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LETTER TO THE EDITOR

The effect of morphological disorder on viscous fingers and diffusion-limited aggregates in a porous medium

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Abstract. We investigate the growth of viscous fingers (VF) and diffusion-limited aggregates (DLA) in a disordered porous medium. We propose that the DLA model and VF are *not* equivalent in a porous medium with topological disorder as represented by a percolation system and suggest new universality classes for VF and DLA in such media. We also present evidence that, for a wide class of porous media with geometrical disorder (i.e. with pores of random sizes), the DLA model and VF are *not* equivalent, even though they have often been characterised by approximately the same fractal dimension. Moreover, VF appear to be sensitive to the local properties of the medium such as the pore size distribution. Therefore, the DLA model cannot be used to simulate VF in disordered porous media and to predict the efficiency of the displacement process.

The displacement of a viscous fluid by a less viscous fluid is often unstable (for a review see Lenormand (1986) and references therein). The instability is manifested by the formation of a multitude of fingers of the displacing fluid (DF), which grow at relatively large speed, leaving behind a large amount of the displaced fluid. The traditional approach for describing such fingers is to use continuum flow equations, based on Darcy's law for creeping flow. Darcy's law states that in a porous medium the fluid flux U is proportional to the gradient of the pressure P:

$$U = -\frac{k}{\mu}\nabla P \tag{1}$$

where μ is the viscosity of the fluid and k is the permeability of the medium. For incompressible fluids $\nabla \cdot U = 0$ and therefore

$$\nabla \cdot (k\nabla P) = 0. \tag{2}$$

For a uniform system k is a constant and, therefore, the pressure P satisfies the Laplace equation. If the viscosity of DF is much smaller than that of the displaced one, then the pressure everywhere in the DF is constant and the pressure in the displaced fluid satisfies (2), with the boundary condition that P is also a constant far from the interface. In the absence of any interfacial tension between the two fluids, the interface would move with the velocity U/ϕ, where ϕ is the porosity of the medium.

Paterson (1984) pointed out the analogy between the problem of displacement of viscous fluids and the diffusion-limited aggregation (DLA) model of Witten and Sander (1981). In this model particles are added to a growing aggregate using random walk trajectories. This analogy between DLA and viscous fingers (VF) is appropriate when

the medium is uniform and the mobility ratio M, i.e. the ratio of the viscosities of the displaced and displacing fluids, is infinite. While there have been several recent studies of VF, when M is finite, and their relation with the DLA model (King and Scher 1985, DeGregoria 1985, Sherwood and Nittmann 1986, Siddiqui and Sahimi 1986), there has not been any attempt to study the relation between VF and the DLA model when the medium is disordered.

If an inviscid fluid displaces a viscous fluid in a porous medium, the interface is unstable even to small fluctuations. The source of the fluctuations is the microstructure of the medium and, therefore, if the DLA or a related model is to be used for simulating such displacement processes and obtaining quantitative predictions about the efficiency of the process, one has to develop an understanding of the relation between the DLA model and the problem of VF in a porous medium. The experiments of Chen and Wilkinson (1985) clearly demonstrate the effect of microstructure of the medium on the displacement process. As the microstructure of the medium changes from an ordered to a highly disordered one, the nature of the displacement changes from ordered (dendritic) patterns to highly unstable with fractal VF. The aim of this letter is to explore the relation between the DLA model and VF in a disordered medium. We consider systems with disordered morphology, i.e. disordered topology (connectedness) and geometry (sizes of the pores), and argue that in such porous media the DLA model and VF may not be equivalent.

Before presenting our analysis, we should develop a criterion for the equivalence of VF and the DLA model. Suppose that after a long time t, N_{VF} pores of the medium have been filled by DF and large VF have been formed. In the absence of surface tension and for $M > 1$, these VF have a fractal structure and, therefore, we can write

$$N_{VF} = a_1 R^{d_{VF}} \tag{3}$$

where d_{VF} is the fractal dimension of VF and R is the radius of gyration. If we grow these VF by the DLA algorithm, after a time t, N_{DLA} pores of the medium would be filled by the displacing fluid, and we can write

$$N_{DLA} = a_2 R^{d_{DLA}}. \tag{4}$$

We now say that VF and the DLA model are equivalent if $N_{DLA} = N_{VF}$. This means that the sweep efficiencies of the two processes, i.e. the volume fraction of the pores filled with the DF, is the same for both processes. For this to be true, we should have $a_1 = a_2$ *and* $d_{VF} = d_{DLA}$; otherwise the two processes are not equivalent and the DLA model cannot be used for quantitative modelling of VF in a porous medium. As we argue below, one or both of these conditions are often not satisfied.

