Non-equilibrium dynamics of fluids in disordered media

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Summary. Fluid flows in disordered media are present in natural and industrial processes such as soil irrigation and secondary oil recovery. These flows display complex spatial and temporal non-equilibrium dynamics arising from the heterogeneities of the medium. Average magnitudes are not sufficient to allow the complexity of their dynamics to be captured. Deeper insight can be gained from a scale-dependent statistical analysis of the fluctuations. Here we introduce the basic laws governing fluid flows in disordered media. Focusing on two-fluid displacements with a well-defined interface, we discuss several non-equilibrium dynamic features that include scale-invariance, avalanches, non-Gaussian fluctuations, and intermittency. [Contrib Sci 11(2): 189-198 (2015)]

Introduction

Fluid flow through disordered media occurs in geological, agricultural, and industrial processes of great importance. Crude oil and gas, for instance, is present in large natural reservoirs and impregnates porous rocks. In secondary oil recovery, water or gas is injected into the oil reservoir in order to displace the oil through the medium and drive it to the production wellbore. Underground water similarly flows through geological formations consisting of porous solids, granular materials, and fractured rocks. Flow in either porous or fractured media is, thus, a central topic in the fields of petrology and hydrology. It is also highly relevant in challenging industrial processes, such as the filtering of chemicals and contaminants.

The complexity of flows in disordered media arises from the heterogeneous structure of these media. The relevant features encompass a very wide range of spatial scales. The smallest correspond to pore sizes, typically ranging from 1 nm (micropores) to > 50 nm (macropores). Flow at pore scales is described by the classical hydrodynamic equations of mass continuity and momentum conservation. A continuum
description, on a coarser scale called the Darcy scale, can be obtained by proper averaging over a representative volume of the pore space. In averaging, the assumption of a random distribution of pore-scale properties is usually not sufficiently accurate. The presence of extended spatial correlations in natural porous formations (e.g., the clustering of low- and high-porosity areas, or the strong correlation between the volume of the throats and the average volume of the pores to which they are connected) must be taken into account. Flow at this scale is described by the equations of continuum mechanics of porous media, of which Darcy’s law (discussed below) plays a central role. The final scale of interest is the field scale, which can extend over kilometers. In this case, the large-scale heterogeneities in the physical properties of the medium are an important consideration. Consistently relating the micro-scale flow quantities to their corresponding Darcy-scale variables is key to understanding transport and transport-controlled processes such as chemical reactions in porous media [20].

Here we focus on two-phase fluid displacements in disordered media. In this kind of flow, a fluid originally residing in the disordered medium is displaced by a second, invading fluid. There is a well-defined (although eventually highly-distorted) interface separating the two fluids. Secondary oil recovery, soil irrigation, and several other relevant fluid displacements belong to this family of flows. As discussed below, both the morphology and the dynamics of the interface are dramatically influenced by the randomness inherent to the heterogeneous structure of the disordered medium. In considering the different features of this problem, several advanced concepts of non-equilibrium statistical physics are encountered, including scale invariance, complex correlations, anomalous fluctuations, and intermittency.

**Darcy’s law and permeability**

Henry Darcy, a pioneer in hydrology studies, carried out systematic experiments on the flow of water through beds of sands. In his monograph “Les fontaines publiques de la ville de Dijon” [4], published in 1856, he established that the discharge rate (volume per unit time) of fluid flowing through a long cylinder filled with sand was proportional to the pressure drop between the two ends of the cylinder (inlet and outlet). This linear relationship is analogous to Ohm’s law of electricity or Fourier’s law of heat conduction. However, like those other phenomenological relations, it has limited validity. Darcy’s law applies to creeping flows, for which the Reynolds number (Re) is $< 1$. Apart from geometrical factors, the proportionality coefficient in Darcy’s law is the ratio of an intrinsic property of the medium, its hydraulic permeability, to an intrinsic property of the fluid, its dynamic viscosity. The concept of hydraulic permeability makes sense only on a coarse-grained spatial scale (the Darcy scale), above the pore scale. Moreover, to have constant permeability a medium will

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1 This technique can result in the recovery of 20–40% of the original oil in place. This is a significant improvement over primary recovery, in which crude oil is driven into the wellbore by the combined action of the natural pressure of the reservoir and gravity, in which case only about 10% of a reservoir’s original oil is typically recovered in place.

