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The stability of immiscible liquid layers in a porous medium

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The stability of liquid layers in a porous medium under the action of viscous and surface forces is described. An extension of previous studies on the stability of a single interface in a porous medium is presented as the basis for solutions to many problems of practical interest where flow in porous media are involved.

Introduction

The stability of liquid–liquid and gas–liquid interfaces in porous media is of considerable interest in a variety of fields. For example, the movement of an oil–water contact is of importance in petroleum production engineering, and the behaviour of a freshwater–brine contact is of interest in ground water hydrology. Thus over the last decade the stability of such interfaces has attracted the attention of many workers. However, the stability of liquid layers in porous media has been considered only recently by Raghavan (1970). This paper represents not only a logical extension of previous theoretical studies but also forms a basis for such problems of practical importance as the flow of liquids of varying viscosity and the mixing of liquids in porous media.

The instability of interfaces in porous media was first considered by Saffman & Taylor (1958). Their principal conclusion was that, when two superimposed fluids of different viscosities are forced through a porous medium and then subjected to small deviations (perturbations), the stability of the interface depends on whether the direction of motion is from the more viscous to the less viscous fluid or vice versa. This is true whatever the relative density of the fluids, provided that the velocity is sufficiently large. They also pointed out that the instability which results due to the viscosity difference is similar to the Rayleigh (1899)–Taylor (1950) instability wherein the interface between two liquids becomes unstable if the acceleration is from the lighter to the heavier liquid and is stable if the acceleration is in the opposite direction.

Saffman & Taylor assumed that the two immiscible fluids remain completely separate at the interface in a porous medium. The description of the normal modes of disturbances from an interface and their rate of growth requires a knowledge of boundary conditions at the interface. Starting with the simplest assumption of a planar interface (surface tension is then neglected and equality of pressures assumed), they showed that when the interface between the two fluids was horizontal, it would be stable for small deviations from steady state if

$$\left(\frac{\mu_2}{k_2}-\frac{\mu_1}{k_1}\right) V+(\rho_2-\rho_1)g > 0, \tag{1}$$

and unstable if

$$\left(\frac{\mu_2}{k_2}-\frac{\mu_1}{k_1}\right) V+(\rho_2-\rho_1)g < 0. \tag{2}$$

Here k is the effective permeability of the medium to the fluids, μ is the viscosity, ρ is the density, V is the velocity normal to the interface, g is the acceleration due to gravity, and subscripts 1 and 2 refer to the upper and lower fluids, respectively.

Under the simplest of assumptions, the effect of surface tension was to dampen wavelengths less than λ, where

$$\lambda = 2\pi\sigma^{\frac{1}{2}}b[12V(\mu_1-\mu_2)+b^2g(\rho_1-\rho_2)]^{-\frac{1}{2}}. \tag{3}$$

Here b is the spacing between the plates in a Hele-Shaw cell and σ is the interfacial tension.

Saffman & Taylor's experiments with air–water systems were somewhat similar to those of Lewis (1950) who studied both accelerated air–liquid and liquid–liquid interfaces. They examined the post-instability situation of fingering—and noted that the development of these fingers was similar to the development of those described by Lewis for the later stages of instability of an accelerated interface. However, in their experiments, Saffman & Taylor noted that the air fingers and the columns of liquid in between them were equally spaced, whereas the air fingers of Lewis's experiment were separated only by very narrow columns of fluid. They also conducted experiments in Hele-Shaw cells using oil and water, which again confirmed theoretical predictions regarding instability, but their results regarding the post-instability situation were inconclusive.

Saffman & Taylor's conclusion that the viscosity ratio is an important factor in fluid displacements in porous media is not new to the oil industry. Until 1958 research in the petroleum production industry had been concerned mainly with the conditions necessary for a steady interface to exist in certain cases, and not explicitly with the stability of the interface (Dietz 1953; Kidder 1956). The analysis of the instability of fluid interfaces has been carried further by Chuoke, van Muers & van der Poel (1959). They considered a planar interface between two immiscible liquids which was initially at an angle ($Z \sim Z_1$) with the vertical plane and which was then displaced at a constant rate, U, normal to the front. Their theoretical results indicated that instability would occur for all rates greater than a critical rate, U_c, given by

$$\left(\frac{\mu_2}{k_2}-\frac{\mu_1}{k_1}\right) U_c+(\rho_2-\rho_1)g\cos(Z \sim Z_1) = 0, \tag{4}$$

and provided that the Fourier decomposition of the spatial perturbation or deformation of the moving displacement front contains modes with wavelengths, λ, which are greater than a critical wavelength, λ_c, given by

$$\lambda_c = 2\pi \left\{ \frac{\sigma}{\left[\left(\frac{\mu_2}{k_2} - \frac{\mu_1}{k_1}\right)(U - U_c)\right]} \right\}^{\frac{1}{2}}. \tag{5}$$

Furthermore, they reported that there is a wavelength of maximum instability given by

$$\lambda_m = 3^{\frac{1}{2}}\lambda_c. \tag{6}$$

Their theoretical expressions are in fair agreement with experiments with both parallel plate channels and unconsolidated glass powder packs.

The non-linear theory of frontal stability in porous media has been considered by Outmans (1962), who points out the error of neglecting the non-linear terms in the boundary conditions. His results are analogous to those of Chang (1959). Outman's analysis includes a detailed discussion of the limitations of the linear theory in studying the phenomena of fingering.