We first consider the effect of random topology on DLA and VF. To this end we consider a percolating system and restrict our attention to a two-dimensional medium represented by, e.g., a square lattice in which only a fraction p of sites are present and the bonds between them have a finite conductance. In three dimensions gravity can significantly alter the behaviour of the displacement process, but if gravity is neglected, our analysis applies equally to 3D systems. For length scales L larger than the correlation length ξ_p of percolation, the system is homogeneous. However, if $L < \xi_p$, the largest cluster (LC) has a fractal character with a fractal dimension $d_p = 91/48 \simeq 1.896$. At the percolation threshold p_c, the LC is a fractal object at all length scales. Thus we first discuss the situation at p_c.

To study the DLA model on the LC at p_c, one occupies a site at the centre of the cluster and releases the random walkers on the *boundaries* of the cluster and restrict

their walks to the sites of the cluster, until they join the growing aggregate. Obviously, the aggregate can grow only on the *backbone* of the cluster, i.e. its multiply connected part, as the dead-end part of the cluster is screened by the growing aggregate. The DLA model on the LC at p_c was first studied by Sahimi and Jerauld (1983), who proposed the model as a way of probing the structure of percolation clusters and their effect on the growth of the aggregates. Meakin (1984) simulated the DLA model on the LC at p_c and estimated that $d_{DLA} \simeq 1.4$ in two dimensions, as compared to a value of about 1.7 for DLA on uniform lattices. However, in Meakin's simulations, random walkers were placed at randomly chosen sites of the cluster (instead of the boundary sites) and, therefore, Meakin's model does not have an obvious relation to what we are discussing here. We expect the true value of d_{DLA} to be even smaller than Meakin's estimate, for reasons discussed below.

To obtain an accurate estimate of d_{DLA} we develop a Flory-like approximation which has been highly successful (Hentschel 1984) in estimating the fractal dimension D of DLA on uniform systems. As usual, we write the 'free energy' F of the system as the sum of an elastic energy and a repulsive energy. To find the contribution of each, we first note that diffusion on the LC at p_c is anomalous (Gefen *et al* 1983), so that after n steps the random walker travels a distance $r \sim n^{\alpha}$, where $\alpha = (d_w)^{-1}$ and d_w is the fractal dimension of the walk, which is about $d_w \simeq 2.87$ in two dimensions. The elastic energy of the system is proportional to $R^{2(D-1)}/n^{\alpha}$, and the repulsive energy is proportional to n^2/R^{d_p}. Therefore, the free energy of the system is given by

$$F = \frac{R^{2D-2}}{n^{\alpha}} + \frac{n^2}{R^{d_p}}. \tag{5}$$

If we minimise F with respect to R and note that $R \sim n^{\beta}$, where $\beta = (d_{DLA})^{-1}$, we obtain

$$d_{DLA} = (2D + d_p - 2)/(2 + 2\alpha). \tag{6}$$

Therefore, with $D \simeq 1.7$, $d_p = 91/48 \simeq 1.896$ and $\alpha \simeq 0.3479$, we obtain

$$d_{DLA} \simeq 1.23 \tag{7}$$

which is much smaller than D. Based on the success of the Flory theory for DLA on uniform lattices, we expect this value to be an accurate estimate of the true value of d_{DLA} and simulations (Siddiqui and Sahimi 1986) also support (7); d_{DLA} is estimated to be about 1.22 (with $a_2 \simeq 1.7$). This low value of d_{DLA} is caused by the fact that for random walks on the LC at p_c the fractal dimensionality d_w is large and the diffusion process is slow. Therefore, the diffusing particles cannot penetrate the aggregate very efficiently. Moreover, the LC at p_c is made of nodes, blobs (of multiply connected bonds) and links that connect the blobs to one another. If a site at the entrance to a blob is occupied by an incoming particle, the blob will be screened and the random walkers can no longer enter the blob. We remark that a different mean-field approximation for d_{DLA} has been developed by Honda *et al* (1986), and is given by

$$d_{DLA} = (d_p^2 + d_w - 1)/(d_p + d_w - 1) \tag{8}$$

which predicts that d_{DLA} $(d = 2) \simeq 1.45$. The derivation of (8) is not based on the standard arguments that are used to derive an equation such as (6). Honda *et al* (1986) derive (8) by considering the flux of incoming particles and that portion of it that would be captured at a distance r from the centre of the aggregate. In addition, they make certain assumptions about the scaling of a screening length (or the linear dimension of the void space in the aggregate) with r.

