Simple Stochastic Model of Spontaneous Imbibition in Hele-Shaw Cells

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The imbibition of water in oil in Hele-Shaw cells is described as an asymmetric random walk of water in a globally disordered medium. The disorder is modeled by an external, nonwhite, dichotomic noise, and the resulting random walk is described in terms of an effective Markovian master equation. The solution to this equation is given, and the mean-squared-displacement (MSD) of fluid particles in an effective ordered medium is calculated. This same quantity was also measured in squared cells with a fixed separation, where Soltrol 170 was displaced by twice-distilled water. The flow generated by spontaneous displacement of oil presented three sequential stages, each with a characteristic speed and advancing front structure. In the first and third stages viscous fingering was not detected, but in the intermediate development stage it was observed. From the recorded time evolution of the front, its average displacement speed and the MSD of water in oil as a function of time were determined. Imbibition of water shows an enhanced diffusion regime in the first stage where MSD varies with t^2, *without any external noise effect. This is followed by a simple diffusion behavior in an effective medium in the second stage, where the external noise effect is large. In the third stage Gaussian diffusion prevails again. Numerical regression techniques were used to determine the best values of model parameters that fit experimental values for different stages, so that the sum of squared errors between theoretical predictions and experimental measurements is minimized. Good agreement was found with average errors of 14.3%, 5.9% and 0.6%, respectively.*

Introduction

Disorder and mixing processes occur in many areas of applied science and engineering. They control the efficiency and output of a diversity of physico-chemical phenomena which vary from the mixing in beds of fluidizing or sedimenting particles, the turbulent mixing occurring in jets or reaction chambers, to the dispersion of miscible or immiscible fluids in porous media. This last phenomenon takes place in a variety of engineering processes such as filtration, chromatography, or in the use of porous catalyzers (Bear, 1972; Matteson and Orr, 1977). In all these processes, the properties of the fluid, as well as those of the media, play an important role and determine the interaction media-fluid, which is responsible for the observed transport properties. Although the usual level of description of these processes is macroscopic and is carried out by using a fluid mechanics description, microscopic and mesoscopic approaches have also been used (Mason and Malinauskas, 1987; Scheiddeger, 1974). Here a mesoscopic description will be understood as one given in terms of macroscopic variables, but which comprises both the deterministic laws and the fluctuations about them (van Kampen, 1992).

The main objective of the present work is to adopt a mesoscopic point of view and to introduce a simple stochastic model to analyze and describe some features of the experimental results on spontaneous imbibition that we have carried out in two-dimensional (2-D) Hele-Shaw cells. Spontaneous imbibition is defined as the immiscible displacement process by which a wetting fluid displaces a nonwetting fluid

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that initially saturates a porous or capillary medium, driven by capillary forces only. In contrast, in forced imbibition the wetting fluid is injected into the medium, pushing the nonwetting fluid mainly by a pressure difference. Although both types of imbibition are important in the oil industry, spontaneous imbibition plays an important role for a natural fractured reservoir and in water-wet rock reservoirs that may be visualized as a network of large matrix rock blocks surrounded by fractures. In a water-flooding process, water invades fractures first and then spontaneously penetrates the matrix rock expelling oil towards the fractures. Spontaneous imbibition is a natural water-flooding process that can be taken advantage of for secondary oil recovery technologies. The exchange of water and oil, from and towards fractures, is known as the matrix-fracture interaction, which has been modeled from experimental results (Aronofsky et al., 1958; Kazemi et al., 1976; Kleppe and Morse, 1974). It was observed in our experiments that the flow generated by the spontaneous displacement of oil presented three sequential and physically distinct stages, each having a characteristic speed and advancing front structure (Hayashi and Soria, 2000; Hayashi, 2000). To our knowledge, there are no previous reported observations of the existence of these stages in spontaneous imbibition processes. However, as early as 1941, (Leverett, 1941) among others (Richardson et al., 1952; Kyte and Rapoport, 1958; Huang, 1996) reported the importance of inlet-and-outlet effects in forced displacement experiments in porous rocks.

It is well known that the fluctuations existing in open systems may be conveniently classified into internal and external fluctuations. The former are self-originated in the system, while the latter are determined by the environment. Internal fluctuations are a consequence of the large number of degrees of freedom averaged out in a macroscopic description; they scale with the size of the system and, therefore, vanish in the thermodynamic limit, except at a critical point where long-range order is established. Their study is an important and well-known part of statistical mechanics. In contrast, external fluctuations exist when a system is under the influence of external noise caused by a natural or induced randomness of the environment of the system. These fluctuations play the role of an external field driving the system, and they do not scale with the system size. Thus, if external noise is present in a macroscopic system, it will dominate over internal fluctuations. Among others, physico-chemical systems where the effect of external noise has been considered include fluids (Gollub and Steinman, 1980; Moss and Welland, 1982); lasers and optical systems (Arecci and Politti, 1979; Dixit and Shani, 1983); chemical reactions (De Kepper and Horsthemke, 1979); nuclear reactors (Williams, 1974), and liquid crystals (Rodríguez et al., 1997). In these applications external noise is usually considered as a stochastic process that is introduced into the parameters of the deterministic equations which describe the macroscopic behavior of a system. In any case, the term fluctuations in a state variable or in a system's parameter shall be identified in this work with the deviations from its average value.