In spite of the superficial resemblance of the Saffman–Taylor instability to the Rayleigh–Taylor instability, there are basic differences. The dynamic boundary condition in the two cases is different and this affects the rate at which the disturbances are amplified. The growth factor for the Saffman–Taylor instability is always real, whereas that for the Rayleigh–Taylor instability can be complex. Also, the number of roots obtained in the solution of the equations are greater for the Rayleigh–Taylor instability.

The physical differences should also be borne in mind. The Rayleigh–Taylor instability corresponds to accelerating inviscid fluids and inertial terms are considered. However, the Saffman–Taylor instability represents movement of fluids at constant velocity where Darcy flow prevails, that is, the inertial terms are negligible and the viscous terms dominate.

Recently the petroleum production industry has begun to take an interest in the formation of emulsions in porous media. Much of the crude oil produced in the world is in the form of stable emulsions. The part played by the porous medium in the formation of emulsions is a controversial question and has been largely overlooked by members of the petroleum production industry. It is often claimed that these emulsions are formed as oil and water flow through the chokes and other flow constrictions in oil field equipment. Even though this view is certainly reasonable, emulsions are still produced in wells which not only lack these constrictions but which also produce at low flow rates. Also in some laboratory experiments the flow of crude oil and brine results in the formation of emulsions. This strongly suggests that at least in some cases the emulsions are produced in the reservoir rock itself and thus there was need for the study of the role of the porous medium in emulsion formation. Such a study has been made by Raghavan & Marsden (1971). They have shown that the study of the stability of fluid interfaces offers a profitable method of investigating emulsification in porous media. Their basic contribution has been to demonstrate that viscosity differences between fluids can play a significant role in the emulsification process.

The emulsification process mentioned above has been visualized by Raghavan & Marsden to occur in a number of stages. The first stage of their model consists of the break-up of a liquid layer. As mentioned earlier, this study should be of interest to a number of research workers in other fields, and so this analysis of the stability of fluid layers is presented in a general manner.

Formulation

In ordinary fluid flow problems the geometry of the conduit can almost always be specified. However, for viscous flow in the extremely complex geometry of porous media, specification of the flow geometry is impossible. The failure of a theoretical analysis to yield a tractable equation of flow has led to the experimental determination of the factors affecting the flow of fluids in porous materials and the results are known as Darcy's law. This law states that the macroscopic velocity of the fluid in the direction of flow at any particular point within the porous medium is proportional to the instantaneous gradient in the fluid head or flow potential at the point. Symbolically this may be expressed as

$$\mathbf{V} = -\frac{k\rho}{\mu}\,\mathrm{grad}\left(zg-\frac{1}{\rho}p\right). \tag{7}$$

Here, p is the pressure, z the distance in the vertical direction which represents the gravitational head, and $\mathbf{V}$ is the macroscopic velocity.

It should be noted that the above velocity is a macroscopic velocity wherein the solid matrix of the porous medium and the fluid occupying the pore channels are envisioned together as a composite conductor of fluids. Miller (1962) has shown that it is also possible to consider only the pore channels in which case a microscopic velocity may be defined and Darcy's law expressed as

$$\mathbf{V} = -\frac{k\rho}{\mu\phi}\,\mathrm{grad}\left(zg-\frac{1}{\rho}p\right), \tag{8}$$

where $\mathbf{V}$ is now a microscopic velocity, and ϕ is a statistically averaged porosity of the matrix. In the present analysis we shall use (8). Miller has also shown that the above equations can be extended to more than one fluid flowing in a porous medium.

Let us now consider the stability of a planar slab or a layer of liquid which separates two liquids at different pressures. The properties of the two surrounding liquids may or may not be different from each other, but they are different from the liquid layer under consideration. The porous medium is assumed to be a homogeneous, isotropic rock matrix with constant porosity and effective permeability to the liquids. We shall assume the liquids to be incompressible, and to have a constant viscosity. The interfacial tension between the liquids is assumed constant. The liquids are also assumed to fill the pore spaces of the rock completely under basic flow conditions.

In a system of this type, an unperturbed immiscible displacement is characterized by a transition zone of steep saturation gradients of the displacing and displaced liquids. This means that a sharp interface does not exist, but that there is an ill-defined transition zone in which the liquids intermingle. In the model the transition zone is replaced by a sharp planar, macroscopic interface to which are assigned pressure discontinuities preserving the capillary properties of the transition zone. The relatively uniform saturation and flow conditions prevailing outside the zone are extrapolated back to this hypothetical interface. Perturbation theory can be directly applied to these models, and finds partial justification

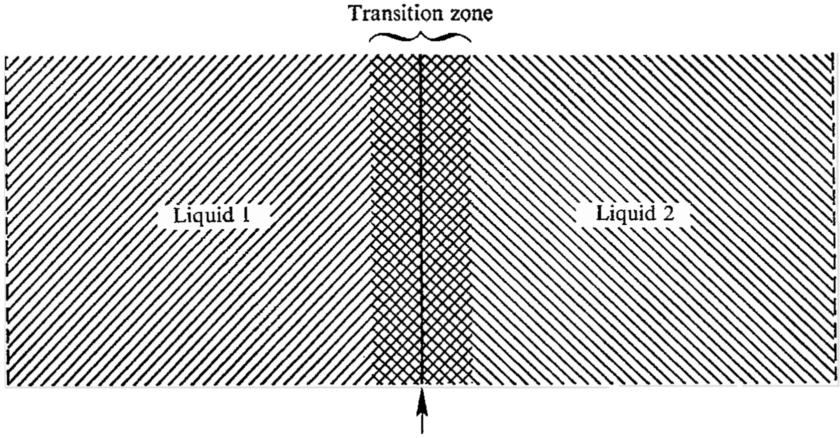

FIGURE 1. Representation of transition zone for immiscible liquid displacements in a porous medium.

