Convective dissolution of carbon dioxide in 2D water-saturated porous media: an experimental study in micromodels

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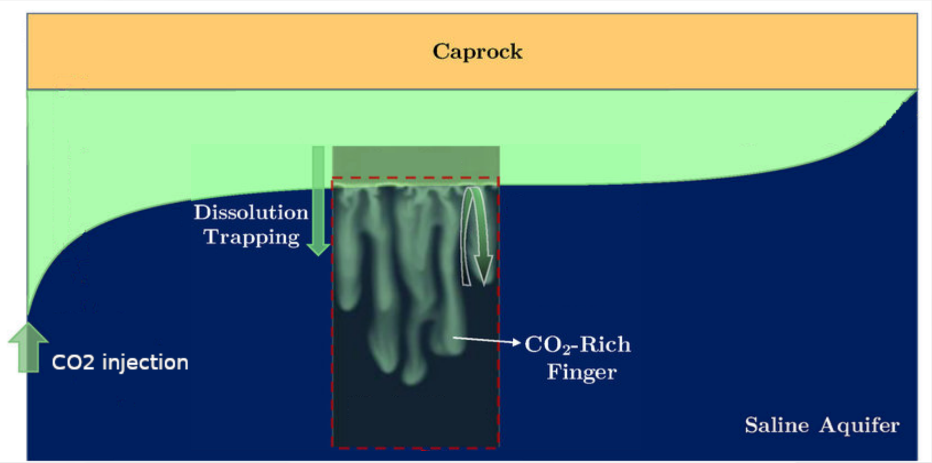
Loughborough University

Aix*Marseille université Socialement engagée

Université de Rennes

Carbon Capture and Storage (CSS)

- Capture CO_2 from industry effluent
- Transport the captured CO_2 to storage site
- Inject CO_2 in geological storage sites – saline aquifers
- Main concern – how CO_2 will stay in the underground reservoirs for millions of years?
- Solubility trapping is an effective mechanism for long term storage



Motivations for the study

So far: many studies in pure Hele-Shaw cells or in rock samples (X-ray tomography), but to our knowledge very few in 2D micromodels.

→ Experiments in 2D micromodels that

- Allow measuring the concentration field of the dissolved CO_2
- Allow for an analysis of the impact of pore scale heterogeneity on the convective dissolution process
- Bridge the gap between pure Hele-Shaw cell and 2D micromodel

Theoretical description

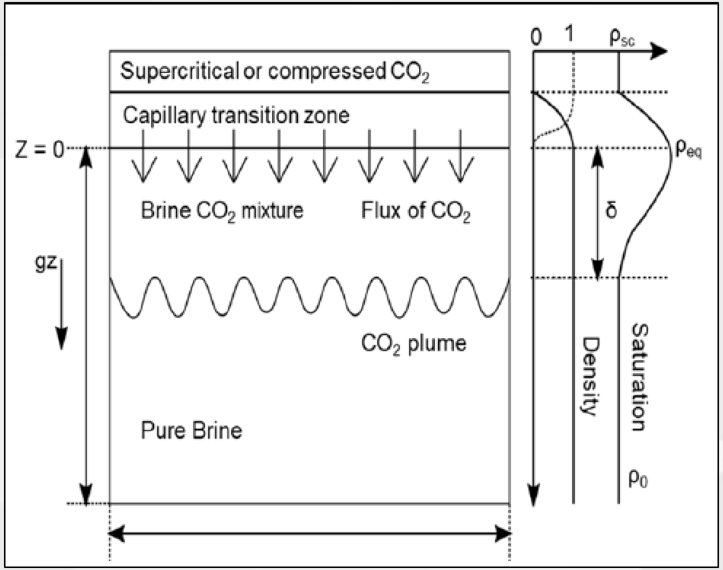
$$\rho\left[\frac{\partial u}{\partial t}+(u\cdot\nabla)u\right]=-\nabla P+\mu\nabla^2 u-c\rho_0\alpha g\hat{z}$$

$$\nabla\cdot\rho u=0 \qquad \frac{\partial c}{\partial t}+(u\cdot\nabla)c=D\nabla^2 c$$

$$c^*=k_H P_{CO_2}\,,\bar{c}=\frac{c}{c^*} \qquad L^*=\frac{D}{u^*},\bar{L}=\frac{L}{L^*}$$

$$u^*=\frac{\triangle\rho g K}{\mu},\bar{u}=\frac{u}{u^*} \qquad t^*=\frac{D}{u^{*2}}\,,\bar{t}=\frac{t}{t^*}$$

Non-dimensional Equations



- D is the CO_2 diffusivity.
- μ and ρ are the viscosity and density of solution.
- K, k_H are permeability of porous media and Henry's constant.