In general, only in the last two decades have the methods of statistical mechanics become a major useful approach in chemical engineering (Deem, 1998). In particular, stochastic methods, which include the effects of external noise on engineering systems and on transport properties of imbibition in particular, have been studied less than the above mentioned systems (Tambe et al., 1985a,b,c,d; Fox and Tan, 1990a,b,c; Salinas-Rodríguez et al., 1998). The properties of diffusion in disorder media are usually described through the dependence of the mean-squared displacement (MSD), $\langle l^2 \rangle$ on time. Whereas, for simple diffusion $\langle l^2 \rangle \approx t$, anomalous diffusion is characterized by $\langle l^2 \rangle \approx t^{\epsilon}$, with $\epsilon \neq 1$. This behavior is found, for example, in the chaotic dynamics of turbulence, which generally leads to enhanced diffusion with $\epsilon \simeq 3$ (Monin and Yaglom, 1971, 1975). On the other hand, in systems with geometric constraints, the diffusion is usually dispersive, $\epsilon < 1$ (Scher and Montroll, 1975). Within this general frame, the main purpose here is to analyze the behavior of the MSD of water within the context of our experimental results on the spontaneous imbibition process occurring in 2-D Hele-Shaw cells. Using a unified treatment of internal and external fluctuations introduced by Sancho and San Miguel (1984), we construct a simple stochastic model that considers the process of imbibition as an asymmetric, 1-D random walk of the water particles on a spatially homogeneous lattice with disorder. The disorder is modeled as an external noise in one of the transition probabilities per unit time. The basic idea is to explore the feasibility of replacing the original disordered medium by an effective ordered one, so that the difference between the calculated effective, and the experimentally measured MSD is minimum for each of the observed stages of imbibition. As shown below, in spite of the simplicity of the model, we find good agreement between both methods.

To this end the organization of this article is as follows. The main features are described of the equipment, experimental procedure and materials used to construct Hele-Shaw cells to study the spontaneous imbibition of water in oil as a function of time. During the first and third stages of the experiment, viscous fingering was not observed, but in the intermediate development stage it was indeed present. The procedure to measure the MSD in these stages, the interpretation of data as well as the error analysis, are also discussed in this section. Next, the Markovian master equation (ME) describing the time evolution of the walker is given and the partial differential equation satisfied by the associated generating function (GF) is derived. The disorder in the medium is modeled by introducing a nonwhite, dichotomic external noise into one of the transition probabilities per unit time of the ME, which also appear as parameters in the equation for the GF. This procedure generates a stochastic partial differential equation for the GF. Averaging this equation over the realizations of the external noise source yields an equation for the effective generating function (EGF), which defines a generalized random walk, in an effective ordered medium (Alexander et al., 1981; Bernasconi et al., 1980; Sahimi et al., 1983; Haus and Kehr, 1987). This equation is solved analytically for appropriate initial and boundary conditions and the MSD is calculated as a function of time and the parameters defining the noise. In the last section, the experimental values for the MSD are compared to those predicted by the stochastic model. For each stage, the best fitting values of the noise parameters are determined and the corresponding interpretation is also provided. For the initial stage where viscous fingering is absent, the average error between theory and experiment turned out to be 14.3%; whereas for the developed and final stages, it was 5.9% and 0.6%, respectively, and good agreement for the different stages of the imbibition process was found. The article concludes by further discussing these results, by pointing out the advantages as well as the limitations of our approach, and by making comments on more realistic generalizations of the model.

Experimental Setup

The experimental research on spontaneous imbibition has focused mainly on studying this process in reservoir rock samples. However, in most of the experiments the rock is considered as a black box and the target goal is to get information about the displacement rates and the sweep efficiency (Brownscombe and Dyes, 1952; Bobek et al., 1958; Mattax and Kyte, 1962; Kleppe and Morse, 1974; Zhang et al., 1996). Although more information has been obtained by using gamma-ray absorption (Lefebvre, 1978) and X-ray computerized tomography scanning (Bourbiaux and Kalaydjian, 1990; Ramirez, 1998; Akin et al., 2000), there are several features of these observations that should be improved in order to understand the spontaneous water-flooding mechanisms, which determine that a nonwetting fluid may be displaced or trapped inside the capillary or porous media.

The spontaneous imbibition process visualization in a real porous media presents many difficulties to observe the motion of fluids *in situ*. Nevertheless, simple models on spontaneous imbibition in 2-D capillary media can provide relevant basic information to understand the basic mechanisms of these processes in real-porous media (Kalaydjian and Legait, 1988; Hayashi and Pérez-Rosales, 1992; Hayashi and Soria, 1995). In this work a 2-D experimental model was built to visualize the spontaneous displacement of oil by water. These simple types of physical models are known as Hele-Shaw cells and are made up with two flat transparent glass sheets assembled parallel, with a thin gap between them. Experimental 2-D setups can be a convenient way to study the fundamental mechanisms of the spontaneous imbibition, since it is easy to visualize the water-flooding process. More realistic 3-D experimental setups should include the influence of buoyant forces on the finger evolution through the capillary media, although this feature may be partially observed in vertically placed 2-D Hele-Shaw cells. A cell was built with 0.9 mm thickness glass sheets with dimensions 19 cm $\times$ 20 cm for the upper glass sheet and 30 cm $\times$ 40 cm for the lower one, with a 0.015 cm gap. The cell had an inlet and an outlet on opposite sides of a square, as shown in Figure 1, where the 2-D capillary system is shown to be formed by the space between the glass sheets.

Twice-distilled water was used as displacing fluid and Phillips Petroleum Soltrol 170 was used as the displaced fluid. Since the Soltrol-Water interface is difficult to distinguish by simple sight, oil soluble pigment was used (Red Oil O BDA from Merck) to emphasize the phase difference, in a concentration of 0.2 g/L. The relevant fluid properties were measured at 25°C and the shear viscosities of water and Soltrol were $\mu_w = 1.00 \pm 0.005$ cp and $\mu_o = 3.80 \pm 0.005$ cp, respectively. Interfacial tension between twice-distilled water and colored Soltrol was $\sigma = 25.85 \pm 0.005$ dyn/cm. The densities were $\rho_w = 1.0002 \pm 0.00005$ g/cm^3 for water and $\rho_o = 0.7735$

