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Capillary displacement of viscous liquids in a multi-layered porous medium

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Capillary driven displacement of viscous liquids into a porous matrix plays a significant role in several porous media applications such as fractured oil reservoirs and paper micro-fluidic devices. The inherent heterogeneity in porous media is known to cause the invading fluid front to lead in narrow pores followed by large pores during spontaneous imbibition. Here, we use experiments in a layered porous medium to show that the leading front is not always in the narrow pores. We observe in a two layered porous medium that the fluid front always advances faster in the narrow pores. However, in a multi-layered porous medium, the front displacement is strongly dependent on the arrangement of the layers, the relative difference between the capillary pressure, the permeability of the layers, and the viscosity ratio of the wetting and the non-wetting fluids. We also develop a one-dimensional lubrication approximation model based on the experimental observations, which predicts the imbibition dynamics in the layers seen in the experiments. Additionally, from our model, we present a scaling law governing the leading front in the porous layers. We also predict using our model that the volume imbibed in the layered porous medium cannot be determined by using the effective porous medium properties and a detailed knowledge of the characteristics of the layers is required to accurately predict the overall imbibition of the layered porous medium.

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1 Introduction

Capillary imbibition is a widely observed phenomenon which uses the surface tension force to drive a viscous fluid through narrow pore spaces. The displacement of a viscous non-wetting liquid by a viscous wetting liquid is important for several applications such as oil recovery from a fractured reservoir^{1–5} and paper micro-fluidics.⁶ One of the earliest studies on capillarity by Lucas⁷ and Washburn⁸ in a cylindrical capillary filled with air shows that impregnation of a wetting fluid follows diffusive behavior, *i.e.*, the distance invaded by the wetting fluid is proportional to the square root of time. The Lucas–Washburn law is obtained from the Hagen–Poiseuille’s law⁹ and is expressed as

$$x^2 = \left(\frac{2KPc}{\mu} \right) t, \quad (1)$$

where $K = \frac{r^2}{8}$ is the permeability for a cylindrical capillary of radius r , $Pc = \frac{2\gamma \cos \theta}{r}$ is the capillary pressure across the meniscus, γ is the surface tension and μ is the viscosity of the wetting fluid, θ is the contact angle of the invading fluid with

the capillary walls, and x is the distance invaded by the imbibing fluid in time t . It should be noted that the distance imbibed by the invading fluid in a capillary of larger radius is more than the distance imbibed in a small radius capillary, as $KPc \propto r$. This is due to the higher viscous resistance offered by the small radius capillary. The deviation from the classical Lucas–Washburn law has also been investigated for non-uniform pore geometries^{10,11} and in capillary transport of polymers.^{12–14}

In a homogenous porous medium also, the capillary driven invasion of a wetting fluid is known to follow the diffusive Lucas–Washburn law.^{15–18} Therefore, a porous medium is often treated as a bundle of independent capillaries,^{19,20} for simplicity. Each capillary in the tube bundle models follows diffusive dynamics and hence the fluid front in the largest radius tube leads. However, the imbibition dynamics observed in real porous media is rather complex and the bundle of non-interacting capillaries is unable to explain the phenomena sufficiently. A diffused imbibition front is observed in porous media, for example, the capillary suction into paper napkins, towels, diapers and fractured oil reservoirs.^{21–24} Bico and Quéré²⁵ described that the imbibing fluid leads in the narrow pores of a porous medium, unlike the tube bundle model.

The diffused fluid front in a porous medium is due to the heterogeneity of the pore sizes, which causes the pores to have different capillary pressure and permeability.^{25,26} Small and large scale heterogeneities in the form of layers within the

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porous medium also lead to a diffused imbibition front. For example, oil reservoirs are made of micro scale layers called laminae to meter scale layers of sediments.²⁷ Similarly, soil comprises interacting porous layers called soil horizons.²⁸ In addition, micro scale layers are also used in polymer electrolyte membrane fuel cells,²⁹ micro reactors^{30,31} and paper based analytic devices.^{32,33} Imbibition in such layered systems is complex and, therefore, a clear understanding of the invasion dynamics in a layered porous medium is important.

Reyssat *et al.*³⁴ explained the imbibition dynamics in detail for a layered porous medium having a permeability gradient in the direction of flow. However, a sharp front was considered in their study. Bico and Quéré²⁵ were the first to describe that spontaneous invasion of a wetting fluid due to capillarity in a porous medium has two fronts, a leading front in the narrow pores and a trailing front, which saturates the porous medium. Bico and Quéré²⁵ explained that the cross-flow or fluid transfer from the saturating large pores to the finer pores having a greater capillary pressure is responsible for the invading liquid to lead in the fine pores, contrary to the observation in non-interacting pores. Ashraf *et al.*²⁶ gave a Lucas–Washburn like solution for a layered porous medium, having a permeability gradient perpendicular to the direction of flow. They observed that for some cases, the invading fluid front will not lead in the narrowest pores as proposed by Bico and Quéré.^{25,26} Ashraf *et al.*²⁶ also gave a scaling law governing this anomalous behavior.

However, in these studies on spontaneous imbibition, a non-viscous resident fluid is considered. Hultmark *et al.*³⁵ experimentally showed, in a cylindrical capillary tube, that viscous effects due to the resident phase viscosity are significant. Moreover, in most of the invasion phenomena, the wetting phase spontaneously displaces a viscous non-wetting phase in a porous medium.^{23,24} Therefore, considering the viscous resistance of the resident fluid is crucial in obtaining an accurate solution to the impregnation behavior in a layered porous medium.

In this work, we use experiments to investigate spontaneous displacement of a viscous fluid due to capillary suction in a layered porous medium. We first perform experiments in two and three layered porous media and track the fronts of the spontaneous imbibition in all the layers. We also develop a quasi one-dimensional lubrication approximation model to predict the wetting fluid invasion in the layers. Our one-dimensional model shows excellent agreement with the experimental observations. In the layered porous medium, we observe using experiments and our model that the leading front is strongly affected by the arrangement of the layers. The front displacement in the layers is governed by the permeability and capillary pressure contrasts between the layers and their arrangement. Using the experiments and model, we give a scaling law using permeability and capillary pressure, which can predict the invasion pattern and the leading front layer. The model developed in this study can be used to design porous media for desired flow behavior, or to predict the flow behavior in naturally occurring layered porous media such as oil reservoirs.

2 Experimental setup

We have performed experiments to visualize the flow behavior in hydrodynamically interacting two and three layered porous media. We use an experimental setup made of parallel acrylic sheets which are 0.15 cm apart, held in place by nuts and bolts as shown in Fig. 1. The inlet and the outlet ends of the porous medium are sealed by a thin layer of paper with negligible resistance. Layers of glass beads are filled between the acrylic sheets through section AB in Fig. 1, with regular tapping to ensure proper packing of the beads. Similar experimental setups were used in previous studies.^{36–38} The open end of the setup is sealed with another acrylic sheet and is made leak proof to avoid spillage of resident and invading fluids. The dimensions of the porous medium are 10 cm × 1.8 cm × 0.15 cm. For the two layered porous medium, each layer is 0.9 cm wide, whereas, for the three layered porous medium, the width of each layer is 0.6 cm. The thickness of the porous layers is 0.15 cm. It can be observed from Fig. 1 that the imbibing fluid flows along the X-axis and the permeability varies along the Y-axis. Gravity acts along the small height of the porous layers (0.15 cm) in the negative Z direction. The Bond number, which is the ratio of the gravitational to the surface tension forces, is of the order of 0.01, indicating that the effect of gravity can be neglected.

Beads of diameters $d_l = 274 \pm 24 \mu\text{m}$ and $d_s = 50 \pm 25 \mu\text{m}$ are used in this study to understand the imbibition phenomena in a two layered porous medium, while, for a three layered porous medium, the bead diameters used are $d_l = 274 \pm 24 \mu\text{m}$, $d_m = 125 \pm 25 \mu\text{m}$ and $d_s = 50 \pm 25 \mu\text{m}$, where the suffixes l, m, and s stand for large, medium, and small, respectively. The capillary pressure for beads of diameter d_i is $P_{c_i} = \frac{c_i \gamma}{d_i}$, where $3 < c_i < 4.5$, according to Reyssat *et al.*³⁴ In this study, similar sizes of bead diameters are chosen in performing experiments on two and three layered porous media, therefore, the value of c_i is considered to be constant for a given bead diameter across

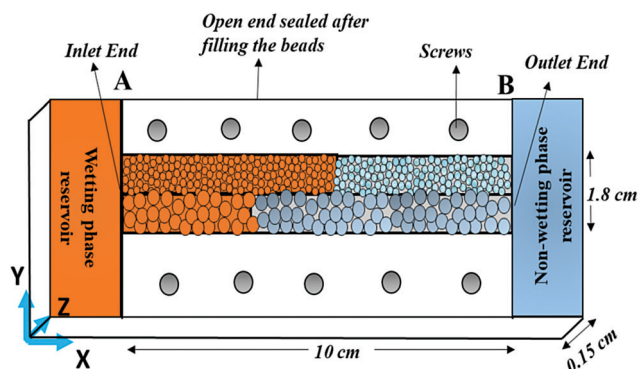


Fig. 1 Schematic of the experimental setup for spontaneous imbibition in a layered porous medium. The dimensions of the porous medium are 10 cm × 1.8 cm × 0.15 cm. The right end of the porous medium is in contact with the non-wetting phase reservoir and the wetting reservoir is on the left side of the porous medium. The two parallel acrylic sheets between which the porous layers are filled are made leak proof by sealing with another acrylic sheet using screws.

all experiments. The permeability of each layer is given by the Kozeny–Carman equation,³⁹ $K_i = \Lambda d_i^2$, where K_i is the permeability of a porous medium made of grains of diameter d_i and Λ is of order 0.0001. The porosity of all the layers is measured and is approximately 0.35.

