



CO₂ Enhanced Storage (CO₂ES) An industrial Chair about CO₂ Storage

Fabrizio CROCCOLO

UniMi Physics Department Seminar – February 10th, 2022



**Roberto
CERBINO
Uni-Wien (AT)**

**Marta
COSENTINO
Industry (IT)**

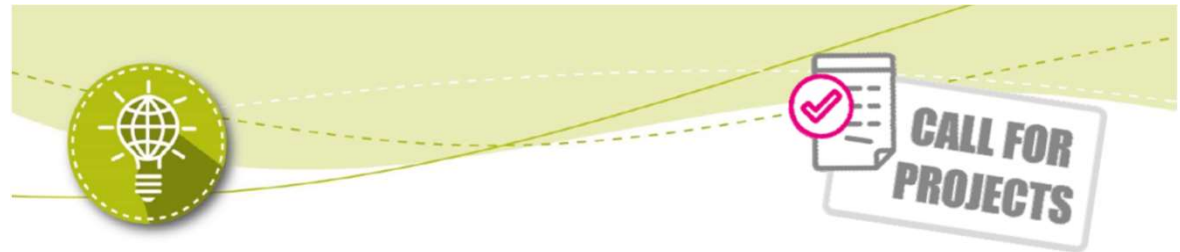
**Fabrizio
CROCCOLO
Uni-Pau (FR)**

**Doriano
BROGIOLI
Uni-Bremen
(DE)**

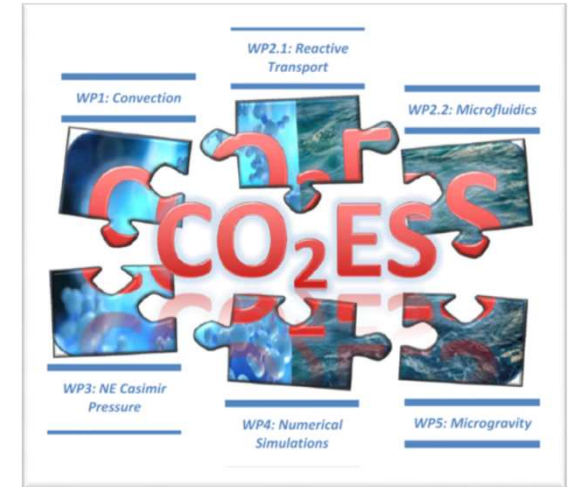
**Pietro
CICUTA
Uni-Cambridge
(UK)**



INDUSTRIAL CHAIR CO₂ES



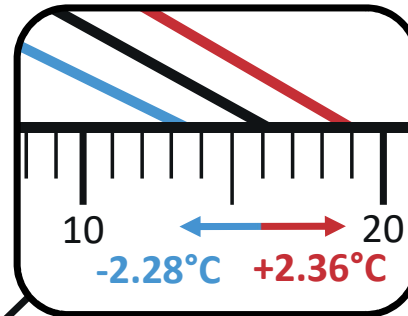
INDUSTRIAL CHAIR CO₂ES



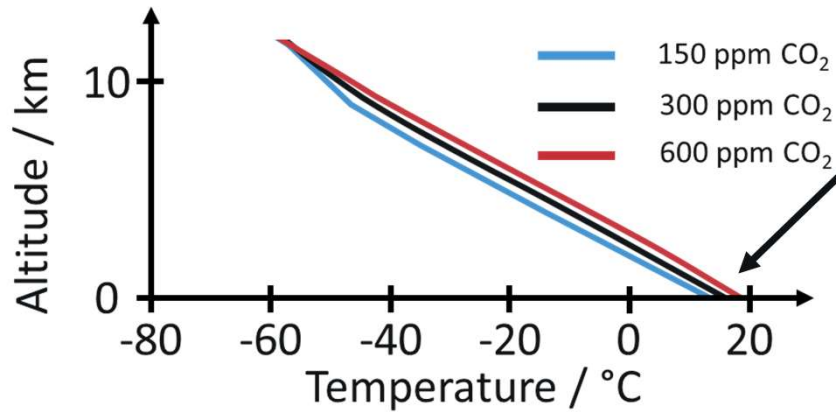
Chaire CO₂ES
Dirigée par Fabrizio Crocolo
Professeur au LFCR

La chaire industrielle CO₂ES a pour objectif d'améliorer la compréhension des différents mécanismes de piégeage du CO₂ dans les réservoirs géologiques.

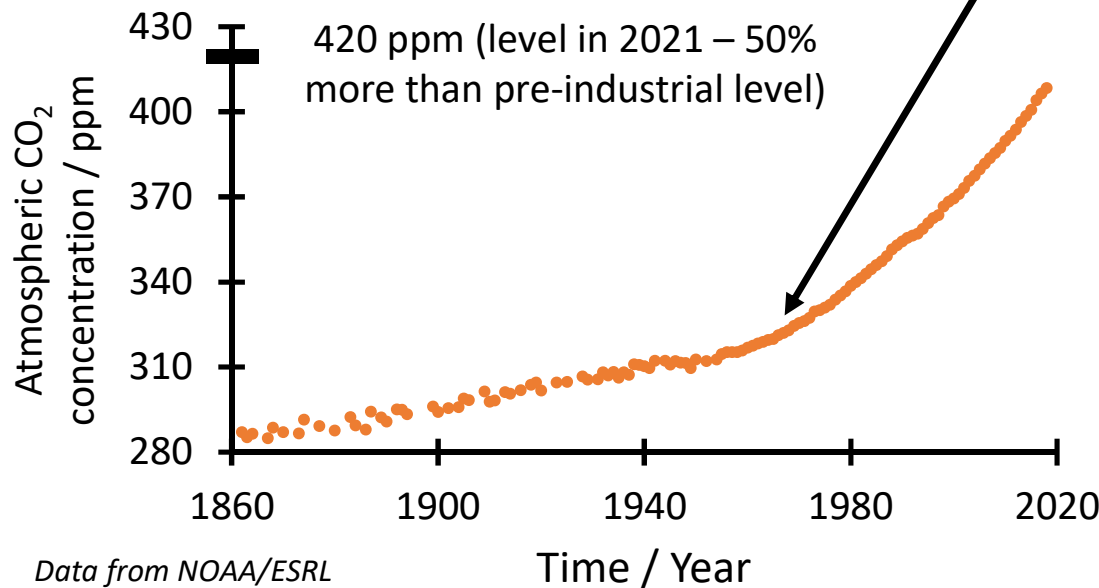
CO₂ IMPACT ON GLOBAL TEMPERATURE



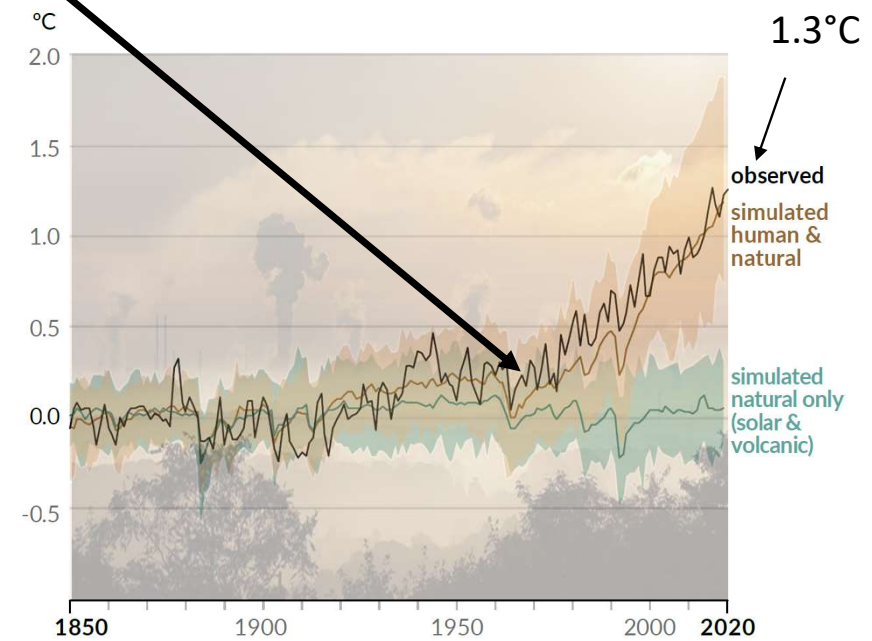
Syukuro Manabe – Nobel price in Physics 2021
 © Nobel Prize Outreach



Manabe and Wetherald
 (1967)



Data from NOAA/ESRL



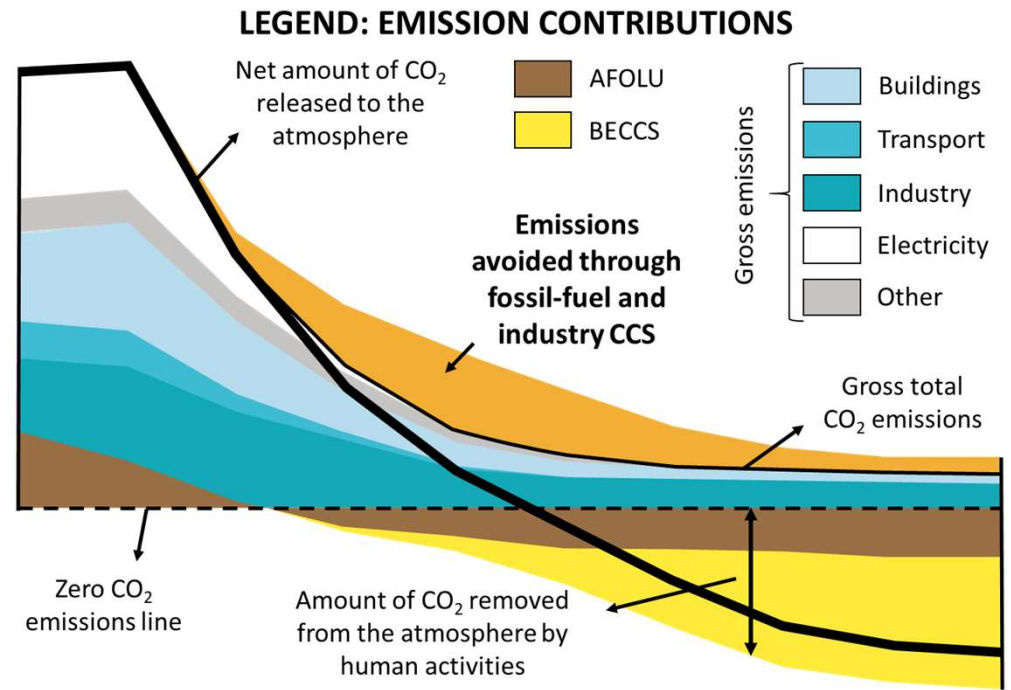
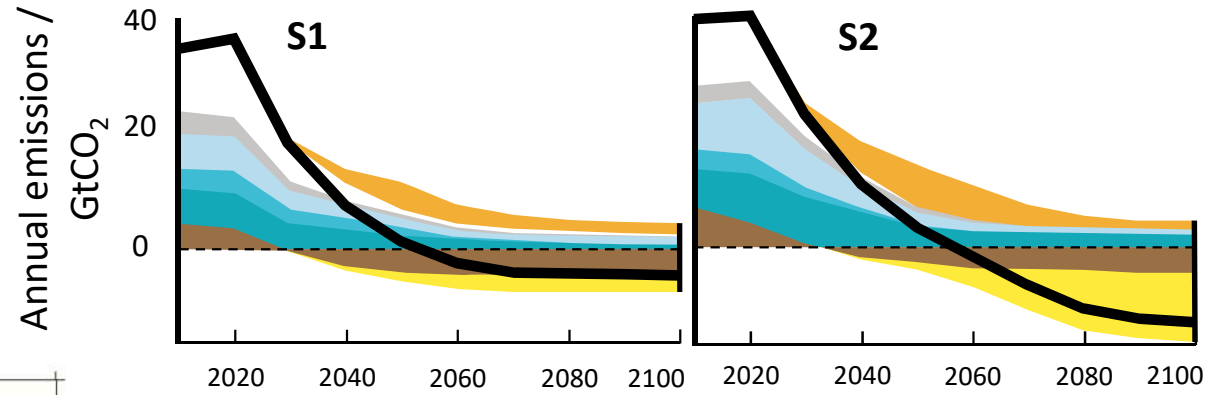
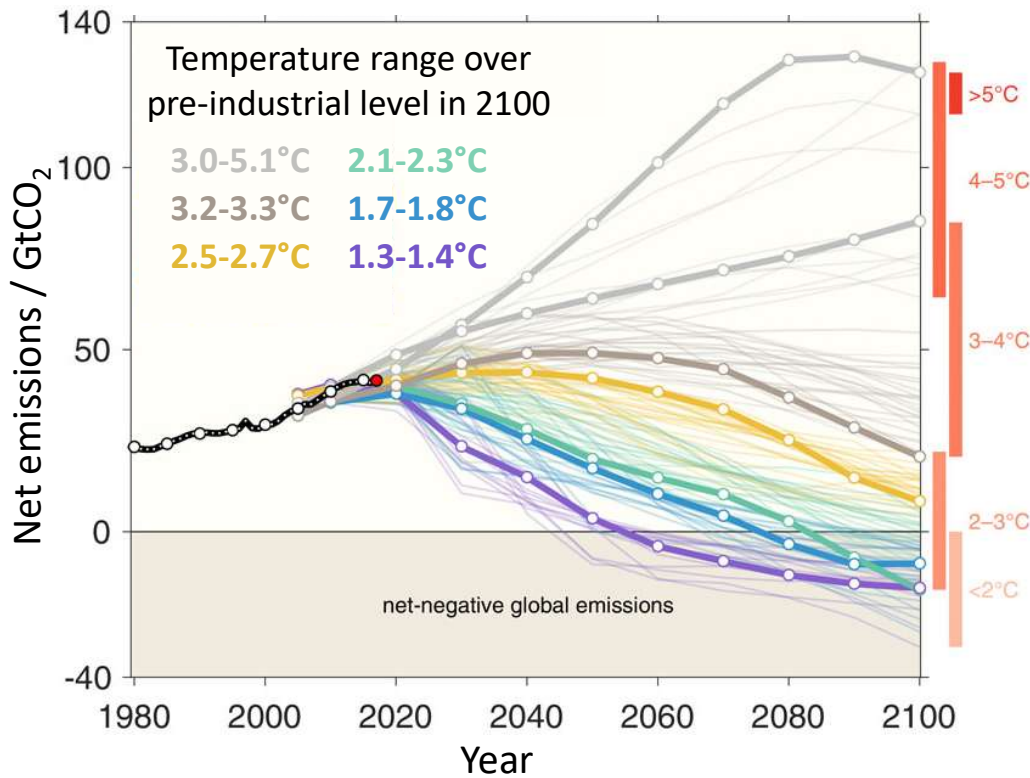
IPCC Climate change 2021 – The physical basis



PATHWAYS TO LIMIT GLOBAL WARMING TO 1.5°C



Simulated carbon net global emissions from the Global Carbon Project



IPCC Special report – Global warming of 1.5°C (2018)

Rai 3 HD

Quale percentuale dei gas serra viene emessa dalle nostre attività?

