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Stability of an acid front moving through porous rock

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As an acid flows through porous rock, it etches the rock and so increases the permeability. This propagating reaction front suffers an instability, rather like the viscous fingering instability, in which the acid prefers to follow high-permeability channels which it has already etched. We have examined the linear stability, obtaining analytic results for small and large wavenumbers and for small variations of the permeability, and obtaining numerical results in other cases.

1. Introduction

During the drilling of oil wells, drilling muds can invade the oil-bearing porous rock and then later impede the production of oil. It is also possible for drilling fluids to react with the original formation fluids and throw a precipitate which blocks the constrictions joining the pores, thereby decreasing the permeability by as much as a thousandfold. Further, some porous sandstones have accumulated over geological times similar blockages with an internal coatings of clays. A common treatment of such damaged wells is to 'stimulate' them by pumping down acid which dissolves away the blockage. In sandstones the rock itself does not dissolve, and so the porosity hardly changes as the permeability increases dramatically. In carbonate rocks, the acid (or even fresh water over geological times) does dissolve the rock, thus increasing the porosity. Williams, Gidley & Schechter (1979) and McCleod (1984) have reviewed the history of the 'acid treatment' of oil fields.

Many papers have been written on the kinetics of the reactions between the acids, e.g. HCl and HCl/HF mixtures, and the minerals, e.g. calcite, dolomite, albite and microcline, see for example Lund, Fogler & McCune (1973), Lund *et al.* (1975, 1976). Lund & Fogler (1976) have studied a mathematical model which predicts the steady movement of the acid front through sandstone cores by applying mass balances for the acid and for the dissoluble minerals both lumped into single quantities. Two parameters characterize the acidization: a *Damköhler length* which is the distance the acid is advected before it is spent, and an *acid capacity number* which is the ratio of the acid present in the pore space to that required to dissolve all the dissoluble mineral in the corresponding solid space. The results of this model are in good agreement with the experimental measurements for the concentration of the effluent acid and for the time for the front to move completely through the core.

The linear stability of the moving acid front has been investigated by Sherwood (1987). He used a chemical reaction law which depended only on the concentration of acid and was independent of the minerals. He also used an unbounded permeability law, inversely proportional to the concentration of the dissoluble minerals. Sherwood showed that the flow was unstable and that for his reaction and permeability laws the growth rate increased logarithmically with wavenumber of wavelengths shorter than the thickness of the reaction front.

In this paper we shall examine the linear stability of the moving acid front using the standard chemical reaction rate proportional to the product of the concentrations of acid and of dissoluble minerals. We must assume that the acid is well mixed within a single pore, although we shall ignore the large-scale hydrodynamic dispersion of the acid which might lead to a dampening of the shorter wavelengths. We also must assume that the surface area for the dissolution of the minerals is proportional to the volume of the minerals. Note that the area of the surface reaction would be independent of the volume of the minerals if the minerals formed a smooth flat coating, and would vary faster than the volume if they were thickly coated to the inside of a circular pipe. Two different bounded examples of the dependence of the permeability on the minerals are considered. We obtain analytic results for the linear growth rate for small variations of the permeability and for small and large wavenumbers. Like Sherwood (1987) we find the flow to be unstable, but for our reaction law we find that the growth rate increases to a limiting value at short wavelengths. The growth rate is larger when the permeability changes are larger.

The stability of a moving acid front has also been studied by Chadam *et al.* (1986). They used more complex reaction equations and included diffusion of the acid as well as significant changes in the porosity. Making a thin-reaction-front approximation, they were not able to examine disturbances with wavelengths comparable with the thickness of the reaction front; wavelengths which we find to be the most unstable. They also include a full numerical simulation of the fingering.

