Rôle des hétérogénéités structurales et minérales dans la formation des réseaux karstiques : approche mécanistique de l'échelle du pore à l'échelle de la carotte

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SNO Karst

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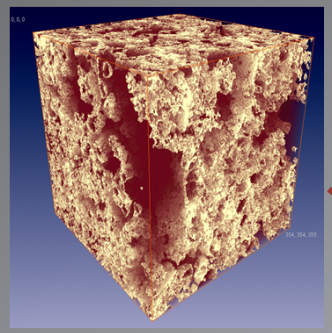
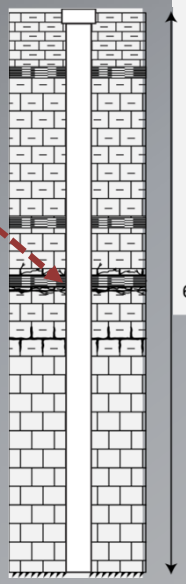
Main Project

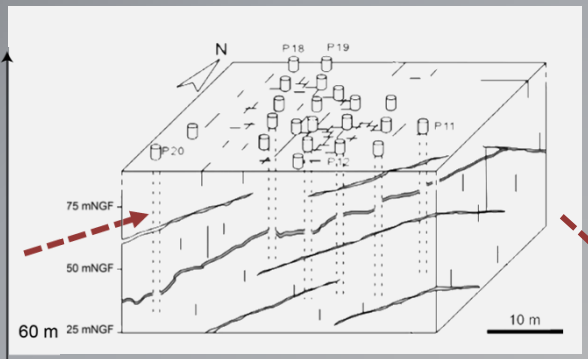
Les hydroroutes souterraines :
De la formation des karsts à l'étude du transport de contaminants