An LED light source is placed below the porous medium to ensure uniform light intensity in each of the layers. In all the experiments, the porous medium is initially filled with dodecane of viscosity 1.35 cP, a non-wetting resident phase. For spontaneous imbibition to occur, the invading fluid should always be preferentially wetting compared to the resident fluid. The producing end of the porous medium is in contact with the non-wetting phase reservoir to avoid capillary back pressure while the other end is in contact with a reservoir, which is slowly filled with the wetting phase, to initiate the spontaneous imbibition. Inertial effects at early times are neglected in the experiments.³⁵ The wetting phase used in the experiments is distilled water, colored with dye, of viscosity 1 cP. The spontaneous imbibition phenomena in the interacting porous layers is recorded using a Nikon D750 camera at approx. 60 frames per second and a resolution of 1280×720 pixels. Each experiment is performed thrice to ensure the reproducibility of the experimental observations. A customized MATLAB[®] program is used to track the fluid front displacement in the porous layers. Images at regular intervals are processed by choosing a suitable threshold of intensity for the imbibing phase in each layer.

3 Two layered porous medium

3.1 Experimental observations

The bead combination used in constructing the two-layered porous medium is $d_l = 274 \pm 24 \mu\text{m}$ and $d_s = 50 \pm 25 \mu\text{m}$, as shown in the snapshots of experiments in Fig. 2(a and b), respectively. The capillary pressures of layers made of beads of diameter d_l and d_s are P_{c_l} and P_{c_s} respectively, such that $P_{c_l} < P_{c_s}$. The porous medium is initially saturated with the non-wetting fluid and the reservoir at the right end of the porous medium is filled with dodecane as shown in Fig. 1. Colored water is introduced in the wetting phase reservoir at the left end of the porous medium. As the imbibition begins, we observe from the experimental snapshots shown in Fig. 2(a and b) that the wetting fluid front in the small permeability layer leads while the wetting fluid front in the large permeability layer lags behind. At early times, the viscous resistance in the porous medium is due to the resident non-wetting fluid only. Although the viscous resistance in the small pores is greater than the viscous resistance in the large pores, the imbibing fluid at the inlet is spontaneously driven first into the small pores due to greater capillary pressure. The residing phase in the small pores near the inlet is driven into the neighboring large pore spaces. The large pores which are already filled with the non-wetting phase expel the excess residing phase in the counter current direction of the flow of the imbibing phase. However, in this study, we neglect the

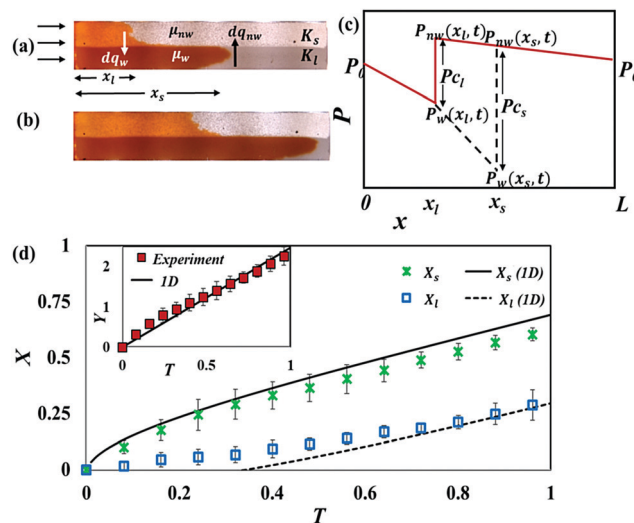


Fig. 2 Spontaneous displacement of a viscous resident fluid in a two layered porous medium. Snapshots of the experiment at dimensionless time (a) 0.5, and (b) 1. (c) Anticipated pressure gradient graph for the imbibition in a two layered porous medium. (d) Comparison of the experimental observations with the one-dimensional model for the average dimensionless length imbibed in the layers with dimensionless time. The inset shows the comparison of the experimental observations with the model for overall imbibition in the porous medium, $Y = (1 - M)(X_l^2 + X_s^2) + 2M(X_l + X_s)$ vs. dimensionless time T , the slope of the solid line is $2(1 + \alpha_s \beta_s) = 2.46$.

counter current imbibition as it is negligible in comparison to the cocurrent imbibition.^{40,41} Therefore, the cocurrent imbibition first begins in the small pores followed by subsequent imbibition in the large pores as observed in the experiments shown in Fig. 2(a and b). Fig. 2(d) shows the length X_i scaled with total length L as $X_i = \frac{x_i}{L}$ with time T , scaled as $T = \left(\frac{P_{c_l} K_l}{\phi \mu_w L^2} \right) t$. The value of $\frac{P_{c_l} K_l}{\phi \mu_w L^2}$ is of order 0.001 s^{-1} , used in all the experiments.

3.2 Mathematical model

Here, we present a quasi one-dimensional, Washburn like model for multiphase capillary driven flow in a two layered porous medium. As the imbibition proceeds, the wetting fluid of viscosity μ_w displaces a non-wetting fluid of viscosity μ_{nw} as shown in Fig. 2(a and b). The layer made with a large bead diameter (d_l) has permeability K_l and capillary pressure P_{c_l} , whereas the layer made with a small bead diameter (d_s) has permeability K_s and capillary pressure P_{c_s} , such that $K_l > K_s$ and $P_{c_l} < P_{c_s}$. The pressure at both the ends of the porous medium is the same, i.e., P_0 . At a time t , the locations of interfaces in the small permeability layer and large permeability layer are at distances x_s and x_l from the inlet, respectively, such that $x_s > x_l$, as shown in Fig. 2(a). To develop the one-dimensional model, we consider the assumptions of Ashraf *et al.*⁴² that:

(1) The pressure and pressure gradient in the two layers are the same when the same fluid is present in both the layers. To explain this assumption, in Fig. 2(c) we show the anticipated width averaged pressure along the length of the layers at a time

when the wetting fluid front is at x_1 in the large permeability layer and at x_s in the small permeability layer. We see that the pressure in both the layers is the same from 0 to x_1 . From x_s to L also, the same fluid is present in both the layers and the pressure in both the layers is the same.

(2) The wetting fluid transfers from the layer having the low capillary suction and high permeability to the layer having the larger capillary suction and lower permeability at the front x_1 . The non-wetting fluid transfers from the small permeability layer to the large permeability layer at the front in the small permeability layer *i.e.*, x_s . This is because, at x_1 , the sudden change in pressure in the large permeability layer due to the capillary pressure jump causes fluid exchange from the large to the small permeability layer. At x_s , the sudden capillary pressure jump in the small permeability layer causes non-wetting fluid exchange. By the lubrication approximation, we can consider the fluid exchanges to occur 'at' the front location only.

Debabbi *et al.*⁴³ mention different cross flow regimes depending on the transverse capillary number and effective aspect ratio. In our model, using definitions of Debbabi *et al.*, the effective aspect ratio is 5.6 and the transverse capillary number is of the order of 10. Therefore, we are in the capillary cross flow regime, *i.e.*, the capillary pressure contrast between the layers is driving the cross flow. We also ignore thin film flow^{38,44,45} and viscous fingering,⁴⁶ which cause the incomplete displacement of the non-wetting fluid behind the front. This is because thin films are not observed in our experiments, consistent with Lago *et al.*, Reyssat *et al.*, Steiakakis *et al.*, and Xiao *et al.*^{15,34,47,48} However, Zhao *et al.*, Tanino *et al.*, and Odier *et al.*^{38,44,45} report thin films during strong and forced imbibition conditions which is not the case in our experiments. In weak imbibition, Zhao *et al.*⁴⁴ also report the displacement by contact-line motion at low capillary numbers where the invading wetting fluid completely saturates the pores behind the front. At a high viscosity ratio, which causes fingering, capillary pressure alone will not be sufficient to displace the highly viscous non-wetting fluid. Therefore, our model would work for the capillary cross-flow regime, contact-line displacement of the non-wetting fluid in a layer and a favorable viscosity ratio. We neglect gravity and assume that the thickness of the layers is much smaller than the length and use the lubrication approximation.

We use Darcy's law for the pressure drop across different sections of the porous medium. The pressure gradient till the first fluid front x_1 from the inlet is the same in both the layers as shown in Fig. 2(c) and the pressure drop is

$$P_w(x_1, t) - P_0 = -\frac{\mu_w x_1}{K_1} v_1(t), \quad (2)$$

where $v_1(t)$ is the Darcy velocity of the fluid in the large permeability layer before x_1 , and P_0 and $P_w(x_1, t)$ are the imbibing phase pressures at the inlet and x_1 , respectively. The same pressure gradient till x_1 in both the layers implies $\frac{v_1(t)}{K_1} = \frac{v_s(t)}{K_s}$, where $v_s(t)$ is the Darcy velocity in the small permeability layer. At x_1 , the pressure jump across the front is equal to P_{c1} as shown

in Fig. 2(c). The sudden increase in the pressure across the fluid front at x_1 causes a volumetric fluid transfer rate of dq_w from the wider pores in the large permeability layer into the fine pores as indicated by the arrow in Fig. 2(a).^{26,42} The fluid transfer phenomenon occurs just before the front at x_1 and by the lubrication approximation, we consider it to occur at the front location. The velocity of the fluid front $\frac{dx_1}{dt}$ is given by

$$\phi_1 \frac{dx_1}{dt} = v_1(t) - \frac{dq_w}{A}, \quad (3)$$

where ϕ_1 is the porosity of the large permeability layer and A is the cross-sectional area of the layer. The fluid transfer causes the wetting fluid velocity in fine pores to increase after x_1 to $v_s(t) + \frac{dq_w}{A}$. The pressure drop of the wetting fluid in the small permeability layer from x_1 till the front at x_s is

$$P_w(x_s, t) - P_w(x_1, t) = -\frac{\mu_w(x_s - x_1)}{K_s} \left(v_s(t) + \frac{dq_w}{A} \right), \quad (4)$$

where $P_w(x_s, t)$ and $P_w(x_1, t)$ are the pressures of the wetting phase at x_s and x_1 , respectively. As the large permeability layer is occupied with the non-wetting fluid, the pressure drop from x_1 to x_s in this layer is given as

$$P_{nw}(x_s, t) - P_{nw}(x_1, t) = -\frac{\mu_{nw}(x_s - x_1)}{K_1} \left(v_1(t) - \frac{dq_w}{A} \right), \quad (5)$$

