Kinetic Roughening and Pinning of Two Coupled Interfaces in Disordered Media

Alexander S. Balankin,^{1,2} Rafael García Paredes,¹ Orlando Susarrey,¹ Daniel Morales,² and Fernando Castrejon Vacio²

¹Grupo "Mecánica Fractal," Instituto Politécnico Nacional, México D.F., 07738 México

²Instituto Mexicano de Petróleo, México D.F., 07730 México

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We studied the kinetic roughening dynamic of two coupled interfaces formed in paper wetting experiments at low evaporation rate. We observed three different regimes of impregnation in which kinetic roughening dynamics of coupled precursor and main fronts belong to different universality classes; nevertheless both interfaces are pinned in the same configuration. Reported experimental observations provide a novel insight into the nature of kinetic roughening phenomena occurring in the vast variety of systems far from equilibrium.

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Kinetic roughening of interfaces occurs in a wide variety of physical situations ranging from thin solid film growth and fracture phenomena to the fluid invasion in porous media [1,2]. Extensive theoretical and experimental studies of the last decade state that in a system with quenched disorder an initially flat interface grows and roughens continuously as it is driven by some external force F [1]. If F is larger than a critical force F_C , the interface moves with a finite velocity, while it remains pinned by the disorder if $F < F_C$. In both cases the interface roughening dynamic displays power-law scaling, characterized by a set of scaling exponents [3]. Furthermore, it is believed that dynamic scaling of interfaces formed in quite different systems can be classified in a few classes of universality characterized by the universal values of scaling exponents [1,2,4]. Specifically, in many cases the global width of moving interface z(x, t) behaves according to the Family-Vicsek dynamic scaling ansatz as [1]

$$W(L,t) = \overline{\langle [z(x,t) - \bar{z}]^2 \rangle}^{1/2} \propto t^{\alpha/z} f(L/\xi(t))$$
(1)

where the overbar denotes average over all x in a system of size L and the brackets denote average over different realizations; $\xi \propto t^{1/z}$ is the horizontal correlation length, and the scaling function behaves as $f(y) \propto y^{\alpha}$ if $y \ll 1$ and it becomes a constant when $y \gg 1$; α , z, and $\beta = \alpha/z$ are so-called roughness, dynamic, and growth scaling exponents. More generally, a moving interface is characterized by the *q*-order height-height correlation functions [1]: $\sigma_q(\Delta, t) = \langle \langle |z(x, t) - z(x + \Delta, t)|^q \rangle_\Delta \rangle_P^{1/q}, \text{ where } \langle \dots \rangle_\Delta$ denotes a spatial average over axis x in a window of size Δ , and $\langle \ldots \rangle_P$ denotes average over different positions. In the absence of any characteristic length, except system size L, the interface is expected to exhibit a self-affine invariance. If so, $\sigma_q(\Delta, t)$ also satisfies the Family-Vicsek dynamic scaling ansatz, $\sigma_q(\Delta, t) \propto t^{\alpha/z} f(\Delta/\xi_q(t))$. However, many rough interfaces in nature exhibit a multiscaling behavior characterized by different scaling exponents for different moments of the height distribution. The multiaffine roughness is characterized by the spectrum of scaling exponent ζ_q defined by scaling relations: $\sigma_q(\Delta, t \gg \xi^z) \propto \Delta^{\zeta_q}$ [5]. Moreover, in the case of socalled anomalous kinetic roughening, the global and the local width of moving interface are characterized by different scaling exponents: $\zeta_2 < \alpha$ and $\beta_{loc} < \beta$ [6].

Another interesting topic is a kinetic roughening and pinning of coupled interfaces moving in a disordered medium [7,8]. Theoretical considerations and numerical simulations predict the strong effects of coupling between moving interfaces on their roughening dynamics [6,7]. However, as far as we know, no experimental studies of these phenomena have been reported. In this work we perform a detailed study of kinetic roughening dynamics and pinning of two coupled interfaces moving in a paper.