Produzione industriale (cemento, acciaio, materie plastiche)	31%
Produzione di energia elettrica	27%
Agricoltura e allevamento	19%
Trasporti (aerei, camion, navi mercantili)	16%
Riscaldamento e condizionamento	7%



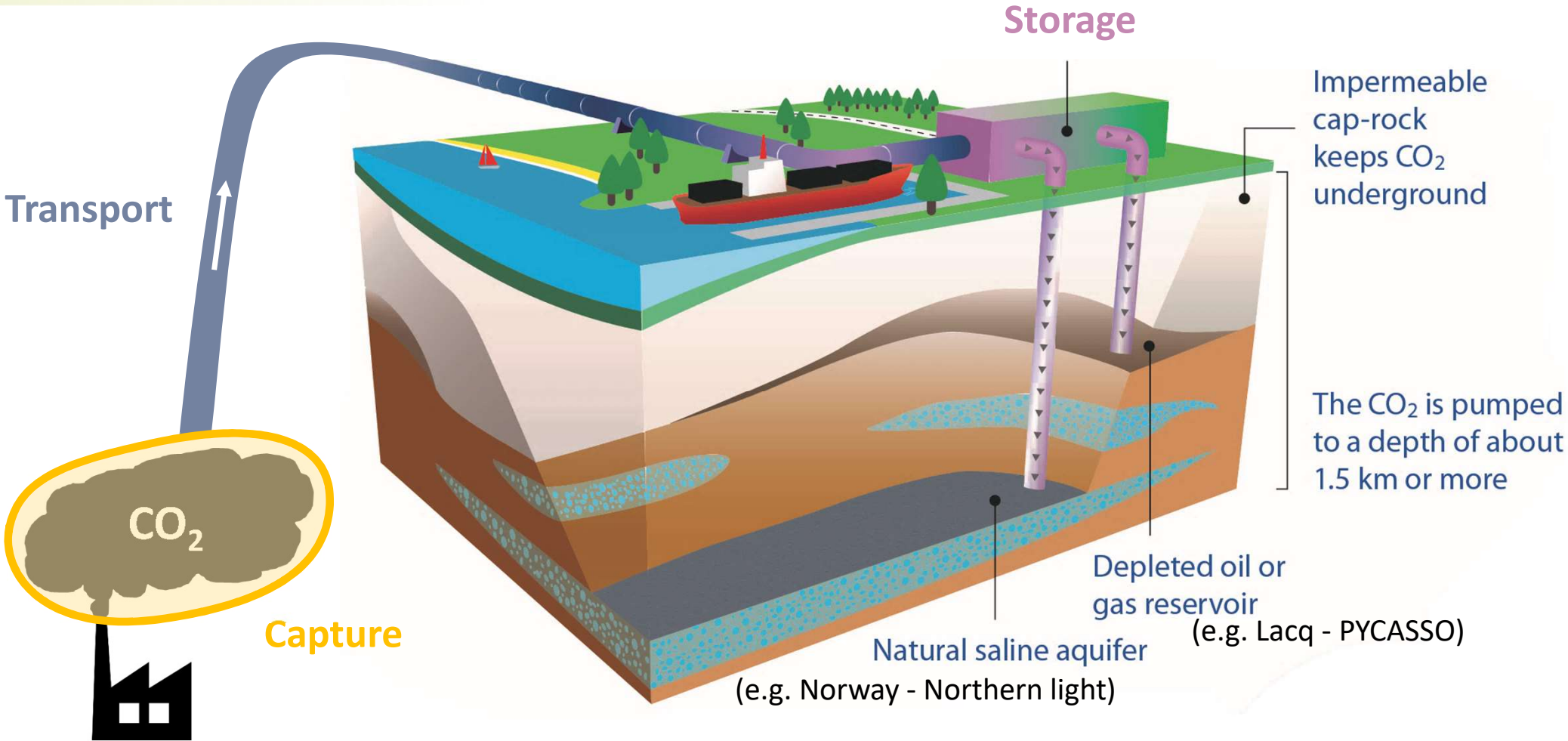
#CTCF

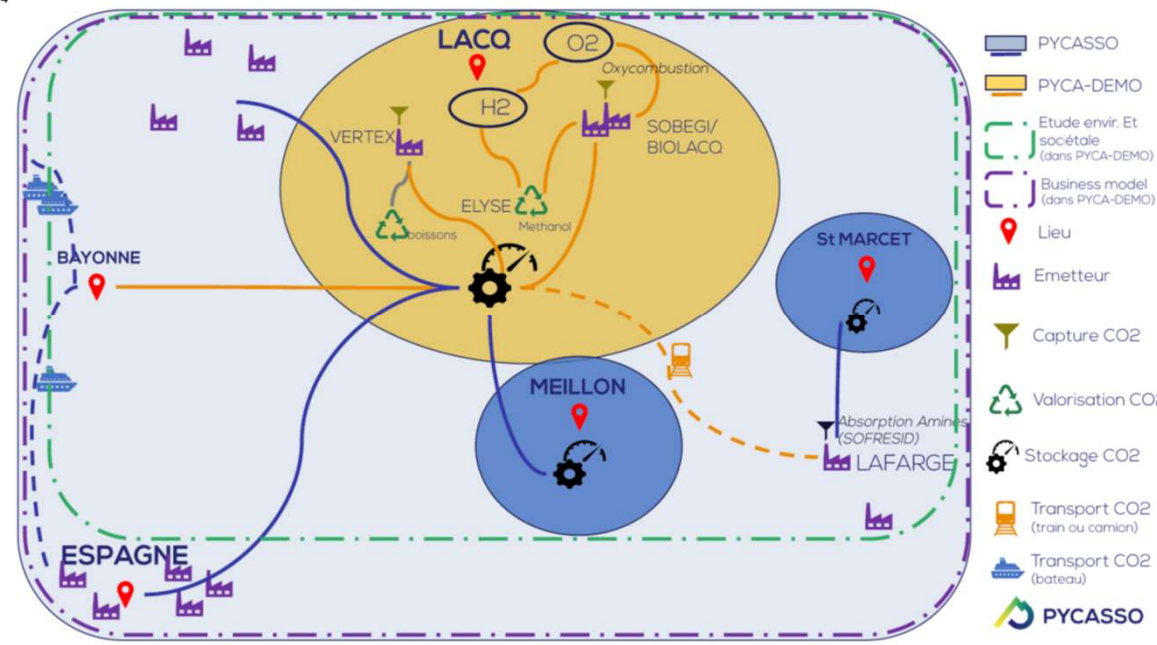
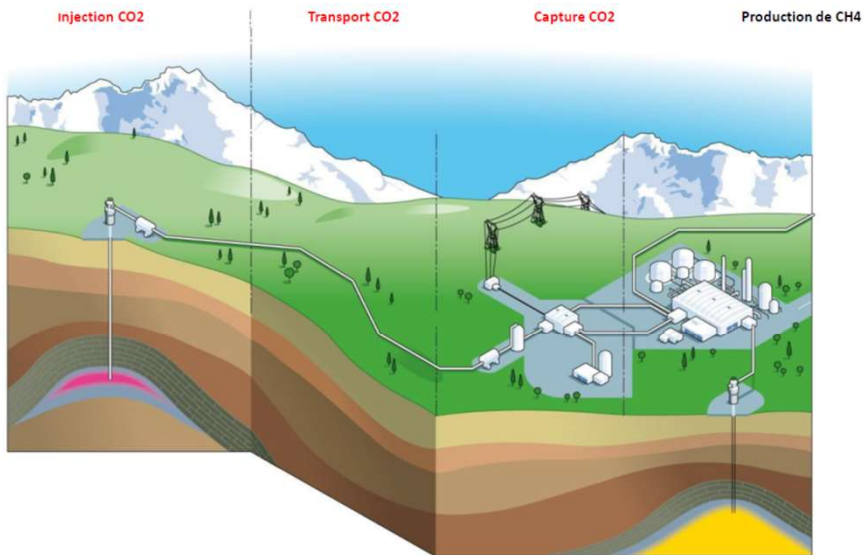
IN COLLEGAMENTO
DA SEATTLE

