Strong Roughening of Spontaneous Imbibition Fronts

Z. Sadjadi[1], H. Rieger[1]
S. Grüner[2], P. Huber[2]

[1] Theoretical Physics, Saarland University
[2] Experimental Physics, Saarland University

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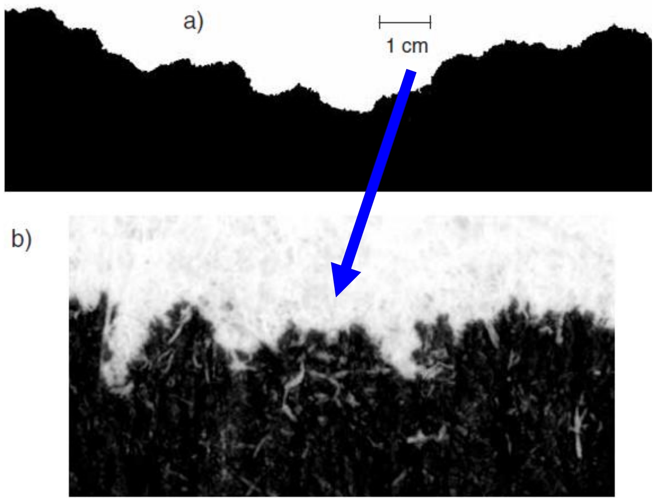
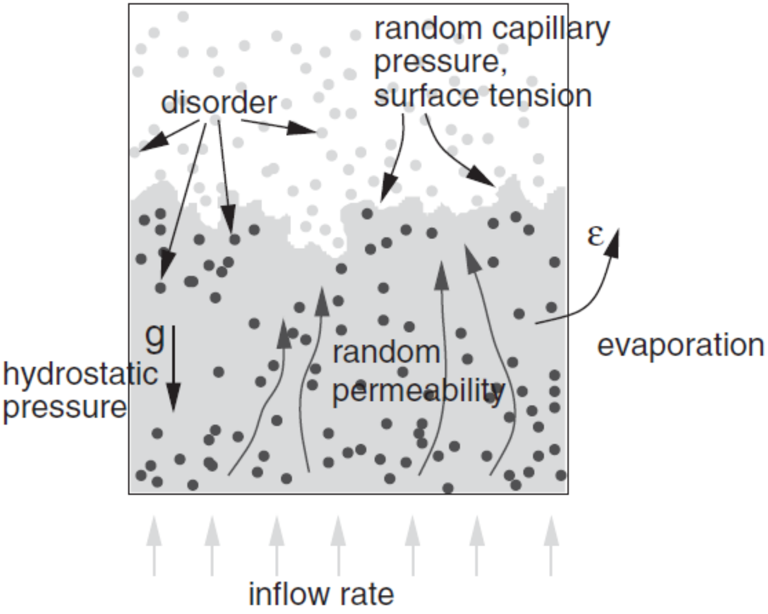
Imbibition:

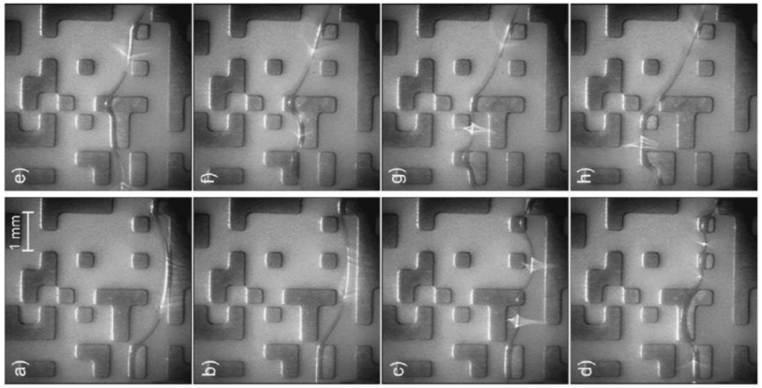
Displacement of one fluid by another immiscible fluid in a porous matrix



Ink in paper towel

Physical processes involved:





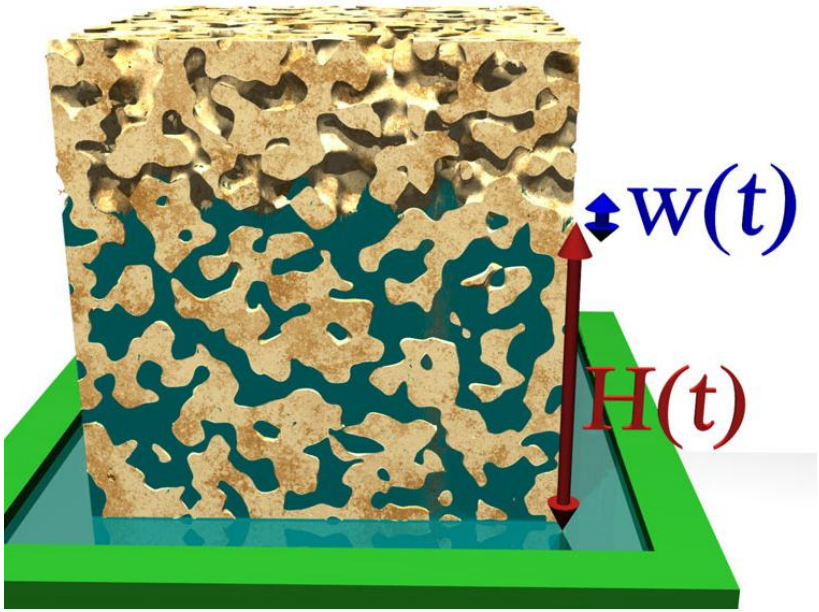
Oil-air interface in an artificial microstructure

