Absorption and Spreading of a Liquid Droplet Over a Thick Porous Substrate

Rachid Chebbi*

ABSTRACT: Spreading over porous substrates occurs in several processes including printing, cleaning, coating, and manufacturing of ceramic structures. For small drops, viscous and capillary forces are ultimately the predominant forces. The process typically undergoes three phases: a first stage in which the droplet spreads, a second phase in which the area of contact with the solid substrate nearly remains constant, and a third stage in which the droplet retracts with its volume reaching zero finally. The objective of the investigation is to find the dynamics of spreading and absorption of the droplet using fundamentals while making relevant approximations to account for both radial and vertical dynamics. The proposed model requires minimal computational work. The results are compared with the published experimental data for the perfect wetting case, and are found to be in good agreement with detailed published experimental data for both droplet dynamics and dynamics of penetration in the porous substrate.

1. INTRODUCTION

The dynamics of spreading and penetration is important in a number of cases including ink-jet and 3D-printing, clean-up processes, coating of porous substrates, irrigation, and manufacture of ceramic structures.1−3 In ink-jet printing, spreading produces blurring and in clean-up processes penetration should be limited.4 The dynamics of spreading over an impermeable wall are reviewed in Rosenholm and the references therein. For the case of perfect wetting, the works of Chebbi and Selim5 and Chebbi,6 extending those of Starov et al.7 and Alleborn and Raszillier.9 The actual contact angle is assumed to be given by the molecular-kinetic theory in Clarke et al.11 and a modified Hoffman–Voinov–Tanner law in Hilpert and Ben-David.12 To model the flow near areas along with the contact angles of the droplet and the imbibed part. The results presented in a dimensionless form show very close profiles for different viscosities of the silicon oil used with the same glass filter material, and for different materials (metal and glass filters) having nearly the same porosity and average pore size for using the same silicon oil viscosity. Differences in the dimensionless curves for the radii of the droplet contact area and the wetted circle at the interface between air and the wetted part of the porous substrate are presented.

Based on the scope of the present work addressing wetting of porous media by a perfectly wetting liquid, the emphasis is on spreading and imbibition of thick and initially dry porous substrates. Different models have been used. They differ in the scope of the work, assumptions used, and simplifications included to allow for more analytical treatment. The dynamics have two competing processes: spreading and penetration. As far as spreading is concerned, the lubrication approximation requiring a small contact angle is used in Davis and Hocking,1 and Alleborn and Raszillier.9 The actual contact angle is assumed constant in Davis and Hocking, and the apparent contact angle is assumed to be given by the molecular-kinetic theory in Clarke et al.11 and a modified Hoffman–Voinov–Tanner law in Hilpert and Ben-David.12 To model the flow near
the contact line, the precursor film and disjoining pressure concepts are used in Alleborn and Raszillier, and the no-slip boundary condition is adopted in Davis and Hocking. As far as the dynamics of penetration is concerned, the flow is modeled as occurring in vertical capillary tubes. The Washburn (also called Lucas–Washburn) equation is used in Holman et al. and Denesuk et al., and Darcy’s law is utilized in Alleborn and Raszillier and Clarke et al. Denesuk et al. considered two cases, CDA and DDA, and Holman included the IDA case. The solutions for the contact area radius and the droplet contact angle in Clarke et al. were found to depend on a friction parameter, with the advancing contact angle ranging between 38.9 and 51.9°, and a third parameter depending on the porous substrate. The three parameters were determined to get the best fit with the experimental data obtained. The power law of a linear function of time for the contact area radius was found by curve fitting against experimental data in Holman et al. The work of Hilpert and Ben-David for the wetting case (excluding perfect boundary condition is adopted in Davis and Hocking. As far as validated against extensive data and conclusions are validated against extensive data and conclusions are

2. COMPUTATIONAL METHODS

The dynamics is subdivided into three phases. First, spreading occurs along with penetration of the liquid into the porous substrate. In the second phase, the droplet radius nearly remains constant. In the last phase, the droplet retracts. There is a continuous loss of mass by continuing penetration during the three phases till the mass of the droplet reaches zero.