2. The governing equations

Let the concentration of the acid be $c(x, y, t)$ per unit volume of pore space and let the concentration of the dissoluble minerals be $w(x, y, t)$ per unit volume of the solid space. Here we are lumping the complicated mixtures of acids and minerals into two single species. Let the volume flux of the acid per unit area of the porous medium (the 'superficial velocity') be $\boldsymbol{u}(x, y, t)$. We assume that the porosity ϕ of the porous rock does not change significantly. On the other hand, we assume that the permeability is a strong function of the concentration of the dissoluble minerals, $\kappa(w)$. Finally we assume that chemical reaction has a rate constant k and a stoichiometric constant ν. The reaction equations are then

$$\phi\frac{\partial c}{\partial t}+\nabla\cdot\boldsymbol{u}c = -kwc = \nu(1-\phi)\frac{\partial w}{\partial t}.$$

Far upstream the acid concentration tends to its constant input value and the minerals are all dissolved,

$$c\rightarrow c_0 \quad \text{and} \quad w\rightarrow 0 \quad \text{as} \quad x\rightarrow -\infty.$$

Far downstream all the acid is spent and none of the minerals are dissolved,

$$c\rightarrow 0 \quad \text{and} \quad w\rightarrow w_0 \quad \text{as} \quad x\rightarrow +\infty.$$

Finally the fluid motion through the porous rock is governed by Darcy's equations

$$\boldsymbol{u} = -\frac{\kappa(w)}{\mu}\nabla p$$

and
$$\nabla\cdot\boldsymbol{u}=0,$$
where p is the pressure in the fluid and μ is the fluid viscosity.

We start by recovering the solution for the steadily moving plane reaction front. Let the acid be flowing along the x-axis at speed U and let the front be moving at a speed V along the x-axis. Thus we seek a solution of the form
$$c=c(x-Vt)\quad\text{and}\quad w=w(x-Vt).$$
The governing reaction equations then become
$$-\phi Vc'+Uc'=-kcw=-\nu(1-\phi)\,Vw'.$$
Integrating once and using the upstream condition, we have
$$(U-\phi V)\,(c-c_0)=-\nu(1-\phi)\,Vw.$$
Applying the downstream condition, we find
$$(U-\phi V)\,c_0=\nu(1-\phi)Vw_0.$$
This gives the speed of the propagating reaction front
$$V=\frac{U}{\phi+\alpha^{-1}},$$
where
$$\alpha=\frac{c_0}{\nu(1-\phi)\,w_0}$$
is the *acid capacity number*. This is the ratio of the input concentration of the acid to the concentration of acid required in a pore space to dissolve all the minerals in the corresponding solid space. Note that the velocity of the front is always less than U/ϕ which is the speed the acid moves through the pore space. Typically the acid capacity is small (Fogler & McCune 1976 gives values of about 0.1 for dissolving minerals from sandstone), and so the front moves much slower that the acid, because much acid is required to dissolve the minerals.

Substituting V back into the reaction equations, we obtain the profiles of the reaction front
$$w=w_0\frac{e^{\beta x-Vt}}{1+e^{\beta x-Vt}}\quad\text{and}\quad c=c_0\frac{1}{1+e^{\beta x-Vt}},$$
where the Damköhler thickness of the reaction front is given by
$$\beta^{-1}=\frac{V}{kw_0\alpha}=\frac{U\nu(1-\phi)}{k(\phi c_0+\nu(1-\phi)\,w_0)}.$$
Fogler & McCune (1976) give values corresponding to 5 cm.

It is convenient to non-dimensionalize the problem, scaling c with c_0, w with w_0, $\boldsymbol{u}$ with V, x and y with β^{-1}, and t with $1/\beta V$. With these scalings the reaction equations become
$$\alpha\left(\phi\frac{\partial c}{\partial t}+\nabla\cdot\boldsymbol{u}c\right)=-wc=\frac{\partial w}{\partial t}.$$

We now look at small disturbances to the above steady solution which have a y-wavenumber k and a growth rate λ and which are moving with the reaction front. Thus introducing a moving coordinate $X = x-t$, we consider solutions of the form

$$\begin{aligned}
c &= \frac{1}{1+\mathrm{e}^X} + \mathrm{e}^{\lambda t+\mathrm{i}ky}\tilde{c}(X),\\
w &= \frac{\mathrm{e}^X}{1+\mathrm{e}^X} + \mathrm{e}^{\lambda t+\mathrm{i}ky}\tilde{w}(X),\\
u &= \phi+\alpha^{-1} + \mathrm{e}^{\lambda t+\mathrm{i}ky}\tilde{u}(X),\\
v &= 0 + \mathrm{e}^{\lambda t+\mathrm{i}ky}\tilde{v}(X),\\
p &= \int^X \frac{\mu(\phi+\alpha^{-1})}{\kappa(\mathrm{e}^X/(1+\mathrm{e}^X))}\,\mathrm{d}X + \mathrm{e}^{\lambda t+\mathrm{i}ky}\tilde{p}(X).
\end{aligned}$$