where $P_{nw}(x_s, t)$ and $P_{nw}(x_1, t)$ are the non-wetting phase pressures at x_s and x_1 , respectively. At x_s , the pressure difference across the fluid front in the small permeability layer is P_{cs} , as shown in Fig. 2(c). The sudden jump in pressure across the interface causes the non-wetting fluid of flow rate dq_{nw} to flow into the large permeability layer immediately after x_s , as shown by the arrow in Fig. 2(a). As the fluid transfer occurs after x_s , the velocity of the front can be equated to the wetting fluid flow in the small permeability layer between x_1 and x_s , *i.e.*,

$$\phi_s \frac{dx_s}{dt} = v_s(t) + \frac{dq_w}{A}, \quad (6)$$

where ϕ_s is the porosity of the small permeability layer and $\frac{dx_s}{dt}$ is the front velocity at x_s . After x_s , the pressure gradients of the large and the small permeability layers are the same, which is

$$P_0 - P_{nw}(x_s, t) = -\frac{\mu_{nw}(L - x_s)}{K_1} v_1(t), \quad (7)$$

where P_0 is the pressure at the exit. $|dq_w|$ and $|dq_{nw}|$ are equal for eqn (7) to be true. Eqn (2)–(7) are rearranged and substituting the capillary pressures as $(P_{nw}(x_i, t) - P_w(x_i, t)) = P_{ci}(i = l, s)$, we obtain the governing equations for the fronts' displacement as

$$P_{c1} = \frac{\mu_w x_1 + \mu_{nw}(L - x_s)}{K_1 + K_s} \left(\phi_1 \frac{dx_1}{dt} + \phi_s \frac{dx_s}{dt} \right) + \frac{\mu_{nw}(x_s - x_1)}{K_1} \left(\phi_1 \frac{dx_1}{dt} \right), \quad (8)$$

$$P_{c_s} - P_{c_l} = (x_s - x_l) \left(\frac{\mu_w}{K_s} \left(\phi_s \frac{dx_s}{dt} \right) - \frac{\mu_{nw}}{K_l} \left(\phi_l \frac{dx_l}{dt} \right) \right). \quad (9)$$

Eqn (8) and (9) are non-dimensionalized as

$$1 = \frac{X_l + M(1 - X_s)}{1 + \alpha_s} \left(\frac{dX_l}{dT} + \frac{dX_s}{dT} \right) + M(X_s - X_l) \left(\frac{dX_l}{dT} \right), \quad (10)$$

$$\beta_s - 1 = (X_s - X_l) \left(\frac{1}{\alpha_s} \left(\frac{dX_s}{dT} \right) - M \left(\frac{dX_l}{dT} \right) \right), \quad (11)$$

where $X_l = \frac{x_l}{L}$ and $X_s = \frac{x_s}{L}$ are the dimensionless lengths of the imbibition fronts in the large and the small permeability layers respectively, $\beta_s = \frac{P_{c_s}}{P_{c_l}}$ is the ratio of capillary pressures in the small and the large permeability layers, $\alpha_s = \frac{K_s}{K_l}$ is the ratio of the permeability of the small permeability layer to the large permeability layer, $M = \frac{\mu_{nw}}{\mu_w}$ is the viscosity ratio of the resident and the imbibing phases, and $T = t \frac{P_{c_l} K_l}{\phi \mu_w L^2}$ is the dimensionless time. The porosity in all the layers is considered the same, *i.e.*, $\phi_l = \phi_s = \phi$.

Eqn (10) and (11) are valid when $\beta_s > 1$, *i.e.*, the capillary suction for the wetting fluid in the small permeability layer is greater than the capillary suction of the large permeability layer due to the wettability of the sediments and smaller pores in the small permeability layer. If $\beta_s < 1$, then the wetting fluid will always travel faster in the large permeability layer due to less resistance to flow and higher capillary suction.

In eqn (11), we know that the value of β_s is greater than unity. If the left hand side of eqn (11) is positive then two solutions are possible. (1) $X_s > X_l$ and $\left(\frac{1}{\alpha_s} \left(\frac{dX_s}{dT} \right) - M \left(\frac{dX_l}{dT} \right) \right) > 0$ or (2) $X_s < X_l$ and $\left(\frac{1}{\alpha_s} \left(\frac{dX_s}{dT} \right) - M \left(\frac{dX_l}{dT} \right) \right) < 0$. If the viscosity ratio M is very high, then the capillary forces will not be sufficient to overcome the viscous resistance for practical purposes. We know that $\alpha_s < 1$, which depends on the permeability of the layers in a porous medium. This indicates that, for a favorable viscosity ratio, $\left(\frac{1}{\alpha_s} \left(\frac{dX_s}{dT} \right) - M \left(\frac{dX_l}{dT} \right) \right)$ is positive and, therefore, X_s is always greater than X_l as shown in the anticipated pressure gradient graph of Fig. 2.

We also observe that by rearranging and adding eqn (10) and (11) and then integrating, we obtain,

$$2(1 + \alpha_s \beta_s)T = (1 - M)(X_l^2 + X_s^2) + 2M(X_l + X_s). \quad (12)$$

Eqn (12) describes the flow behavior in the hydrodynamically interacting layered porous medium as a whole.

3.3 Comparison of experiments with the mathematical model

In Fig. 2(d), we compare the solution of the one-dimensional model with the experimental results of capillary driven imbibition in the two layered porous medium. For the model we use, $\alpha_s = 0.033$, $\beta_s = 7$ and $M = 1.35$. We see from the comparison of

Fig. 2(d) that the average length invaded by the wetting fluid with time is predicted with excellent accuracy by our one-dimensional model.

The inset of Fig. 2(d) compares the experimental observations for the two layered porous medium with eqn (12) by plotting $Y = (1 - M)(X_l^2 + X_s^2) + 2M(X_l + X_s)$ vs. dimensionless time T . The slope of Y vs. T is $2(1 + \alpha_s \beta_s)$, which matches with our experimental observations as shown in the inset of Fig. 2(d). Budaraju *et al.*¹¹ give the spontaneous imbibition equation for a homogeneous porous medium as

$$2T = (1 - M)(X^2) + 2MX, \quad (13)$$

where X is the average scaled length of displacement of a viscous non-wetting fluid by a viscous wetting fluid in scaled time T . Comparing eqn (13) with eqn (12), we note that the total volume or average length imbibed by the wetting fluid in a layered porous medium cannot be predicted by eqn (13) even if the effective permeability $K_{\text{eff}} = K_l + K_s$ and the effective capillary pressure $P_{c_{\text{eff}}} = \frac{(P_{c_l} K_l + P_{c_s} K_s)}{K_l + K_s}$ are used in the equation for scaling time t to T . This is because, for a layered porous medium, the square of the individual layers' front displacement is needed for eqn (12).

4 Three layered porous medium

We perform experiments using a three layered porous medium in which the layers are made of beads of diameters d_l , d_m and d_s , respectively. The permeability in a layer made of beads of diameter d_i is $K_i = A d_i^2$ ($i = l, m, s$), hence $K_l > K_m > K_s$. The capillary pressure for bead diameter d_i is $P_{c_i} = \frac{c\gamma}{d_i}$, therefore $P_{c_l} < P_{c_m} < P_{c_s}$. The porosity in the three layers is ϕ_l , ϕ_m , and ϕ_s , which is measured to be approximately 0.35. In a hydrodynamically interacting three layered porous medium, the three layers can be positioned in three ways and the possible arrangements are: (i) the small permeability layer at the center, as shown in Fig. 3(a), (ii) the medium permeability layer at the center, as shown in Fig. 4(a) and (iii) the large permeability layer at the center, as shown in Fig. 5(a). The spontaneous imbibition in the three porous media proceeds as explained earlier. We deal with these porous media separately to investigate the effect of the arrangement of the layers on the imbibition behavior.

4.1 Small permeability layer at the center

4.1.1 Experimental observations. We first perform experiments using a three layered porous medium where the layer made of beads of diameter $d_s = 50 \pm 25 \mu\text{m}$ is packed between the porous layers made of beads of diameters $d_l = 274 \pm 24 \mu\text{m}$ and $d_m = 125 \pm 25 \mu\text{m}$ as shown in Fig. 3(a and b). Fig. 3(d) shows the invaded length in each of the layers with time from the start of the imbibition. The length X_i is scaled as $X_i = \frac{x_i}{L}$ and the time is scaled as $T = t \left(\frac{P_{c_l} K_l}{\phi \mu_w L^2} \right)$. We see that the front

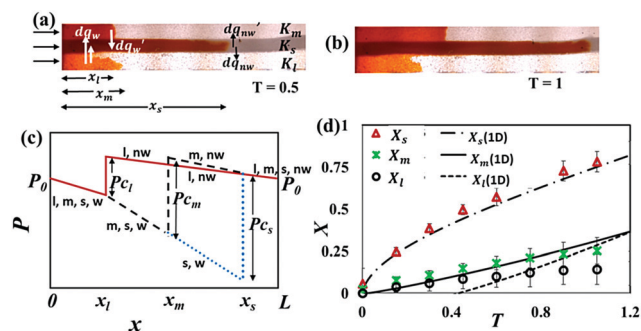


Fig. 3 Spontaneous displacement of a viscous resident fluid in a three layered porous medium with the small permeability layer in the center. Snapshots of the experiment at T (a) 0.5, and (b) 1. (c) Anticipated pressure gradient graph for the imbibition in a three layered porous medium with the small permeability layer at the center. Each pressure gradient line is annotated with the corresponding permeability and the phase flowing in the layer(s). The annotations l, m, and s represent large, medium and small permeability layers, and w and nw represent the wetting and the non-wetting phases, respectively. (d) Comparison of the experimental observations and the one-dimensional model for the dimensionless imbibition lengths in the layers vs. dimensionless time.

