Nonuniversality in Imbibition

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We report an imbibition experiment in 2D random porous media in which height - height correlation function grows with a nonuniversal exponent; rather it depends on evaporation. We present an imbibition model which is consistent with the experiment. The model also shows self-organisation of the interface. To our knowledge this is the first model which explicitly allows for evaporation.

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Recently there has been a lot of activity in understanding interface growth phenomena \[1\], particularly in deposition \[2\] and imbibition \[3,4\]. Imbibition experiments in which a suspension (for example coffee or ink) is imbibed into paper have aroused a great deal of interest in this area. These simple experiments could help in understanding the nature of diffusion through random porous media. This is of importance in chromatography. They could also provide as good "table top" systems to study pattern formation \[5,6\]. In the experiments a paper is fixed with the bottom end dipped into a suspension. The suspension is imbibed into the pores of the paper by capillary action. The fluid rises through the pores carrying the suspended particles with it. An interface is formed by the wet front which rises steadily. There is some amount of randomness present in the medium due to the blocking of the pores. This randomness and evaporation of the fluid tries to pin this wetting front. The motion of the wet front is impeded by the evaporation rate, concentration of the suspension, the ratio of the size of the suspended particles to the pore size and viscosity of the fluid. The front stops moving when the fluid has completely evaporated. The roughness of this interface is solely due to the disorder in the paper. If there is no blocking of the pores the interface will be smooth at any rate of evaporation. However the disorder experienced by the fluid is affected by the evaporation. For smaller evaporation the fluid can go around the obstacles and thus not see the blocks than when the evaporation is high. As a result the suspended particles carried by the fluid get deposited at this interface causing a darkening of the boundary. It should be mentioned that this imbibition case differs from the deposition models in two ways (i) there is a time scale set by the evaporation (ii) the randomness in imbibition is quenched and not fluctuating in time.

Till now most of the understanding of the interface growth phenomena in imbibition comes through computer simulations. So far no simulations have been done taking care of evaporation, size of the particles and concentration of the suspension explicitly. In the treatment by Amaral et al \[3\] evaporation was incorporated phenomenologically by a steady increase ($\Delta p$) at each time step in the probability $p$ of blocking the pores. This increase drives the system into a percolation threshold wherein they obtained a connected cluster of
blocked pores. This treatment assumes that the randomness is fluctuating in time.

The correlations of the fluctuations in the height difference between two points $x$ and $x + l$ after the wetting front stops is given by

$$W(l) = \left\langle (h(x) - h(x + l))^2 \right\rangle_x^{1/2} \sim l^\alpha. \quad (1)$$

The time correlation of the height growth at any point $x$ is defined as

$$C(\delta t) = \left\langle (h(t) - h(t + \delta t))^2 \right\rangle_t^{1/2} \sim \delta t^\beta \quad (2)$$

In reference [3] the exponents $\alpha$ and $\beta$ obtained from simulations are .63 and .68 respectively. They claimed these exponents to be universal. In the experiment reported in reference [3] also $\alpha$ was found to be .65. But more experiments are necessary to establish the universality of these exponents (i.e. the independence of the exponents on various factors). And more theoretical work as well as simulations taking account of these factors influencing interface growth are also required. Since in the imbibition experiments described above the noise is due to the disorder present in the paper a quenched randomness is more appropriate.

In this paper we present the results of a preliminary experimental study. We find that the exponent $\alpha$ depends on the evaporation rate. A model for the imbibition in 2D random porous media which takes care of the effect of evaporation explicitly is described. We have carried out a simulation study of this model which gives results in good agreement with our preliminary experiments. We address ourselves to the static and dynamic behaviour of the rough interface.

The experiments are carried out using Whatman No:1 filter paper as the porous medium and ink as the suspension. The evaporation rate was varied by changing the room humidity and temperature. The ink rises through the paper and stops at a particular height. The darkening of the interface is indicative of this stoppage of growth. The interface was then digitized using a CCD camera and a frame grabber with a resolution of 260 pixels per inch. Figure 1 shows the behaviour of $W(l)$ against $l$ for two different values of evaporation rate. The data are averaged over 10 experiments. We find that the exponent $\alpha$ is very different for the two cases. This shows that unlike the results of reference [3] $\alpha$ is not universal.
To understand the dependence of the various parameters affecting the growth of interface we present a model for studying the imbibition of a liquid into 2D random porous media. Before discussing the model used in this simulation let us try to understand the problem from a microscopic point of view. In the experiments described above paper was used as the random medium. At a microscopic level one can regard the paper as a randomly disordered medium with a fixed probability $p$ for the pores to be blocked. So, the interface growth phenomena is nothing but the propagation of fluid particles through this disordered medium. The wetting front of the fluid particles propagates due to the capillary forces. The disorder in the medium and the evaporation tries to pin this growth. Evaporation constantly decreases the number of fluid particles in the wetting front. This makes it more difficult for the fluid to pass the obstacles. The front stops moving at a critical height due to this pinning. It is true that the smaller the evaporation rate, larger will be the critical height.