Substituting into the reaction equations and linearizing, we obtain

$$\alpha\phi\lambda\tilde{c} + \tilde{c}' - \alpha\frac{\mathrm{e}^X}{(1+\mathrm{e}^X)^2}\tilde{u} = -\frac{\tilde{c}\,\mathrm{e}^X+\tilde{w}}{1+\mathrm{e}^X} = \lambda\tilde{w} - \tilde{w}'.$$

The Darcy flow equations give

$$\begin{aligned}
\tilde{u} &= -\frac{\kappa}{\mu}\tilde{p}' + (\phi+\alpha^{-1})\frac{\mathrm{d}\kappa/\mathrm{d}w}{\kappa}\tilde{w},\\
\tilde{v} &= -\frac{\kappa}{\mu}\mathrm{i}k\tilde{p},\\
\tilde{u}' &+ \mathrm{i}k\tilde{v} = 0.
\end{aligned}$$

Eliminating $\tilde{p}$ and $\tilde{v}$, these give a vorticity equation

$$\kappa\left(\frac{1}{\kappa k^2}\tilde{u}'\right)' - \tilde{u} = -(\phi+\alpha^{-1})\frac{\mathrm{d}\kappa/\mathrm{d}w}{\kappa}\tilde{w},$$

where κ and $\mathrm{d}\kappa/\mathrm{d}w$ are evaluated with the steady solution $w = \mathrm{e}^X/(1+\mathrm{e}^X)$.

3. Small changes in permeability

Let the permeability vary little with the concentration of the minerals:

$$\kappa = \kappa_0(1-\delta w) \quad \text{with} \quad \delta \ll 1.$$

(in dimensional form, $\kappa = \kappa_0(1-\delta w/w_0)$). The minus sign is because we expect the permeability to be decreased by the presence of the minerals. We shall seek an expansion in the small parameter δ.

Now the instability is driven by the right-hand side of the vorticity equation, which is $O(\delta)$. So we expect to find a flow and a growth rate also $O(\delta)$. For the $O(1)$ terms in the concentrations, we therefore need a neutrally stable solution with no flow. Fortunately a simple candidate can be obtained by noting that the above steady solution had its origin set arbitrarily to $X = 0$. Small changes in the origin are equivalent to differentiating that solution with respect to X, and differentiation is like the stability linearization. Hence we can pose an expansion

$$\tilde{c} = \left(\frac{1}{1+\mathrm{e}^X}\right)' + \delta c_1(X),$$

$$\tilde{w} = \left(\frac{\mathrm{e}^X}{1+\mathrm{e}^X}\right)' + \delta w_1(X),$$

$$\tilde{u} = 0 + \delta u_1(X),$$

$$\lambda = 0 + \delta \lambda_1(X).$$

At $O(\delta)$ the reaction equations become

$$-\alpha\phi\lambda_1 \frac{\mathrm{e}^X}{(1+\mathrm{e}^X)^2} + c_1' - \alpha u_1 = -\frac{c_1\,\mathrm{e}^X + w_1}{1+\mathrm{e}^X} = \lambda_1 \frac{\mathrm{e}^X}{(1+\mathrm{e}^X)^2} - w_1'.$$

Hence

$$c_1' + w_1' = \alpha(\lambda_1(\phi + \alpha^{-1}) + u_1)\frac{\mathrm{e}^X}{(1+\mathrm{e}^X)^2}.$$

Integrating from $-\infty$ to $+\infty$ where c_1 and w_1 vanish, we obtain an expression for the growth rate:

$$\lambda_1 = -\frac{1}{\phi + \alpha^{-1}} \int_{-\infty}^{\infty} \frac{\mathrm{e}^X}{(1+\mathrm{e}^X)^2} u_1 \,\mathrm{d}X.$$

The vorticity equation at $O(\delta)$ is

$$u_1'' - k^2 u_1 = k^2(\phi + \alpha^{-1}) \frac{\mathrm{e}^X}{(1+\mathrm{e}^X)^2}.$$

While it is possible to write down an integral expression for u_1 which decays at $\pm\infty$, it is not possible to evaluate explicitly this integral or the double integral for λ_1. Hence we tackle the vorticity equation for u_1 in the two limits of small and large wavenumber k, and also solve the equation numerically. We can, however, note that λ_1 will be independent of the porosity ϕ and the acid capacity α.

