Quantitative Analysis of an Invading-Fluid Invasion Front under Gravity

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(Received 18 April 1988)

We report a detailed quantitative comparison between experimental measurements of the structure of a nonwetting fluid invasion front into a porous medium and a gradient percolation model accounting for the influence of gravity. The 2D correlation function of the invaded area in horizontal cuts follows a universal behavior at short distances in the fractal domain in good agreement with numerical predictions. Variations with the cut height of the mean saturation and of the crossover length between the fractal and the Euclidean domains can also be fitted precisely with theoretical gradient percolation curves.

PACS numbers: 47.55.Mh, 64.60.Ak

The flow of two nonmiscible fluids through porous media is one of the real-world problems where the techniques of statistical and disordered media physics have proved to be most successful. Such concepts as percolation, diffusion-limited aggregation, and fractal geometries have been applied to these phenomena and verified accurately in 2D model networks.[1,2] Up to recently, however, few experiments on more realistic 3D porous media have been performed[3-8] and no full quantitative comparisons with a theoretical model have been realized to our knowledge. In this paper, we present a quantitative analysis of an experimental invasion front using a "gradient percolation" model to account for the influence of gravity. We shall show that the numerical results permit a detailed analysis of the experimental front structures.

The "invasion percolation" model has been previously suggested[9-15] to describe the invasion of a porous medium by nonwetting fluid at a low velocity and under zero gravity. In this approach, the pore volume is modeled as a disordered lattice of sites (pores) linked by bonds (connecting channels). At an injection pressure Π, only a fraction of $p(\Pi)$ of all channels (or pores) can be invaded: Those with a radius r large enough so that Π overcomes the corresponding capillary pressure $\Pi(r)$. The sample only gets invaded in volume above a minimum value Π_c of Π (equivalent to the threshold value p_c for the percolation parameter p). For $\Pi=\Pi_c$, the invaded volume has a fractal geometry analogous to that of a percolation cluster.

In real life, it is not possible to neglect the influence of gravity in most 3D systems. The hydrostatic component adds to the applied injection pressure Π_a and creates a vertical gradient in the effective injection pressure Π. Therefore, the fraction of accessible pores decreases with height and the experimental results cannot be compared with classical percolation. This missing gradient term has been recently introduced[16,17] and the concept of gradient percolation has been proposed to analyze 2D and 3D diffusion processes. We show in this paper that gradient percolation (GP) describes precisely invasion fronts obtained under gravity (IFG). In particular, we show that experimental IFG measurements are very similar to numerical data obtained from GP simulations.

The GP model differs from ordinary percolation on periodic lattices by a linear variation (Δp per intersite distance) of the percolation parameter p between 0 and 1 along a lattice axis. As a consequence, there is a "frontier region" connected to the $p=0$ or $p=1$ lines by a continuous chain of free or occupied sites, respectively. This "frontier" is equivalent to the front surface limiting the invaded volume in the invasion experiment. This is an essential point in the comparison between GP and IFG.

The experimental injection system and image analysis procedure in the IFG has been described elsewhere[5,6]: A low-melting-point liquid alloy (Wood's metal) is injected at the bottom of a vertical evacuated crushed-glass column of 10 cm inner diameter. The flow velocity U is low (a few millimeters per hour) so that viscous pressure losses can be neglected and the capillary number Ca is $\ll 1$ ($\mathrm{Ca}=\eta U/\gamma$, η is the liquid viscosity and γ the surface tension). When the front has reached a given height, the injection is stopped and the liquid is solidified; then horizontal sections of the front corresponding to various heights z are analyzed. The pictures are digitized into a square lattice of pixels and "invaded" or "empty" pixels are discriminated against by a threshold procedure. We then determine the correlation function $C(r)$ of the metal distribution in horizontal planes; $C(r)$

is the fraction of invaded pixels at a distance r from an occupied origin site [$C(r)$ is averaged over the origin pixels for which *all* points at the distance r are within the picture frame.]

In the same manner $C(r)$ is determined for the numerical simulation for sites located on the GP frontier: It is the key tool for analysis of GP and IFG data. In both cases, close to p_c in a range of r values between the individual grain size d (which approximately corresponds to the lattice constant a in the GP model) and an upper limit λ (crossover length),[18] $C(r)$ varies roughly as

$$C(r) \propto r^{D_{\rm fr}-2}. \tag{1}$$

This seems to show that the cut structure is fractal with a dimension $D_{\rm fr}$ at short distances. When r is between λ and the sample size, $C(r)$ has a constant value S proportional to the mean metal saturation $S_{\rm mw}$. As in similar 3D experiments,[4-8] the values of $D_{\rm fr}$, λ, and S cannot be directly compared with classical percolation predictions since the percolation parameter gradient must be taken into account.