We now consider VF on the LC at p_c. The standard procedure (King and Scher 1985, DeGregoria 1985, Sherwood and Nittmann 1986) has been to inject the displacing fluid into the origin of a large system. One solves (2) with the boundary conditions mentioned above and obtains the pressure field, based on which the interface advances into one of the adjacent pores (bonds) with a probability proportional to the flux in the bond. The pressure field is then recalculated and the procedure is repeated until a large cluster is formed. In uniform media, this is exactly equivalent to the DLA model and to the dielectric breakdown model of Niemeyer *et al* (1984). But even if the system is perfectly ordered, the random choice of the invaded pore introduces noise and fluctuations into the process and fractal VF are formed. However, the fact that even a pore with extremely small flux might be invaded by the DF means that an extremely high degree of noise dominates the system, which is in fact absent in an ordered medium. This model is also incorrect in the microscopic sense. That is, the pressure field is determined assuming the entire interface is instantaneously moving, yet only one pore is filled with the DF at a time. In reality, if several pores adjacent to the interface have similar velocities, they will be filled by the same amount, but the interface will also advance into the other pores with smaller velocities and fill a portion of them. Thus, on a uniform square grid only dendritic patterns are observed (Chen and Wilkinson 1985) which are *not* similar to DLA in the same systems. To circumvent this difficulty, one may adopt a method similar to that of Tang (1985) or Kertész and Vicsek (1986): the perimeter sites do not become part of the aggregate if they are hit only once by the incoming particles. One counts the number n_i of random walkers hitting site i of the perimeter. If n_i reaches a preassigned value m, i becomes part of the aggregate. In the method of Tang (1985), once i becomes part of the aggregate, n_j is *not* set to be zero for all other sites j of the perimeter, whereas in the method of Kertész and Vicsek (1986) one sets $n_j = 0$, once i is part of the aggregate. From a microscopic point of view the latter method is more appropriate, since setting $n_j = 0$ and recalculating them as new walkers come in correspond to recalculating a new pressure field once a pore has been filled with the DF. However, we have found (Siddiqui and Sahimi 1986) that the long-time behaviour of both models is the *same*: as m increases, the growing aggregate becomes more regular and approaches the pattern obtained by Chen and Wilkinson (1985).

We carried out simulations of VF on the LC at p_c. Most of our calculations were performed for 81×81 and 101×101 samples and the results were averaged over 10 different realisations. The pore radii were distributed uniformly in the interval $(1-\lambda, 1+\lambda)$, with λ being an adjustable parameter, which is used to change the broadness of the pore size distribution (PSD). We first solve for the pressure field. Then, in a time Δt, an interface in the pore i will move a distance, $l_i = (Q_i/\pi r_i^2)\Delta t$, where Q_i is the flow rate in the pore and r_i is its radius. Δt is chosen to be the time that it takes for exactly one interface to reach a node. We thus move *all* the interfaces according to this rule and recalculate the pressure field, and so on (see Chen and Wilkinson 1985). Figure 1 represents our results for $\lambda = \frac{1}{3}$, from which we obtain

$$\begin{aligned} d_{VF} &\simeq 1.54 \pm 0.10 \\ a_1 &\simeq 2.17 \end{aligned} \tag{9}$$

which do not agree with the DLA results, which means that the two systems are not equivalent. Similar results were obtained for other values of λ. We found that, for smaller values of λ, d_{VF} decreases which means that VF may be sensitive to the broadness

