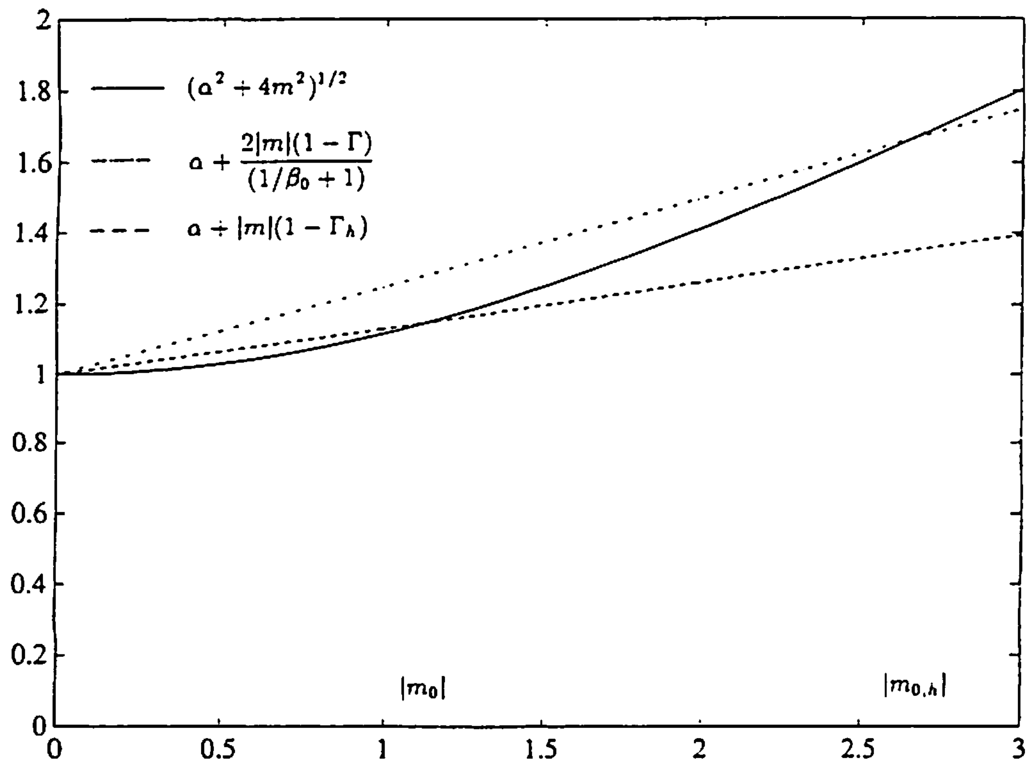

Fig. 4.

the last expression with equality being the determining equation for $|m_{0,h}|$ [1, 2]. We see that $|m_0|$ is the intersection of the branch of a hyperbola and a line (as in Fig. 4) whereas $|m_{0,h}|$ is the intersection of the same hyperbola (which is an increasing function of m) and a steeper straight line. Therefore, $|m_0| < |m_{0,h}|$ so that the horizontal layering ahead of the front stabilizes the front because the interval of unstable modes $(0, |m_0|)$ is shorter than that for the corresponding homogeneous situation $(0, |m_{0,h}|)$. By exactly the same type of analysis, one can show that vertical layering ahead of the front has a destabilizing effect.

b) *Layering ahead and behind the front*

We consider the situation in which the reactive fluid impinges on a layered porous medium dissolving out a fixed proportion at every location and leaving behind a similarly layered medium with higher porosity

$$\varphi_0(x, y) = \theta \varphi_f(x, y), \quad 0 < \theta < 1, \tag{4.3.5}$$

where θ is the fixed proportion dissolved out. Again, we shall only treat the horizontally layered situation – the similar vertically layered case can be treated analogously.

In this case

$$\beta_0^2 = \frac{\langle \varphi_0^{-(k+1)} \rangle^{-1}}{\langle \varphi_0^{k+1} \rangle} = \frac{\theta^{k+1} \langle \varphi_f^{-(k+1)} \rangle^{-1}}{\theta^{k+1} \langle \varphi_f^{k+1} \rangle} = \beta_f^2 \tag{4.3.6}$$

and

$$\Gamma = \lambda_0 / \lambda_f = \frac{\langle \varphi_0^{-(k+1)} \rangle}{\langle \varphi_f^{k+1} \rangle} = \theta^{k+1} \frac{\langle \varphi_f^{k+1} \rangle}{\langle \varphi_f^{k+1} \rangle} = \theta^{k+1} = \Gamma_h. \tag{4.3.7}$$