2.1. Penetration in a Capillary Tube. Flow in small capillaries involves both inertia forces and viscous forces initially, with viscous forces remaining as the ultimate dominating resisting force in the remaining process. The Lucas–Washburn equation (also called Washburn equation) assumes a constant contact angle, which is strictly valid at small contact line velocities. The equation provides the penetration depth $x$ into one straight capillary tube as

$$x = \sqrt{\frac{a \sigma \cos \alpha}{2 \mu}} t$$

where $a$ is the inner radius of the capillary tube and $\alpha$ is the static contact angle. A treatment involving the effect of the dynamic contact angle on the dynamics of penetration shows excellent agreement with the published experimental data. The use of the concept of disjoining pressure shows excellent agreement with the experimental data for the relation between capillary number and dynamic contact angle in the case of perfect wetting case.

2.2. Porous Medium Permeability. The porous substrate is characterized by its permeability defined by Darcy’s equation for the superficial liquid velocity $v_s$ (volumetric flow rate per unit area of porous medium) as a function of the pressure gradient $dp/dx$

$$v_s = -\frac{k}{\mu} \frac{dp}{dx}$$

In the case of penetration, the pressure gradient is obtained from the Young–Laplace equation as

$$-\frac{dp}{dx} = \frac{2 \sigma \cos \alpha / a}{x}$$

To account for the porosity $\phi$ of the bed, we have

$$\phi \frac{dx}{dt} = \frac{k}{\mu} \frac{2 \sigma \cos \alpha / a}{x}$$

Integrating eq 4 and comparing the expression for $x(t)$ with eq 1 provides the following expression for $k$

$$k = \frac{a^2 \phi}{8}$$

The result is in agreement with ref 9.

Using the above expression for $k$ along with the static contact angle approximation, the liquid depth takes the following form, consistent with ref 3

$$x = \zeta \sqrt{t} \quad \zeta = \sqrt{\frac{4 \kappa \sigma \cos \alpha}{\mu a \phi}}$$

2.3. Relations Between Apex, Radius, and Volume. In the present work, the liquid droplet and the volume of the porous medium imbibed by the liquid are both assumed spherical in shape. The following relation applies to the spherical cap volumes of apex $h_a$ and radius $L$

$$h_a^3 + 3L^2h_a = \frac{6V}{\pi}$$

The following relations including the radius of curvature $R_c$ and the contact angle $\theta$ can be easily derived
2R₀ = hₐ + \frac{L^2}{hₐ} \tag{8}

\cos \theta = 1 - \frac{h₁}{R₀} \tag{9}

In the case of thin droplets, the spherical cap can be approximated as a paraboloid. The angle and apex expressions can be simplified and eqs 7–9 reduce to

\[ L^2hₐ = \frac{2V}{\pi} \tag{10} \]

\[ 2R₀ = \frac{L^2}{hₐ} \tag{11} \]

\[ \frac{\theta^2}{2} = \frac{hₐ}{R₀} \tag{12} \]

In the present treatment, the general equations 7–9 are used for the droplet. The approximations 10 and 11 can be used for the imbibed part of the porous medium since the contact angles considered are not large.

### 2.4. Combined Capillary Spreading with Penetration into a Permeable Substrate

A schematic figure of a drop spreading on a thick porous medium is given in Figure 1.

![Figure 1. Schematic of a spreading drop on a porous substrate.](image)

The fraction of the porous medium filled with liquid is denoted by ε. Using eqs 5 and 6, the effective \( kₐ \) and \( ζₑ \) values are estimated as

\[ kₐ = k \frac{εₐ}{ε}; \quad ζₑ = Cᵦε \frac{εₐ}{ε} \tag{13} \]

where \( Cᵦ \) is a correction factor. The effective \( ζₑ \) is used instead of \( ζ \) in the following treatment.

