Geometrical instability in the imbibition of a sphere
Supplementary Information
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1 List of symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$t$</td>
<td>Time</td>
</tr>
<tr>
<td>$R_0$</td>
<td>Radius of the aggregate</td>
</tr>
<tr>
<td>$\ell_0$</td>
<td>Length of the cylinder</td>
</tr>
<tr>
<td>$R_0$</td>
<td>Radius of the cylinder</td>
</tr>
<tr>
<td>$R$</td>
<td>Radius of the dry core</td>
</tr>
<tr>
<td>$\ell$</td>
<td>Length of the dry droplet in cylinder geometry</td>
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<tr>
<td>$\phi$</td>
<td>Volume fraction</td>
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<tr>
<td>$\epsilon$</td>
<td>Porosity</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>Permeability</td>
</tr>
<tr>
<td>$n$</td>
<td>Refractive index of the solvent</td>
</tr>
<tr>
<td>$\eta$</td>
<td>Viscosity of the solvent</td>
</tr>
<tr>
<td>$P_0$</td>
<td>Atmospheric pressure</td>
</tr>
<tr>
<td>$P_c$</td>
<td>Capillary pressure</td>
</tr>
<tr>
<td>$P_{in}$</td>
<td>Initial pressure in the aggregate</td>
</tr>
<tr>
<td>$R$</td>
<td>Nondimensional radius</td>
</tr>
<tr>
<td>$\Pi$</td>
<td>Nondimensional pressure</td>
</tr>
<tr>
<td>$t$</td>
<td>Nondimensional time</td>
</tr>
</tbody>
</table>

2 Kinetics of imbibition: theoretical description

2.1 Imbibition of a sphere

The air pressure inside the aggregate increases according to the Boyle-Mariotte law:

\[ P_{in}R_0^3 = P_{in}R^3 \]  \hspace{1cm} (1)

where $P_{in}$ is the initial air pressure, $R_0$ the radius of the aggregate and $R$ the radius at time $t$. By equating the charge loss with the difference of pressure, we get:

(1)
Figure 1: Imbibition of a sphere. The wetting fluid (blue) imbibes a sphere of porous material. The air (white) is entrapped at the center of the sphere.

\[ P_0 + \frac{\eta \epsilon R_0^2 \frac{d}{dt} \left( \frac{R}{R_0} - \frac{R^2}{R_0^2} \right)}{\kappa} + P_c = \frac{P_{in}}{R_0^3} \]

(2)

where \( \eta \) is the viscosity of the solvent, \( \epsilon \) the porosity of the medium and \( \kappa \) its permeability. It can be normalized as:

\[ \frac{d\tilde{R}}{dt}(\tilde{R} - \tilde{R}^2) + 1 = \frac{P_{in}}{P_0 + P_c} \frac{1}{\tilde{R}^3} \]

(3)

using the dimensionless quantities:

\[ \tilde{R} = \frac{R}{R_0} \quad \tilde{t} = \frac{\kappa (P_0 + P_c)}{\eta \epsilon R_0^2} t \]

(4)

A stationary state at long time is obtained:

\[ \tilde{R}_{\text{plateau}} = \Pi^{1/3} \]

(5)

where \( \Pi \) is defined as \( \frac{P_{in}}{P_0 + P_c} \). The solution is given by:

\[ \tilde{t} = \frac{1}{6} - \frac{\tilde{R}^2}{2} + \frac{\tilde{R}^3}{3} + \frac{1}{6} \Pi^{2/3} \log \left( \frac{\Pi^{2/3} \tilde{R}^2 + \Pi^{1/3} \tilde{R} + 1}{\Pi^{2/3} + \Pi^{1/3} + 1} \right) \]

\[ - \frac{1}{\sqrt{3}} \Pi^{2/3} \left( \arctan \left( \frac{2\Pi^{1/3} \tilde{R} + 1}{\sqrt{3}} \right) - \arctan \left( \frac{2\Pi^{1/3} + 1}{\sqrt{3}} \right) \right) \]

\[ - \frac{1}{3} \Pi^{2/3} \log \left( \frac{\Pi^{1/3} \tilde{R} - 1}{\Pi^{1/3} - 1} \right) + \frac{1}{3} \log \left( \frac{\Pi \tilde{R}^3 - 1}{\Pi - 1} \right) \]

(6)

This equation has been used to model the evolution of the imbibed radius as a function of time (Fig. 1 of the MS).

The front acceleration is then obtained by derivation of Eq. 4:
\[
\frac{d^2 \hat{R}}{d\hat{R}^2} = \frac{1}{(\hat{R}^2 - \hat{R})^3} \left( 1 - \frac{\Pi}{\hat{R}^3} \right) \left( -\frac{4\Pi}{\hat{R}^3} + \frac{5\Pi}{\hat{R}^2} + 1 - 2\hat{R} \right) \quad (7)
\]

and is plotted in Fig. 2 of the Letter. The first two terms of the product are negative. The sign of the acceleration is thus governed by the last term, and a straightforward analysis shows that the acceleration is positive for a range of \(\hat{R}\) when:

\[
\Pi < \frac{4}{625} = 6.4 \cdot 10^{-3} \quad (8)
\]
2.2 Imbibition kinetics of an infinite cylinder aggregate

Let us now consider an infinite cylinder geometry.

\[
P_0 + \frac{\eta \epsilon}{\kappa} \frac{dH}{dt} (H_0 - H) + P_e = P_m \frac{H_0}{H}
\]

or, with the dimensionless quantities defined in Eq. 4 :

\[
\frac{d\tilde{R}}{dt} \tilde{R} \log \tilde{R} + 1 = \frac{\Pi}{\tilde{R}^2}
\]

The acceleration of the front is then given by :

\[
\frac{d^2 \tilde{R}}{dt^2} = \frac{\left(\Pi \tilde{R}^{-2} - 1\right) \left(3\Pi \tilde{R}^{-2} \log(\tilde{R}) + \Pi \tilde{R}^{-2} - 1 - \log(\tilde{R})\right)}{(\tilde{R} \log(\tilde{R}))^3}
\]

The front accelerates over a range of \( \tilde{R} \) values when :

\[
\Pi < \frac{e^{-8/3}}{9} = 5.5 \cdot 10^{-3}
\]

3 Planar geometry

The pressure balance now becomes :

\[
P_0 + \frac{\eta \epsilon}{\kappa} \frac{dH}{dt} (H_0 - H) + P_e = P_m \frac{H_0}{H}
\]

Defining \( \tilde{H} = \frac{H}{H_0} \) it may be written in dimensionless units :

\[
\frac{d\tilde{H}}{dt} (1 - \tilde{H}) + 1 = \frac{\Pi}{\tilde{H}}
\]
Figure 3: Imbibition of a planar geometry. The wetting fluid (blue) imbibes a plane of porous material. The air (white) in entrapped above the fluid.

and the second derivative writes:

\[
\frac{d^2 \hat{H}}{dt^2} = \frac{\left(\Pi \hat{H}^{-1} - 1\right)\left(-\Pi \hat{H}^{-2} + 2\Pi \hat{H}^{-1} - 1\right)}{(1 - \hat{H})^3} \tag{15}
\]

The sign of \(\frac{d^2 \hat{H}}{dt^2}\) remains constant for \(\hat{H} \in [0, 1]\) and the front never accelerates.