

Conserved dynamics and interface roughening in spontaneous imbibition: A critical overview

M. Dubé^{1,2,a}, M. Rost^{1,2}, and M. Alava²

¹ Helsinki Institute of Physics, P.O. Box 9 (Siltavuorenpenger 20 C), 00014 University of Helsinki, Helsinki, Finland

² Laboratory of Physics, Helsinki University of Technology, P.O. Box 1100, 02150 HUT, Espoo, Finland

Received 17 February 1999 and Received in final form 24 November 1999

Abstract. Imbibition phenomena have been widely used experimentally and theoretically to study the kinetic roughening of interfaces. We critically discuss the existing experiments and some associated theoretical approaches on the scaling properties of the imbibition front, with particular attention to the conservation law associated to the fluid, to problems arising from the actual structure of the embedding medium, and to external influences such as evaporation and gravity. Our main conclusion is that the scaling of moving interfaces includes many crossover phenomena, with competition between the average capillary pressure gradient and its fluctuations setting the maximal lengthscale for roughening. We discuss the physics of both pinned and moving interfaces and the ability of the existing models to account for their properties.

PACS. 47.55.Mh Flows through porous media – 05.40.-a Fluctuation phenomena, random processes, and Brownian motion – 68.35.Ct Interface structure and roughness

1 Introduction

A considerable amount of effort has been spent in the field of kinetic roughening over the last two decades. Apart from technological interests in crystal growth, a good part of the fascination for this field comes from the possibility of describing many different types of interfaces by a few distinct universality classes, in terms of scaling exponents and scaling functions [1, 2].

However, if the theoretical and numerical aspects of the field are extremely rich and varied, the experimental backing of these ideas is quite lacking. This is for instance the case in kinetic roughening in crystal growth due to the great variety of atomistic processes. Much of the experimental attention has rather concentrated on simpler one-dimensional “toy-systems”, in particular driven interfaces in random media. A definite advantage of these systems is that the interface configuration is directly observable in the experiment (while, it must often be deduced, from some probe-surface interaction, in crystal growth). Even these simple systems are challenging. They are most commonly grouped in universality classes described by the Kardar-Parisi-Zhang [3] (KPZ) or Edwards-Wilkinson [4] (EW) equations, with either quenched or thermal noise, depending on the driving regime. In the case of quenched noise the exact low-dimensional scaling behaviour is however still somewhat contradictory [5–9].

Examples of experimental studies in these systems are slow combustion fronts [10] (for which KPZ scaling was

recently demonstrated), shock fronts, fluid-gas interfaces in Hele-Shaw cells [11–15], or in paper [16–24] and fracture surfaces [25] such as one-dimensional fracture lines [26, 27], to name a few. In this paper, we concentrate on a particular system, namely the spontaneous imbibition of a porous medium by a liquid [16–24]. Our goal is to critically review the experiments and theories that exist in the literature and to indicate a direction for further investigation and comprehensive understanding of imbibition front roughening. The key questions are (i) whether imbibition should present any kind of scaling behaviour at all, and (ii), if so, under which conditions can one of the – possibly several – scaling regimes become observable.

Our motivations in writing this paper are twofold. First, the fluid dynamics aspect of imbibition is itself very complex, and should not be ignored in any discussion of the statistical fluctuations of the interface. We feel that this has not been properly done so far. Secondly, we present in a companion paper the results of a line of investigation of spontaneous imbibition based on a phase field formalism [28]; the present paper is intended to lay down the aspects which we believe essentials to any models of imbibition.

We start in Section 2 by reviewing the macroscopic properties of imbibition and show that even the simple propagation of an imbibition front may have several dynamical regimes, depending on the design of the experiment. We also show that the macroscopic progression of the interface will have a very strong influence on the roughening process. Section 3 outlines the models that have been proposed for imbibition experiments as well

^a e-mail: dube@pcu.helsinki.fi

as their predictions. In Section 4, we discuss the existing experimental data, and point along the way to several features missed by these experiments. Finally we conclude with a discussion and proposals concerning future experiments.

2 Macroscopic features of imbibition

Although extremely familiar to researchers working in the field of flow in porous media, the details of imbibition would seem to be relatively unknown to the statistical physics community. It is generally defined in reference to two-fluid flow in porous media, and corresponds to the displacement of the lesser wetting fluid by the more wetting one [29,30]. Notice that this definition is irrespective of whether the flow of the fluids is spontaneous or induced (*e.g.*, by a pump). In this work, we will restrict ourselves to the case of spontaneous imbibition. The flow of the fluids is thus driven solely by capillary forces, with gravity and/or evaporation being the only external influences on the fluids' motion.

2.1 Capillary rise

The simplest example of spontaneous imbibition is capillary rise: a part of a capillary tube, of radius R , is immersed into a reservoir, exposed to an ambient atmospheric pressure P_0 . We assume that the fluid wets the capillary so that a meniscus, described by the surface $z - h(x, y, t) = 0$ is formed. At equilibrium, this surface is characterised by a contact angle θ , obtained from Young's law [31]

$$\gamma_{lg} \cos \theta = \gamma_{sg} - \gamma_{sl}, \quad (1)$$

where γ_{ij} is the surface tension between the phases i and j , and s , l and g stand for solid, liquid and gas respectively. If the meniscus is in motion, the contact angle differs from its equilibrium value [32], but we neglect this effect here. The important point is that the curvature causes a pressure difference $\Delta P \equiv P_c = P_0 - P(z = h^-) = 2\gamma_{lg} \cos \theta / R$ where γ is the surface tension of the liquid-gas interface and $R/\cos \theta$ is the curvature of the meniscus. We refer to P_c as the capillary pressure.