Rai



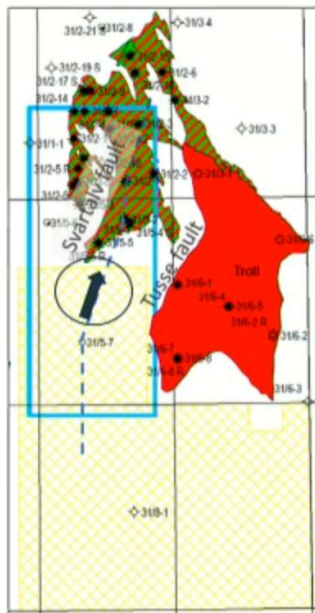
CARBON CAPTURE TRANSPORT & STORAGE



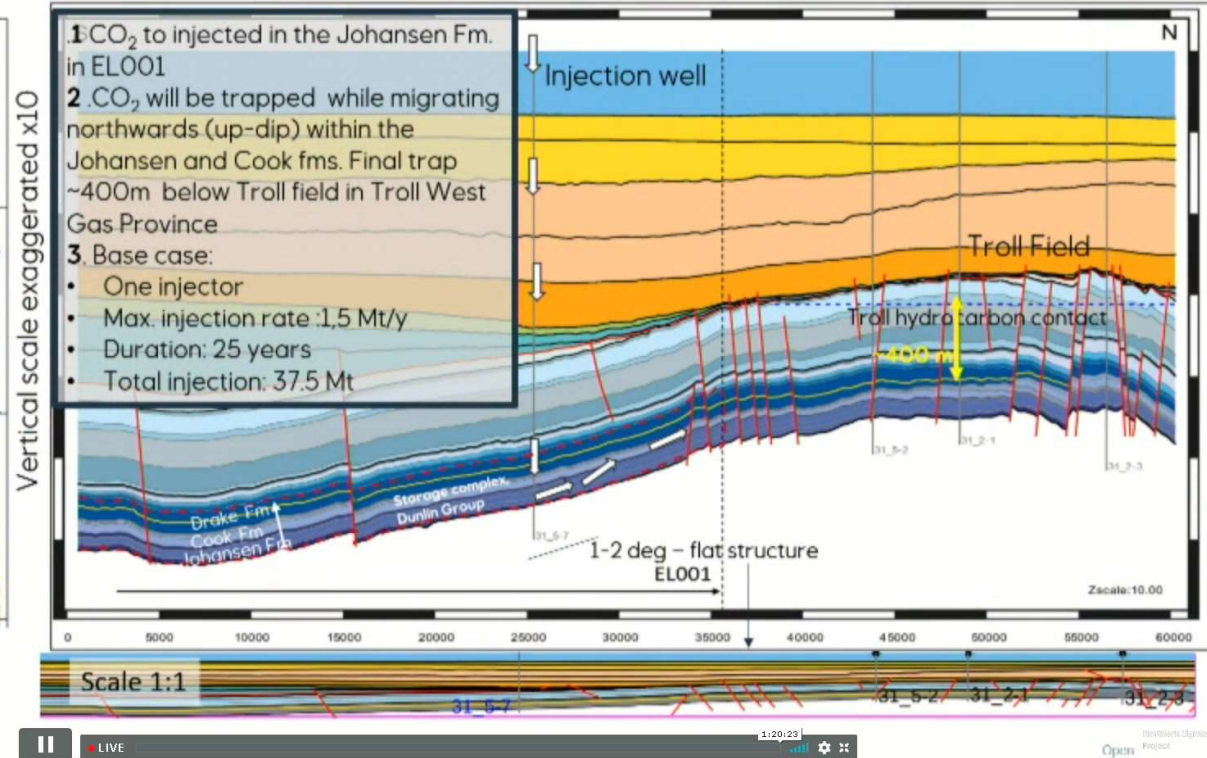


NORTHERN LIGHTS

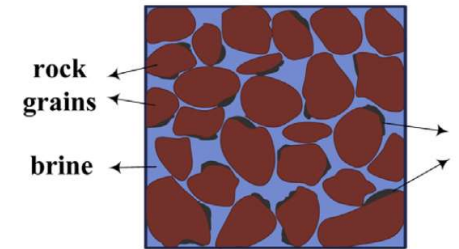
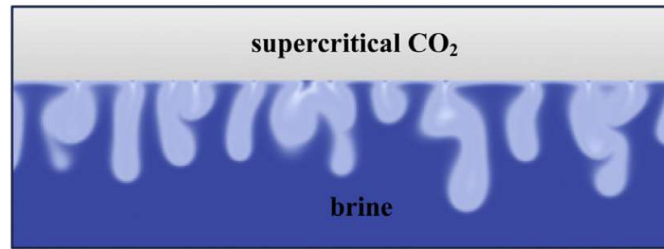
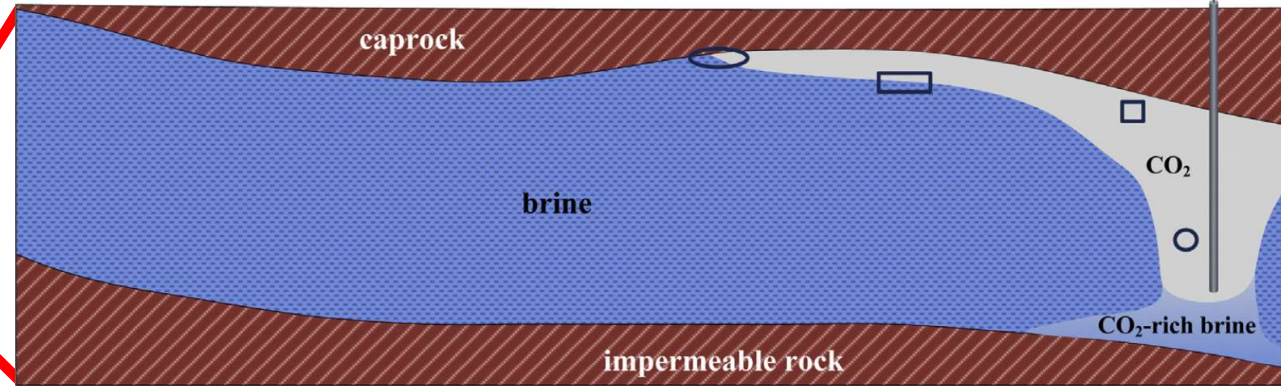
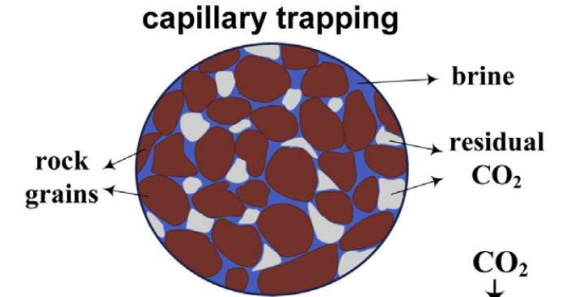
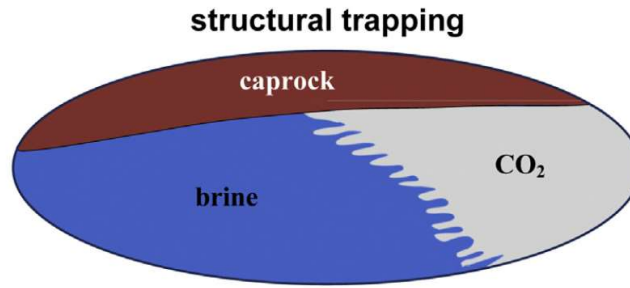
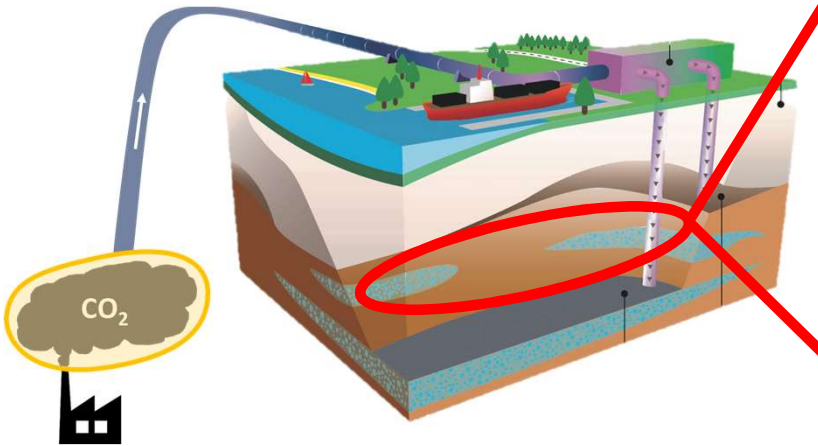
The CO₂ storage concept – confirmed now by the Eos well (31/5-7).



TWOP - Troll Vest ojeprovin
 TWGP - Troll Vest gasprovin



CARBON CAPTURE TRANSPORT & STORAGE



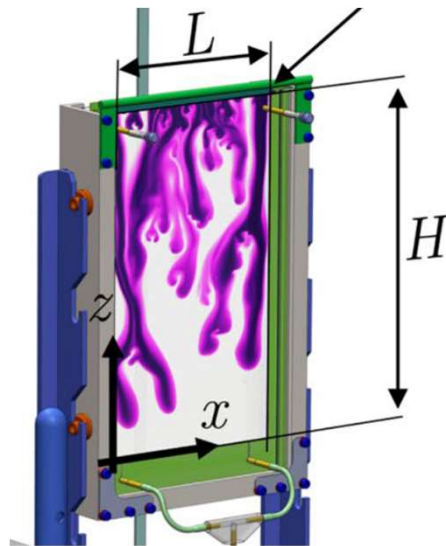
Emami-Meybodi et al. (2015))

STATE-OF-THE-ART

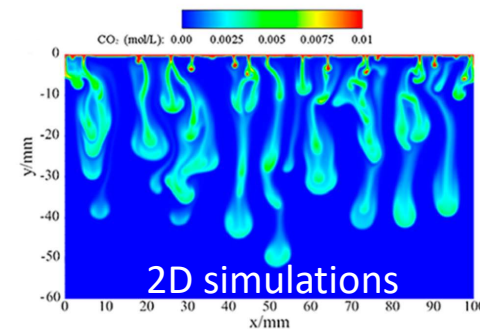
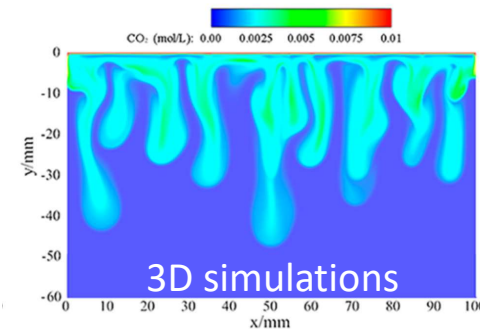
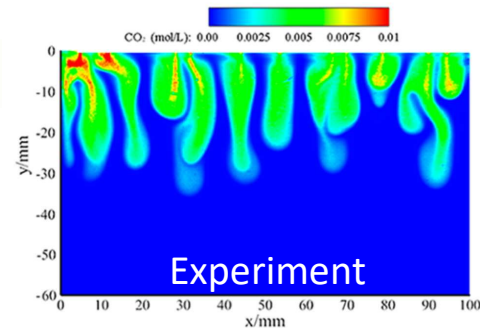


Hele-Shaw cell

- Quasi-2D
- Mimics a porous medium
- Rayleigh-Darcy regime
- Atmospheric pressures

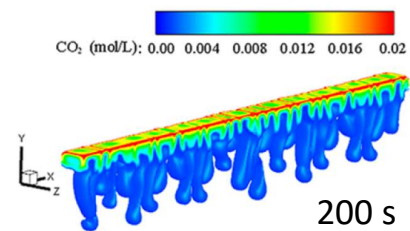
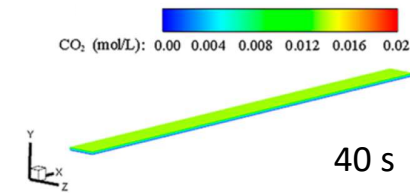


Alipour et al. (2020)

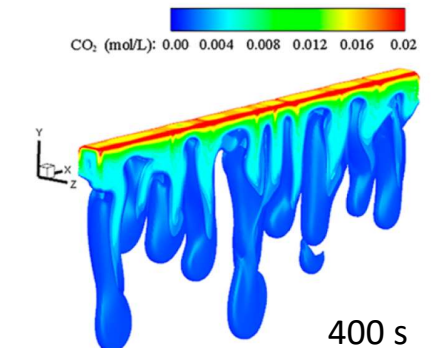
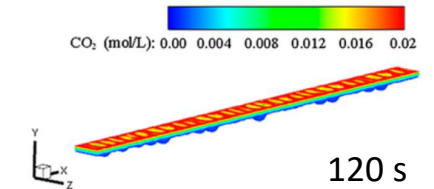


Zhang et al. (2020)

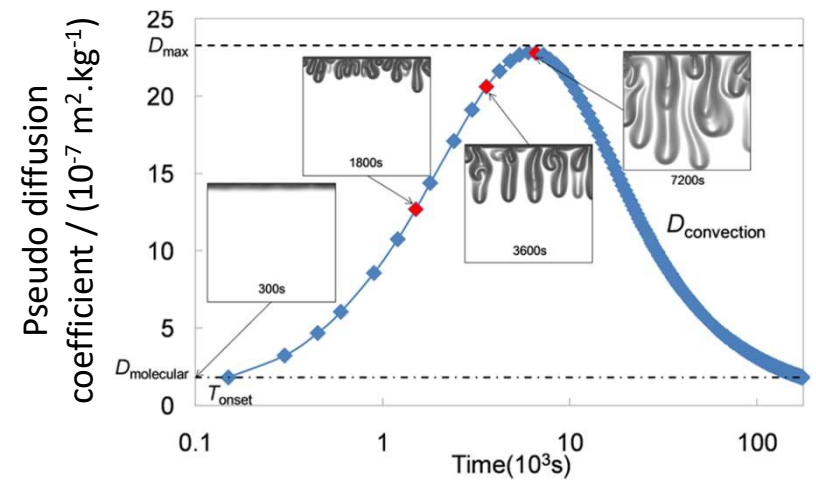
3D simulations



Zhang et al. (2020)



PVT cell



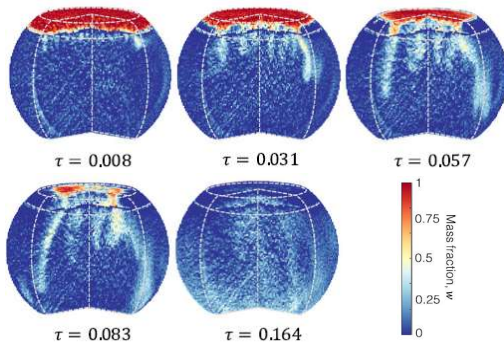
Tang et al. (2020)

STATE-OF-THE-ART



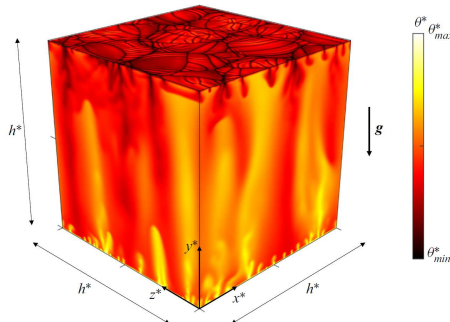
Rayleigh-Darcy
(3D porous medium)

$10^{-19} - 10^{-7}$



Liyanage et al. (2018)

Convective dissolution experiments in porous medium with X-ray tomography

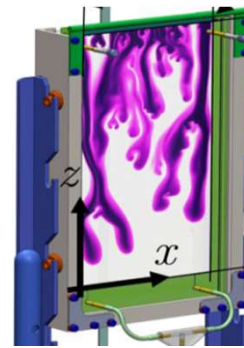


Pirozzoli et al. (2020)

Simulations of heat convection in porous media

Rayleigh-Darcy – Hele-Shaw
(2D free medium)

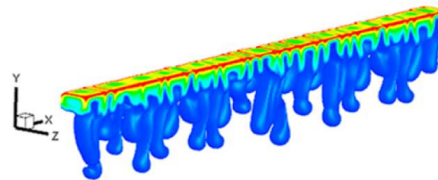
$10^{-9} - 10^{-7}$



Alipour et al. (2020)

Convective dissolution experiments in Hele-Shaw cell

CO₂ (mol/L): 0.00 0.004 0.008 0.012 0.016 0.02



Zhang et al. (2020)

Simulations of convective dissolution in Hele-Shaw-like configurations

Rayleigh-Taylor
(3D free medium)

1

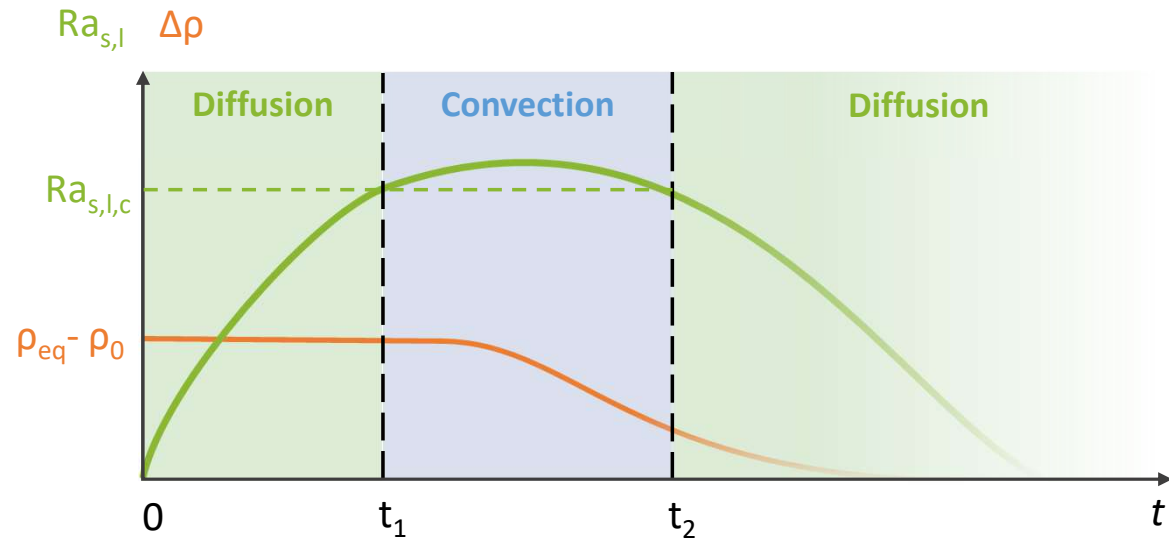
Regime
→
Permeability / m²

- Development of a reliable experimental apparatus and methodology
- Shadowgraph using visible radiations
- Better understand convective dissolution in free medium (e.g., CO₂ capture, safety of nuclear generation, protein crystallisation, etc.)
- Starting point in CO₂ES project before porous medium

FUNDAMENTALS



- p_0, p_{eq} Initial and equilibrium pressures
- c_0, c_{eq} Initial and equilibrium concentrations of CO_2 in brine
- ν, η Kinematic and dynamic viscosities
- ρ_b, ρ_{BL} Densities of brine and BL
- D Fick diffusion coefficient
- g Gravitational acceleration
- d, h Thicknesses of the boundary and brine layers
- $Ra_{s,l}$ Local solutal Rayleigh number

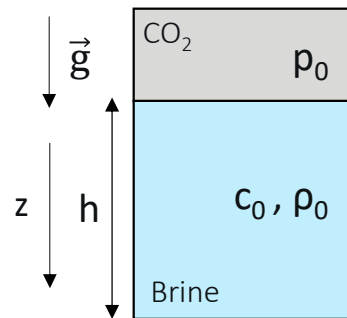


GRAVITATIONAL INSTABILITY

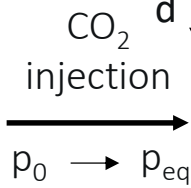
$$Ra_{s,l} = \frac{\beta \vec{g} \cdot \vec{\nabla} c d^4}{\nu D}$$