Spontaneous imbibition in nanoporous Vycor glass

[S. Grüner, P. Huber (2010)]

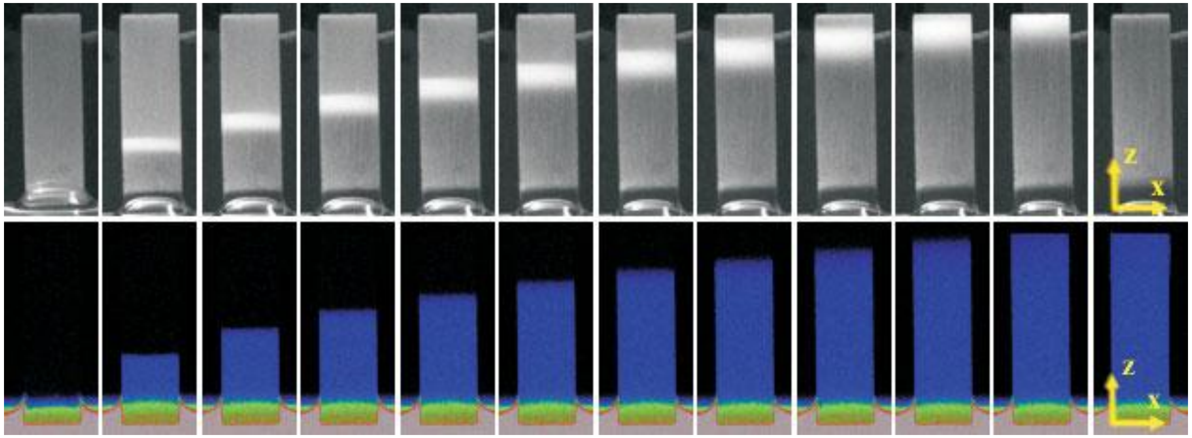
Network of
nano-pores

$R_0 \sim 5 \cdot 10^{-9}$m

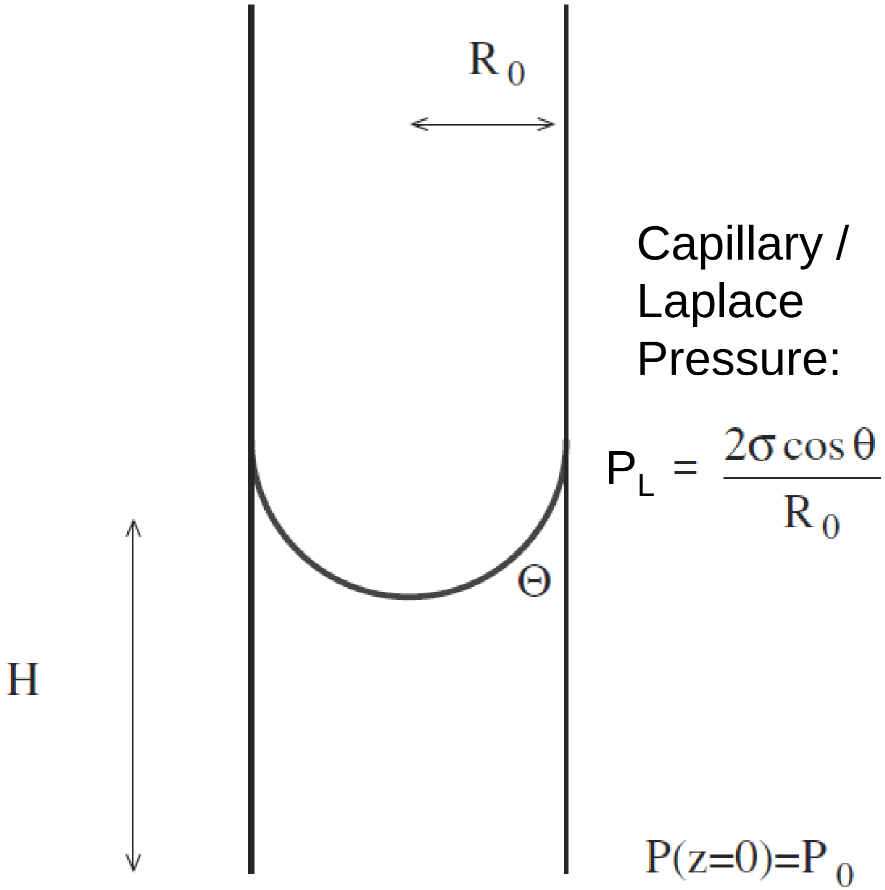


Lightscattering
Experiment:

front height
front width



Lucas-Washburn law for spontaneous capillary rise



Capillary / Laplace Pressure:

$$P_L = \frac{2\sigma\cos\theta}{R_0}$$

$$Q = \frac{\pi}{8}\frac{R_0^4}{\eta}\cdot\nabla P\,, \quad \nabla P = \frac{P_0 - P_L}{H(t)}$$

