STABILITY OF REACTIVE FLOWS IN POROUS MEDIA: COUPLED POROSITY AND VISCOSITY CHANGES*

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Abstract. The infiltration flow of a reactive fluid in a porous medium is investigated. The reaction causes porosity/permeability changes in the porous medium as well as viscosity changes in the fluid. The coupling of the associated reaction-infiltration and Saffman–Taylor instabilities are considered. A mathematical model for this phenomenon is given in the form of a moving free-boundary problem. The morphological instability of a planar dissolution front is demonstrated using a linear stability analysis. An unexpected simplification occurs in that the resulting fourth-order equation can be solved explicitly.

Key words. modelling geological dissolution fronts, porous media, travelling waves, moving free boundaries, bifurcation and stability, reaction-infiltration instability, Saffman–Taylor instability

AMS(MOS) subject classifications. 35R35, 35B32, 86A60, 73H

1. Introduction. If water is forced through a porous medium, the soluble component will be dissolved upstream, and the water will become saturated sufficiently far downstream. Between these extremes there is a dissolution zone (usually thin) across which the soluble mineral content—and hence the porosity/permeability—changes from its original downstream value to the altered value upstream. Note that if this reaction zone protrudes into the unaltered region at some time, the flow of the undersaturated water tends to be focused to the tip of the protrusion, because behind it (on the upstream side) the permeability is greater than in the neighboring regions. Thus dissolution is enhanced at the tip of the protruding zone, and it therefore advances more rapidly than the rest of the zone. This is the reaction—infiltration instability. On the other hand, diffusion from the sides of the tip raises the concentration of the dissolved solute that is focusing at the tip and hence will decelerate its advancement. The competition between these two mechanisms will lead either to restabilization to a more complicated dissolution zone (fingering), or to the original planar zone. This phenomenon was modelled and studied mathematically by us in a recent set of papers [1], [2].

In this note, we allow for the possibility that the solute can increase the viscosity of the fluid when it dissolves in the water. This process must certainly contribute in an important way in the secondary recovery of oil (by acidifying the field to uniformly increase permeability and hence the yield) or in the leaching of tar sand reserves. Since the water is solute-free at the inlet and concentrated far downstream, this corresponds to the unstable situation of forcing a less viscous fluid into one with a higher viscosity [3]-[5]. On the other hand, the present phenomenon should be distinguished from these Saffman–Taylor-like instabilities because here it is coupled and interacting with the more fundamental reaction-infiltration instability. Chemical reactions, which are not present in the Saffman–Taylor situation, are central here, causing the instability through porosity/permeability changes. As a result, the front velocity is slower than the fluid velocity in contrast to the Saffman–Taylor situation in which they are identical.

In § 2, we present a mathematical model for the coupling of the above phenomena, which in the geochemically relevant limit of large solid density asymptotics reduces to a moving free-boundary problem. In § 3, the planar solution is given and the linearization of the problem with respect to this solution is obtained. This problem can be written as a fourth-order, linear, variable coefficient ordinary differential equation that, quite surprisingly, can be solved explicitly in terms of Whittaker or Kummer functions. An explicit form can thus be obtained for the spectrum, allowing for comparisons with previous work that did not include viscosity variations and for precise determination of the intervals of instability and the most unstable modes.

2. The mathematical model. A complete derivation of the nonlinear partial differential equations that model the above phenomenology can be found in §§ 2 and 3 of [1]. For completeness, we state these model equations. In terms of the space-time functions φ, c, and p– the porosity, solute concentration, and pressure, respectively, the governing equations are:

$$(2.1a) \qquad \frac{\partial(c\varphi)}{\partial t} = \nabla \cdot \left[\varphi D(\varphi)\nabla c + c\varphi \frac{\kappa(\varphi)\nabla p}{\mu(c)} \right] + \rho \frac{\partial\varphi}{\partial t},$$

$$(2.1b) \qquad \frac{\partial\varphi}{\partial t} = +kG(\varphi, c),$$

$$(2.1c) \qquad \nabla \cdot \left[\varphi \frac{\kappa(\varphi)}{\mu(c)} \nabla p \right] = \frac{\partial\varphi}{\partial t}.$$