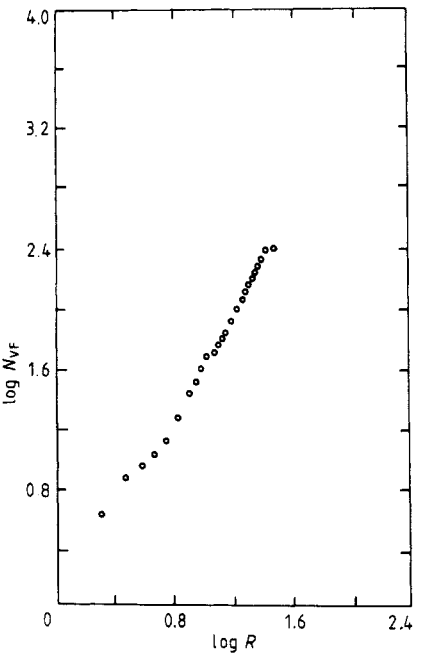

Figure 1. The log-log plot of N_{VF} against the radius R. The simulations were done at the percolation threshold and the pore radii were distributed uniformly in the interval $(1-\lambda, 1+\lambda)$; here $\lambda = \frac{1}{3}$.

of the PSD. However, within our error estimates various values of d_{VF} appeared to be consistent with one another, the details of which will be given elsewhere (Siddiqui and Sahimi 1986). The reason for the difference between d_{DLA} and d_{VF} may be due to the fact that VF fill the LC more efficiently than the DLA, because at each step several pores are invaded, whereas according to the DLA algorithm only one pore is invaded. Note that (8), within the error estimates, is very close to the fractal dimension d_{BB} of the backbone of LC at p_c, $d_{BB} \simeq 1.6$ (Sahimi 1984, Herrmann *et al* 1984).

What happens above p_c? Slightly above p_c the correlation length ξ_p is very large, and if the root-mean-squared displacement (RMSD) of the diffusing particles is less than ξ_p, one is still in the regime of anomalous diffusion. Therefore, unless $1 > p \gg p_c$ (where ξ_p is small), one would still have many random walkers whose RMSD are less than ξ_p, so that the overall effective fractal dimensionality of all random walks may be between 2 and $d_w \simeq 2.87$. Thus even for length scales $L > \xi_p$, one may have $d_{DLA} < 1.7$, and the crossover from a regime characterised by (7) to a uniform system may not be sharp, but a slow one. In contrast, for VF and for $L > \xi_p$ the system is homogeneous and there is a sharp crossover from that characterised by (8) to that of a uniform system.

Next, we consider the effect of geometrical disorder (i.e. the sizes of the pores) on VF and DLA, and pose the following question which is quite general. Are VF and DLA exactly equivalent (in the sense defined above) in a disordered porous medium (with or without percolation effects) characterised by a PSD? Most porous media are characterised by a PSD $f(r)$ (Collins 1961) which can be measured by, e.g., the automated serial sectioning technique. A heterogeneous porous medium is characterised by a distribution of permeabilities, which can drastically change the behaviour of the displacement process. It is not clear at all that the DLA model is equivalent to VF in

such media. In the percolation system discussed above, we evidently used a PSD of the following form:

$$f(r) = (1-p)\delta(r) + ph(r) \tag{10}$$

and our analysis suggested that DLA and VF are not equivalent for certain values of p and length scales L. This means that there are certain classes of $f(r)$ for which DLA and VF are qualitatively different. Therefore, a more relevant question is the following: for what class of $f(r)$ are DLA and VF equivalent (in the sense defined above)? If there is such a class of $f(r)$, one would also like to know whether it depends on the value of p.

We recall that, according to the DLA algorithm, the probability that any one pore is invaded by the displacing fluid is proportional to the flux of fluid in the pore. However, an actual displacement process in a geometrically disordered porous medium does not proceed stochastically (see above) and, even if it does, it is not clear that the interface would invade a pore with such a probability. However, our goal here is not to propose a stochastic or deterministic growth law for displacement processes in a porous medium, but to devise a method based on which one can distinguish between a DLA-like growth and an actual displacement process in a disordered porous medium.