(implied in all the following work) in that only fundamental perturbation modes with wavelengths that are large relative to the width of the transition zone at the time of application of perturbation will be considered. Figure 1 is a sketch showing the transition zone as well as the idealization mentioned above.

As a further assumption, we shall neglect the motion of the surrounding fluids, in order to consider the stability of a single layer in detail. It may be suggested that the utility of this analysis is limited because the surrounding liquids are then assumed to have a negligible density and mobility (ratio of permeability to viscosity). But inertial effects are usually neglected while considering flow through porous media as exemplified in the analysis made by Saffman & Taylor. In fact, it should be noted that Saffman & Taylor's conclusions are independent of the relative density of the fluids.

The assumption that the surrounding fluids have a negligible mobility also requires some mention. In oil-field practice, mobility differences between fluids undergoing immiscible displacement (either naturally such as the movement of an oil–water contact, or artificially in the case of a water-flooding operation) can be fairly large. Thus from a practical point of view the assumption regarding mobility ratios is satisfactory.

Having considered the flow equation as well as the principal assumptions in detail we can now consider further the formulation of the problem. Darcy's law, together with the assumption of incompressibility, implies the existence of a velocity potential, Φ, defined by

$$\Phi(x,y,z,t) = (k/\mu\phi)(p+\rho g z), \tag{9}$$

which satisfies Laplace's equation. Thus

$$\mathbf{V} = -\nabla\Phi \tag{10}$$

and

$$\nabla^2\Phi = 0. \tag{11}$$

The boundary surface is given by

$$z = \eta_j(x,y,t) \quad (j = 1,2). \tag{12}$$

The subscript $j = 1$ and 2 corresponds to the upper and lower surfaces of the liquid layer respectively.

The dynamic and kinematic boundary conditions can be written, respectively, as

$$\frac{\Phi\phi\mu}{k\rho} - g\eta_j - \frac{(-1)^j}{\rho}\sigma 2H_j = \frac{p_j}{\rho} \quad \text{on} \quad z = \eta_j \quad (j = 1,2) \tag{13}$$

and

$$\Phi_z - \Phi_x\eta_{jx} - \Phi_y\eta_{jy} + \eta_{jt} = 0 \quad \text{on} \quad z = \eta_j \quad (j = 1,2), \tag{14}$$

where p_j is the pressure outside the layer, H is the curvature of the surface, and the subscripts x, y, z and t represent partial derivatives in the space and time.

Equation (13) states that the pressure discontinuity across the interface is equal to the capillary pressure. Equation (14) expresses the kinematic boundary condition that the substantial time derivative on the interface is equal to zero. This means that there is no relative motion between the interface and any fluid particle at the interface, i.e. that any fluid particle at the interface moves in the direction of and at the speed of the interface. The initial conditions will be imposed as needed.

In the present instance the flow is unbounded in the x, y plane (plan form). If the flow is bounded by a cylindrical tube which has an axis parallel to the z axis the condition on the circumference of the cylinder is

$$\partial\Phi/\partial N = 0, \tag{15}$$

where N is the direction normal to the circumference.

Solution

The proposed model assumes that a zero-order flow exists. The first-order perturbation of the same flow which satisfies linear equations is represented by a series, or integrals of normal modes. Some of the modes are found to be unstable in the sense that their amplitude increases indefinitely with time. Of these unstable modes, the one which has the largest exponent is the least stable, and is assumed to be responsible for the break-up of the layer into segments. The number of segments depends upon the number of positive and negative regions into which this mode is decomposed by its nodal lines. These segments, after break-up of the layer, could become spherical. The number and size of the resultant droplets are then determined.

Zero-order solution

Let the bounding surfaces be planes normal to the z axis and independent of x and y. The solution will then be independent of x and y. Then if U denotes the z component of velocity, we have, from (10) and (11),

$$\Phi^0 = -Uz+b, \tag{16}$$

where the superscript 0 denotes the zero-order solution.

Now, as η_j is dependent only on t, from (13), (14) and (16) we have

$$[-U\eta_j^0+b]\frac{\phi\mu}{k\rho}-g\eta_j^0-\frac{(-1)^j 2H_j}{\sigma}=\frac{p_j}{\rho} \quad (j=1,2) \tag{17}$$

0

and
$$U = \eta_{j,t}^0 \quad (j=1,2). \tag{18}$$

If h denotes the initial separation between the two surfaces, from (12) we have

$$\eta_2^0 = \eta_1^0 - h. \tag{19}$$

Solving for U, we have
$$U = \frac{k}{\phi\mu}\left[\frac{p_2-p_1}{h}-\rho g\right]. \tag{20}$$

First-order perturbation

Suppose that the initial bounding surfaces differ from planes, and that the initial velocity differs slightly from the constant z component assumed in the above solution, which we will henceforth call the 'unperturbed solution'. Then we may expect the subsequent solution to differ slightly from the above solution. If ϵ is a measure of maximum deviations from the initial data, we assume that the solution may be written in the form

$$\Phi = \Phi^0+\epsilon\Phi^1+\ldots, \quad \eta_j = \eta_j^0+\epsilon\eta_j^1+\ldots. \tag{21}$$