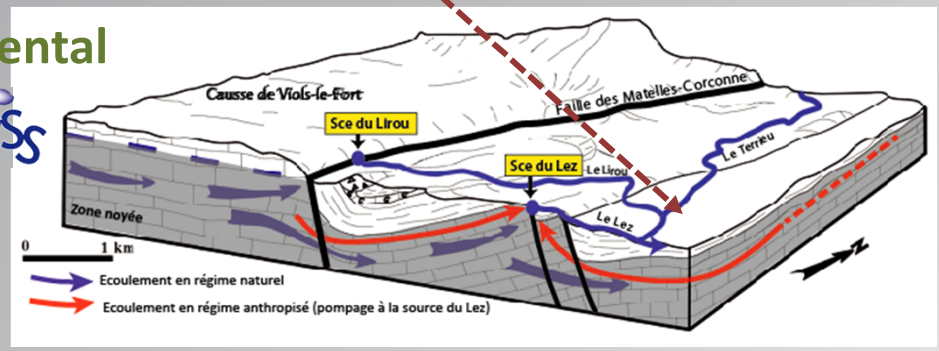
Echelle de la carotte



Echelle du forage



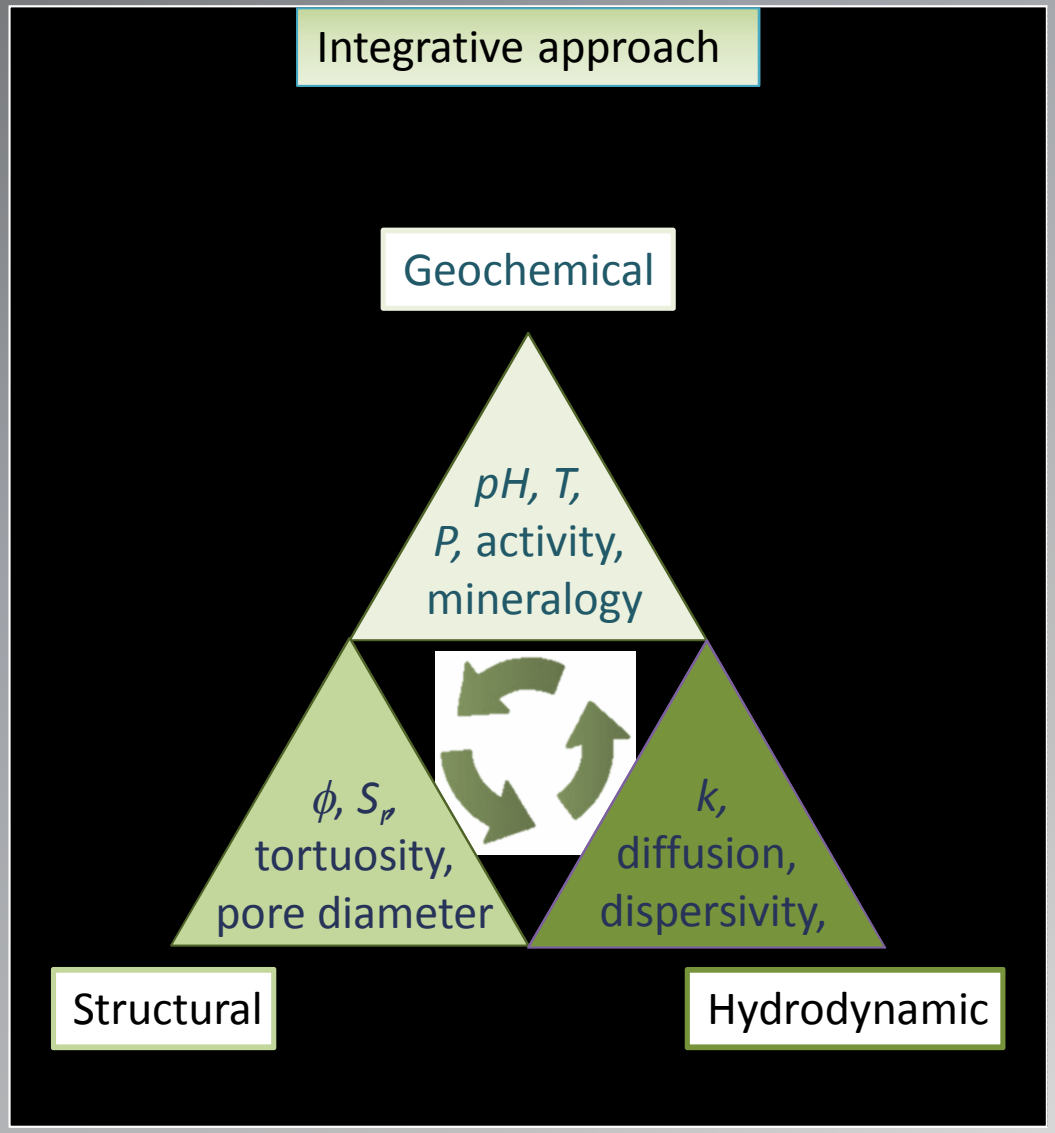
Echelle du site expérimental



Echelle régionale

RBV RÉSEAU DES BASSINS VERSANTS

Aim: Coupled approach



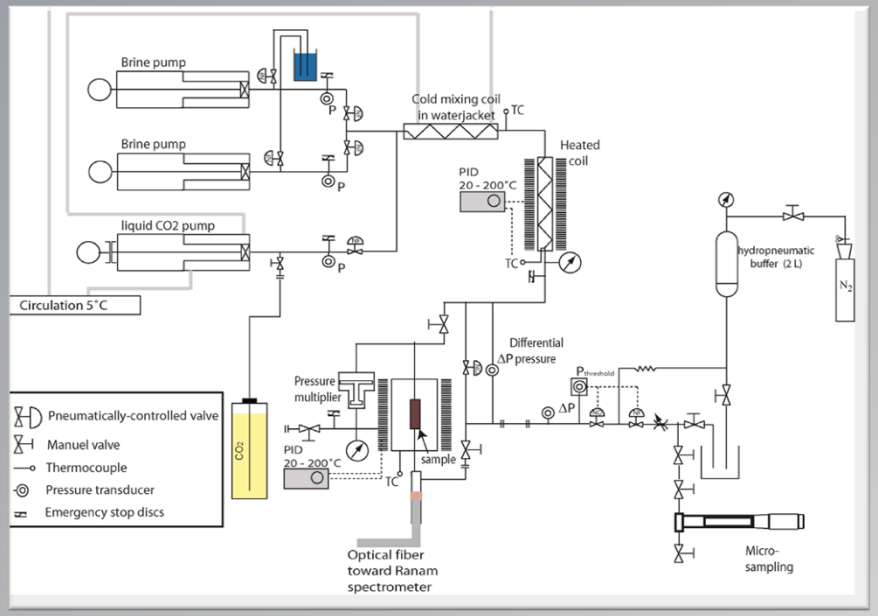
Take home message

To characterize and predict chemical processes (dissolution, precipitation, redox reactions, …), it is necessary to take into account the coupled reactive-transport processes and the local structural, hydrodynamic and mineralogical heterogeneities.