Different kinds of paper were widely used as model media in experimental studies of kinetic roughening of moving interfaces [1,2,9]. While the initial hope of most studies was to determine the universality class corresponding to different phenomena (fracture [10], imbibition [2,11], fire propagation [12]), experimental results were far from theoretical expectations [1,2,9–13]. Specifically, in paper impregnation experiments it was found that scaling exponents are dependent on the paper structure [9,14] and, probably, on the evaporation rate [15]. Furthermore, generally, liquid invasion in porous medium generally involves two simultaneous flows: film flow, which propagates along the pore surfaces, and bulk flow, which saturates the pore spaces [16]. Accordingly, we can distinguish between two interfaces moving through the medium: the main impregnation front and the precursor front [17]. Film flow is especially important in paper wetting experiments [18]. However, no experimental studies of two front roughening dynamics in wetted paper were performed.

In this work we perform paper impregnation experiments by clipping a sheet $(500 \times 200 \text{ mm}^2)$ of "Filtro" paper with open porosity [19] to a ring stand, and allowing it to dip into a reservoir filled with black Chinese-ink suspension [20]. Experiments were performed in a climate box with controlled temperature and humidity. The size of

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the reservoir was large enough to assure the free surface of ink at a constant level z = 0 [21]. The moving fronts were monitored with a high resolution (6.5 megapixels) video recorder [22]. All snapshots (see Fig. 1) were digitized and plotted as single-valued functions z(x) [23] with the help of Scion-Image software [24]. To avoid the effect of sheet borders on the front configurations, the scaling analysis was performed only on the central parts of interfaces, leaving the edges of width 0.2L (see Refs. [2,11,12]).

First of all, we note that paper impregnation behavior depends on the air humidity and temperature during the experiment. Namely, at high evaporation rates (low humidity <25%, $T = 25 \pm 3$ °C) the ink is absorbed into the paper, forming a unique rough interface between the wet and dry regions which rises along the paper and roughens until it is pinned by the disorder in paper structure. In the opposite case of low evaporation (higher humidity >35%, $T = 25 \pm 3$ °C) after a short initial period ($t < t_1$) the filtered water front overtakes the main impregnation front (see Fig. 1). Accordingly, in the transient stage ($t_1 < t <$ $t_{\rm S}$) the distance between precursor (water film flow) and impregnation (bulk flow) fronts increases and achieves the maximum at $t = t_S$. At the final stage $(t > t_S)$ the distance between precursor and main fronts decreases until both fronts coincide in the pinning position at $t = t_P$ (see Figs. 1 and 2). The coefficient of correlations between precursor $z_W(x, t)$ and main front $z_I(x, t)$ configurations, C(t) = $\operatorname{Cov}[z_W(x, t); z_I(x, t)] / \sigma_2(z_W) \sigma_2(z_I)$, which characterizes the strength of coupling between two moving interfaces, decreases from $C(t_1) = 1$ to the minimum $C(t_S) =$ $\min_{t} C(t)$ and then increases until both interfaces coincide in the same configuration and $C(t_P) = 1$ [see Fig. 2(d)].



FIG. 1. Digitized snapshots of wetting fronts in paper at times: (a) $t = t_1 = 115$ sec, (b) t = 150 sec, (c) t = 500 sec, (d) t = 1000 sec, (e) t = 2000 sec, (f) $t = t_s = 3600$ sec, (g) t = 4000 sec, (h) t = 4500 sec, (i) t = 5000 sec, (j) t = 5600 sec, (k) $t = t_p = 6100$ sec.

We found that the time intervals corresponding to different stages, as well as $C(t_S)$ and pinning height [25], are strongly dependent on the evaporation rate controlled by the air humidity in the climate box (see Ref. [15]). Accordingly, to study the coupled roughening of two interfaces, we perform 30 experiments at fixed humidity of 45% and temperature $T = 25 \pm 3$ °C; all data reported below are averaged over 30 measurements.

In these conditions, we found that at the initial stage ($t \le t_1 = 115 \pm 25 \text{ sec}$) the unique wetting front rises in the Washburn regime, i.e., $h(t) = \overline{z(x, t)} \propto \sqrt{t}$ [see Fig. 2(a)] and the global interface width grows as $W \propto t^{\beta}$ with $\beta = 0.40 \pm 0.05$ [see inset in Fig. 2(c)]. The spatial roughness of interface moving in the Washburn regime is characterized by the local roughness exponent $\zeta = 0.50 \pm 0.02$ [see Figs. 3(a) and 2(b)]. These findings indicate that roughening dynamics belongs to a directed percolation universality class in (2 + 1) dimensions, as it is expected for imbibition in a 3D medium (see Ref. [26]).