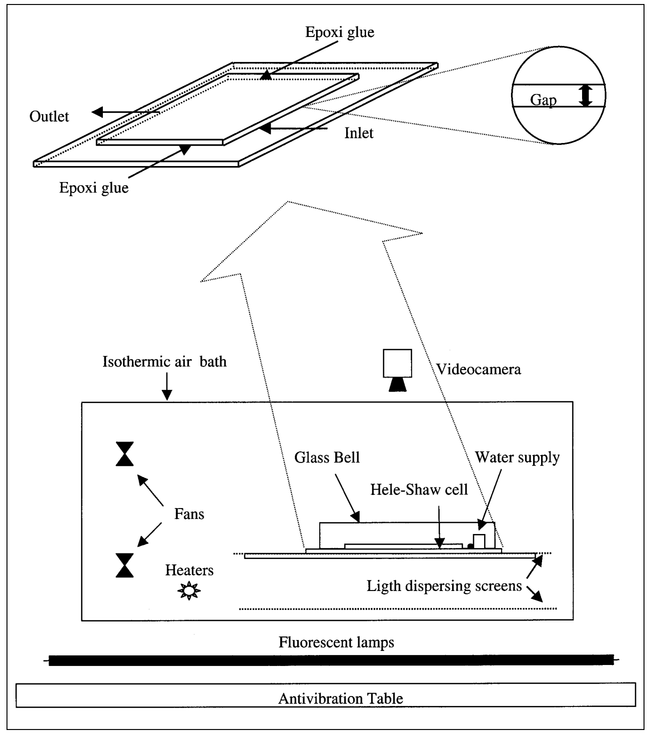

Figure 1. Squared Hele-Shaw cell and experimental setup.

± 0.00005 g/cm^3 for Soltrol. The experiments were conducted at vapor pressure saturation conditions at 25 ± 0.5°C. The experimental setup is also shown in Figure 1.

The capillary space between the glass sheets was initially saturated with colored Soltrol. The cell was set horizontally to avoid gravitational effects and the water supplier was approached to the cell inlet. As the surrounding water reached the cell edge, the water began gradually to invade the capillary space occupied by the Soltrol. At the same time, the oil was expelled through the opposite side of the cell. The displacement of oil by water began at the inlet. The water continuously entered through the inlet, generating an advancing front and expelling an equal oil volume by the outlet. The displacement front was made up by the water-oil interface, whose shape and evolution depended on the relationship between the capillary forces and the viscosities of both fluids. The front initially ran throughout the inlet and evolved in time, leaving in its way fragments of residual oil that remained trapped as islands. These islands could stay fixed, be distorted, grow-up, migrate or even shrink and disappear, depending on the local wetting conditions and on their sizes, shapes, and positions within the cell. The sequential advance of the front is shown in Figure 2. A Panasonic M9000 videocamera with super-VHS format was placed above the cell and the images were captured and digitally processed with the help of a Matrox card and VisioLab 2000 software from Biocom.

At the beginning of the experiment, it was observed that the water-oil interface was uniform-like developing an irregular front advancement up to 60 min, as shown in Figure 2.

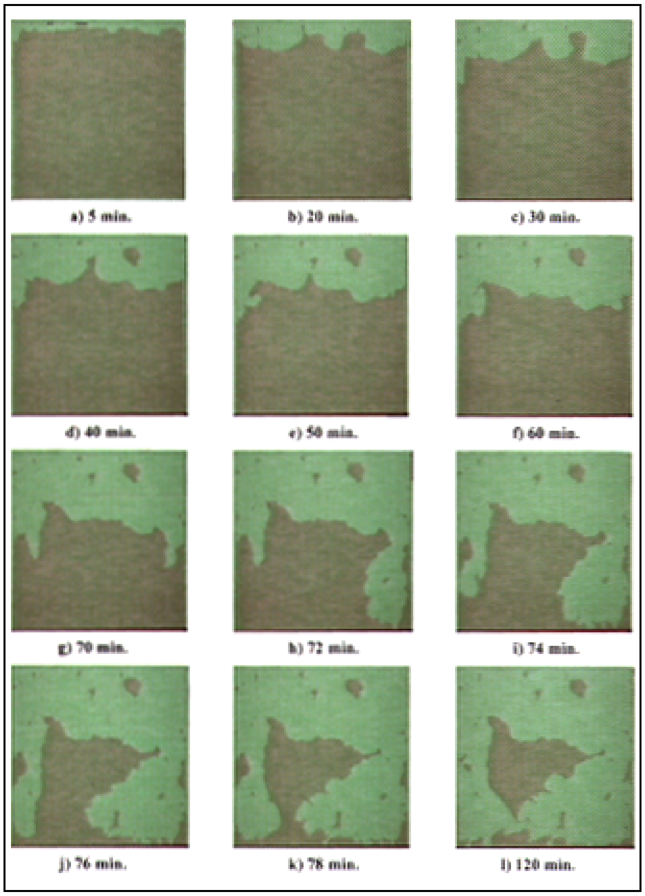

Figure 2. Waterflooding advancement for the spontaneous imbibition process in the Hele-Shaw cell.

During this initial period, the front advancing speed was slow. Once this initial stage was surpassed, the speed increased and the advancement of the front was through viscous fingering.

Experimental Results

The final oil recovered volume is shown in Table 1.

As mentioned above, three characteristic stages for the behavior of the displacement speed were found. The averaged displacement speed $\langle v \rangle$ in each stage was computed from the experiment by using two procedures. In the first one, the average displacement speed for each time interval considered was evaluated according to the relation

$$\langle v_i \rangle = \frac{A_i - A_{i+1}}{(t_{i+1} - t_i)L}, \tag{1}$$

where $\langle v_i \rangle$ is the forward difference average displacement speed at time t_i, A_i is the oil area at time t_i, and L is the inlet length of the cell. Each area A_i was estimated by using the VisioLab 2000 image processing software. $\langle v \rangle^I$ is obtained as the mean value of $\langle v_i \rangle$ over the corresponding time interval. In the first stage from 0–70 min, $\langle v \rangle^I$ remained approximately constant. For the second stage, 70–79 min, $\langle v \rangle^I$ first showed a continuous increase characterized by the occurrence of a water breakthrough with a pronounced acceleration and the sudden appearance of viscous fingering; afterwards, an average speed decay was observed accompanied by fingering lateral evolution towards the center of the cell. Finally, the third stage occurred between 79–120 min, when water reached the outlet and fingering stopped. As water drained out of the cell, the contact between water and the outer edge of the cell was more extensive; some edge sites were in contact with oil up to the end of the experiment, which occurred when no further changes in the spatial configuration were observed. In this stage $\langle v \rangle^I$ became approximately constant once more. The corresponding values of $\langle v \rangle^I$ are given in Table 2.