Theoretical description (2)

$$\frac{1}{Sc}\left[\frac{\partial \bar{u}}{\partial t} + (\bar{u}\cdot\nabla)\bar{u}\right] = -\nabla P + \nabla^2 \bar{u} - \frac{1}{Da\, Ra^2}\bar{c}\hat{z}$$

$$\frac{\partial \bar{c}}{\partial t} + (\bar{u}\cdot\nabla)\bar{c} = \nabla^2 \bar{c} \qquad \nabla\cdot\bar{u} = 0$$

→ σ and q relation, dispersion equation

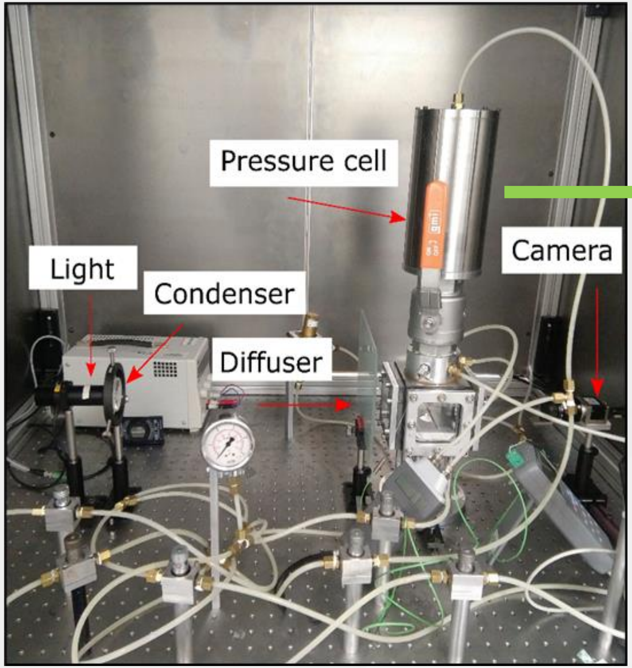
$$Ra = \frac{\triangle\,\rho g K H}{\mu D},\ Da = \frac{K}{H^2},\ Sc = \frac{\mu}{\rho D}$$

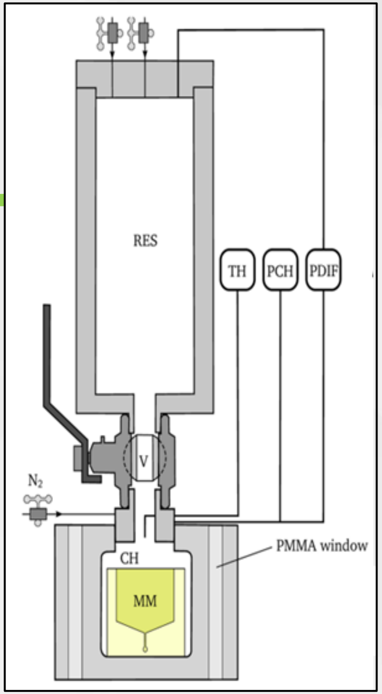
$$c = c_0 + \tilde{c}$$
$$w = w_0 + \tilde{w}$$
$$\tilde{c} = c_s(z)e^{iqx}e^{\sigma t}$$
$$\tilde{w} = w_s(z)e^{iqx}e^{\sigma t}$$

- Sc is high.
- H is the height of the reservoir.
- Flow is parabolic along the thickness of the cell

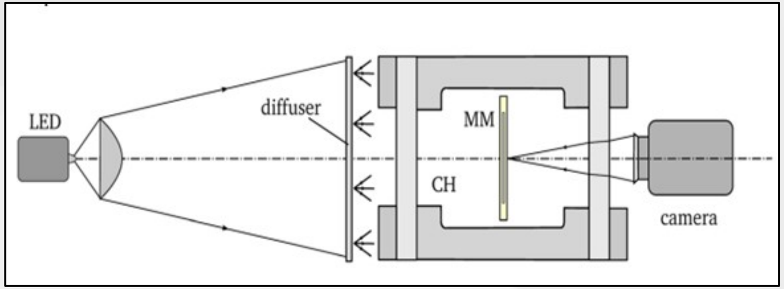
- q and σ are wavenumber and the growth rate of instability.
- c_0, w_0 are base state, and $\tilde{c}, \tilde{w}$ are perturbation state.