Let us now discuss the equivalence between the different parameters used in the IFG and GP studies. The invading-fluid saturation S, the cut height z (related to the fraction of accessible pores), and the grain size d correspond respectively to the front concentration p_f, the percolation parameter p, and the lattice constant a. More precisely, the lattice constant a of the cubic lattice considered in GP is associated with an average distance between pores in IFG. This distance is, in practice, proportional to, and of the same order of magnitude as d. Unless otherwise stated, in the following we shall consider all distances to be measured in lattice units a. The relation between z and p introduces two parameters: the critical height z_c corresponding to the value $p=p_c$ of the percolation parameter and the local slope dp/dz in the vicinity of p_c. At this point an important question is the relation between the cut height z and the local value of the percolation parameter p. At low velocities, the effective injection pressure Π varies linearly with z because of the hydrostatic pressure gradient. The relation between Π and the fraction p of accessible pores depends on the pore size distribution: It cannot be determined easily and is almost certainly not linear over the whole range $p=0$ to 1. We shall, however, assume that the relation is linear close to $p=p_c$. Hence

$$p-p_c \cong (dp/dz)_c(z-z_c) = (\nabla p/a)(z-z_c).$$

Finally the local fraction p_f of sites located on the frontier in GP must be proportional to the fraction S of occupied pixels in the digitized pictures. The proportionality coefficient between S and p_f depends on the image thresholding procedure and on the pore geometry. Let us now make scaling-type predictions for the dependence of $C(r)$ on r and the cut height z. In the presence of gravity, there are two different characteristic lengths in the problem[16]:

In horizontal planes, $z=$const, the characteristic length is the percolation correlation length

$$\xi(p) \propto (p-p_c)^{-\nu} \tag{2}$$

corresponding to the value of p in this plane. The critical exponent ν has the value 0.88 of classical 3D percolation. Relation (2) is equivalent to $\xi(p) \propto (z-z_c)^{-\nu}$ if the relation between z and p is linear close to $p=p_c$.

In the vertical direction parallel to the flow, the gradient percolation front has a scaling behavior depending on a single length δz, which (to a constant) represents the maximum spatial extent of the fractal domain.[16] δz varies with the probability gradient $|\nabla p|$ as (a being taken as the unit length)

$$\delta z \cong |\nabla p|^{-\nu/(1+\nu)}. \tag{3}$$

This result can also be obtained from the assumed linearity between z and p,

$$\delta p/\delta z = |\nabla p|/a,$$

and, from Ref. 16,

$$\delta p = C|\nabla p|^{1/(1+\nu)}. \tag{4}$$

The existence of the length scale δz is due to a self-limitation of the divergence of the correlation length ξ because of the variation of p with z. If we use the two scales δz and ξ to define reduced variables, $(z-z_c)/\delta z$ and $r/\xi C(r)$ can be written from Eq. (26) in (Ref. 16)

$$C_2 r^{3-D_f} C(r) = \Gamma\left[\frac{C_1 r}{(z-z_c)^{-\nu}}, \frac{z-z_c}{\delta z}\right], \tag{5}$$

where r is measured in pixel units R. $C_1 = R|\nabla p|^{\nu}/a^{(1+\nu)}$ and $C_2 = (R/a)^{3-D_f} p_f/S$ are taken as adjustable parameters.

A key result obtained through the GP[16] is that, at short distances r, $r^{d-D_f}C(r)$ is determined by the local fractal structure of the "central" cluster inside which the origin occupied site is located. Then, at small r, $r^{d-D_f}C(r)$ depends only on r/ξ with a universal law independent of $p-p_c$. Figure 1 shows the variation of the corresponding quantity $C_2 r^{d-D_f}C(r)$ for the IFG as a function of $C_1 r/(z-z_c)^{-\nu}$ for 25 different cut heights. There is a striking similarity with results from the GP simulation (see Fig. 7 in Ref. 16). As in the GP, the experimental value of $r^{d-D_f}C(r)$ indeed follows a universal law independent of z up to a crossover length λ which increases with the cut height z (when the mean saturation gets lower). In the limit $r \to 0$, $r^{d-D_f}C(r)$ becomes (as expected) a nonzero constant and $C(r) \propto r^{D_f-3}$ independently of the value of ∇p. D_f should therefore be equal to the fractal dimension of the 3D percolation cluster; the best fit is obtained for $D_f=2.4$ which is slightly lower than the value $D_f=2.5$ for classical 3D percola-