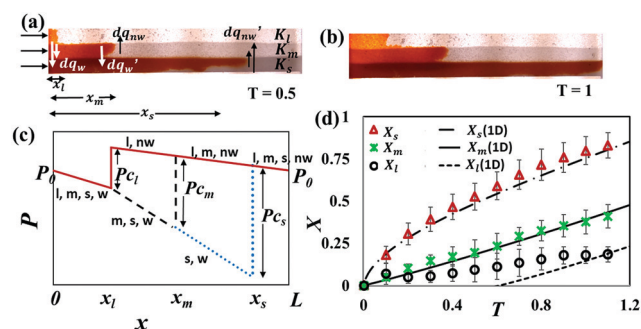


Fig. 4 Spontaneous displacement of a viscous resident fluid in a three layered porous medium with the medium permeability layer in the center. Snapshots of the experiment at T (a) 0.5, and (b) 1. (c) Anticipated pressure gradient graph for the imbibition in a three layered porous medium with the medium permeability layer in the center. Each pressure gradient line is annotated with the corresponding permeability and the phase flowing in the layer(s). The annotations l, m, and s represent large, medium and small permeability layers, and w and nw represent the wetting and the non-wetting phases, respectively. (d) Comparison of the experimental observations and the one-dimensional model for the dimensionless imbibition lengths in the layers vs. dimensionless time.

in the small permeability layer leads, followed by the front in the medium permeability layer, which is followed by the large permeability layer. We develop a mathematical model for the observed phenomenon.

4.1.2 Mathematical model. Fig. 3(c) shows the anticipated pressure gradients in different sections of the porous medium. Each pressure gradient line in the figure is annotated with the corresponding permeability and the phase flowing in the layer(s). The annotations l, m, and s represent large, medium and small permeability layers, and w and nw represent the wetting and the non-wetting phases, respectively. For example, annotation l, m, nw would represent the pressure gradient due

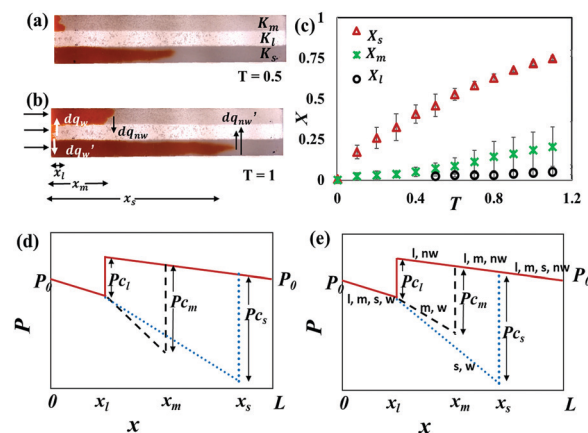


Fig. 5 Spontaneous displacement of a viscous resident fluid in a three layered porous medium with the large permeability layer at the center. Snapshots of the experiment at T (a) 0.5, and (b) 1. (c) Dimensionless distances imbibed in the layers with dimensionless time. (d) Anticipated pressure gradient graph for which the pressure gradient in the medium permeability layer is more than the pressure gradient in the small permeability layer. (e) Anticipated pressure gradient graph for which the pressure gradient in the small permeability layer is more than the pressure gradient in the medium permeability layer. Each pressure gradient line is annotated with the corresponding permeability and the phase flowing in the layer(s). The annotations l, m, and s represent large, medium and small permeability layers, and w and nw represent the wetting and the non-wetting phases, respectively.

to the non-wetting fluid in the large and the medium permeability layers. The same notation is used in the subsequent pressure gradient graphs. After time t , the fronts in the small, the medium and the large permeability layers have traveled x_s , x_m and x_l distances from the inlet, respectively. From Fig. 3(c), we see that the pressure drop in all the three layers till x_l is the same, given by Darcy's law as

$$P_w(x_l, t) - P_0 = -\frac{\mu_w x_l}{K_l} v_l(t), \quad (14)$$

implying that

$$\frac{v_l(t)}{K_l} = \frac{v_m(t)}{K_m} = \frac{v_s(t)}{K_s}, \quad (15)$$

where $v_i(t)$ is the Darcy velocity of the wetting fluid in layer i ($= l, m, s$) till x_l . Just before x_l , the wetting fluid transfers from the large permeability layer to the other two layers as shown by arrows in Fig. 3(a). The velocity of the front at x_l is given by

$$\phi_l \frac{dx_l}{dt} = v_l(t) - \frac{dq_w}{A}, \quad (16)$$

where dq_w is the rate of fluid transfer at x_l as shown in Fig. 3(a), A is the cross-sectional area of the layer and $\frac{dx_l}{dt}$ is the velocity of the fluid front in the large permeability layer. After x_l , the non-wetting fluid flows in the large permeability layer and the wetting fluid flows in the other two layers. The volumetric fluid transfer rate dq_w is distributed between the small and the medium permeability layers such that the pressure drop from x_l to x_m is the same for the medium and the small permeability layers.

If ηdq_w is the rate of fluid transfer into the medium permeability layer (K_m), the velocities of the wetting fluid in the medium and the small permeability layers after the fluid transfer dq_w are related as

$$\frac{v_m(t) + \frac{\eta dq_w}{A}}{K_m} = \frac{v_s(t) + \frac{(1-\eta)dq_w}{A}}{K_s}. \quad (17)$$

Substituting eqn (15) in eqn (17), we obtain $\eta = \frac{K_m}{K_m + K_s}$, which is the fraction of fluid transfer into the medium permeability layer. The pressure drop of the wetting fluid from x_1 to x_m in the medium permeability layer is given as

$$[P_w(x_m, t) - P_w(x_1, t)]_{K_m} = -\frac{\mu_w(x_m - x_1)}{K_m} \left(v_m(t) + \frac{\eta dq_w}{A} \right), \quad (18)$$

where $P_w(x_m, t)$ is the wetting phase pressure at x_m . The pressure drop of the non-wetting phase in the large permeability layer from x_1 to x_m is

$$[P_{nw}(x_m, t) - P_{nw}(x_1, t)]_{K_l} = -\frac{\mu_{nw}(x_m - x_1)}{K_l} \left(v_l(t) - \frac{dq_w}{A} \right). \quad (19)$$

In all the equations, $P_{nw}(x_i, t)$ is the non-wetting phase pressure at x_i and $P_w(x_i, t)$ is the wetting phase pressure at x_i . At x_m also, the wetting fluid transfers from the medium permeability layer to the small permeability layer with a volumetric flow rate dq_w' . The velocity of the front at x_m is given by

$$\phi_m \frac{dx_m}{dt} = v_m(t) + \eta \frac{dq_w}{A} - \frac{dq_w'}{A}, \quad (20)$$

where $\frac{dx_m}{dt}$ is the velocity of the fluid front in the medium permeability layer. The fluid transfer at x_m causes an increment in the velocity of the small permeability layer, which is $\left(v_s(t) + (1-\eta) \frac{dq_w}{A} + \frac{dq_w'}{A} \right)$. From x_m to x_s , the small permeability layer is occupied with the wetting fluid, while the medium and the large permeability layers are occupied with the non-wetting fluid. However, the non-wetting fluid in the two layers is not in contact as shown in Fig. 3(a). Therefore, the pressure in these two layers can be different as shown in the schematic of Fig. 3(c). The pressure drop in the large permeability layer from x_m to x_s is given by

$$[P_{nw}(x_s, t) - P_{nw}(x_m, t)]_{K_l} = -\frac{\mu_{nw}(x_s - x_m)}{K_l} \left(v_l(t) - \frac{dq_w}{A} \right). \quad (21)$$

The pressure drop in the medium permeability layer from x_m to x_s is given by

$$[P_w(x_s, t) - P_w(x_m, t)]_{K_m} = -\frac{\mu_w(x_s - x_m)}{K_m} \left(v_m(t) + \eta \frac{dq_w}{A} - \frac{dq_w'}{A} \right). \quad (22)$$

The pressure drop of the wetting fluid in the small permeability layer from x_m to x_s is

$$[P_w(x_s, t) - P_w(x_m, t)]_{K_s} = -\frac{\mu_w(x_s - x_m)}{K_s} \left(v_s(t) + (1-\eta) \frac{dq_w}{A} + \frac{dq_w'}{A} \right), \quad (23)$$

where $P_w(x_s, t)$ is the wetting phase pressure at x_s . The velocity of the fluid front in the small permeability layer $\frac{dx_s}{dt}$ is given by

$$\phi_s \frac{dx_s}{dt} = v_s(t) + (1-\eta) \frac{dq_w}{A} + \frac{dq_w'}{A}. \quad (24)$$

After x_s , the non-wetting fluid distributes such that the pressure in all the three layers is the same. The pressure drop of the non-wetting fluid after x_s till L is

$$P_0 - P_{nw}(x_s, t) = -\frac{\mu_{nw}(L - x_s)}{K_l} v_l(t), \quad (25)$$

where P_0 is the pressure at the non-wetting fluid reservoir. Now, across the fronts x_1 , x_m and x_s , the pressure difference between the non-wetting and the wetting phases is equal to the capillary pressure of the respective layer which is $P_{nw}(x_i, t) - P_w(x_i, t) = P_{ci}$ where $i = l, m, s$. By eliminating the terms $P_{nw}(x_i, t)$, $P_w(x_i, t)$, dq_w , dq_w' , and $v_i(t)$ ($i = l, m, s$), we obtain the governing equations for fluid fronts as

$$P_{cl} = \frac{\mu_w x_1 + \mu_{nw}(L - x_s)}{K_l + K_m + K_s} \left(\phi_l \frac{dx_1}{dt} + \phi_m \frac{dx_m}{dt} + \phi_s \frac{dx_s}{dt} \right) + \frac{\mu_{nw}(x_s - x_1)}{K_l} \left(\phi_l \frac{dx_1}{dt} \right), \quad (26)$$

$$P_{cm} - P_{cl} = (x_m - x_1) \frac{\mu_w}{K_m + K_s} \left(\phi_m \frac{dx_m}{dt} + \phi_s \frac{dx_s}{dt} \right) + (x_s - x_m) \frac{\mu_{nw}}{K_m} \left(\phi_m \frac{dx_m}{dt} \right) - (x_s - x_l) \frac{\mu_{nw}}{K_l} \left(\phi_l \frac{dx_l}{dt} \right), \quad (27)$$

$$P_{cs} - P_{cm} = (x_s - x_m) \left(\frac{\mu_w}{K_s} \left(\phi_s \frac{dx_s}{dt} \right) - \frac{\mu_{nw}}{K_m} \left(\phi_m \frac{dx_m}{dt} \right) \right). \quad (28)$$

Eqn (26)–(28) are non-dimensionalized by scaling lengths as $X_i = \frac{x_i}{L}$ ($i = l, m, s$), and time T is scaled as $T = t \left(\frac{P_{cl} K_l}{\phi_l \mu_w L^2} \right)$.