The motion of the fluid, of density ρ and viscosity η , can be treated within the assumption of Poiseuille flow [33], *i.e.* the full Navier-Stokes equation is replaced by the simpler Stokes equation for an incompressible fluid under the influence of the gravitational force $\rho \mathbf{g}$. As usual, the pressure field is found from Laplace's equation $\nabla^2 P = 0$, but with boundary condition $P(z = 0) = P_0$ and $P(z = h) = P_0 - P_c$. (We can neglect corrections to the pressure field close to the meniscus, if its height is a lot larger than the radius of the tube). This yields $P(x, y, z) = P(z) = P_0 - P_c z/h(t)$, and it is straightforward to obtain the progression of the interface,

$$\frac{dh}{dt} = \frac{\kappa}{\eta} \rho g \left(\frac{h_{\text{eq}}}{h(t)} - 1 \right) \quad (2)$$

a classical result derived by Washburn [34] and Rideal [35]. Washburn's and Rideal's equation includes the permeability of the tube $\kappa = R^2/8$ and the equilibrium height of the meniscus $h_{\text{eq}} = P_c/\rho g$. Studies of capillary rise including the inertial term of the Navier-Stokes equation [36] show that equation (2) is essentially correct, with the exception of very short times. Notice also that this equation of motion also neglects completely the problem of the actual motion of the contact line between the gas-solid and liquid phases [32].

The (transcendental) solution of equation (2) has the following asymptotic properties: Defining $h(t_0)$ as the initial height of the column and $\tau_{\text{eq}} \equiv h_{\text{eq}}\eta/\kappa(\rho g)^2$ as an equilibration time, for low heights $h \ll h_{\text{eq}}$ (and also in the absence of gravity where $h_{\text{eq}} = \infty$) the rise is of the form

$$h^2(t) - h^2(t_0) = \frac{\kappa P_c}{\eta} (t - t_0). \quad (3)$$

The equilibrium height is approached by h exponentially

$$h(t) \sim h_{\text{eq}}(1 - e^{-t/\tau_{\text{eq}}}). \quad (4)$$

This was examined experimentally by Washburn [34] and Rideal [35] as well as numerous others, who found very good agreement between theory and experiment. Capillary rise in a liquid-liquid system was also examined by Mumley *et al.* [37]. They confirmed Washburn's result in the case of perfect wetting, but reported discrepancies for systems with non-zero contact angle. In this case, the rise was slower than $t^{1/2}$, a behaviour attributed to the motion of the contact line itself [32].

2.2 Capillary rise in porous media

With the basic capillary rise phenomenon understood, we can now turn to spontaneous imbibition in porous media. The flow of liquids in porous media is generally described in terms of Darcy's equation [29,30]

$$\langle \mathbf{Q} \rangle = -\rho A \frac{\kappa}{\eta} (\nabla P - \rho \mathbf{g}), \quad (5)$$

where $\langle \mathbf{Q} \rangle$ is the average mass of fluid transported per unit time through the cross-section A of the porous medium, and κ and η are the average permeability and viscosity respectively. Darcy's equation arises from an averaging procedure of the porous medium and ignores all details on length scales smaller than the pores. If we assume the porous medium to be homogeneous, such that the permeability is a constant κ_0 independent of the fluid concentration, we can still solve Laplace's equation for the pressure and obtain a coarse-grained pressure gradient, $\nabla P = \hat{\mathbf{z}} P_c/h(t)$, with P_c an effective capillary pressure, and h the average height of the fluid column. This is of course valid only when the notion of an interface is itself well-defined, *i.e.* it should not be too "fuzzy" up to macroscopic scales. Under these assumptions, and with the identification $\langle Q \rangle = \rho A dh/dt$, Darcy's equation leads

directly to the Washburn-Rideal result, equation (2) with a dynamical rise given by equations (3) and (4).

This form of Darcy's equation has been used to study many experiments of fluid propagation in porous media, including some fibrous materials (see below). The agreement is not always perfect – we shall come back to this in Section 2.5. At this point, a more complete system of equations [30,38] would only obscure the physics.

2.3 Evaporation

Considering the typical experimental setups for imbibition front propagation [20], evaporation effects should also be included to Washburn's equation. As far as we are aware, no detailed studies of fluid motion through thin porous media including the effects of evaporation have been performed. A natural assumption is to introduce an evaporation rate proportional to the area of the fluid exposed to air, *i.e.* the net loss of fluid mass per unit time due to evaporation is $Q_e = -2\mathcal{E}\rho Lh(t)$ where L is the lateral width, and \mathcal{E} is a phenomenological evaporation rate per unit area. Thus equation (2) is modified to

$$\frac{dh}{dt} = \frac{\kappa}{\eta} \left(\frac{p_c}{h} - \rho g \right) - \epsilon h \quad (6)$$

where $\epsilon = 2\mathcal{E}\rho$ is the evaporation rate. This form obviously neglects changes in the concentration profile towards the surface, as well as the evaporation at the interface itself, which is both justified for thin media such as a sheet of paper.

An immediate consequence of equation (6) is an equilibrium height depending on the evaporation rate

$$h_{\text{eq}} = \frac{h_e^2}{2h_g} \left(\left(1 + \frac{4h_g^2}{h_e^2} \right)^{1/2} - 1 \right) \quad (7)$$

where $h_g = P_c/\rho g$ and $h_e = (P_c\kappa/\epsilon\eta)^{1/2}$ are respectively the equilibrium heights in the cases where only gravity or evaporation have a significant influence, with a crossover defined by $2h_g \sim h_e$. When gravity can be neglected, the height $h(t)$ behaves as

$$h^2(t) = h^2(t_0) e^{-2\epsilon(t-t_0)} + h_e^2 \left(1 - e^{-2\epsilon(t-t_0)} \right). \quad (8)$$

Well below h_e the rise still follows $h(t) \sim t^{1/2}$, and it again approaches exponentially the equilibrium height.

2.4 Addition of a solution

Many experiments on the roughness of the interface in an imbibition context have studied the behaviour of a dye, added to the pure fluid [16,20,23]. In the literature on porous media this is referred to as the hydrodynamic dispersion phenomenon [30]. At the simplest level, it can be

treated by the introduction of a dye concentration field $K(\mathbf{x}, t)$ advected by the macroscopic velocity field \mathbf{v} ,

$$\frac{\partial K}{\partial t} + \mathbf{v} \cdot \nabla K = D \nabla^2 K, \quad (9)$$

where D is a diffusion constant (in some cases, it may be necessary to introduce a diffusion tensor D_{ij} [30]).

