Spontaneous Imbibition Experiment in Newspaper Sheets

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We study the behavior of an ink-paper interface in a spontaneous imbibition experiment as a function of time and paper orientation. To characterize the interface roughness the growth β and Hurst H exponents (calculated using the root mean square interface width W) and the H_q scaling exponent (calculated using the qth order height-height correlation function) are used. Our results indicate that the values of H and H_q depend on the orientation of the paper sheets, while β does not, and that the interface exhibits a multiaffine character during all its evolution.

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The study of rough interfaces is a subject of intensive research and has attracted much scientific and industrial interest. Rough interfaces can be obtained from the fracture process in disordered materials [1–5], fluid flow in porous media [6–9], growth of bacterial colonies [10], etc.

The irregularity of the interface is often characterized by its roughness, or width, $W(\varepsilon)$, defined as the rms value of the fluctuations of the surface height $h_i = h(x_i)$ over a length scale ε [1]: $W^2(\varepsilon) = \langle\langle[h(x) - \langle h\rangle_\varepsilon]^2\rangle_\varepsilon\rangle_x$, where $\langle\cdots\rangle_\varepsilon$ denotes an average over x in windows of size ε and $\langle\cdots\rangle_x$ denotes average over all x in a system of size N.

An important characteristic of the rough interface is its self-affine character. For this type of interface the function $h(x)$ has the same statistical properties as $k^{-H}h(kx)$, where k is a constant and H is known as the Hurst exponent, which satisfies $0 < H < 1$. For a self-affine interface, the roughness W over a length scale ε satisfies the scaling law $W(\varepsilon) \sim \varepsilon^H$. Another interesting behavior is the dependence of the roughness W with time t which obeys the power law $W(t) \sim t^\beta$, where β is the growth exponent. Together, the β and H scaling exponents can be used to characterize the universality class of a system. Different systems with the same value of H and β have the same universality class.

There are rough surfaces which are not characterized by a single scaling exponent H, but by many values of H [11]. In this case the surface exhibits multiaffine properties which can be investigated by calculating the qth order height-height correlation function as a function of the length scale Δ [10]: $C_q(\Delta) = \langle[h(x) - h(x + \Delta)]^q\rangle_x$.

For many rough surfaces, it has been found that the qth order height-height correlation function obeys the power law $C_q(\Delta) \sim \Delta^{qH_q}$, where H_q is a scaling exponent which changes continuously with the qth order of the moments of the height-height correlation function.

Nowadays there is an intense discussion, both theoretical and experimental, about the universality of the scaling exponents H and H_q regarding roughening dynamics studies in many different areas. Also, many works found in the literature have used the C_2 height-height correlation function to calculate the "Hurst exponent" instead of $W(\varepsilon)$ [7,10].

Many of these works have paid particular attention to the physics of fluid flow in a porous medium, which involves many interesting phenomena including imbibition, i.e., the displacement of a viscous fluid by a more viscous one. In this process the propagation of the fluid interface into the medium can be either spontaneous or forced. In the spontaneous imbibition process the propagation of the fluid is due to capillary forces alone, while for forced imbibition there is also an external force applied in the system. An important factor which influences the dynamic of propagation of the interface is the disorder. It acts as a contrary force to the movement, pinning the interface. The disorder can either change with time or not, in which case it is called quenched disorder.

Several experiments have been developed in order to investigate the kinetics of the interface produced in a spontaneous imbibition process under influence of capillary forces alone [2,6,7,12–17]. Many of these experiments have studied the interface roughness in terms of the β and H exponents. Among them are the experiments that used paper as a disordered medium [2,7,14–17]. Paper is a good candidate to investigate the spontaneous imbibition process in a medium with quenched disorder, due to its structural nonuniformity. Generally, the fiber network structure is highly disordered since the fibers are randomly positioned and oriented. The random nature of the fiber network acts as a quenched disorder obstructing the propagation of the interface. The roughness of this interface is solely due to the disorder in the paper. Nevertheless, in some types of paper, the fabrication process is such that the fibers acquire some order, having a tendency to align along one direction. This might affect the roughness of the moving interface.

In this work we have carried out experiments aiming the investigation of the universality of the exponents H and H_q and the correctness of the use of the height-height correlation function to calculate the Hurst exponent.

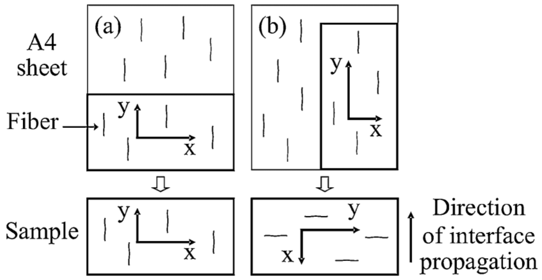

FIG. 1. Schematic representation of the process of orientation of the paper: (a) vertical sample and (b) horizontal sample. For the vertical sample the direction of interface propagation is parallel to the fibers while for the horizontal sample it is perpendicular to the fibers.