INITIAL EQUILIBRIUM (t=0)

$$\rho_{CO_2,g} < \rho_b$$

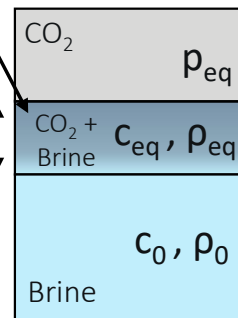


Boundary layer (BL)

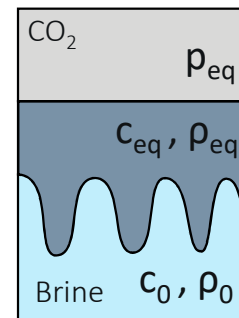


DIFFUSION (0 < t < t_1)

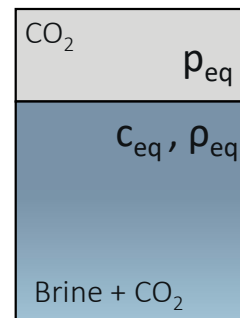
$$\rho_{BL} > \rho_b$$



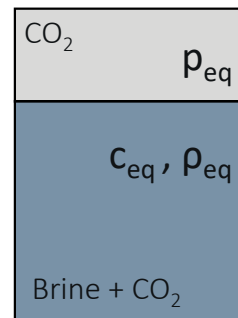
CONVECTION (t_1 < t < t_2)



DIFFUSION (t > t_2)

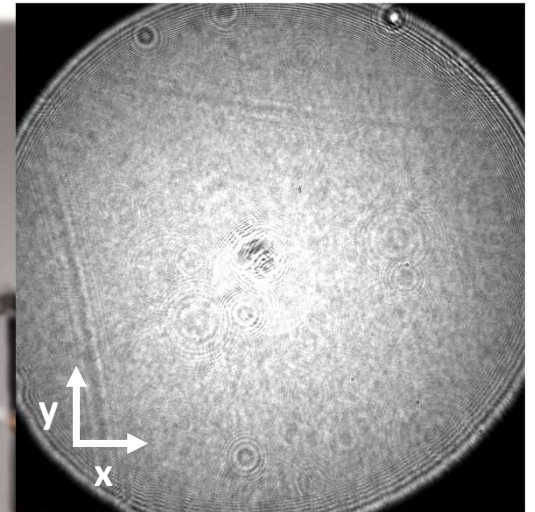
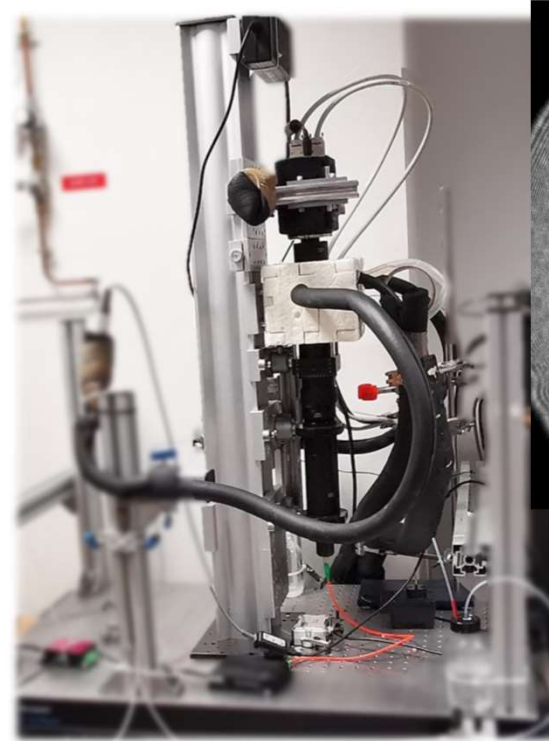
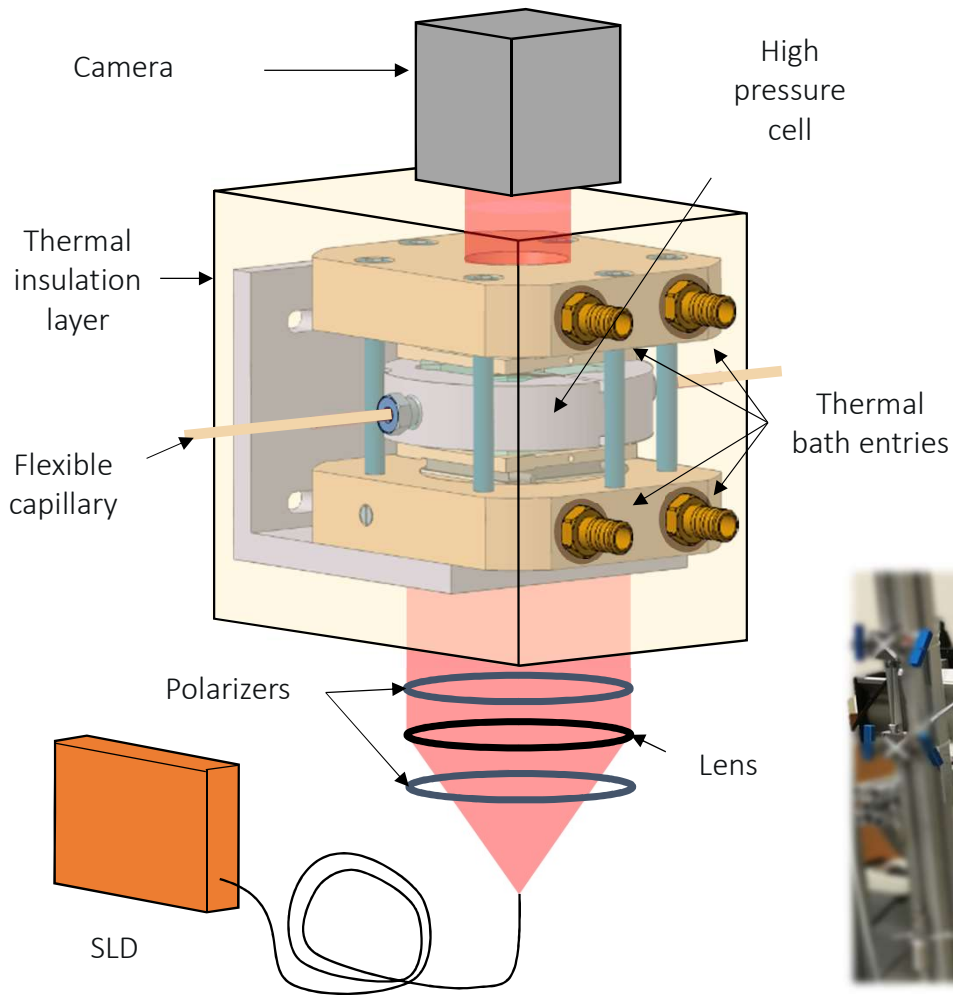


FINAL EQUILIBRIUM

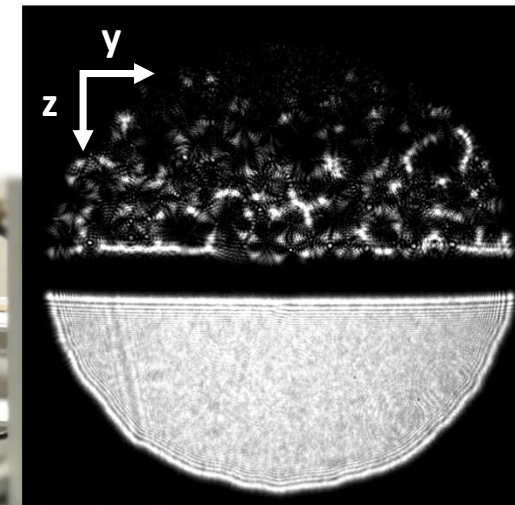
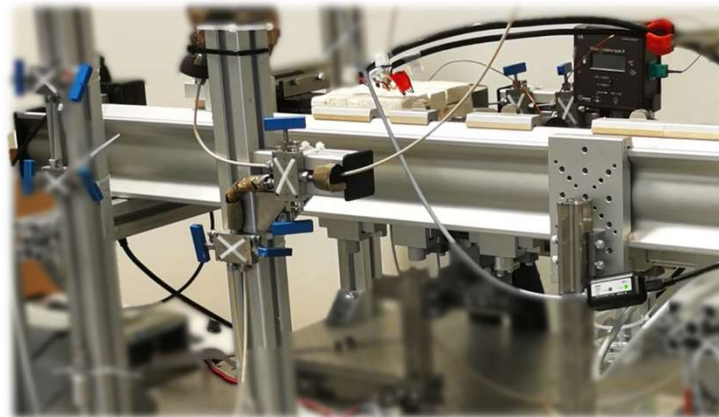


$\delta\rho \rightarrow \delta n \rightarrow$ Intensity variations

EXPERIMENTAL SET-UP



Vertical configuration



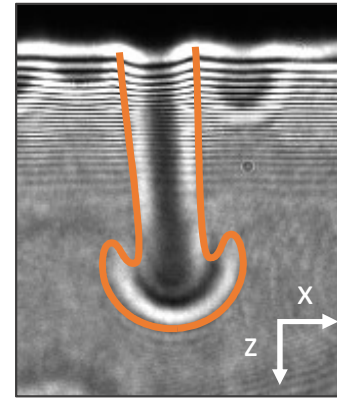
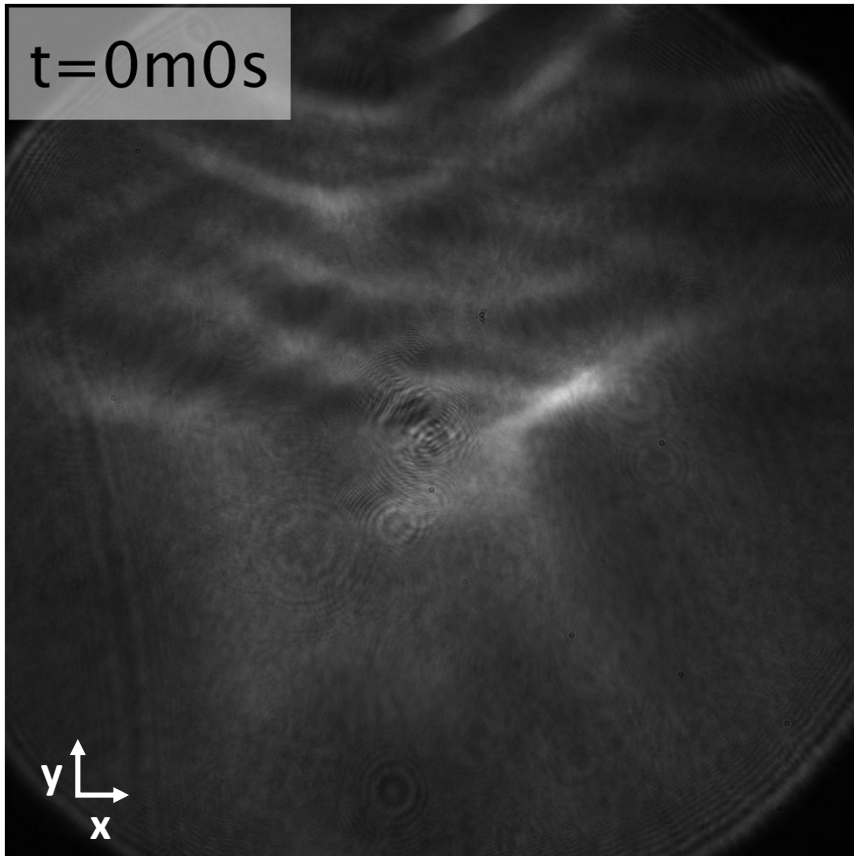
Horizontal configuration

PRELIMINARY ANALYSIS



Observation from above

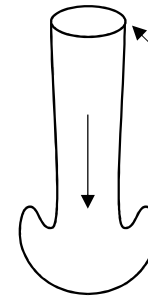
$p_0 = 0.1 \text{ MPa}$ $p_{eq} = 2.1 \text{ MPa}$
Pure water