Here the diffusion coefficient $D(\varphi)$ and the permeability $\kappa(\varphi)$ depend on the porosity, and $G(\varphi, c)$ is the reaction rate. These functions are left completely general to stress that none of the subsequent analysis depends on their specific form. The constants k and ρ are the dissolution rate constant and the molar density of the dissolved solid. In these equations, we have used Darcy's law for the velocity in the form

$$(2.2) \qquad \underline{v} = -\frac{\kappa(\varphi)}{\mu(c)} \nabla p,$$

where the viscosity μ depends on the concentration of the dissolved solute. In the following analysis, we have chosen a fairly typical form (see, e.g., [5]) for this dependence,

$$(2.3) \qquad \mu(c) = A e^{\Lambda c}.$$

Problem (2.1) is completed by imposing the following boundary conditions:

$$(2.4a) \qquad c = 0, \quad \varphi = \varphi_f, \quad \frac{\partial p}{\partial x} = p_f' \quad \text{as } x \to -\infty,$$

$$(2.4b) \qquad c = c_{eq}, \quad \varphi = \varphi_0 \quad \text{as } x \to +\infty.$$

At the inlet $(x = -\infty)$, (2.4a) indicates that the water is solute free and that the flow is driven by a pressure gradient in the x-direction. Furthermore, all of the soluble mineral has been previously dissolved, and the porous medium has reached its final porosity φ_f. Far downstream $(x = +\infty)$, the water has reached its equilibrium concentration of the solute c_{eq}. Since this precludes any reaction with the porous medium, it remains at its initial unaltered porosity φ_0. Note that the downstream pressure gradient (or equivalently the downstream velocity) must be determined as part of the solution. In the following we take $\varphi_0, \varphi_f, p_f'$ to be constant to stress that instabilities and

subsequent pattern selection (shapes of the fronts of porosity/permeability change) can result from this simplest, homogeneous (unpatterned) situation. This model differs from our previous work [1], [2] in that it allows for concentration-dependent viscosity (2.3), which had previously been taken constant to focus exclusively on the then newly discovered reaction-infiltration instability. Here we investigate the effect of the competition of this instability with the more widely known (see [3]-[5] and references therein) viscosity-change instability, which occurs when a less viscous fluid ($\mu_0 = \mu(0) = A$) is being driven into a more viscous fluid ($\mu_f = \mu(c_{eq}) = A\, e^{\Lambda c_{eq}}$).

In typical geological situations, the ratio of the equilibrium solute concentration to the density is quite small. This leads to a very narrow reaction zone since the water becomes saturated quite rapidly as it comes into contact with the solid. Since it is the shape of this reaction zone, and not its internal structure, which is of interest, it is reasonable to study the problem (2.1)-(2.4) in the limit $c_{eq}/\rho \to 0$, the so-called solid density asymptotic limit. Matched asymptotics to order $O(\sqrt{c_{eq}/\rho})$ results (see [1, § 4]) in a Stefan-like moving free-boundary problem for the dissolution interface in the slow time variable $t_{new} = (kc_{eq}/\rho)t_{old}$. If the further standard scalings are made,

$$x' = (v_f/D_f)x, \quad y' = (v_f/D_f)y, \quad t' = (v_f^2/D_f)t$$

$$\gamma = c/c_{eq}, \quad v' = v/f_f, \quad \mu' = \mu/\mu_f$$

$$p' = (k_f/\mu_f)p, \qquad R'(y', t) = (v_f/D_f)R(y, t),$$

then, dropping the primes, the resulting problem can be written in the following convenient form:

(2.5a) $$\left. \begin{array}{l} \Delta\gamma - v \cdot \nabla\gamma = 0, \\[2mm] v = -\dfrac{\nabla p}{\mu(\gamma)}, \\[2mm] \nabla \cdot v = 0, \end{array} \right\} \quad \text{in } x < R(y, t),$$

(2.5b)

(2.5c)

and

(2.6a) $$\left. \begin{array}{l} \gamma \equiv 1, \\[2mm] \Delta q = 0, \end{array} \right\} \quad \text{in } x > R(y, t).$$

(2.6b)

Here $x = R(y, t)$ is the location of the unknown dissolution interface across which the porosity changes discontinuously; i.e.,

(2.7) $$\varphi = \begin{cases} \varphi_f, & x < R(y, t), \\ \varphi_0, & x > R(y, t). \end{cases}$$

For notational convenience, the pressure behind and ahead of the front is denoted by p and q, respectively. The asymptotic analysis ([1, Appendix]) reveals that the boundary conditions at the interface are:

(2.8a) $$\left. \begin{array}{l} \gamma = 1, \\[2mm] p = q, \\[2mm] \dfrac{\partial p}{\partial n} = \Gamma\dfrac{\partial q}{\partial n}, \\[2mm] \dfrac{\partial\gamma}{\partial n} = (1 - \varphi_0/\varphi_f)R_t/(1 + R_y^2)^{1/2} \end{array} \right\} \quad \text{on } x = R(y, t),$$

(2.8b)

(2.8c)

(2.8d)

where $\partial/\partial n$ is the normal derivative at the surface (into the unaltered zone) and $\Gamma = \varphi_0\kappa_0/\varphi_f\kappa_f$ with κ_0 and κ_f being, respectively, the initial and final permeabilities of

the medium. These equations are to be solved for γ (behind the front, since $\gamma \equiv 1$ ahead of the front), p, q, and R subject to the asymptotic conditions

$$(2.9a) \qquad \gamma = 0, \qquad \frac{\partial p}{\partial x} = -1, \qquad \text{for } x \to -\infty,$$

$$(2.9b) \qquad \frac{\partial q}{\partial x} = ?, \qquad \text{for } x \to +\infty,$$

where the downstream pressure gradient is to be determined as part of the solution. On the transverse boundaries, which can be taken at $y = \pm\pi$ by scaling, zero-flux conditions are used throughout.

Note that $\Gamma = \varphi_0\kappa_0/\varphi_f\kappa_f$ is a measure of the porosity/permeability change, while $\mu_f/\mu_0 = e^\Lambda$ is a measure of the maximum viscosity change. In this problem, they have decoupled, allowing us to study the effects of the interaction of these two phenomena on the shape stability of the reaction interface. Normally, they appear combined through the mobility ratio $(\varphi_0\kappa_0/\mu_0)/\varphi_f\kappa_f/\mu_f$. The factor $(1 - (\varphi_0/\varphi_f))$, on the other hand, in the Stefan condition (2.8d) will not effect the determination of which modes are unstable (the sign of the spectrum of the linearized problem) but will adjust the strength of their instability (amplitude of the spectrum), since it can be removed with a rescaling of time in (2.8d).

3. Shape stability of the dissolution interface. In this section we give the planar, travelling wave solution of problem (2.5)-(2.9) and examine its linear stability to a complete set of perturbations of the form $\cos my$, $m = 0, 1, 2, \cdots$. These form a complete set because the channel has finite width 2π with zero flux boundary conditions. If a is the velocity of the front, it is straightforward to check that, in a coordinate system moving along with the front, $x' = x - at$ (and dropping primes), the travelling wave planar solution is

$$(3.1a)$$
$$(3.1b) \qquad \bar\gamma(x) = \begin{cases} e^x, & x < 0 \\ 1, & x > 0, \end{cases}$$

$$(3.2a) \qquad \bar p(x) = \int_x^0 \mu(\bar\gamma(\xi))\, d\xi, \qquad x < 0,$$

$$(3.2b) \qquad \bar q(x) = -e^\Lambda \Gamma^{-1} x, \qquad x > 0,$$

and the velocity of the front is

$$(3.3) \qquad a = (1 - \varphi_0/\varphi_f)^{-1}.$$

From the scaled version of Darcy's law (2.5b), we obtain the fluid velocity to be in the positive x-direction of magnitude

$$(3.4a)$$
$$(3.4b) \qquad v = \begin{cases} 1, & x < 0, \\ \Gamma^{-1} & x > 0. \end{cases}$$