Experimental Setup



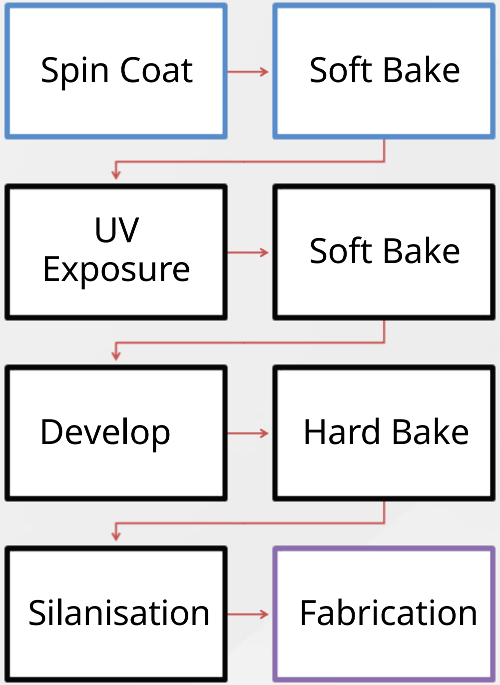


Schematic of Optical Diagram

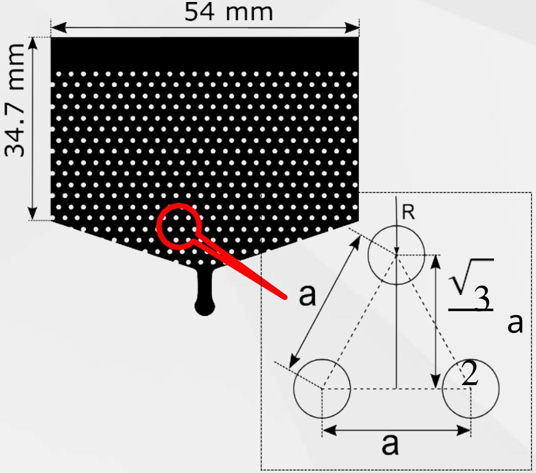


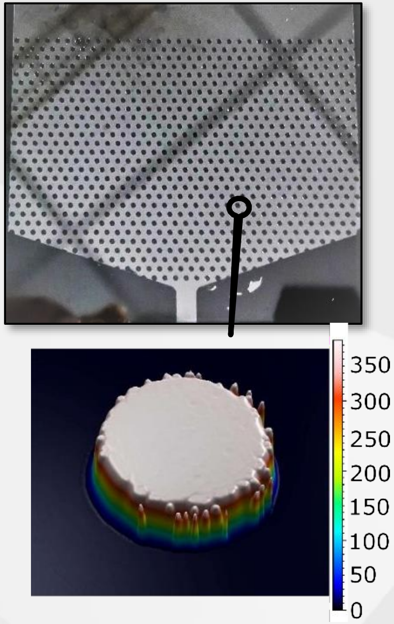
- Direct injection
  → thermal instability
- Perform experiment at isothermal condition
- $P_{CO_2} = (V_c + V_V + V_R)^{P_T}/_{V_R}$
- Mixing time <30 s