Experimental methodology

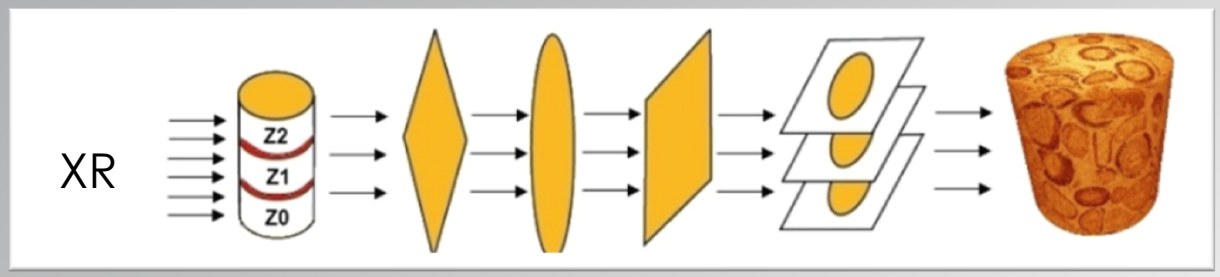
Percolation experiments on natural core samples:

- Pore scale heterogeneities
- Mineral heterogeneities
- Flow and transport effects
- Fluid chemistry controled



X-ray microtomography images:

- 3D visualization of the localization of the processes
- Structural heterogeities
- Mineral, grain and pore distributions
- Transport properties



Contents

- Effect of structure heterogeneities

- Effect of mineral heterogeneities

$\longrightarrow$ on dissolution processes (localization and rate)

Effect of structure heterogeneities on dissolution processes

CO_2-rich brine percolation through limestone samples

Same T, P, P_{CO2}, Pe and Da numbers

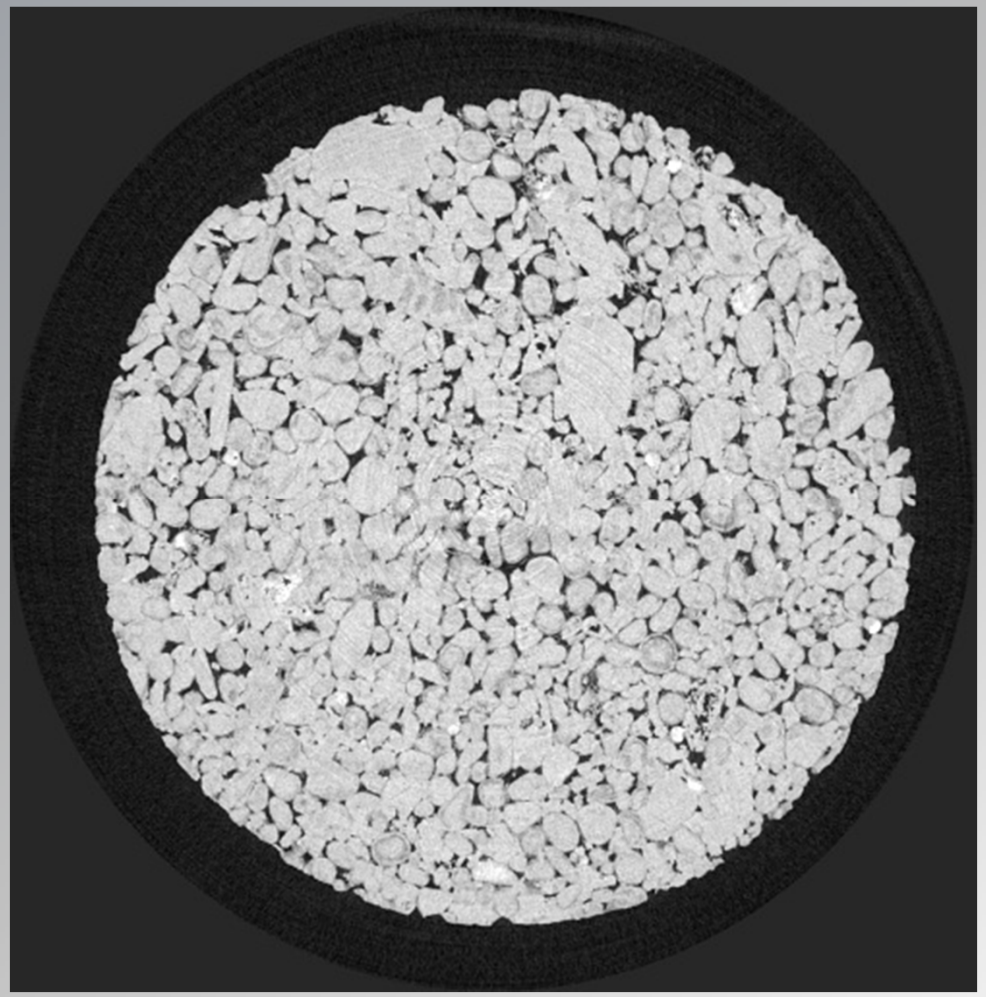
Two different rock structures

Effect of structure heterogeneities on dissolution processes

CO_2-rich brine percolation through limestone samples

Same T, P, P_{CO2}, Pe and Da numbers

Two different rock structures



High permeability (connectivity through the macro-porosity)

Luquot and Gouze, 2009. Chem. Geol.
Gouze and Luquot, 2011. J. Cont. Hydro.

