

DYNAMICS OF FORCED IMBIBITION IN INTERACTING PORES

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ABSTRACT

Imbibition of viscous fluids in capillaries is important in diagnostics, design of microfluidic devices and enhanced oil recovery. The imbibition of a viscous wetting fluid in a capillary follows Lucas-Washburn law. The Lucas-Washburn regime is only observed when the viscous forces are balanced by the capillary forces. This has been previously described for capillary driven flow as a function of the Ohnesorge number(Oh), the length imbibed by the fluid(x) and the radius(r), for a capillary initially filled with fluid of negligible viscosity, i.e., $Oh(\frac{x}{r}) \sim 1$. We show using VOF simulations that, in a capillary of length L initially filled with a viscous fluid, the modified Lucas-Washburn law is observed only if the criterion $Oh(\frac{L}{r}) \sim 1$ is fulfilled. We use VOF simulations to show the deviation of capillary driven flow from the classical Lucas-Washburn behavior for $Oh(\frac{L}{r}) \sim 0.1$. VOF simulations for forced imbibition in the regime preceding the Lucas-Washburn regime for a single capillary show that with increase in the applied pressure, the advancement of the meniscus is faster. Forced imbibition dynamics in the interacting capillary geometry are also investigated in this study using VOF simulations. We observe that the leading meniscus in the interacting capillaries is significantly dependent on the applied pressures. We also show using VOF simulations that the wettability of the imbibing fluid plays a crucial role in determining the dynamics in an interacting capillary system.

NOMENCLATURE

ρ density of the fluid
 μ viscosity of the fluids

P pressure
 $\mathbf{v}$ velocity vector
 $\mathbf{F}$ Force due to capillary pressure
 α volume fraction of the wetting phase
 γ interfacial tension
 $\mathbf{n}$ vector normal to the interface
 κ curvature of the interface
 $\hat{\mathbf{n}}$ unit vector normal to the interface
 $\hat{\mathbf{n}}_1$ unit vector normal to the capillary wall
 $\hat{\mathbf{n}}_2$ unit vector tangential to the capillary wall
 t time
 x interface location from the capillary inlet
 r radius of the capillary
 L total length of the capillary
 θ contact angle of the invading fluid with the capillary wall
 r radius of the capillary
 ρ_{nw} density of the resident fluid
 ρ_w density of the invading fluid
 μ_{nw} viscosity of the resident fluid
 μ_w viscosity of the invading fluid
 P_c capillary pressure
 Oh Ohnesorge number

1 INTRODUCTION

Imbibition of viscous fluids in narrow pore spaces is important in understanding the ascent of cell sap in trees [1, 2], oil recovery from reservoirs [3–6] and in performing bioassays [7, 8]. The imbibition in a cylindrical capillary was first described by Lucas [9] and Washburn [10]. For a capillary initially filled with

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air, the imbibition dynamics are diffusive, where the distance impregnated by the invading phase is proportional to the square root of time. The Lucas-Washburn law is obtained from the Hagen-Poiseuille's law and is expressed as $x^2 = \left(\frac{r^2 P_c}{4\mu}\right)t$, where r is the capillary radius, P_c is the capillary pressure at the meniscus, μ is the viscosity of the invading fluid and x is the advancement of meniscus with time t . The Lucas-Washburn law states that the distance imbibed by the invading fluid in a capillary of larger radius is more than the small radius capillary. It was observed that the imbibition dynamics in pore spaces of varying geometry deviates from the classical Lucas-Washburn law. The studies by Reyssat et al., [11], Gorce et al. [12], Bartley and Ruth [13] show that capillary driven flow in irregular geometries cannot be predicted by the Lucas-Washburn's law, due to the geometric variations of the capillaries.

