Salt crystallization in rocks: probing its physics with X-ray (& neutron) imaging

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DMEX

Développement de Méthodologies Expérimentales

01 Context & motivation

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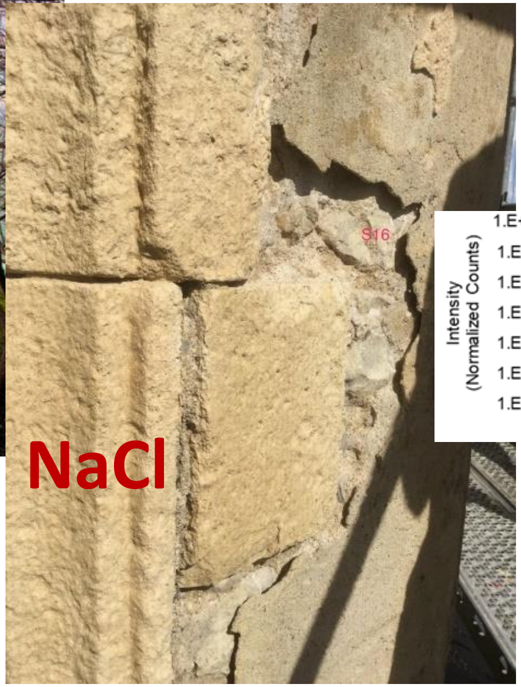
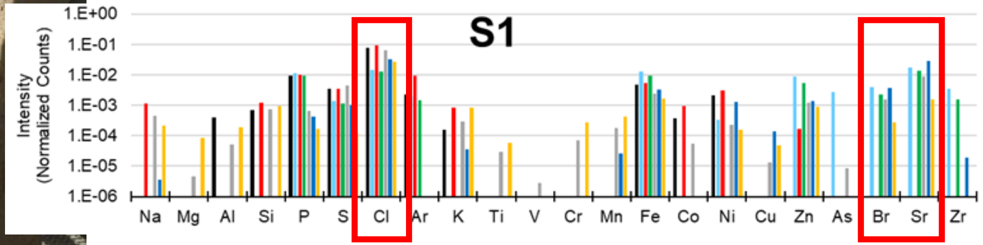
Salt crystallization in a building context

- Sea spray & marine aerosol:

Villa Belza, Biarritz (France)



in-situ XRF analysis:



- Cl: mainly NaCl-induced damage
- Br & Sr: sea is NaCl source

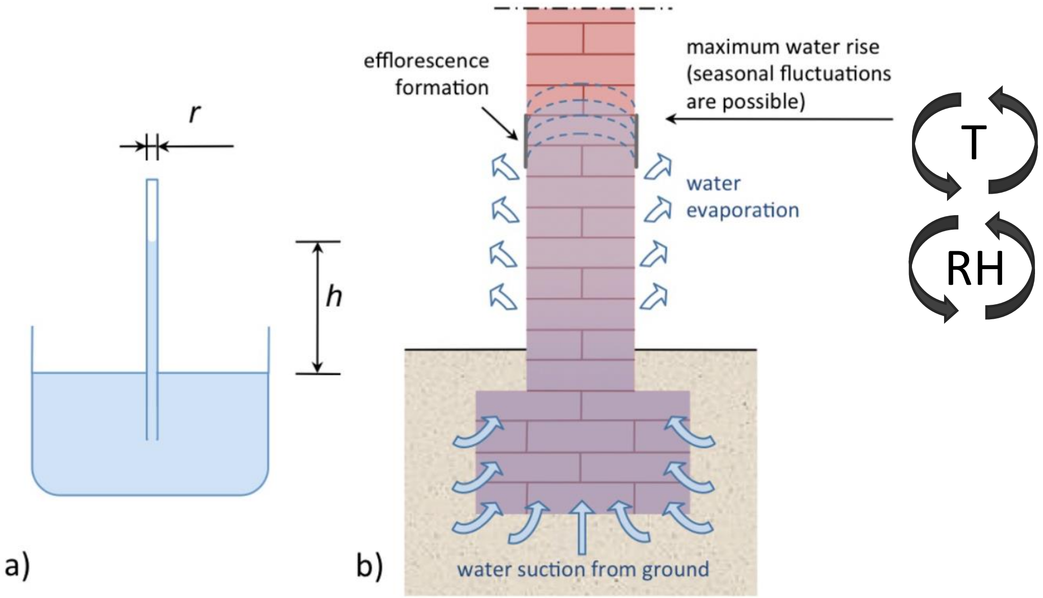
⇒ salt damage!

Mendonça Filho et al, 2019, *Microchemical Journal*
Morillas et al, 2020, *Science of the Total Environment*

Salt crystallization in a building context

- Rising damp:

City Hall, Huesca (Spain)

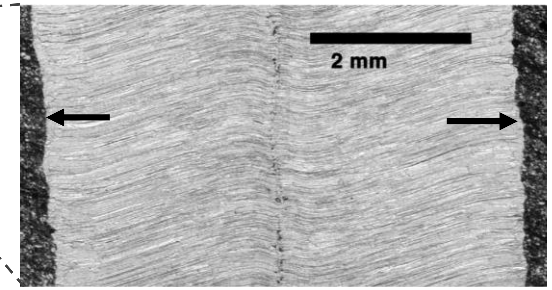


Franzoni, 2014, *Construction and Building Materials*

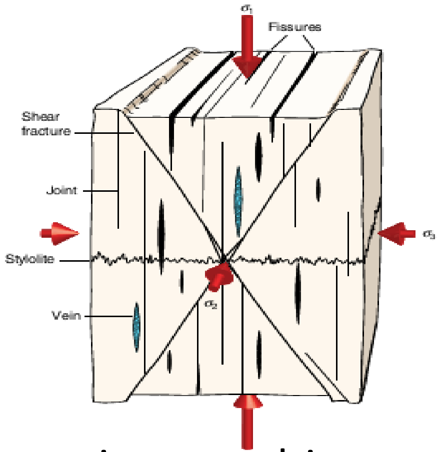
⇒ salt damage!

Salt crystallization in a geological context

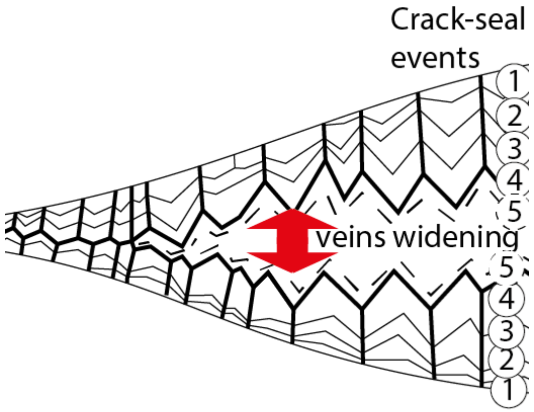
- Natural fractures driven by crystallization pressure under confinement?
  (Taber, 1916; Wiltschko & Morse, 2001)



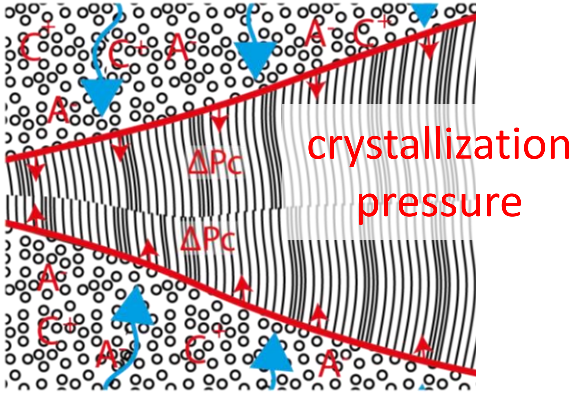
antitaxial fibrous veins



Tectonic stress driven crack propagation?



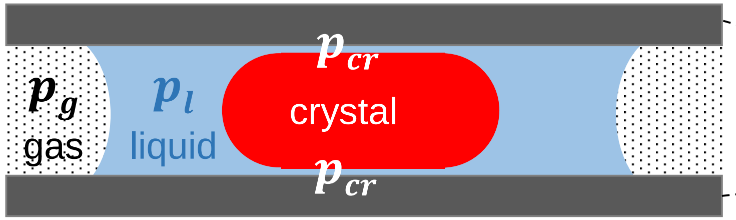
Hydraulic fracturing driven crack widening/propagation?