At large k, we have

$$u_1 \sim -(\phi + \alpha^{-1}) \frac{\mathrm{e}^X}{(1+\mathrm{e}^X)^2}$$

and so evaluating the integral for λ_1:

$$\lambda_1 \sim \tfrac{1}{6}.$$

At small k, there are three regions to consider. Near to the front

$$u_1 \sim kA + k^2[(\phi + \alpha^{-1}) \ln(1+\mathrm{e}^X) + BX] \quad \text{when } |X| \text{ is } O(1),$$

where A and B are two constants. Far from the front

$$u_1 \sim kA\,\mathrm{e}^{-|kX|} \quad \text{when } |kX| \text{ is } O(1),$$

using the matching of u_1 at the leading $O(k)$ constant term. Matching the next $O(kX)$ term, we find

$$B = -A \quad \text{and} \quad -(\phi + \alpha^{-1}) + B = A.$$

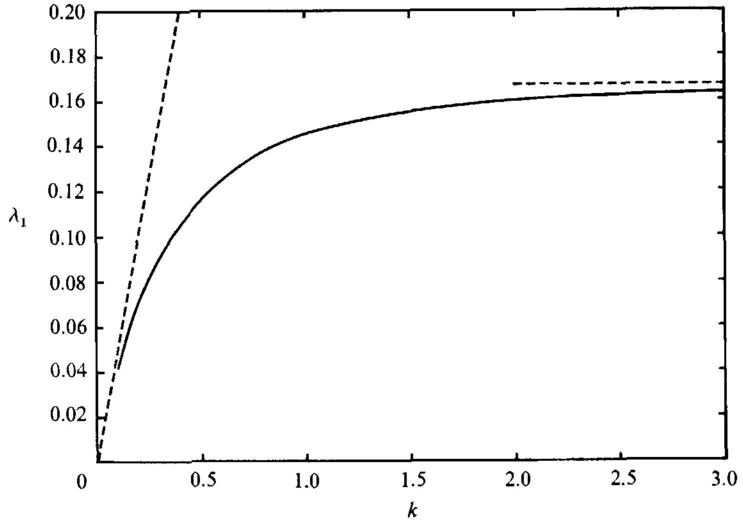

FIGURE 1. The growth rate $\lambda = \delta\lambda_1$ at small changes in the permeability, $\kappa = 1-\delta w$, $\delta \ll 1$, as a function of the y-wavenumber k. The results are independent of the porosity ϕ and acid capacity α.

Thus $$A = -\tfrac{1}{2}(\phi+\alpha^{-1})$$

and so $$\lambda_1 \sim \tfrac{1}{2}k.$$

Figure 1 compares these asymptotic results for λ_1 at small and large wavelengths with a numerical evaluation. The small-k result over-estimates the value by 20% by $k = 0.1$. The large-k result is within 20% by $k = 1.0$. In between the numerical solution increases monotonically.

We thus find in the case of small changes in the permeability that the front is unstable (if $\delta > 0$, i.e. the permeability decreases with the minerals). The growth rate is small, proportional to the wavenumber, at wavelengths long compared with the width of the front. The growth rate increases to a limiting value at short wavelengths.

4. Numerical results

For more general variations of the permeability it is necessary to solve the eigenvalue problem for the growth rate λ numerically. We have considered two particular forms for the permeability, an exponential decrease and a reciprocal decrease with minerals

$$\kappa(w) = e^{-\delta w} \quad \text{and} \quad \kappa(w) = \frac{1}{1+\delta w},$$

with $\delta = 1, 2, 3$ and 5.