2 The Reynolds number Re is a dimensionless ratio of inertial to viscous forces. It is typically used to distinguish between laminar (low Re) and turbulent (high Re) flows.

3 In a medium with a permeability of 1 darcy, a pressure gradient of 1 atm/cm on water (dynamic viscosity 1 mPa·s) produces a discharge rate of 1 cm$^3$/s through a cross-section of 1 cm$^2$. 
have to exhibit rather homogeneous and isotropic porosity on the Darcy scale. In geology and petroleum engineering permeability is measured in darcy units, with 1 darcy equal to 0.9869233 (µm)$^2$. Typical permeabilities of soils are 105 darcy for gravel, 1 darcy for sand, and 0.01 darcy for granite.

Even though Darcy derived his law phenomenologically, on the basis of his own experimental observations, it can be derived also from the Navier-Stokes equations of fluid mechanics as applied to stationary, creeping, incompressible flow, assuming that the viscous resisting force depends linearly on the fluid velocity. Analytic calculations can be carried out on simple geometries. Thus, an analysis of laminar flow through a long cylindrical capillary of radius $R$ shows that the permeability of the capillary is given by $R^2/8$. Similarly the hydraulic permeability of a Hele-Shaw cell, consisting of two large parallel plates separated by a narrow gap spacing $b$, is given by $b^2/12$. Combining simple geometries of this kind it is possible to derive approximate expressions of the hydraulic permeability of more complex materials, such as bundles of capillaries and beds of closely packed spheres. This approach to modeling the geometrical complexity of actual disordered media is an active field of research (Fig. 1).

Two-phase displacements

Many flows of interest involve the presence of an interface separating two different phases. Oil displacement by water in secondary oil recovery is an important example of two-phase displacement in a porous medium, in which the dynamics of the front between the two immiscible phases determine the efficiency of the recovery process. Printing, coating, impregnation, soil irrigation, and the rise of sap in plants are also examples of two-phase displacements.

If a cube of sugar or a biscuit is dipped in a cup of coffee or tea, the liquid quickly invades the pore spaces of the solid material, displacing the air initially present. The physical phenomenon behind this process of spontaneous fluid invasion, which seems to defy Earth's gravitational attraction, is capillarity. The interface separating the invading fluid from the displaced air touches the walls of the material on each pore. Depending on the relative interfacial energies of the three phases present (solid, liquid, and gas), there is a non-zero capillary force acting on the fluid menisci on each pore and throat of the material. In the case of tea or coffee invading a cube of sugar or a biscuit, this force drives the liquid into the medium. With mercury in glass, however, the capillary force would drive the menisci in the opposite direction, because glass is not wetted by liquid mercury, i.e., the contact angle is $>90^\circ$ (Fig. 2). The dynamics of the invasion process indeed depend on the relative ability of the two fluids (displacing and displaced) to wet the walls of the disordered medium. If the invading fluid preferentially wets the medium, the displacement is favored by capillary forces and is referred to as imbibition (from the Latin verb *imbibere*, to drink in). Conversely, when the preferentially wetting fluid is the displaced one, capillary forces oppose the displacement of the menisci inside the pores. The corresponding process is called drainage. It plays a central role in hydrology when surface and subsurface water are removed, either naturally or artificially, from an area thus preventing erosion and the leaching of nutrients.

A second important issue is the stability of the interface separating two fluids. The displacement may be either stable or unstable, depending on the relative viscosity of the fluids involved. The displacement is stable when the displacing fluid is more viscous than the displaced fluid. Because of

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4 Len Fisher won the Ig Nobel prize in Physics in 1999 for his experiments on the optimal way of dipping a biscuit into tea or coffee, an experience that enhances flavor release by up to ten fold. For a full account, see L. Fisher (1999) Nature 397:469.
capillary pressure fluctuations at the pore scale, the front at large scales is slightly irregular, but front disturbances cannot increase because the viscous pressure gradient on the side of the displacing fluid is larger than on the side of the displaced fluid. When the displacing fluid is less viscous than the displaced fluid (e.g., when water displaces oil) the situation is the opposite. Small front disturbances become amplified and rapidly growing fingers of the displacing fluid invade the displaced fluid, limiting the effectiveness of the displacement process. This is a very serious difficulty in secondary oil recovery and has motivated many research efforts [11].