Crystallisation driven crack widening/propagation?

Salt crystallization in rocks

Crystallization induces pressure on pore walls



$$p_x = p_{cr} - p_l = \frac{RT}{\bar{V}_{cr}} \ln \frac{a}{a_0} - \sigma_{cr,l}\kappa_{cr,l} - p_c \frac{\Delta \bar{V}}{\bar{V}_{cr}}$$

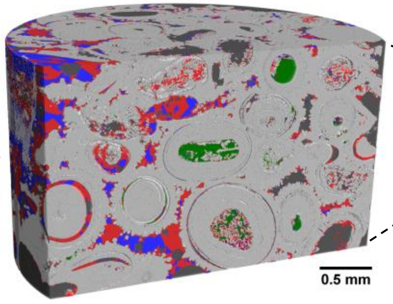
supersaturation $a(c,T)/a_0(T)$

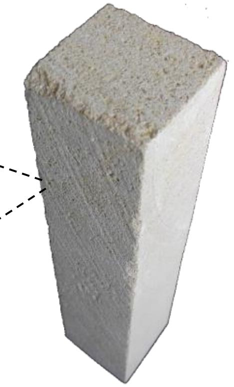
crystal-liquid surface tension & curvature

solubility change in unsaturated conditions

Scherer, 2004, *Cement and Concrete Research*
Steiger, 2005, *Journal of Crystal Growth*

How does this propagate in a porous rock?





Diffusive & advective ion transport:

$$\frac{\partial c}{\partial t} = \nabla \cdot (D \nabla c) - \nabla \cdot (c\vec{v}) + S$$

Crystallization kinetics:

$$\frac{\partial R_{cr}}{\partial t} = K \left(\frac{a}{a_0} - 1 \right)^g$$

Stoichiometry:

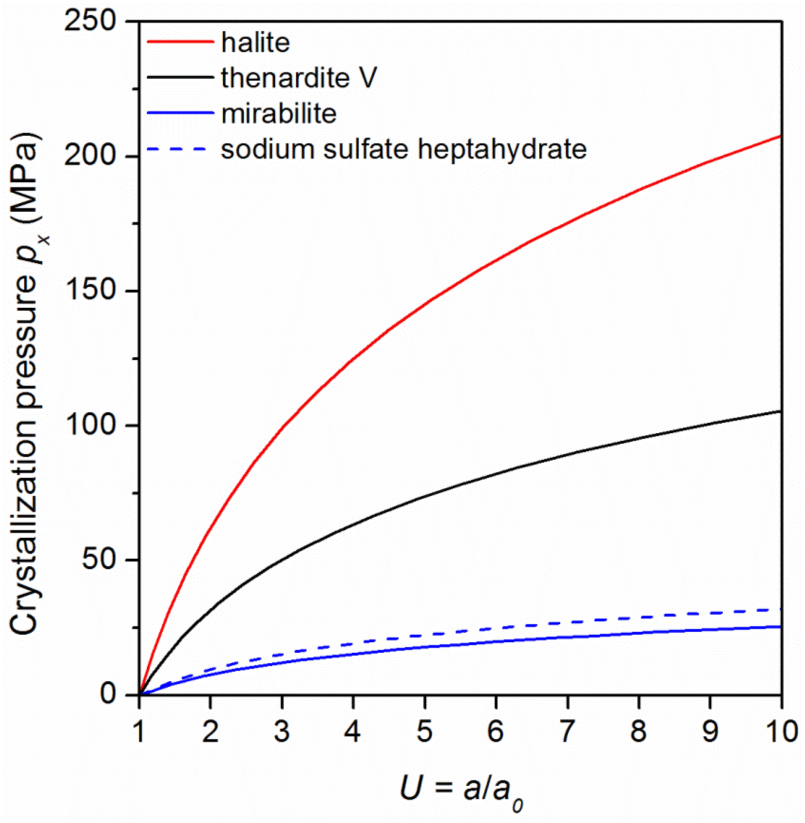
$$\nu_M M + \nu_X X + \nu_0 H_2O \leftrightarrow M_{\nu_M} X_{\nu_X} \cdot \nu_0 H_2O$$

Poromechanical response:

$$\boldsymbol{\sigma}_s = \mathrm{D}(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}_T) - b \int_{p_{c,ref}}^{p_c} (S_l + S_{cr}) dp_c \mathbf{I} - b S_{cr} (p_x - p_{x,ref}) \mathbf{I}$$

Salt crystallization in rocks

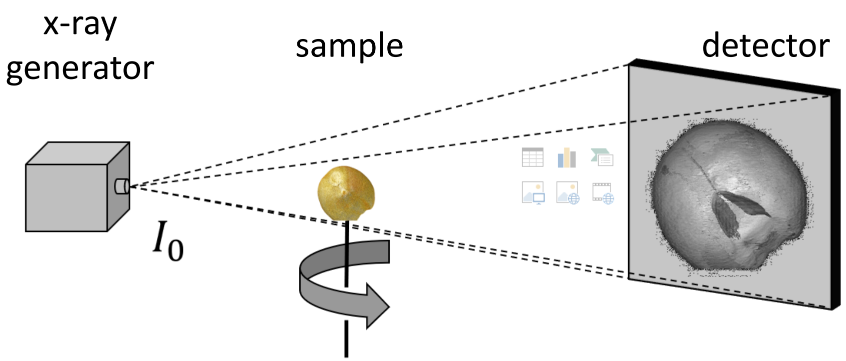
Crystallization induces pressure on pore walls



Derluyn, 2012, *PhD thesis*

How does this propagate in a porous rock?

- Transport – precipitation - mechanics



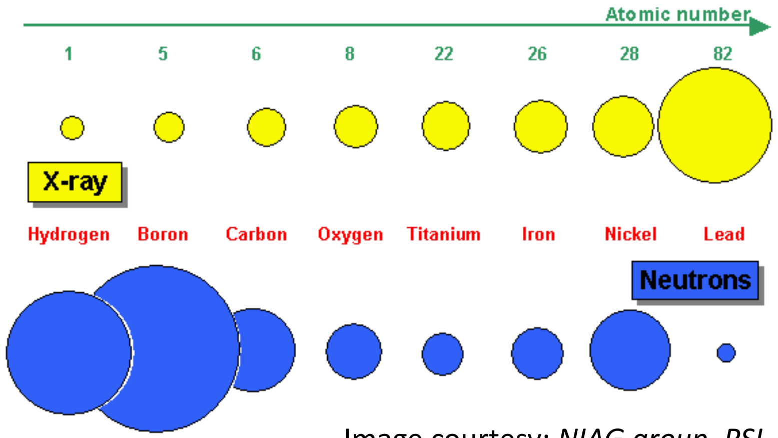
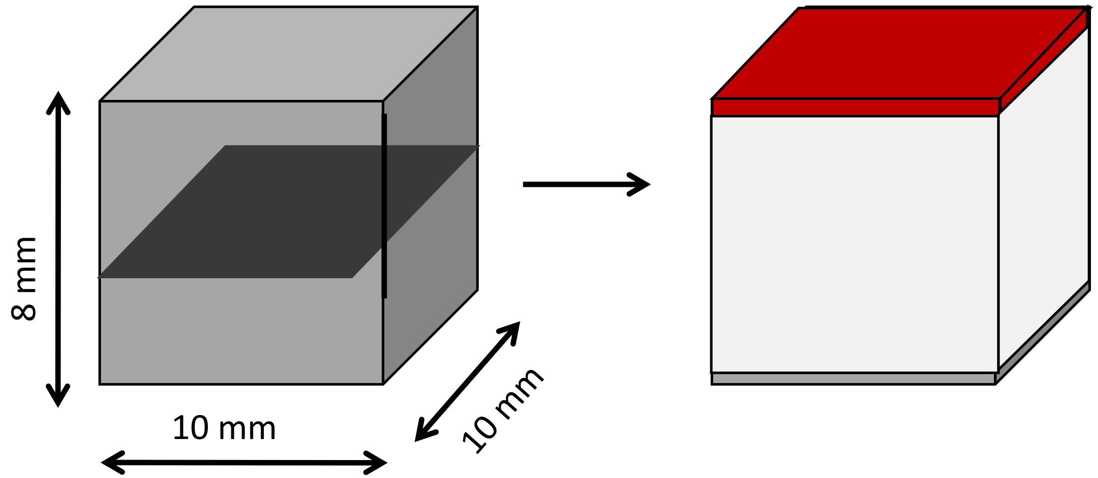