During the imbibition phenomenon in a capillary initially filled with a negligible viscosity fluid, the viscous forces in the early regimes are negligible. In this regime, the inertial and capillary forces counter balance each other. Das et al., [14] explained that the classical Washburn's regime is observed for $Oh\left(\frac{x}{r}\right) \sim 1$, where Oh is the Ohnesorge number, which is the ratio of viscous to capillary forces. For $Oh\left(\frac{x}{r}\right) \ll 1$, the viscous forces are negligible. Usually for most of the commonly used liquids, for $Oh\left(\frac{x}{r}\right) \sim 1$, the ratio $\frac{x}{r}$ should be approximately 100 to 1000 so that the Washburn regime is observed [14]. However, naturally occurring porous media consists of interacting pores having different radii and cross-sections. The design of lab-on-chip devices and microfluidic devices also involves capillaries where, the ratio $\frac{x}{r}$ is less than 100. In such cases, the Lucas-Washburn law cannot predict the imbibition dynamics in the capillaries. Therefore, understanding the imbibition dynamics preceding the Lucas-Washburn regime in a capillary is important in microfluidic applications.

In this work, we use Computational Fluid Dynamics (CFD) to understand the imbibition dynamics in capillaries. Using Ansys Fluent[®], 19.1, we first validate the Volume of Fluid (VOF) simulation in a single capillary for $Oh\left(\frac{x}{r}\right) \sim 1$. We show that the imbibition dynamics from the VOF simulations excellently agrees with the modified Lucas-Washburn law. Then, using VOF simulations, we predict the imbibition behavior in a single capillary for $Oh\left(\frac{x}{r}\right) < 1$, to understand the flow dynamics before the Lucas-Washburn regime. We show that the capillary driven flow for a single capillary deviates from the modified Lucas-Washburn law. We also use VOF simulations in a single capillary for $Oh\left(\frac{x}{r}\right) < 1$ to understand the effect of pressure applied at the inlet. Then, we use a two interacting capillary system to understand the imbibition dynamics for complex geometries. By considering different cases of forced imbibition, we show that the imbibition dynamics in the capillaries changes significantly with change in the inlet pressure. For low inlet pressures, we observe that the small radius capillary will have the leading meniscus.

With increase in the inlet pressure, the meniscus in the small radius capillary initially leads, but at later times, the meniscus in the large radius capillary leads. The time at which this cross-over phenomena is observed is found to decrease with increase in the inlet pressure. We also show the affect of wettability of the imbibing fluid and observe that the imbibition dynamics in the interacting capillary system significantly changes with a change in wettability.

2 VOF SIMULATIONS

Volume of Fluid (VOF) method solves the volume averaged Navier-Stokes equation for multiphase flow. This method, introduced by Hirt and Nicholas [15], tracks the fluid-fluid interface by determining the volume fraction of each phase in a grid block. Here, we consider a capillary of radius r and length L , initially filled with a non-wetting fluid of viscosity μ_{nw} and density ρ_{nw} . When the inlet end of the capillary is brought in contact to a wetting fluid of viscosity μ_w and density ρ_w , the wetting fluid begins to imbibe into the capillary. The outlet end of the capillary is considered to be in contact with the non-wetting phase to exclude the capillary end effects. The contact angle of the imbibing wetting fluid with the capillary wall is θ . The capillary geometry is modeled by solving the Navier-Stokes equation and the equation of continuity shown in eq (1) and (2). By solving these equations, the velocity and the pressure of the volume averaged mixture of the fluids is obtained.

$$\frac{\partial}{\partial t}(\rho \mathbf{v}) + \nabla \cdot (\rho \mathbf{v} \mathbf{v}) = -\nabla P + \nabla \cdot [\mu (\nabla \mathbf{v} + (\nabla \mathbf{v})^T)] + \mathbf{F}, \quad (1)$$

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \mathbf{v} = 0, \quad (2)$$

where, ρ is the density, μ is the viscosity of the mixture, $\mathbf{v}$ is the velocity vector and P is the pressure. The term describing the force due to capillary pressure is $\mathbf{F}$ in eq (1), which is due to the surface forces. The volume fraction of grid cells occupied by the imbibing wetting phase are considered to be unity, whereas the volume fraction of the cells filled with the non-wetting phase are considered to be zero. All the cells except the cells at the interface are filled with completely wetting or non-wetting fluid. Therefore, the density and viscosity for the cells at the interface is determined as,

$$\rho = (1 - \alpha)\rho_{nw} + \alpha\rho_w, \quad (3)$$

$$\mu = (1 - \alpha)\mu_{nw} + \alpha\mu_w, \quad (4)$$

where, α is the volume fraction of the wetting phase, which is solved using the phase continuity equation as,

$$\frac{\partial}{\partial t}(\alpha \rho_w) + \nabla \cdot (\alpha \rho_w \mathbf{v}) = 0, \quad (5)$$