The interfacial instability leading to viscous fingering is known as Saffman-Taylor instability [19]. In 1958, these authors studied two-phase displacements in a Hele-Shaw cell. The narrow gap of the cell results in high-friction (inertialess) bulk fluid motion that follows Darcy’s law, in analogy with flow in a disordered medium. Performing a linear stability analysis of the unperturbed front between the two fluids, Saffman and Taylor found that the interfacial tension along the front always damped infinitesimal perturbations of small wavelength; but, depending on the relative viscosity of the two fluids, viscous pressure could either damp or amplify infinitesimal perturbations of large wavelength. The linear stability of the front was thus controlled by two dimensionless ratios: the capillary number, \( \text{Ca} = \frac{\mu_1 v}{\gamma} \), which compares the strength of viscous and interfacial forces, and the viscosity ratio \( M = \frac{\mu_2}{\mu_1} \). Here \( \mu_1 \) and \( \mu_2 \) are the dynamic viscosities of displaced and displacing fluid, \( v \) the average velocity of the front, and \( \gamma \) its interfacial tension. Viscous fingers are formed when \( M < 1 \), covering a band of wavelengths that widens with \( \text{Ca} \). Saffman and Taylor also performed experiments in which air (of negligible viscosity) slowly displaced a viscous fluid, and thereby confirmed the predictions of their linear stability analysis. They found that a dynamic competition between air fingers of different sizes that formed at the onset finally gave rise to a stationary single finger that occupied half of the cell width at large \( \text{Ca} \). Their seminal work has been the starting point for a great deal of research activity on all possible aspects of viscous fingering and pattern formation resulting from this interfacial instability [2] (Fig. 3).

Differences in the wettability and viscosity of the fluids therefore provide a rationale for classifying two-phase fluid displacements in disordered media, as shown in Table 1.

Using quasi-two-dimensional transparent micromodels of porous media, Lenormand, Zarcone, and others explored these scenarios systematically and compiled their main findings in schematic phase diagrams, with \( \text{Ca} \) and \( M \) as the controlling parameters [13].

Very slow drainage is governed by capillary fingering. A fraction of the available channels are invaded by the less-wetting fluid, following the order dictated by the values of the capillary pressure jump across the meniscus in each channel. This sequential invasion is well described by the model of invasion percolation, and the resulting pattern is a self-similar fractal. At larger \( \text{Ca} \), unstable drainage results in a highly ramified pattern of invaded pores, also a self-similar fractal but of smaller fractal dimension. Its morphology and growth dynamics correspond to a process of diffusion limited aggregation, which describes the pattern formed by a growing unstable interface in an external field that obeys Laplace’s equation (Fig. 4). Finally, stable drainage displacements at large \( \text{Ca} \) produce compact patterns.

Fast imbibition displacements lead to the same morphologies as fast drainage, because the dynamic contact angle increases with \( \text{Ca} \) and makes the injected

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fluid invade the central part of the channels, as in drainage. However, slow *imbibition* displacements depend on the pore-to-channel aspect ratio. For a large aspect ratio, the injected fluid first wets the walls of the channels and then progressively invades the network by a sequence of collapses in the channels. At even slower displacements, the invasion mechanism is the flow of a precursor film ahead of the menisci, so that apparently disconnected filled channels may appear anywhere. For a small aspect ratio, invasion at moderate Ca takes place by a sequence of channel by channel pore invasions that lead to a faceted domain whose shape is dictated by the underlying network topology. Finally, very slow displacements combine this invasion mechanism with the flow of the precursor film, so that compact clusters appear anywhere in the network. Imbibition is therefore complicated by the underlying geometry of the model porous medium and by a new mechanism of invasion, the flow of a precursor film, at very low flow rates.

