Memory properties of fluid invasion in disordered media

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Abstract: Fluid invasion through a porous medium exhibits interesting physical properties such as hysteresis and memory. In the ideal case where the invasion is driven quasistatically, and the passage from one equilibrium state to another is considered instantaneous, hysteresis cycles exhibit a property called Return Point Memory (RPM). This paper shows how to take into account the viscous pressure of the fluid and proves that the corresponding hysteresis cycles still exhibit RPM. In addition, a qualitative study of this viscous pressure based on a numerical solution of the balance equation is presented.

I. INTRODUCTION

There are different systems in physics, chemistry and biology that can encode and store information during a dynamic process [1]. This physical phenomenon is known as memory. In most cases this property appears in systems out of equilibrium. This work will focus on a memory-related property called Return Point Memory (RPM).

RPM is directly related to hysteresis phenomena such as those found in condensed matter and materials physics. In some systems, when the order parameter is plotted against the control parameter, a hysteresis cycle arises. This hysteresis cycle is generated by the tendency of the system to retain information from the applied external stimulus. This external stimulus is what we call the control parameter. If the control parameter is varied cyclically, a hysteresis cycle is generated.

When the control parameter is changed between two values, a hysteresis cycle is obtained. If we modify the control parameter appropriately during the hysteresis cycle, we can generate a small hysteresis cycle called internal cycle. This cycle starts and ends in the same state. When the internal cycle ends, the system continues along the main curve following the same states as if the internal cycle had not occurred. These system features are correlated with the RPM.

These memory and hysteresis properties can be modelled in a magnetic system with a random-field Ising model (RFIM) at zero temperature. When an external magnetic field is applied, the system presents a hysteresis cycle with RPM. During the variation of this external magnetic field, it is possible to produce internal cycles [2]. Apart from ferromagnetic systems [3], other interesting systems that show hysteresis with memory properties are deterministic cellular automata [4], charge-density waves [5] [6], and capillary condensation in nanoporous materials [7].

In this paper, we will focus on the study of hysteresis and RPM of two-phase flow in disordered media. In this system, fluids can be imbibed or drained into the porous media. Imbibition takes place when the more wetting fluid displaces the less wetting fluid, and drainage is the opposite situation. As the external pressure on the more wetting fluid is increased or decreased, the porous medium becomes more or less saturated, respectively. This process produces pressure-saturation trajectories that exhibit hysteresis and RPM. The outline of this study will be the following: In Sec. II we will present the experimental system used to model fluid invasion in porous media, and the physics behind this system. In Sec. III a physical model for pressure-driven quasistatic displacements introduced in Ref. [8] will be explained. The existence of the RPM in the ideal case in which the relaxation of the fluid front between metastable equilibrium states is considered instantaneous will also be demonstrated, following the aforementioned paper. Finally, in Sec IV, knowing all of the above and adding a viscous term to the balance equation, we will demonstrate theoretically that the system still exhibits RPM in the case where a nonzero relaxation time is at play. We define the relaxation time as the characteristic time that must elapse to increase the fluid saturation by a factor $(1 - 1/e)$. In addition, we will do a qualitative study of the viscous pressure solving the balance equation with a Runge-Kutta method.

II. HELE-SHAW CELL AND INTERFACE MODEL

The hysteresis properties and the RPM of two-phase flow in disordered media are studied using an "imperfect" Hele-Shaw cell [9]. This cell consists of two plates separated by a distance b_0 as illustrated in Fig. 1. The upper glass plate confines the fluid between it and the lower plate. The lower plate is filled with randomly distributed obstacles of height δb. This disordered Hele-Shaw cell will be imbibed and drained, quasistatically, with a viscous fluid (in our case a silicone oil) stored in a reservoir of height H.