The volumetric capillary force $\mathbf{F}$ in eq (1) is computed using the Continuum Surface Force(CSF) method proposed by Brackbill et al. [16]. In this model, the capillary force is a function of the curvature of the fluid-fluid interface in the capillary, given as,

$$\mathbf{F} = -\gamma \kappa \mathbf{n}. \quad (6)$$

Here, γ is the interfacial tension between the wetting and the non-wetting phases, $\mathbf{n}$ is the vector normal to the interface defined as $\mathbf{n} = \nabla \alpha$ and κ is the curvature of the interface calculated as,

$$\kappa = \frac{1}{|\mathbf{n}|} \left[\left(\frac{\mathbf{n}}{|\mathbf{n}|} \cdot \nabla \right) |\mathbf{n}| - \nabla \cdot \mathbf{n} \right]. \quad (7)$$

The unit normal vector $\hat{n}$ is calculated at the wall boundary by CSF method as,

$$\hat{n} = \hat{n}_1 \cos \theta + \hat{n}_2 \sin \theta, \quad (8)$$

where, θ is the contact angle, $\hat{n}_1$ and $\hat{n}_2$ are unit vectors normal and tangential to the wall, respectively.

To initiate the imbibition in the geometry considered here and to keep the inlet end of the capillary in contact with the wetting fluid, all the cells at the inlet boundary are considered to be occupied with the wetting phase, i.e., the volume fraction of the cells at the inlet is one. For the formulation used in this study, eq (5) is first solved implicitly and the value of α is then used in eqs (1) and (2), which solves for P and $\mathbf{v}$. The coupling of P and $\mathbf{v}$ is carried out using PISO (Pressure Implicit with Splitting Operators) algorithm by Ubbink and Issa [17]. A second order upwind scheme and Iterative Time Advancement Scheme available in Fluent are used to implicitly solve the unsteady state equations. A piecewise linear approach, called as the Geo-reconstruct is used to reconstruct the fluid-fluid interface. The geometric construction and meshing of the capillaries is done in ANSYS Workbench 19.1. The geometry was spatially discretized using 300,000 and 600,000 quadrilateral elements to study the effect of grid resolution on the dynamics of interface position. The spatially discretized geometry is shown in Figure 1(a). Figure 1(b) shows that the grid with 300,000 elements is sufficient to model the two phase phenomena.

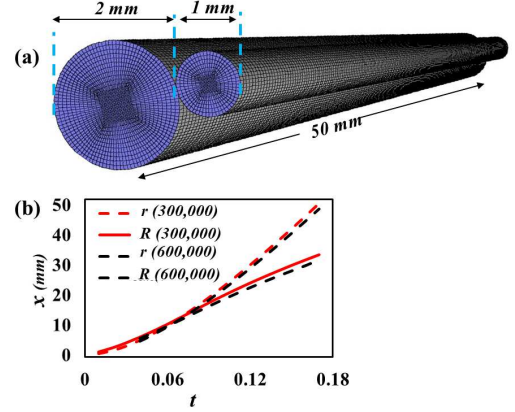


FIGURE 1: (a) The spatially discretized geometry, (b) Distances imbibed with time for 300,000 and 600,000 grid cells for an applied pressure of 420 Pa.

3 VALIDITY OF VOF SIMULATIONS

We now consider a single capillary of radius $r = 0.5$ mm and length $L = 100$ mm. The viscosity of the invading and the residing fluid are same, i.e., $\mu_{nw} = \mu_w = 0.001$ Pa.s. Similarly, we consider the invading and the residing fluids to have same density, i.e., $\rho_{nw} = \rho_w = 1000$ kg.m⁻³. The capillary is meshed into 225×531 grid cells in cross-sectional and axial directions, respectively. Initially, the capillary is filled with the resident fluid. The invading fluid at the inlet end is spontaneously driven into the capillary, displacing the resident fluid. The contact angle of the imbibing fluid with the capillary wall $\theta = 0^\circ$. The interfacial tension is 0.05 Nm⁻¹. The Volume of Fluid(VOF) simulation solves for the advancement in interface of the two fluids with time.