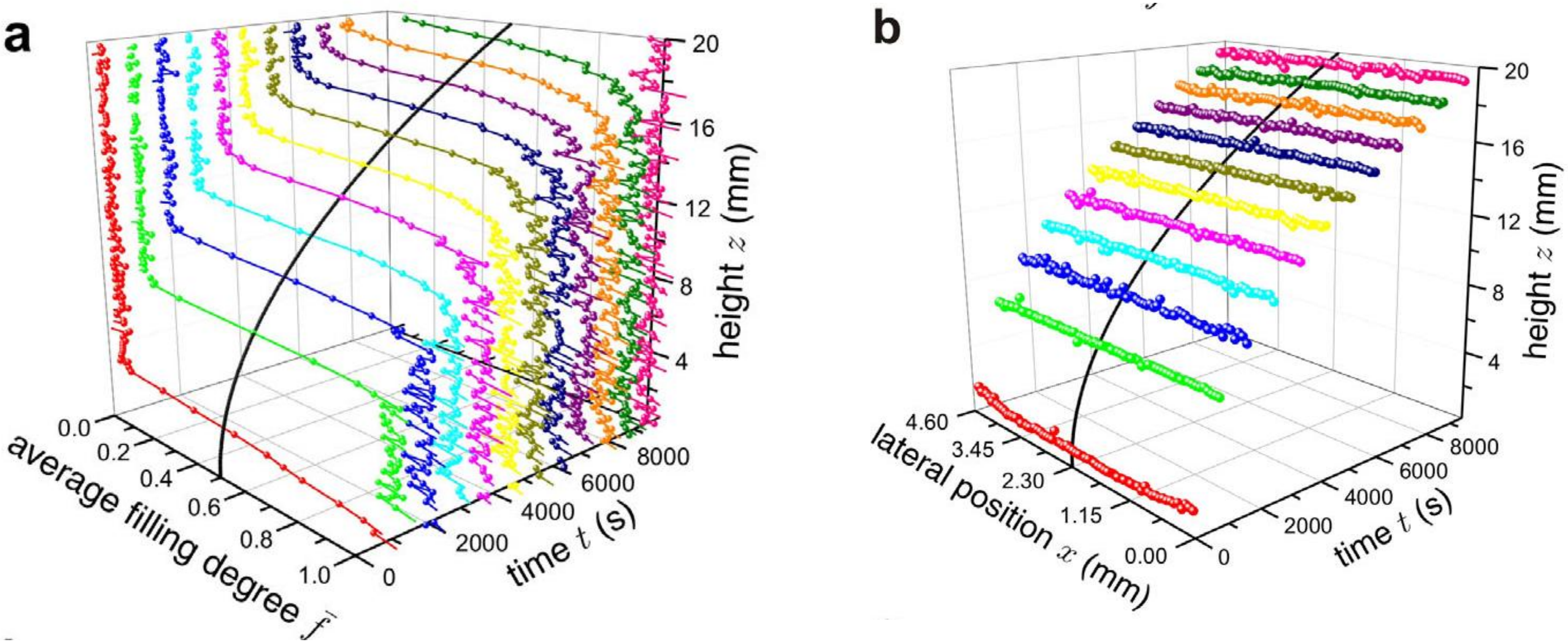
$$Q = \frac{dV}{dt} = \pi R_0^2 \cdot \frac{dH}{dt}$$

$$\Rightarrow dH/dt \propto 1/H(t)$$

$$\boxed{\Rightarrow H(t) \propto \sqrt{t}}$$

Lucas-Washburn

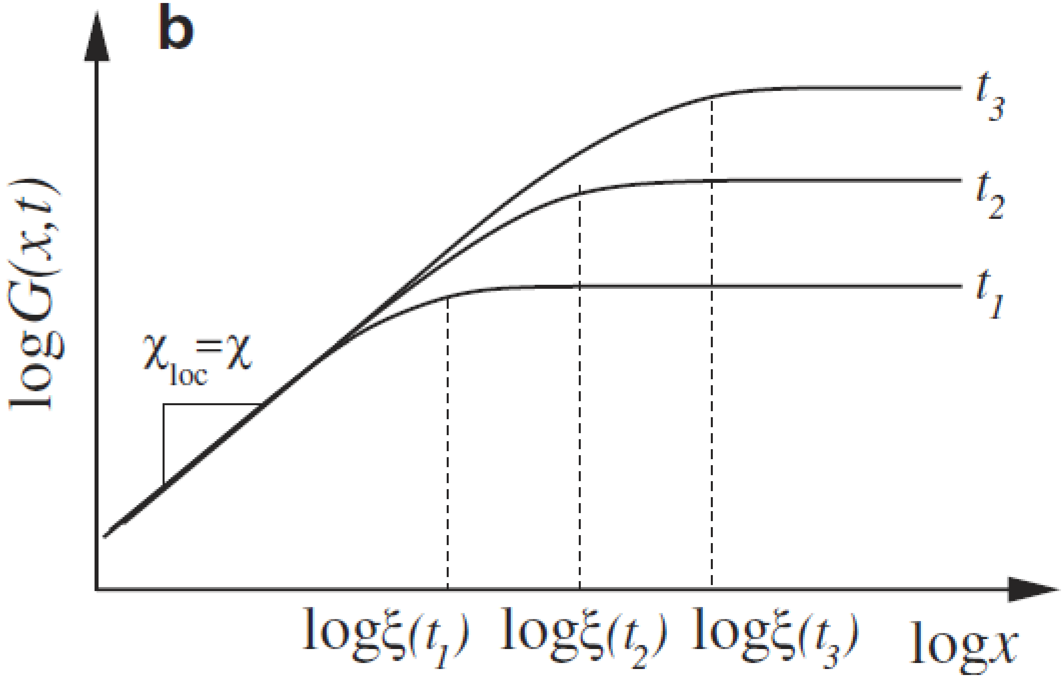
Height evolution of imbibition front in nano-porous Vycor glass