The fourth-order linear system of differential equations for c, w, u and u' was solved by a fourth-order Runge–Kutta scheme. Far upstream there are two decaying solutions like $e^{(1+\lambda)X}$ and e^{kX} and two growing solutions like $e^{-\alpha\phi\lambda X}$ and e^{-kX}. Far downstream there are two decaying solutions like $e^{-(1+\alpha\phi\lambda)X}$ and e^{-kX} and two growing solutions like $e^{\lambda X}$ and e^{kX}. The numerical method adopted was to take each of the decaying solutions in turn and shoot from the far field to $X = 0$, noting the resulting values at $X = 0$ of c, w, u and u'. Solving a linear system of equations one

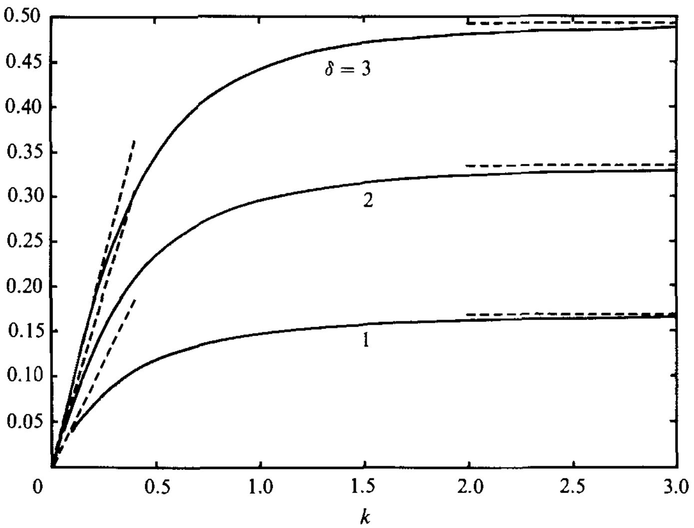

FIGURE 2. The growth rate λ for an exponential dependence of the permeability $\kappa = e^{-\delta w}$ for $\delta = 1$, 2 and 3 as a function of the y-wavenumber k. The porosity $\phi = 0.11$ and the acid capacity $\alpha = 1.0$.

can select the amplitudes of these four solutions that make c, w and u continuous at $X = 0$, and that are normalized in some way (one amplitude was set to unity). Finally we adjusted the guess for the value of λ until u' was continuous at $X = 0$.

We found that a step size of $\Delta X = 0.1$ gave four-figure accuracy. The need for a large value of X_∞ could be avoided by adding in a correction term $e^{-(1+k)|X|}$ to the solution $e^{-k|X|}$. With this correction good accuracy was achieved with $X_\infty = 10$ for most k. A larger value for X_∞ was required for the slower decaying solutions when $k < 0.2$, and for $k > 2$ a smaller value of X_∞ is needed to avoid an ill-conditioned matrix for the amplitudes.

The numerical results for the growth rate λ are given as a function of the y-wavenumber k in figure 2 for the exponential decrease in the permeability, and in figure 3 for the reciprocal decrease. The porosity has been taken to be $\phi = 0.11$ and the acid capacity $\alpha = 1.0$.

First we note that the propagating reaction front is unstable if the permeability is decreased by the dissoluble minerals, $\delta > 0$. The growth rate is proportional to the wavenumber at small wavenumbers, and increases monotonically to a limiting value at high wavenumbers, coming close to the plateau by $k = 2$.

For moderate changes in the permeability, say by a factor of 2 when δ is about 1, the growth rate is small, about 0.1. This means that the front has to propagate through 10 of its thicknesses before the instability increases by e-fold. The growth rate becomes large, about 0.5, for the larger changes in the permeability, by a factor of 150 for $\delta = 3$ in the exponential case. This high growth rate would mean that the instability would double as the front advanced through just twice its thickness and so the instability would quickly overwhelm the front. The above dependence of the growth rate on the magnitude of the changes in the permeability may explain why acid fronts can be seen in experiments with sandstone, but the acid advances through 'worm-hole' instabilities in limestones.