Table 1. Recovered Oil Volumes

Initial oil volume, cm^3	5.38
Recovered oil volume, cm^3	4.18
Recovered oil volume, %	77.7
Total elapsed time in experiment, min	120

Table 2. Average Displacement Speed $\langle v \rangle$ and Capillary Number C_a for the Different Experimental Stages of Imbibition

Stage, min.	0–70	70–79	79–120
$\langle v \rangle^{I}$, cm/min.	0.111	0.663	0.028
C_a^{I} $(\times 10^{-6})$	2.739	16.24	0.679
$\langle v \rangle^{II}$, cm/min	0.112	0.685	0.018
C_a^{II} $(\times 10^{-6})$	2.748	16.77	0.434

The relationship between viscous and capillary forces was determined by evaluating the capillary number C_a defined as

$$C_a \equiv \frac{\langle v \rangle \mu}{\sigma}. \tag{2}$$

For the different stages the values of C_a^I are also given in Table 2.

A second procedure to get $\langle v \rangle$ and C_a was also developed. This was done by partitioning the cell into parallel stripes of 20 cm × 1 cm. In each section we measure the distance l_i between the inlet and the front position defined by each intersection with the partitioning lines for each observation time t_i. The average value of l_i^2 for all the stripes at the same time yields the mean value $\langle l_i^2 \rangle$. In the future comparison with the theoretical MSD, we use for convenience the notation $\langle l_i^2 \rangle \equiv \langle l^2(t) \rangle$. The plot of the experimental values of $\langle l^2(t) \rangle$ vs. t for the whole process is shown in Figure 3. From the root-mean square (rms) value of $\langle l_i^2 \rangle$, $\langle v \rangle$ is estimated from the relation

$$\langle v \rangle = \sum_i f_i \frac{\sqrt{\langle l_{i+1}^2 \rangle} - \sqrt{\langle l_i^2 \rangle}}{t_{i+1} - t_i}, \tag{3}$$

where the statistical weight f_i of the different time intervals is given by

$$f_i = \frac{t_{i+1} - t_i}{\sum_i (t_{i+1} - t_i)}. \tag{4}$$

This leads to the values of $\langle v \rangle^{II}$ and C_a^{II} given in Table 2, which shows that both estimations yield quite a good agreement.

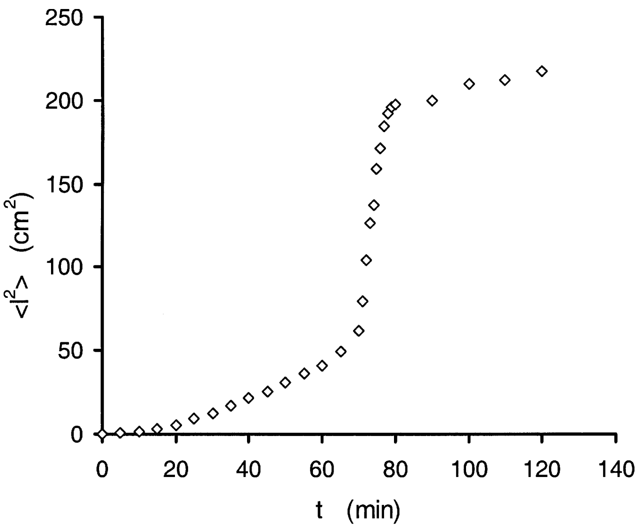

Figure 3. Experimentally determined values of $\langle l^2 \rangle$ vs. t for the whole imbibition process.

Cell volume $V = 5.38$ cm^3.

Model Formulation

New methods to study transport phenomena in porous media have been developed recently. Among these, the method of volume averaging is particularly powerful and has been validated to include complex structures in a porous medium (Whitaker, 1999; Trinh et al., 2000). This continuum approach yields volume-averaged transport equations by phase-averaging the local hydrodynamic variables over an appropriately defined volume. Equations for the deviations of these variables from their intrinsic phase-averaged-value may also be obtained in this way. It is essential to stress that since the state variables are not considered to be stochastic variables in this approach, by performing the intrinsic average, their probability distribution is never needed. It is worth mentioning that the concept of deviation in this context is different from that of "fluctuations" (defined in the Introduction), of the hydrodynamic variables considered in other macroscopic approaches such as fluctuating hydrodynamics (Landau and Lifshitz, 1968).

As already mentioned, we do not follow a continuum approach in this work; instead, we adopt a "mesoscopic" point of view to describe the dynamics of the process of imbibition. More specifically, we shall identify a particle of the displacing fluid (water) with a sufficiently small material volume that does not subdivide when moving through the displaced fluid (oil) (Scheiddeger, 1974). We assume that the interaction between the water particles and the oil occurs only through the capillary forces associated with the imbibition process, and that this interaction generates a spatially homogeneous, asymmetric random walk of the water particles through the oil. It is assumed to be asymmetric, because the experiments show that in the initial stage, only water penetrates into the cell. However, since the oil also flows and changes its configuration, we represent it as a disordered medium with dynamic disorder. Thus, in this model the state variable is the position of the walker (water particle) at time t, which is considered to be a stochastic variable defined through its range of values and its probability density over them. The "fluctuations" of this variable will be identified with the deviations from its average value calculated with the corresponding probability density. Clearly, these "deviations" are different from those mentioned in the average volume method.

The stochastic approach that we use allows us to take advantage of the simple features of the well-studied random-walk type models and their ability to generate relatively simple mathematical expressions for various quantities of interest. Here, the random walk of water is represented by a 1-D, stochastic one-step Markovian process, whose time evolution is described by a Markovian master equation of the general standard form (van Kampen, 1992; Rodríguez et al., 1994)

$$\frac{\partial P(l,t)}{\partial t} = \alpha V P(l-1,t) + \beta V P(l+1,t) - V(\alpha+\beta)P(l,t). \quad (5)$$

Here, $P(l,t) \equiv P(l,t;l_0,t_0)$ denotes the conditional probability of finding the walker at position l at time t, given that the walker started at l_0 at time t_0, if all the steps are of the same (unit) size. V denotes the volume of the Hele-Shaw cell and the transition probabilities per unit time α and β, for an advancing or receding step, respectively, are assumed to be constant. It should be pointed out at this point that in contrast to the continuum methods where the spatial dependence of the state variable is explicitly considered, we have only taken into account here its time dependence. The reason for this simplification is the following. For a spatially inhomogeneous system, the concentration of fluctuations of water particles is, indeed, a local phenomenon and their description requires knowing the mass density at different points in the system. However, if the number density is a continuous random variable, the system would be described by an infinite set of stochastic variables, whose description requires the use of a probability defined in a function space. To keep the model mathematically simple and manageable, we avoid in a first attempt all these spatial complications.