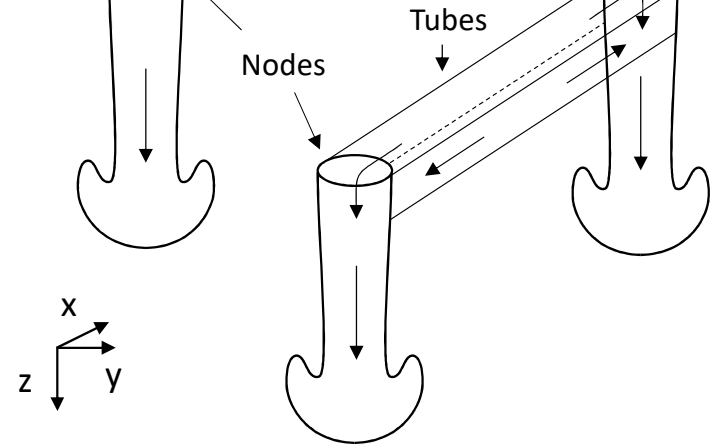
Hele-Shaw like configuration

Transversal observation

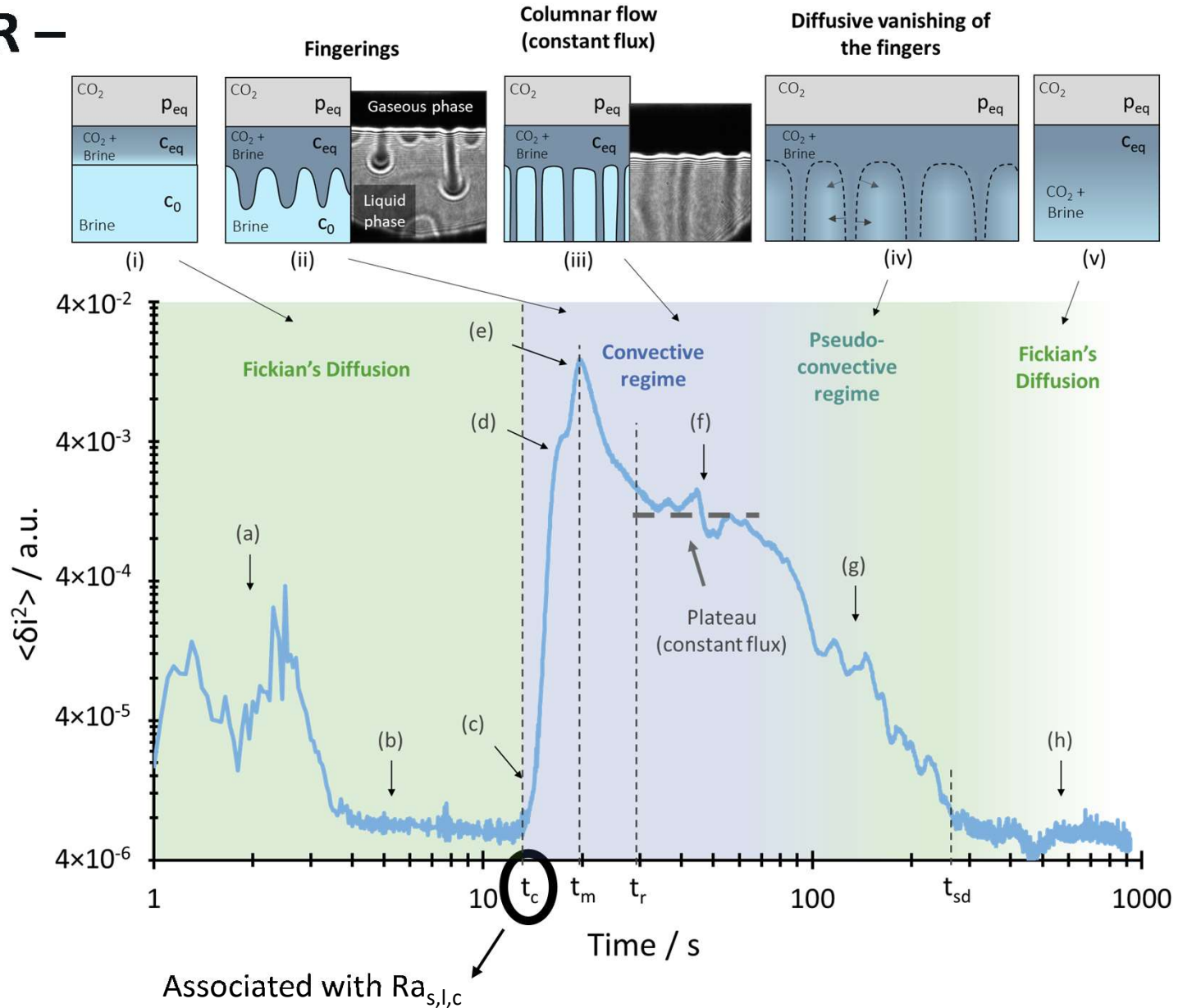
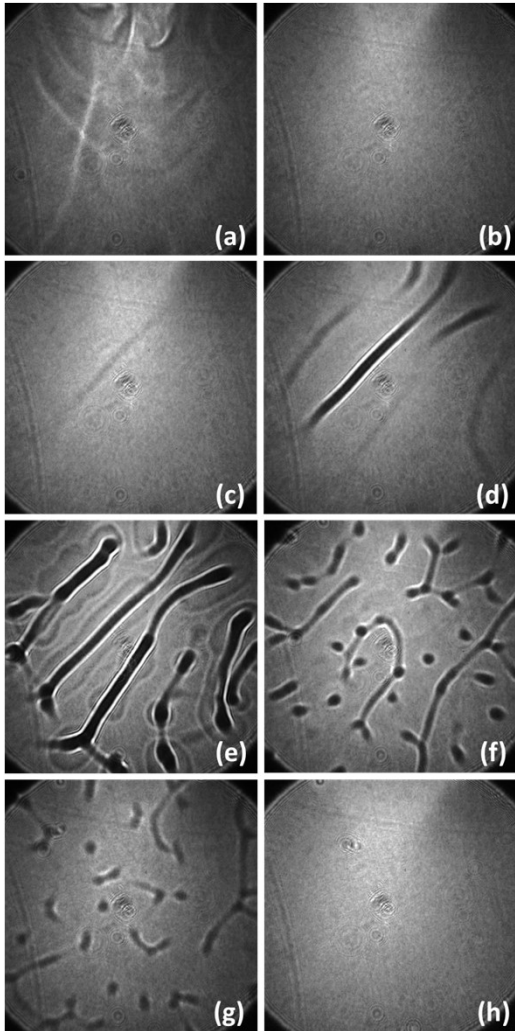
Isolated Finger



Linked Fingers



TEMPORAL BEHAVIOUR – VARIANCE ANALYSIS



GENERAL BEHAVIOUR

$$t_c = \frac{h^2}{\pi D} Ra_{s,l,c}^{2/3} Ra_s^{-2/3}$$

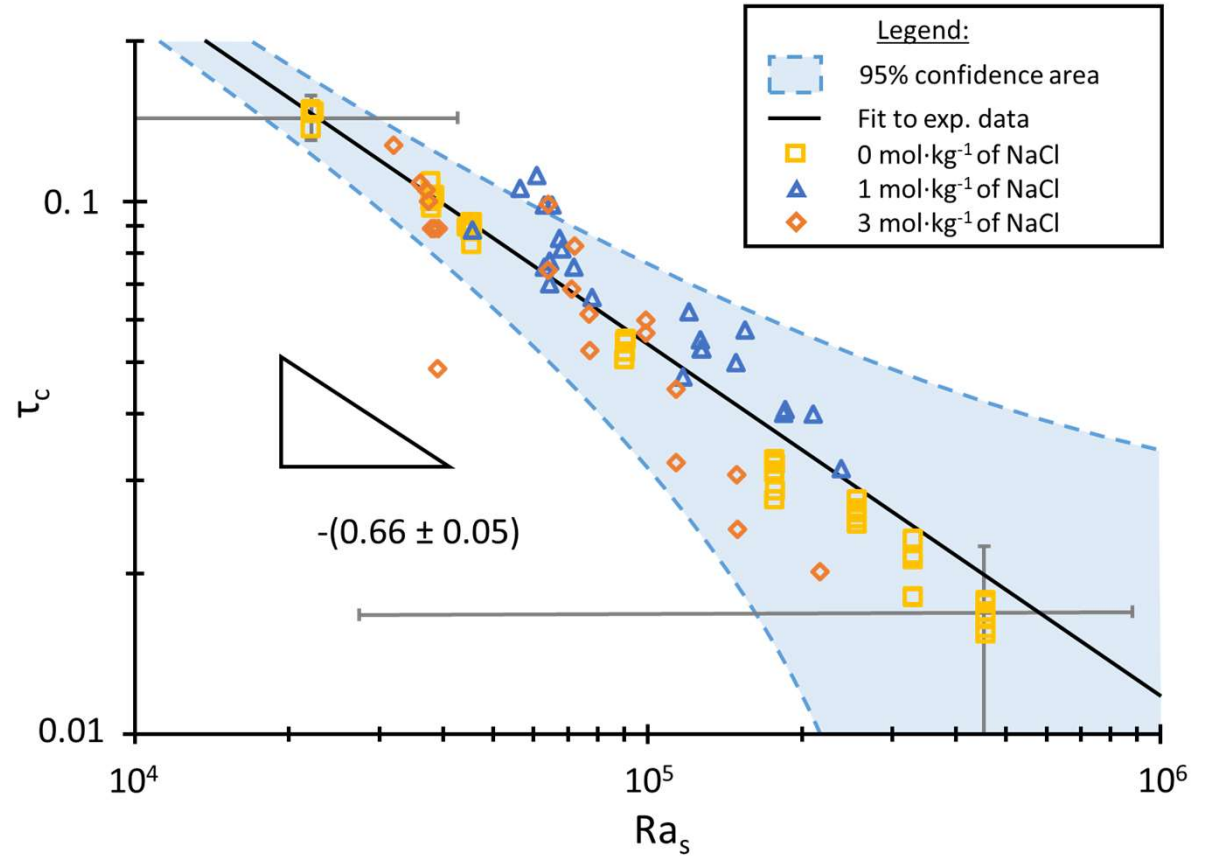
Diffusive time over the entire brine layer $t_D = \frac{h^2}{\pi D}$

Dimensionless critical time $\tau_c = \frac{t_c}{t_D}$

Universal law in free media

$$\tau_c = \left(\frac{Ra_s}{Ra_{s,l,c}} \right)^{-2/3}$$

P. Fruton *et al*, Nature Comm. submitted (2021)



Cédric GIRAUDET
postdoc



Paul FRUTON
PhD and postdoc



Happiness IMUETINYAN
PhD



Aziza NAURUZBAEVA
stage MS

PERSPECTIVES

Development of a new sample cell

Larger & thicker

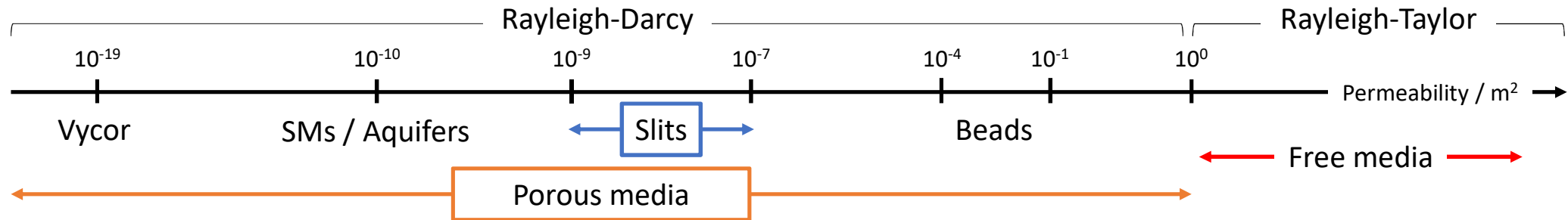
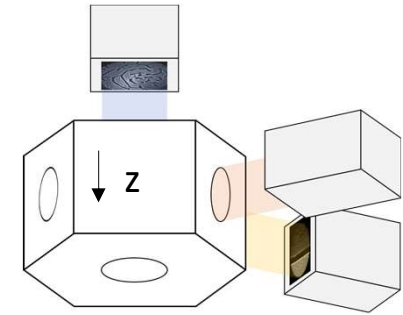


Avoid meniscus effects
Better control of h

Multiple observations

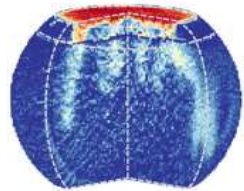


Reconstruction of the concentration-field



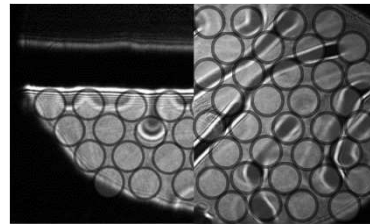
IN PREPARATION

Core of water permeable rocks



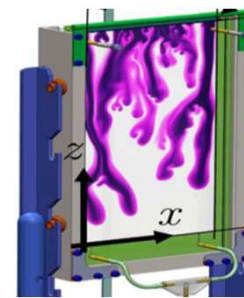
Platform DMEX
X-ray tomography
(3D)

Beads / Silica monoliths (SMs) / Vycor glass

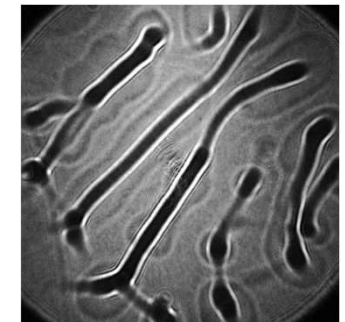


Refractive index matching
(3D)

Thin slits



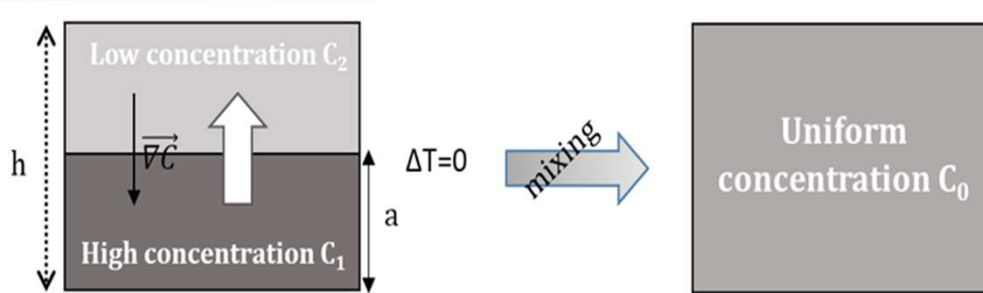
Hele-Shaw
(2D)



CO2ES
(3D)