Two remarks on equation (9) are in order. First, in everyday life, it is easily seen that the fluid front is always faster than the dye front. This clogging phenomenon, which may be due to a smaller permeability for the dye, may easily be incorporated phenomenologically by introducing a constant $\lambda < 1$ in the front of the convective term, *i.e.*

$$\mathbf{v} \cdot \nabla K \rightarrow \lambda \mathbf{v} \cdot \nabla K. \quad (10)$$

The second point concerns the stopped interface. If the pinning of the interface, at a distance h_e from the reservoir, is due to evaporation, there will nevertheless be a fluid motion, with approximate velocity $v_e \sim \kappa P_c/\eta h_e$, in order to compensate the losses. This implies that more and more dye particles will be brought to the fluid interface, thus creating a band of high dye concentration, analogous to the coffee rings examined recently [39]. The increase of the width of this band with time can be used as an alternative way of measuring the relevant macroscopic parameters.

2.5 Validity of macroscopic description

We now discuss the validity of the macroscopic results for the specific case of spontaneous imbibition in paper. Ordinary paper is made out of wood fibers and, in many cases, chemical additives and filler materials like talc and clay, arranged in a disordered structure. Not only is there a wide distribution of pore sizes, with a high effective tortuosity but the surface of the network is extremely uneven. This gives rise to all the standard problems in defining a static/dynamical contact angle for inhomogeneous substrates.

Darcy's equation, equation (5), is relatively well-established (to the point that it is even often referred to as a law) for general porous media. However, paper, as well as other fibrous materials present the peculiarity that the fiber structure may be modified by the contact with the liquid, a phenomenon known as swelling [40]. Cellulose fibers show a great affinity to water and can absorb large quantities in millisecond timescales, giving rise to concomitant changes in fiber volume and pore structure (this is however not the case for many organic fluids and oils). Thus one's intuitive picture of fibers as capillary tubes is false: the pore structure is highly non-trivial and in some cases time-dependent. There are two different effects that play a role: the volume to be filled with liquid increases, and the flow resistance of the pore network changes.

There can also be an exchange of liquid between the inside of a fiber and the "surface" pores, which complicates enormously the fluid flow, since there are no well

defined “structures” (either the pores, or the fibers) responsible for the capillary forces [41,42]. This is of course a very serious impediment to any kind of attempt at a “microscopic description” of the structure, say in terms of a percolation network [43].

A more serious problem with water is the fact that it is not actually known whether a pressure balance argument is always valid. On short time scales (smaller than a few seconds), it has been proposed that front penetration would proceed in pores first with the establishment of a prewetting layer through diffusion [45], in which case the front position could advance as t^k with k varying up to unity. Experiments have demonstrated that clear, Washburn-like behaviour can be obtained in conditions that amount to basically no interaction (swelling, prewetting) between the material and the penetrating liquid [44]. Note that minute applications of chemicals, *e.g.* during paper manufacture, may induce drastic changes in the effective viscosity or surface tension of the invading liquid.

There are however some advantages to paper. The very high permeability of the fibers will reduce the formation of overhangs in the interface. Darcy’s equation cannot treat trapped air bubbles in bulk porous media [38], which in paper only play a minor role: paper is thin and the pores should be connected well to the surface. Moreover spontaneous imbibition is slow and allows more time for the removal of overhangs and trapped bubbles.

In conclusion, one would believe that a well-defined fluid-air interface should exist, and that Darcy’s equation should be valid on length scales larger than the interface width, provided that interactions between the liquid and the fibers are minimal. In that sense, a full hydrodynamical treatment of the problem may not be necessary (as noted in [28]). In many cases, it may also be necessary to introduce a concentration dependent permeability, *i.e.* $\kappa = \kappa(\rho)$ [44]. In any case a Washburn approach may be a first step. In that respect, recent experimental work with paper and deionised water, showing front propagation consistent with Washburn’s equation is quite encouraging [46].

2.6 Statistical fluctuations of the interface

So far, we have considered a flat interface in a completely homogeneous medium. In general, the disorder in the paper causes fluctuations which will accumulate to roughen the advancing front. In the standard picture of kinetic roughening [1,2] the fluctuations of the interface are correlated up to a distance $\xi_{\parallel}(t)$, a lateral correlation length, increasing in time as $\xi_{\parallel} \sim t^{1/z}$ and described by the dynamical exponent z . The vertical extent of the interface fluctuations, its width W , is related to ξ_{\parallel} through the roughness exponent $W \sim \xi_{\parallel}^{\chi}$. Thus the width increases with time as $W \sim t^{\beta}$ with $\beta = \chi/z$, until the interface fluctuations have saturated, *i.e.*, until $\xi_{\parallel}(t) = L$, which defines the crossover time $t_{\times} \sim L^z$.

The initial increase of the width and eventual saturation are comprised in a (Family-Vicsek) scaling form

$$W(L, t) = L^{\chi} f(t/L^z) \quad (11)$$

with the scaling function $f(u) \sim u^{\beta}$ for $u \ll 1$ and $f(u) \sim \text{const}$ for $u \gg 1$.

The spatial height difference correlation function (of the q th moment)

$$G_q(\mathbf{r}, t) = \langle |h(\mathbf{r} + \mathbf{r}', t) - h(\mathbf{r}', t)|^q \rangle^{1/q} \quad (12)$$

generally scales similarly to the total width, *i.e.* $G_q \sim r^{\chi}$ for small r , and approaching a constant as $r \rightarrow \infty$. However, care has to be taken: In the case of anomalous scaling the height differences for fixed $|\mathbf{r}|$ do not saturate with time, and G_q shows the local roughness exponent χ_{loc} [2,48]. The height difference distribution for fixed $|\mathbf{r}|$ may also have a long tail, causing intermittent or “turbulent” jumps in the height configuration in which case each moment has a different exponent χ_q , *i.e.*, the interface exhibits multiscaling [47]. Also a temporal height difference correlation function

$$C_q(t) = \langle |h(\mathbf{r}, t + t') - h(\mathbf{r}, t') - (\bar{h}(t + t') - \bar{h}(t'))|^q \rangle^{1/q} \quad (13)$$

can be considered (*e.g.* in [21]), which is of particular use in the case the system exhibits time-translational invariance.