Photolithography Technique

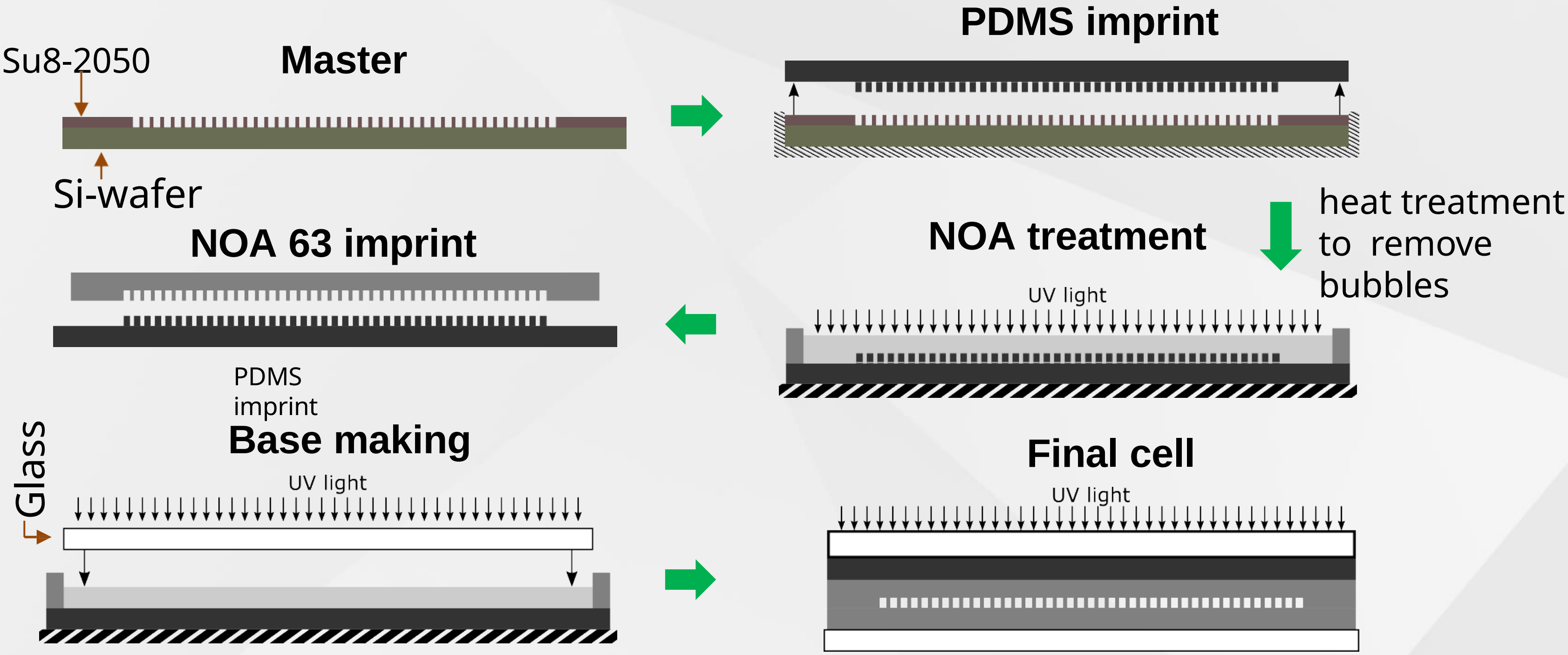


- Repeat 2 times
- 300 μm thickness

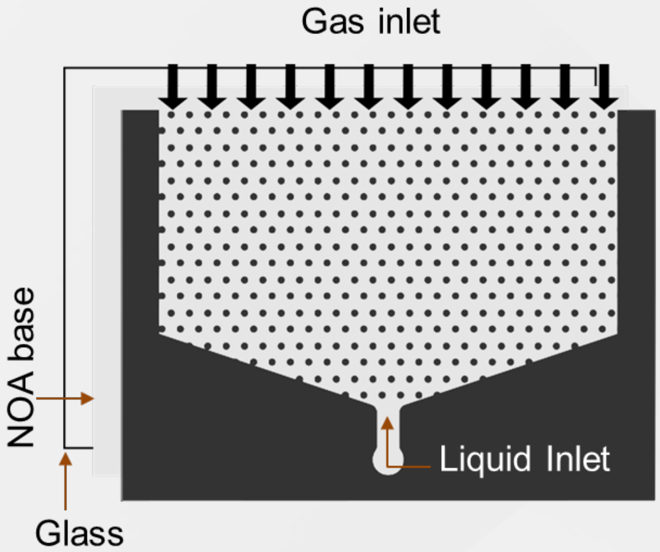


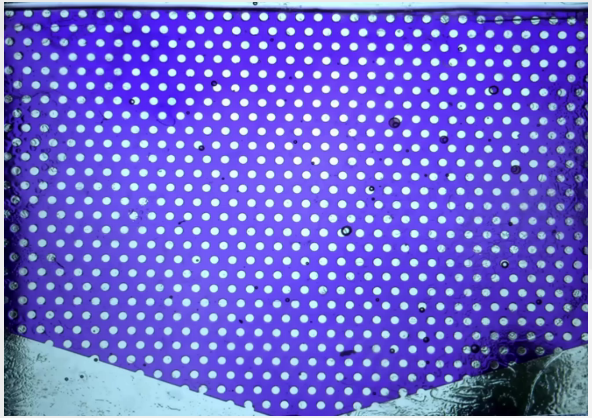


NOA 63 device fabrication



Experimental protocol





- Schematic of NOA 63 device
- Cell filled before experiment

- $P_{CO_2} = 7$ bar, $BP_{conc} = 10^{-3}$ mol l^{-1}
- FPS = 7, total time = 81 min