In order to achieve this we performed spontaneous imbibition experiments of ink into newspaper samples and investigated the dependence of the interface roughness on the direction of ink propagation along the newspaper. The interface roughness was characterized by the β and H exponents. Two directions, x and y, perpendicular to each other, were assigned to A4 sheets of newspaper ($(36.8 \pm 0.2)\ \mathrm{g/m^2}$), along the width and the length of the sheets, respectively (Fig. 1). For the A4 sheets used the fibers have a tendency to align themselves along the y direction. Samples of 21.6×6 cm were cut from the newspaper sheets and the orientation of the directions x and y were marked on them. Because of the orientation of the fibers in each of the two types of samples we refer to the sample oriented as shown in Fig. 1(a) as vertical and to the one oriented as shown in Fig. 1(b) as horizontal.

The samples were maintained vertically with the upper extremity fixed and the lower extremity dipped into a reservoir filled with a black ink suspension (Pilot TC 42). The ink reservoir was large enough to keep the free surface of ink at a constant level for the duration of the experiment. The ink was absorbed into the paper by capillary forces, creating a rough interface that moved upward. At determined time intervals the samples were photographed with a Canon A620 digital camera with 7.1 megapixels. To avoid any effects on the propagating interface that might arise from the borders of the samples, only the central 15 cm of the samples was photographed. Figure 2 shows some of the photographs obtained. All photographs were analyzed by a home made computer program in order to obtain the functions $h(x)$ which represents the patterns of the investigated interface lines. All overhangs present in the interfaces were removed considering, for a given point x, only the highest value of the function $h(x)$. After some days the interface becomes pinned due to the disorder in the paper structure. All experiments were performed in a box with controlled temperature ($32 \pm 1\,^{\circ}\mathrm{C}$) and humidity ($71 \pm 1\%$).

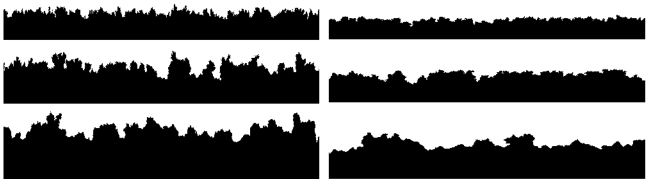

FIG. 2. Time evolution of the interface for the vertical (left) and horizontal (right) samples, after 1 (top), 5 (middle), and 25 h (bottom) from the beginning of the experiment.

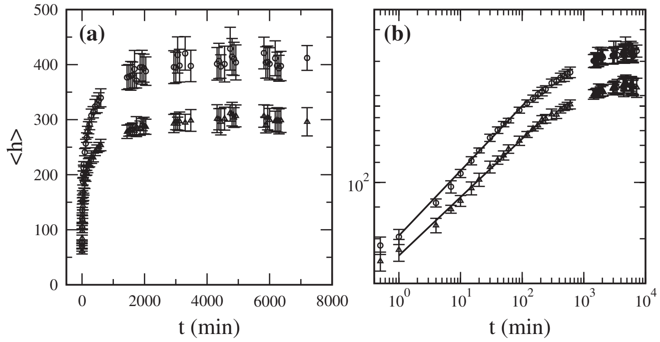

FIG. 3. (a) Average height $\langle h \rangle$ as a function of time t for the two different samples: vertical (circle) and horizontal (triangle). (b) Log-log plot of the data shown in (a). The slope of the straight line is 0.25 ± 0.03 for the vertical and 0.23 ± 0.03 for the horizontal sample.

All error bars presented in the figures were obtained averaging 10 independent realizations of the same experiment performed under the same conditions.