To our knowledge none of the works studying interface fluctuations in imbibition have checked for anomalous (as determined by the higher moments of the correlation functions) types of scaling behaviour [16–24], although this is in principle difficult, since an exact diagnosis for higher moments $q > 2$ is severely hampered by large statistical fluctuations. These concepts are nevertheless important when the randomness of the medium is described with random-field disorder [28].

Another peculiarity of imbibition is that, apart from the standard kinetic lateral correlation length $\xi_{\parallel}(t)$, arising from the accumulating history of fluctuations, another lateral length scale is to be seen: local conservation of the fluid determines a lateral length ξ_{\times} related to the average motion of the interface. Intuitively, it is clear that such a length scale must exist, since fluctuations ahead and behind the average position of the imbibition front have respectively a slower and faster instantaneous velocity. To make a quantitative argument, we introduce an effective surface tension γ^* , representing the energy cost of a curved air-liquid interface on a macroscopic scale [54]. Any curvature at the interface – the typical size of fluctuations being W vertically and ξ_{\times} laterally – modifies the pressure by an amount $\Delta P = \gamma^* W / \xi_{\times}^2$ (the Laplace pressure effect). This should be compared with the difference in the pressure field across the same vertical distance W due to the pressure gradient derived in Section 2.1, namely $\Delta P = P_c W / H$, from which we obtain

$$\xi_{\times}^2 \sim \gamma^* H / P_c, \quad (14)$$

relating the parallel length scale ξ_{\times} to the height of the interface. Beyond this length scale we do not expect any

correlated roughness of the interface, since those fluctuations would be suppressed by the overall gradient in the pressure field P_c/H .

Let us recall once again that the interface continuously slows down [58], without gravity nor evaporation $H \sim t^{1/2}$. Thus the slow increase of $\xi_x \sim H^{1/2} \sim t^{1/4}$ with time leads to an increase of the width $W \sim t^\beta$ with $\beta = \chi/4$. However, we stress that this increase is conceptually different from the increase of the lateral correlation length with a dynamic exponent z in standard models of kinetic roughening.

3 Models of imbibition

In parallel with experimental work, many theoretical models of imbibition have been developed. These models fall either in discrete (cellular automata) or continuous classes. Most of the theoretical discussion on imbibition was done in terms of the DPD model, which was shown to belong to the same universality class as the quenched KPZ equation. Other types of models or continuum equations have also been proposed.

3.1 Cellular automata models

The earliest theoretical investigations of imbibition phenomena were based on the directed percolation depinning (DPD) model [16,17,20]. It is a cellular automaton, with space discretized in cells which are either blocked or wettable, with the blocked cells being a fraction p . The particularity of the model is that overhangs in the interface are removed as soon as they occur, which thereby introduces anisotropy in the interface. Growth takes place by invasion of available cells until the interface comes across a percolating directed path of blocked cells. Clearly, the statistical properties of the interface are related to the properties of the percolating path. Close to the percolation threshold $p_c \simeq 0.47$, the directed percolating path is characterized by the parallel and perpendicular length scales $\xi_{\parallel} \sim |p-p_c|^{-\nu_{\parallel}}$ and $\xi_{\perp} \sim |p-p_c|^{-\nu_{\perp}}$, with values $\nu_{\parallel} \simeq 1.7$ and $\nu_{\perp} \simeq 1.09$. so the width $W(L) \sim \xi_{\perp} \sim L^{\nu_{\perp}/\nu_{\parallel}}$, yielding $\chi \simeq 0.63$.

To treat evaporation, the DPD model was modified by the introduction of height dependent fraction of blocked cells, *i.e.* $p = p(h) \sim h/h_0$ with an equilibrium height $h_0 \sim (\nabla p)^{-1}$ defined by $p(h_0) = p_c$ [20]. In this model, the interface is described by the standard DPD roughness exponent up to a length $l_x \sim (\nabla p)^{-\gamma/\chi_{\text{DPD}}}$ after which it saturates to a constant value $w_{\text{sat}} \sim (\nabla p)^{-\gamma}$. The identification $w_{\text{sat}} \sim \xi_{\perp}$ then yields $\gamma = \nu_{\perp}/(1 + \nu_{\perp}) \sim 0.52$, a value comparable to the experimental results.

As already noted [21], there is a physical justification to the main feature of the DPD model, namely the erosion of overhangs. It is plausible that propagation of the fluid in a direction parallel to the reservoir, if it has a chance to occur, will be favoured over the motion of the fluid away from it, since this does not result in any change in the

average capillary pressure. In that sense, any overhangs will eventually be removed, although the question of the different time scales involved in the dynamics is certainly extremely complex [28].

The DPD model in its simplest form then probably describes well the statistical properties of the pinned interface. It is however difficult to believe that this model can describe the whole dynamical motion of the interface, since it does not account for liquid conservation. It already fails by predicting a constant average interface velocity. As we saw in Section 2, one of the principal features of imbibition is a constant slowing down of the front, a result intimately related to fluid conservation. It can be argued that the DPD refers specifically to the motion of the dye, but the rise of the dye front is necessarily bounded by the liquid-air interface, and the conservation law governing the motion of the fluid must be reflected on the motion of the dye particles. From that point of view, simply replacing $\mathbf{v} \sim t^{-1/2}$ in equation (9) certainly would be incorrect.