Characteristic non-dimensional numbers

$$Da = \frac{k_{Da}}{H^2}$$

$$Ra = \frac{\Delta p g k_{Da} H}{\mu \emptyset D}$$

$$Da = \frac{k_{HS}}{a^2}$$

$$k_{HS} = {E^2}/{12}$$

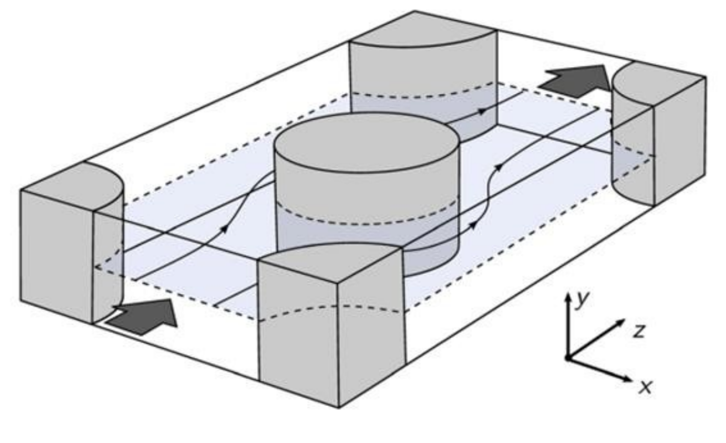
$$Ra\sqrt{Da} = \frac{\Delta\rho g}{\mu D}\kappa^{\frac{3}{2}} = A \times P_{CO_2} \times k_{comsol}{}^{3/2}$$

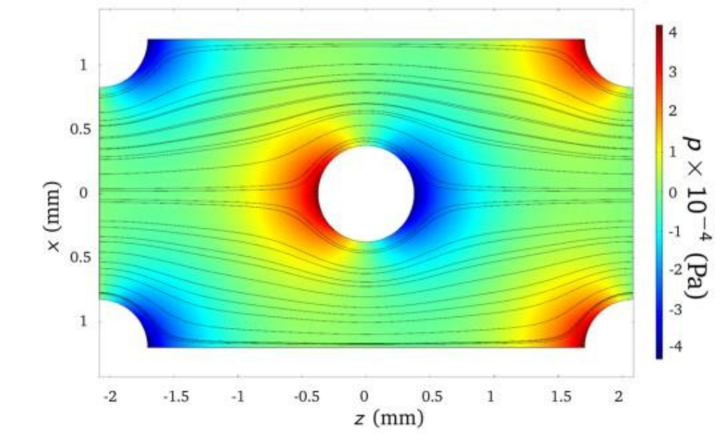
$$q^* = q \times \mathcal{L} = \frac{2\pi N}{L} \times \frac{1}{A \times P_{CO_2} \times k_{comsol}}$$

$$A = \frac{\alpha K_H}{\left(\frac{\mu}{\rho_0}\right) D}$$

- Low-porous- $\emptyset \sim 0.6$, $k_{Da} \ll k_{HS}$
- Intermediate-porous - $\emptyset \sim 0.8$, $k_{Da} < k_{HS}$
- High-porous - $\emptyset \sim 0.9$, $k_{Da} \approx k_{HS}$