$h(t) \sim t^{1/2}$ - Lucas-Washburn as expected

since on average $$\frac{d\langle h\rangle}{dt} \propto \frac{1}{\langle h\rangle}$$

Kinetic Roughening: Standard scaling



Height difference fluctuations:

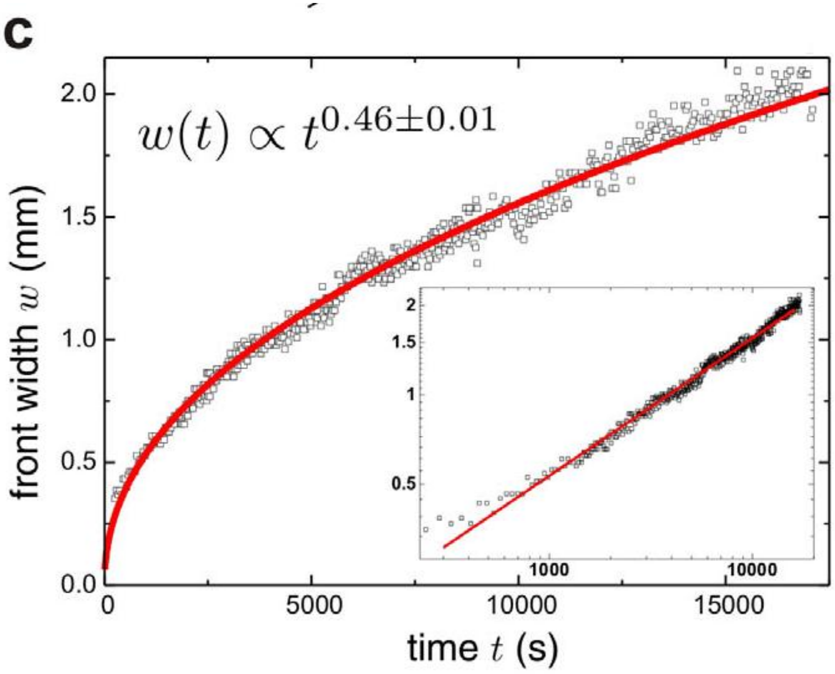
$$G(x,t) = \langle (h(x_0,t) - h(x_0+x,t))^2 \rangle^{1/2} = \begin{cases} c \cdot x^{\chi} & \text{for} \quad x \ll \xi(t) \\ w(t) \sim t^{\beta} & \text{for} \quad x \gg \xi(t) \end{cases}$$

$$\xi(t) \sim t^{1/z} \text{ with } \; z = \chi/\beta$$

Roughness of the imbibition front

Front Width / Roughness

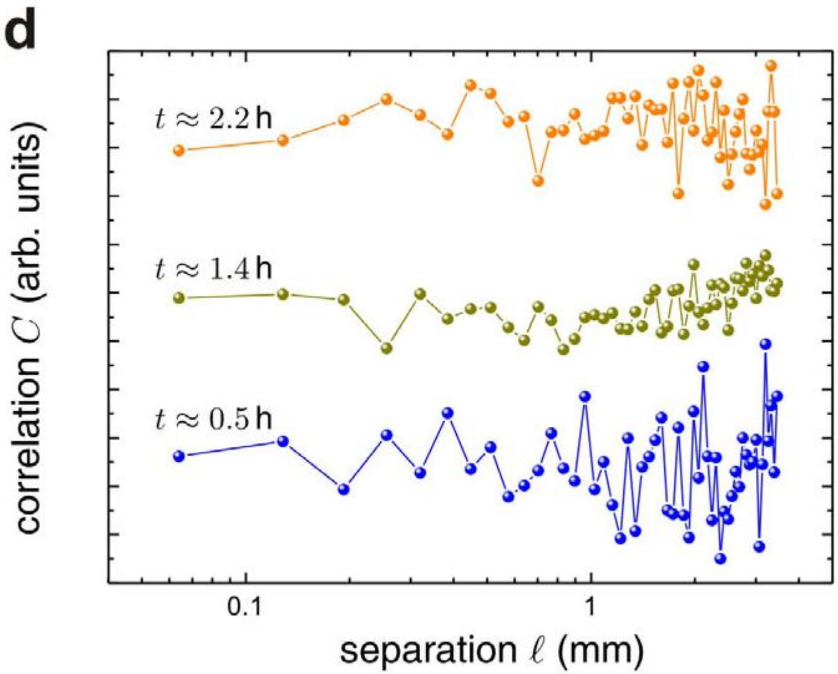
$$w(t) = \sqrt{\langle h^2 \rangle - \langle h \rangle^2}$$



$$\beta = 0.46 \pm 0.01$$

Height difference fluctuations

$$G(\ell, t) = \langle (h(x_0, t) - h(x_0 + \ell, t))^2 \rangle^{1/2}$$



ξ ≈ const. (time independent)