Instead of working with the differential-difference ME, it is easier to use the complete representation provided by the generating function (GF) defined by

$$F(s,t) \equiv \sum_{l=0}^{\infty} s^l P(l,t). \quad (6)$$

$F(s,t)$ contains all the statistical information on the system, since the probability distribution $P(l,t)$ and its moments are obtained through the general relations (Goel and Richter-Dyn, 1974)

$$P(l,t) = \frac{1}{l!}\left[\frac{\partial^l}{\partial s^l}F(s,t)\right]_{s=0} \quad (7)$$

and

$$\langle l^m \rangle \equiv \sum_{l=0}^{\infty} l^m P(l,t) = \left[\left(s\frac{\partial}{\partial s}\right)^m F(s,t)\right]_{s=1}. \quad (8)$$

From Eqs. 5 and 6, it can be readily shown that $F(s,t)$ satisfies the following partial differential equation

$$\frac{\partial F(s,t)}{\partial t} = V\left[\alpha(s-1) + \beta\left(\frac{1}{s}-1\right)\right]F(s,t). \tag{9}$$

External noise

We shall now introduce external noise into Eq. 9 by substituting the fixed constant value of one of the parameters in Eq. 9 by a random variable. For instance, if α is replaced by

$$\alpha = \bar{\alpha} + \zeta(t), \tag{10}$$

where $\bar{\alpha}$ is the mean value of α and $\zeta(t)$ is its fluctuation around the average; then, α is no longer constant and is a time-dependent random variable. Consequently, Eq. 9 becomes now the following stochastic partial differential equation

$$\frac{\partial F(s,t)}{\partial t} = V\left[\bar{\alpha}(s-1) + \beta\left(\frac{1}{s}-1\right) + (s-1)\zeta(t)\right]F(s,t), \tag{11}$$

which defines $F(s,t)$ as a *functional* of $\zeta(t)$. Averaging this equation over the external noise yields an equation for the effective generating function (EGF), defined as

$$\bar{F}(s,t) \equiv \overline{F(s,t)}, \tag{12}$$

where the overbar denotes an average over the realizations of $\zeta(t)$. Clearly, the nature of the resulting equation for $\bar{F}(s,t)$ depends on the specific form of $\zeta(t)$. In order to keep the model mathematically simple and analytic, we shall follow Sancho and San Miguel (1984) and make the rather unrealistic assumption that $\zeta(t)$ is a two-state or dichotomic Markov process. This means that the stochastic variable N is a stepwise constant process which jumps between two discrete values $\pm\Delta$ with equal probability at instants randomly distributed and with a correlation time λ^{-1}. More explicitly, $\zeta(t)$ is defined by the properties

$$\overline{\zeta(t)} = 0, \tag{13}$$

$$\overline{\zeta(t)\zeta(t')} = \Delta^2 e^{-\lambda|t-t'|}. \tag{14}$$

Although it is indeed difficult to appropriately represent a natural random environment by this dichotomic noise, it should be pointed out that this model has the advantage of being simple enough for easy, explicit mathematical manipulation. On the other hand, it may also be produced in the laboratory through a noise generator and applied to real systems (Horsthemke and Lefever, 1983). Note that since $\bar{\alpha}$ and V are positive quantities, the positive character of α imposes the condition $[\bar{\alpha} - \Delta] \geq 0$ on the values of Δ; otherwise, the starting Eq. 5 would be meaningless.

Averaging Eq. 11 over the realizations of $\zeta(t)$ yields the following partial differential equation for the EGF

$$\frac{\partial \bar{F}(s,t)}{\partial t} = V\left[\bar{\alpha}(s-1) + \beta\left(\frac{1}{s}-1\right)\right]\bar{F}(s,t) + V(s-1)F_1(s,t), \tag{15}$$

where we have defined

$$F_1(s,t) \equiv \overline{\zeta(t)F(s,t)}. \tag{16}$$

Since Eq. 15 is not a closed equation for $F(s,t)$ it is necessary to derive an independent equation for $F_1(s,t)$. This is accomplished by using the formula of Shapiro and Loginov (1978)

$$\frac{\partial F_1(s,t)}{\partial t} = -\lambda F_1(s,t) + \overline{\zeta(t)\frac{\partial F(s,t)}{\partial t}}, \tag{17}$$

which shows that the last term on the righthand side should be calculated to get a closed system of equations for $\bar{F}(s,t)$. Substitution of Eq. 11 into this term leads to

$$\overline{\zeta(t)\frac{\partial F(s,t)}{\partial t}} = V\left[\bar{\alpha}(s-1) + \beta\left(\frac{1}{s}-1\right)\right]\overline{\zeta(t)F(s,t)} + V(s-1)\overline{\zeta^2(t)F(s,t)}. \tag{18}$$

The last term on the righthand side $\overline{\zeta^2(t)F(s,t)}$, can be evaluated by using Novikov's theorem (Novikov, 1965). This leads to the result

$$\overline{\zeta^2(t)F(s,t)} = \Delta^2\bar{F}(s,t). \tag{19}$$

When this expression is substituted into Eq. 18 and the resulting equation is in turn inserted into Eq. 17, we arrive at

$$\frac{\partial}{\partial t}F_1(s,t) = \left\{-\lambda + V\left[\bar{\alpha}(s-1) + \beta\left(\frac{1}{s}-1\right)\right]\right\}F_1(s,t) + V\Delta^2(s-1)\bar{F}(s,t). \tag{20}$$