From (11) and (21), we see that Φ^1 also satisfies the Laplace equation. From (13), (14), (16) and (21), we obtain the first-order perturbation equations for Φ^1 and η_j^1:

$$\frac{\phi\mu}{\rho k}\Phi^1-\left(\frac{\phi\mu}{\rho k}U+g\right)\eta_j^1-\frac{(-1)^j\sigma}{\rho}\nabla^2\eta_j^1 = 0 \quad \text{on} \quad z=\eta_j^0 \quad (j=1,2) \tag{22}$$

and
$$\Phi_z^1+\eta_{j,t}^1 = 0 \quad \text{on} \quad z=\eta_j^0 \quad (j=1,2). \tag{23}$$

To solve Laplace's equation for Φ^1, assume that Φ^1 is a product of a function of z and t multiplied by a function $\Psi(x,y)$. Thus, let

$$\Phi^1 = [G_1(t)\,e^{mz}+G_2(t)\,e^{-mz}]\,\Psi(x,y) \tag{24}$$

and
$$\eta_j^1 = A_j\,e^{\alpha t}\,\Psi(x,y), \tag{25}$$

where m is the wave-number of the disturbance in the z direction, G is a function of time, A_j is an arbitrary constant, and α is the growth factor which governs the amplification of the interface.

Our prime interest in this analysis is the study of the behaviour of α. Substituting the right-hand sides of (24) and (25) for Φ^1 and η^1 in (22) and (23) and simplifying, we have:

$$\alpha = -\frac{\sigma k}{\phi\mu}\left\{m^3\coth(mh) \pm m\left[\frac{\rho^2}{\sigma^2}\left[\frac{\phi\mu}{\rho k}U+g\right]^2 + m^4[\coth^2(mh)-1]\right]^{\frac{1}{2}}\right\}, \quad (26)$$

or

$$\frac{\phi\mu h^3}{\sigma k}\alpha = -\theta^3\coth\theta \pm \theta[B^2+\theta^4\sinh^{-2}\theta]^{\frac{1}{2}}, \quad (27)$$

where

$$\theta = mh \quad (28)$$

and

$$B = \frac{\rho h^2}{\sigma}\left(\frac{\phi\mu}{\rho k}U+g\right). \quad (29)$$

There are two values of α and each α is the sum of two terms of the form of (27).

Marginal stability

It has been shown that there are two values of the growth factor α. If both values of α are negative, then the perturbation of the surface is such that no break-up of the interface occurs. The negative values of α are not of significance in the present study, and in practice cannot be seen as they would be damped out. However, when the positive sign before the square root sign is chosen for a certain range of θ, α is positive and real, leading to a growing exponential. As the system passes from stability to instability, the value of α will pass through zero. The results obtained have established that there is no oscillating motion with respect to time; consequently, attention may be confined to modes associated with real exponential time factors, and limiting conditions of stability are in fact obtained when all time variations are zero. The study of these limiting conditions—marginal stability (Jeffreys 1926)—is of importance to understand the departure from stable to unstable flow.

To determine the condition for the onset of instability, the growth factor, α, is set equal to zero in (27) and then the following results:

$$m^2 = (p_2-p_1)/\sigma h. \quad (30)$$

Defining

$$\beta = (p_2-p_1)/\sigma h, \quad (31)$$

we have

$$m^2 = \beta. \quad (32)$$

The critical velocity for the onset of instability is, after some manipulation,

$$U_c = \frac{k}{\phi\mu}(\sigma m^2 - \rho g), \quad (33)$$

which can be further expressed as

$$U - U_c = \frac{k\sigma}{\phi\mu}\left(\frac{p_2-p_1}{\sigma h} - m^2\right). \quad (34)$$

The neutral stability curve represented by (30) is shown in figure 2. The plot delineates the stable and unstable regions for ratios of pressure gradient to interfacial tension as a function of wave-numbers. (Note that any disturbance should contain some of all wave-numbers shown.) Large values of m (small wavelengths, $m = 2\pi/\lambda$), are stabilized for any particular ratio of pressure gradient to interfacial tension, β, i.e. there is a cutoff wave-number above which instability does not occur for a particular value of β. This is due to the stabilizing influence of interfacial tension. Figure 2 also indicates that the cutoff wave-number increases as the ratio for the viscous to surface forces increases for a given disturbance, thus demonstrating the relationship between the stabilizing and destabilizing forces.

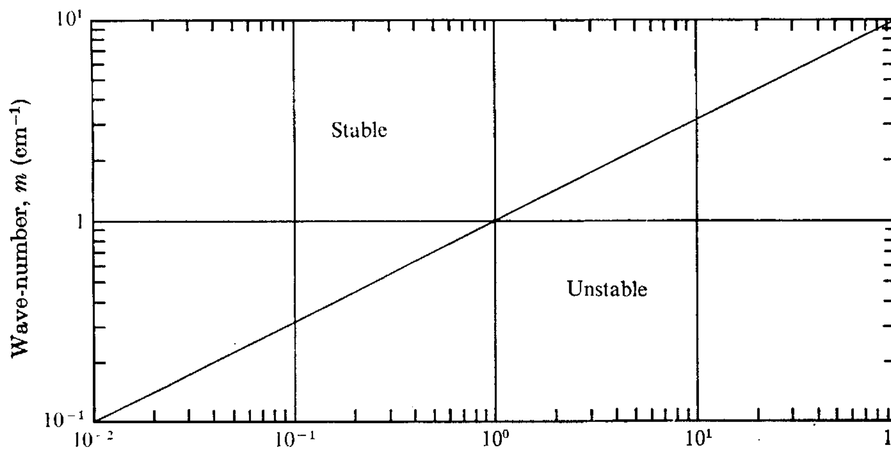

FIGURE 2. Neutral stability curve: wave-number versus ratio of pressure gradient to interfacial tension.