In our model the porous medium is considered as a square lattice with disorder being incorporated by blocking some cells randomly with a constant probability $p$ (see figure 2). The maximum capacity of each cell is fixed to $N_0$ number of fluid particles. At time $t=0$, at the bottom edge of the lattice a horizontal line of wet cells with $N_0$ particles is created. At $t+1$ the particles are imbibed into all unblocked cells which are nearest neighbors to the wet region. When a cell transfers to its nearest neighbours the number of particles it contains remains the same due to the source below. If a cell has more than one wet nearest neighbour it gets particles from all of them subject to a maximum number $N_0$. At every time step, evaporation was explicitly modelled by the loss of certain number of particles $n$ in the transfer. We also apply the rule that every cell blocked or unblocked below a new wet cell become wet as well to avoid the presence of overhangs and islands. We use periodic boundary condition in $x$ by indentifying the cells at the edge of the lattice.

This model is different from the model used previously, in the sense that here the effect of evaporation is incorporated in an explicit way. Unlike the directed percolation models where the interface is pinned by the connected cluster of blocks here it stops because it runs out of fluid. The model is also different from the Eden growth in the sense that all
the sites in the boundary moves at the same time *i.e.* here the growth process by itself does not cause interface roughness. The concentration of the suspension and the pore size are incorporated in the model in the following way. For a given concentration the number of fluid particles to the number of suspended particles is fixed. Thus for a fixed pore size the change in concentration of the suspension or *Vice Versa* is same as changing the maximum number of fluid particles $N_0$ which a cell can hold. Also it is possible to include the effect of other factors like gravity, which can induce a bias to the propagation of the fluid particles through the disordered medium. The model can be used to study for example *thin layer chromatography* wherein a mixture of different chemical compounds are made to diffuse through a porous medium resulting in their separation.

The simulations are done on a lattice of length 6000 lattice units. The results are averaged over 500 realisations. The simulations show the existence of a crossover length $l_x$ such that the height - height correlation function $W \sim l^\alpha$ for $l \ll l_x$. Whereas for $l \gg l_x$ $W$ saturates to a constant value $W_{sat}$ as shown in figure 3. This saturation width depends upon the number $n$ of the fluid particles evaporated as

$$W_{sat} \sim (n)^{-\gamma}$$

So, one can define a scaling form given by

$$W(l, n) \sim l^\alpha f(ln(l^{\frac{\alpha}{\gamma}}))$$

where $f(x) \rightarrow$ a constant as $x \rightarrow 0$ and $f(x) \rightarrow x^{-\alpha}$ as $x \rightarrow \infty$.

We find that there are two critical values of evaporation $n_1$ and $n_2$ for a given value of $N_0$. These critical values change in such a way that the ratios $n_1/N_0$ and $n_2/N_0$ remain constant. For low values of evaporation the mean height increases without stopping (regime I). Above the critical evaporation $n_1$ the mean height stops after a finite time. For $n > n_1$ (regime II) the $log(W) - log(l)$ plots fall on to a single curve through the scaling form given in equation (3) (see figure 4). We find in this region $\alpha = 0.5$ and $\gamma = 3.0$. Note that this is very different from the exponent obtained in [3]. At the second critical evaporation rate
this scaling breaks down. For \( n > n_2 \) (regime III) the exponent \( \alpha \) decreases continuously with \( n \) as can be seen from figure 3. The critical values \( n_1/N_0 \) and \( n_2/N_0 \) decreases with \( p \). For \( p = .45 \) we get \( n_1/N_0 = .124 \) and \( n_2/N_0 = .134 \).

We can get more insight by looking at how the height - height correlation exponent \( \alpha \) changes with time. This is depicted in figure 5. At short times \( \alpha \) shows a rapid increase. This region is independent of evaporation. However the long time behaviour shows \( \alpha \) saturating to a value \( \alpha_{sat} \). This establishes the fact that the roughness of the interface is controlled by evaporation and shows that the reason for dependence of \( \alpha \) on \( n \) is not due to the lack of time for the interface to saturate. We see from figure 5 that for \( n < n_1 \) \( \alpha \) is greater than .5. However in this low evaporation regime the front does not get pinned but moves at a constant velocity. This dependence of \( \alpha \) on evaporation is consistent with the experimental results described above. However more experiments are necessary to confirm the smooth dependence of \( \alpha \) on evaporation predicted by the model. To establish the universal dependence of \( \alpha \) on evaporation more experiments using different kinds of papers and suspensions are being pursued.

