Drying as an Immiscible Displacement Process with Fluid Counterflow

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(Received 26 February 1987)

Experiments with refractive-index–matched materials show that evaporation of a liquid from a porous medium produces a modified form of immiscible displacement. In this process the displaced fluid is removed from the pore structure by counterflow in the same pore channels as the displacing fluid. Pressure gradients that arise from fluid flow result in displacement fronts that, unlike conventional displacement fronts, are inherently stable irrespective of the relative viscosities of the fluids.

PACS numbers: 47.55.Mh

Immiscible displacement (ID) of liquids in porous media has been extensively studied as an interesting problem in hydrodynamics and as a simple model for secondary oil recovery. In ID, a liquid is forced into the medium, displacing and pushing ahead a second immiscible liquid that originally fills the pores. At slow displacement rates, local capillary effects control the movement of the menisci separating the two liquids, and ID can be modeled as invasion percolation.[1-4] At larger displacement rates, the pressure gradients that arise in ID affect the stability of the displacement front. Pressure gradients developed when a viscous phase is displaced by a less viscous phase produce instability and fingering of the front.[5,6] With a more viscous displacing liquid, the displacement front stabilizes and forms a linear front with characteristic width.[7-9] The transition from capillary to viscous-flow–dominated behavior can be characterized by the dimensionless capillary number ($N_{\rm cap}$), a measure of the relative importance of viscous and capillary forces. Expressed in terms of the Darcy velocity (v) and viscosity (η) of the viscous phase, $N_{\rm cap}=\eta v/\gamma$ where γ is the liquid-liquid interfacial tension. On the basis of percolation effects, Wilkinson has argued that the width of the front should scale as $N_{\rm cap}$ to a power that depends on whether or not the displacing phase wets the particles.[9]

Another process that might be modeled as ID is the drying of a rigid granular medium.[10] In the drying process, evaporation causes a liquid-air interface to recede into the pore channels at the surface of the drying body creating a negative capillary pressure $P_{\rm cap}$ in the liquid. At a critical $P_{\rm cap}$, a single meniscus is forced through a restriction in the porosity and the meniscus advances to the next existing restriction. Liquid in the pores that the meniscus passes through is transported back to the external surface. Thus air invades the body from the external surface, displacing liquid back toward the surface through the partially drained region of the body.

In the present paper, it is demonstrated that the displacement front formed by a drying process is different from that formed by the usual ID process. The drying front is stable with a characteristic width even though the displacing phase (air) is the less viscous phase. The apparent anomaly is explained in terms of the pressure gradients that arise near the drying front.

The drying cell used for the experiments consisted of a homogeneous, dense, random packing of 0.5-μm-diameter silica spheres, sandwiched in a 15–20 μm×2.5 cm×4.0 cm gap between two glass slides.[10] After packing the cell with spheres and filling it with distilled water, three edges of the cell were sealed with epoxy. The drying was initiated by allowing the water to evaporate from the remaining open edge. Close matching of the refractive index of water and the spheres allowed the progress of drying to be monitored visually. When filled with water, the bed of particles was transparent. As air invaded the packing during drying, the cell became opaque. This allowed the propagation of the drying front to be directly imaged in an optical microscope using transmitted light and either photographed or recorded with a video camera. For analysis, photographs were digitized with a Data Copy 910 camera driven by an IBM PC/XT microcomputer.

A typical portion of the drying front is shown in Fig. 1, where the front is seen advancing from the top to the bottom of the image. The dark regions are areas of drained porosity that have emptied by menisci advancing into the pores from the open edge of the cell. At this magnification, the drained porosity has a highly irregular fractal structure that resembles a two-dimensional percolation cluster. In the lower-magnification image, Fig. 1(b), an upper limit to the length scale of irregularities in the front is apparent. Above this length scale, the drained porosity forms a linear front. The linearity of the drying front indicates that it is stable and resistant to viscous fingering.