It is interesting to show how the global behaviour of the modified DPD model may be related to the macroscopic Washburn behaviour, (Eq. (6)). The density of open cells $1 - p(h)$ can be thought of as an effective force acting on the interface, similar to the gradient in pressure on fluid motion. Since the Washburn equation is by essence a dissipative equation, we can associate, away from pinning

$$p(h) \rightarrow \frac{\kappa}{\eta} \left(\frac{p_c}{h} - \rho g \right) - \epsilon h. \quad (15)$$

The assumption of constant gradient thus effectively corresponds to an evaporation rate. It is also possible, at the macroscopic level, to include gravity, and to mimic the slowing down of the interface. Note however that we predict a pinning height $h_e \sim \epsilon^{-1/2}$ while the assumption of Amaral *et al.* implies $h_e \sim (\nabla p)^{-1}$, since they neglect the capillary driving term.

Another model of evaporation was introduced by Kumar and Jana [22]. The model is essentially similar to the DPD model, with the modification that each cell is not necessarily “full” or “empty”, but may contain many “subunits”. Evaporation is modeled by a loss of n subunits in the transfer between cells. It is found that below a critical n_{c1} , the interface propagates indefinitely. Between this value and a second critical loss rate n_{c2} , they observe an interface behaviour similar to the one observed by Amaral *et al.* [20], but with value $\chi = 0.5$ and $\gamma = 3.0$. For $n > n_{c2}$, this scaling regime breaks down and the roughness exponent becomes n -dependent. These results were backed experimentally, but not in any consistent way.

It is rather difficult to believe that indefinite front propagation under evaporation (obtained for $n < n_{c1}$) is physically realistic, nor seems the way of including evaporation convincing. In this model, the concentration of fluid molecules decreases continuously from a maximum N_0 to a value of 0 at the interface, which seems physically unrealistic.

We terminate by presenting invasion models based on the random field Ising model [49], which predict a depinning transition, as well as a change of morphology of

the interface at some length scale related to the capillary pressure. It seems however unsuited for spontaneous imbibition, because any advance of the front requires the increase of an externally applied pressure.

A variation of this idea, introduced by Sneppen [50], allows invasion always at sites of lowest resistance. An interesting aspect of this model is temporal multiscaling [51], due to avalanche motion [52,53]. Avalanches are also present in the process of spontaneous imbibition, although the conservation law imposes a natural cutoff on their size and distribution [15,28]. It is nevertheless interesting that a similar lack of temporal scaling is also seen in the phase field model of imbibition, thus surviving the introduction of a conservation law.

3.2 Continuum description

The DPD model (in its original version without “evaporation”) belongs to the same universality class as the quenched KPZ equation (see *e.g.* Chapter 10 in [1] and references therein, as well as [9])

$$\frac{\partial h(x,t)}{\partial t} = \nu \nabla^2 h(x,t) - \frac{\lambda}{2} (\nabla h(x,t))^2 + \eta(h(x,t), x). \quad (16)$$

A related equation with multiplicative noise has been introduced by Csa ok *et al.* [9] in order to model the random porosity of the medium. Since these equations are local at the interface, they do not contain a conservation law for the fluid.

With these considerations in mind, a continuum equation was introduced by Zik *et al.* [24] following Krug and Meakin [54]. In Fourier space, it has the form

$$\frac{dh_k}{dt} = -\frac{\kappa p_c}{\eta} |k| \frac{h_k}{h_0(t)} + \eta_k(t) \quad (17)$$

where k is the wave-vector, $h_0(t)$ is the mean height of the interface at time t (*cf.* Eq. (3)), and η_k is the noise term, assumed to be annealed, with correlations $\langle \eta_k(t) \eta_{k'}(t') \rangle \sim \delta_{k+k'} \delta(t-t')$. This equation is essentially similar to the one derived by Krug and Meakin for the roughness of stable Laplacian fronts with noise and corresponds to the leading term of a Saffmann-Taylor analysis of the problem. Note that $|k|$ is nonlocal in space.

The other difficulty with respect to spontaneous imbibition, the quenched nature of the disorder, remains. As long as the interface sweeps fast enough through the medium, the disorder acts as annealed, time dependent noise [6,7], although the question of the effective noise correlator is far from trivial [28] — equation (17), as it stands, gives the unphysical results of an interface that never saturates. In any cases, at late times under slow average interface propagation in a continuum model one is confronted with both the difficulties of a nonlocal interface equation with quenched disorder.

4 Experimental efforts in imbibition

Most of the experiments concerned with front propagation in random media seem to produce a self-affine interface, but the numerical value of the exponents often bears little resemblance to the standard universality classes and/or associated models. Imbibition of porous media would seem like an obvious first experimental choice, since the associated time and spatial scales are easily accessible in the laboratory. Many such imbibition experiments were performed, but it turns out that there is very little in common between the different experiments. This is partly due to the fact that different experiments had different goals, but it is also a reflection of the great complexity of the processes involved in imbibition.

4.1 Experiments on pinned interfaces

The earliest statistical physics imbibition experiments were done by Buldyrev *et al.* [16,17,19] and Family *et al.* [18]. The first experiment was performed with a dye solution in a vertical capillary rise setup. The rising front moved from the reservoir and the roughness of the interface developed during the process. Eventually, the dye front stopped, due to gravity and/or evaporation (no dynamical measurements, either micro- or macroscopic, were done in this experiment), and the roughness of the pinned interface was measured. The main experimental conclusion of the paper was a roughness exponent with value $\chi = 0.63$, consistent with the DPD model (see above, Sect. 3). However, the length scale over which the scaling behaviour was observed is extremely small. For a sheet of paper of total lateral extent $L = 40$ cm, $C(l) \sim l^\chi$ only for length scales l smaller than $l_{\max} \sim 1$ cm, after which it levels off to a constant or to a logarithmic function of l [55]. Since this scaling region is only a few times larger than the length scale of the fibers themselves, it is not at all clear whether the exponent χ really is a universal value, or simply results from the microscopic fiber structure. As a comparison, the scaling region for the paper burning experiment [10] was from 1.5 cm to about 10 cm, for a total length 30 cm [56]. A similar experiment was done in a three-dimensional sponge-like material [19]. Again, the stopped interface was observed, yielding a roughness exponent $\chi^{(2d)} \sim 0.5$, a result consistent with the higher-dimensional version of the DPD model. No other experimental details were however given.