Comparing the results in figures 2 and 3 for the two different dependencies of the permeability, we see that for larger values of δ the exponential law is more unstable

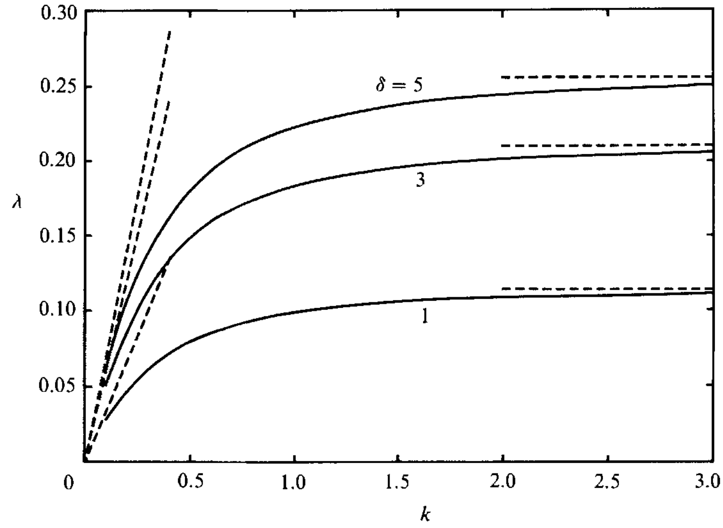

FIGURE 3. The growth rate λ for an reciprocal dependence of the permeability $\kappa = 1/(1+\delta w)$ for $\delta = 1$, 3 and 5 as a function of the y-wavenumber k. The porosity $\phi = 0.11$ and the acid capacity $\alpha = 1.0$.

than the reciprocal law. The exponential law with $\delta = 2$ has a higher growth rate than the reciprocal with $\delta = 5$. For these two cases the permeability decreases by a factor $\kappa(1)/\kappa(0) = 0.135$ for the exponential and a factor 0.167 for the reciprocal. Clearly the total change in the permeability is a better measure for the growth rate than δ which is the rate of change at no minerals, $\kappa'(0)$. Looking at the vorticity equation one can see that κ only occurs in the combination κ'/κ, and so some weighted average of this quantity is the real measure.

5. Long wavelengths

The above results suggest that the growth rate might be small at small wavenumbers. Thus for a general permeability $\kappa(w)$ we seek an expansion in the small parameter k;

$$\tilde{c} = \left(\frac{1}{1+e^X}\right)' + kc_1(X),$$

$$\tilde{w} = \left(\frac{e^X}{1+e^X}\right)' + kw_1(X),$$

$$\tilde{u} = 0 + ku_1(X) + k^2u_2(X),$$

$$\lambda = 0 + k\lambda_1(X).$$

As in the case of small changes in permeability we can obtain from the reaction equations the integral expression for the growth rate:

$$\lambda_1 = -\frac{1}{\phi+\alpha^{-1}}\int_{-\infty}^{\infty}\frac{e^X}{(1+e^X)^2}u_1\,dX.$$

To obtain the flow, we again have to consider three regions. Far from the front we have

$$u_1 = A\,e^{-|kX|} \quad \text{when } |kX| \text{ is } O(1),$$

where A is a constant. Near to the front

$$u_1 = A \quad \text{when } X \text{ is } O(1),$$

while u_2 is governed by

$$\left(\frac{1}{\kappa}u_2'\right)' = -(\phi+\alpha^{-1})\frac{\mathrm{d}\kappa/\mathrm{d}w}{\kappa^2}\left(\frac{\mathrm{e}^X}{1+\mathrm{e}^X}\right)'$$

$$= (\phi+\alpha^{-1})\left(\frac{1}{\kappa(w(X))}\right)'.$$

Integrating, $$u_2' = (\phi+\alpha^{-1})+B\kappa(w(X)),$$

where B is a constant of integration. Matching downstream, we have

$$u_2' \to (\phi+\alpha^{-1})+\kappa(1)B = -A,$$

while matching upstream

$$u_2' \to (\phi+\alpha^{-1})+\kappa(0)B = A.$$

Hence $$A = -(\phi+\alpha^{-1})\frac{\kappa(0)-\kappa(1)}{\kappa(0)+\kappa(1)}$$

and so the integral for the growth rate gives

$$\lambda = k\frac{\kappa(0)-\kappa(1)}{\kappa(0)+\kappa(1)}.$$

This result is independent of the porosity ϕ and the acid capacity α and most of the details of the variation of the permeability $\kappa(w)$. If the permeability without the minerals is very much larger than the original with the minerals, $\kappa(0) \gg \kappa(1)$, the growth rate is independent of the permeability. In the special case of small variations in the permeability, $\kappa(0) \approx \kappa(1)$, we recover the previous result for long wavelengths in the case of small changes in permeability. The above asymptotes for small k are included in figures 2 and 3. When δ is large the small-k solution is only applicable at quite small k.