In order to identify the mechanism by which liquid is transported to the open edge of the cell, the rate of advance of the front was compared with the rate of advance of a liquid-vapor meniscus formed in a cell that contained no particles. Under comparable conditions, the meniscus in the empty cell moved 1 order of magnitude slower than the drying front in the filled cell. The primary difference between the two experiments is that in the empty cell the liquid can only be transported to the open edge by vapor transport, whereas in the particle layer the liquid retained in crevices at particle contacts

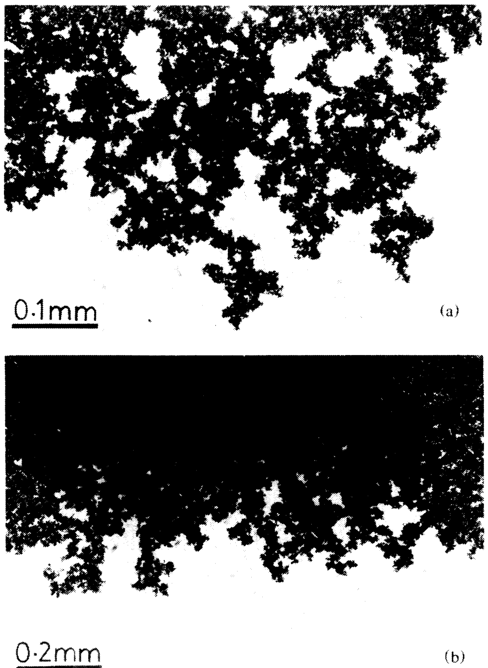

FIG. 1. (a) Portion of a drying front moving through a thin layer of silica spheres. Drained porosity appears dark in the transmitted light image. (b) In the lower-magnification image, it is evident that the front is stable.

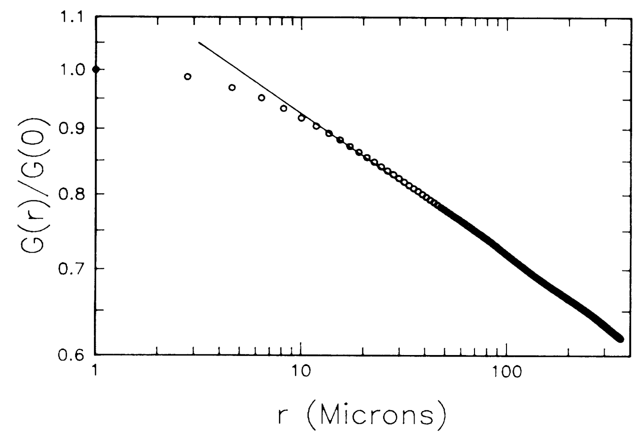

FIG. 2. Averaged two-point correlation function obtained from ten gray-scale images of portions of a drying front. A typical example of one of the images used in the analysis is the one shown in Fig. 1.

and in filled pores can transport liquid through the partially drained region by fluid flow. The more rapid movement of the front in the filled cell implies that fluid flow plays an important role in the transport of liquid from the front to the open edge of the cell. Near the front, where the saturation is highest, fluid flow is probably the dominant transport mechanism. The movement of the front is thus envisioned to take place by a counterflow process in which air mixed with some water vapor flows in from the open edge of the cell to replace the water drained from pores on the front. The displaced water then flows back out to near the open edge of the cell where evaporation occurs. The counterflow process is analogous to the removal of a wetting liquid from a square capillary tube that is plugged at one end, by a process in which a central finger of gas displaces the liquid to the open end of the tube along channels of liquid retained at the corners of the tube.

The mass fractal dimension of the drained pores near the leading edge of the drying front was determined from their autocorrelation plots. The plots were obtained from ten 512×512 digitized gray-scale images of the front from which the background contrast had been subtracted and in which larger values corresponded to darker image contrast. With the assumption that the light scattering is roughly proportional to the number of pores drained through the thickness of the particle layer, intensity in the gray scale provides a measure of the number of pores drained through the thickness of the sample. The maximum length scale used for the analysis was selected so that it was below the cutoff length scale associated with the width of the front. A typical example of an image used for the analysis is the largest square section of image that can be extracted from Fig. 1(a). The averaged autocorrelation plot, Fig. 2, is linear over a wide range of length scales. From its slope, which is equal to the difference between the Euclidean and fractal dimensions,[11] the mass fractal dimension for the drained pores is determined to be 1.89 ± 0.03. The nonlinearity of the correlation plot at short length scales indicates a transition to three-dimensional behavior at length scales on the order of the thickness of the particle layer. The edge of the front was also located from digitized images by setting a threshold gray level to select the cluster of drained pores and then by the determination of the external perimeter of the cluster. Autocorrelation plots, similar to the one in Fig. 2, indicate that the leading edge of the front has a fractal dimension that is insensitive to the threshold used to locate the perimeter with a value of 1.38 ± 0.02.