Equations 15 and 20 form a closed system which can be solved with appropriate initial and boundary conditions. We choose the following initial conditions

$$\bar{F}(s,t=0) = s^{l_0}, \tag{21}$$

which amounts to assume

$$\bar{P}(l,t=0) = \delta_{l,l_0}, \tag{22}$$

where δ_{ij} denotes the Kronecker's delta, and

$$F_1(s,t=0) = 0. \tag{23}$$

We shall use a boundary condition that preserves the normalization of $P(l,t)$, namely,

$$\bar{F}(s=1,t)=1, \tag{24}$$

The solution of this system of equations gives for $\bar{F}(s,t)$

$$\bar{F}(s,t)=\frac{s^{l_0}}{2\Lambda}\, e^{\left(a-\frac{\lambda-\Lambda}{2}\right)t}\left[(\lambda+\Lambda)+(\lambda-\Lambda)e^{-\Lambda t}\right] \tag{25}$$

and for $F_1(s,t)$

$$F_1(s,t)=\frac{\lambda^2-\Lambda^2}{4b\Lambda}\, e^{\left(a-\frac{\lambda-\Lambda}{2}\right)t}(1-e^{-\Lambda t}). \tag{26}$$

Here, the following abbreviations have been used

$$\Lambda \equiv (\lambda^2+4\Delta^2 b^2)^{1/2}, \tag{27}$$

$$a\equiv V\left[\bar{\alpha}(s-1)+\beta\left(\frac{1}{s}-1\right)\right] \tag{28}$$

and

$$b\equiv V(s-1).$$

From the above expression for the EGF and Eq. 8, the first two moments of $\bar{P}(l,t)$ are explicitly calculated with the result

$$\langle \bar{l}(t)\rangle = V(\bar{\alpha}-\beta)t \tag{29}$$

and

$$\langle \overline{l^2}(t)\rangle = V\left[\left(2V\Delta^2\lambda^{-1}+\bar{\alpha}+\beta\right)t+V(\bar{\alpha}-\beta)^2t^2\right]. \tag{30}$$

As a consequence, the relative fluctuation χ turns out to be

$$\chi\equiv\frac{\langle\overline{l^2}\rangle-\langle\bar{l}^2\rangle}{\langle\bar{l}^2\rangle}=\frac{1}{(\bar{\alpha}-\beta)^2t}\left(\frac{2\Delta^2}{\lambda}+\frac{\bar{\alpha}+\beta}{V}\right). \tag{31}$$

Note that these results show that in the effective ordered medium, the mean value $\langle\bar{l}(t)\rangle$ is independent of the external noise, whereas $\langle\overline{l^2}(t)\rangle$ and χ have terms that depend on both internal and external fluctuations as well. The external noise contributions depend on the amplitude Δ and correlation time λ^{-1} of the dichotomic noise through the combination $\gamma\equiv\Delta^2\lambda^{-1}$, and remain finite in the thermodynamic limit. On the other hand, the contribution due to internal fluctuations vanishes in this limit, as expected. However, both contributions vanish as $t\to\infty$.

The full probability distribution function $\bar{P}(l,t)$ associated with $\bar{F}(s,t)$ may also be obtained explicitly from the relation

$$\bar{F}(s,t)\equiv\sum_{l=0}^{\infty}s^l\bar{P}(l,t) \tag{32}$$

and by expanding $\bar{F}(s,t)$ in powers of s. To carry out this procedure in detail, let us first consider the special case of pure imbibition $\beta=0$ in the static limit of the external noise defined by $\lambda^{-1}\to\infty$. In this case, from Eq. 25 $\bar{F}(s,t)$ reduces to

$$\bar{F}(s,t)=\frac{s^{l_0}}{2}e^{-V(\bar{\alpha}+\Delta)t}e^{V(\bar{\alpha}-\Delta)st}+e^{-V(\bar{\alpha}-\Delta)t}e^{V(\bar{\alpha}-\Delta)st} \tag{33}$$

and by using the series expansions of an exponential function we get

$$\bar{F}(s,t)=\frac{s^{l_0}}{2}\left\{e^{-V(\bar{\alpha}+\Delta)t}\sum_{n=0}^{\infty}\sum_{l=0}^{\infty}\frac{[V(\bar{\alpha}+\Delta)]^{n+l}t^{n+2l}}{(n+l)!l!}s^n\right.$$
$$\left.+e^{-V(\bar{\alpha}-\Delta)t}\sum_{n=0}^{\infty}\sum_{\nu=0}^{\infty}\frac{[V(\bar{\alpha}-\Delta)]^{n+\nu}t^{n+2\nu}}{(n+\nu)!\nu!}s^n\right\}. \tag{34}$$

If, for simplicity, we assume that $l_0=0$, comparison with Eq. 6 yields

$$\bar{P}(l,t)=\frac{1}{2}\frac{[V(\bar{\alpha}+\Delta)t]^l}{l!}e^{-V(\bar{\alpha}+\Delta)t}$$
$$+\frac{1}{2}\frac{[V(\bar{\alpha}-\Delta)t]^l}{l!}e^{-V(\bar{\alpha}-\Delta)t}. \tag{35}$$

Thus, in the static limit of the noise $\bar{P}(l,t)$ is the sum of two Poisson distributions with mean values $V(\bar{\alpha}+\Delta)t$ and $V(\bar{\alpha}-\Delta)t$, respectively. This feature clearly shows that the previously mentioned condition $[\bar{\alpha}-\Delta]\geq 0$ on the values of Δ, guarantees the positivity of $\bar{P}(l,t)$.