Viscosity ratio $M = \frac{\mu_{nw}}{\mu_w}$, capillary pressure ratio $\beta_j = \frac{P_{cj}}{P_{cl}}$, and permeability ratio $\alpha_j = \frac{K_j}{K_l}$ ($j = m, s$) are used and $\phi_l = \phi_m = \phi_s = \phi$.

The non-dimensionalized governing equations are

$$1 = \frac{X_l + M(1 - X_s)}{1 + \alpha_m + \alpha_s} \left(\frac{dX_l}{dT} + \frac{dX_m}{dT} + \frac{dX_s}{dT} \right) + M(X_s - X_l) \left(\frac{dX_l}{dT} \right), \quad (29)$$

$$\beta_m - 1 = (X_m - X_l) \frac{1}{\alpha_m + \alpha_s} \left(\frac{dX_m}{dT} + \frac{dX_s}{dT} \right) + (X_s - X_m) \frac{M}{\alpha_m} \left(\frac{dX_m}{dT} \right) - (X_s - X_l) M \left(\frac{dX_l}{dT} \right), \quad (30)$$

$$\beta_s - \beta_m = (X_s - X_m) \left(\frac{1}{\alpha_s} \left(\frac{dX_s}{dT} \right) - \frac{M}{\alpha_m} \left(\frac{dX_m}{dT} \right) \right). \quad (31)$$

We see that the left hand side of eqn (31) is positive, *i.e.*, $\beta_s - \beta_m > 0$, indicating that the right hand side should also be positive. This implies that both $(X_s - X_m)$ and $\left(\frac{1}{\alpha_s} \left(\frac{dX_s}{dT} \right) - \frac{M}{\alpha_m} \left(\frac{dX_m}{dT} \right) \right)$ are either greater than zero or less than zero. We see from Fig. 3(c) that $\left(\frac{1}{\alpha_s} \left(\frac{dX_s}{dT} \right) - \frac{M}{\alpha_m} \left(\frac{dX_m}{dT} \right) \right)$ is the difference between the pressure gradients of the small permeability layer and the medium permeability layer from x_m to x_s , *i.e.*, $\left(\frac{dP}{dx} \right)_{K_s, w} \Big|_{x_m}^{x_s} - \left(\frac{dP}{dx} \right)_{K_m, nw} \Big|_{x_m}^{x_s}$, where $\left(\frac{dP}{dx} \right)_{K_i, y}$ is the pressure gradient in the layer having permeability K_i , occupied by phase y ($= w, nw$). For a favorable viscosity ratio, $\left(\frac{dP}{dx} \right)_{K_s, w} \Big|_{x_m}^{x_s} > \left(\frac{dP}{dx} \right)_{K_m, nw} \Big|_{x_m}^{x_s}$, due to the fluid transfer to the small permeability layer. Therefore $\left(\frac{1}{\alpha_s} \left(\frac{dX_s}{dT} \right) - \frac{M}{\alpha_m} \left(\frac{dX_m}{dT} \right) \right)$ is always positive implying that $X_s > X_m$ at all times. Therefore, the position of fluid fronts is $X_s > X_m > X_l$ at all times.

4.1.3 Comparison of experiments with the mathematical model. We use $\beta_s = 7$, $\beta_m = 1.8$, $\alpha_s = 0.033$, $\alpha_m = 0.20$ and $M = 1.35$ to solve eqn (29)–(31) for X_s , X_m , and X_l with T . In Fig. 3(d), we compare the experimental observations with the solution to eqn (29)–(31). We see that the invading lengths predicted by the numerical solution are in excellent agreement with the experimental observations for the small and the medium permeability layers. However, we see that the imbibition in the large permeability layer is slightly different from that predicted by our model. This is because, initially, when the fluid fronts in all the layers are close to the inlet, the non-wetting fluid in narrow pores is displaced into the larger pores. This creates a small counter-current flow in the experiments, which vanishes very soon. The small counter-current flow causes the front in the large permeability layer to move slowly in the experiments which is not captured by our one-dimensional model. This causes the slight variation in the front displacement in the large permeability layer between the experimental observations and predictions by our one-dimensional model. Therefore, in a three layered porous medium with the small permeability layer as the middle layer, the fluid front in the small permeability layer travels ahead, followed by the fluid front in the medium permeability layer, while the fluid front in the large permeability layer lags behind.

4.2 Medium permeability layer at the center

4.2.1 Experimental observations. In this arrangement, the layer made of beads of diameter $d_m = 125 \pm 25 \mu\text{m}$ is placed

between the porous layers made of beads of diameters $d_l = 274 \pm 24 \mu\text{m}$ and $d_s = 50 \pm 25 \mu\text{m}$ as shown in Fig. 4(a and b). Experimental observations of the dimensionless invaded length $X_i = \frac{x_i}{L}$ in each of the layers with dimensionless time $T = t \left(\frac{Pc_l K_l}{\phi \mu_w L^2} \right)$ are shown in Fig. 4(d).

We see that the leading front is in the small permeability layer, followed by the front in the medium permeability layer, which is followed by the front in the large permeability layer. To develop a mathematical model for the observed phenomena, we consider the anticipated pressure gradient graph of Fig. 4(c), which shows the pressure gradient in sections of each of the layers occupied with the invading and the resident phases. We now develop a mathematical model in a similar fashion as explained in Section 4.1.2.

4.2.2 Mathematical model. We determine the pressure drop in all the sections from the anticipated pressure gradient graph shown in Fig. 4(c), for each of the layers. We obtain the equations governing the front displacement for all the layers in this arrangement as

$$Pc_l = \frac{\mu_w x_l + \mu_{nw}(L - x_s)}{K_l + K_m + K_s} \left(\phi_l \frac{dx_l}{dt} + \phi_m \frac{dx_m}{dt} + \phi_s \frac{dx_s}{dt} \right) + \frac{\mu_{nw}(x_m - x_l)}{K_l} \left(\phi_l \frac{dx_l}{dt} \right) + \frac{\mu_{nw}(x_s - x_m)}{K_l + K_m} \left(\phi_l \frac{dx_l}{dt} + \phi_m \frac{dx_m}{dt} \right), \quad (32)$$

$$Pc_m - Pc_l = (x_m - x_l) \left(\frac{\mu_w}{K_s + K_m} \left(\phi_m \frac{dx_m}{dt} + \phi_s \frac{dx_s}{dt} \right) - \frac{\mu_{nw}}{K_l} \left(\phi_l \frac{dx_l}{dt} \right) \right), \quad (33)$$

$$Pc_s - Pc_m = (x_s - x_m) \left(\frac{\mu_w}{K_s} \left(\phi_s \frac{dx_s}{dt} \right) - \frac{\mu_{nw}}{K_m + K_l} \left(\phi_l \frac{dx_l}{dt} + \phi_m \frac{dx_m}{dt} \right) \right). \quad (34)$$

Eqn (32)–(34) are non-dimensionalized by scaling lengths as $X_i = \frac{x_i}{L}$ ($i = l, m, s$), and time as $T = t \left(\frac{Pc_l K_l}{\phi \mu_w L^2} \right)$, and using viscosity ratio $M = \frac{\mu_{nw}}{\mu_w}$, capillary pressure ratio $\beta_j = \frac{Pc_j}{Pc_l}$, permeability ratio $\alpha_j = \frac{K_j}{K_l}$ ($j = m, s$), and $\phi_l = \phi_m = \phi_s = \phi$. The governing equations become,

$$1 = \frac{X_l + M(1 - X_s)}{1 + \alpha_m + \alpha_s} \left(\frac{dX_l}{dT} + \frac{dX_m}{dT} + \frac{dX_s}{dT} \right) + M(X_m - X_l) \left(\frac{dX_l}{dT} \right) + \frac{M(X_s - X_m)}{1 + \alpha_m} \left(\frac{dX_l}{dT} + \frac{dX_m}{dT} \right), \quad (35)$$

$$\beta_m - 1 = (X_m - X_l) \left(\frac{1}{\alpha_m + \alpha_s} \left(\frac{dX_m}{dT} + \frac{dX_s}{dT} \right) - M \left(\frac{dX_l}{dT} \right) \right), \quad (36)$$

$$\beta_s - \beta_m = (X_s - X_m) \left(\frac{1}{\alpha_s} \left(\frac{dX_s}{dT} \right) - \frac{M}{1 + \alpha_m} \left(\frac{dX_l}{dT} + \frac{dX_m}{dT} \right) \right). \quad (37)$$

From eqn (37), we see that the left hand side is greater than zero, implying that the right hand side should also be positive. Fig. 4(c) shows that in the region x_m to x_s , the pressure gradient of the wetting fluid in the small permeability layer must be greater than the pressure gradient of the non-wetting fluid in the other two layers, so that the capillary pressure condition $P_{c_s} > P_{c_m}$ is satisfied. Using Darcy's law for the pressure gradients of the wetting and the non-wetting fluids from x_m to x_s , we get $\left(\frac{1}{\alpha_s} \left(\frac{dX_s}{dT}\right) - \frac{M}{1 + \alpha_m} \left(\frac{dX_l}{dT} + \frac{dX_m}{dT}\right)\right) > 0$, indicating that $(X_s - X_m) > 0$. Therefore, for the three layered porous medium with the medium permeability layer at the center, the fluid front in the small permeability layers travels faster than the fluid front in the medium permeability layer while the fluid front in the large permeability layer lags behind.