In the drying process, undrained regions cannot become trapped, as in a conventional ID process,[4] because the continuity of the liquid in the partially drained pores allows isolated clusters of undrained pores to drain. Without trapping, invasion percolation produces clusters with the same fractal dimensions as a percolation cluster.[4] My measured dimensions are in excellent agreement with those expected for the mass[12] and external perimeter[13] of a 2D percolation cluster. The agreement indicates that, at short length scales at least, movement of the drying front is dominated by local capillary effects as in a conventional ID process.

The drying front advances by the abrupt addition of large clusters of drained pores to the front, rather than single pores as in the invasion percolation. This behavior suggests that the pressure on breaking though a pore channel does not drop to a low value and then gradually build up again (as implied in the invasion-percolation model), but rather is retained at a high value that allows the menisci to sweep through a large number of restrictions before becoming pinned again. A possible source for locally maintaining pressure is the connected liquid in the partially drained pore channels. Although this phenomenon would alter the sequence of pore drainage, eventually, after the pressure builds back up to that which initiated the original breakthrough, the same pores are drained as would be in normal invasion percolation.

The most interesting feature of the drying front is its stability. Drying is a process where a less viscous nonwetting fluid (air) displaces a more viscous wetting fluid (water). In our experiments the viscosity ratio of water to air is approximately 50 and $N_{cap} \simeq 1 \times 10^{-4}$ at the fastest drying rates. At comparable N_{cap} and viscosity ratios, ID produces unstable displacement fronts that finger.[14,15] The formation of a stable displacement front in the drying experiments, therefore, differs markedly from the behavior in conventional ID experiments.

The stability of the drying front can be attributed to the counterflow of air and water in the pores. In the drying process, liquid displaced from pores on the front flows from the pores back towards the edge of the cell where it evaporates. As a result the pressure in the liquid, on average, decreases from the leading edge of the front to the open edge of the cell. The pressure gradient in the air, if any, is such that the pressure increases from the leading edge of the front to the open edge of the cell as the air flows from the edge of the cell to the drained pores on the front. The capillary pressure, defined as the difference in pressure between the air and the liquid, must, therefore, decrease from the edge of the cell to the leading edge of the drying front. The driving force to advance menisci on the front, P_{cap}, is consequently lower for menisci at more advanced positions on the drying front. A gradient in P_{cap} of this form is known to produce a stable displacement front with a characteristic width[8,9] as observed in the drying experiment. It is interesting to note that in this discussion no assumption is made about the relative viscosities of the phases. It is expected that when ID occurs by a counterflow process, the displacement front is stable irrespective of the relative viscosities of the fluids.

Stabilization of the drying front by fluid flow would be expected to cause the width of the front to decrease as the average front velocity and, hence, the gradient in P_{cap} across the front increases. During an experiment, the average velocity of the drying front decreases as the square root of time, and so the velocity of the front drops by 1 order of magnitude during an experiment. Images taken at different times in a single experiment can therefore be used to examine how velocity affects the width of the front. The perpendicular distance between the most advanced and least advanced points on the leading edge of a front was used as a measure of the width. A 5-mm-long segment of the front, corresponding to the width of the video image, was used for each measurement. A scatter plot of the width of the front versus its average velocity in single experiment is shown in Fig. 3. A least-squares fit indicates that the width of the front varies as the velocity to the power -0.48 ± 0.1. Qualitatively, the trend of decreasing width with increasing velocity is that which is expected.

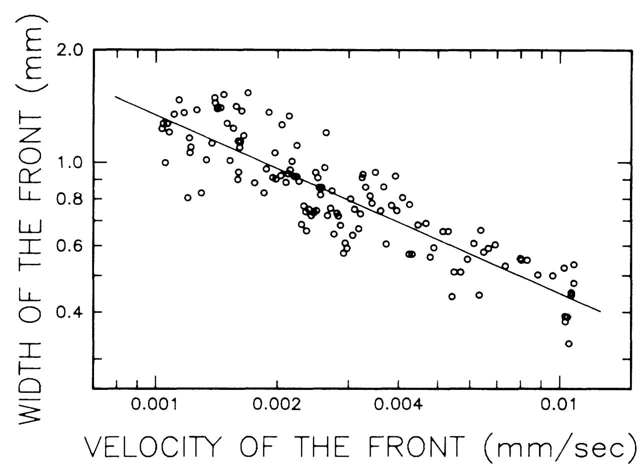

FIG. 3. Variation of the width of the drying front with its average velocity. A least-squares fit to the data gives an exponent of -0.48 ± 0.1.