Theory

- Interface roughening, KPZ
- Cellular automata: Directed percolation depinning
- Microscopic simulations (lattice Boltzmann)
- Phase Field Models
- …

Models with surface tension in the imbibition front
display a growing correlation length
and a roughening exponent $\beta << 0.5$

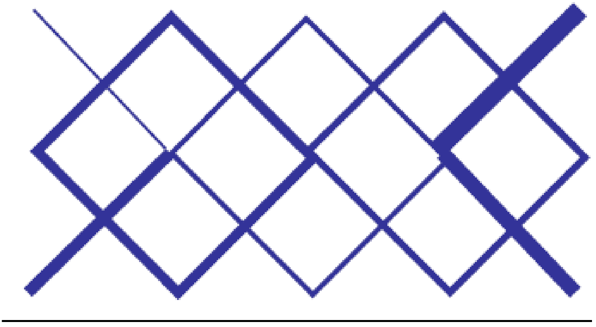
- independent pores with constant (not ok) radius: $\beta = 1/2$
- independent pores with randomly varying (ok) radius: $\beta = 1/4$

- lattice gas model with in porous medium with high porosity (not ok): $\beta \approx 0.5$

Ergo: Theoretical description must contain
A) pore aspects
B) network aspects

Pore network model with quenched disorder

Network of cylindrical pipes with random diameter (<10nm)



Pipe radii:

$$r \in \left[r_{av} - \delta_r , r_{av} + \delta_r \right]$$

← Liquid reservoir in contact with lower system boundary

Filling height in each pipe: $\Delta h_{\mathrm{pipe}}(t)$

Hagen-Poiseuille $$Q_{\mathrm{pipe}}(t) = -\frac{\pi r^4_{\mathrm{pipe}}}{8\eta} \cdot \frac{\Delta p_{\mathrm{pipe}}(t)}{\Delta h_{\mathrm{pipe}}(t)}$$

mass balance at each node i $$\sum_j Q_{\mathrm{pipe}\,(ij)}(t) = 0$$

Boundary conditions:

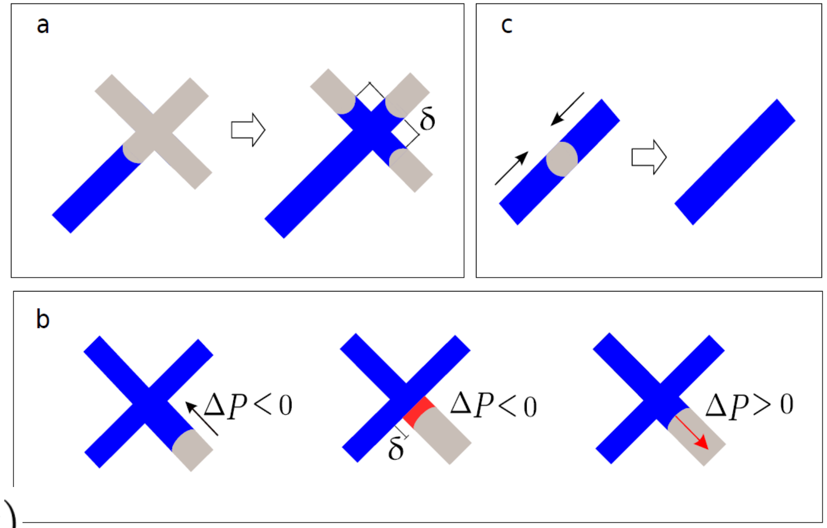
$p_i = 0$ for bottom node layer

$p_{\mathrm{meniscus}} = p_{\mathrm{laplace}} = 2\sigma/r$

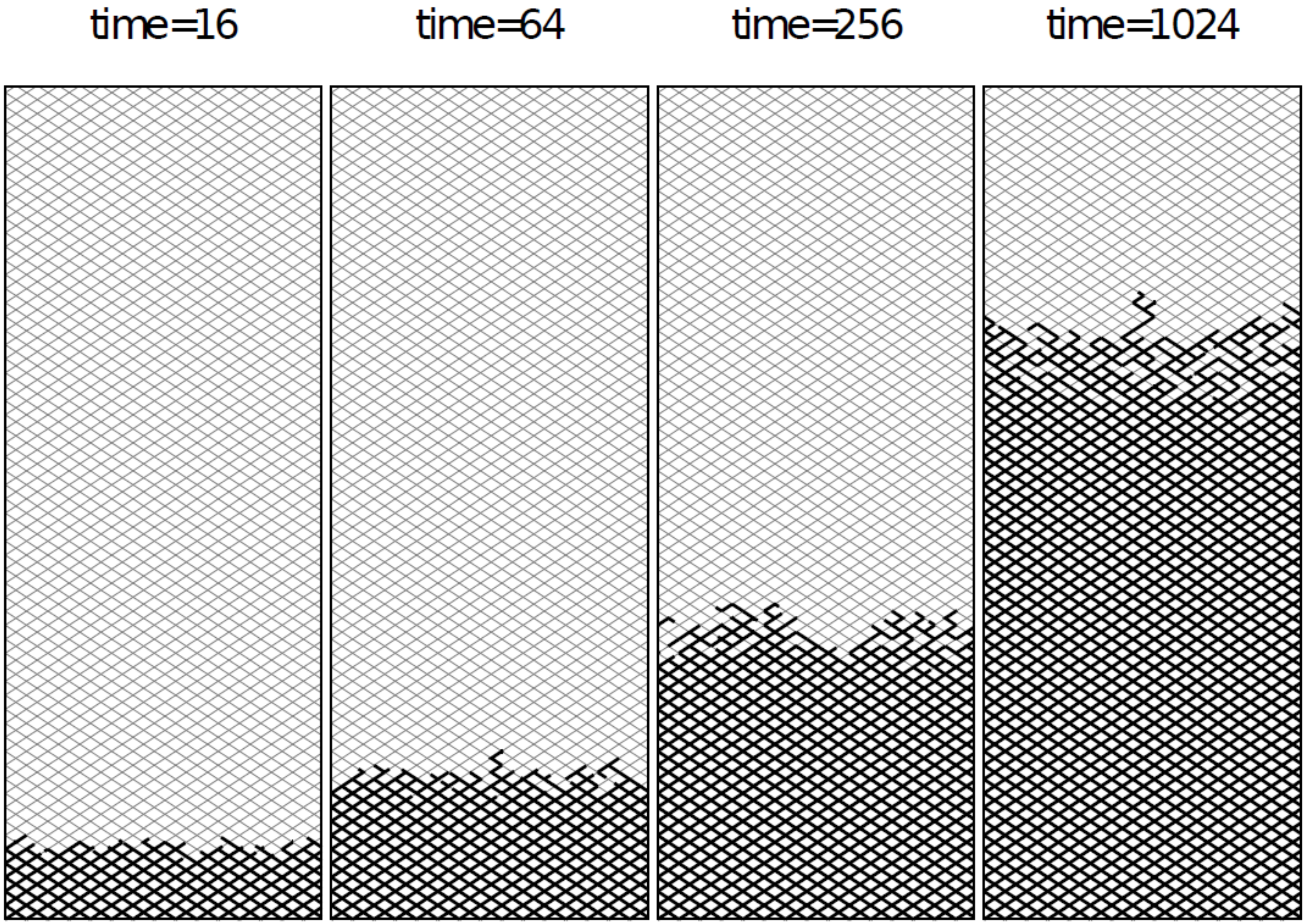
$\Rightarrow$ Compute all node pressures $p_i(t)$

Integrate for step Δt: $$\frac{d\Delta h_{\mathrm{pipe}}(t)}{dt} = \frac{r^2_{\mathrm{pipe}}}{8\eta} \cdot \frac{\Delta p_{\mathrm{pipe}}(t)}{\Delta h_{\mathrm{pipe}}(t)}$$

Rules at junctions / collisions



Time evolution snapshots:

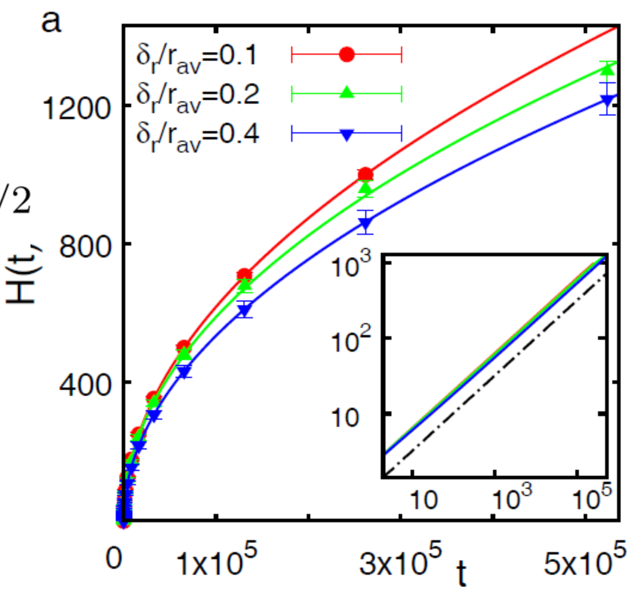


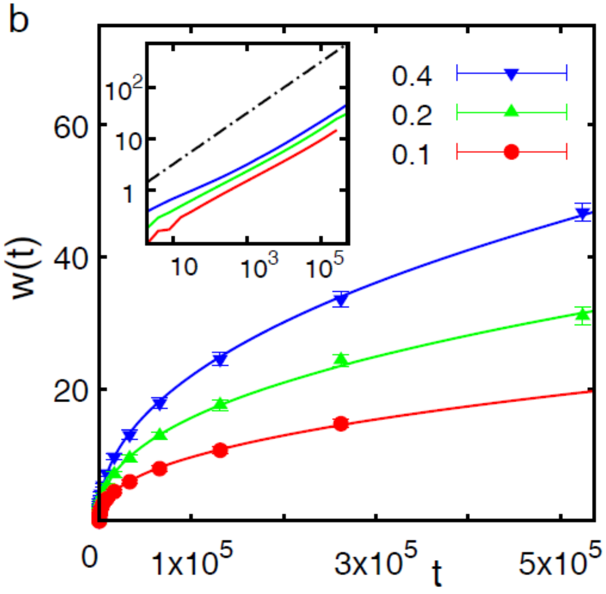
Define height h(x,t) at lateral coordinate x: Average over all menisci in column x

Computer simulation results (disorder averaged):