**Dynamics of capillary invasion in porous media**

An often-studied scenario of stable imbibition is the invasion of a porous medium by a liquid that preferentially wets the walls and displaces the air initially present, as in the case of the biscuit dipped in coffee. This process occurs spontaneously and is driven by capillary forces.

An equation for the average position of the invading front versus time can be derived in a continuous (hydrodynamic) framework at the Darcy scale. The equation is obtained by combining Darcy’s law for the viscous pressure drop in the bulk flow with the average capillary pressure that acts on the different menisci, assuming a constant contact angle between the invading fluid and the inner walls of the porous medium. The solution \( h(t) \) is known as *Washburn’s law*. In the absence of gravity (e.g., for horizontal displacements), this law predicts that the average position of the imbibition front grows as the square root of time, \( h \sim t^{1/2} \), so that the front slows down in time but never stops. When gravity resists capillary invasion, there is a crossover between this behavior at early times and an exponential slowing down at late times that finally brings the front to rest, although theoretically over an infinite time. The average position followed a power law over time, rather than the predicted exponential dynamics, and did not seem to approach an equilibrium height asymptotically (Fig. 5).

Washburn’s law provides a very good description of capillary-driven fluid invasion of simple capillaries. Traditionally it has also been considered as an appropriate description of capillary invasion of disordered media, since it well describes the fast stages of invasion, up to the dramatic slowing down theoretically associated with approaching Jurin’s height.

Several years ago, however, capillary rise in porous media over very long durations was investigated. Using vertical cylindrical columns packed with glass spheres, several groups \[6,12\] showed that Washburn behavior was indeed observed in the initial stages of invasion, which lasted a few minutes. The slow advancement of the front at the end of this Washburn invasion, however, switched to a motion of small-amplitude jumps on the pore scale that continued for hours. The average position followed a power law over time, rather than the predicted exponential dynamics, and did not seem to approach an equilibrium height asymptotically (Fig. 5).

The reasons for these unexpected observations remained elusive until recently, when a new theory of capillary rise in disordered media was proposed. This theory considers the different modes of motion that menisci go through on the pore scale, in the framework of a macroscopic (Darcy scale) description. Three main modes are considered. The first is a *wetting* mode, which describes the motion of the contact line on the pore scale driven by capillary forces, essentially in the...
Washburn regime, with a shape that is practically unchanged but with a velocity-dependent contact angle. The second is a threshold mode, which describes the local pinning of the contact line upon reaching a pore, followed by deformation of the meniscus until the contact angle reaches a critical value at which the contact line can resume its motion. These two modes alone reproduce the essential features of Washburn dynamics, although the final height is attained in finite time and depends only on the threshold value of the contact angle [23]. The third mode, called subcritical depinning, introduces a new mechanism of motion. On the pore scale there are pressure fluctuations, due to the mutual influence of menisci that are in different modes of motion at a given moment. These fluctuations are unimportant when the local pressure is capable of pushing the meniscus through the threshold mode. But when this is not the case the front remains locally pinned, so that the effectiveness of the fluctuations grows over time until, finally, the interface may depin subcritically due to random fluctuations. This mode of motion, responsible for the long-term behavior of capillary invasion, is associated with avalanches of the invading menisci that had been observed experimentally indeed in the long-term regime of capillary rise. The theory, moreover, predicts that capillary rise eventually comes to a halt, albeit at very long times that fall outside the range of currently available measurements [22].

### Kinetic roughening

Interfacial growth driven by competing forces at different length scales is known to result in rough interfaces that exhibit scale-invariant properties. The morphology of the interface looks the same (i.e., it has the same statistical properties) at different magnifications, at least within a wide range. Scale-invariance is ubiquitous in nature (Fig. 6).