The behavior of the average interface height $\langle h \rangle$ as a function of time t for the two types of samples is presented in Fig. 3. It can be clearly observed that the orientation of the paper affects the interface dynamics. Notice that for a given time, the average hight is always larger for the vertical samples than for the horizontal ones. The saturation of the interface seems to occur after the same period of time for both orientations. It can also be observed in Fig. 3(b) that the slope of the straight region does not depend on the orientation of the samples. For the horizontal samples the direction of propagation of the interface is perpendicular to the fibers. In this case the fibers act as obstacles hindering

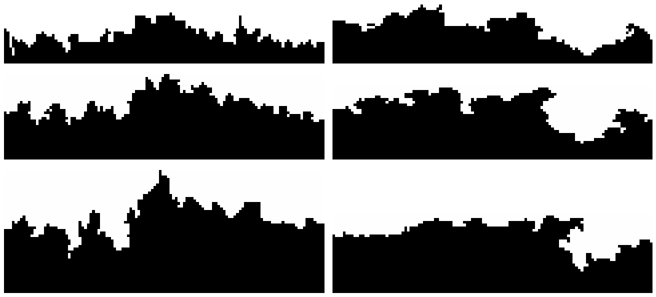

FIG. 4. Detailed view of the interfaces shown in Fig. 2 for the vertical (left) and horizontal (right) samples, after 20 (top), 30 (middle), and 40 min (bottom) from the beginning of the experiment. The horizontal size of all samples is 1.4 cm.

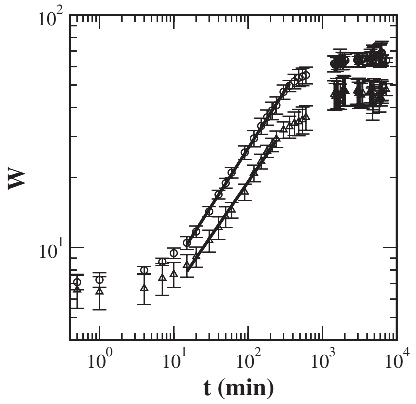

FIG. 5. Plot of the interface roughness as a function of time for the vertical (circle) and horizontal (triangle) samples.

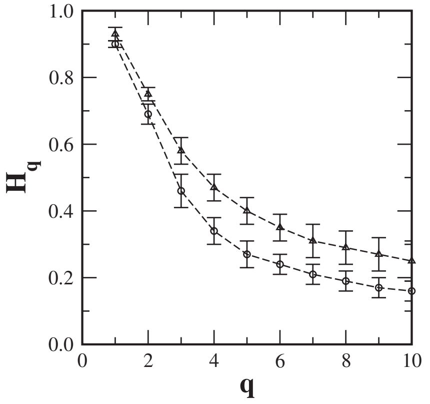

FIG. 7. The H_q spectrum for a time of 104 h from the beginning of the experiment for the vertical (circle) and horizontal (triangle) samples.

the upward movement of the interface: after a small "finger" has been formed at some place of the interface, the upward flow of ink is redirected along the direction of the fibers, forming an overhang (Fig. 4). This almost never happens for the vertical samples. It is possible to see in the experiments that when the average interface height is reaching saturation the movement of the interface occurs by means of jumps in some isolated regions, while the rest of the interface remains stationary. The sizes of the jumps are larger for the vertical samples than for the horizontal ones.

Our results indicate that the ink-paper interface does not move according to the Washburn law [18], where the average interface height obeys the power law $\langle h\rangle \propto t^{1/2}$. As observed in the Ref [9], Washburn-like scaling is not observed in many experiments of spontaneous imbibition due to a variety of factors (e.g., swelling of the paper fibers, non-Newtonian character of the ink, etc.).

Figure 5 shows the log-log plot of the interface roughness W as a function of time t for the two orientations of the paper. The roughness W was calculated using eq. 1 with ϵ equal to the size of the interface. The slope of the linear region is equal to the growth exponent β. Our results indicate that $\beta = 0.51 \pm 0.02$ for the vertical samples and $\beta = 0.49 \pm 0.03$ for the horizontal ones. These results suggest that the growth exponent β does not depend on the orientation of the paper sheet. Soriano *et al.* [19] studied, for different disorder configurations, a forced imbibition experiment using a Hele-Shaw cell. They also found $\beta \approx 0.5$ and verified that this value was independent of the disorder configuration. Note that, as for $\langle h\rangle$, the roughness for any time interval is higher for the vertical samples. This result also indicates that the orientation of the paper samples affects the process of propagation of the interface.

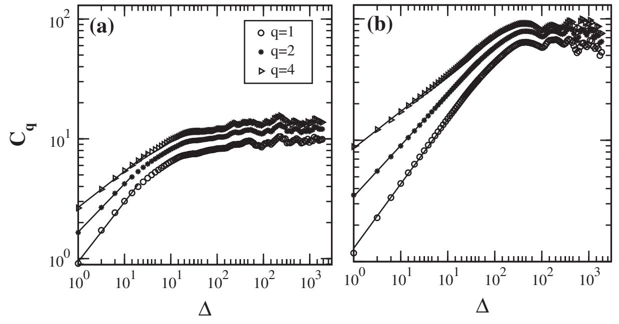

FIG. 6. Correlation function C_q as a function of the length scale Δ (a) before the pinned regime and (b) in the pinned regime. These plots were obtained for the horizontal samples. The same behavior is observed for the vertical samples.