Lucas-
Washburn

$$H(t) \propto t^{1/2}$$





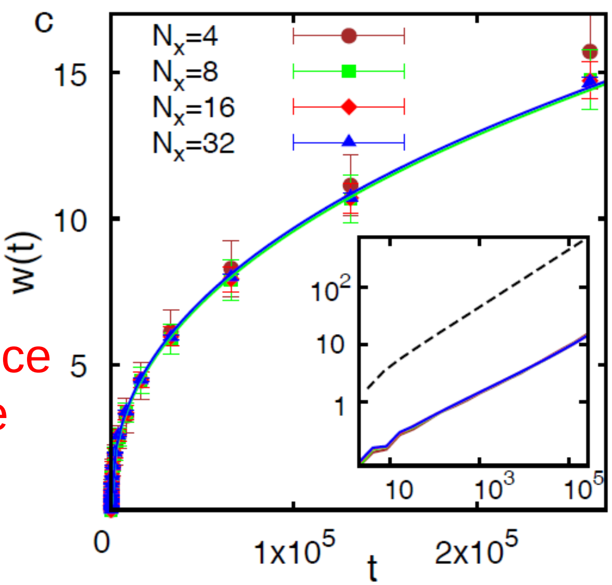
Kinetic
Roughening

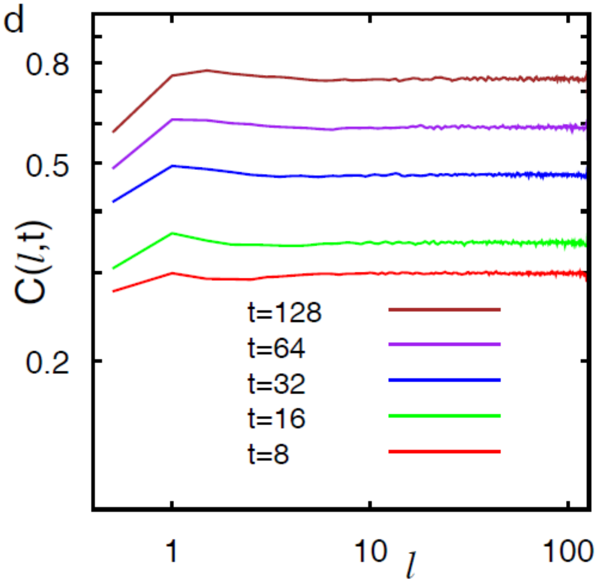
$$w(t) \propto t^{\beta}$$

β = 0.45
± 0.02

„Finite size
scaling“:

No dependence
on lateral size





Height-
difference
Fluctuations
C(l,t)
for fixed t

Correlation
length
ξ=O(1)

Conclusion

- Roughening of imbibition front in vycor glass $\beta = 0.46 \pm 0.01$ …
- … in pore network model $\beta = 0.45 \pm 0.02$.
- Absence of lateral correlations ⇒ uncorrelated meniscus propagation
- Low porosity materials, pore aspect ratio > 2:
  New (?) universality class
- Expected crossover for pore aspect ratio < 1:
  hydrodynamics in junctions important,
  influence of surface tension within imbibition front important

Titel

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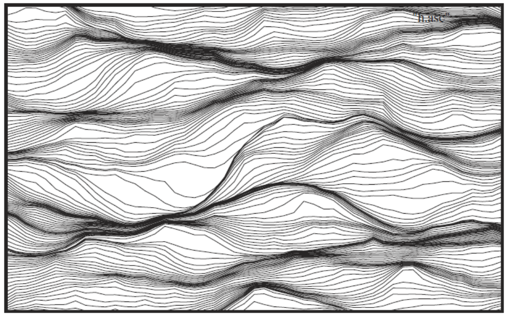
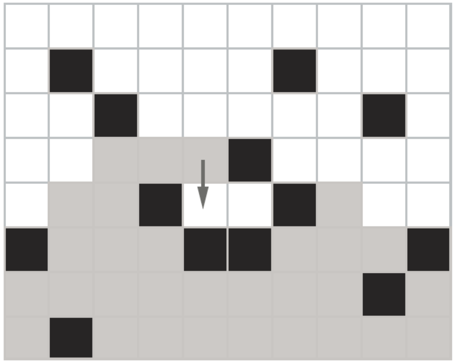
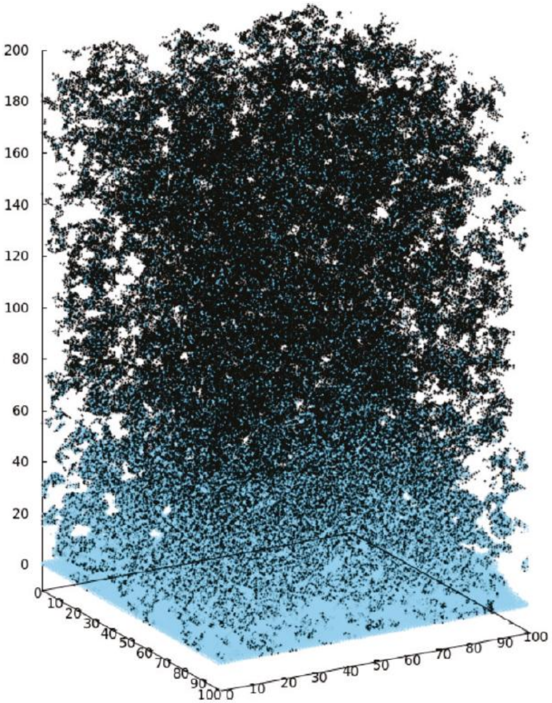
Imbibition: theory



Phase field models (Alava et al.)