Assuming one-dimensional penetration as an approximation, the rate of change in the volume of porous medium imbibed by the liquid is given by

\[ \frac{dVₚ}{dt} = \epsilon_i \int_0^{R} \frac{\partial H}{\partial t}(2\pi r)dr + \epsilon_f \frac{dR}{dt}H(t, R)\frac{(2\pi R)}{2} \tag{15} \]

The last term in eq 15 is zero, as the depth of penetration \( H \) is zero at \( L \).

As in Clarke et al., \(^{11} \) we assume penetration to start at \( τ(r) \) defined as the time at which the leading edge reaches the radial distance \( r \). Using eq 6 along with the unidirectional penetration approximation yields

\[ \frac{\partial H}{\partial t} = \frac{ζₑ}{2\sqrt{t - τ(r)}} \tag{16} \]

Then, substituting into eq 15 while discarding the last term gives

\[ \frac{dVₚ}{dt} = \epsilon_i \int_0^{L} \frac{ζₑ}{2\sqrt{t - τ(r)}}(2\pi r)dr \tag{17} \]

#### 2.4.1. IDA Phase

Spreading over a permeable solid is approximated by the following expression

\[ L = L₀ + \frac{\ln(t/t₀)}{\ln(t_{CDA}/t₀)}(L_{CDA} - L₀) \tag{18} \]

or piecewise by a linear function of \( \ln(t) \) as needed. Using a mass balance, the remaining droplet volume, \( Vᵢ \) and the liquid volume imbibed by the porous substrate, \( Vₚ \), as

\[ V = Vᵢ - Vₚ \tag{19} \]

The values of \( Vₚ \) can be obtained numerically through a series of time steps \( tₙ \left( \text{n starting at 1} \right) \) using the discretized form of eq 17

\[ Vₚₙ = Vₚₙ₋₁ + \Delta t \times \epsilon_i ζₑ \sum_{i=1}^{n} \frac{\pi (L⁻²₋ L⁻²_{i₋₁})}{2 \sqrt{tₙ₋₁ - τₙ₋₁}} \tag{20} \]

Discretizing eqs 18 and 19 provides the radius of the droplet at \( tₙ \)

\[ Lₙ = L₀ + \frac{\ln(tₙ/t₀)}{\ln(t_{CDA}/t₀)}(L_{CDA} - L₀) \tag{21} \]

and the updated value \( Vₙ \)

\[ Vₙ = Vᵢ - Vₚₙ \tag{22} \]

Using eqs 7–9 yields the following expressions for the apex \( hₐ \) and the contact angle \( θ \)

\[ hₐ = \sqrt{\frac{3V}{π} + \frac{9V^2}{π²} + L^6} + \sqrt{\frac{3V}{π} - \frac{9V^2}{π²} + L^6} \]

\[ \theta = \arccos\left(\frac{L² - hₐ²}{L² + hₐ²}\right) \tag{23} \]

Making use of eq 23 along with the updated values \( Lₙ \) and \( Vₙ \) provides the updated values of \( hₐ \) and the contact angle \( θ \).

For small contact angles, the following simpler equations can be used

\[ hₐ = \frac{2V}{πL³}; \quad θ = \frac{4V}{πL³} \tag{24} \]

The imbibed part of the porous medium, \( Vₚ \), is partially filled with air, while the rest, \( Vₚʰ \), is filled with liquid. The volume, \( Vₚʰ \), is obtained as \( Vₚ = Vₚʰ/εₚ \) where \( εₚ \) is calculated as
\[ \psi = \frac{V_1}{\pi R_{\text{max}}^3, \psi_{\text{max}, c}} \times 1/4 \]  

(25)

using the final data for the wetted area radius at the interface between the porous medium and the air and the contact angle of the imbibed part of the liquid.