Image courtesy: *NIAG group, PSI*

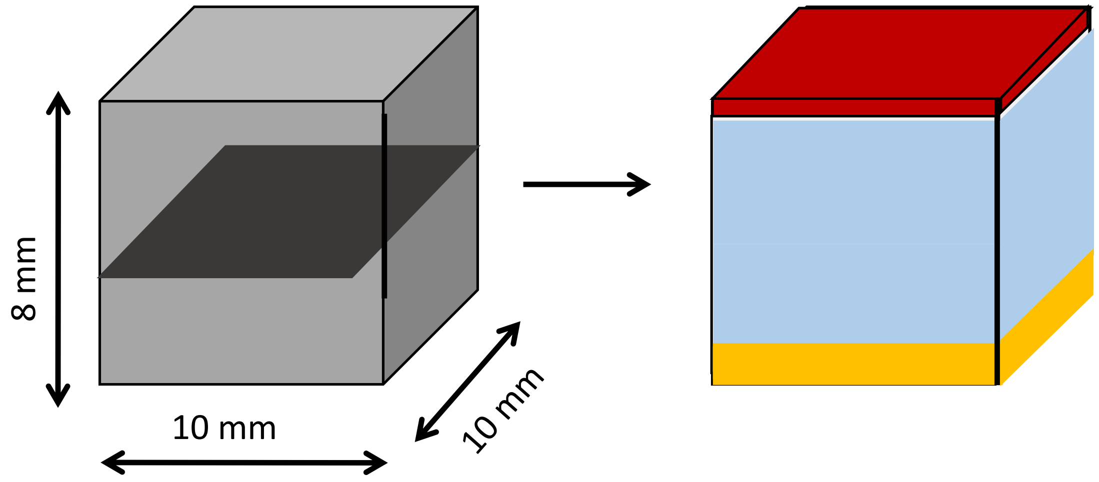
02 Drying-induced NaCl-damage

Experimental protocol



lab X-ray tomography
initial dry state

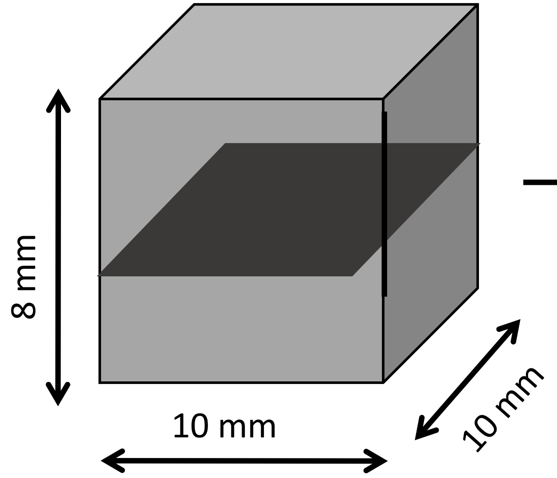
Experimental protocol



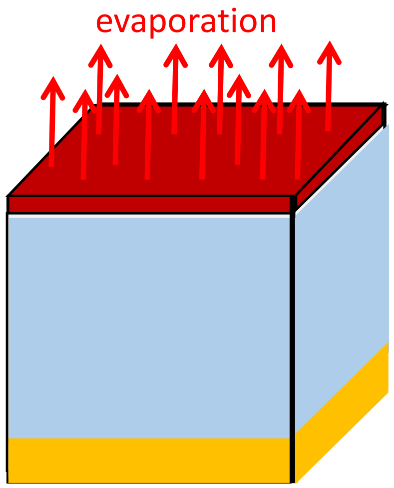
lab X-ray tomography
initial dry state

capillary uptake @ room T of
5.8 molal NaCl-solution

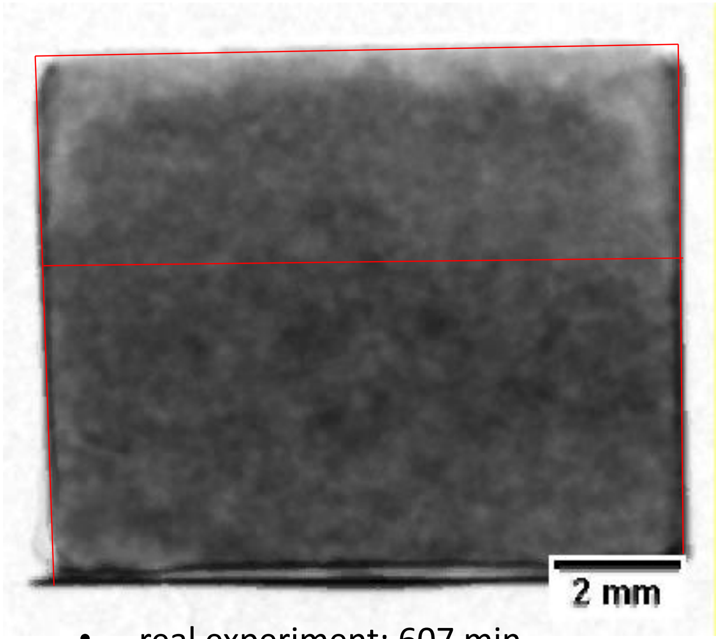
Experimental protocol



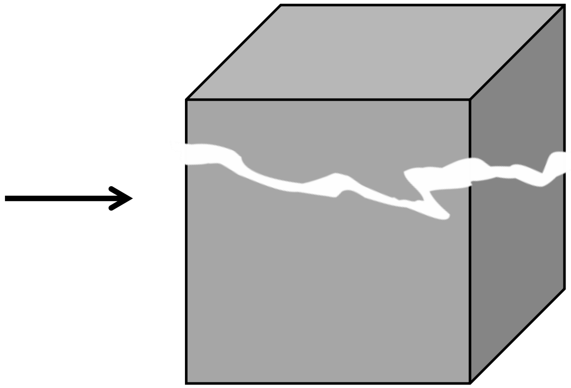
lab X-ray tomography initial dry state



neutron radiography of 1D saline drying @ 45°C – 5% RH



- real experiment: 607 min.
- movie: 25700 x faster; looped



lab X-ray tomography final dry state

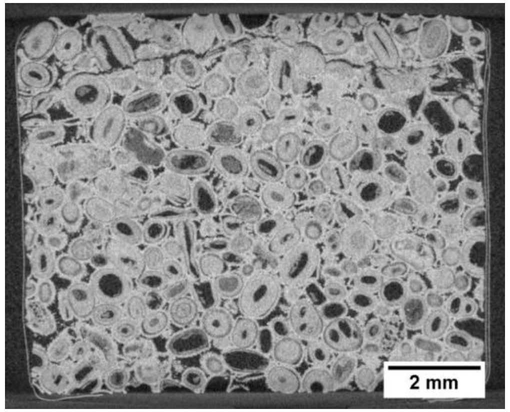
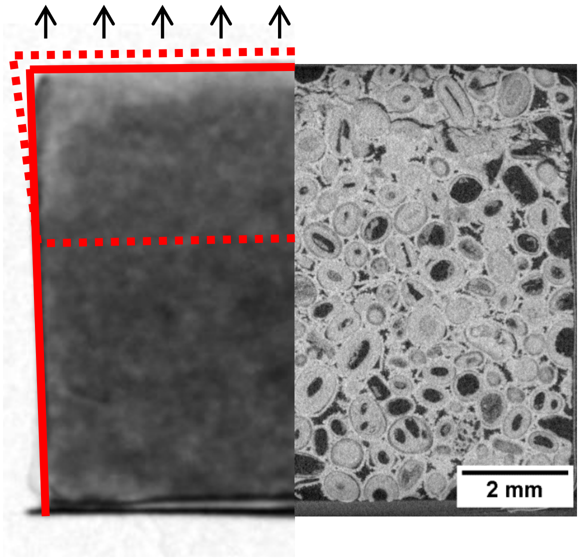
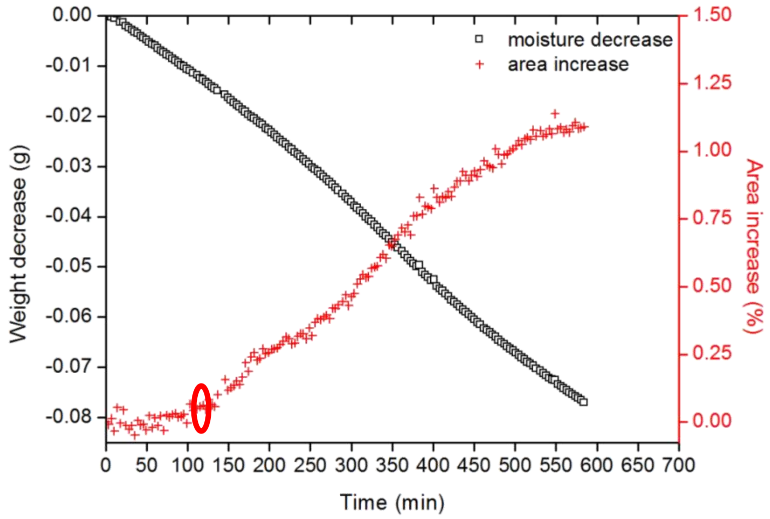