Initially, the cell will be filled with air at atmospheric pressure. Subsequently, the oil will be injected from the tank creating an oil-air interface. The interface will travel through the interior of the cell encountering various ob-

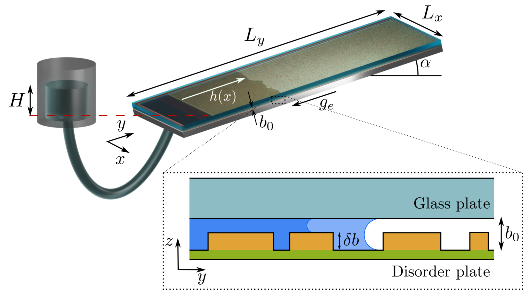

FIG. 1: Design of the Hele-Shaw cell used to study hysteresis and RPM of two-phase flow in disordered media. The inset shows the interior of the cell. Inside the cell, we can see the opening through which the oil will flow and the obstacles distributed throughout the cell. *Reprinted from Ref. [10].*

stacles. Each time the interface encounters an obstacle, there will be a change of capillary pressure at the interface in that point. This change of capillary pressure originates from a change in the curvature of the interface, and results in an increase in the pressure difference between the oil and the air, given by the Young-Laplace equation. The increase in pressure induces a local increase of the interface velocity over the obstacle. The oil-air interface will show the irregularities shown in Fig. 1. For each value of x, we will have a different interface height $h(x)$ because each point of the interface will have passed through a different number of obstacles. If the Hele-Shaw cell were unobstructed, all points in the interface would have the same velocity at all times and the interface would be straight. It is interesting to notice that the cell is tilted at an angle α to induce an effective gravity. This effective gravity is necessary to avoid viscous fingering instabilities in drainage.

As explained in detail in Ref. [8], we can model the system using a pressure equilibrium equation. This equation will take into account the pressure at which the oil is imbibed or drained, $\rho g H$, the gravitational pressure due to the inclination of the cell, $\rho g \sin\alpha \ h(x)$, and the pressure due to capillary contributions at the interface.

As discussed before, the capillary pressure contributions are given by the Young-Laplace equation. This equation gives the pressure jump across a curved interface:

$$[\![p]\!] = \gamma\kappa = \gamma(\kappa_\perp + \kappa_\parallel). \quad (1)$$

As we can see in Eq. (1), the pressure jump across the interface will depend on the surface tension γ between the fluids and on a parameter κ that refers to the curvature of the interface. To study the curvature term, we separate κ into a perpendicular ($\kappa_\perp$) and a parallel ($\kappa_\parallel$) component. The $\kappa_\perp$ component refers to the curvature of the oil-air meniscus in the $y-z$ plane and the $\kappa_\parallel$ component to the curvature in the $x-y$ plane. Taking into account the linear approximation $|dh/dx| < 1$, we have that $\kappa_\parallel \approx d^2h/dx^2$. For the perpendicular term, taking into account the geometry of the meniscus, we know that $\kappa_\perp = 2\cos\theta/b(x,y)$ [11]. The $\kappa_\perp$ term introduces an increase in pressure (which translates into an increase in velocity) at the point where the interface meets an obstacle. The $\kappa_\parallel$ term introduces an additional pressure that depends on its neighbouring points.

It is important to note that we need to imbibe and drain quasistatically so that the displacement goes through the metastable equilibrium states represented by the model. The equation of mechanical equilibrium is as follows:

$$\gamma\frac{d^2h(x)}{dx^2} - \rho g \sin\alpha \ h(x) + \rho g H + p_c[x, h(x)] = 0, \quad (2)$$

where $p_c[x, h(x)] = 2\gamma\cos\theta/b(x,y)$ and g the Earth's gravitational acceleration. The expression $b(x,y)$ is equal to b_0 when the interface is not over an obstacle at that point and is equal to $b_0 - \delta b$ when the interface is over an obstacle. The terms $\gamma d^2h(x)/dx^2$ and $p_c[x, h(x)]$ are the pressure terms of capillary origin, which refer to the curvature of the system and are derived from the Young-Laplace equation.