Using Darcy's law in the horizontal direction yields

\[ \frac{dR}{dt} = \frac{k_e \frac{p_v}{\mu R - L}} \]  

(26)

where the capillary pressure \( p_v \) for zero contact angle is given by

\[ p_v = \frac{2 \sigma}{a} \]  

(27)

Substituting for the capillary pressure \( p_v \) and using the definition of \( \psi \), yields \( R \) by a simple explicit integration of

\[ \frac{dR}{dt} = \frac{\psi^2}{2(2 - \psi^2)} \text{ for } \psi \leq \psi_{\text{max}} \]  

(28a)

where \( \psi_{\text{max}} \) is max \( \psi_v \), with \( \psi_v = 4V_p/(\pi R^3) \), or simple calculation using

\[ R = \left( \frac{4V_p}{\pi \psi_{\text{max}}} \right)^{1/3} \text{ for } \psi = \psi_{\text{max}} \]  

(28b)

The new radius of the imbibed part \( R \) is obtained by a simple explicit integration of eq 27.

The apex \( H_{\text{a}} \) of the imbibed part is obtained by integration of

\[ \frac{dH_{\text{a}}}{dt} = \frac{\zeta_e}{2\sqrt{t}} \]  

(29)

Substituting into eq 30 gives the updated value of the contact angle \( \psi \)

\[ \psi = \max (2H_{\text{a}}/R, \psi_{\text{max}}) \]  

(30)

The IDA phase starts at \( t_0 \) and ends at \( t_{ee} \). The spreading dynamics during this phase is obtained from a fit to experimental data. The contact angle of the droplet and the dynamics of penetration, including the contact angle and radius, are modeled.

2.4.2. CDA Phase. In case an intermediate constant drawing area phase is included in the analysis, \( L \) is approximately constant. Assuming that \( \tau_{sw} \) is small compared to \( t_{ee} \) and integrating eq 37 (derived in Section 2.4.3) yields

\[ V = V_{\text{CDA}} - 2\pi L^2 \zeta_e (\sqrt{t} - \sqrt{t_{\text{CDA}}}) \]  

(31)

For a recommended and more accurate integration, the following discretization equation can be used along with eq 21 to obtain \( V_{\text{pl}} \).

\[ V_{\text{pl}} = V_{\text{pl-1}} + \Delta t \times \frac{\pi (L_i^2 - L_{i-1}^2)}{2 \sqrt{t_i - \frac{1}{4} t_{ee}}} \sum_{i=1}^{N} \]  

(32)

\( L \) does not change during the second phase. Therefore, the summation does not include the terms \( N + 1 - n \) since the corresponding terms are equal to zero.

The values of \( h_{\text{a}} \) and \( \theta \) can be calculated using eqs 23 or 24 for a small contact angle. Using the approximate expression 31, we have for small \( \theta \)

\[ \theta = 4 \frac{V_{\text{CDA}} - 2\pi L^2 \zeta_e (\sqrt{t} - \sqrt{t_{\text{CDA}}})}{\pi L_{\text{CDA}}^3} \]  

(approximate)

Integrating eq 28a yields for \( L \) constant (second phase)

\[ R = L + \sqrt{L^2 + R_{\text{CDA}}^2} - 2LR_{\text{CDA}} + \zeta_e (t - t_{\text{CDA}}) \]  

(34)

To get more accurate results, eq 32 is recommended instead of eq 34, and used in the Results and Discussion Section. Using the volume of the imbibed part \( V_{\text{pi}} \) (obtained from \( V \) and \( R, H_{\text{a}}, \) and \( \psi \) are obtained from eqs 29 and 30).

The time interval for the CDA phase is \( t_{\text{CDA}} - t_{\text{DDA}} \). In the present model, the value of \( t_{\text{DDA}} \), the time at which the CDA phase ends, is postulated to be the time at which \( \psi_{\text{f}} \) reaches \( \psi_{\text{max}} \) as discussed in Section 2.4.1. The value, \( \theta_{\text{SW}} \) as defined in refs 13, 22, is determined at \( t_{\text{CDA}} \).