Directed percolation
Depinning (DPD)

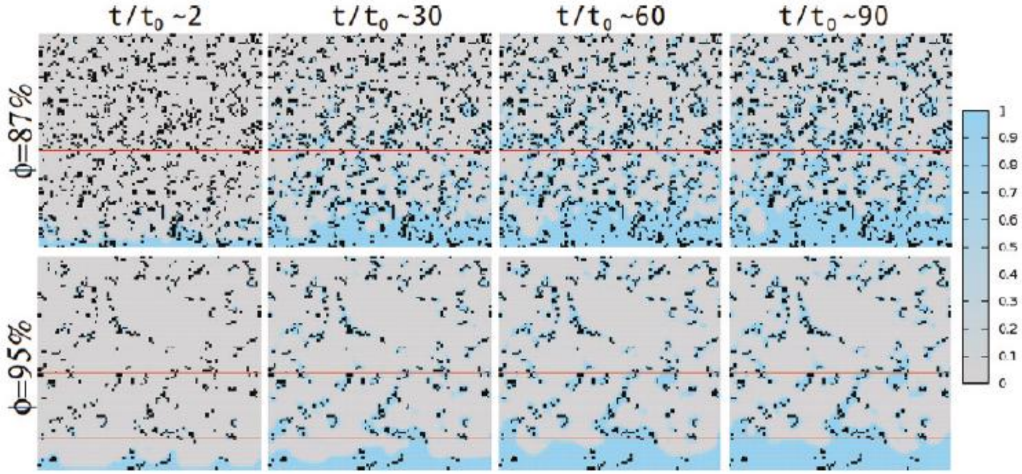


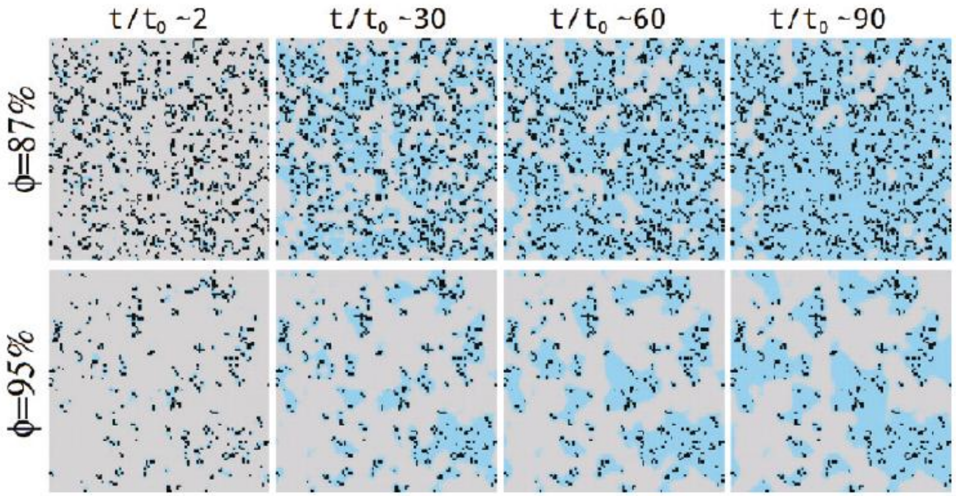
Lattice gas model (Tarjus et al)

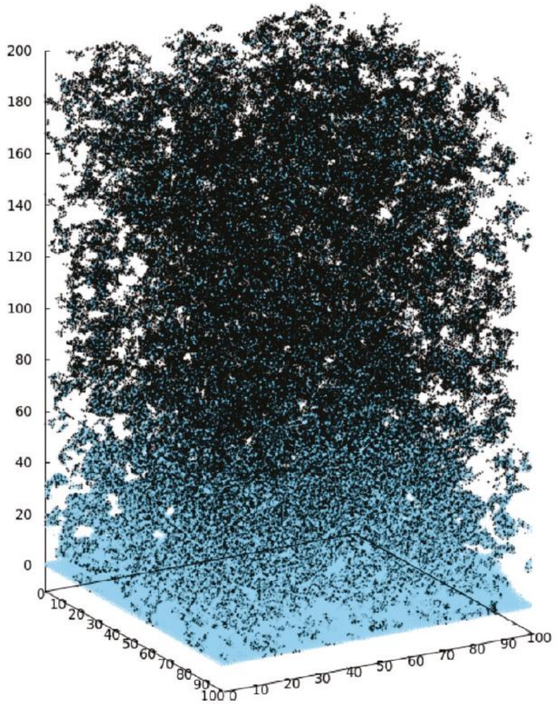
$$H = -w_{\mathrm{ff}} \sum_{\langle ij \rangle} \tau_i \tau_j \eta_i \eta_j - w_{\mathrm{sf}} \sum_{\langle ij \rangle} [\tau_i \eta_i (1 - \eta_j) + \tau_j \eta_j (1 - \eta_i)]$$

Lattice gas model for an aero-gel

$$H = -w_{\mathrm{ff}} \sum_{\langle ij\rangle} \tau_i \tau_j \eta_i \eta_j - w_{\mathrm{sf}} \sum_{\langle ij\rangle} \left[\tau_i \eta_i (1-\eta_j) + \tau_j \eta_j (1-\eta_i)\right]$$







$\beta \approx 0.5$

BUT: High porosity

$\Rightarrow$ surface tension relevant

$\Rightarrow$ lateral correlations