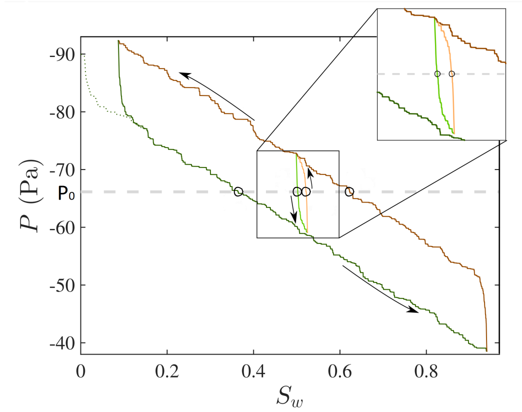

FIG. 2: Representation of the external pressure as a function of the wetting-phase saturation of the disordered Hele-Shaw cell. The main hysteresis cycle can be seen together with a small internal cycle. Four possible configurations of the system for a given pressure P_0 have been shown in the graph. *Adapted from Ref. [8]*

By increasing or decreasing the height H of the reservoir, the oil will be imbibed or drained into the cell, respectively. During this imbibition and draining process, the pressure-saturation curves show a hysteresis cycle and RPM (Fig. 2). It is important to notice that, if we create an internal cycle, the start and end state will be the same. Moreover, when the internal cycle is completed, the system follows the same trajectory that it would have followed if the internal cycle had not been created. It is interesting to notice also that, for the same given pressure, the system can present an infinite number of different configurations, since one can make as many internal cycles as one wishes. The curious thing is that, even with an infinite number of available states, the system knows which trajectory is following in the pressure-saturation curves and always remembers the configuration it has to take every time. This phenomenon is due to the existence of the RPM.

III. DEMONSTRATION OF RPM WITH ZERO RELAXATION TIME

We will demonstrate the existence of RPM in quasistatically-driven two-phase flow in disordered media, following Ref. [8]. First of all, a system that presents RPM must satisfy three properties: the dynamics of the front must be deterministic, rate-independent, and must obey the no-passing rule (NPR) [5].

Firstly, the system must be deterministic because it must be possible to predict the state that will be acquired after a particular driving protocol. This property is fulfilled because, for a given realization of the disorder, the defects in the Hele-Shaw cell are located in the same position and have a macroscopic size. Therefore, the system will not be subjected to external fluctuations, thus ignoring stochastic effects. Secondly, the dynamics of the interface is rate-independent as long as the external pressure varies quasistatically. Finally, the NPR holds as well, as proved next.

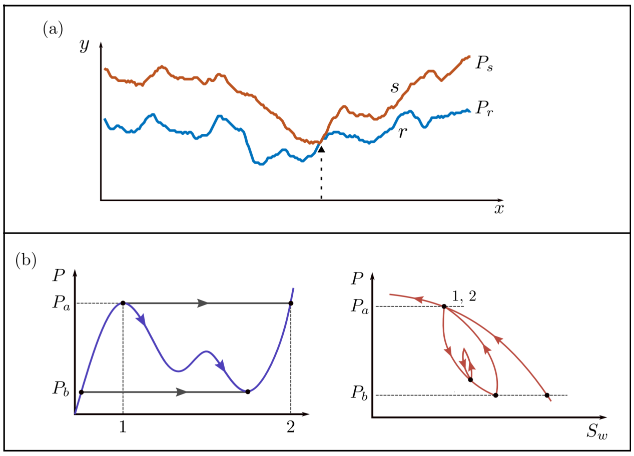

FIG. 3: (a) Representation of two fronts in a Hele-Shaw cell, where the y-axis represents the height of the cell and the x-axis its width. (b) The left diagram represents the temporal protocols followed by the external pressure (in a.u.) to carry out the cycles. The right diagram represents the applied external pressure (in a.u.) as a function of the cell saturation (in a.u.). *Adapted from Ref. [8].*

We will assume that the relaxation time of the fluid after a variation of the external pressure is negligible. Strictly speaking, this would be true only if the invading fluid was inviscid. A reductio ad absurdum demonstration will be used to prove the NPR. We start by defining the parameter p_e:

$$p_e(x) = \gamma \frac{d^2 h(x)}{dx^2} - \rho g \sin\alpha \; h(x) + P + p_c[x, h(x)]. \quad (3)$$

The parameter $p_e(x)$ represents the net external pressure applied in each point of the interface. If $p_e(x) > 0$ the oil will be imbibed, if $p_e(x) < 0$ the fluid will be drained, and if $p_e(x) = 0$ the fluid will remain in mechanical equilibrium at that point x.