In a further set of experiments, Amaral *et al.* [20] studied the role of evaporation in more detail. They again considered the stopped interface, but changed the height where pinning occurred through the evaporation rate (presumably by modifying the humidity during the experiment, although this is not specified). Their main result is that the width of the pinned interface is related to the pinning height H_p (and thus to the evaporation strength) through a new exponent γ , namely $w_{\text{sat}} \sim H_p^\gamma$. The experiments gave a value $\gamma = 0.49$, which – as reported in Section 3 – can also be obtained from a modified version of the DPD model [20]. (We will come back to this in

Sect. 4.3). On the other hand, Kumar and Juma [22] also performed an experiment in presence of various evaporation rates and claimed that the roughness exponent was not universal but strongly dependent on the evaporation rate, *i.e.* $\chi = \chi(\epsilon)$, although no systematic experimental investigation of this breakdown of universality was done.

4.2 Experiments on moving interfaces

The experiment of Family *et al.* was performed in an horizontal capillary setup with water only, and the position of the air-water interface was recorded both temporally and spatially [18]. The main experimental results were an average interface progression $\bar{h} \sim t^\delta$, with $\delta = 0.7$, and a self-affine interface described by a Family-Vicsek scaling relation, and characterized by the exponents $\beta = 0.38$ and $\chi = 0.76$. The first result $h(t) \sim t^{0.7}$ is certainly intriguing. However, it should be noticed that a reservoir was placed underneath the piece of paper, in order to prevent evaporation. It is of course possible that this caused condensation instead, thus increasing the velocity of the front. It is also in line with the results of non-Washburn water penetration in paper [45]. Also here the spatial scaling regime was rather small, for distances below $l_{\max} \sim 2$ cm for a 40 cm wide sheet of paper. It should also be noticed that these experiments were made with chinese paper, having a thickness of only a few fiber layers, which may also influence the results.

The temporal scaling of the interface was studied in details by Horváth and Stanley [21]. Moving a piece of paper such that the interface always remained at a fixed distance H above a reservoir, they found a power law behaviour for the time correlation function $C_2(t) \sim t^\beta$ with $\beta = 0.56$. Another result is that the velocity V at which the the piece of paper must be moved towards the reservoir in order to keep the interface at a constant H varies as $V \sim H^{-\Omega}$, where $\Omega = 1.6$. Notice how this implies an interface propagating such that $H(t) \sim t^{2/5}$, since $V = dH/dt$. This is slower than $\delta = 1/2$ expected from Darcy's equation, but consistent with our earlier discussion. Unfortunately, the scaling behaviour of the interface as a function of space was not discussed and/or measured in this otherwise very careful work.

It is certainly interesting that the value of β is constant for all heights considered. The different heights did however affect the time at which the time correlation functions saturated, and the values at which it did so. Horváth and Stanley found for $C(t)$ a scaling form

$$C(t) \sim V^{-\Theta_L} f(tV^{(\Theta_t + \Theta_L)/\beta}), \quad (18)$$

where $f(y)$ is a scaling function such that $f(y) \sim y^\beta$ for $y \ll 1$ and $f(y) \sim \text{const}$ for $y \gg 1$. The values of the independent exponents were $\Theta_L = 0.48$ and $\Theta_t = 0.37$ [57].

Another experiment was performed by Kwon *et al.* [23]. They deposited a paper towel on an inclined glass plate and followed the capillary rise of a dye solution, giving a mean interface dynamical propagation $\bar{h}(t) \sim t^{0.37}$. Again, two scaling regimes were present in

the saturated width; on small length scales (≤ 2 cm), $\chi = 0.67$ while on larger length scales (up to 20 cm) $\chi \sim 0.2$. Within the simple Family-Vicsek picture they obtained $\beta = 0.24$ on the short lengthscale regime.

Finally, Zik *et al.* [24] performed a horizontal capillary front experiment. They obtained rough interfaces only with highly anisotropic paper, for which they found $\chi = 0.4$. For isotropic paper, the roughness was at best logarithmic. It is remarkable, that the scaling for the anisotropic paper was observed through a large range of length scales, not only up to fiber length.

4.3 Analysis of the experiments: influence of fluid conservation

As the previous paragraphs show, there is a large amount of experimental work on the subject of spontaneous imbibition and front roughening, but a coherent picture does not emerge naturally. The results are quite contradictory and difficult to analyse properly.

We have already mentioned a few experimental difficulties inherent to imbibition in paper, the most important being the complicated flow profile of a fluid inside the paper, due to fiber swelling and dissolution. Certainly all experiments suffer from these difficulties at least to some degree. Apart from the work of Family *et al.* [18], all experiments are consistent with an average front propagating roughly according to the Washburn-Rideal result, (Eq. (2)). They do not agree perfectly, but definitely reflect the influence of the conservation of fluid on the propagation process.

One consistent feature to emerge from the experiments is an interface which has developed roughness only on very short length scales. A partial remedy to this problem is, as done by Amaral *et al.* [20], to introduce a length scale l_p above which the interface saturates. In their experiment and model (see also Sect. 3) it becomes visible in the width the interface takes at the height where it gets pinned by evaporation, and they establish the relation $w \sim H_p^\gamma \sim l_p^\chi$.

The idea of such a length scale has of course a wider range of applicability than in the context of imbibition with evaporation. Indeed, the experiment of Horváth and Stanley [21], done with little influence of evaporation, and no discernable influence of gravity, showed the existence of a similar length scale, in this case, related to the velocity V at which paper was being pulled down (or, equivalently, to the average height of the interface).