FREE DIFFUSION OF SALT MIXTURES



Initial conditions

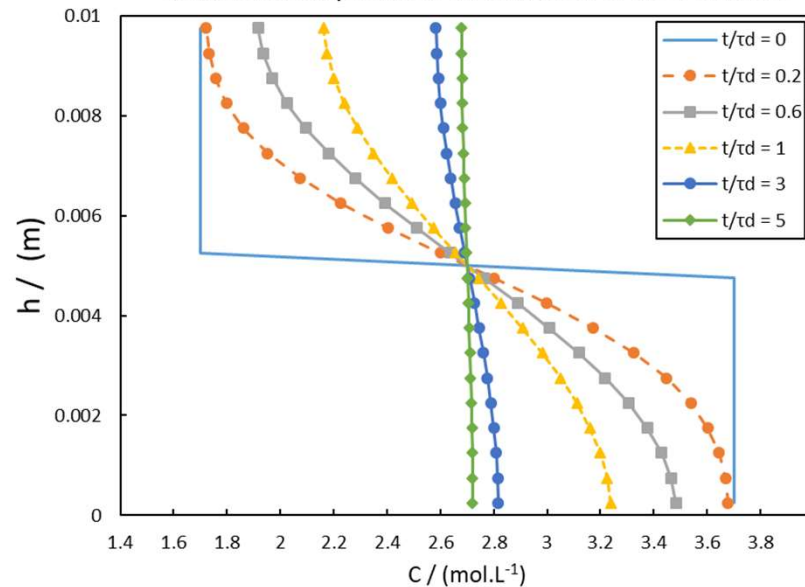
$$C(z, 0) = \begin{cases} C_1, & 0 < z < a \\ C_2, & a < z < h \end{cases}$$

$$C_{\text{mean}} = (C_1 + C_2) / 2$$

$$\Delta C = C_1 - C_2$$

Fickian diffusion: $\vec{j} = -\rho D \vec{\nabla} C$

Concentration profile of the diffusion of NaCl in water

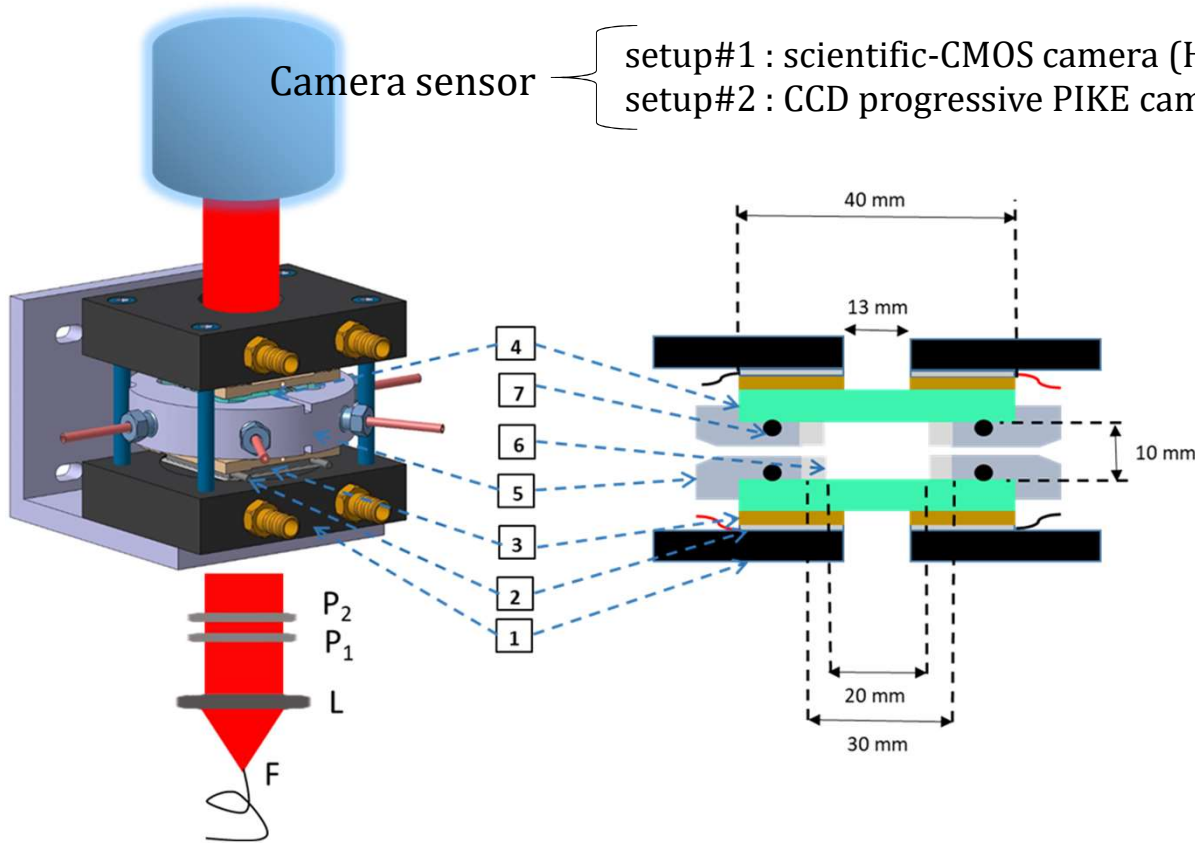


$$\tau_d = \frac{(h/2)^2}{(\pi D)} \approx 1.43 \text{ hours}$$

- $C_1 = 3.7 \text{ mol.L}^{-1}$
- $C_2 = 1.7 \text{ mol.L}^{-1}$
- $C_0 = 2.7 \text{ mol.L}^{-1}$
- $D = 1.55 \times 10^{-5} \text{ cm}^2 \cdot \text{s}^{-1}$
- $h = 1 \text{ cm}$

Plot of the concentration profile with PHREEQC

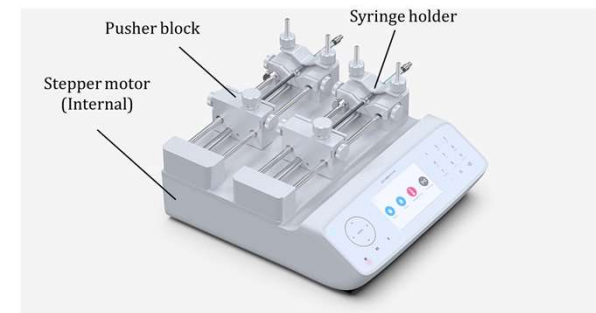
DIFFUSION CELL & SHADOWGRAPH SET-UP



1	Water circulation
2	Peltier elements
3	Aluminium plates
4	Sapphire windows
5	Stainless steel annulus
6	Teflon ring
7	Viton O-rings

Experimental setup

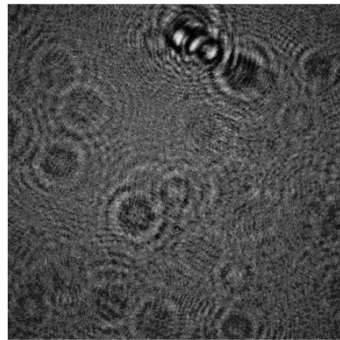
A. T. Ndjaka *et al*, Eur. Phys. J. E submitted (2021).



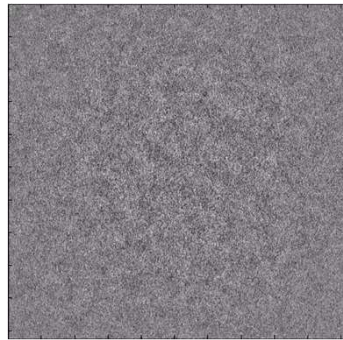
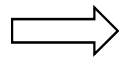
Fusion 4000 independent dual-channel infusion and withdrawal syringe pump from Chemyx.

DIFFERENTIAL DYNAMIC ALGORITHM

Free diffusion experiment of NaCl into water ($C = 2.7 \text{ mol.L}^{-1}$ and $\Delta C = 2 \text{ mol.L}^{-1}$).

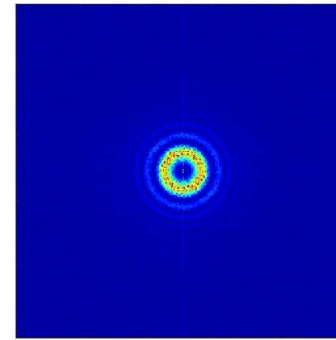
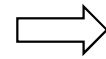


Near field image 1024x1024
pix² recorded image
 $I(\vec{x}, t)$



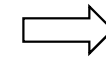
Difference between two images
separated by $\Delta t = 2 \text{ s}$

$$\Delta i(\vec{x}, t, \Delta t) = i(\vec{x}, t) - i(\vec{x}, t + \Delta t)$$



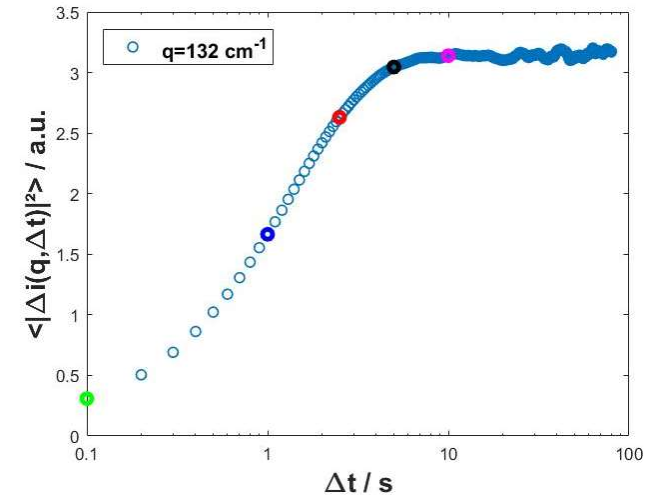
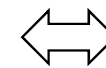
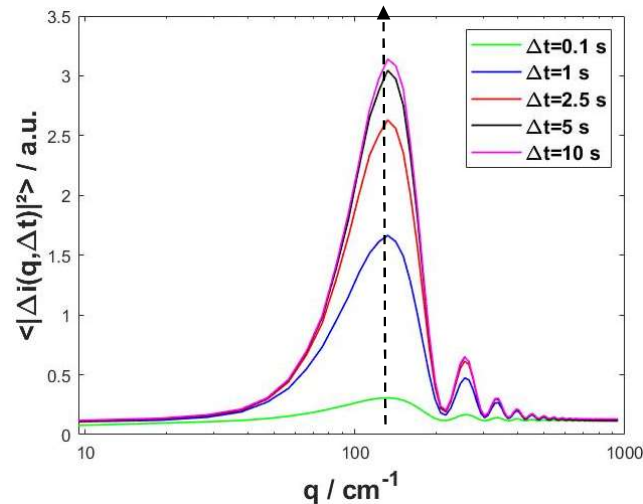
2D FFT of image differences

$$|\Delta i(\vec{q}, t, \Delta t)|^2 = |i(\vec{q}, t) - i(\vec{q}, t + \Delta t)|^2$$

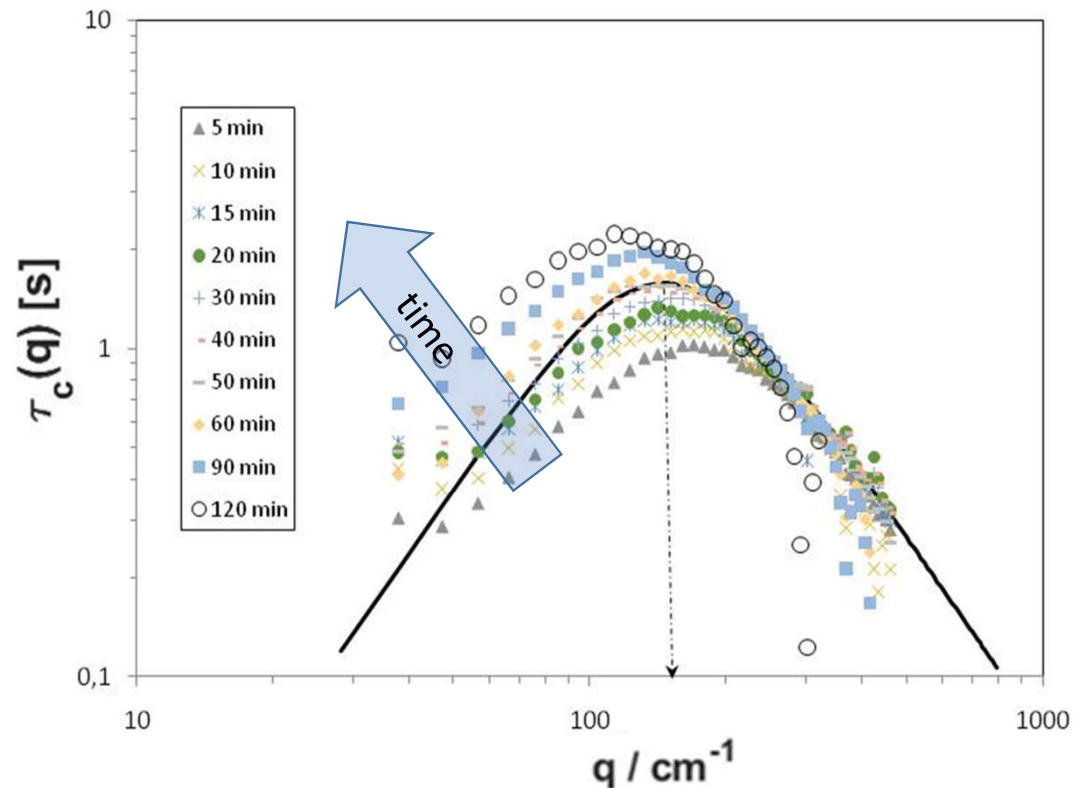


STRUCTURE FUNCTION

$$SF(q, \Delta t)$$



DYNAMIC ANALYSIS METHOD OF C-NEFS



Decay times of the c-NEFs as a function of the wave numbers and time after closing the inlet/outlet valves for the free-diffusion experiment :NaCl/water at $C = 2.7 \text{ mol.L}^{-1}$, $\Delta C = 2 \text{ mol.L}^{-1}$ and $T=25 \text{ }^\circ\text{C}$.

Fitting the SFs in the wave number range from 30 to 500 cm^{-1}

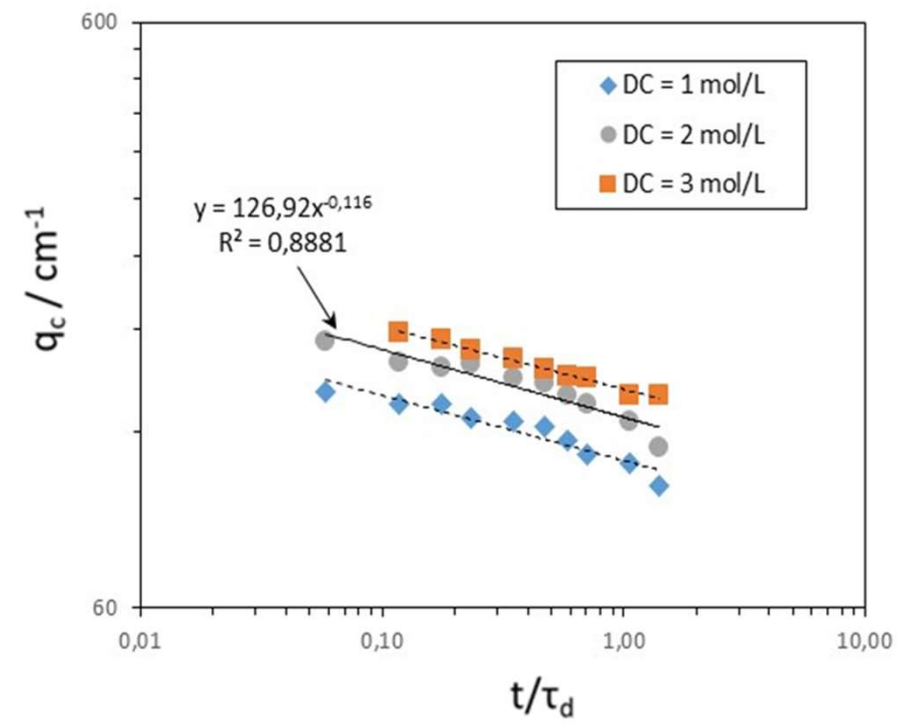
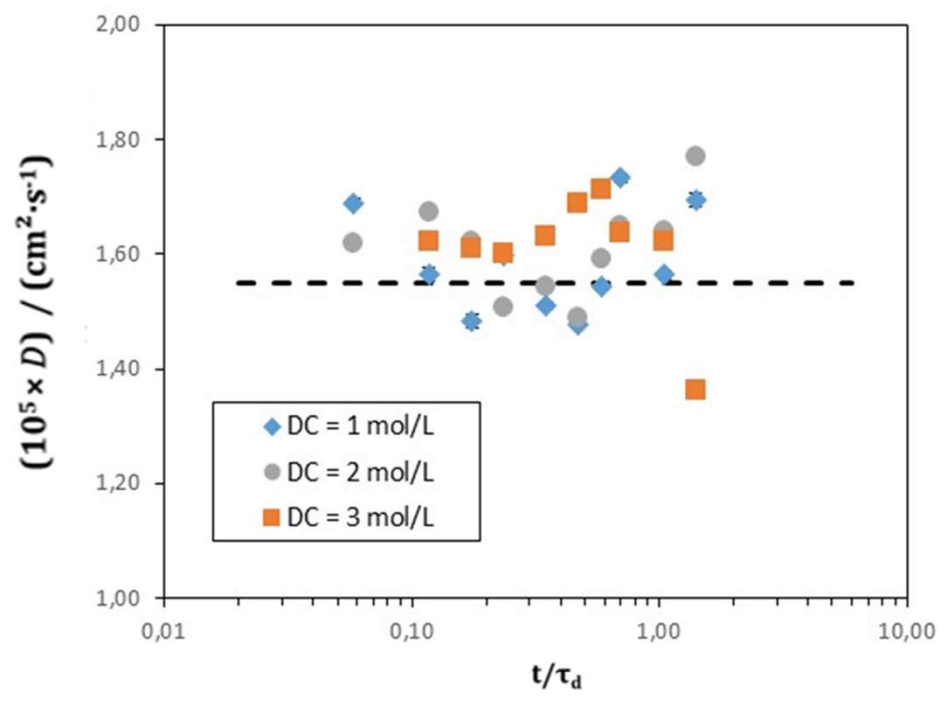
$$\tau_c(q) = \frac{1}{Dq^2 \left[1 + \left(\frac{q_c}{q} \right)^4 \right]}$$