In the transient regime, $t_1 < t < t_S = 3600 \pm 300$ sec, the precursor film flow and the bulk impregnation both exhibit deviation from the Washburn behavior. Specifically, we find that both fronts rise as $h \propto t^{\delta}$ [see Fig. 2(a)], but with different exponent δ [27]: $\delta_W =$ 0.41 ± 0.03 for precursor film flow and $\delta_I = 0.38 \pm 0.02$



FIG. 2. (a) The plots of suspension (1), precursor (2, 3), and ink (4, 5) height versus time (curves are power-law fits of experimental data); inset shows the transition from Washburn to transient regime of imbibition. (b) Time dependence of roughness exponents of suspension (1) precursor (2, 3), and ink (4, 5)fronts. (c) Log-log plots of suspension (1), precursor (2, 3), and ink (4, 5) front widths versus time (dashed and dotted lines (5) correspond to the logarithmic $W(t_S \le t \le t_P) = 161.6 -$ 16.46 Int ($R^2 = 0.99$) and power law $W(t_S \le t \le t_P) \propto t^{-0.74}$ $(R^2 = 0.94)$ fittings, respectively); inset shows the time behavior W(t) in the Washburn regime of imbibition. (d) Log-log plot of correlation coefficient C(t) versus time [the slopes of straight lines are $\mu = 0.154$ (I) and $\mu = 0.98$ (II)]. (e) Log-log plots of $\Delta \zeta$ versus C at the transient (full circles) and saturation (open circles) stages of impregnation; straight line is data fitting by Eq. (3).



FIG. 3. Roughness scaling plots of (a) ink front at t = 60 sec (1), t = 1300 sec (2), and t = 4200 sec (3); (b) ink front at t = 1300 sec for q = 1 and $\zeta_1 = 0.8 \pm 0.04$ (1), q = 2 and $\zeta_2 = 0.7 \pm 0.03$ (2), and q = 4 and $\zeta_4 = 0.6 \pm 0.04$ (3); (c) precursor water front at t = 1300 sec and $\zeta = 0.5 \pm 0.02$ (1), t = 4920 sec and $\zeta = 0.65 \pm 0.03$ (2), and t = 6000 sec and $\zeta = 0.78 \pm 0.03$ (3); (d) pinned front for q = 1, 2, and 4. All graphs in this figure are shifted for clarity (1 pixel = 0.078 mm).

for ink suspension flow [28]. The growth exponent of precursor front in the transient regime is found to be $\beta_W =$ 0.24 ± 0.02 and the roughness exponent is a constant $\zeta =$ 0.50 ± 0.02 [see Fig. 2(b)]. So, in the transient regime, the roughening dynamics of film flow front is consistent with the Edwards-Wilkinson universality class in (1 + 1) dimensions [2]. In contrast to this, the moving ink front in the transient regime is characterized by $\beta_W = 0.46 \pm 0.04$ and the continuously varying roughness exponent $\zeta(t)$, which increases from $\zeta(t_1) = 0.5 \pm 0.02$ to $\zeta(t_s) =$ 0.80 ± 0.02 [see Figs. 2(b) and 2(c)]. Moreover, we find that a moving ink front in a transient regime does not exhibit self-affine invariance; rather it is multiaffine [see Fig. 3(b)]; i.e., the kinetic roughening of main impregnation front exhibits the generalized multiscaling with timedependent roughness exponent (see Ref. [29]). The latter can be attributed to the time dependence of pore capillarity forces due to the swelling of fibers wetted by a precursor water film (see Ref. [2]). Accordingly, the observed behavior of the impregnation front is consistent with the (suggested in Ref. [30]) universality class with absorbing states, characterized by continuously varying critical exponents.

