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

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01 Context & motivation
Salt crystallization in a building context

• Sea spray & marine aerosol:

Villa Belza, Biarritz (France)

⇒ salt damage!

in-situ XRF analysis:

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

Mendonça Filho et al, 2019, Microchemical Journal
Morillas et al, 2020, Science of the Total Environment
Salt crystallization in a building context

- Rising damp:

  \( \text{Na}_2\text{SO}_4 \)

City Hall, Huesca (Spain)

⇒ salt damage!

Franzoni, 2014, *Construction and Building Materials*
Salt crystallization in a geological context

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

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

\[ p_x = p_{cr} - p_l = \frac{RT}{V_{cr}} \ln \frac{a}{a_0} - \sigma_{cr,l} \kappa_{cr,l} - p_c \frac{\Delta V}{V_{cr}} \]

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:
\[ \sigma_s = D(\epsilon - \epsilon_T) - b \int (S_l + S_{cr})dp_c I \]
\[ -bS_{cr} (p_x - p_{x,ref}) I \]
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

evaporation

lab X-ray tomography
final dry state

- real experiment: 607 min.
- movie: 25700 x faster; looped
Image quantification & poromechanical prediction

Derluyn et al, 2013, *J Build Physics*
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
Rewetting-induced Na$_2$SO$_4$-damage
Experimental protocol

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

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Okumko et al, in preparation
**X-ray – neutron μCT comparison**

⇒ “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
Rewetting-induced $\text{Na}_2\text{SO}_4$-damage under reservoir conditions
Experimental protocol

- Adamswiller sandstone with a localized $\text{Na}_2\text{SO}_4$ zone inducing crystallization pressure due to thenardite – mirabilite transition:

A) sample after injection of 4 ml of solution ($2.8 \text{ m Na}_2\text{SO}_4$)

B) sample impregnated with thenardite

C) after imbibition with pure water
No confining pressure

• Acoustic emission:
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)
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 31\textsuperscript{st} August 2022
https://excite-network.eu/excite-transnational-access-call-open/

National access: **IMAGINE\textsuperscript{2}**
First call expected end of 2022
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Thank you for your attention!