Let us suppose that there are two different and independent fronts: $r(x)$ and $s(x)$. Furthermore, $r(x)$ is below $s(x)$, and their external driving pressures satisfy that $P_r \leq P_s$. The NPR says that the front $r(x)$ can never reach $s(x)$, i.e. $r(x) < s(x)$. To prove that the NPR is satisfied in our system, we proceed by assuming the opposite, i.e. that $r(x) = s(x)$ at some point x, as shown in Fig. 3a. When the overtaking takes place at x, $\gamma d^2 r(x)/dx^2 \leq \gamma d^2 s(x)/dx^2$. This condition applies because the front $r(x)$ must have a lesser curvature than $s(x)$ around the point x. Furthermore, $P_r \leq P_s$ by assumption. Bearing in mind that we are studying a point where $s(x) = r(x)$, we can affirm that we will have $p_c[x, r(x)] = p_c[x, s(x)]$. Also, $\rho g \sin\alpha \; r(x) = \rho g \sin\alpha \; s(x)$ because both interfaces will have the same effective gravity due to the inclination of the cell. It is essential to note that overtaking can only occur if $p_e^r(x) > p_e^s(x)$. But we show next that this condition can never occur. Starting from the overtaking condition we have that:

$$p_e^r(x) > p_e^s(x) \quad \Rightarrow \quad \gamma \frac{d^2 r(x)}{dx^2} + P_r > \gamma \frac{d^2 s(x)}{dx^2} + P_s.$$

But $\gamma d^2 r(x)/dx^2 \leq \gamma d^2 s(x)/dx^2$ and $P_r \leq P_s$, so that

$$p_e^r(x) \leq p_e^s(x).$$

This contradiction proves that overtaking can never occur. This proves the existence of NPR in quasistatically driven two-phase flow in disordered media for inviscid fluids (negligible relaxation time).

Let us imagine now that we want to move the interface from a pressure P_b to a pressure P_a. We will apply three different pressure protocols illustrated in the left figure in Fig. 3b. The first protocol will consist of maintaining the pressure in P_b and increasing the pressure at the end until reaching P_a. The second protocol will consist of increasing the pressure at the beginning to P_a and staying at that pressure. The third protocol shall consist of a bounded non-monotonic trajectory between P_a and P_b. If we perform this bounded pressure variation protocol between P_a and P_b the fact that the system satisfies the NPR, ensures that the configuration of the interface at P_a will be the same in the three protocols. Taking into account this and knowing that the system is deterministic and rate-independent, we can say with certainty that the system will present RPM. It is interesting to note that if NPR would not be fulfilled, we could not state that the final configuration of the interface was independent of the pressure protocol used. The right figure in Fig. 3b shows the pressure-saturation curves of the pressure protocols used, taking into account that the NPR holds.

In summary, we have demonstrated that the NPR, together with the fact that the system is deterministic and rate-independent, implies that it verifies the RPM property.