Thus the front's velocity is adjusted to accommodate the porosity change, and the region of changing viscosity is carried along behind it in a region where the fluid velocity is the same as that at the inlet. This situation, therefore, is different from the classical Saffman-Taylor instability [3]-[5].

Denoting the perturbation of the planar front by $r(y, t) = R(y, t) - at$, we consider perturbations of the form

(3.5a) $\gamma(x, y, t) = \bar{\gamma}(x) + \delta\gamma_1(x)\, e^{\sigma t} \cos my,$

(3.5b) $p(x, y, t) = \bar{p}(x) + \delta p_1(x)\, e^{\sigma t} \cos my,$

(3.5c) $q(x, y, t) = \bar{q}(x) + \delta q_1(x)\, e^{\sigma t} \cos my,$

(3.5d) $r(y, t) = 0 + \delta\, e^{\sigma t} \cos my,$

where δ is small. This perturbation problem can be transformed to a more standard form by the change of variables $x' = x - at - r(y, t)$. The free boundary then becomes fixed at $x' = 0$ so that the linearized versions of the boundary conditions (2.8) are the same, while equations (2.5) become horribly nonlinear, involving $r(y, t)$. The method of linearizing such equations is straightforward though tedious. The resulting linearized equations (with d/dx denoted by a prime and with the subscript 1 deleted) are (see [2, §§ 2 and 3] for more details)

(3.6a) $\gamma'' - \gamma' - m^2\gamma + \Lambda\bar{\gamma}'\gamma + \bar{\gamma}'\, e^{-\Lambda\bar{\gamma}}p' + m^2\bar{\gamma}' = 0, \qquad x < 0,$

(3.6b) $p'' - \Lambda\bar{\gamma}'p - m^2p - \Lambda\bar{p}'\gamma + m^2\bar{p}' = 0, \qquad x < 0,$

(3.6c) $q'' - m^2q - m^2\alpha\Gamma^{-1} = 0, \qquad x > 0,$

subject to the interface and asymptotic conditions,

(3.7) $\gamma = 0, \quad p = q, \quad p' = \Gamma q', \quad \gamma' = (1 - \varphi_0/\varphi_f)\sigma, \quad \text{on } x = 0,$

(3.8a) $\gamma \to 0, \qquad p' \to 0, \quad \text{as } x \to -\infty,$

(3.8b) $\gamma \to 0, \qquad q' \to 0, \quad \text{as } x \to +\infty.$

It is convenient and simpler to write these equations in terms of velocity rather than pressure using the linearized versions of (2.5b) and (2.5c). Specifically, writing the x-component of the velocity as

(3.9a) $u(x, y, t) = 1 + \delta u_1(x)\, e^{\sigma t} \cos my, \qquad x < 0,$

(3.9b) $w(x, y, t) = \Gamma^{-1} + \delta w_1(x)\, e^{\sigma t} \cos my, \qquad x > 0$

with δ small, (3.6)–(3.8) are replaced by (again dropping the subscript 1)

(3.10a) $\gamma'' - \gamma' - m^2\gamma - \bar{\gamma}'u + m^2\bar{\gamma}' = 0, \qquad x < 0,$

(3.10b) $u'' + \Lambda\bar{\gamma}'u' - m^2u - m^2\Lambda\gamma + \Lambda m^2\bar{\gamma}' = 0, \qquad x < 0,$