Suppose that at any given time there are N pores available for the invasion of the displacing fluid. According to the DLA model the probability q_{DLA} that a pore i is invaded by the displacing fluid is given by

$$q_{\mathrm{DLA}} = U_i / A \tag{11}$$

where $A = \sum_{j=1}^{N} U_j$. Thus, this growth law does not directly use any information about the local microstructure of the medium. It also fills only one pore at a time. In an actual displacement, if two pores adjacent to the interface have similar velocities, then the interface will move comparable distances in both of them in any given time interval Δt, it does not select anyone of them randomly. To make further progress, we attribute all the randomness of the medium to r, the effective radius of a pore. That is, given a porous medium with a PSD $f(r)$, we can deterministically calculate all other quantities of interest. A pore i with the largest velocity among the N available pores will be the first to be filled by the DF in time Δt, i.e. if Q_i/s_i is maximum, where $s_i = \pi r_i^2$. During this time, the DF will also invade the remaining pores adjacent to the interface and fill a portion of each. If we assume that these portions are small compared to the completely filled pore, then we can derive a relation for q_{VF}, the probability that a pore i is invaded during the displacement process. Under this assumption, a simple probabilistic argument yields

$$q_{\mathrm{VF}} = \int_0^\infty S(s_i)\,\mathrm{d}s_i \prod_{j\neq i}^{N} \int_a^\infty S(s_j)\,\mathrm{d}s_j \tag{12}$$

where S is the probability density function of s and $a = s_j Q_j / Q_i$. Thus if the growth laws (11) and (12) are to be the same, i.e. if a DLA-like simulation is to yield exactly the same predictions as in an actual displacement, there has to be a probability density S (which can be easily obtained from $f(r)$) which would reduce (12) to (11). However, it is easy to see that for PSD such as (10), equation (12) cannot be reduced to (11), unless $h(r) = \delta(r - r_0)$ and $p = 1$, which explains to some extent our results with the percolating media discussed above. Of course, equation (12) is only an approximation, since we neglected the fact that the DF invades several pores and took into account

only that pore which is first filled by the DF. On the other hand, this approximation strongly indicates that the DLA model and VF are not equivalent in a disordered porous medium, because if the two models are not identical even when only one pore (with the largest velocity) is invaded at each step of the displacement process (which the above analysis indicates), we expect them to be quite different for the more general case of invading several pores. We have carried out simulations with many other PSD $f(r)$, and in all cases the predictions of the DLA models do not agree with those of VF. The results, together with further development of equation (12) when all the invaded pores are taken into account, will be given elsewhere (Siddiqui and Sahimi 1986). We may conclude that, for a general type of geometrical disorder, the DLA model may not be exactly equivalent to VF. Therefore, the exact analogy that has been asserted to exist between the DLA model and VF (see, e.g., Paterson 1984, Kadanoff 1985) does not hold for disordered porous media. As a result, for a wide class of disordered porous media, the DLA model cannot be used to predict quantitatively the properties of interest in a displacement process, such as the sweep efficiency. Moreover, the problem of developing a random walk solution to the VF problem in a disordered porous medium remains unsolved.

After the submission of this letter, and during the STATPHYS 16 conference in Boston, we became aware of two papers on the problem studied here. Murat and Aharony (1986) have simulated the DLA model and VF on percolating networks, in which all open pores have the same radius. Their estimate of d_{DLA} is consistent with ours, but they find that $d_{VF} \simeq d_{DLA}$. However, there are two differences between their simulations of VF and those discussed here. As a boundary condition, Murat and Aharony (1986) assume that the *pressure gradient* is zero on the boundary of the LC (we specify *pressure* on the boundary of the LC). More significantly, Murat and Aharony (1986) use the dielectric breakdown model to simulate VF which, as discussed above, may not be totally appropriate for simulating VF. They distinguish between the DLA model and VF by noting that the *ratio* a_1/a_2 deviates from unity. We cannot rule out the possibility that, with larger networks, our results will become consistent with theirs, although we have some indications that this will not be the case. Chan *et al* (1986) consider a model of porous media with narrow and equal tubes which are connected to one another at chambers with random volumes very much larger than the volumes of narrow tubes. They argue that the DLA model and VF are not equivalent in such a medium, unless the distribution of the volumes of the chambers has a particular form. They derive an equation somewhat similar to equation (12), which is exact for their model.

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Note added in proof. More recently, we have simulated VF and DLA in 151×151 lattices at and above p_c with distributed pore radii. We have found lower values of d_{VF} than that given by (9). However, the value of d_{DLA} appears to be in complete agreement with the prediction of the Flory theory, equations (6) and (7), and our simulations with smaller lattices. This further indicates that, in disordered porous media, VF and DLA belong to different universality classes. We have also carried out simulations with pore conductance distributions that are very broad. The results indicate that d_{VF} and d_{DLA} are very sensitive to the details of the distribution. More details are given in Siddiqui and Sahimi (1986).

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