4.2.3 Comparison of experiments with the mathematical model. We use $\beta_s = 7$, $\beta_m = 1.8$, $\alpha_s = 0.033$, $\alpha_m = 0.20$ and $M = 1.35$, to solve eqn (35)–(37) for the dimensionless lengths imbibed with dimensionless time. We see from Fig. 4(d) that the experimental observations are in excellent agreement with the invading lengths predicted by our model. Therefore, in a three layered porous medium with the medium permeability layer as the center layer, the fluid front in the small permeability layer travels ahead, followed by the fluid front in the medium permeability layer, while the fluid front in the large permeability layer lags behind.

4.3 Large permeability layer at the center

4.3.1 Experimental observations. In this arrangement, the large permeability layer made of beads of diameter $d_l = 274 \pm 24 \mu\text{m}$ is placed between the porous layers made of beads of diameters $d_m = 125 \pm 25 \mu\text{m}$ and $d_s = 50 \pm 25 \mu\text{m}$ as shown in Fig. 5(a and b). Fig. 5(c) shows the length $X_l = \frac{x_l}{L}$ invaded in each of the layers with time $T = t \left(\frac{P_{c_l} K_l}{\phi \mu_w L^2}\right)$. We see that the fluid front in the small permeability layer leads, followed by the fluid front in the medium permeability layer, which is followed by the fluid front in the large permeability layer. We proceed with the positions of the fluid fronts observed in experiments and develop a one-dimensional model for this arrangement.

4.3.2 Mathematical model. For this case, where the large permeability layer is placed at the center, the wetting fluid is continuous in all the three layers till x_l as shown in Fig. 5(b). After x_l , although the medium and the small permeability layers are filled with the wetting fluid, their pressure gradients are different as the wetting fluid in the two layers is not in contact. The pressure gradient of the wetting phase after x_l in the medium permeability layer can be greater than or less than the pressure gradient in the small permeability layer as shown in Fig. 5(d and e). From Fig. 5(d), the pressure gradient in the medium permeability layer is more than the pressure gradient in the small permeability layer from x_l to x_m , i.e.,

$$\frac{\mu_w \left(\phi_m \frac{dx_m}{dt}\right)}{K_m} > \frac{\mu_w \left(\phi_s \frac{dx_s}{dt}\right)}{K_s} \text{ using Darcy's law. As } \frac{1}{K_m} < \frac{1}{K_s},$$

therefore $\left(\phi_m \frac{dx_m}{dt}\right) > \left(\phi_s \frac{dx_s}{dt}\right)$. This means that at any given time $x_m > x_s$ which is a contradiction. This precludes the possibility of the case in Fig. 5(d). Therefore, for this arrangement, Fig. 5(e) is the anticipated pressure gradient when $x_s > x_m > x_l$.

By using the inferences from the pressure vs. distance graphs of Fig. 5(e), we develop the one-dimensional model for this arrangement, similar to the one-dimensional model explained in Section 4.1.2. The equations governing the spontaneous imbibition in the layers are

$$P_{c_l} = \frac{\mu_w x_l + \mu_{nw}(L - x_s)}{K_l + K_m + K_s} \left(\phi_l \frac{dx_l}{dt} + \phi_m \frac{dx_m}{dt} + \phi_s \frac{dx_s}{dt} \right) + (x_m - x_l) \frac{\mu_{nw}}{K_l} \phi_l \frac{dx_l}{dt} + (x_s - x_m) \frac{\mu_{nw}}{K_l + K_m} \left(\phi_l \frac{dx_l}{dt} + \phi_s \frac{dx_m}{dt} \right), \quad (38)$$

$$P_{c_m} - P_{c_l} = (x_m - x_l) \left(\frac{\mu_w}{K_m} \left(\phi_m \frac{dx_m}{dt} \right) - \frac{\mu_{nw}}{K_l} \left(\phi_l \frac{dx_l}{dt} \right) \right), \quad (39)$$

$$P_{c_s} - P_{c_l} = (x_s - x_l) \frac{\mu_w}{K_s} \left(\phi_s \frac{dx_s}{dt} \right) - \frac{\mu_{nw}}{K_l} (x_m - x_l) \left(\phi_l \frac{dx_l}{dt} \right) - (x_s - x_m) \frac{\mu_{nw}}{K_m + K_l} \left(\phi_l \frac{dx_l}{dt} + \phi_m \frac{dx_m}{dt} \right). \quad (40)$$

Eqn (38)–(40) in dimensionless form are

$$1 = \frac{X_l + M(1 - X_s)}{1 + \alpha_m + \alpha_s} \left(\frac{dX_l}{dT} + \frac{dX_m}{dT} + \frac{dX_s}{dT} \right) + (X_m - X_l) M \frac{dX_l}{dT} + (X_s - X_m) \frac{M}{1 + \alpha_m} \left(\frac{dX_l}{dT} + \frac{dX_m}{dT} \right), \quad (41)$$

$$\beta_m - 1 = (X_m - X_l) \left(\frac{1}{\alpha_m} \left(\frac{dX_m}{dT} \right) - M \left(\frac{dX_l}{dT} \right) \right), \quad (42)$$

$$\beta_s - 1 = (X_s - X_l) \frac{1}{\alpha_s} \left(\frac{dX_s}{dT} \right) - M \left((X_m - X_l) \left(\frac{dX_l}{dT} \right) + (X_s - X_m) \frac{1}{1 + \alpha_m} \left(\frac{dX_l}{dT} + \frac{dX_m}{dT} \right) \right). \quad (43)$$

In eqn (41)–(43), the lengths are scaled as $X_i = \frac{x_i}{L}$ ($i = l, m, s$), time T is scaled as $T = t \left(\frac{P_{c_l} K_l}{\phi \mu_w L^2} \right)$, the viscosity ratio is $M = \frac{\mu_{nw}}{\mu_w}$, the capillary pressure ratios are $\beta_j = \frac{P_{c_j}}{P_{c_l}}$, the permeability ratios are $\alpha_j = \frac{K_j}{K_l}$ ($j = m, s$), and $\phi_l = \phi_m = \phi_s = \phi$.

4.3.3 Comparison of experiments with the mathematical model. Fig. 6(a) shows the solution to eqn (41)–(43), considering $\alpha_m = 0.20$, $\alpha_s = 0.03$, $\beta_m = 1.8$, $\beta_s = 7$ and $M = 1.35$. We see from Fig. 6(a) that X_m and X_s are very close to each other contradictory to the experimental results shown in Fig. 5(c). This is because of the packing of beads of diameter d_m above the beads of diameter d_l . When we fill the porous layers between the vertical acrylic sheets,

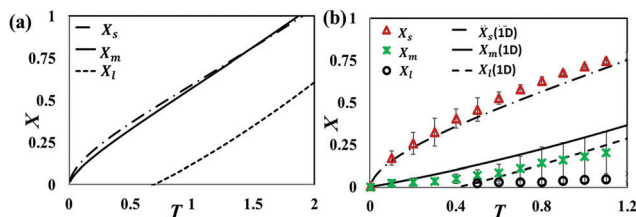


Fig. 6 The spontaneous displacement of a viscous resident fluid in a three layered porous medium with the large permeability layer at the center. (a) Numerical solution for the dimensionless distances imbibed in the layers with dimensionless time for $\alpha_m = 0.20$, $\alpha_s = 0.03$, $\beta_m = 1.8$, $\beta_s = 7$ and $M = 1.35$. (b) Comparison of the experimental observations with the numerical solution for the dimensionless distances imbibed in the layers with dimensionless time for $\alpha_m = 0.20$, $\alpha_s = 0.03$, $\beta_m = 1.1$, $\beta_s = 7$ and $M = 1.35$.

the small beads (d_s) are filled first with regular tapping to ensure proper packing of the layer, then the large beads (d_l) are also filled by tapping of beads, and form the large permeability layer at the center. Then, on the top, the medium beads (d_m) are filled, which is the medium permeability layer. The large diameter beads create pore spaces large enough for the medium beads to slip into the large permeability layer. To minimize the number of medium beads slipping into the pores of the large permeability layer, we do not tap the porous medium after the medium permeability layer is filled. Therefore, the medium permeability layer is loosely packed causing the capillary pressure to decrease. In Fig. 6(b), we show the comparison of the model with the experiments using $\alpha_m = 0.20$, $\alpha_s = 0.03$, $\beta_m = 1.1$, $\beta_s = 7$ and $M = 1.35$ in the model, which shows a good match. Therefore, if the capillary pressure and the permeability of the layers are known for any arrangement, our one-dimensional model can predict the flow behavior in the layered porous medium.

We note from the model predictions for $\beta_m = 1.8$, and $\beta_s = 7$ in Fig. 6(a) that the fluid front in the small permeability layer leads initially, but the fluid front in the medium permeability layer leads at later times. From the governing equations, we infer that the model does not predict the relative location of the fluid fronts in the small and the medium permeability layers. Therefore, $(X_s - X_m)$ can be greater than or less than zero. Fig. 7(a) shows an anticipated pressure gradient graph for a case when $X_m > X_s$. We now develop a mathematical model for this case.