(3.10c) $w'' - m^2w = 0, \qquad x > 0$

with the interface conditions,

(3.11a) $\gamma = 0,$

(3.11b) $u = \Gamma w,$

(3.11c) $u' = w' + \dfrac{(1 - \Gamma)}{\Gamma}\, m^2,$ $\left.\begin{array}{c} \\ \\ \\ \\ \end{array}\right\}$ on $x = 0,$

(3.11d) $\gamma' = (1 - \varphi_0/\varphi_f)\sigma,$

and the asymptotic conditions

(3.12a) $\gamma \to 0, \qquad u \to 0, \quad \text{as } x \to -\infty,$

(3.12b) $\gamma \to 0, \qquad w \to 0, \quad \text{as } x \to +\infty.$

If $\Lambda = 0$ (no viscosity change), (3.6) and (3.10) decouple and can be solved as a sequence of second-order, constant coefficient equations giving the previous results [1], [2] for the porosity/permeability change instability. If $\Lambda \neq 0$, (3.10a) and (3.10b) are a coupled pair of variable coefficient, second-order equations that can be most conveniently treated by eliminating γ to obtain a fourth-order, variable coefficient equation for u. Specifically, solving (3.10c) for $w(x) = A e^{-|m|x}$ and using this in the interface conditions (3.11b) and (3.11c) we obtain (with d/dx written as D)

$$(3.13) \qquad (D^2 - D - m^2)(D^2 - m^2)u + \Lambda e^x (D+1)(D^2 - m^2)u = 0, \qquad x < 0$$

with the interface conditions

$$(3.14a) \qquad u' = -|m|u/\Gamma + m^2(1-\Gamma)/\Gamma, \qquad x = 0,$$

$$(3.14b) \qquad u'' = \Lambda|m|(u - |m|)/\Gamma + m^2 u, \qquad x = 0,$$

and the asymptotic conditions

$$(3.15) \qquad u, u'' \to 0, \qquad \text{as } x \to -\infty.$$

The fact that (3.13) factors as indicated allows us to obtain an analytical solution rather than being forced to resort to a numerical treatment at this stage. Specifically, letting $z = (D^2 - m^2)u$ and making the change of variables $t = \Lambda e^x$ we obtain (with a super dot indicating d/dt)

$$(3.16) \qquad \ddot{z} + \dot{z} + \left(\frac{1}{t} - \frac{m^2}{t^2}\right)z = 0, \qquad 0 < t < \Lambda.$$

The solution of this equation can be written in terms of Whittaker or Kummer functions (e.g., by writing it in terms of $y(t) = e^{t/2}z(t)$ and using formulae 13.1.31–13.1.33 of [6, p. 505]). That is,

$$(3.17) \qquad z(t) = e^{-t}t^{\nu+1/2}[C_3 M(\nu - \tfrac{1}{2}, 1 + 2\nu, t) + C_4 U(\nu - \tfrac{1}{2}, 1 + 2\nu, t)],$$

where $\nu = (m^2 + 1/4)^{1/2}$. Now (3.15) implies that $z(0) = 0$. Using the asymptotic behavior of U as $t \to 0$ ([6, p. 508, 13.5.6]) we find that $C_4 = 0$. Solving $(D^2 - m^2)u = z$ using variation of parameters and the fact (3.15) that $u(t = 0) = 0$, we find

$$(3.18) \qquad u(t) = C_1 t^{|m|} + \frac{C_3}{2|m|} \int_0^t (t^{|m|}s^{-(|m|+1)} - t^{-|m|}s^{|m|-1})z(s) \, ds.$$

The constants C_1 and C_3 can be determined from the conditions (3.14) most conveniently by writing them in the form

$$(3.19a) \qquad z = \Lambda|m|(u - |m|)/\Gamma,$$
$$(3.19b) \qquad \dot{u} = |m|(|m|(1-\Gamma) - u)/\Gamma\Lambda.$$
$$\left.\right\} \text{on } t = \Lambda$$