Computation of permeability





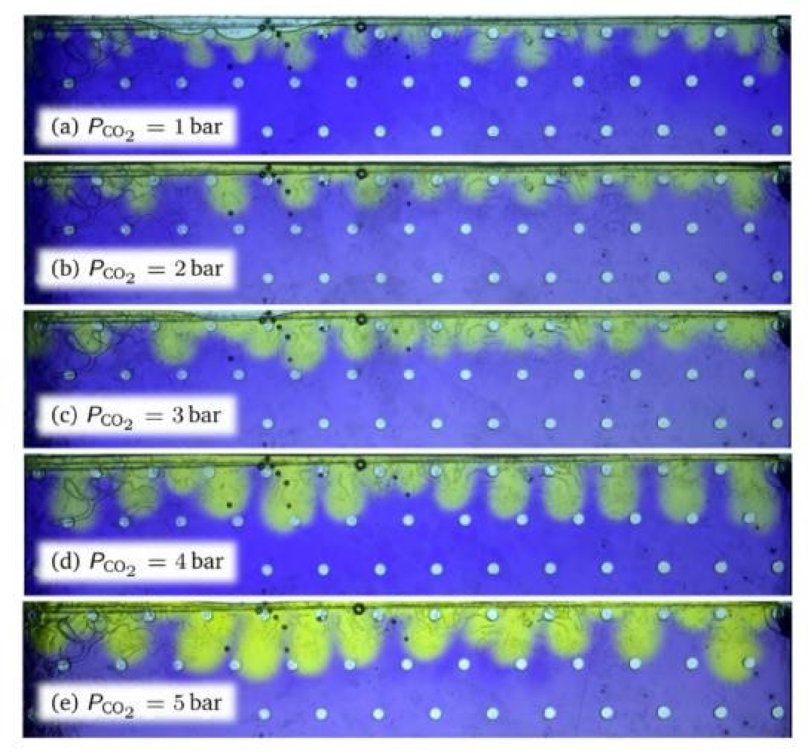
- k_{Da} is determined for the unit cell by COMSOL simulation.

From classical Hele-Shaw to porous micromodel

MM type	E (μm)	a (μm)	R (μm)	ϕ	$\kappa_{HS} \times 10^{10}$ (m^2)	$\kappa_{MM} \times 10^{10}$ (m^2)
a3R200	171	601	176	0.69	24.4	7.9
a3R300	230	900	275	0.66	44.1	16.2
a3R400	227	1202	373	0.65	43.0	14.5
a4R200	255	796	175	0.82	54.2	26.2
a4R400	278	1608	364	0.81	64.4	35.8
a6R200	168	1205	185	0.91	23.5	17.7
a6R400	206	2400	375	0.91	35.4	27.7
a8R200	226	1600	175	0.96	42.6	35.9
a8R400	203	3200	375	0.95	34.3	30.0
a10R200	311	2000	175	0.97	80.6	70.5
a10R400	225	4000	375	0.97	42.2	38.6

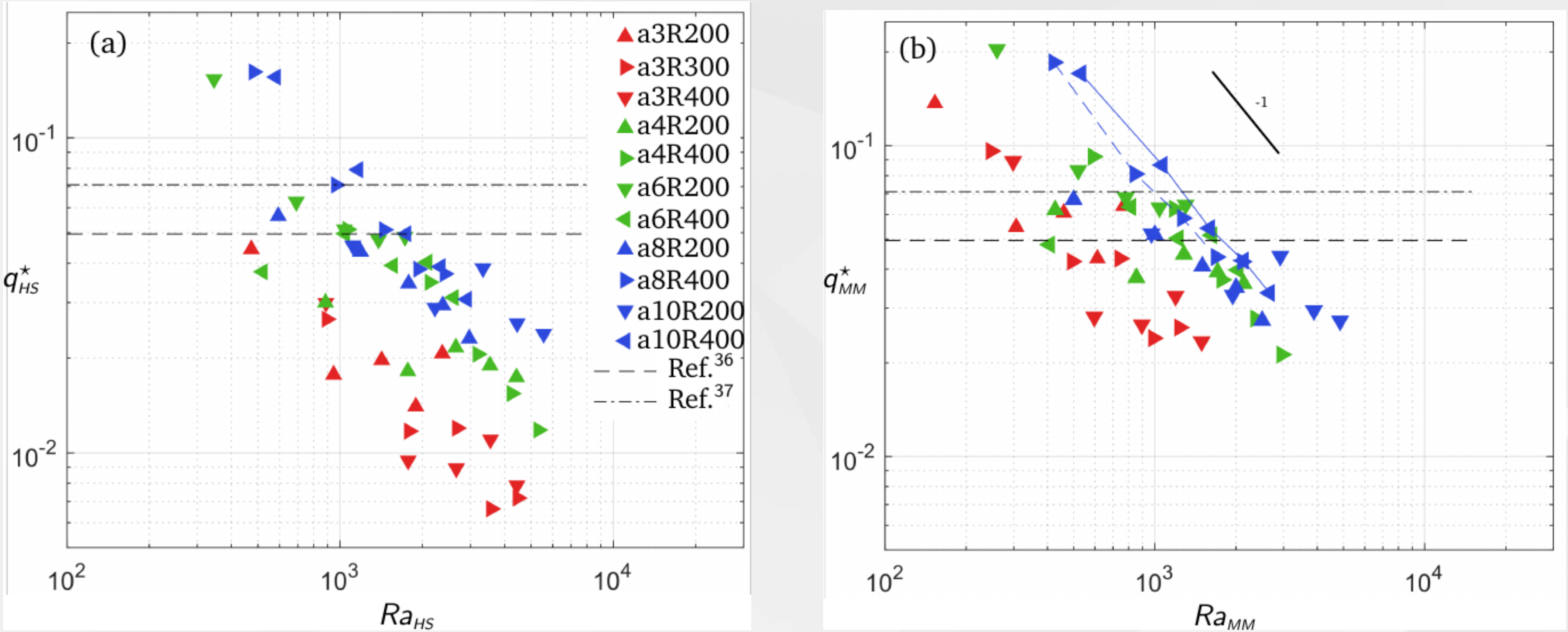
- $\varnothing = 1 - \frac{\pi}{2\sqrt{3}}\left(\frac{d}{a}\right)^2$
- k_{MM} directy inferred from 3D comsol Multiphysics
- Close to $\varnothing$=1 Darcy and pore scale are equal, Hele-Shaw approximation works
- $\varnothing$~0.5 pore scale is much higher than that of Darcy scale, hele-shaw approximation is invalid