4.3.4 Mathematical model ($X_m > X_s > X_l$). In this arrangement, the large permeability layer is placed at the center, as shown in the schematic of Fig. 7(b). The equations governing the flow behavior for the large permeability layer in the center and $x_m > x_s > x_l$ as shown in Fig. 7(a and b) are given by

$$P_{c_l} = \frac{\mu_w x_l + \mu_{nw}(L - x_m)}{K_l + K_m + K_s} \left(\phi_l \frac{dx_l}{dt} + \phi_m \frac{dx_m}{dt} + \phi_s \frac{dx_s}{dt} \right) + (x_s - x_l) \frac{\mu_{nw}}{K_l} \phi_l \frac{dx_l}{dt} + (x_m - x_s) \frac{\mu_{nw}}{K_l + K_s} \left(\phi_l \frac{dx_l}{dt} + \phi_s \frac{dx_s}{dt} \right), \quad (44)$$

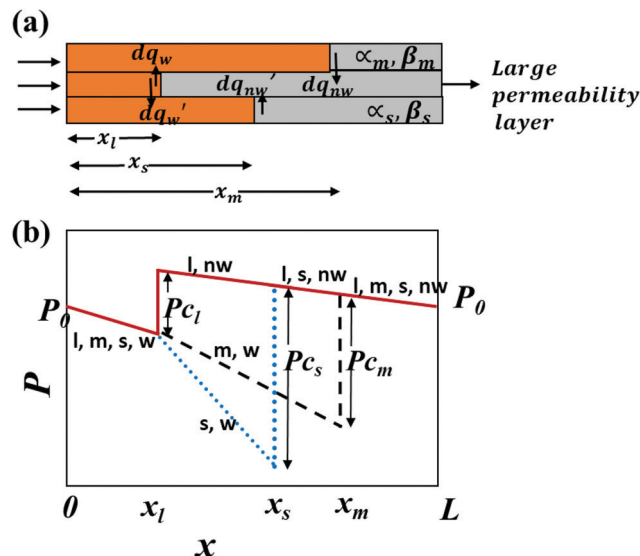


Fig. 7 (a) Schematic of fluid front positions in the layers. (b) Anticipated pressure gradient graph for spontaneous imbibition in a three layered porous medium with the large permeability layer at the center for $x_m > x_s > x_l$. Each pressure gradient line is annotated with the corresponding permeability and the phase flowing in the layer(s). The annotations l, m, and s represent large, medium and small permeability layers, and w and nw represent the wetting and the non-wetting phases, respectively.

$$P_{c_m} - P_{c_l} = (x_m - x_l) \frac{\mu_w}{K_m} \left(\phi_m \frac{dx_m}{dt} \right) - \frac{\mu_{nw}}{K_l} \left((x_s - x_l) \phi_l \frac{dx_l}{dt} + (x_m - x_s) \frac{K_l}{K_l + K_s} \left(\phi_l \frac{dx_l}{dt} + \phi_s \frac{dx_s}{dt} \right) \right), \quad (45)$$

$$P_{c_s} - P_{c_l} = (x_s - x_l) \left(\frac{\mu_w}{K_s} \left(\phi_s \frac{dx_s}{dt} \right) - \frac{\mu_{nw}}{K_l} \left(\phi_l \frac{dx_l}{dt} \right) \right). \quad (46)$$

Converting eqn (44)–(46) to dimensionless form, we get

$$1 = \frac{X_l + M(1 - X_m)}{1 + \alpha_m + \alpha_s} \left(\frac{dX_l}{dT} + \frac{dX_m}{dT} + \frac{dX_s}{dT} \right) + (X_s - X_l) M \frac{dX_l}{dT} + (X_m - X_s) \frac{M}{1 + \alpha_s} \left(\frac{dX_l}{dT} + \frac{dX_s}{dT} \right), \quad (47)$$

$$\beta_m - 1 = (X_m - X_l) \frac{1}{\alpha_m} \left(\frac{dX_m}{dT} \right) - M \left((X_s - X_l) \left(\frac{dX_l}{dT} \right) + (X_m - X_s) \frac{1}{1 + \alpha_s} \left(\frac{dX_l}{dT} + \frac{dX_s}{dT} \right) \right), \quad (48)$$

$$\beta_s - 1 = (X_s - X_l) \left(\frac{1}{\alpha_s} \left(\frac{dX_s}{dT} \right) - M \left(\frac{dX_l}{dT} \right) \right). \quad (49)$$

In eqn (47)–(49), the lengths are scaled as $X_i = \frac{x_i}{L}$ ($i = l, m, s$), time T is scaled as $T = t \left(\frac{P_{c_l} K_l}{\phi \mu_w L^2} \right)$, the viscosity ratio is

$M = \frac{\mu_{nw}}{\mu_w}$, the capillary pressure ratios are $\beta_j = \frac{Pc_j}{Pc_l}$, the permeability ratios are $\alpha_j = \frac{K_j}{K_l}$ ($j = m, s$), and $\phi_l = \phi_m = \phi_s = \phi$. Eqn (47)–(49) also do not suggest if $X_m > X_s$ or $X_s > X_m$.

We see that for the case $X_s > X_m > X_l$, the section $|X_s - X_m|$ is filled with the wetting fluid in the small permeability layer, while the medium permeability layer is filled with the non-wetting fluid. It is opposite for the case $X_m > X_s > X_l$, in which the non-wetting fluid occupies the small permeability layer and the wetting fluid occupies the medium permeability layer, in the section $|X_s - X_m|$. We now use eqn (41)–(43) and (47)–(49) to understand the effect of the arrangement and viscosity ratio on the capillary pressure driven displacement behavior in the layers. We consider three cases of the three layered porous medium, namely (A), (B) and (C), having the large permeability layer at the center as shown in Fig. 7(b). For (A), we consider $\alpha_m = 0.20$, $\alpha_s = 0.03$, $\beta_m = 1.1$, and $\beta_s = 7$ as in our experiments. For (B), we consider $\alpha_m = 0.40$, $\alpha_s = 0.20$, $\beta_m = 1.5$, and $\beta_s = 1.8$. For (C), we consider $\alpha_m = 0.40$, $\alpha_s = 0.13$, $\beta_m = 1.57$, and $\beta_s = 2.74$. In Fig. 8, we show the numerical solutions for the porous media (A), (B) and (C), for different viscosity ratios.

If we analyze eqn (41)–(43) and (47)–(49) for $M = 0$, we get the same set of governing equations, which are,

$$1 + \alpha_m + \alpha_s = X_l \left(\frac{dX_l}{dT} + \frac{dX_m}{dT} + \frac{dX_s}{dT} \right), \quad (50)$$

$$\alpha_m(\beta_m - 1) = (X_m - X_l) \left(\frac{dX_m}{dT} \right), \quad (51)$$

$$\alpha_s(\beta_s - 1) = (X_s - X_l) \left(\frac{dX_s}{dT} \right). \quad (52)$$

For this case, $X_m > X_s$ when $[\alpha_m(\beta_m - 1)] - [\alpha_s(\beta_s - 1)] > 0$ and $X_s > X_m$ when $[\alpha_m(\beta_m - 1)] - [\alpha_s(\beta_s - 1)] < 0$ as shown in Fig. 8(a and b) for the porous media (A) and (B), respectively. For the porous medium (C), $[\alpha_m(\beta_m - 1)] - [\alpha_s(\beta_s - 1)] = 0$, we get $X_s = X_m$ as shown in Fig. 8(c). We also observe from Fig. 8(a–c) that, for $M = 0$, the imbibition is faster at early times. The imbibition rate decreases with time due to the increase in viscous resistance in the pore spaces, caused due to the invasion of the viscous wetting fluid.

For a favorable and non-zero viscosity ratio M , the same trend is observed, i.e., $X_m > X_s$ when $[\alpha_m(\beta_m - 1)] - [\alpha_s(\beta_s - 1)] > 0$ and $X_s > X_m$ when $[\alpha_m(\beta_m - 1)] - [\alpha_s(\beta_s - 1)] < 0$. In Fig. 8(d–f), we show the capillary driven flow for $M = 0.5$, for porous media (A), (B), and (C), respectively. Due to $M < 1$, the imbibition in the layers is initially fast, slowing down as the fronts advance, due to the increase in viscous resistance.

For the case where the imbibing and residing fluids have the same viscosity, i.e., $M = 1$, the viscous resistance in all the layers remains the same throughout the imbibition process once the imbibition in the large permeability layer begins. Although the condition $[\alpha_m(\beta_m - 1)] - [\alpha_s(\beta_s - 1)] = 0$ is satisfied for the porous medium (C), X_m leads for low viscosity ratios ($M = 0.5, 1$), as shown in Fig. 8(f and i). The lower viscous resistance in the

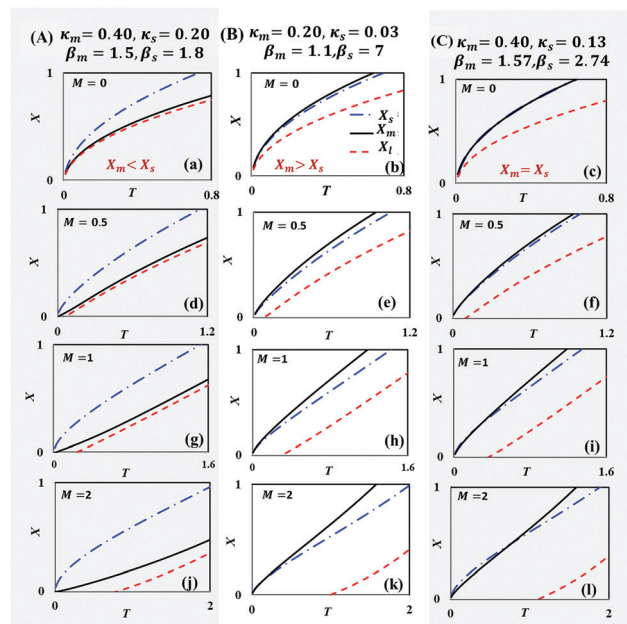


Fig. 8 Dimensionless time T vs. X_i ($i = l, m, s$) for three layered porous media (A), (B) and (C) having the large permeability layer in the center. The case name is mentioned in the graphs. For (A), we consider $\alpha_m = 0.20$, $\alpha_s = 0.03$, $\beta_m = 1.1$, and $\beta_s = 7$. For (B), we consider $\alpha_m = 0.40$, $\alpha_s = 0.20$, $\beta_m = 1.5$, and $\beta_s = 1.8$. For (C), we consider $\alpha_m = 0.40$, $\alpha_s = 0.13$, $\beta_m = 1.57$, and $\beta_s = 2.74$. The viscosity ratio M is mentioned at the top left corner in the figures for each of the porous media (A), (B), and (C).

medium permeability layer for low viscosity ratios causes a faster displacement of the resident fluid as compared to the small permeability layer, where the viscous resistance is high, as shown in Fig. 8(f and i).