The dependence of the dynamical exponent \( \beta \) on evaporation is the same as that of \( \alpha \). In figure 6 we show the behaviour of \( C(\delta t) \) for various values of \( n \) for a fixed values of \( p \). We see that in regime I all the curves have a slope greater than .5. This slope changes continuously to .5 in regime II. In regime III the slope decreases with \( n \).

In this model we can apply an external bias which could be present due to gravity or anisotropy in the medium. This bias was incorporated in the model by introducing a difference in the number of particles transfered to the vertical and horizontal neighbours of a given cell. The different regimes described above were also observed in this case for a fixed value of \( n \) and \( p \) as the bias was varied.

The correlation functions \( W(l) \) and \( C(\delta t) \) can be directly mapped to the root mean square displacement in a random walk, with \( l \) taking the place of time in \( W(l) \). The critical behaviour at \( n_2 \) described above is similar to the dynamical transition in biased random walk in a random medium [1], wherein the exponent changes continuously with the applied
bias is above a critical value. Since the behaviour found with the external bias and that due to evaporation are the same one can assume that the evaporation effectively introduces a bias. The detailed nature of this bias is however not clear.

In the model the particles have more than one path to reach a particular cell. The effective number of paths available to reach a cell decreases since increased evaporation suppresses the longer paths. For \( n < n_1 \) the number of fluid particles that a cell loses through evaporation is more than compensated by the inflow because of the many paths available. Hence this regime becomes super diffusive. The value of exponents being greater than 0.5 support this argument. As the number of paths become less the particles get stuck at obstacles for a longer time inducing a transition into a normal diffusive regime with exponents 0.5 \( \frac{\hat{\beta}}{3} \). On further increase of evaporation the system becomes subdiffusive. In the case of biased random walk this region is known to have exponents which change continously with bias \( \hat{\beta} \).

The effect of evaporation on the roughness exponent is similar to the dependence of exponents on amount of nonconservation in cellular-automaton. These systems are known to exhibit self-organisation \( \frac{\hat{\beta}}{3} \). We establish the self-organisation of the interface in imbibition by looking at the dependence of \( W_{\text{sat}} \) on the system size \( L \). This is shown in fig.7. We find \( W_{\text{sat}} \sim L^{0.12} \). A detailed analysis of the effect of parameters on the self-organisation will be published elsewhere.

In conclusion we have shown the breakdown of universality in imbibition from direct experiments. A model for imbibition which includes the effect of evaporation in a very natural way is presented. The model shows that the static height - height correlation exponent \( \alpha \) depends on the evaporation which is consistent with our experiments. The interface is shown to exhibit self-organised criticality.
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FIGURES

FIG. 1. The experimental values of height - height correlation function W(l) plotted against
the distance of separation l after the interface stopped growing. Points marked with * fall onto a
curve with exponent $0.45 \pm 0.004$. For higher evaporation the points (marked o) correspond to an
exponent $0.67 \pm 0.004$.

FIG. 2. Example of the multiple connectivity of the model for a 6X6 lattice. The blocked cells
are shown black. At time 0 all the cells corresponding to $i=1$ and $j=1,6$ are filled with $N_0$ particles.
At each time step the particles are transfered to nearest neighbours. Note that the cell (4,4) gets
particles from both (4,3) and (4,5). Also when a cell (i,s,j) is wet all the other cells (i,j) with $i < i_s$
are wet as well.

FIG. 3. The simulated height - height correlation function W(l) plotted against the distance
of separation l after the columns stopped growing. The length is measured in units of lattice
parameter. The parameters are $p = 0.45$, $n = 18.1$ to 22.3 in equal intervals of 0.6. The saturation
value of $W(l)$ decreases monotonically with $n$. The simulations are done with a lattice of length
6000 applying periodic boundary condition.

FIG. 4. The $W(l)$ plotted against $l$ in the scaling form defined in the text for the parameters
given in figure 4.

FIG. 5. The growth of height - height correlation exponent $\alpha$ as a function of time for $p = 0.2$.
The three regimes are marked I, II and III. The lines are for $n = 43, 44.2, 45$ respectively. The
behaviour of $\alpha_{sat}$ with evaporation n in this three regimes is shown in the inset.

FIG. 6. The plot of dynamic correlation function $C(\delta t)$ for various values of $\delta t$. The dotted
line corresponds to a slope of 0.5. Other parameters are same as figure 4.

FIG. 7. The simulated height - height correlation function W(l) plotted against the distance
of separation l after the columns stopped growing for system sizes $L=6000,4000,2000,500$. $p=0.45$,
$n/N_0 = 0.126$. 

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