Experiments at different partial pressures



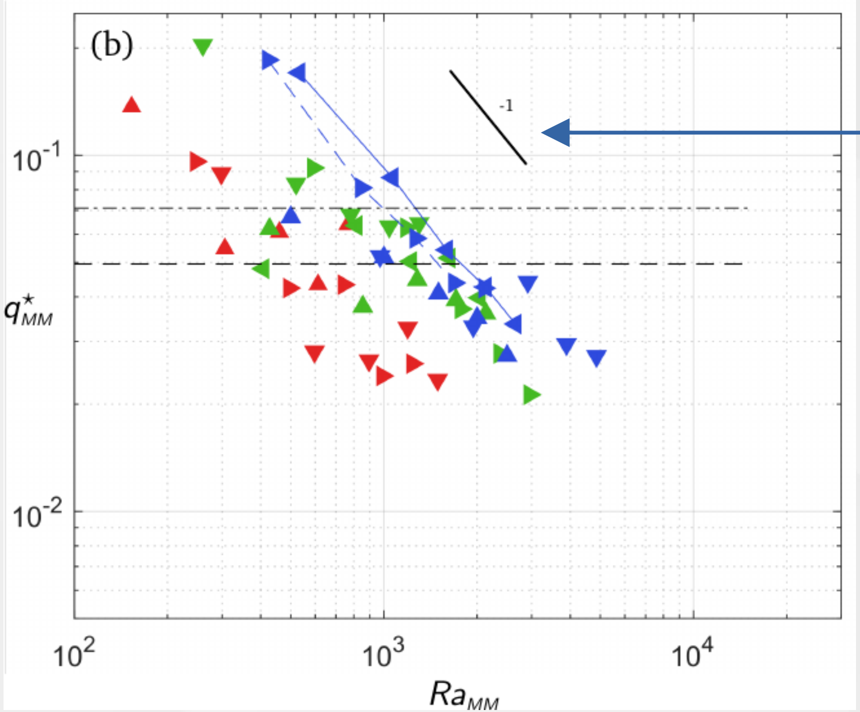
- Porous micromodel with center-to-center distance a = 4 mm
- PCO2 = 1 to 5 bar (end of the linear regime, t = 2 min).
- The wavelength Λ* does not show obvious dependence on the pressure, so that the dimensionless number q^*_{MM} is proportional to Ra_{MM}^{-1}

Wavenumber as a function of Ra



Ref:- Two-dimensional micromodels for studying the convective dissolution of carbon dioxide in 2D water-saturated porous media, De et al, Lab Chip, 2022, 22, 4645