In a similar fashion and for the same static limit, it is possible to derive the expression for the probability distribution $\bar{P}(l,t)$ in the general case when $\beta\neq 0$. Following a similar procedure that leads to Eq. 34, one arrives at

$$\bar{F}(s,t)$$
$$=\frac{s^{l_0}}{2}\left\{e^{-V(\bar{\alpha}+\beta+\Delta)t}\sum_{n=0}^{\infty}\sum_{l=0}^{\infty}\frac{[V(\bar{\alpha}+\Delta)]^{n+l}(V\beta)^l t^{n+2l}}{(n+1)!l!}s^n\right.$$
$$\left.+e^{-V(\bar{\alpha}+\beta-\Delta)t}\sum_{n=0}^{\infty}\sum_{\nu=0}^{\infty}\frac{[V(\bar{\alpha}-\Delta)]^{n+\nu}(V\beta)^\nu t^{n+2\nu}}{(n+\nu)!\nu!}s^n\right\}. \tag{36}$$

If we again assume $l_0=0$ and follow the same procedure that led to Eq. 35, we arrive at the more involved expression

$$\bar{P}(l,t)=\frac{1}{2}\left(\frac{\bar{\alpha}+\Delta}{\beta}\right)^{l/2}e^{-V(\bar{\alpha}+\Delta+\beta)t}I_l\left(2V\sqrt{(\bar{\alpha}+\Delta)\beta t}\right.$$
$$+\frac{1}{2}\left(\frac{\bar{\alpha}-\Delta}{\beta}\right)^{l/2}e^{-V(\bar{\alpha}-\Delta+\beta)t}I_l\left(2\sqrt{(\bar{\alpha}+\Delta)\beta t}\right), \tag{37}$$

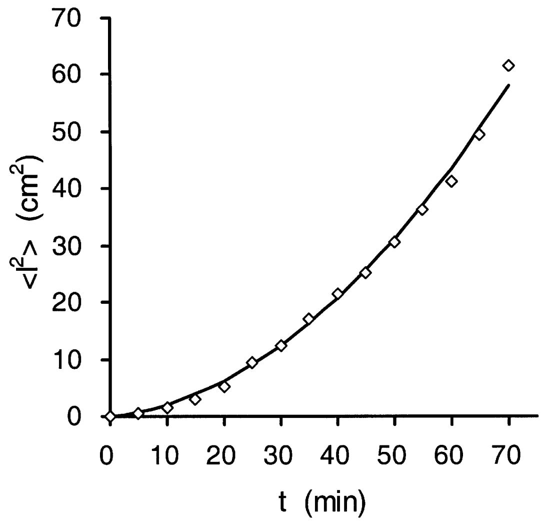

Figure 4. Experimental and theoretical estimations of $\langle l^2 \rangle$ vs. t for the stage 0–70 min.

(◇) Experiment; (—) theory.

where $I_\nu(x)$ is the ν-th modified Bessel function whose infinite series definition is

$$I_\nu(x) \equiv \sum_{s=0}^{\infty} \frac{1}{(s+\nu)!s!}\left(\frac{x}{2}\right)^{2s+\nu}. \tag{38}$$

The corresponding expressions for $\overline{P}(l,t)$ in the nonstatic limit may also be derived from Eq. 36.

Model predictions and discussion

We shall now make a numerical regression analysis in Eq. 30 to determine the best adjusted values of $\overline{\alpha}$, β, and $\gamma \equiv \Delta^2/\lambda$ in each imbibition stage, so that the sum of squared errors between the experimental values and the theoretical predictions is minimized. Carrying out this procedure for the same experimental values of $\langle l^2(t)\rangle$ and times indicated in Figure 3, we find the continuous curves shown in Figures 4–6 for

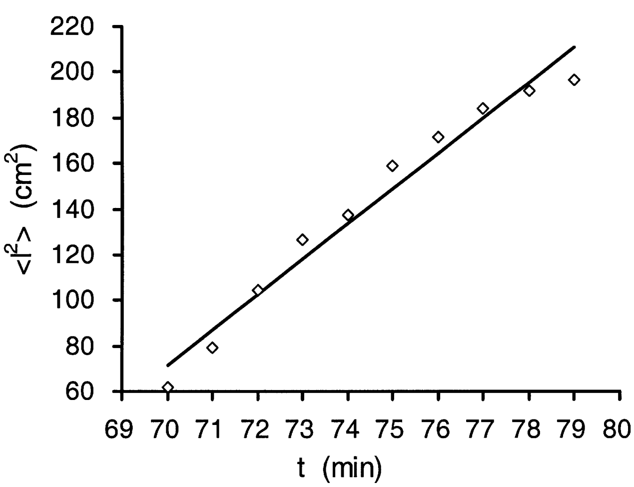

Figure 5. Experimental and theoretical estimations of $\langle l^2 \rangle$ vs. t for the stage 70–79 min.

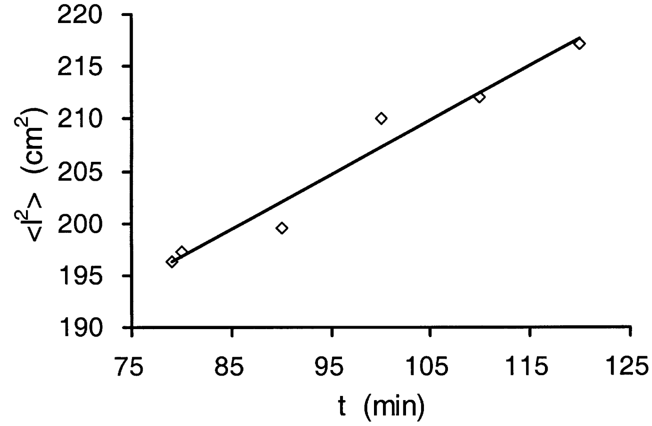

Figure 6. Experimental and theoretical estimations of $\langle l^2 \rangle$ vs. t for the stage 79–120 min.

each stage. From these theoretical estimations of $\langle \overline{l^2(t)} \rangle \equiv \langle l^2(t)\rangle$, we can estimate $\langle v \rangle$ from Eq. 3 with the results indicated in Figures 4–6. As before, the capillary number C_a is determined from Eq. 2. For a cell volume of $V = 5.38$ cm^3, the best fitted values of $\overline{\alpha}$, β and γ, with the corresponding averaged error ϑ, the resulting values $\langle v \rangle_{\text{theo}}$, and $(C_a)_{\text{theo}}$, are given in Table 3.