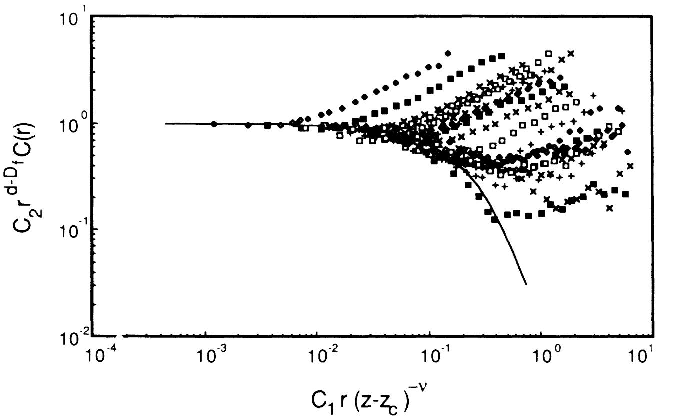

FIG. 1. Variation of $r^{d-D_f}C(r)$ for experimental data obtained from 25 horizontal cuts of a Wood-metal invasion front in crushed glass: We have used five different symbols to separate data at different cut heights. For the parameter values we used $C_1=4.5\times10^{-3}$, $C_2=1.5$, $z_c=25.5$ mm (absolute height for which $p=p_c$), and $D_f=2.4$.

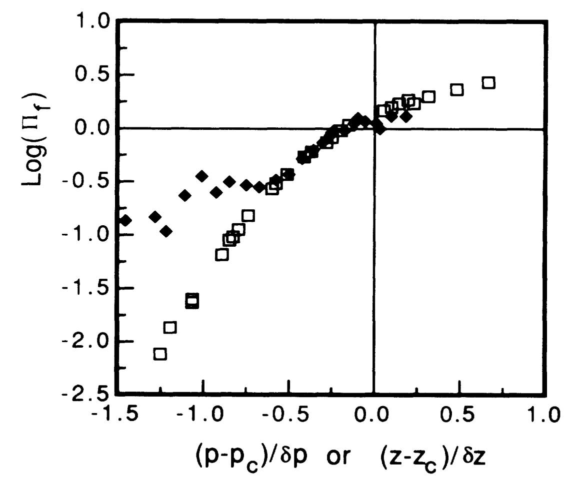

FIG. 2. Variations of the mean invading-fluid saturation S in the experimental horizontal cuts (diamonds) and the fraction p_f of sites located on the numerical gradient percolation front (squares). The variations are plotted as functions of a reduced variable corresponding to the cut height. An adjustable parameter has been introduced to represent the proportionality constant between the actual saturation and the digitized images.

tion.[19] At longer distances $r>\lambda$, the value of $C(r)$ is determined by farther parts of the clusters in the cut. As in the GP, $C(r)$ tends towards an asymptotic constant value of $C(\infty)$ and all curves in Fig. 1 become parallel straight lines of slope $d-D_f$. $C(\infty)$ corresponds (within a constant factor) to the fraction of occupied pixels at the corresponding height (in the GP, the limit value is the fraction $p_f<p$ of sites located on the frontier).

The correlation function $C(r)$ provides a detailed characterization of the front structure; on the other hand, averages over horizontal sections of such physical quantities as the invading-fluid concentration or the crossover length λ are more easily measured experimentally and more relevant to practical applications. Let us take for instance the mean metal saturation S_{mw} in the IFG (proportional to the fraction S of "occupied" pixels). It is equivalent to the fraction p_f ($<p$) of sites located on the frontier at a given height in the GP. The only relevant reduced variable is $(z_c-z)/\delta z$ [or $(p-p_c)/\delta p$] since r/ξ has been averaged out. In the limit $|\nabla p|\to 0$, $p>p_c$, one retrieves the results of classical percolation[16] and p_f' must be independent of ∇p and proportional to $(p-p_c)^\beta$. Thus, $p_f'=p_f/p$ should verify[16]

$$p_f'=p_f/p=|\nabla_p|^{\beta/1+\nu}\Pi_f[(p-p_c)/\delta p]. \qquad (6)$$

The values of S obtained from the experimental system must follow similar scaling laws. In order to compare the variations of p_f and S we plot p_f as a function of the reduced coordinate $(p-p_c)/\delta p$ for the GP, and S as a function of the corresponding quantity $(z_c-z)/\delta z$ for the IFG. Then we assume that

$$p_f'/(\delta p)^\beta=KS. \qquad (7)$$

The fit shown in Fig. 2 gives $K=9.5$ (in good agreement with the value $C_2=1.5$ found above) and $\delta z=14$ mm. We have used the same value for z_c (25.5 mm) obtained from the fit on $C(r)$. One observes that the experimental and numerical curves are in good agreement for $-0.5\le(p-p_c)/\delta p\le 0.1$; for $(p-p_c)/\delta p<-0.5$ S is approximately constant whereas p_f decreases to 0. We interpret this discrepancy as arising from the presence of a few channels of high permeability inside the crushed glass. The Wood metal can more easily rise through them than the rest of the material: This can explain the presence of a small residual saturation ahead of the main front.