Wavenumber as a function of Ra (2)



Locking of the wavelength on the distance between adjacent pillars

Ref:- Two-dimensional micromodels for studying the convective dissolution of carbon dioxide in 2D water-saturated porous media, De et al, Lab Chip, 2022, 22, 4645

Conclusions

- New type of NOA-based microfluidic cell to investigate convective dissolution of CO_2 in deep aquifers
- Experiments were performed with CO_2 pressures ranging from 1 to 5 bars
- **The convective dissolution process is much slower than in a pure Hele-shaw cell**, despite the large porosity (small size of the pillars)
- Measured values of the most unstable wavelength in the linear instability regime exhibit values that are up to 10 times different from those predicted for a standard Hele-Shaw (HS) cell
- When considering in that prediction the medium's permeability (not that of the corresponding HS cell), the discrepancy is down to a factor 3
- A new phenomenon was observed: **locking of the wavelength** on the distance between adjacent pillars

Funding and paper

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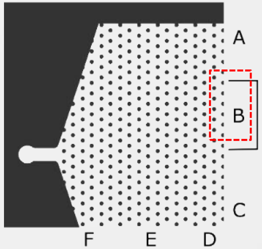
Reference:

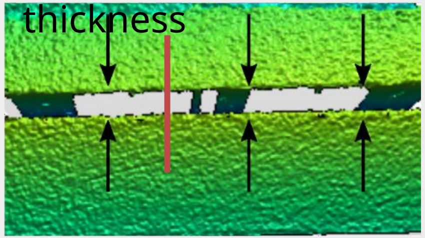
N. De, N. Singh, R. Fulcrand, Y. Méheust, P. Meunier & F. Nadal (2022), Two-dimensional micromodels for studying the convective dissolution of carbon dioxide in 2D water-saturated porous media, *Lab Chip* **22**(23), 4645-4655.

Thank you for your attention

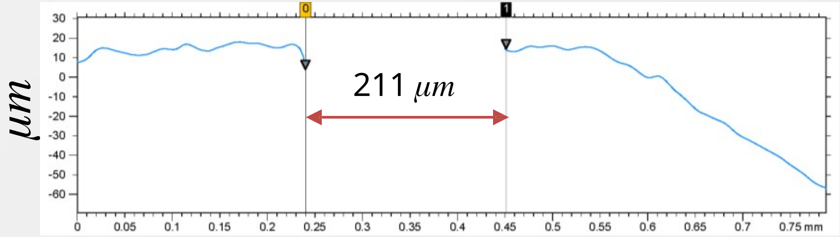
Measurement of cell parameters



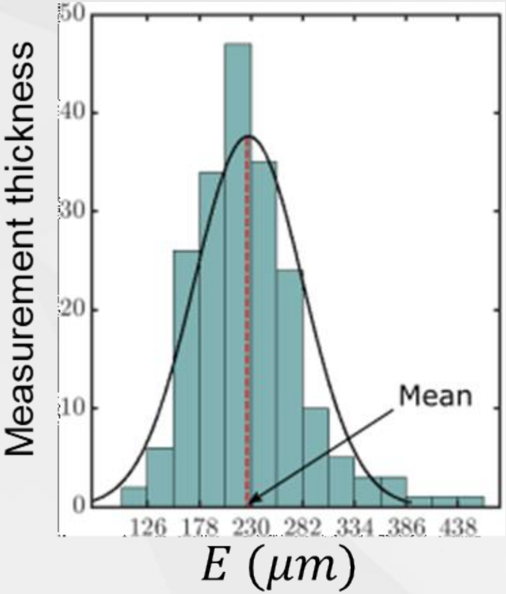
Gap thickness

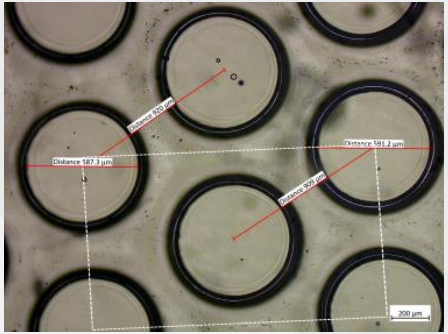


Profilometric measurement



Protocol has good repeatability





- Microscopic image for a = 1200 μm , R = 400 μm,
- Final data are average,
- Measured E is used.