We have already shown that such a length scale $\xi_\times(H)$, connected to the existence of a conservation law existed. It can provide a natural connection between the work of Amaral *et al.* and Horváth and Stanley, provided that one is willing to revise the role of evaporation. If the sole role of evaporation is to stop the macroscopic progression of the interface, with little or no influence on the statistical fluctuations of the interface itself, then the width $w(t) \sim \xi_\times^\chi(H_p) \sim H_p^{\chi/2}$ and therefore $\gamma = \chi/2$. This length should also influence the setup examined by Horváth and Stanley [28]. Indeed, it provides an alternative explanation to the velocity (or alternatively height)

dependence of the saturated value of the time correlation function, since

$$C_2^2(\tau \rightarrow \infty) \sim \int_{1/\xi_x(H)}^{1/a} \frac{dk}{k^{2\chi+1}} \quad (19)$$

which yields $C_2(\tau \rightarrow \infty) \sim H^{\chi/2} \sim V^{-\chi/2}$. Note that as long as $\xi_x < L$, the correlation function $C_2(t)$ is independent of the total length of the system. Since only one length of paper was considered experimentally, it is unfortunately impossible to check this hypothesis.

However, in order to demonstrate convincingly the existence of such a length scale (or of any other length scales) and the scaling below it, it is imperative to extend the region of scaling. We fail to see how a sub-centimeter description of the roughness might be considered universal. It is quite possible that the crossover ξ_x associated with paper really is in the sub-centimeter range, in which case, the alternative way to check for scaling is in the scaling of the width. Even in the freely rising case, the total width of the interface should show some early time power law behaviour, with some exponent β .

One should also note the fact the structure of paper does have short-scale power-law correlations [59]. These are manifest in the two-point-correlation function of the areal mass density of the paper, with the cut-off of the effective power-law correlations extending in practice to a few times the fiber length. It is by no means clear what role such correlations play in imbibition, since penetration of the liquid may take place both inside the sheet, and on the outer surface. Also, it is not clear how these correlations map into correlations in the hydraulic conductivity or local permeability. They however remind of the fact that scaling exponents established on lengthscales that are also relevant from the microstructural viewpoint should be taken with a grain of salt. Still, it is quite possible that the effective noise correlator at the interface will display power law behaviour, as in the case of forced flow through Hele-Shaw cells [60].

5 Summary and perspective

In this paper, we have tried to extract a coherent picture of all previous studies about interface roughening in spontaneous imbibition experiments. Concerning theoretical understanding, the DPD picture plausibly reproduces the properties of the pinned interface. We strongly feel that not enough attention has been paid to the macroscopic dynamical features and their connection to the underlying microscopic structure. The flow of fluid in paper is influenced by many microscopic mechanisms, and a study of the average front velocity already indicates which ones may or may not be relevant. The main experimental characteristic of imbibition certainly is the slowing down of the front, a phenomenon not seen in other systems. Some very simple predictions concerning evaporation and gravity would already allow a solid comparison between the expected behaviour and the experimental results.

An important reason to consider the fluctuations of the propagating front is the presence of a lateral length scale ξ_x , for which experimental evidence already exists [20,21], and which has a very strong influence on the roughening process.

Finally, we believe that a complete description of imbibition (including the dynamic interface) must include a conservation law for the whole fluid, not simply at the interface level. This is for two reasons, first the average interface ought to slow down continuously, and secondly the dynamics of the interface need to have long range interactions. Also, any model must deal the quenched nature of the noise in a suitable way. These requirements make it impossible to describe the interface roughening with a local equation of motion. To answer these criteria, we introduce in the next paper [28] a phase-field model of imbibition, which incorporates explicitly a conservation law for the liquid in a disordered medium, and thus produces the correct macroscopic physics.

We finish this overview by pointing out a few experimental directions. The use of “ordinary” paper for the medium creates some problems that are usually neglected. In this respect it comes to mind that other liquids than water or water/ink mixtures might well prove advantageous, in that they are less susceptible to evaporation and/or do not interact with the fibers themselves. Another option is to monitor the roughness of the front using an Hele-Shaw cell. This eliminates the problem of fiber-liquid interaction.

We wish to acknowledge interesting and stimulating discussions with J. Kertész, T. Ala-Nissila, K. R. Elder, S. Majaniemi, and J. Lohi. This work has been supported by the MATRA program of the Academy of Finland.

References

1. See *e.g.*, A.-L. Barabási, H.E. Stanley, *Fractal Concepts in Surface Growth* (Cambridge University Press, Cambridge, 1995).
2. J. Krug, *Adv. Phys.* **46**, 139 (1997).
3. M. Kardar, G. Parisi, Y.-C. Zhang, *Phys. Rev. Lett.* **56**, 889 (1986).
4. S.F. Edwards, D.R. Wilkinson, *Proc. R. Soc. London A* **281**, 17 (1982).
5. H. Leschhorn, *Phys. Rev. E* **54**, 1313 (1996).
6. T. Nattermann, S. Stepanow, L.-H. Tang, H. Leschhorn, *J. Phys. II France* **2**, 1483 (1992).
7. H. Leschhorn, T. Nattermann, S. Stepanow, L.-H. Tang, *Ann Phys.* **6**, 1 (1997).
8. O. Narayan, D.S. Fisher, *Phys. Rev. B* **48**, 7030 (1993).
9. Z. Csahók, K. Honda, E. Somfai, E. Vicsek, T. Vicsek, *Physica A* **200**, 136 (1993).
10. J. Maunuksela, M. Mylly, O.-P. Kähkönen, J. Timonen, N. Provatas, M.J. Alava, T. Ala-Nissila, *Phys. Rev. Lett.* **79**, 1515 (1997).
11. M.A. Rubio, *Phys. Rev. Lett.* **63**, 1695 (1989).