The critical velocity according to (33) and (34) is a function of the wave-number and the ratio β. The relationship between the critical velocity and the wave-number for ratios of pressure gradient to interfacial tension is shown in figure 3. It is seen that at least some wave-numbers of a disturbance are unstable for any value of β, i.e. instability always exists. This velocity is insensitive to the wave-number when $m < 1$ for all β considered, and is considerably influenced by the wave-number for $m > 1$.

Instability

For conditions when $\alpha > 0$, there is a greatest value of α, corresponding to a particular value of θ (say $\theta_{\max}$). The exponential corresponding to this value of θ will grow most rapidly. An approximate value of $\theta_{\max}$ may be obtained if we first obtain an expansion for α when B is large. In this case (27) becomes

$$\phi\mu h^3\alpha/\sigma k = \theta[B - \theta^2 \coth\theta], \tag{35}$$

and α is positive between $\theta = 0$ and $B^{\frac{1}{2}}$.

It should be noted that (35) implies that

$$B > \frac{1}{2^{\frac{1}{2}}}\frac{\theta^2}{\sinh\theta} > \frac{1.3}{2^{\frac{1}{2}}}.$$

The maximum value of α occurs when

$$\theta_{\max} = \left(\frac{B}{3}\right)^{\frac{1}{2}} = h\left[\frac{(\phi\mu/k)\,U + \rho g}{3\sigma}\right]^{\frac{1}{2}}. \tag{36}$$

Recalling that by (28) $\theta = mh$, we may introduce m_{max}, the value of m corresponding both to θ_{max} and to maximum instability. From (36) we have

$$m^2_{max} = \frac{(\phi\mu/k)\,U + \rho g}{3\sigma}. \qquad (37)$$

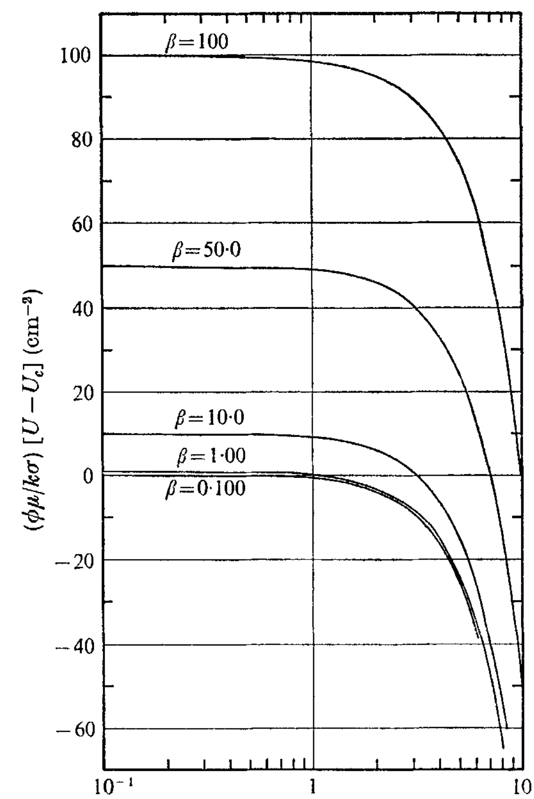

FIGURE 3. Calculated difference in interfacial and critical velocity versus wave-numbers for various ratios of pressure gradient to interfacial tension. $\beta = \Delta p/\sigma h$.

The relationship between a growth factor, χ_n, defined by

$$\chi_n = (\phi\mu/\sigma k)\alpha \qquad (38)$$

and a wave-number, m, for various ratios of pressure gradient to interfacial tension is given in figure 4. Some of the modes for a given β are unstable. The dominant mode would be expected to grow at the fastest rate, and be responsible for the break-up of the interface. As the ratio β increases, the value of the cutoff wave-number m and the magnitude of the growth factor χ_n corresponding to each wave-number increase. It is also of interest to note that wave-numbers just below the cutoff wave-number grow at a rate faster than those of larger wave-lengths.

The dependence of the growth factor χ_n on the ratio of pressure gradient to interfacial tension β for the range of wave-numbers considered can be seen in figure 5.

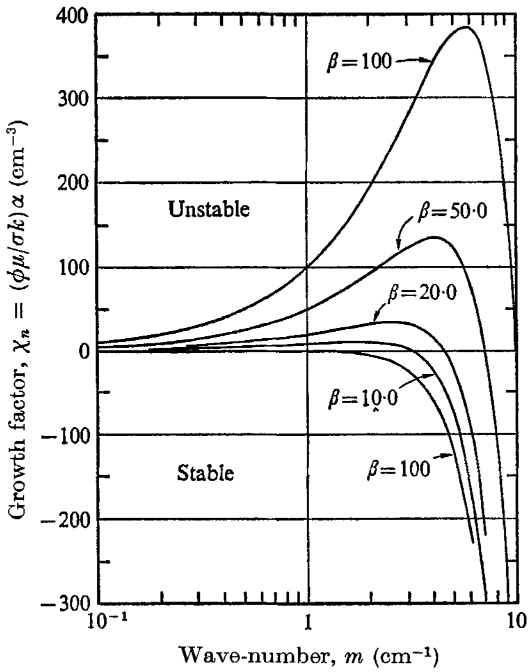

FIGURE 4. Calculated growth factors versus wave-numbers for various ratios of pressure gradient to interfacial tension. $\beta = \Delta p/\sigma h$.