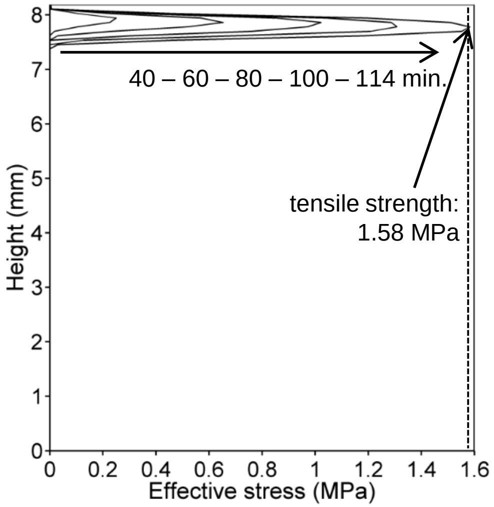
Image quantification & poromechanical prediction



neutron radiography
X-ray tomography



quantitative image analysis

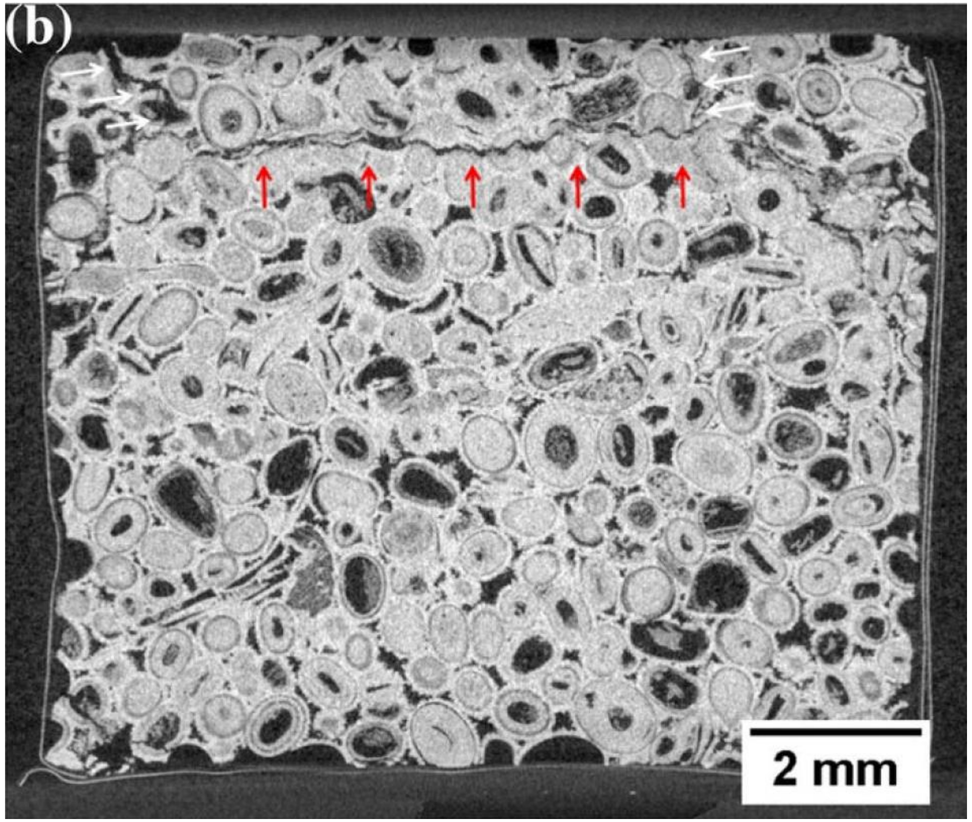


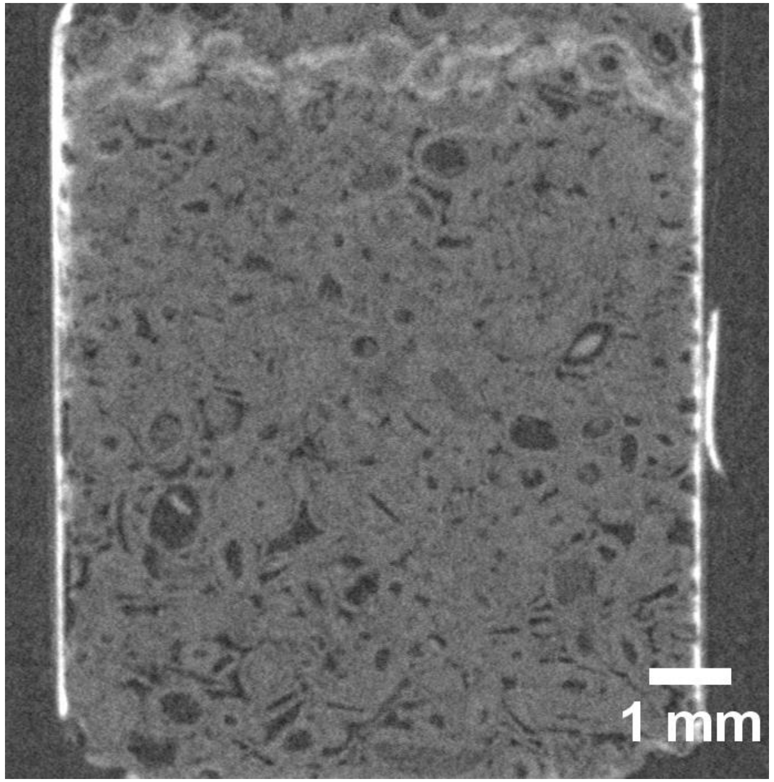
poromechanical FEM model

Derluyn et al, 2013, *J Build Physics*
Derluyn et al, 2014, *J Mech Phys Solids*
Derluyn et al, 2019, *Transport in Porous Media*

Looking deeper

- X-ray & neutron tomography (µCT):