Indeed only C_3 is required because the object of our study, the spectrum of the linearized problem, can be expressed using (3.11d) and (3.10b) as

$$\sigma(m) = [\dot{z}(\Lambda) + z(\Lambda)(1 + |m|\Gamma/\Lambda - 1/\Lambda) + |m|^3]/m^2(1 - \varphi_0/\varphi_f)$$

$$(3.20) \qquad = (1 - \varphi_0/\varphi_f)^{-1}C_3 m^{-2} e^{-\Lambda}\left[\Lambda^{\nu-1/2}M(\nu - \tfrac{1}{2}, 1 + 2\nu, \Lambda)(\nu - \tfrac{1}{2} + |m|\Gamma)\right.$$

$$\left. + \frac{\Lambda^{\nu+1/2}}{2}\left(\frac{\nu - 1/2}{\nu + 1/2}\right)M(\nu + \tfrac{1}{2}, 2 + 2\nu, \Lambda)\right] + |m|,$$

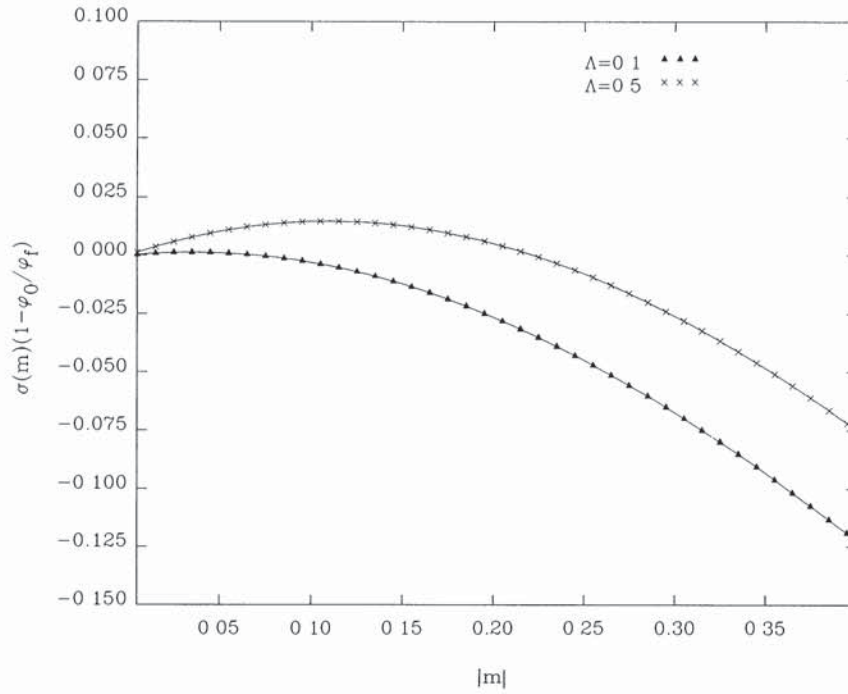where the Kummer functions and their integrals can be conveniently evaluated using [7].