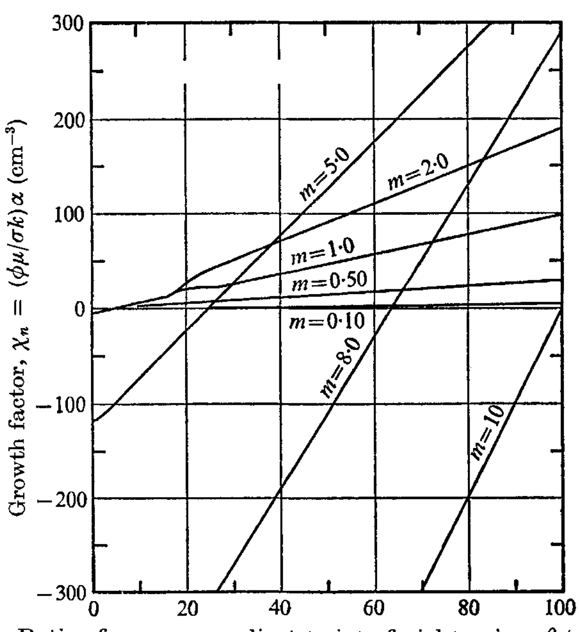

FIGURE 5. Calculated growth factors versus ratios of pressure gradient to interfacial tension for various wave-numbers, m.

Plan form

To determine the number of pieces into which the layer breaks due to the growth of the term corresponding to $m_{\max}$, the function $\Psi(x,y)$ must be considered in detail. Again the geometry of the medium plays an important part. If the layer is unbounded in the x, y plane, there will be many functions of $\Psi(x,y)$ corresponding to $m_{\max}$. On the other hand, if the region is bounded, there may be no solution corresponding exactly to $m_{\max}$, although there will be many functions of $\Psi(x,y)$ near $m = m_{\max}$. Thus there will be many functions Ψ with either exactly, or approximately, the same value ($m \approx m_{\max}$); and the corresponding terms will grow at the same maximum rate. Consequently the exact manner of break-up will depend upon the extent to which these terms are excited by the initial perturbation.

An estimate of the number of pieces into which the layer breaks can be made as follows. For an unbounded region, one solution of $\Psi(x,y)$ corresponding to $m_{\max}$ is

$$\Psi(x,y) = \exp\left[2\pi i\left(\frac{x}{\lambda_1}+\frac{y}{\lambda_2}\right)\right], \tag{39}$$

where

$$\left(\frac{2\pi}{\lambda_1}\right)^2+\left(\frac{2\pi}{\lambda_2}\right)^2 = m_{\max}^2.$$

Consider regions in the x, y plane in which the real part of Ψ is positive (or negative). These are rectangles bounded by nodal lines, and the dimensions of the rectangle are $\frac{1}{2}\lambda_1$ and $\frac{1}{2}\lambda_2$. The area for a fixed perimeter is minimum and is given by $\lambda_1 = \lambda_2 = 2\pi 2^{\frac{1}{2}}/m_{\max}$. Thus the minimum area is

$$\frac{\lambda_1^2}{4} = \frac{2\pi^2}{m_{\max}^2} = \frac{6\pi^2\sigma}{(\phi\mu/k)\,U+\rho g}. \tag{40}$$

Equation (40) can be expected to represent roughly the area into which a segment of the layer will break because all parts of the surface in this region move in the same direction. The volume of the segment will be given by this area determined above, multiplied by the thickness h. The radius r of the sphere into which the surface volume will ultimately deform is

$$r = \left[\frac{9\pi\sigma h}{2((\phi\mu/k)\,U+\rho g)}\right]^{\frac{1}{3}}. \tag{41}$$

Using (20), we have

$$r = \left[\frac{9\pi\sigma h^2}{2(p_2-p_1)}\right]^{\frac{1}{3}}. \tag{42}$$

Somewhat similar results are obtained for a tube of rectangular or any other bounded shape.

On the basis of (42) three means are available for diminishing droplet size, namely: (i) reducing the thickness of the layer h; (ii) reducing interfacial tension by use of surface-active agents; (iii) increasing the pressure difference, (p_2-p_1).

Another point to be noted is that reducing h has the twofold effect of (*a*) increasing velocity and (*b*) creating thinner pieces.

The time required for break-up can be approximately estimated if it is assumed that the layer breaks up when the perturbation reaches a value of h. The time for break-up is given by

$$t = \frac{1}{\alpha_{max}} \ln \frac{h}{A}, \tag{43}$$

where A is the initial perturbation and α_{max} is the value of α corresponding to θ_{max}.

From (35) and (36), the value of α_{max} can be obtained as

$$\alpha_{max} = \frac{2kh}{(27)^{\frac{1}{2}} \phi\mu\sigma^{\frac{1}{2}}} \left[\frac{\phi\mu}{k} U + \rho g\right]^{\frac{3}{2}}. \tag{44}$$

Thus,

$$t = \frac{(27)^{\frac{1}{2}} \phi\mu\sigma^{\frac{1}{2}}}{2k} \left(\frac{h}{p_2 - p_1}\right)^{\frac{3}{2}} \ln \frac{h}{A}. \tag{45}$$

Effect of surface tension

The preceding analysis has been carried out for a constant value of surface tension. The effect of the spatial variation of the surface tension is now examined in a simplified manner. Also, the variation in surface tension should of itself cause a relative movement of the interface. The model proposed is essentially an extension of the Saffman–Taylor analysis.