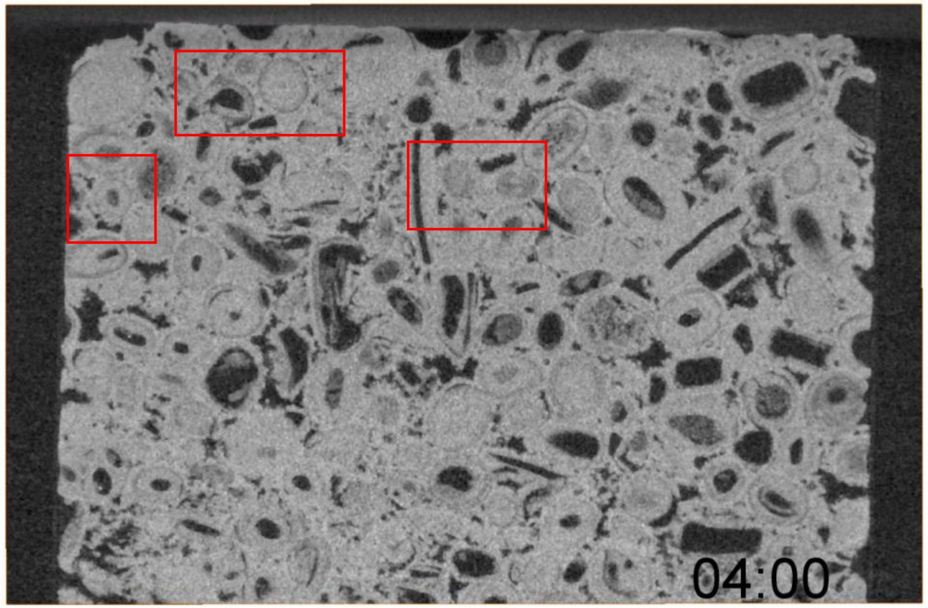


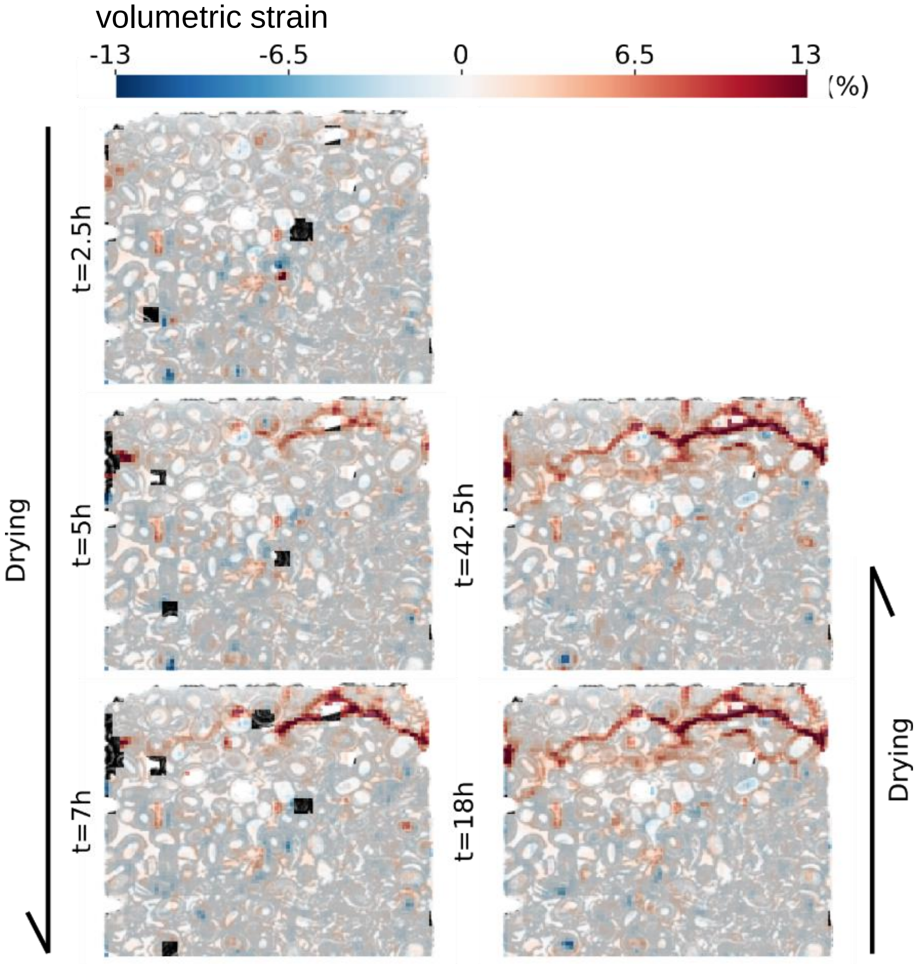


⇒ main horizontal crack following crystal precipitation
+ secondary cracks

Looking deeper

- Time-resolved X-ray µCT:





Derluyn et al, *in preparation*

03 Rewetting-induced Na_2SO_4-damage

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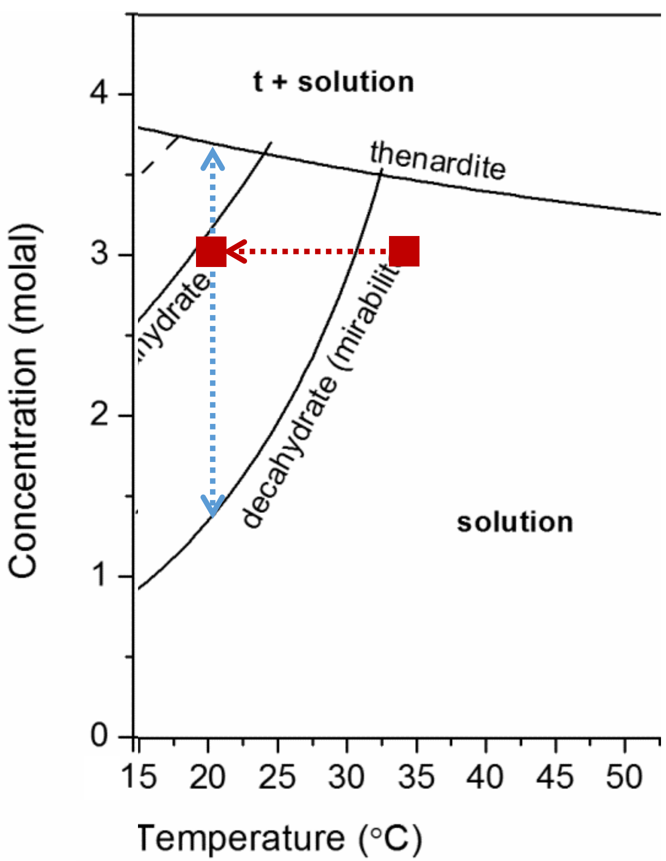
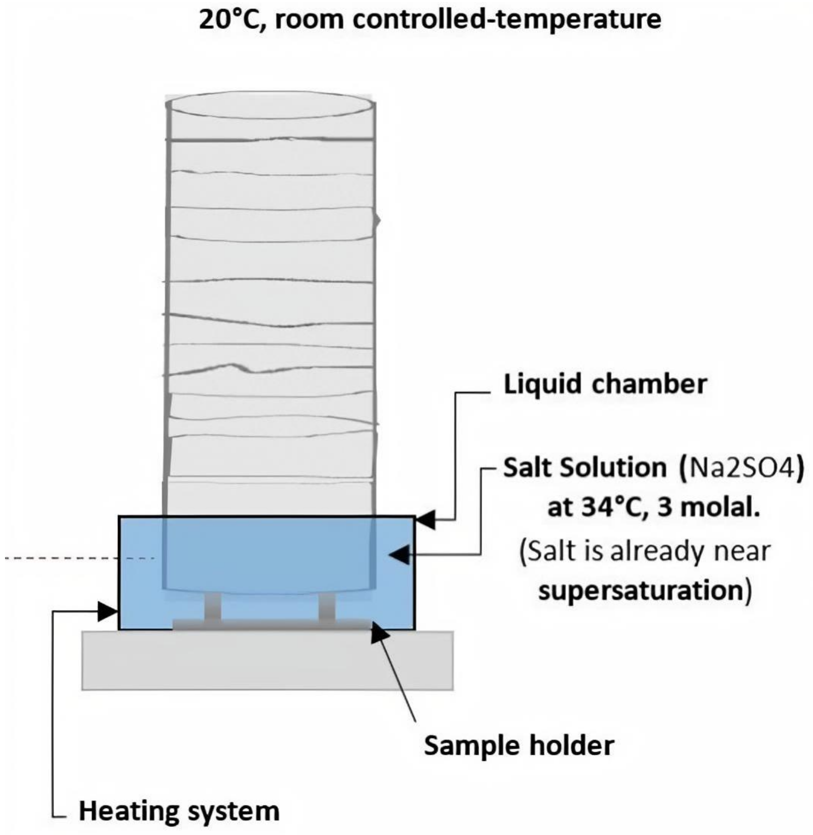
Experimental protocol

- Vosges sandstone during capillary rise – 3D evaporation inducing crystallization pressure due to thenardite – mirabilite transition:



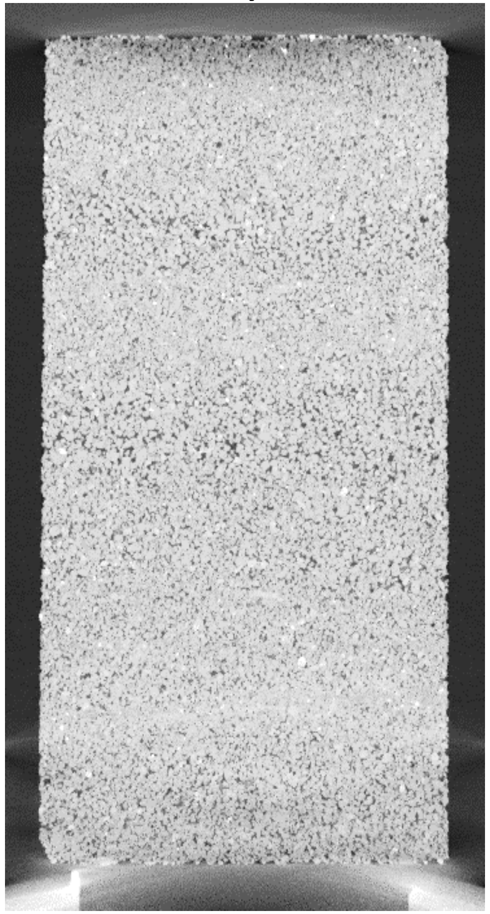
Flatt, 2002, *Journal of Crystal Growth*
Shahidzadeh et al, 2010, *Physical Review E*



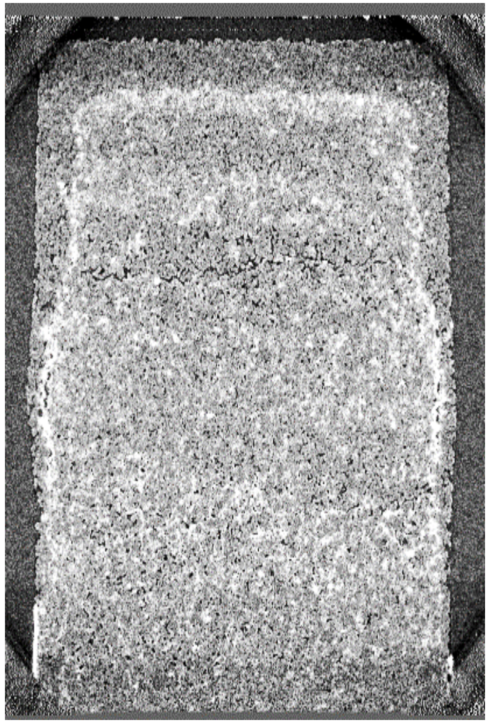
Okumko et al, *in preparation*

X-ray – neutron µCT comparison

X-ray 0h



neutron 17h 15 min

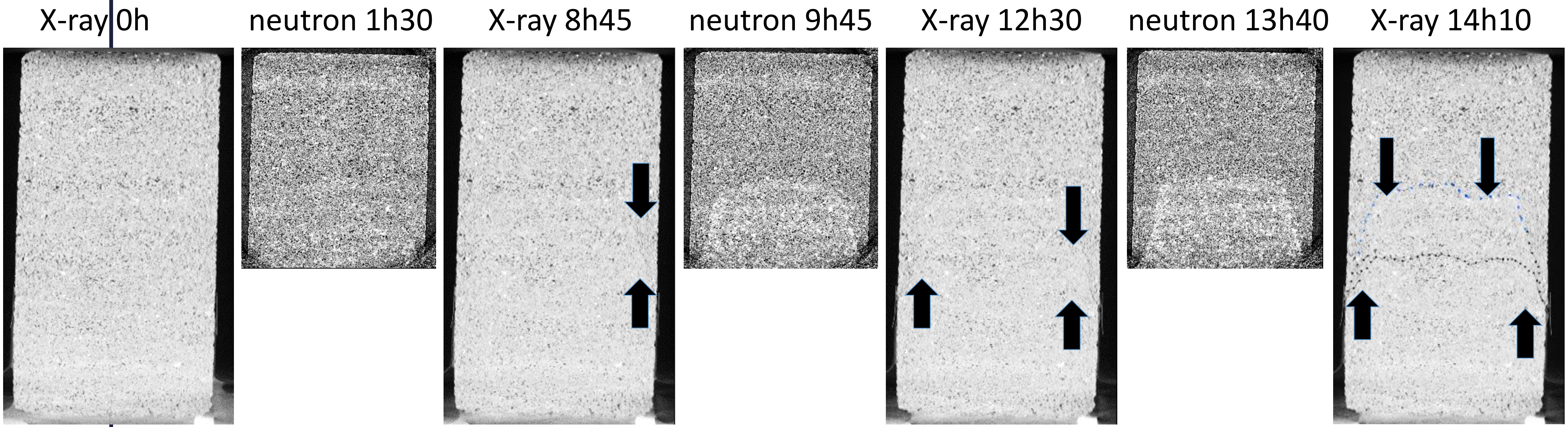


X-ray 19h 15 min

⇒ “vertical” evaporation front = vertical “scaling” cracks

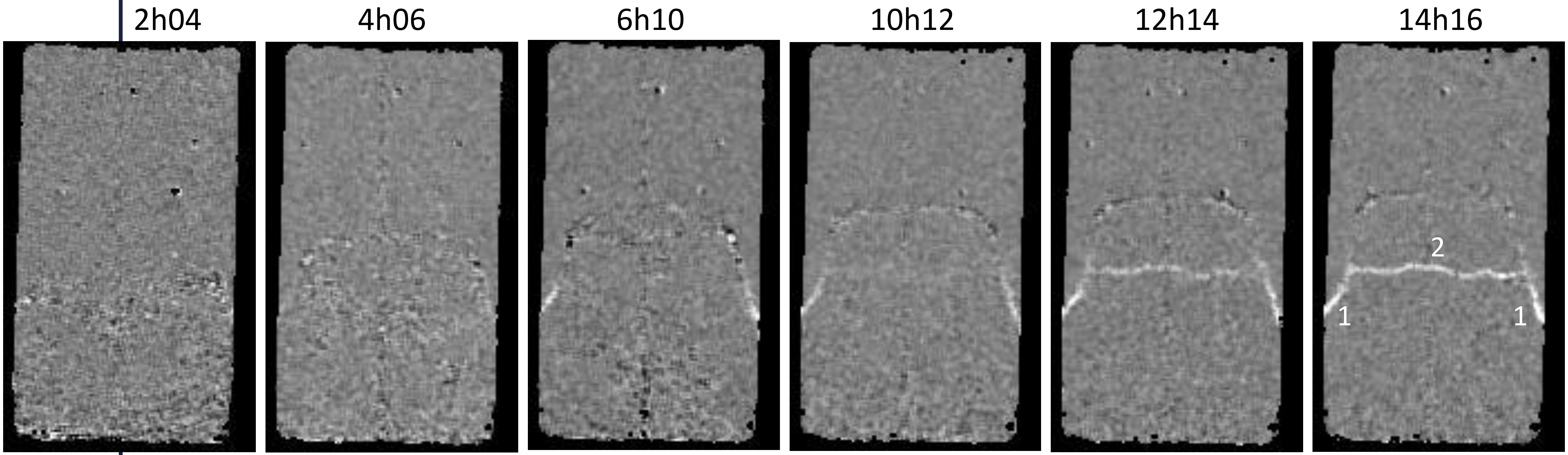
⇒ “horizontal” evaporation front ≠ horizontal cracks

X-ray – neutron µCT comparison



X-ray µCT analysis

- Digital volume correlation: volumetric strain



⇒ largest deformations and cracking correspond to
(1) zones of salt precipitation close to surface
(2) mechanically weaker zones