Let us now physically interpret these results. For instance, during the first stage, where $\overline{\alpha} = 0.019$, $\beta = 0$, imbibition is pure, in the sense that the advancing steps dominate over the receding ones; this is a result consistent with the observations, as shown in Figure 2. The fact that $\gamma = 0$ indicates that in this stage the disorder of the medium (oil) has no effect on imbibition. This would correspond either to an external noise of vanishing amplitude, or with an instantaneous correlation time (white noise), in which case the model is not applicable. To show the nature of diffusion it is convenient to rewrite Eq. 30 in the form $\langle l^2(t)\rangle = A_I t + B_I t^2$, where $A_I \equiv V\overline{\alpha}$, $B_I = A_I^2$, and estimate the relative values of the linear and quadratic contributions for this stage. These values are given in Table 4.

The above values show that for times smaller than 10 min, $\langle l^2(t)\rangle$ varies linearly with time, indicating a linear (Gaussian) diffusion process. However, as time increases, the quadratic term dominates, $\langle l^2(t)\rangle$ varies as t^2, and diffusion is enhanced.

In contrast, for the developed intermediate stage, imbibition is no longer pure since $\beta = 0.056$. The presence of noise is significant, $\gamma = 0.26$, but averaging over the external noise yields an effective ordered medium, where diffusion is normal (Gaussian), since $\overline{\alpha} = \beta$ and the coefficient of the

Table 3. Best Fitted Values of $\overline{\alpha}$, β and γ in Each Imbibition Stage

Stage, min	0–70	70–79	79–120
$\overline{\alpha}$	0.019	0.056	0.017
β	0	0.056	0.017
γ	0	0.26	0.01
ϑ (%)	14.3	5.9	0.6
$\langle v \rangle_{\text{theo}}$, cm/min	0.109	0.675	0.018
$(C_a)_{\text{theo}}(\times 10^{-6})$	2.669	16.540	0.446

Table 4. Relative Values of the Linear and Quadratic Terms in Eq. 30 for the First Imbibition Stage

t (min)	5	10	20	30	45	55	70
$A_I t$	0.5	1	2	3.1	4.6	5.6	7.1
$B_I t^2$	0.3	1	4.2	9.4	21.1	31.5	51
$B_I t/A_I$	0.5	1	2	3.1	4.6	5.6	7.1

quadratic term in Eq. 30 vanishes. This means that, in spite of the fact that at this stage fingering occurs, this complex process as a first approximation could be replaced by normal diffusion in an equivalent medium with a small average error (5.9%).

Finally, diffusion is also linear in the third stage and imbibition is not pure since $\overline{\alpha} = \beta = 0.017$. The effect of noise is much smaller than in the previous stage, $\gamma = 0.01$. Actually, this fact could have been anticipated, since at this stage a large fraction of the oil has already been recovered and, therefore, the effect of noise on water should decrease, as indicated in Table 1.

Concluding Remarks

In summary, experimental results and have been presented and a stochastic model was developed for spontaneous imbibition of water in oil in squared 2-D Hele-Shaw cells. To elaborate on these results and on the aforementioned comparison between theory and experiment, the following comments may be useful.

To begin with, it is important to stress again that our approach is mesoscopic, in the sense that it allows the description of the dynamics of the fluctuations of the stochastic state variables, and not only of the average values. The model is able to reproduce the observed behavior of breakthrough speed, as observed from the quadratic displacement as a function of time. The displacement speed was also measured from the increase in the waterflooding area. The mean values of the displacement speed in all three regions were close to those obtained by the mean quadratic displacement approach. On the other hand, of particular interest is the possible physical interpretation of the nature of the diffusion processes occurring in the different stages of imbibition, arising from the optimized values of the noise parameters. Accordingly, one is allowed to hypothesize that the first stage can be understood as a pure imbibition process, corresponding to an enhanced diffusive mechanism. A pure imbibition process in the context of this 1-D model is understood as due to the motion of the water particles with just forward steps and without the occurrence of backward steps. In the second stage, where the appearance of a fingered front was noticed, the dichotomic noise correlation influence was higher. This stage was characterized by the fastest front advance with a first forward and then lateral finger growth. The model suggests that, in this stage, the complex diffusion process in the real disordered system may be replaced by a linear diffusion in an effective ordered medium. Moreover, the imbibition process at the third stage is surely affected by the near outlet zone and the closer end of the experiment, which happens when the static force equilibrium between viscous and capillary forces is reached or just by the absence of a contact line and the line tension as driving force at the cell outlet. It should be noticed that, at this third stage, the external noise influence is very low. This fact could be due to a smaller contact line length. The model is able to separately fit each one of the three stages, but it is not possible to represent the transitions between them. Therefore, the characterization of the three stages as a first enhanced diffusion mechanism followed by two Gaussian diffusion processes was proposed.

It should be pointed out that the random walk model considered here is idealized in many respects, for example, it is 1-D, asymmetric, spatially homogeneous, and contains dichotomic noise. However, in spite of these simplifications, it serves as a nontrivial example of a highly complex process for which a measurable property, the mean-squared displacement, can be analytically calculated in a rather simple way. The disorder in the oil, which is enhanced by its displacement by water, was modeled as an external noise represented by a two-level jump stochastic process acting on the probability per unit time for an advancing step (α). The use of regression techniques rendered the best estimated values of the walk parameters $\overline{\alpha}$, β and the noise properties $\gamma \equiv \Delta^2/\lambda$ that minimized the average error between the theoretical and measured values of the MSD as given in Figures 4–6. Although a more realistic description should represent the dynamics of imbibition by a more complex and spatially-dependent noise, it is likely that the analytic simplicity of the model will be lost and one has to resort to the use of numerical methods to solve the stochastic equations. In this sense the approach used in this work has been exploratory and indicative, but applies well-established methods to describe well-known phenomena, such as the Brownian motion, to the description of complex transport processes in porous media. The 1-D random walk model considered here may be generalized to 2-D or 3-D when the interaction between the water particles may be neglected.

The essential point to stress, though, is that our approach allowed us to replace a complex imbibition process in a real system, by simpler diffusion processes in an equivalent, but fictitious, medium, with a reasonable small error. Whether this substitution is feasible for other transport properties or processes taking place in a porous medium, remains to be assessed.

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