To validate the VOF simulations in a single capillary, we compare the numerical model with the modified Washburn's law, where the resident fluid is viscous. In Figure 2, we show a comparison of the meniscus displacement with time from the VOF simulations with the analytical solution given by Hultmark et al. [18]. The length traveled by the meniscus, x with time t is given by,

$$(\mu_w - \mu_{nw})x^2 + 2\mu_{nw}Lx = \frac{c\gamma r}{4}t. \quad (9)$$

where, $c = 2 \cos \theta$. As $\mu_{nw} = \mu_w = \mu$, in our simulations, eq (9) reduces to, $x = \frac{\cos \theta \gamma}{4\mu L}rt$. We see that VOF excellently simulates the flow in a single capillary, as shown in Figure 2.

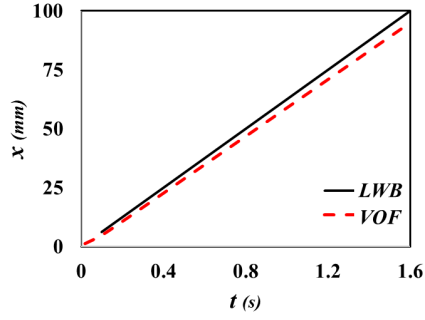


FIGURE 2: Comparison of VOF simulations and theoretical results from modified Washburn's law for distance vs time for spontaneous imbibition in a single capillary of length $L = 100$ mm and radius $r = 0.5$ mm. LWB is the Lucas-Washburn solution, which is compared to the Volume of Fluid (VOF) simulation.

For this case, we see that the viscous forces remain same in the capillary as the viscosity of both the invading and residing fluids is same throughout the imbibition phenomena. Therefore, the viscous forces cannot be neglected at the start of imbibition. We now determine the factor $Oh\left(\frac{x}{r}\right)$ as a criterion to determine the flow regime. As the viscous forces act along the entire length of the capillary L at all times, we find that $Oh\left(\frac{x}{r}\right)$ is of the order 1, for $x = L$. Therefore, the Washburn regime is observed for the parameters used in the VOF simulation.

4 IMBIBITION BEFORE THE WASHBURN REGIME

We now consider a capillary of length $L = 50$ mm to understand the flow phenomena in the regime preceding the Washburn regime. We consider the viscosity and densities of the invading and residing fluids same as the earlier case and demonstrate the effect of applied inlet pressure for a fine mesh geometry of length $L = 50$ mm and radius $r = 0.5$ mm. For this case, $Oh\left(\frac{x}{r}\right)$ is of the order 0.1 for $x = L$, indicating that the Washburn's regime could never be observed in this capillary. We consider $x = L$ because, Das et al., [14] used $Oh\left(\frac{x}{r}\right)$ where, x is the length of the viscous fluid in the capillary. In our case, we are using a viscous resident fluid which has viscosity of the order of the imbibing fluid. Therefore, the length of the viscous fluid will always be L .

We show in Figure 3 the VOF simulation for the capillary driven flow in the regime preceding the Washburn's behavior, i.e., for $Oh\left(\frac{L}{r}\right) \sim 0.1$. We also show in Figure 3 the dynamics predicted by Lucas-Washburn's law for comparison. We see that the imbibition observed from the VOF simulation is slower than the imbibition given by Lucas-Washburn law. Therefore, we show that if $Oh\left(\frac{L}{r}\right) \sim 1$ is not satisfied, the imbibition dynamics

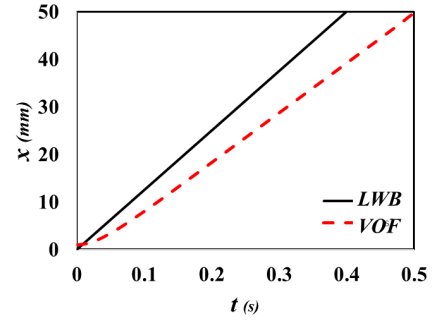


FIGURE 3: Comparison of VOF simulations and theoretical results from modified Washburn's law for distance vs time for spontaneous imbibition in a single capillary of length $L = 50$ mm and radius $r = 0.5$ mm. LWB is the Lucas-Washburn solution, and VOF is the imbibition from the Volume of Fluid (VOF) simulation.