Typical plots of the correlation function C_q as a function of the length scale Δ for two different time intervals are shown in Fig. 6. It can be observed that the slope of the straight regions of the correlation function are different for different values of q. This shows that the interface exhibits a multiaffine character during all its evolution. Our results are, then, different from the ones presented in Ref [7], where the multiaffine behavior was only observed before the pinned regime.

Figure 7 shows, for the vertical and horizontal samples, the H_q spectrum for a time of 104 h from the beginning of the experiment. Notice that the multiaffine behavior of the rough interface does not depend on the orientation of the

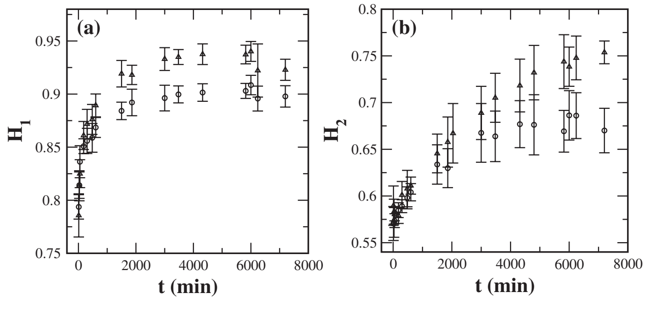

FIG. 8. Plot of the scaling exponents (a) H_1 and (b) H_2 as a function of time. The data were obtained for the two different samples: vertical (circle) and horizontal (triangle).

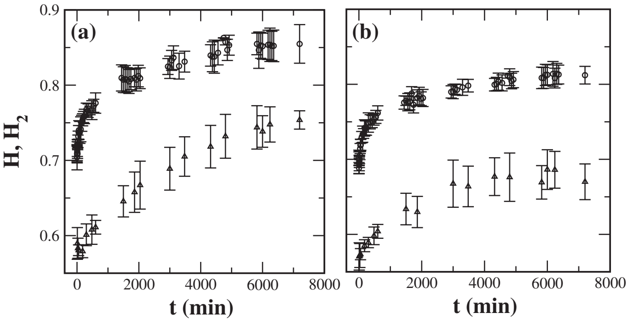

FIG. 9. Scaling exponent H (circle) and H_2 (triangle) as a function of the time t for two different samples: (a) horizontal and (b) vertical.

paper. However, the value of the scaling exponent H_q depends on the orientation of the paper.

The behavior of the exponents H_1 and H_2 as a function of time is presented in Fig. 8. It can be observed two regions: one where H_1 and H_2 are time dependent (in this region the interface has not yet reached the pinned state) and another where H_1 and H_2 are constant in time (here the interface is pinned). The time dependence of H_1 and H_2 can be attributed to the time dependence of the capillary forces due to the swelling of the fibers wetted by the ink. A similar result was observed by Balankin *et al.* [7] in the study of the kinetic roughening dynamics of two coupled interface formed in paper wetting experiments. Notice that the values of H_1 and H_2 depend on the direction of propagation of the ink-paper interface for all time intervals (this is true even for $0 < t < 1000$ min). This is due to the different orientation of the fibers.

Figure 9 shows the behavior of the Hurst exponent H, calculated using $W(\varepsilon) \sim \varepsilon^H$, and the scaling exponent H_2, calculated using the C_2 height-height correlation function. It is clear that although the behavior of both exponents is the same for each type of sample their values are not, even though it is expected they should be. These results show that H and H_q are not universal. Also, the values of H and H_2 depend on the method used to calculate them. Several works have used the height-height correlation function $C_q(\Delta)$ to calculate the "Hurst exponent H". Actually, what is calculated with $C_q(\Delta)$ is the H_2 scaling exponent, the Hurst exponent being calculated using the interface width. Since our results show that the values of H and H_2 may be different, care must be taken in using $C_q(\Delta)$ to calculate the Hurst exponent.

The dependence of H with the orientation of the paper was also observed by Menezes-Sobrinho *et al.* [1] in the study of the crack propagation in paper samples. It is interesting to comment that in the fracture process performed in the same type of paper as the present work the roughness profile displays a self-affine character.

In conclusion, our results show that for the imbibition of ink in paper the dynamics of the rough interface depends on the orientation of the paper fibers in relation to the direction of the interface propagation. We have found that the interface exhibits a multiaffine character during all its evolution. It was shown, also, that H and H_q are not universal. They depend on the orientation of the paper sheets while the growth exponent β does not. To our knowledge, this is the first experimental evidence of the dependence of the orientation of the paper for the kinetic roughening.

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