Stable imbibition fronts in disordered media are subject to the competing influence of surface tension, viscous pressure drop, capillary pressure fluctuations, and permeability variations. While surface tension and viscous pressure drop keep the front smooth at different length scales, capillary pressure fluctuations and permeability variations distort the front. The result is that an initially smooth front undergoes a kinetic roughening process [1], in which front fluctuations grow over time due to the progressive correlation of different points in the front, with increasing correlation length $\xi \sim t^{\frac{\beta}{z}}$. The root-mean-square fluctuations of the front position grow over time according to $W \sim t^{\frac{\beta}{z}}$, until they saturate. The resulting front is scale-invariant and verifies $W \sim L^{\alpha}$, where $L$ is the lateral size of the system. The exponents $\alpha$, $\beta$, and $z$ are called the roughness, growth, and dynamic exponent, respectively, and they satisfy the scaling relation $\alpha / \beta = z$. Since $\alpha$ is usually a non-integer, the rough front is a self-affine fractal object. This is called Family-Vicsek scaling, the simplest possible scenario of scale-invariant growth. In a pioneering work, Rubio et al. [18] found that it applied to stable imbibition displacements in two-dimensional models of porous media consisting of Hele-Shaw cells packed with glass beads. The actual values of the scaling exponents are important because, following the concept of scale invariance in equilibrium critical phenomena, there are solid arguments to believe that the long-term, large-scale asymptotic behavior of growing interfaces does not depend on the microscopic details of the systems under study but
only on general properties, such as the space dimensionality and the interaction range. Hence a few basic models are sufficient to identify universality classes of kinetic roughening and their corresponding scaling exponents. This framework is well established for interfacial growth problems with local interactions. However, stable imbibition displacements in disordered media are intrinsically non-local: the dynamics of one point of the front depend on all other points, because of mass continuity. Accordingly, in spite of serious theoretical efforts [8–10,15], it is not yet clear to which universality class of kinetic roughening this problem belongs and which scaling exponents define it.

The Family-Vicsek scaling scenario can be extended by considering the way in which front fluctuations scale with the lateral size of the window of observation, i.e., $W \sim \ell^{\alpha}$. If they scale in the same way as they do with the lateral system size $L$, Family-Vicsek scaling is recovered. If not, three additional scaling scenarios (defined by the values of five scaling exponents that verify two scaling relations) are possible [17]. These anomalous scaling scenarios appear in interfacial problems in which the mean local slope of the front diverges in time, thus introducing a new correlation length in the growth direction [14]. A few years ago we showed that tailoring the disorder properties of the medium can lead to stable imbibition displacements with anomalous kinetic roughening [24,25]. The experiments were carried out in a laboratory model of an open fracture, a Hele-Shaw cell with quenched disorder consisting of random dichotomic variations in the gap thickness. When this disorder is persistent in the direction of displacement, it gives rise to rough fronts with very large local slopes and, consequently, to anomalous scaling (Fig. 7).

**Avalanches, non-Gaussian velocity fluctuations and intermittency**

Fluid invasion of porous and fractured media at low velocity (low $\text{Ca}$) is dominated by fluctuations in capillary
Fluids in disordered media

The slow advancement of the front takes place by localized velocity bursts or avalanches. Avalanches represent the dominant mode of motion in the late stages of capillary rise in porous media, as discussed earlier, and they are also at the origin of the scale-invariant properties of invasion fronts.

In slow stable imbibition displacements, avalanches are triggered by capillary pressure fluctuations and suppressed by the interfacial tension of the invading front and by fluid viscosity. In the case of a porous medium modeled by a Hele-Shaw cell packed with glass beads, Dougherty and Carle [7] showed that the areas swept by the avalanches followed an exponential distribution with the characteristic size of the pore spaces. The burst dynamics in porous media are therefore controlled by pore-scale dynamics. This is in striking contrast to the case of an open fracture, where the length scale \( \lambda = \sqrt{\kappa / C_a - (\mu v)^{-2}} \) at which the two damping mechanisms (interfacial tension and fluid viscosity) cross over plays a very relevant role. Considering an initially flat front, the lateral correlation length grows in time until it reaches \( \lambda_c \), and the front then reaches a statistically stationary state of saturated roughness. In this stationary state, the motion of the front is composed of local avalanches with a very wide distribution of lateral sizes, from the lower cutoff imposed by the characteristic size of the disorder to an upper cutoff given by \( \lambda_c \). This upper cutoff may be tuned by controlling the average velocity of the front, \( v \). As \( v \) approaches zero, \( \lambda_c \) diverges and so does the lateral correlation length, revealing that the limit of zero velocity (pinning) corresponds to a nonequilibrium critical point. Near this point relevant quantities...
such as lateral size, area and duration of local velocity bursts should be scale invariant; that is, they will follow power-law probability distributions over a very wide range of values, within the upper and lower bounds imposed by the cutoffs just discussed. An example of the probability distribution of avalanche amplitudes in slow imbibition displacements at constant flow rate, based on our model open fracture with random (non persistent) dichotomic gap thickness [21], is shown in Fig. 8.