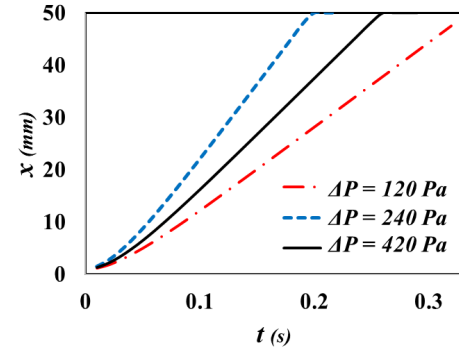


FIGURE 4: Distance vs time plot for forced imbibition in a single capillary of length $L = 50$ mm and radius $r = 0.5$ mm for applied pressures of (a) 120 Pa, (b) 240 Pa and (c) 420 Pa.

cannot be predicted using Washburn's law.

We now use VOF simulations to understand the imbibition phenomena when pressure is applied at the inlet. The properties of the invading and the residing fluids are same as described earlier. Three cases of applied pressure are considered, namely, 120 Pa, 240 Pa and 420 Pa. We show in Figure 4 that the imbibition rate in the single capillary increases with increase in the applied pressure.

5 IMBIBITION IN INTERACTING CAPILLARIES

We now construct a geometry of two capillaries that hydrodynamically interact along their axes. The length of the capillaries is $L = 50$ mm and the radii of the capillaries are $r = 0.5$ mm, referred as the small capillary, and $R = 1$ mm, referred as the large capillary. The cross-section of the geometry is shown in Figure 5(a). For the interacting capillaries, the viscosity of the non-wetting fluid is $\mu_{nw} = 0.002$ Pa.s while the wetting fluid has a viscosity of $\mu_w = 0.001$ Pa.s. The density of both the wetting and non-wetting fluids is 1000 Kg.m^{-3} . The interfacial tension is 0.05 Nm^{-1} . We consider that the imbibing fluid is wetting and makes a contact angle $\theta = 0^\circ$ with the capillary wall. To initiate

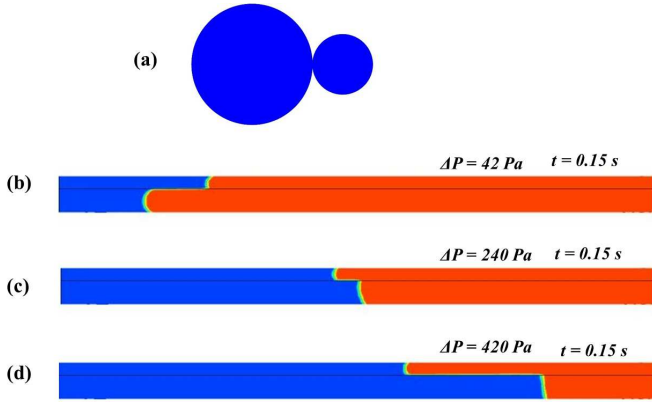


FIGURE 5: (a) The cross-section of the interacting capillary system. The interface locations in the VOF simulations at 0.15 s for applied pressures of (b) 42 Pa, (c) 240 Pa and (d) 420 Pa.

the imbibition, the inlet end of the interacting capillary system is placed in contact with the wetting fluid while the outlet end is in contact with the non-wetting fluid. The wetting fluid is driven into the capillaries by applying a pressure at the inlet. The imbibing fluid flows at a higher flow rate in the capillaries as compared to the spontaneous imbibition in the interacting capillary system as described by Ashraf et al., [19]. The interface locations at 0.15 s for 42 Pa, 240 Pa and 420 Pa are shown in Figure 5 (b), (c) and (d), respectively. We see that an increase in applied pressure causes an increase in the imbibed distance at the same time.

