Geometrical instability in the imbibition of a sphere
Supplementary Information

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1 List of symbols

Time	t	
Radius of the aggregate	R_0	$1--3.5$ mm
Length of the cylinder	ℓ_0	7 mm
Radius of the cylinder	R_0	1.5 mm
Radius of the dry core	R	
Length of the dry droplet in cylinder geometry	ℓ	
Volume fraction	ϕ	63 %
Porosity	ϵ	0.36
Permeability	κ	73 nm^2
Refractive index of the solvent	n	1.48
Viscosity of the solvent	η	0.1 Pa.s
Atmospheric pressure	P_0	10^5 Pa
Capillary pressure	P_c	$4.5 \cdot 10^5$ Pa
Initial pressure in the aggregate	P_{in}	10 mbar - 1 bar
Nondimensional radius	$\tilde{R}$	$\frac{R}{R_0}$
Nondimensional time	$\tilde{t}$	$\frac{\kappa(P_0+P_c)}{\eta\epsilon R_0^2}t$
Nondimensional pressure	Π	$\frac{P_{in}}{P_0+P_c}$

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 :

$$P_{in}R_0^3 = P_{in}R^3 \tag{1}$$

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 :

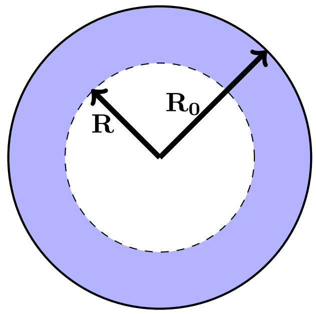

Figure 1: Imbibition of a sphere. The wetting fluid (blue) imbibes a sphere of porous material. The air (white) in entrapped at the center of the sphere.

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

where η is the viscosity of the solvent, ϵ the porosity of the medium and κ its permeability. It can be normalized as :

$$\frac{d\tilde{R}}{d\tilde{t}} (\tilde{R} - \tilde{R}^2) + 1 = \frac{P_{in}}{P_0 + P_c} \frac{1}{\tilde{R}^3} \tag{3}$$

using the dimensionless quantities :

$$\tilde{R} = \frac{R}{R_0} \qquad \tilde{t} = \frac{\kappa (P_0 + P_c)}{\eta \epsilon R_0^2} t \tag{4}$$

A stationary state at long time is obtained :

$$\tilde{R}_{plateau} = \Pi^{1/3} \tag{5}$$

where Π is defined as $\frac{P_{in}}{P_0+P_c}$. The solution is given by :

$$\begin{aligned} \tilde{t} = {} & \frac{1}{6} - \frac{\tilde{R}^2}{2} + \frac{\tilde{R}^3}{3} + \frac{1}{6}\Pi^{2/3} \log \left(\frac{\Pi^{2/3}\tilde{R}^2 + \Pi^{1/3}\tilde{R} + 1}{\Pi^{2/3} + \Pi^{1/3} + 1} \right) \\ & - \frac{1}{\sqrt{3}}\Pi^{2/3} \left(\arctan \left(\frac{2\Pi^{1/3}\tilde{R} + 1}{\sqrt{3}} \right) - \arctan \left(\frac{2\Pi^{1/3} + 1}{\sqrt{3}} \right) \right) \\ & - \frac{1}{3}\Pi^{2/3} \log \left(\frac{\Pi^{1/3}\tilde{R} - 1}{\Pi^{1/3} - 1} \right) + \frac{1}{3}\Pi \log \left(\frac{\Pi\tilde{R}^3 - 1}{\Pi - 1} \right) \end{aligned} \tag{6}$$

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 :

$$\frac{d^2\tilde{R}}{d\tilde{t}^2} = \frac{1}{(\tilde{R}^2 - \tilde{R})^3}\left(1 - \frac{\Pi}{\tilde{R}^3}\right)\left(-\frac{4\Pi}{\tilde{R}^3} + \frac{5\Pi}{\tilde{R}^2} + 1 - 2\tilde{R}\right) \tag{7}$$

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 $\tilde{R}$ when :

$$\Pi < 4/625 = 6.4 \cdot 10^{-3} \tag{8}$$

2.2 Imbibition kinetics of an infinite cylinder aggregate

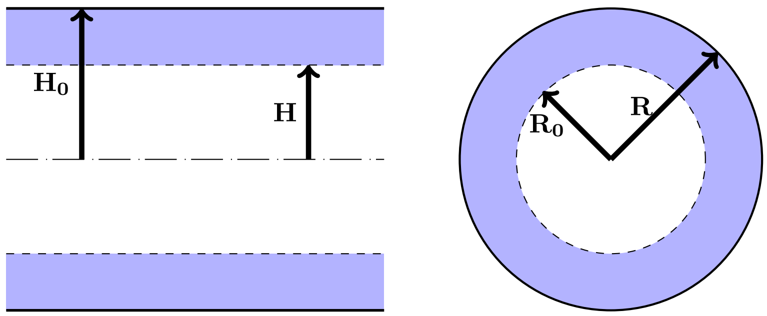

Figure 2: Imbibition of an infinite cylinder. The wetting fluid (blue) imbibes a cylinder of porous material. The air (white) in entrapped at the center of the cylinder.

Let us now consider an infinite cylinder geometry.

$$P_0 + \frac{\eta\epsilon}{\kappa}\frac{dR}{dt}R\log\left(\frac{R}{R_0}\right) + P_c = P_{in}\frac{R_0^2}{R^2} \tag{9}$$

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

$$\frac{d\tilde{R}}{d\tilde{t}}\tilde{R}\log\tilde{R} + 1 = \frac{\Pi}{\tilde{R}^2} \tag{10}$$

The acceleration of the front is then given by :

$$\frac{d^2\tilde{R}}{d\tilde{t}^2} = \frac{\left(\Pi\tilde{R}^{-2} - 1\right)\left(3\Pi\tilde{R}^{-2}\log(\tilde{R}) + \Pi\tilde{R}^{-2} - 1 - \log(\tilde{R})\right)}{(\tilde{R}\log(\tilde{R}))^3} \tag{11}$$

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

$$\Pi < \frac{e^{-8/3}}{9} = 5.5\cdot 10^{-3} \tag{12}$$

3 Planar geometry

The pressure balance now becomes :

$$P_0 + \frac{\eta\epsilon}{\kappa}\frac{dH}{dt}(H_0 - H) + P_c = P_{in}\frac{H_0}{H} \tag{13}$$

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

$$\frac{d\tilde{H}}{d\tilde{t}}(1 - \tilde{H}) + 1 = \frac{\Pi}{\tilde{H}} \tag{14}$$

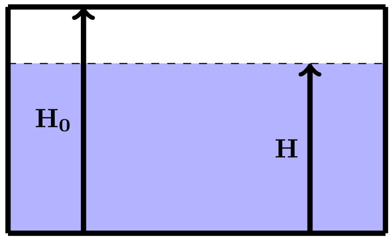

Figure 3: Imbibition of a planar geometry. The wetting fluid (blue) imbibes a plane of porous material. The air (white) in entrapped above the fluid.

and the second derivative writes :

$$\frac{d^2\tilde{H}}{d\tilde{t}^2} = \frac{\left(\Pi\tilde{R}^{-1} - 1\right)\left(-\Pi\tilde{H}^{-2} + 2\Pi\tilde{H}^{-1} - 1\right)}{(1-\tilde{H})^3} \tag{15}$$

The sign of $\frac{d^2\tilde{H}}{d\tilde{t}^2}$ remains constant for $\tilde{H} \in [0,1]$ and the front never accelerates.