2.4.3. DDA Phase. The droplet starts receding at a constant angle, \( \theta_{\text{DDA}} \), during the last phase.\(^{13,22} \) In the present model, \( \theta_{\text{DDA}} \) is determined at \( t_{\text{DDA}} = (t_{\text{DDA}} + t_{\text{DDA}})/2 \) and used to determine \( L(t) \) in the last phase as shown later. The average time considered corresponds to the case where the drop dynamics is considered as consisting of two stages only\(^{13,22} \) (rather than three stages), in which the drop initially spreads, reaches a maximum value for \( L(t_{ee} + t_{ee}) \), and then starts retracting at a constant angle. We can write eq 17 in the following form

\[ \frac{dV_{\text{pl}}}{dt} = -\epsilon_1 L^2 \zeta_e \frac{1}{2\sqrt{t - \tau_{sw}(t)}} \]  

(35)

where the average \( \tau_{sw} \) is given by

\[ \frac{1}{\sqrt{t - \tau_{sw}(t)}} = \epsilon_1 \int_0^t \zeta_e \left( \frac{1}{\sqrt{t - \tau(r)}} - \frac{1}{(2\pi)dr} \right) \]  

(36)

In the third phase, \( t \) is large compared to \( \tau_{sw} \). Using this approximation, differentiating the mass balance eq 19 and combining it with eq 35 yields

\[ \frac{dV}{dt} = -\epsilon_1 L^2 \zeta_e \frac{1}{2\sqrt{t}} \]  

(37)

Substituting for \( V \) as a function of the constant contact angle \( \theta_{\text{DDA}} \) and \( L \) yields

\[ \frac{dL}{dt} = -\frac{2\epsilon_1 \zeta_e}{3\theta_{\text{DDA}} L_{\text{DDA}}} \sqrt{t - \tau_{sw}(t)} \]  

(38)

Neglecting \( \tau_{sw} \) and integrating the above equation provides

\[ L = L_{\text{DDA}} + \frac{4\epsilon_1 \zeta_e}{3\theta_{\text{DDA}} L_{\text{DDA}}} (\sqrt{t} - \sqrt{t_{\text{DDA}}}) \]  

(39)

As in Denesuk et al.,\(^{15} \) \( L \) varies as a linear function of \( \sqrt{t} \) in the DDA phase. The droplet radius and the apex \( h_{\text{a}} \) are given by

\[ V = \frac{\pi \theta_{\text{DDA}} L_{\text{DDA}}^3}{4} \]  

(40)

\[ h_{\text{a}} = \theta_{\text{DDA}} \frac{L_{\text{DDA}}}{2} \]  

(41)

The radius of the wetted area at the solid substrate surface \( R \) is determined using eqs 28a and 28b. The volume of the porous medium \( V_{\text{pi}} \) is obtained as \( V_{\text{pi}}/\epsilon_1 \). Using eqs 29 and 30, we can determine the apex \( H_{\text{a}} \) and \( \psi \). The final time reached when \( L \) is zero. Using eq 39 provides
The model results are compared with the experimental data for the case of perfect wetting of silicone oil droplets on thick porous glass filters in Starov et al.13,22 The physical properties and characteristics of the porous medium are presented in Table 1.

The correction factor \(C_f\) for the cases referred to in Table 1 (ref 13) is taken as 0.707 (\(\approx 1/\sqrt{2}\)), assuming the average pore size in ref 13 represents the average pore diameter, which is typically the case, and \(C_f\) is taken as 1 if the average pore size in ref 13 represents the average pore radius, which is less common. In both cases, \(\zeta\) has the same value, and the results obtained and presented below are the same. The values of permeability \(k\) in Table 1 were obtained using the data in Starov et al.13 for air flowing at a flux of 1.9 \(L/(min\ cm^2)\) and a pressure of 0.1 bar across a glass filter of 1.9 mm thick. The fraction of the porous medium filled with the liquid within the imbibed volume, \(\varepsilon_0\) was calculated using eq 25 along with the experimental end values of \(\psi_{\text{run}}\) for Run 2 and 3.50 mm for Run 3. The values of permeability \(k\) were found to be 0.374 and 0.369 for Runs 2 and 3, respectively. The values of \(\zeta\) and \(\zeta_e\) were calculated using eqs 6 and 13 and are shown in Table 1. To calculate \(\zeta\), a surface tension value of 0.02 N/m was estimated.7