The capillary pressure in the small and the large capillaries, determined using Young-Laplace equation are 200 Pa. and 100 Pa., respectively. In Figure 6(a), we show the interface advancement with time for 42 Pa of applied pressure. We see that the interface in the small capillary leads at all times. For low applied pressures, as the capillary pressures are greater than the applied pressure, the meniscus in the small radius capillary advances faster. This is similar to the qualitative imbibition behav-

ior of spontaneous imbibition in interacting capillaries described by Ashraf et al., [19]. For the case where 240 Pa pressure is applied, the interface in the small capillary leads initially, but, at later times, the imbibition in the large radius capillary leads, as shown in Figure 6(b). As the applied pressure at the inlet increases, the viscous resistance in the capillaries also increases. When a pressure 240 Pa. is applied, which is comparable to the capillary pressure of small radius capillary, the interface in the small radius capillary initially leads. However, the increase in viscous resistance with time in the small radius capillary causes a decrease in the imbibition rate, which can be observed from Figure 6(b). This causes the interface in the large radius capillary to advance faster at later times. In Figure 6(c), we show the imbibition in the interacting capillary system for an applied pressure of 420 Pa and we see that the interface in the large capillary leads at all times. When the applied pressure is greater than the capillary pressures of both the capillaries, i.e., 420 Pa., the high viscous resistance in the small radius capillary reduces the advancement of meniscus significantly and therefore, the interface in the large radius capillary advances faster. Therefore the interface dynamics in the interacting capillary system are strongly dependent on the pressure causing the flow.

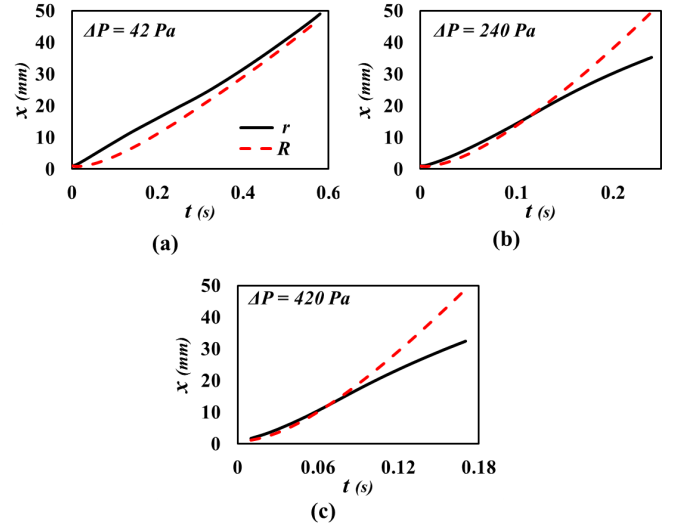


FIGURE 6: Distance vs time for forced imbibition in an interacting capillary system for applied pressures of (a) 42 Pa, (b) 240 Pa and (c) 420 Pa.

5.1 EFFECT OF WETTABILITY

As the flow dynamics is significantly affected by the applied pressure, we now show the impact of wettability during forced imbibition in the interacting capillary system. Here, the applied pressure is 240 Pa and we consider contact angles of the wetting fluid with the capillary wall to be θ as 0° , 60° , 90° and 120° . The viscosity and density of the wetting and non-wetting phases are $0.001 \text{ Pa}\cdot\text{s}$ and $1000 \text{ Kg}\cdot\text{m}^{-3}$, respectively. We show in Figure 7(a) that, for $\theta = 0^\circ$, the interface in the small capillary leads at all times. For this case, the capillary pressures in the small and the large capillaries are 200 Pa. and 100 Pa., respectively. For $\theta = 60^\circ$, the capillary pressures in the small and the large capillaries become 100 Pa. and 50 Pa., respectively. Here, the applied pressure is more than the capillary pressure in the small capillary. We observe that interface in the small capillary leads initially due to the high applied pressure, but at later times the viscous resistance in the small capillary increases. This causes the interface in the large capillary to advance faster at later times, as shown in Figure 7(b). As the contact angle increases to $\theta = 90^\circ$, the leading meniscus is in the large capillary, as shown in Figure 7(c). For this case, the contact angle causes the capillary pressures in both the small and the large capillaries to be zero. The high viscous resistance in the small capillary decreases the rate of meniscus advancement and therefore, the meniscus in the large capillary leads. In Figure 7(d) we show the imbibition for $\theta = 120^\circ$, which implies that the invading fluid is non-wetting, which is a special case of drainage. For this case, the capillary pressures in the small and the large capillaries are -100 Pa. and -50 Pa. , respectively. This implies that the net pressure at the inlet is 140 Pa. for small capillary and 190 Pa. for the large capillary. As the viscous resistance is high in the small capillary and the pressure causing the force is also lower and the imbibition rate significantly decreases. Therefore, the meniscus in the large capillary leads while the small capillary meniscus has a very little advancement with time. Therefore, the wettability of imbibing fluid plays a crucial role in the flow dynamics of an interacting capillary system.