From these relations it follows that

$$s = \frac{\frac{\beta_f}{\beta_0} + 2 - \Gamma}{\Gamma + \frac{\beta_f}{\beta_0}} = (3 - \Gamma)/(\Gamma + 1) = s_h$$

so that from (4.2.19) it follows that the magnitude of $|m_0|$ compared to that of $m_{0,h}$ is determined by the values of t and α/δ_f in the two cases. If $t = 1 = t_h$ and $\alpha/\delta_f = \alpha_h$, then (4.2.19) reduces to the expression for the homogeneous cutoff $|m_{0,h}|$. Since the right-hand side of (4.2.19) is a decreasing function of t, it follows that if $t < t_h = 1$ (i.e., the diffusion term δ_f is smaller than the flow term β_f) then the proportional horizontal layering either side of the front has a destabilizing effect if $\alpha/\delta_f > \alpha_h$. In order to motivate these assumptions on t and α/δ_f we assume a final porosity function that is a small perturbation (with zero average) of the homogeneous porosity $\overline{\varphi}$:

$$\varphi_f = \overline{\varphi}_f + \varepsilon \widetilde{\varphi}_f = \overline{\varphi}_f (1 + \varepsilon \chi_f(x, y)) \tag{4.3.8}$$

where $\langle \widetilde{\varphi}_f \rangle = 0$ so that $\overline{\varphi}_f = \langle \varphi_f \rangle$ and $\chi_f = \widetilde{\varphi}_f / \overline{\varphi}_f$. Now assuming the phenomenological function $\lambda(\varphi) = K \varphi^{k+1}$, using the definitions for λ_f and Λ_f, expanding in Taylor series and average remembering that $\langle \chi_f \rangle = 0$ and letting p denote the second moment (i.e., $p = \langle \chi_f^2 \rangle$), we obtain the following expression for β_f:

$$\beta_f^2 = \frac{\Lambda_f}{\lambda_f} = \frac{K\overline{\varphi}_f^{k+1}\left(1 - \frac{(k+1)(k+2)}{2}\varepsilon^2 p\right)}{K\overline{\varphi}_f^{k+1}\left(1 + \frac{(k+1)k}{2}\varepsilon^2 p\right)} \approx \left(1 - (k+1)^2\varepsilon^2 p\right). \tag{4.3.9}$$

Similarly, if we assume the phenomenological function $d(\varphi) = \varphi D(\varphi) = D\varphi^{d+1}$, we obtain:

$$\delta_f^2 = \frac{D_f}{d_f} = \frac{D\overline{\varphi}_f^{d+1}\left(1 - \frac{(d+1)(d+2)}{2}\varepsilon^2 p\right)}{D\overline{\varphi}_f^{d+1}\left(1 + \frac{(d+1)d}{2}\varepsilon^2 p\right)} \approx \left(1 - (d+1)^2\varepsilon^2 p\right). \tag{4.3.10}$$

From the above, we have

$$\begin{aligned}
\frac{\alpha}{\delta_f} &= -p_f' \frac{\lambda_f}{(d_f D_f)^{1/2}} \\
&= -p_f' \frac{K\overline{\varphi}_f^{k+1}}{D\overline{\varphi}^{d+1}}\left(1 + \frac{(k+1)k}{2}\varepsilon^2 p\right)\left(1 + \frac{(d+1)d}{2}\varepsilon^2 p\right)^{-1/2} \\
&\quad \times \left(1 - \frac{(d+1)(d+2)}{2}\varepsilon^2 p\right)^{-1/2} \\
&= \alpha_h\left(1 + \frac{1}{2}\left((k+1) + d + 1\right)\varepsilon^2 p\right) \\
&> \alpha_h
\end{aligned} \tag{4.3.11}$$

and

$$\begin{aligned}
\delta_f/\beta_f &= \left(1 - (d+1)^2\varepsilon^2 p\right)^{1/2}\left(1 - (k+1)^2\varepsilon^2 p\right)^{-1/2} \\
&= \left(1 + \frac{1}{2}\left((k+1)^2 - (d+1)^2\right)\varepsilon^2 p\right)
\end{aligned} \tag{4.3.12}$$

which is smaller than the homogeneous value 1 if $d > k$; i.e., if diffusion (φ^{d+1}) is dominated by flow (φ^{k+1}). The converse ($d < k$), is more complicated since, by (4.3.11), local analysis always implies that $\alpha/\delta_j > \alpha_h$ so that the effect of proportional horizontal layering now depends on a competition between the effects of α/β_f and δ_f/β_f. Analogous results can be obtained if the layering is vertical.

5. Conclusions

We have presented a novel form of homogenization applicable in the context of free boundary problems. We considered a model of reactive flow in layered porous media in which the layering is represented by small-scale periodic structure. The homogenization technique uses a combination of the methods of geometric optics, multiple scales and matched asymptotics to derive the equations for an effective free boundary problem for the reactive flow. The effective free boundary equations are cast in terms of macroscopic variables which account for the effect of the fine-scale layering on the movement of the reaction front.

As an application of the effective free boundary equations, we summarize the analysis [6] for the spectrum of the linearized shape stability problem. This is used to compare the effect of the layered medium on the onset of instability with that for homogeneous medium with the same average *porosity/permeability* in two typical situations: i) layering ahead of the front and homogeneous behind, and ii) layering ahead and behind the front subject to the physically reasonable assumption that the layering in the altered and unaltered medium are related through a proportionality constant ($\varphi_0(x, y) = \theta\varphi_f(x, y)$).

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