Let us now put $\delta z=14$ mm in Eq. (3). We obtain $|\nabla p|\cong 1/8700$ for $\nu=0.88$ (classical 3D percolation value) for $a=200$ μm (the mean grain size is taken here to be the lattice constant a). This $|\nabla p|$ value would correspond to a height variation $\Delta z\cong 1.75$ m for the overall variation from $p=0$ to $p=1$. This value is unrealistically high compared with the overall vertical extent H of the front (a few centimeters). The most probable origin for the discrepancy is that the above evaluation assumes that the fraction p of accessible pores increases linearly with the effective injection pressure (and therefore with z). Δz is in fact the inverse of the local derivative $dp(z)/dz$ of the fraction of accessible pores around $p=p_c$. Because of the complex geometry of the crushed-glass grains, the function $p(z)$ is certainly not linear and very large variations of $dp(z)/dz$ may exist. Some information on this quantity could be obtained from a porosimetry-type measurement and by recording the pressure variations during the invasion of a similar sample.

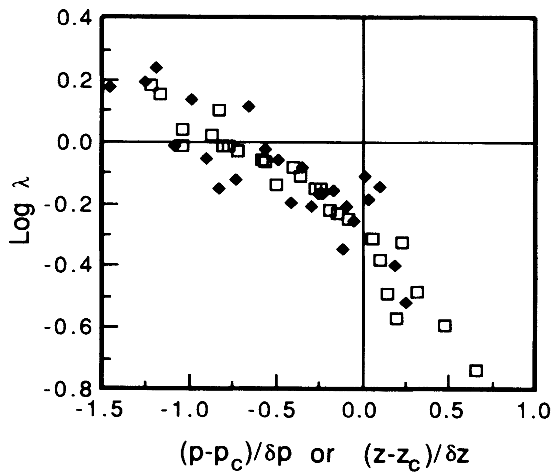

FIG. 3. Compared variations of the crossover length λ between the Euclidean and fractal domains in horizontal cuts of the theoretical GP front (squares) and the experimental invasion front (diamonds).

We have also compared the experimental and numerical variations of the crossover length λ with height. As above, the reduced variables $(p-p_c)/\delta p$ for the GP and $(z_c-z)/\delta z$ for the IFG will be used. λ verifies scaling relations similar to Eqs. (6) and (7). Figure 3 shows the compared variations of λ in the experimental and numerical situations after they have been normalized to the same value for $p=p_c$. Here again, good agreement is obtained between the two series of data.

In Refs. 5 and 6 values of a "fractal dimension" $D_{\rm fr}$ obtained from the mean slope (in log-log coordinates) of the variation of $C(r)$ with r in the domain $r<\lambda$ are also reported. It appears from Fig. 1 that the slope of $C(r)$ when $r\to 0$ is a constant equal to the fractal dimension of a classical 3D percolation cluster. $D_{\rm fr}$ as defined in Refs. 5 and 6 is then just a mean slope of the universal (decreasing) variation of $r^{d-D_f}C(r)$ between $r=0$ and $r=\lambda$ (λ is itself a decreasing function of the percolation parameter p). $D_{\rm fr}$ reported in Ref. 6 is a complex parameter which is equal to D_f only when $p=p_c$ (or $z=z_c$).

In conclusion, we have shown in this paper that the gradient percolation model accounts *quantitatively* for experimental observations of the structure of a Wood-metal invasion front in homogeneous crushed glass. The variations of the 2D correlation function of the metal repartition in horizontal cuts performed at different heights have been studied. They are in excellent agreement with the numerical simulation results for realistic values of the geometric adjustable parameters of the model; however the value $D_f=2.4$ for the fractal dimension of the front in the limit of small distances is smaller than the value 2.5 expected for classical 3D percolation. The variations of the mean fluid saturation with the cut height in the medium and high saturation zones are also in good agreement with the predictions, in particular for the crossover length λ between the fractal and the constant saturation domains. More experiments with different values of the percolation parameter gradient will, however, be needed to further establish the model's validity. They could be obtained, for instance, by use of fluids with a lower density contrast.

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