04

Rewetting-induced Na_2SO_4-damage under reservoir conditions

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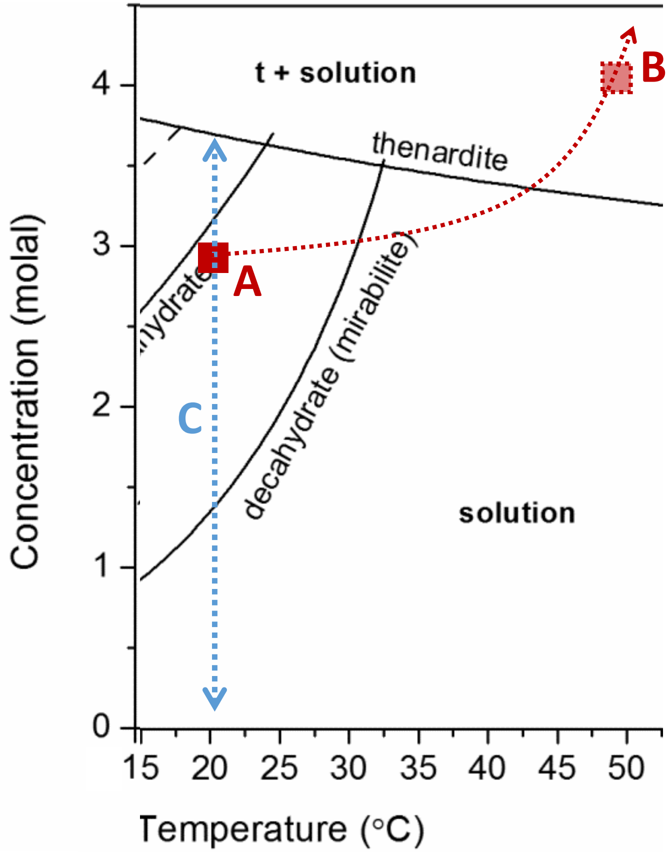
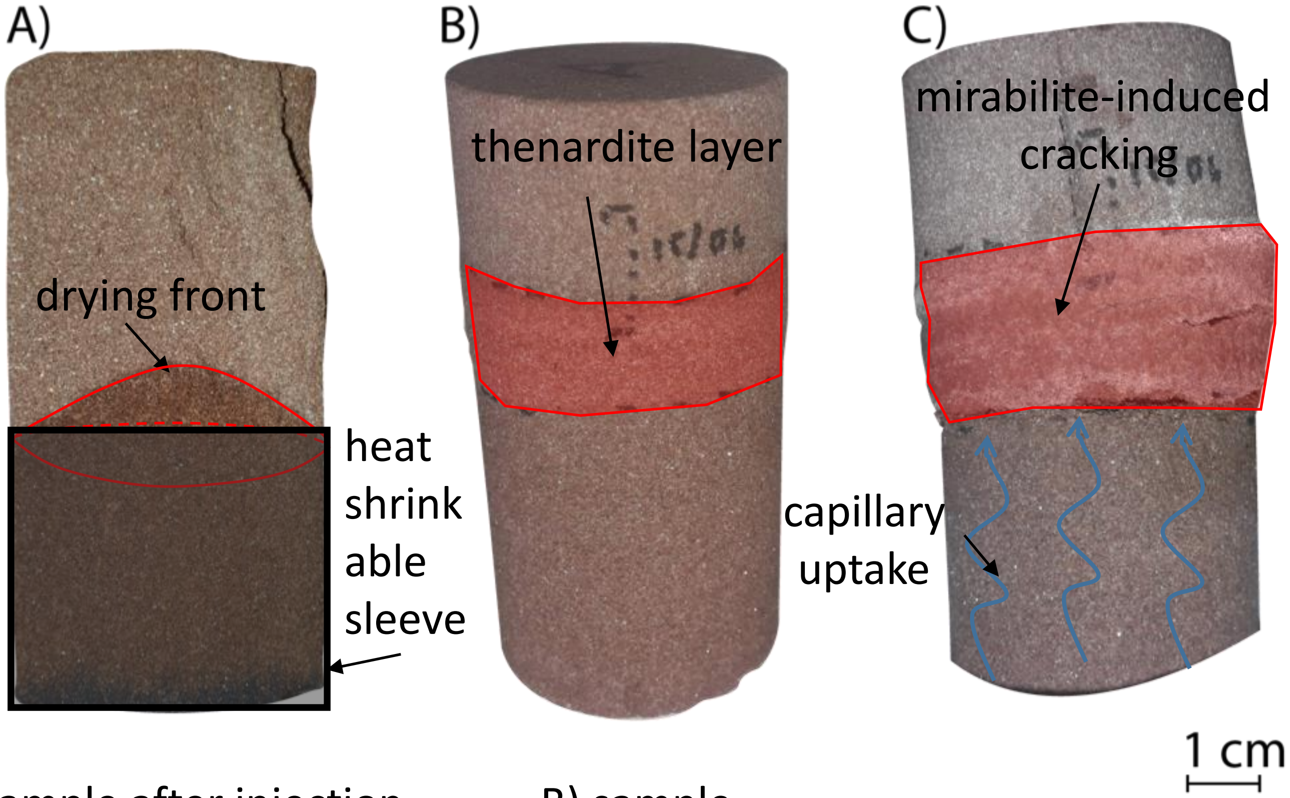
Experimental protocol

- Adamswiller sandstone with a localized Na_2SO_4 zone inducing crystallization pressure due to thenardite – mirabilite transition:



Flatt, 2002, *Journal of Crystal Growth*
Shahidzadeh et al, 2010, *Physical Review E*



A) sample after injection of 4 ml of solution (2.8 m Na_2SO_4)