Wilkinson[8,9] has analyzed the effect that a pressure gradient has on the width ξ_f of a stable displacement front using a mean-field approach. For ID by a more viscous nonwetting liquid, he predicts scaling of the form

$$\xi_f \propto V_f^{-\nu/(t-\beta+1+\nu)}, \tag{1}$$

where V_f is the average velocity of the front, and t, β, and ν are the usual conductivity, percolation-probability, and correlation-length exponents for percolation, respectively. For accepted values[12] of t, β, and ν, the exponent has a value of 0.4 in two dimensions. The dependence of the scaling exponent on t and β results from the fact that the gradient in P_{cap} across the front is controlled by fluid flow in the percolation cluster formed by the displacing fluid. With counter flow, similar scaling would be expected if the displacing phase is the more viscous phase. In the case of the present experiments, the gradient in P_{cap} across the drying front is controlled by fluid flow in the higher-viscosity nonpercolating phase (water) and not the percolating fluid (air). We therefore expect a different scaling behavior from that in Eq. (1). If we assume as a first approximation that the pressure gradient in the water is constant across the width of the front and that the pressure gradient in the air can be neglected, then we expect $dP_{\text{cap}}/dx \propto V_f$. Wilkinson has shown[8] that for a constant pressure gradient, as in displacement in the presence of gravity, the width of the front scales as

$$\xi_f \propto (dP_{\text{cap}}/dx)^{-\nu/(1+\nu)};$$

hence,

$$\xi_f \propto V_f^{-\nu/(1+\nu)}.$$

The scaling exponent $-\nu/(1+\nu)$ has a somewhat larger value of -0.57 (2D). The scatter in the experimental data in Fig. 3 is too large to identify the exponent unequivocally; however, the value of -0.57 lies within the experimental uncertainty of the data. The somewhat lower exponent of -0.48 ± 0.1 obtained by a least-squares fit suggests that pressure gradient does vary across the width of the front but with a weaker power dependence than that implied in Eq. (1).

In summary, we have found experimentally that the process of drying a granular porous material can be modeled as a modified form of ID in which the displaced liquid is removed from the pore structure by flow counter to the displacing fluid. At slow drying rates and short length scales, capillary pressure effects dominate the movement of the front as in a conventional ID process, and the movement of the front can be modeled as invasion percolation without trapping. At faster drying rates and long length scales, capillary pressure gradients, arising from the counterflow of the fluids in the pore structure, stabilize the displacement front even though the displacing fluid is the less viscous of the two fluids. As fluid flow is no longer controlled by the percolating phase in the case of a drying front, we expect the scaling behavior to differ from that of a stable front produced by a conventional displacement process.

I am grateful to Richard Voss for allowing me to adapt his programs for the analysis and to George Onoda, David Clarke, and Robert Cook for critically reviewing the manuscript.

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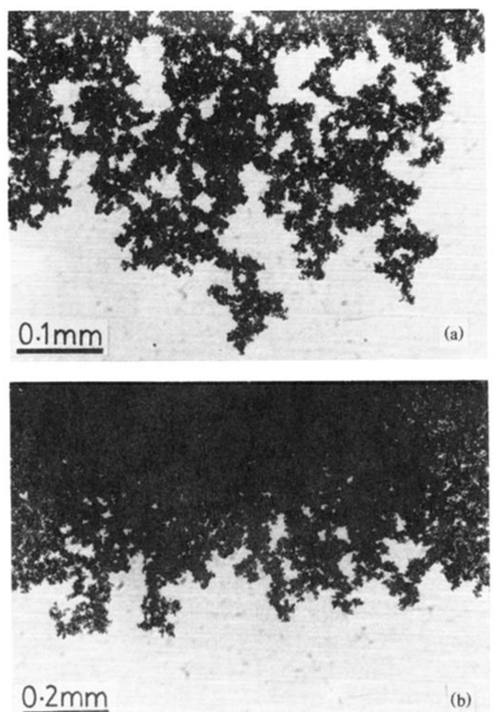

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