The experimental data for \(L(t)\) during the IDA phase are approximated using the following expressions

\[
L(t) = 1.23 + \frac{\log(t/0.04)}{\log(1/0.04)} (2.27 - 1.23), \quad \text{for } 0.04 s \leq t(s) \leq 1 s
\]

Run 2:

\[
L(t) = 1.60 + \frac{\log(t/0.35)}{\log(0.5/0.35)} (2.57 - 1.60), \quad \text{for } 0.35 s \leq t(s) \leq 5 s
\]

Run 3:

Integation was performed using a time step \(\Delta t\) of 0.001 s for Run 2 and 0.01 s for Run 3. The initial conditions were obtained using the initial data at \(t_0 = 0.04 s\) for \(R\) and \(\psi\). The value of \(R\) at \(t_0\) was taken as 1.002 \(L\) to avoid singularity while using eq 28a. The values of \(\psi_{\text{max}}\) are found to be 0.613 rad (39.0 grad) for Run 2 and 0.614 rad (39.1 grad) for Run 3, compared to the experimental data: 39 grad and 42 grad for Runs 2 and 3, respectively (Figure 2).

The results for \(L(t)\) and \(R(t)\) for Runs 2 and 3 are shown in Figures 3 and 4, respectively.

The dotted lines represent the approximate profiles for \(L(t)\) using the above expressions. The results (continuous lines) are found to be in good agreement with the experimental data. The profiles for \(L\) are approximated as flat (CDA phase) over the range 1–4 s for Run 2 and 5–20.9 s for Run 3 (Figures 3 and 4).

The sharp change occurring in the profile for \(R\) is due to the use of eq 28b instead of eq 28a when \(\psi_{\text{max}}\) reaches \(\psi_e\) (Figure 2), at which time \(\psi\) is considered as constant at larger times according to the present model. The change in the trend for the experimental values of \(R\) is more visible in Figure 3.

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Figures 3–6 show that the present results are in good agreement with the experimental data in refs 13, 23. In the case of low viscosity (Run 1, viscosity = 0.05 P), the second stage is fast and is more difficult to observe experimentally.

The droplet profiles and the profiles for the porous medium part \( V_l \) containing the imbibed liquid \( V_p \) in the pores (Figure 7) are obtained using

\[
h = \frac{L^2}{\sin^2 \theta} - \frac{r^2}{2} - \frac{1}{\sin \theta}\cos \theta
\]

(43)

\[
H = \frac{R}{2} \left( 1 - \frac{r^2}{R^2} \right)
\]

(44)

4. CONCLUSIONS

The three phases occurring during the absorption of a liquid over a thick porous medium (increasing, constant, and decreasing drawing area phases) are considered using an analytical model. The present model was developed assuming no or small changes in the properties including surface tension. The present model applies to the case of perfect wetting, small size drops (so that gravity effects can be dropped), and no/low volatility cases. In addition, the model does not include the very first stage of the process as inertia terms are neglected. Both axial and radial dynamics are included in the analysis. The results are found in good agreement with the extensive experimental data for both the drop dynamics and liquid penetration in the perfect wetting case of silicone oil droplets on thick porous glass filters.13,22 The results include the droplet dynamics during the CDA and DDA phases and the dynamics of penetration during the IDA, CDA, and DDA phases. Investigation of the dynamics of spreading of the droplet over thick porous substrates during the first phase (IDA) is recommended in the future.

AUTHOR INFORMATION

Corresponding Author
Rachid Chebbi – Department of Chemical Engineering, American University of Sharjah, Sharjah 26666, United Arab
Complete contact information is available at:
https://pubs.acs.org/10.1021/acsomega.0c05341

Notes
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