6. Short wavelengths

At short wavelengths, $k \gg 1$, the vorticity equation gives

$$\tilde{u} \sim (\phi+\alpha^{-1})\frac{\mathrm{d}\kappa/\mathrm{d}w}{\kappa}\tilde{w}.$$

The reaction equation then becomes

$$\alpha\phi\lambda\tilde{c}+\tilde{c}'-(\alpha\phi+1)\frac{\mathrm{d}\kappa/\mathrm{d}w}{\kappa}\frac{\mathrm{e}^X}{(1+\mathrm{e}^X)^2}w = -\frac{\tilde{c}\,\mathrm{e}^X+\tilde{w}}{1+\mathrm{e}^X} = \lambda\tilde{w}-\tilde{w}'.$$

This equation contains no k, and so one can anticipate the growth rate λ increasing to a limiting value at short wavelengths. It is necessary to solve the above equation numerically, shooting from $-X_\infty$ with $c = \mathrm{e}^{(\lambda+1)X}$ and $w = -(\alpha\phi\lambda+\lambda+1)\,\mathrm{e}^{(\lambda+1)X}$ to $+X_\infty$, and adjusting the value of λ until $c \to 0$ at $+X_\infty$. (These particular forms in the far field can be obtained by examining the equations there.)

The results of the numerical solution are given in figure 4. The values from this calculation were used for the high-wavenumber asymptotes in figures 2 and 3. Comparing the results for the exponential with those for the reciprocal in figure 4, we

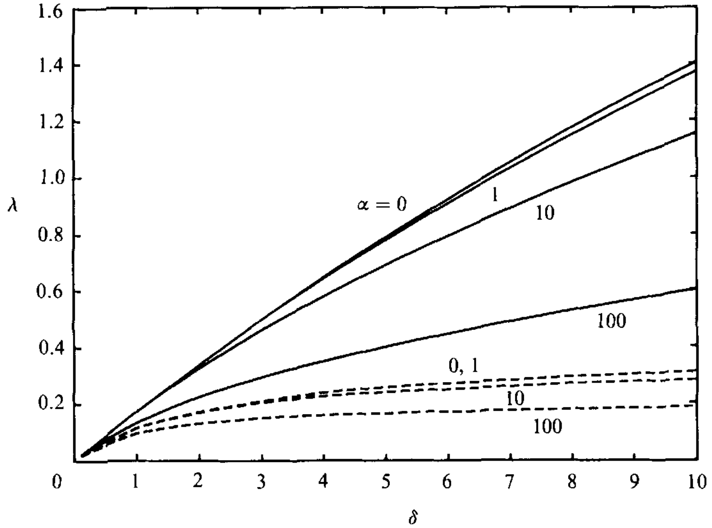

FIGURE 4. The short-wave limiting value of the growth rate λ as a function of the change in the permeability δ at acid capacity $\alpha = 0, 1, 10$ and 100 and at porosity $\phi = 0.11$. The continuous curves are for the exponential dependence of the permeability $\kappa = e^{-\delta w}$ and the broken curves are for the reciprocal dependence $\kappa = 1/(1+\delta w)$.

see again that the total change in the permeability is a better measure of the growth rate than the parameter δ.

The acid capacity is seen to have little effect until its value is large. Looking at the equations governing the short-wave limit we can see that α only occurs in the combination $\alpha\phi$. As the porosity ϕ is always small, the acid capacity will only have an effect on the growth rate if it is correspondingly large. One might suppose that at high values of the acid capacity the concentrated acid dissolves all the minerals more uniformly and so the permeability is also more uniform, thus reducing the scope for the preferential growth of the instability.

The nearly linear increase in the growth rate with the parameter δ for the exponential permeability law in figure 4 may be connected with the logarithmic increase in the growth rate which Sherwood (1987) found at short wavelengths with an unbounded permeability law.

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