12. V.K. Horváth, F. Family, T. Vicsek, J. Phys. A **24**, L25 (1991).
13. S. He, G.L.M.K.S. Kahanda, P.-z. Wong, Phys. Rev. Lett. **69**, 3731 (1992).
14. T. Delker, D.B. Pengra, P.-z. Wong, Phys. Rev. Lett. **76**, 2902 (1996).
15. A. Dougherty, N. Carle, Phys. Rev. E **58**, 2889 (1998).
16. S.V. Buldyrev, A.-L. Barabási, F. Caserta, S. Havlin, H.E. Stanley, T. Vicsek, Phys. Rev. A **45**, R8313 (1992).
17. A.-L. Barabási, S.V. Buldyrev, S. Havlin, G. Huber, H.E. Stanley, T. Vicsek, in *Surface Disordering: Growth, Roughening and Phase Transitions*, edited by R. Jullien, J. Kertész, P. Meakin, D.E. Wolf (Nova Science, Commack, 1992).
18. F. Family, K.C.B. Chan, J. G. Amar, in *Surface Disordering: Growth, Roughening and Phase Transitions*, edited by R. Jullien, J. Kertész, P. Meakin, D.E. Wolf (Nova Science, Commack, 1992).
19. S.V. Buldyrev, A.-L. Barabási, S. Havlin, J. Kertész, H.E. Stanley, H.S. Xenias, Physica A **191**, 220 (1992).
20. L.A.N. Amaral, A.-L. Barabási, S.V. Buldyrev, S. Havlin, H.E. Stanley, Phys. Rev. Lett. **72**, 641 (1994).
21. V.K. Horváth, H.E. Stanley, Phys. Rev. E **52**, 5166 (1995).
22. P.B.S. Kumar, D. Jana, Physica A **224**, 199 (1996), see also `cond-mat/9509047`.
23. T.H. Kwon, A.E. Hopkins, S.E. O'Donnell, Phys. Rev. E **54**, 685 (1996).
24. O. Zik, E. Moses, Z. Olami, I. Webman, Europhys. Lett. **38**, 509 (1997).
25. E. Bouchaud, J. Phys. Cond. Matt. **9**, 4319 (1997).
26. J. Kertész, V.K. Horváth, F. Weber, Fractals **1**, 67 (1993).
27. T. Engøy, K.J. Måløy, A. Hansen, S. Roux, Phys. Rev. Lett. **73**, 834 (1994).
28. M. Dubé, K.R. Elder, M. Rost, M.J. Alava, S. Majaniemi, T. Ala-Nissila, Eur. Phys. J. B **15**, 701 (2000). See also M. Dubé *et al.*, Phys. Rev. Lett. **83**, 1628 (1999).
29. A.E. Scheidegger, *The Physics of Flow through Porous Media* (MacMillan Co, New-York, 1957).
30. M. Sahimi, Rev. Mod. Phys. **65**, 1393 (1993).
31. J. S. Rowlinson, B. Widom, *Molecular Theory of Capillarity*, (Clarendon Press, Oxford, 1982).
32. P.G. de Gennes, Rev. Mod. Phys. **57**, 827 (1985), other references may be found in, *e.g.*, J.F. Joanny, L. Léger, Rep. Prog. Phys. **55**, 431 (1992).
33. L.D. Landau, *Fluid Mechanics* (Pergamon Press, NY, 1979).
34. E.W. Washburn, Phys. Rev. **17**, 273 (1921).
35. E.K. Rideal, Philos. Mag. **44**, 1152 (1992).
36. J. Szekely, A.W. Neumann, Y.K. Chuang, J. Colloid Interface Sci. **35**, 273 (1971).
37. T.E. Mumley, C.J. Radke, M.C. Williams, J. Colloid Interface Sci. **109**, 398 (1986), *ibid* **109**, 413 (1986).
38. R. Hilfer, Phys. Rev. E **58**, 2090 (1998).
39. R.D. Deegan *et al.*, Nature **389**, 827 (1997).
40. J.A. Bristow, Svensk Papperstidning **20**, 645 (1971).
41. J.S. Aspler, S. Davis, M.B. Lyne, J. Pulp and Paper Sci. **13**, J55 (1987).
42. I. Pérezron, G. Bourgain, D. Quéré, J. Colloid Interface Sci. **173**, 319 (1995).
43. R. Lenormand, E. Touboul, C. Zarcone, J. Fluid Mech. **189**, 185 (1988).
44. T. Gillespie, J. Coll. Sci. **13**, 32 (1958); **14**, 123 (1959).
45. See, *e.g.*, D.E. Eklund, P.J. Salminen, Tappi J. **69**, 116 (1986).
46. O. Zik, T. Kustanovich, E. Moses, Z. Olami, Phys. Rev. E **58**, 689 (1998).
47. J. Krug, Phys. Rev. Lett. **72**, 2907 (1994).
48. J.M. Lopez, M.A. Rodriguez, R. Cuerno, Phys. Rev. E **56**, 3993 (1997).
49. N. Martys *et al.*, Phys. Rev. **44**, 12294 (1991).
50. K. Sneppen, Phys. Rev. Lett. **69**, 3539 (1992).
51. K. Sneppen, M.H. Jensen, Phys. Rev. Lett. **71**, 101 (1993).
52. Z. Olami, I. Procaccia, R. Zeitak, Phys. Rev. E **49**, 1232 (1994).
53. H. Leschhorn, L.-H. Tang, Phys. Rev. E **49**, 1238 (1994).
54. J. Krug, P. Meakin, Phys. Rev. Lett. **66**, 703 (1991).
55. This can be seen easily from Figure (2) of reference [16].
56. On length scales smaller than 1.5 cm, there may be signs of scaling, with a $\chi \sim 0.7$, but again, it is difficult to attach any universal significance to length scales of the order of the fiber length.
57. Horvath and Stanley also included the total system size L in a scaling hypothesis, but since only one paper size was used, their hypothesis could not be fully tested.
58. As far as we are aware, only the experimental groups of references [18], [23] and [24] have realised this physical fact, but the implications of it were not discussed in any elaborate ways.
59. N. Provatas, M.J. Alava, T. Ala-Nissila, Phys. Rev. E **54**, R336 (1996).
60. V.K. Horváth, F. Family, T. Vicsek, Phys. Rev. Lett. **67**, 3207 (1991).