IV. DEMONSTRATION OF RPM TAKING INTO ACCOUNT THE VISCOUS RELAXATION TIME

Our objective now is to demonstrate the existence of RPM in a more realistic case where the viscosity of the fluid is taken into account. First of all, a time-dependent term must be added to the balance equation that will characterise the relaxation of the fluid after a change in applied presure to its new equilibrium configuration. This term is derived from the Navier-Stokes equations. Working in the Stokes limit, where inertial effects are negligible compared to viscous effects, and taking into account that, for the most of the motion of the fluid is in the $x-y$ plane, the gap-averaged 2D velocity field in the $x-y$ plane of the cell is given by:

$$\vec{v} = -\frac{\kappa}{\mu}\vec{\nabla}p,$$

where κ is the permeability of the cell and μ is the dynamic viscosity of the fluid. To introduce this expression in the pressure balance equation, we assume that the pressure gradient has its dominant component in the y-axis direction, where $\partial p/\partial y = \Delta p/h$. The balance equation with the viscous pressure term will be then:

$$\gamma\frac{\partial^2 h}{\partial x^2} - \rho g \sin\alpha\ h + \rho g H + p_c[x,h] - \frac{\partial h}{\partial t}\frac{\mu}{\kappa}h \simeq 0. \quad (4)$$

This equation takes into account the viscous relaxation of the front velocity. The relaxation will depend on the fluid velocity $\partial h/\partial t$, the interface height h, the cell permeability κ, and the fluid dynamic viscosity μ. It is important to notice that now the fluid height depends on the position x and the time t.

To prove the RPM property in this case, let us suppose again that we have the fronts $r(x,t)$ and $s(x,t)$. If (under driving pressures $P_r \leq P_s$) we assume that we have a point where both interfaces are equal ($r(x,t) = s(x,t)$), the NPR will have been violated. From here on, we will prove that this does not happen.

For an overtaking to occur,

$$p_e^r(x) > p_e^s(x).$$

In this case we have the same conditions than in Sec. III: $p_c[x, r(x,t)] = p_c[x, s(x,t)]$ and $\rho g \sin\alpha\ r(x,t) = \rho g \sin\alpha\ s(x,t)$. Applying these conditions the previous inequality implies:

$$\gamma\frac{\partial^2 r}{\partial x^2} + P_r - \frac{\partial r}{\partial t}\frac{\mu}{\kappa}r > \gamma\frac{\partial^2 s}{\partial x^2} + P_s - \frac{\partial s}{\partial t}\frac{\mu}{\kappa}s.$$

Furthermore, $\gamma\partial^2 r/\partial x^2 \leq \gamma\partial^2 s/\partial x^2$, $P_r \leq P_s$, so that

$$-\frac{\partial r}{\partial t}\frac{\mu}{\kappa}r > -\frac{\partial s}{\partial t}\frac{\mu}{\kappa}s.$$

Taking into account that $\mu s/\kappa = \mu r/\kappa$ at point x, we can say that:

$$\frac{\partial s}{\partial t} > \frac{\partial r}{\partial t}. \quad (5)$$

Based on the hypothesis that the interface $r(x,t)$ has overtaken the $s(x,t)$, we achieve the expression (5). This generates a contradiction because the velocity of $s(x,t)$ would be greater than $r(x,t)$, and an overtaking could not occur. It is interesting to note also that if we applied the condition that the velocity of $r(x,t)$ was greater than that of $s(x,t)$, i.e. $\partial r/\partial t > \partial s/\partial t$, in order to impose an overtaking, we would obtain that $p_e^r(x,t) < p_e^s(x,t)$ and we would be falling into another contradiction. In conclusion, the validity of the NPR has been demonstrated also when the viscous pressure of the fluid is taken into account. Moreover, since the system is also deterministic and rate-independent, we can state that it presents RPM.

In order to understand the importance of viscous pressure, the nonlinear pressure balance equation (4) has been solved using a Runge-Kutta method programmed in Fortran. To keep things simple, we did not take into account the capillary terms (flat interface case). The parameters used to solve Eq. (4) numerically are used in Ref. [14] for laboratory experiments. Using these realistic parameters and applying correctly the initial conditions, the results shown in Fig. 4 are obtained. The equation has been solved for a given initial height $h_0 = 0.25$ m and with different angles. In order to move the interface forward, the reservoir has been raised by only $\Delta H = 0.1$ mm. In Fig. 4 we can see that the relaxation time of the fluid decreases with increasing cell inclination. This result makes sense since the more inclination the cell has more effective gravity and it is easier to slow down the fluid. During the relaxation, two different regimes can be distinguished [13]. At short times the viscous term dominates and $h \sim \sqrt{t}$ (Washburn regime). At long times the gravitational term starts to gain importance and the