$$q_c(t) = \sqrt[4]{\frac{\beta g(C_1 - C_2)}{\nu D \sqrt{4\pi D t}}}$$

RESULTS: MEASUREMENTS OF D SALTS IN WATER

• Sodium Chloride / water mixture: $C = 2,7 \text{ mol.L}^{-1}$, $T = 25 \text{ }^\circ\text{C}$ & $\Delta T = 0 \text{ }^\circ\text{C}$ & $P=1 \text{ atm}$

Diffusive time $\tau_d = \frac{(h/2)^2}{(\pi D)} \approx 86 \text{ min.}$



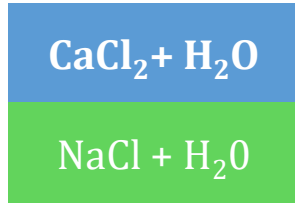
Rard and Miller (1979), *Journal of Solution Chemistry*, **8**(10), 701-716

M. Schraml *et al*, *Eur. Phys. J. E Soft Matter*. 2021 Oct 18;44(10):128.

$$q_c(t) = \sqrt[4]{\frac{\beta g (C_1 - C_2)}{\nu D \sqrt{4\pi D t}}} \propto t^{-0,125}$$

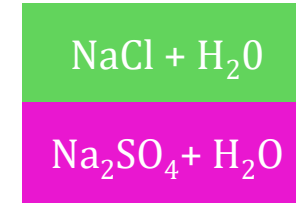
SUPERIMPOSITION OF TWO AQUEOUS LAYERS OF NON-REACTIVE SALTS (OBSERVATIONS PARALLEL TO THE GRAVITY)

$T = 25\text{ }^{\circ}\text{C}$ & $\Delta T = 0\text{ }^{\circ}\text{C}$ & $P = 1\text{ atm}$



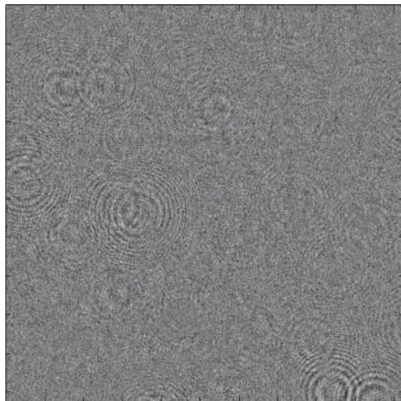
$C_{\text{CaCl}_2} = 0.46\text{ mol.L}^{-1}$
($\rho = 1.038962\text{ g.cm}^{-3}$)

$C_{\text{NaCl}} = 2.637\text{ mol.L}^{-1}$
($\rho = 1.098622\text{ g.cm}^{-3}$)

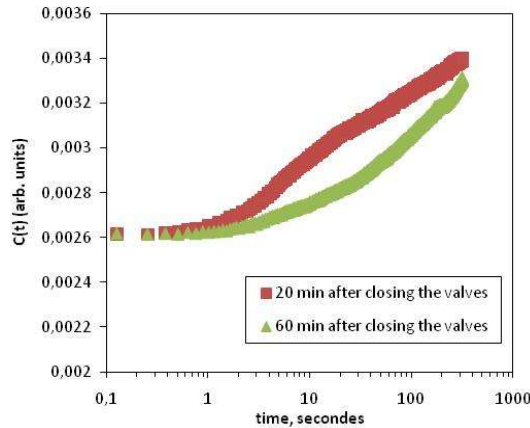


$C_{\text{NaCl}} = 2.637\text{ mol.L}^{-1}$
($\rho = 1.098622\text{ g.cm}^{-3}$)

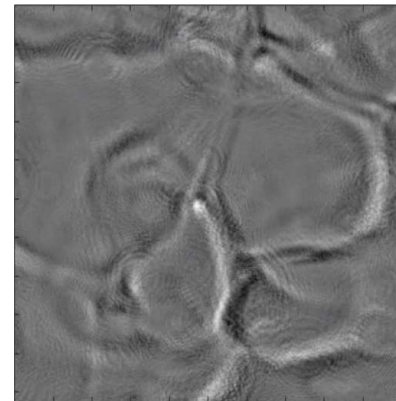
$C_{\text{Na}_2\text{SO}_4} = 1.484\text{ mol.L}^{-1}$
($\rho = 1.168653\text{ g.cm}^{-3}$)



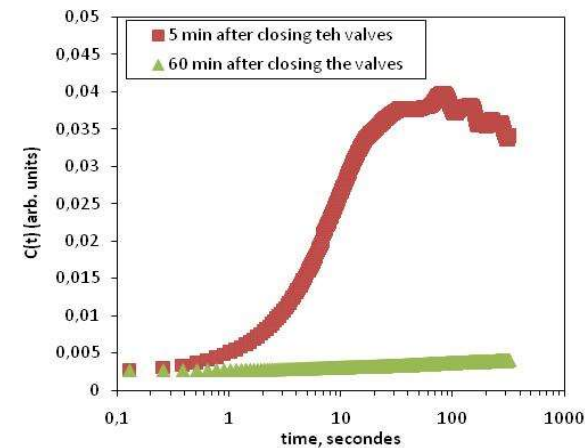
Normalized image differences
20 minutes after closing



Contrast of shadowgraph image sequences
 $C(t)$ as a function of time and for different
moments after closing the valves



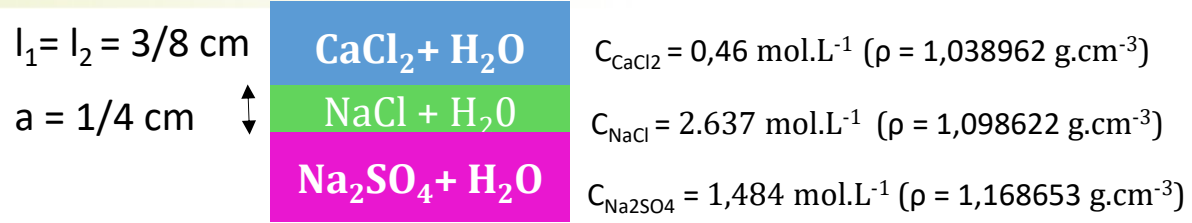
Normalized image differences
20 minutes after closing



Contrast of shadowgraph image sequences
 $C(t)$ as a function of time and for different
moments after closing the valves

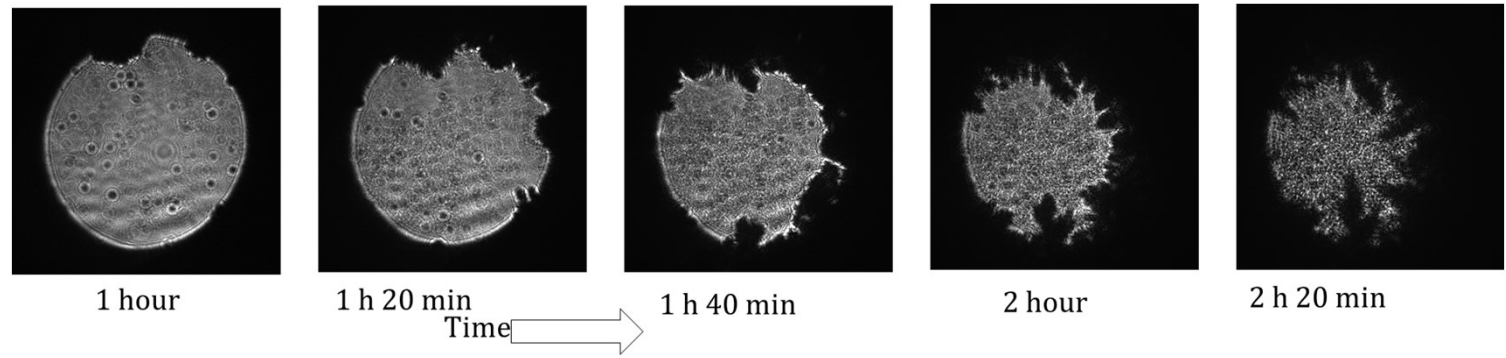


SUPERIMPOSITION OF TWO AQUEOUS LAYERS OF REACTIVE SALTS: (OBSERVATIONS PARALLEL TO THE GRAVITY)

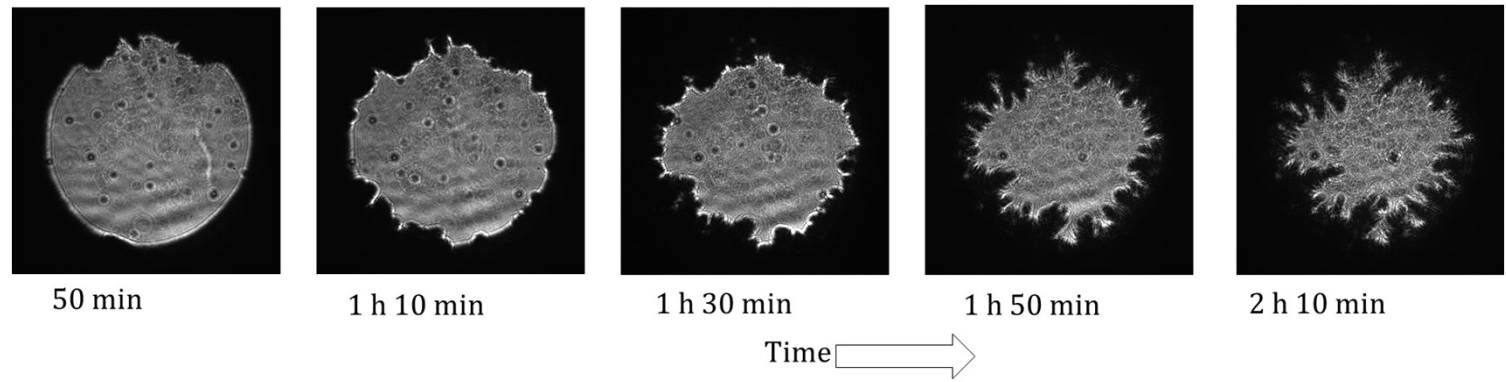


Shadowgraph images during the reactive transport

$T = 25 \text{ }^\circ\text{C}$ & $\Delta T = 0 \text{ }^\circ\text{C}$



$T = 25 \text{ }^\circ\text{C}$ & $\Delta T = 20 \text{ }^\circ\text{C}$



SUPERIMPOSITION OF TWO AQUEOUS LAYERS OF REACTIVE SALTS (OBSERVATIONS PARALLEL TO THE GRAVITY) : IMPACT OF ΔT

$$l_1 = l_2 = 3/8 \text{ cm}$$

$$a = 1/4 \text{ cm}$$



$$C_{\text{CaCl}_2} = 0,46 \text{ mol.L}^{-1} (\rho = 1,038962 \text{ g.cm}^{-3})$$



$$C_{\text{NaCl}} = 2.637 \text{ mol.L}^{-1} (\rho = 1,098622 \text{ g.cm}^{-3})$$

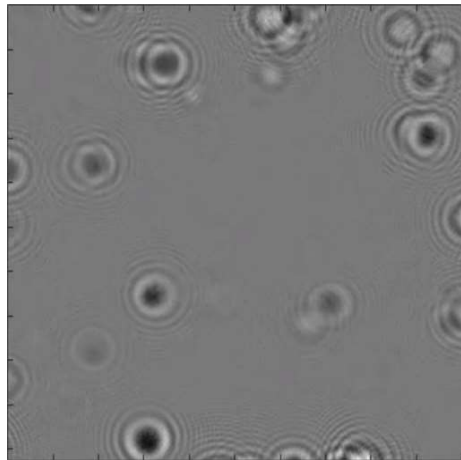
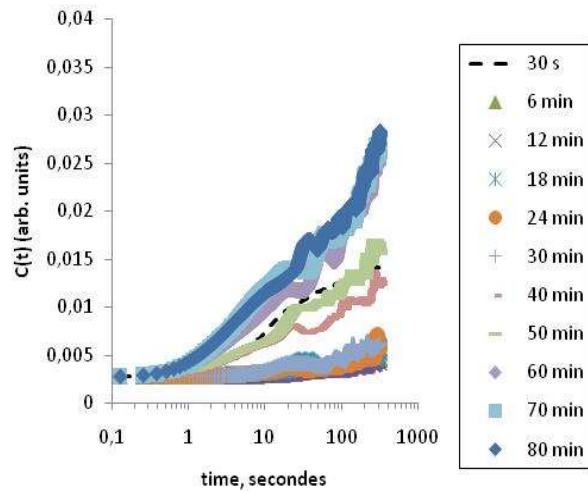


$$C_{\text{Na}_2\text{SO}_4} = 1,484 \text{ mol.L}^{-1} (\rho = 1,168653 \text{ g.cm}^{-3})$$



Light beam

- Precipitation speed depends on the thickness of the buffer solution : $t_p = \frac{a^2}{\pi D}$ and $\Delta t_p = 2t_p \frac{\Delta a}{a}$



Henri
BATTERER
Ass. Pr.