6 CONCLUSIONS

In summary, we use VOF simulations to understand the flow in capillaries. We first consider a single capillary of length $L = 100 \text{ mm}$, where $Oh(\frac{L}{r}) \sim 1$, so that the Washburn regime can be determined. The wetting and non-wetting fluids have a viscosity of 0.001 cP . We compare the VOF simulation with the modified Lucas-Washburn model and show that VOF simulations can accurately predict the flow phenomena in a single capillary. We then use a capillary of length $L = 50 \text{ mm}$, where $Oh(\frac{L}{r}) \sim 0.1$, which is the regime preceding the Lucas-Washburn solution. We show that the Lucas-Washburn model cannot be applied for $Oh(\frac{L}{r}) < 1$. We perform simulations in a single capillary for three different cases of applied pressure and

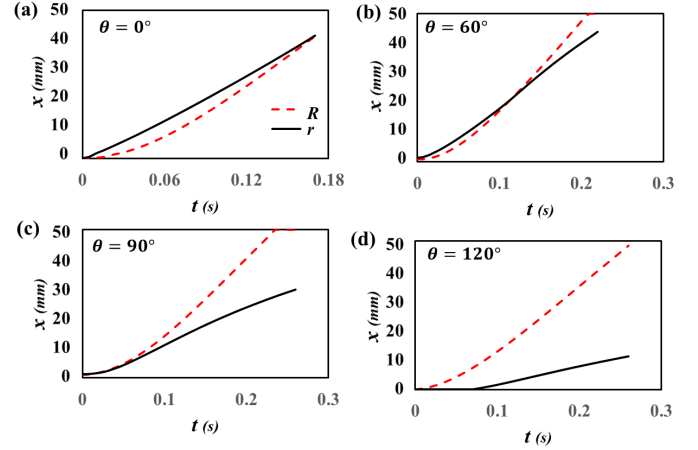


FIGURE 7: Distance vs time for forced imbibition in an interacting capillary system for applied pressures of 240 Pa for contact angles (a) $\theta = 0^\circ$, (b) $\theta = 60^\circ$, (c) $\theta = 90^\circ$ and (d) $\theta = 120^\circ$.

show that with the increase in the applied pressure, the interface in the capillary advances faster.

Then, we consider an interacting capillary system consisting of two capillaries that hydrodynamically interact with each other. We simulate the interacting capillary system for three cases of applied pressures and show different flow phenomena. For the simulations in interacting capillaries, the wetting and non-wetting fluids have viscosity of 0.001 cP and 0.002 cP , respectively. For low applied pressures, the interface in the small radius capillary advances faster at all times. As the applied pressure increases, the small radius capillary has the leading interface initially. However, the large radius capillary will have the leading interface at later times. It is observed that with further increase in applied pressure, the interface in the large radius capillary advances faster at all times.

We also explore the effect of wettability by controlling the contact angle of the imbibing fluid with the capillary wall. The wetting and non-wetting fluids have a viscosity of 0.001 cP . We see that when the imbibing fluid is perfectly wetting, the imbibition is faster in the small radius capillary. We show that as the wettability of the imbibing fluid decreases, the imbibition is initially faster in the small radius capillary but at later times the meniscus in the large radius capillary advances faster. For the case where the contact angle defined the imbibing fluid to be non-wetting, the meniscus in the large radius capillary advances faster at all times. This study provides a basis for understanding the complex imbibition processes occurring at the pore scale.

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