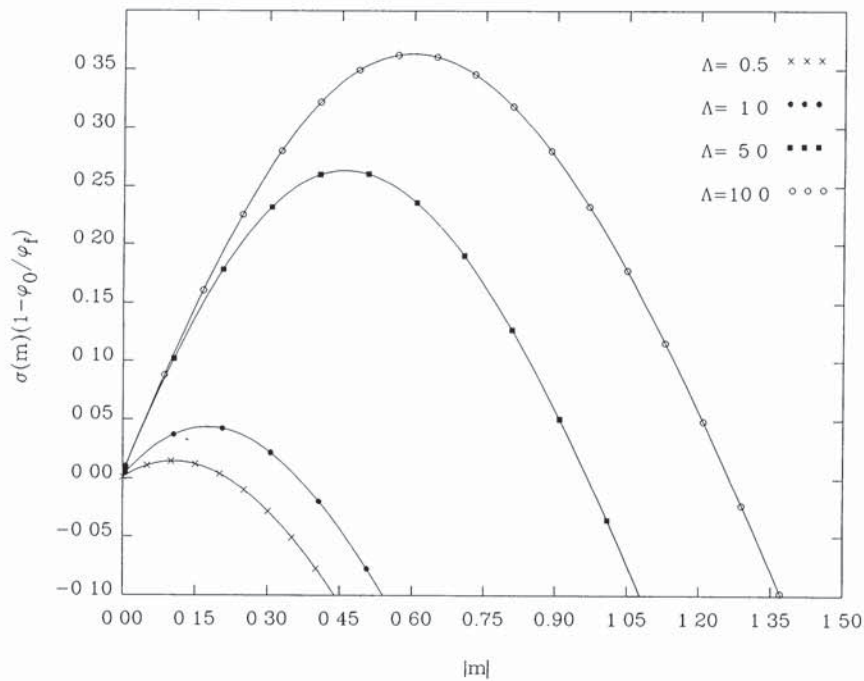FIG. 1(a). *Graph of dispersion relation for* $\Gamma = 0.95$, *with* $\Lambda = 0.1$ *and* 0.5.

FIG. 1(b). *Graph of dispersion relation for* $\Gamma = 0.95$, *with* $\Lambda = 0.5$ *to* 10.0.
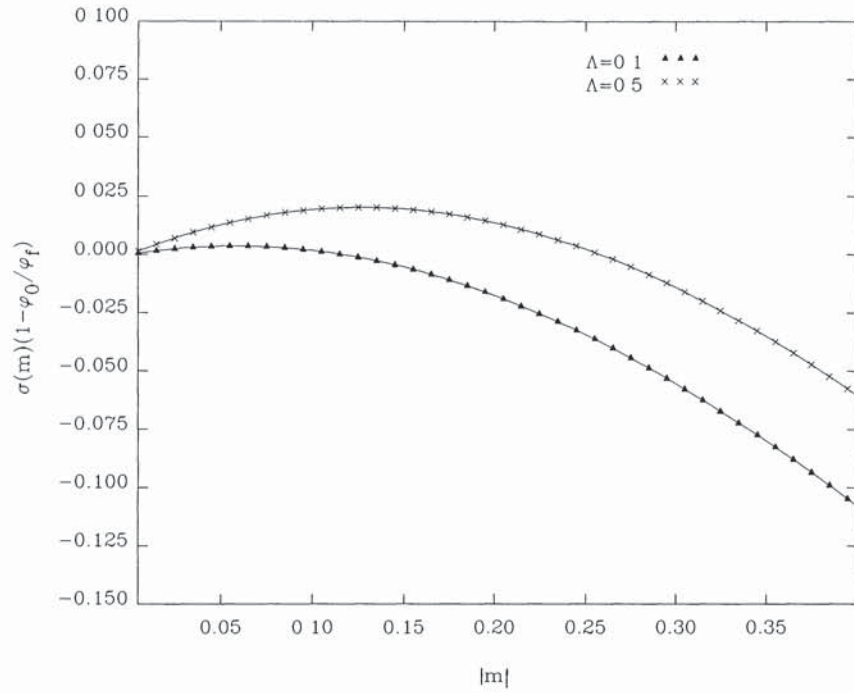
FIG. 2(a). *Graph of dispersion relation for* $\Gamma = 0.85$, *with* $\Lambda = 0.1$ *and* 0.5.
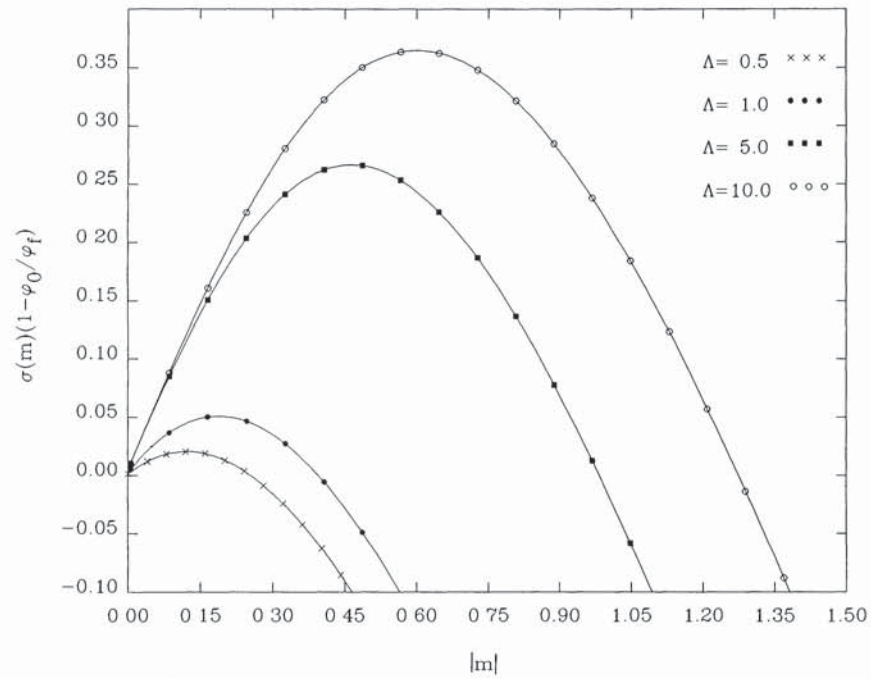