Ange Tatiana
NDJAKA
PhD and
postdoc

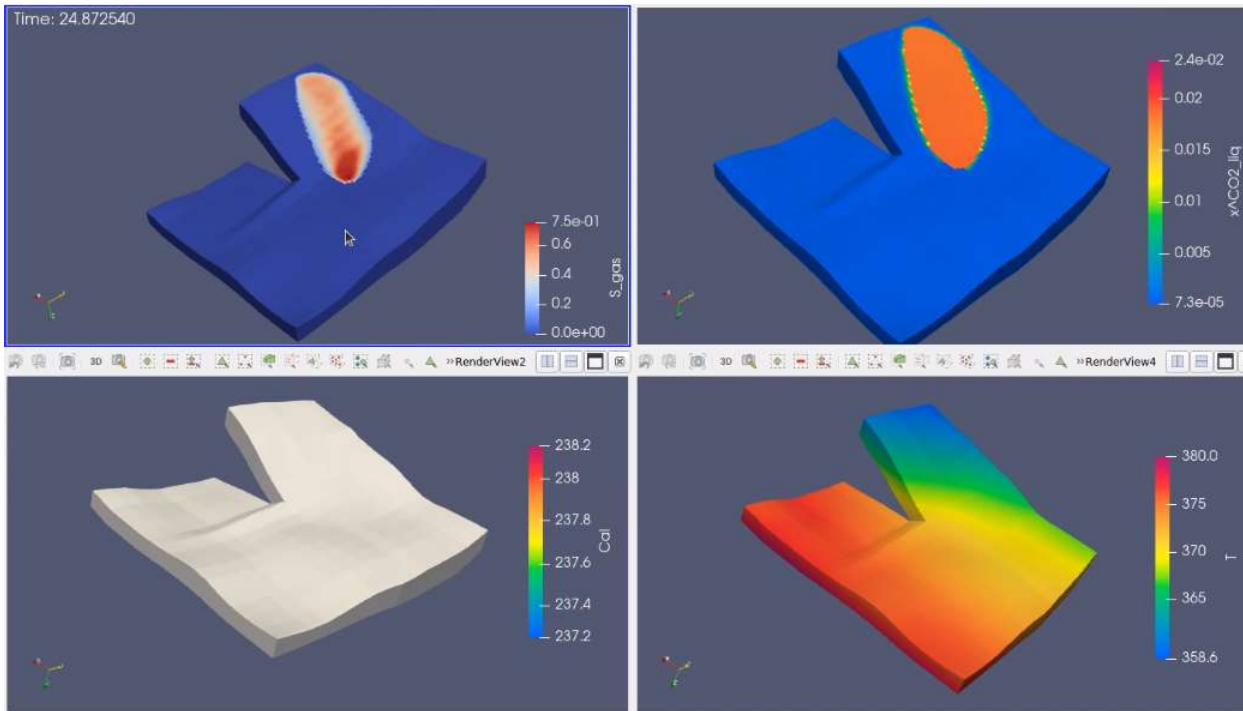


Christian
OKO
PhD



Rizwan
MINHAS
stage MS

NUMERICAL SIMULATIONS AT BASIN SCALE



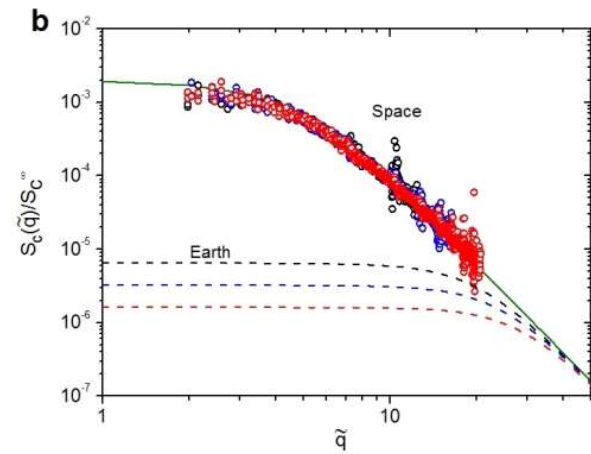
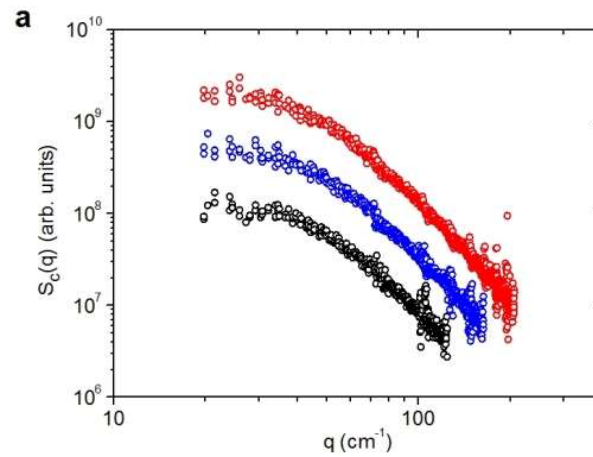
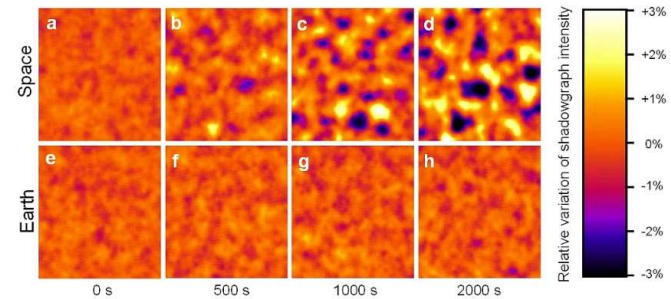
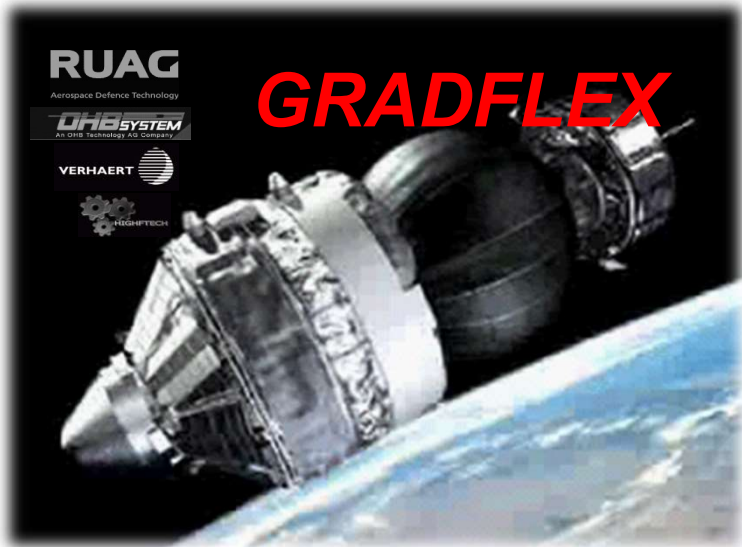
**Brahim
AMAZIANE**
Ass. Pr.



**Nicolas
PILLARDOU**
PhD



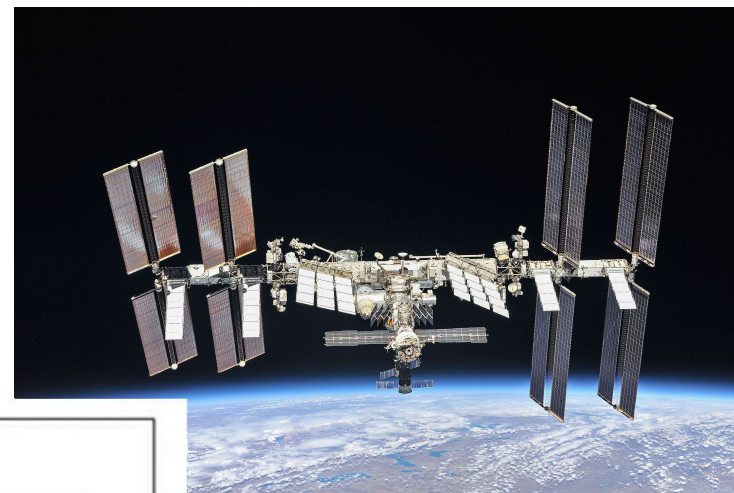
GRADFLEX – FOTON M3 (2007)



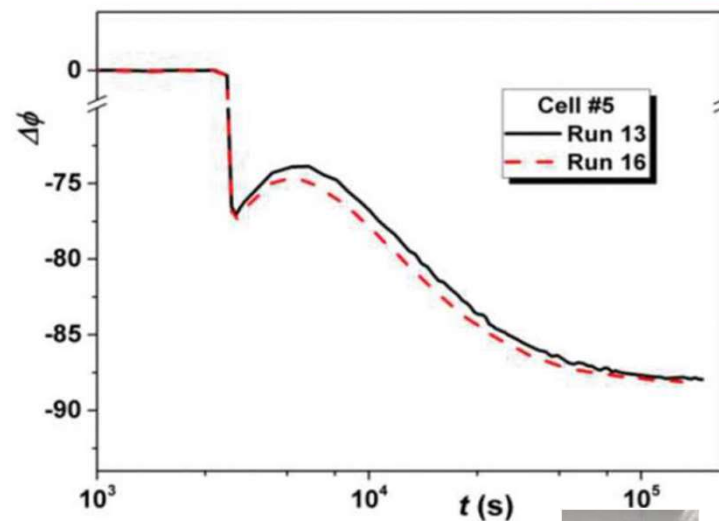
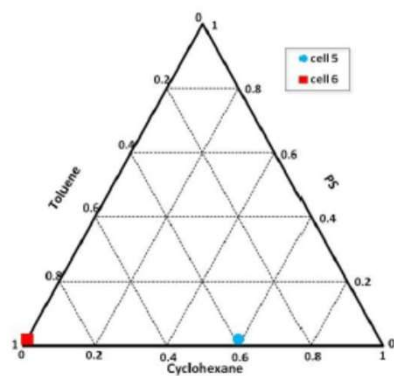
Vailati et al. (2011)

Croccolo et al. (2016)

DCMIX #4 – ISS (2018)



Polystyrene
+ Toluene + Cyclo-hexane



Mialdun *et al.*, EPJE

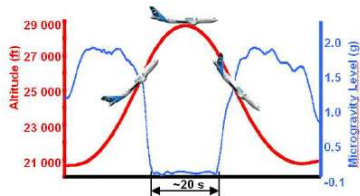


Henri
BATALLER
Ass. Pr.

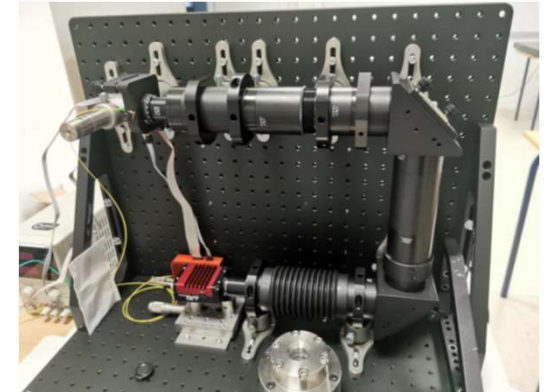


Loreto
GARCIA-
FERNANDEZ
postdoc

CO2EX – PARABOLIC FLIGHT (2019 AND 2020)



C. Giraudet et al. To be submitted



**Cédric
GIRAUDET**
postdoc



**Paul
FRUTON**
PhD and
postdoc



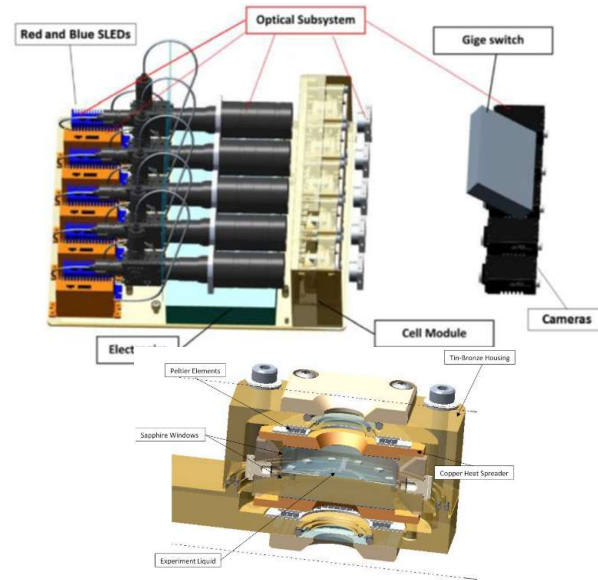
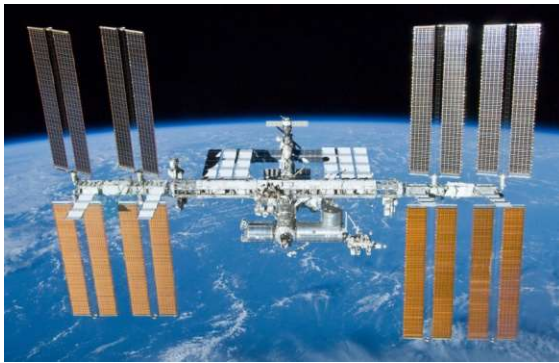
**Mohammed
CRAHA**
PhD



**Emma
LISOIR**
stage BS

GIANT FLUCTUATIONS – ISS (2025 - 2028)

Giant Fluctuations, ISS, 2024



A Vailati et al. Microgravity Sci. and Technol. (2020)



**Henri
BATALLER**
Ass. Pr.



**Cédric
GIRAUDET**
postdoc



**Dan Esli
BOUYOU
BOUYOU**
PhD



**Mohammed
CRAGA**
PhD



**Stefano
CASTELLINI**
visiting PhD



**Mathilde
SAN
BAUDELIO**
stage BS

THANK YOU! QUESTIONS?