Consider two liquids to be at rest in a porous medium with the lighter fluid (represented by carets) to be above the denser fluid with an unperturbed interface, $z = 0$. The model proposed considers the variation of interfacial tension at random spots representing situations where local reductions are few and far apart. Surface tension is considered to be a step function (which may be periodic) and is given by

$$\sigma_1 \quad \text{for} \quad |x| > \delta, \tag{46}$$

$$\sigma_1 - \sigma_0 \quad \text{for} \quad |x| < \delta. \tag{47}$$

The velocity potential and flow equations are those which have been defined previously.

The dynamic condition at the interface is (following Landau & Lifshitz 1959, p. 234).

$$p = \hat{p} - \frac{\partial}{\partial x}\left(\sigma \frac{\partial \eta}{\partial x}\right). \tag{48}$$

The kinematic boundary condition for small disturbances at the interface is

$$\frac{\partial \eta}{\partial t} = -\frac{\partial \Phi}{\partial z} = -\frac{\partial \hat{\Phi}}{\partial z}, \tag{49}$$

where the boundary surface is given by

$$z = \eta(x, t). \tag{50}$$

It will be shown that, for a given disturbance, the interface in the two regions moves at different speeds and thus hydrodynamic instability results.

Solution

Case A, $\sigma = \sigma_1$ *and* $|x| > \delta$. A small perturbation of the form $\cos sx$ is impressed at $t = 0$. Let

$$\hat{\Phi} = A \cos sx \exp(-mz) f(t), \tag{51}$$

$$\Phi = C \cos sx \exp(mz) f(t), \tag{52}$$

$$\eta = A_1 \cos sx f(t), \tag{53}$$

where A, C and A_1 are arbitrary constants and $f(t)$ is a function of time.

Substituting the right-hand sides of (51) and (52) for $\hat{\Phi}$ and Φ in (48) and (49), and simplifying, we have

$$f(t) = D \exp\left\{-\left[\frac{\sigma_1 ms^2 + (\rho - \hat{\rho}) gm}{(\mu/k) + (\hat{\mu}/\hat{k})}\right] t\right\}, \tag{54}$$

where D is an arbitrary constant. Noting that $m = s$, we have

$$f(t) = D \exp(-\alpha_1 t), \tag{55}$$

where

$$\alpha_1 = \frac{\sigma_1 m^3 + (\rho - \hat{\rho}) gm}{(\mu/k) + (\hat{\mu}/\hat{k})}. \tag{56}$$

Thus we have

$$\eta_1 = A_1 \cos mx \exp(-\alpha_1 t). \tag{57}$$

Case B, $\sigma = \sigma_1 - \sigma_0$ *and* $|x| < \delta$. By an analysis similar to that used for case A, it can be shown that

$$\eta_2 = A_2 \cos mx \exp(-\alpha_2 t), \tag{58}$$

where

$$\alpha_2 = \frac{(\sigma_1 - \sigma_0) m^3 + (\rho - \hat{\rho}) gm}{(\mu/k) + (\hat{\mu}/\hat{k})}. \tag{59}$$

In order to study the relative motion of the interface, (57) and (58) would have to be matched at $|x| = \delta$. The conditions to be satisfied are that at $|x| = \delta$, there should not be a sudden jump in physical parameters such as (i) displacement and (ii) velocity. That is, at $|x| = \delta$, $\eta_1 = \eta_2$ and $\eta_{1,t} = \eta_{2,t}$.

Continuity of displacement gives rise to $\cos m\delta = 0$, or

$$m\delta = \tfrac{1}{2}(2q+1)\pi \quad (q = 0, 1, 2, \ldots), \tag{60}$$

the case $q = 0$ being of primary interest.

Continuity of velocity implies that

$$A_2 = A_1 \frac{\alpha_1 \exp(-\alpha_1 t)}{\alpha_2 \exp(-\alpha_2 t)}, \tag{61}$$

i.e.

$$\eta_2 = A_1 \left[\frac{\sigma_1 m^3 + (\rho - \hat{\rho}) gm}{(\sigma_1 - \sigma_0) m^3 + (\rho - \hat{\rho}) gm}\right] \cos mx \exp(-\alpha_1 t). \tag{62}$$

From the comparison of (57) and (62), it can be seen that the two regions are displaced differently.

There are certain points which have to be considered further. Examination of (62) shows that the component of time is a decaying function and that the tendency for spontaneous displacement will decrease with time, while (62) also indicates that density differences of two fluids will influence the movement of the interface. If the density difference is extremely small, then changes in interfacial tension would be dominant. This would be so in two-dimensional horizontal flow, where the 'gravity' terms would drop out and changes (lowering) of interfacial tension would have a marked effect.