FIG. 2(b). *Graph of dispersion relation for* $\Gamma = 0.85$, *with* $\Lambda = 0.5$ *to* 10.0.

Several graphs of $\sigma(m)$ are given for various values of Γ and Λ that show that the instability is enhanced in the same qualitative way as Γ becomes smaller and Λ becomes greater. The sequence in Figs. (1a) and (1b) shows that for fixed $\Gamma = .95$ the interval of instability grows as Λ increases. Similar conclusions follow for $\Gamma = .85$ from Figs. (2a) and (2b). Indeed from the calculations used to generate Fig. (2b), we find, for example, that $|m_0|$ (the value where $\sigma(m)$ changes sign) is .39 for $\Lambda = 1$, .975 for $\Lambda = 5$, and 1.275 for $\Lambda = 10$. By comparing Fig. (1a) with (2a) and Fig. (1b) with (2b) we see the same behavior for various Λ's as Γ decreases from 1. For $\Lambda \to 0+$ in (3.20) we recover the results of (3.15) in [2]:

$$(3.21) \qquad\qquad |m_0| = \frac{2(1-\Gamma)}{(3-\Gamma)(1+\Gamma)}.$$

The limit $\Gamma \to 1-$ (i.e., no porosity/permeability change) is a singular limit, which requires including nonstationary diffusion. The calculation of the spectrum (3.20) is a nonlinear problem that requires numerical treatment. This analysis will be presented elsewhere and the results compared with the related results of Tan and Homsy [5].

4. Conclusions. A mathematical model is proposed that incorporates not only the porosity/permeability changes that occur when a reactive fluid infiltrates a porous medium, but also the possible viscosity changes that might result when the solid dissolves in the fluid. In a geochemically relevant limit, that is, the large solid density limit when $c_{eq/\rho} \to 0$, the problem reduces to a moving free-boundary problem. In this framework, the problem of the shape stability of the dissolution interface can be solved analytically. As one might expect on physical grounds, the number of modes that lose stability increases as the porosity/permeability change increases and as the viscosity change increases. We find that the spectrum of the linearized problem is linear with slope 1 at $|m| = 0$ and tends linearly to $-\infty$ as $|m| \to \infty$. The interval of instability and the most unstable mode can be determined with arbitrary precision.

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