For a viscosity ratio greater than unity as shown in Fig. 8(j–l), we see that the imbibition in the small and the medium permeability layers is much greater than the imbibition in the large permeability layer. This is due to the low capillary suction of the large permeability layer and the high viscous resistance offered by the resident fluid. The effect of the viscous resistance becomes clear from Fig. 8(l), where initially the capillary pressure dominates and the front in the small permeability layer leads. At later times, the effect of the viscous resistance dominates and the front in the medium permeability layer leads. We use our model for $M > 0$ and conclude that, for a range of values $-|\delta| < [\alpha_m(\beta_m - 1)] - [\alpha_s(\beta_s - 1)] < |\delta|$, the front in the small permeability layer leads initially and at later times the fluid front in the medium permeability layer leads. The imbibition behavior $X_m > X_s > X_l$ is observed if $[\alpha_m(\beta_m - 1)] - [\alpha_s(\beta_s - 1)] > |\delta|$ and the fluid front positions $X_s > X_m > X_l$ are observed for $[\alpha_m(\beta_m - 1)] - [\alpha_s(\beta_s - 1)] < -|\delta|$. The value of $|\delta|$ depends on the viscosity ratio, capillary pressure and permeability of the layers, which needs to be explored further.

4.3.5 Experimental observations and comparison with the mathematical model for $X_m > X_s > X_l$. To observe the anomalous behavior $X_m > X_s > X_l$ in a layered porous medium, we perform experiments with beads of diameters $d_l = 274 \pm 24 \mu\text{m}$, $d_m = 175 \pm 25 \mu\text{m}$ and $d_s = 125 \pm 25 \mu\text{m}$, such that $\alpha_m = 0.40$,

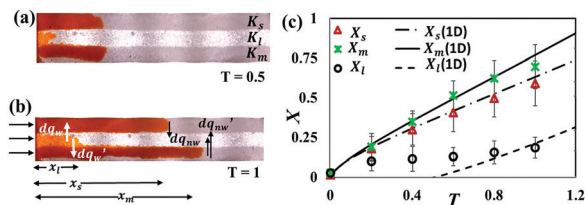


Fig. 9 Spontaneous displacement of a viscous resident fluid in a three layered porous medium with the large permeability layer in the center for $x_m > x_s > x_l$. Snapshots of the experiment at dimensionless times (a) 0.5, and (b) 1. (c) Comparison of the experimental observations with the one-dimensional model for the dimensionless imbibed length in the layers vs. dimensionless time for $d_l = 274 \pm 24 \mu\text{m}$, $d_m = 175 \pm 25 \mu\text{m}$ and $d_s = 125 \pm 25 \mu\text{m}$, such that, $\alpha_m = 0.40$, $\alpha_s = 0.20$, $\beta_m = 1.5$ and $\beta_s = 1.8$.

$\alpha_s = 0.20$, $\beta_m = 1.5$ and $\beta_s = 1.8$ as in case (B) in the previous section. We see that the leading front is in the medium permeability layer, followed by the front in the small permeability layer, which is followed by the front in the large permeability layer as shown in Fig. 9(a and b). A plot of dimensionless length $X = \frac{x}{L}$ vs. dimensionless time $T = \frac{Pc_l K_l}{\phi \mu_w L^2} t$ shows that $x_m > x_s > x_l$ as in Fig. 9(c). Fig. 9(c) shows that the experimental observations are in excellent agreement with the solution to the one-dimensional model given by eqn (47)–(49).

4.4 Equation governing the imbibition in a layered porous medium

For all the arrangements of a three layered porous medium, by rearranging the dimensionless governing equations of each arrangement and adding them, we obtain

$$2(1 + \alpha_m \beta_m + \alpha_s \beta_s)T = (1 - M)(X_l^2 + X_m^2 + X_s^2) + 2M(X_l + X_m + X_s), \quad (53)$$

which describes the spontaneous imbibition dynamics of a hydrodynamically interacting three layered porous medium, irrespective of the arrangement of the layers.

Eqn (53) explains that, for all the three arrangements, a plot of $Y = (1 - M)(X_l^2 + X_m^2 + X_s^2) + 2M(X_l + X_m + X_s)$ vs. dimensionless time T is a straight line of slope $2(1 + \alpha_m \beta_m + \alpha_s \beta_s)$, which is 3.21 for the experiments in Sections 4.1.2 and 4.2.2 as shown in Fig. 10(a). For the experiments in Section 4.3.2, the value of $2(1 + \alpha_m \beta_m + \alpha_s \beta_s)$ is 2.92 as shown in Fig. 10(b). Fig. 10(c) shows that for the experiments in Section 4.3.4, the value of $2(1 + \alpha_m \beta_m + \alpha_s \beta_s)$ is 3.97.

We observe that eqn (53) is analogous to the modified Lucas-Washburn law as in eqn (13), which predicts the fluid front position when the porous medium is initially filled with a viscous fluid for a homogeneous porous medium. However, we cannot predict the total volume or the average length imbibed in the layered porous medium even if the effective permeability $K_{\text{eff}} = K_l + K_m + K_s$ and capillary pressure $Pc_{\text{eff}} = \frac{Pc_l K_l + Pc_m K_m + Pc_s K_s}{K_l + K_m + K_s}$ are used in eqn (13). This is because the squares of the imbibed length are needed for eqn (53).

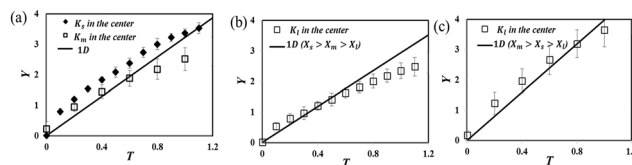


Fig. 10 Comparison of the experimental observations with the one-dimensional model for overall imbibition in a porous medium for $Y = (1 - M)(X_l^2 + X_m^2 + X_s^2) + 2M(X_l + X_m + X_s)$ vs. dimensionless time T for the arrangements: (a) the small permeability layer in the center and the medium permeability layer in the center. The slope of the solid line is $2(1 + \alpha_m \beta_m + \alpha_s \beta_s) = 3.21$. (b) The large permeability layer at the center, where $x_s > x_m > x_l$. The slope of the solid line is 2.92. (c) The large permeability layer at the center, where $x_m > x_s > x_l$. The slope of the solid line is 3.97.

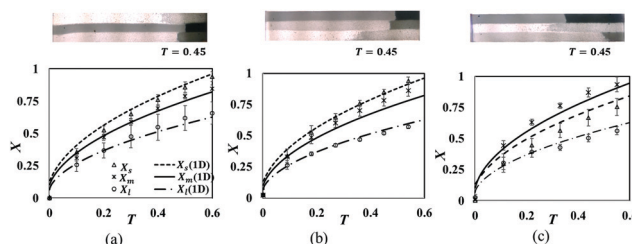


Fig. 11 Dimensionless length vs. time comparison of the experimental observations and one-dimensional model for three layered porous media (a) small permeability at the center, (b) medium permeability at the center and (c) large permeability at the center for viscosity ratio $M = 0$.

To verify our model for other viscosity ratios, we performed experiments for viscosity ratio $M = 0$ where dodecane of viscosity 1.35 cP displaces air, and $M = 2.6$ where colored water of viscosity 1 cP displaces hexadecane of viscosity 2.6 cP. Fig. 11 compares the experimental observations for the imbibed dimensionless length vs. dimensionless time with our one-dimensional model for $M = 0$. The bead diameters used in the experiments are $d_l = 274 \pm 25 \mu\text{m}$, $d_m = 125 \pm 25 \mu\text{m}$ and $d_s = 50 \pm 25 \mu\text{m}$. The ratios of capillary pressure are $\beta_m = 2.19$ and $\beta_s = 5.48$. The ratios of permeability are $\alpha_m = 0.20$ and $\alpha_s = 0.03$. We see that the invading fluid front in the small permeability layer leads for the arrangements (i) the small permeability layer at the center and (ii) the medium permeability layer at the center, as shown in Fig. 11(a and b), respectively. We see from

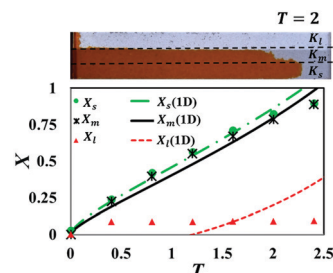


Fig. 12 Dimensionless length vs. time comparison of the experimental observations and one-dimensional model for the arrangement (ii) the medium permeability layer at the center of the three layered porous medium for viscosity ratio $M = 2.6$.

Fig. 11(c) that the fluid front in the medium permeability layer leads as $[\alpha_m(\beta_m - 1)] - [\alpha_s(\beta_s - 1)] > 0$ for the bead diameters used. Fig. 12 compares spontaneous imbibition for the arrangement (ii) the medium permeability at the center, for $M = 2.6$. Here, $d_l = 274 \pm 25 \mu\text{m}$, $d_m = 175 \pm 25 \mu\text{m}$ and $d_s = 125 \pm 25 \mu\text{m}$. The ratios of capillary pressure are $\beta_m = 1.35$ and $\beta_s = 1.4$. The ratios of permeability are $\alpha_m = 0.40$ and $\alpha_s = 0.20$. We see from Fig. 11 and 12 that the one-dimensional model accurately predicts the experimentally observed imbibition dynamics. Thus, in this study, we have developed a model which can accurately predict the front advancement in a layered porous medium given the characteristics of the layers.

5 Conclusions

We have analyzed the capillary driven displacement of a viscous non-wetting fluid with a viscous wetting fluid in a layered porous medium. In the two layered porous medium, the fluid front in the high capillary pressure layer will always lead for a favorable viscosity ratio. In the three layered porous medium, three arrangements are possible. For two arrangements, the front in the small permeability and high capillary pressure layer leads, followed by the front in the medium permeability layer, followed by the front in the large permeability layer. However, when the large permeability layer is in the middle, then the pressure in the wetting phase in all the layers does not equilibrate. This leads to an anomalous behavior and, in many cases, the front in the medium permeability layer leads. We developed a quasi one-dimensional model which excellently mimics the imbibition behavior in a layered porous medium. Using this reduced order model, we could explain the scaling law that governs the anomalous behavior of the front displacement in the three layered porous medium. We also show that the model for a homogeneous porous matrix is unable to estimate the volume imbibed in a layered porous medium.

The one-dimensional model can be generalized for multiple layers of an oil reservoir, ground water remediation and water aquifers. The model and the results of this study can be used in optimizing the layer properties for multi-layered micro reactors, lab-on-chip devices, and membranes of poly-electrolyte membrane fuel cells.

Conflicts of interest

There are no conflicts to declare.

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