It has already been mentioned that the effect decays as time increases. There is one point which must be remembered, however. It has been assumed that the system is at rest. If the interface were moving at a constant velocity U, then, following Chuoke *et al.* (1959),

$$-\alpha_1 = \left\{\left(\frac{\hat{\mu}}{\hat{k}}-\frac{\mu}{k}\right) Um + (\hat{\rho}-\rho)gm - \sigma_1 m^3\right\} \Big/ \left(\frac{\mu}{k}+\frac{\hat{\mu}}{\hat{k}}\right) \tag{63}$$

and

$$-\alpha_2 = \left\{\left(\frac{\hat{\mu}}{\hat{k}}-\frac{\mu}{k}\right) Um + (\hat{\rho}-\rho)gm - (\sigma_1-\sigma_0) m^3\right\} \Big/ \left(\frac{\mu}{k}+\frac{\hat{\mu}}{\hat{k}}\right), \tag{64}$$

and instabilities will now result depending on the viscosities, and the effect of differential displacement will be enhanced. The time component can be changed from a decaying function to an increasing function for a critical velocity U_c, where U_c is given by

$$\left(\frac{\hat{\mu}}{\hat{k}}-\frac{\mu}{k}\right) U_c + (\hat{\rho}-\rho)g = 0, \tag{65}$$

provided that perturbation wavelengths are greater than

$$\lambda = 2\pi\left\{\sigma_1 \Big/ \left[\left(\frac{\hat{\mu}}{\hat{k}}-\frac{\mu}{k}\right)(U-U_c)\right]\right\}^{\frac{1}{2}}. \tag{66}$$

This derivation has several limitations. For example, factors such as interfacial films have not been considered. As a result, quantitative agreement with real systems may not be possible. Nevertheless, the qualitative conclusions regarding this problem are valid for the model envisaged, which is simple mathematically and physically realistic.

One further point bears mention: that is, the phenomenon of negative interfacial tension (Davies & Haydon 1957). In this case, $\sigma_1-\sigma_0$ is negative and the interface could be unstable even if the system is at rest.

Discussion

As already mentioned, the stability of liquid layers in porous media is of interest in the study of a number of practical problems. In the present instance, particularly for studies regarding emulsification, it was felt that surface tension was a more dominant parameter. Thus the analysis presented here considers in detail the motion of a slab of liquid with the movement of the surrounding liquids being neglected. The motion of these liquids can be taken into account. Modifications of the above analysis to include the effects of the motion of the surrounding liquids have been considered by Raghavan & Marsden. This then would lead to the consideration of fluids of varying viscosity in a porous medium.

Comparison must also be made of the results presented here and those obtained by Keller & Kolodner (1954). Even though the expression for the radius of the droplet as given by (42) is the same, there are many basic differences. Both the physical situations and the dynamic boundary conditions are different. Keller & Kolodner analyze the situation for an inviscid fluid and thus consider inertial terms whereas here we consider Darcy flow which implies viscous flow with neglect of inertial terms. The growth factors obtained in both analyses are also different.

The drop formation aspect of the study needs further clarification. Due to the non-linear character of the problem, this study like all similar ones has been based on a linearization technique. As a consequence, the results obtained during the post-instability situation are only approximate. However, the utility of the higher-order analysis is not apparent at this time.

The present analysis has indicated that, as instability sets in, liquids are in tangential motion relative to each other. This could lead to instabilities of the Helmholtz type. Also the stability of these lamallae (fingers) which are formed can be further studied by approximating them to cylindrical threads. Studies of the two instabilities mentioned above have also been made by Raghavan and have been described elsewhere.

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REFERENCES

CHANG, C. T. 1959 Dynamic instability of accelerated fluids. *Phys. Fluids*, **2**, 656.

CHUOKE, R. L., VAN MUERS, P. & VAN DER POEL, C. 1959 The instability of slow, immiscible, viscous liquid–liquid displacements in permeable media. *Trans. AIME* **216**, 188.

DAVIES, J. T. & HAYDON, D. A. 1957 Spontaneous emulsification, gas–liquid and liquid–liquid interface. In *Proceedings of the 2nd International Congress of Surface Activity*, vol. I, p. 417. Butterworth.

DIETZ, D. W. 1953 A theoretical approach to the problem of encroachment and by-passing edge water. *Proc. K. Ned. Akad. Wet.* B **56**, 83.

JEFFREYS, H. 1926 The stability of a layer of fluid heated below. *Lond. Edinb. Dubl. Phil. Mag.* II, Ser. 7, 833.

KELLER, J. B. & KOLODNER, I. 1954 Instability of liquid surfaces and formation of drops. *J. Appl. Phys.* **25**, 913.

KIDDER, R. E. 1956 Motion of the interface between two immiscible liquids of unequal density in a porous solid. *J. Appl. Phys.* **27**, 867.

LANDAU, L. D. & LIFSHITZ, E. M. 1959 *Fluid Mechanics*, vol. 6: *Course on Theoretical Physics*. Pergamon.

LEWIS, D. J. 1950 The instability of liquid surfaces when accelerated in a direction perpendicular to their planes. II. *Proc. Roy. Soc.* A **202**, 192.

MILLER, F. G. 1962 Theory of unsteady state influx in linear reservoirs. *J. Inst. Petrol.* **48**, 365.

OUTMANS, H. D. 1962 Nonlinear theory for frontal stability and viscous fingering in porous media. *Soc. Petr. Engrs J.* **8**, 165.

RAGHAVAN, R. 1970 The instability of liquid–liquid interfaces and model systems for emulsion formation in porous media. Ph.D. Dissertation, Stanford University.

RAGHAVAN, R. & MARSDEN, S. S. 1971 Theoretical aspects of emulsification in porous media. To be published in *Soc. Petr. Engrs J.* **11**.

RAYLEIGH, LORD. 1899 On stability and instability of certain fluid motions. *Scientific Papers*, vol. 1. Cambridge University Press.

SAFFMAN, P. G. & TAYLOR, G. I. 1958 The penetration of a fluid into a porous medium or Hele-Shaw cell containing a more viscous liquid. *Proc. Roy. Soc.* A **245**, 312.

TAYLOR, G. I. 1950 The instability of liquid surfaces when accelerated in a direction perpendicular to their planes. I. *Proc. Roy. Soc.* A **201**, 192.