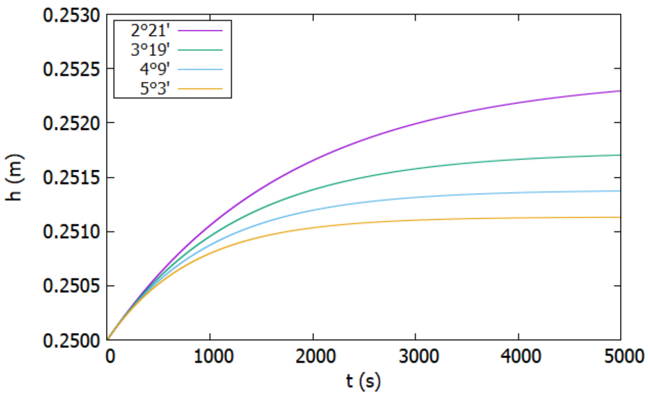

FIG. 4: Diagram of the interface relaxation in the case of a defect-free Hele-Shaw cell for different angles and with the same initial height. The x-axis shows the elapsed time and the y-axis the height h acquired by the interface.

fluid height tends to saturate. The relaxation of the fluid at long times has a negative exponential behaviour [14]. It is important to note that, for the angles studied, in order for the fluid to reach the new equilibrium height we would have to wait between 3000 and 5000 s (of the order of one hour!). The steeper the Hele-Shaw cell is inclined, the faster stability will be reached.

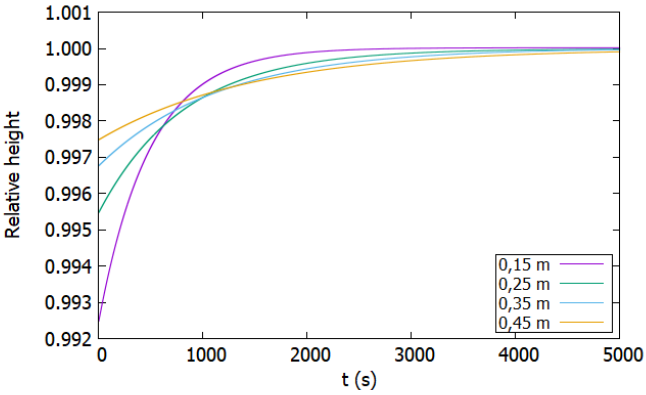

FIG. 5: Diagram showing the relaxation of the fluid at different heights and for a fixed angle ($\alpha = 5°3'$). The x-axis represents the elapsed time and the y-axis represents h/h_{sat}, where h_{sat} is the theoretical height that the fluid should reach to stabilise.

In addition, the model predicts that a higher interface will take more time to reach the equilibrium height. This makes sense because a higher interface will have more oil column under it and it will be harder for gravity to stabilise the fluid. As can be seen in Fig. 5, the numerical simulation confirms this hypothesis. For a given angle, as the initial fluid height is increased, the fluid takes a longer time to stabilise.

V. CONCLUSION

We have studied the hysteresis and memory properties of a two-phase flow in a disordered medium (an “imperfect” Hele-Shaw cell [9]). Following Ref. [8], we have shown that, in the case where the viscous pressure of the fluid is not taken into account, the system presents RPM. Furthermore, we have extended this result for the first time to viscous fluids, and we have proved that the system still exhibits RPM. From the numerical solutions of fluid relaxation in a defect-free cell, we have seen that the instantaneous relaxation approximation is very crude. For a viscous fluid (about 50 times more viscous than water), the times that must be waited to reach the equilibrium height for the angles studied are around one hour approximately. Due to this, we conclude that the viscous pressure is an important factor to take into account to reproduce the experimentally measured pressure-saturation hysteresis cycles.

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