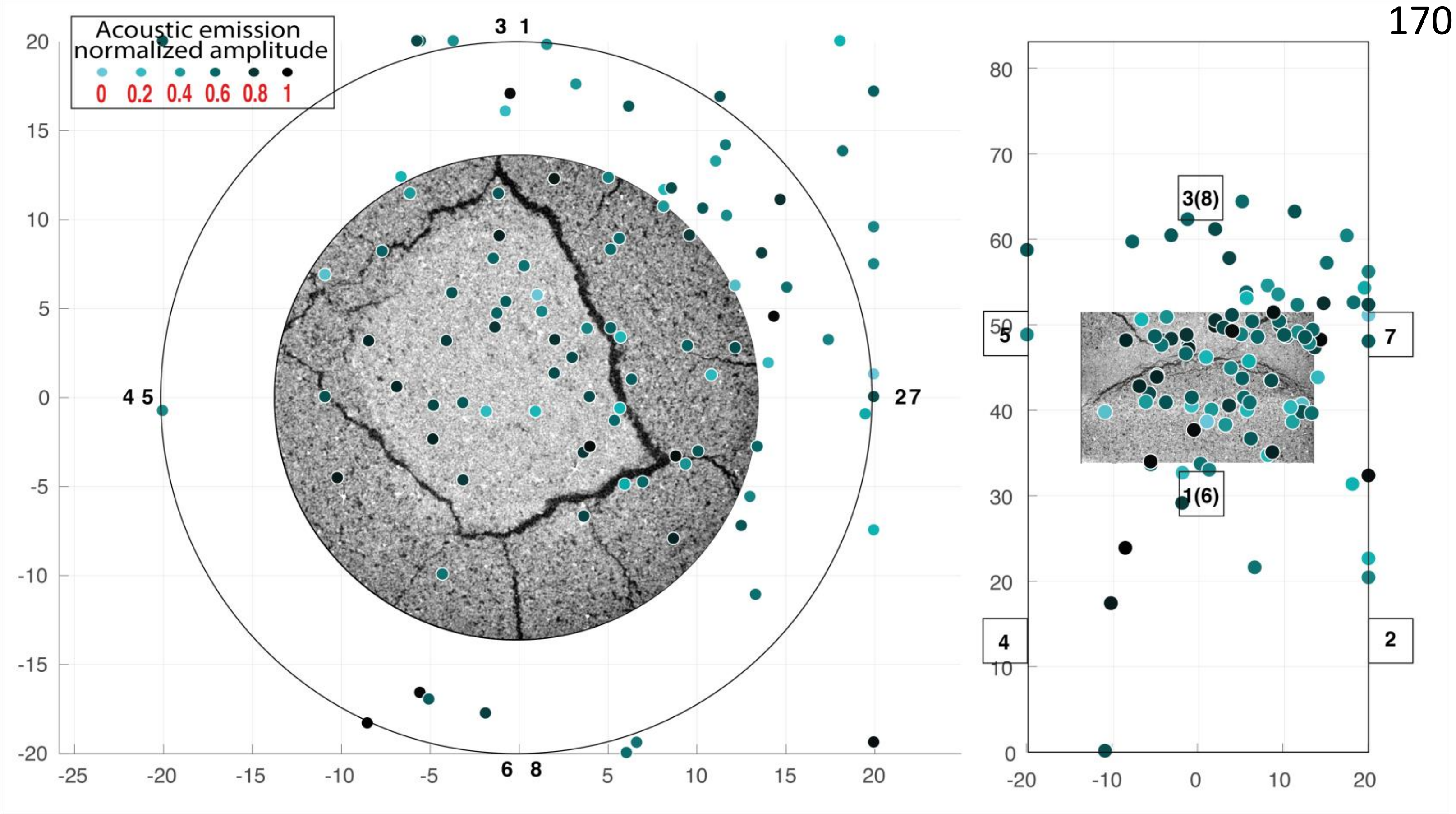
B) sample impregnated with thenardite

C) after imbibition with pure water

No confining pressure

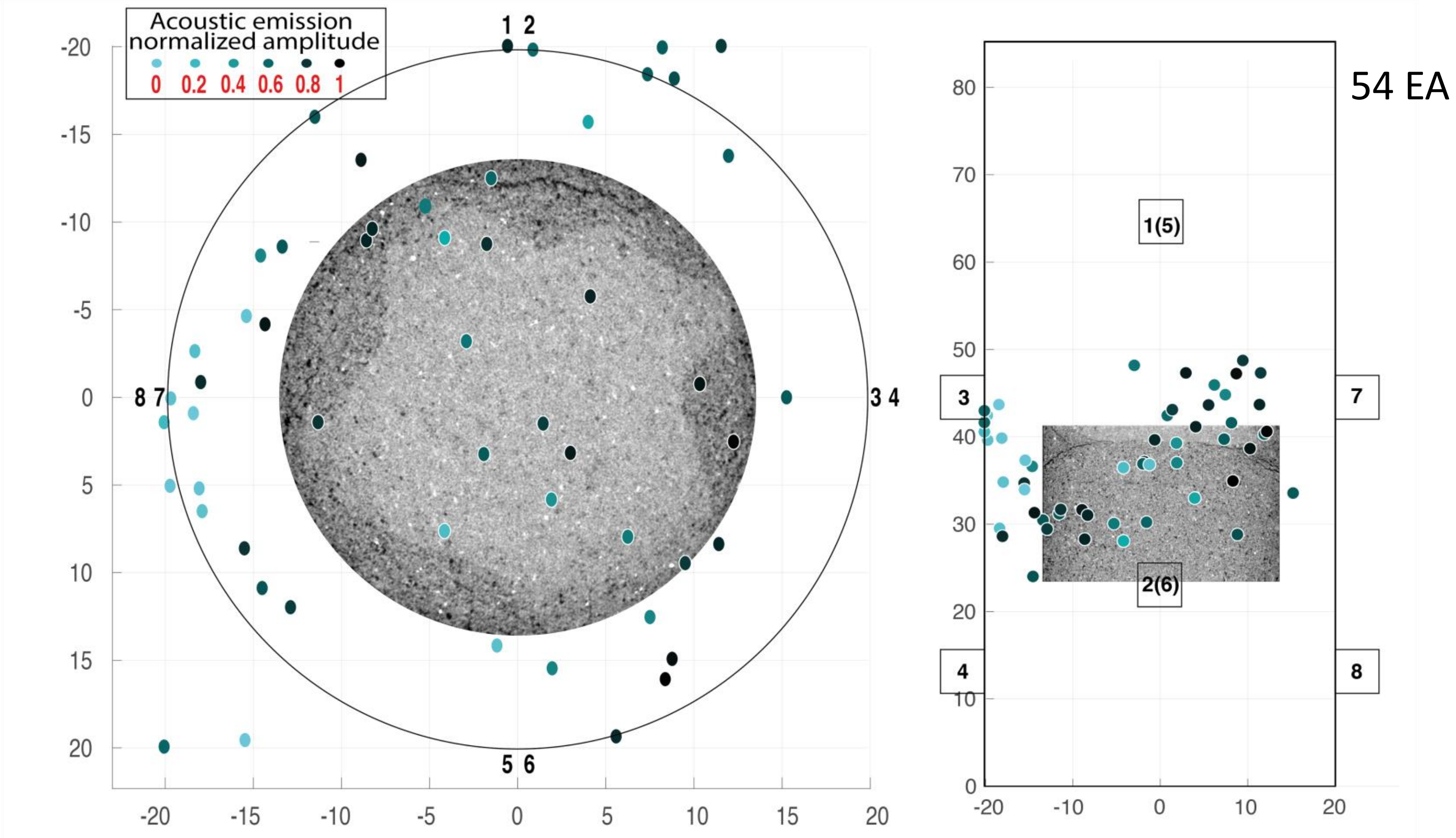
- Acoustic emission:

170 EA



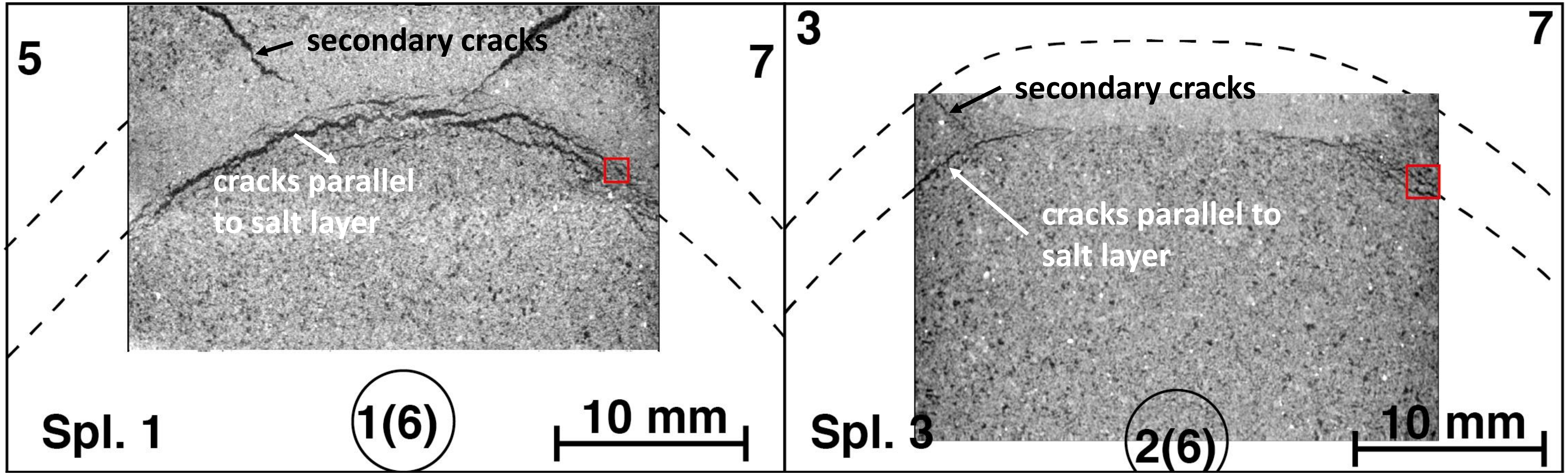
Confined at 10 MPa (-400 meter)

- Acoustic emission:



X-ray µCT comparison

no confining pressure   confined at 10 MPa



⇒ crystallization pressure also works under confined reservoir conditions! (B. Leclère, 2021, *PhD Thesis*)

05 Conclusions

Conclusions

- Through non-destructive and time-resolved imaging techniques we can reconstruct the story of salt crystallization and its hydro-mechanical processes inside rocks

- Seal-crack versus crack-seal: reality is more complex
  - damage not only where crystals precipitate
  - crystallization needed for crack initiation
  - crack propagation also in mechanically weaker layers of heterogeneous rock or as secondary cracks due to differential stresses
  - seal-crack also occurs under confinement

Transnational access: Excite Network

Second call closes 31st August 2022

https://excite-network.eu/excite-transnational-access-call-open/

National access: **IMAGINE^2**

First call expected end of 2022

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Thanks to LFCR *salt* collaborators:


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Prof. D. Grégoire


Prof. J.P. Callot


F. F. Mendonça Filho


Dr. B. Leclère


Dr. F. Thierry

Thank you for your attention!

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