At the saturation stage ($t_S < t \le t_P = 6100 \pm 600 \text{ sec}$) the ink front becomes self-affine [$\zeta_q(t \ge t_S) = \zeta(t_S) = 0.80 \pm 0.02$; see Fig. 3(d)] and rises with the same rate as in the transient regime [see Fig. 2(a)]; nevertheless, its global width decreases logarithmically with time [see Fig. 2(b)] [31]. In contrast to this, the precursor rise rate drastically decreases, whereas the precursor front growth exponent increases (see Table I). Furthermore, the local roughness exponent of the precursor front linearly increases from $\zeta(t_S) = 0.50 \pm 0.02$ to $\zeta(t_P) = 0.80 \pm 0.02$ [see Figs. 3(c) and 2(b)]. The correlations between two fronts also increase from $C(t_S) = 0.59 \pm 0.05$ to $C(t_P) =$ 1 [see Fig. 2(d)]. We find that in both regimes of impregnation C(t) behaves as

$$C(t) \propto t^{\mu}, \tag{2}$$

but with different scaling exponents [see Fig. 2(d)]. Namely, in the transient regime $\mu = -0.154$ ($R^2 = 0.9997$), whereas at the saturation stage $\mu = 0.987$ ($R^2 = 0.9983$).

The large values of C(t) > 0.5 indicate a strong coupling between precursor and main fronts. The asymmetric effect of coupling on the kinetic roughening dynamics of two interfaces in disordered media can be described by the set of coupled stochastic continuum equations suggested in Ref. [7]. In the transient regime the swelling of paper wetted by precursor water influences the fluctuations of the main impregnation front, whereas at the saturation stage the main front drives the kinetic roughening of precursor. The most surprising result of the present studies is that coupled interfaces are pinned in the same configuration [32]. Moreover, we found that the difference between roughness exponents of precursor and main fronts $\Delta \zeta(t) = |\zeta_W(t) - \zeta_I(t)|$ in both regimes depends on the coupling strength, characterized by C(t), as

$$\Delta \zeta = k(1 - C), \tag{3}$$

where $k = 0.73 \pm 0.02$ [33] [see Fig. 2(e)].

As far as we know, this work presents the first experimental study of kinetic roughening and pinning of coupled interfaces in disordered media. So, results of this work offer a new insight into the problem of kinetic roughening phenomena.

TABLE I. Values of scaling exponents characterized different regimes of kinetic roughening of interfaces formed in paper wetting experiments.

	δ		β		ζ	
Stage	Precursor	Ink front	Precursor	Ink front	Precursor	Ink front
$t < \tau_1$	0.50 ± 0.02		0.40 ± 0.05		0.50 ± 0.02	
$\tau_1 < t < \tau_S$	0.41 ± 0.03	0.38 ± 0.02	0.24 ± 0.02	0.46 ± 0.04	0.50 ± 0.02	$\zeta(t)$
$\tau_S < t < \tau_p$	0.19 ± 0.03	0.38 ± 0.02	0.48 ± 0.02	no scaling	$\zeta(t)$	0.80 ± 0.02
$t \ge \tau_p$	0		0		0.80 ± 0.02	

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- [21] All height measurements were made taking into account this reference level.
- [22] Additionally, all pinned fronts were scanned from both sides of the paper with resolution 300 pp. From these images we found that two configurations of wetting front obtained from the left and right sides of the paper do not coincide—the difference between configurations $\Delta z(x) = z_L(x) z_R(x)$ is much larger than the paper thickness $a = 0.32 \pm 0.03$ mm. Namely, we found $\max_{x \in L} \Delta z(x) = 3.5 \pm 0.5$ mm $\gg a$. So, for the purpose of this work, the Filtro paper should be treated as a three-dimensional porous medium.
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- [32] It should be emphasized that the roughness exponent of pinned interfaces ($\zeta = 0.80 \pm 0.02$) is found to be independent on the evaporation rate; nevertheless, the characteristic times t_1 , t_s , and t_p , as well as the final position of interface $h(t_p)$, strongly depend on the evaporation rate and so the roughness exponent values at equal times or equal heights are functions of evaporation rate. Moreover, the roughness of pinned interfaces is characterized by the same exponent that was found for pinned interfaces in the same paper in experiments with high evaporation rates [9], when the precursor front was not presented.
- [33] Best fits give k = 0.75 for the transient regime and k = 0.71 for the saturation stage.