Since local velocities of contiguous points in the front are laterally correlated up to a distance \( \ell_c \), additional information about the dynamics of slow imbibition in our model open fracture can be obtained from scale-dependent statistics. The mean velocity of the front within a window of observation of lateral size \( \ell \) can be computed by taking the spatial average of the local velocities over a lateral distance \( \ell \). The statistical distribution of this new quantity, \( V_\ell \), is sensitively dependent on how \( \ell \) compares with \( \ell_c \). When \( \ell \lesssim \ell_c \), the normalized statistical distribution of \( V_\ell \) is heavily skewed towards values above the ensemble average and clearly non-Gaussian (Fig. 9). Remarkably, the experimental distributions are accurately represented by generalized Gumbel distributions of the same skewness, with no other fitting parameters. The origin of this behavior is the fact that, for \( \ell \lesssim \ell_c \), \( V_\ell \) is an average over strongly correlated local velocities. For larger windows of observation or larger capillary numbers, for which \( \ell / \ell_c > 1 \), a Gaussian distribution of \( V_\ell \) is progressively attained as the average involves more and more uncorrelated local velocities, in correspondence with the result of the central limit theorem. The skewness is controlled by the ratio \( \ell / \ell_c \), which can be thought of as counting the effective number of independent degrees of freedom of the invading front [16].

Furthermore, the velocity \( V_\ell \) is intermittent. This refers to the presence of anomalous temporal correlations, such that periods of low velocities and small accelerations alternate with periods of very large velocities and highly fluctuating accelerations (Fig. 10). Intermittency has been a key concept in hydrodynamic turbulence. It also has been found numerically in the Lagrangian velocities of fluid particles flowing through porous media [5].

A characteristic signature of intermittency is the observation of fat-tailed probability distributions of the mean-velocity increments \( \Delta V_\ell (\tau) = V_\ell (\tau + \tau) - V_\ell (\tau) \).

The scale-dependent analysis is now carried out in terms of the spatial scale \( \ell \), introduced earlier, and a new temporal scale \( \tau \). Our results for slow imbibition displacements in laboratory models of open fractures [3] show that the intermittent dynamics of the front are controlled again by the lateral correlation length of the front through the ratio \( \ell / \ell_c \), as before, but also by a new time scale in the direction of invasion, \( \tau_c \), such that intermittency depends only on \( \ell / \ell_c \) and \( \tau / \tau_c \) (Fig. 10). Not surprisingly, experimental results show that \( \tau_c = \ell_c / \nu \), where \( \ell_c \) is the characteristic extent of the disorder in the direction of front advancement. Since both \( \ell_c \) and \( \tau_c \) vanish at high flow rates, intermittency is present only in slow imbibition displacements, dominated by capillary pressure fluctuations.

**Conclusions**

Understanding the complex spatiotemporal dynamics of fluid flows in disordered media is relevant for natural and industrial processes of importance, such as soil irrigation, water filtering, and secondary oil recovery. Progress in this direction is being achieved through the combination...
of hydrodynamic models and analytical tools from non-equilibrium statistical physics.

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References

About the image on the first page of this article. This photograph was made by Prof. Douglas Zook (Boston University) for his book Earth Gazes Back [www.douglaszookphotography.com]. See the article “Reflections: The enduring symbiosis between art and science,” by D. Zook, on pages 249-251 of this issue [http://revistes.iec.cat/index.php/CtS/article/view/142178/141126]. This thematic issue on “Non-equilibrium physics” can be unloaded in ISSUU format and the individual articles can be found in the Institute for Catalan Studies journals’ repository [www.cat-science.cat; http://revistes.iec.cat/contributions].