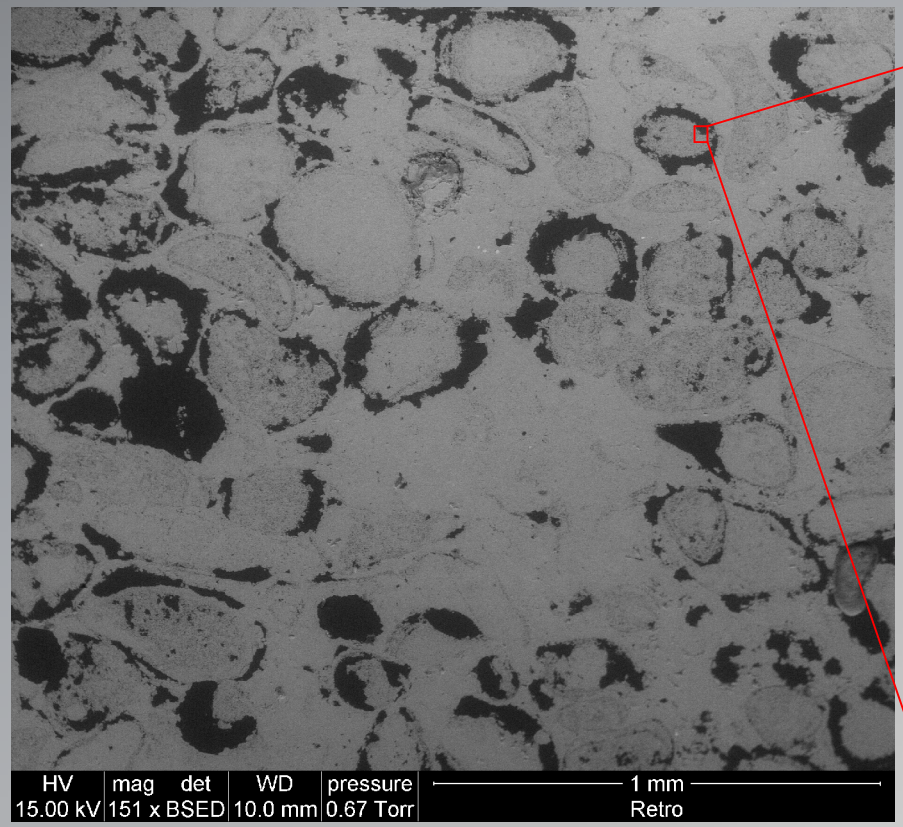
Effect of structure heterogeneities on dissolution processes

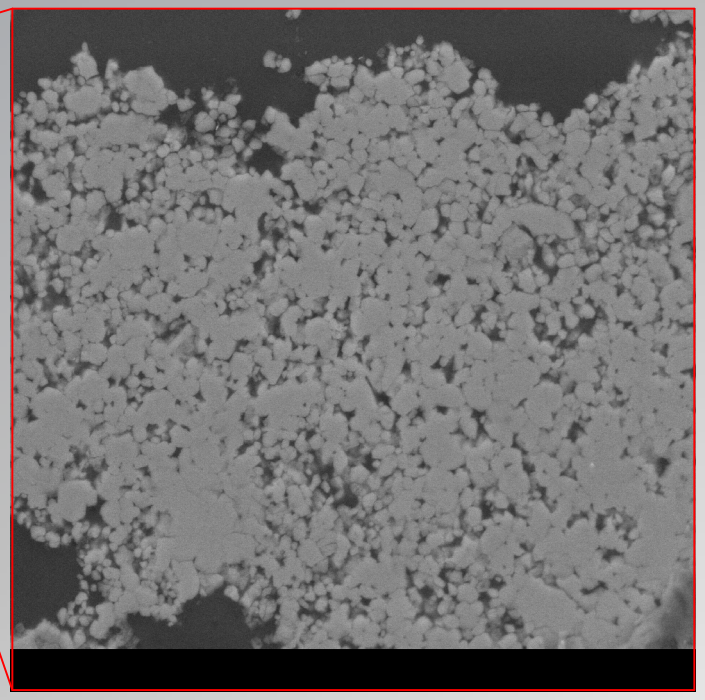
CO_2-rich brine percolation through limestone samples

Same T, P, P_{CO2}, Pe and Da numbers

Two different rock structures

Low permeability (connectivity through the micro-porosity)



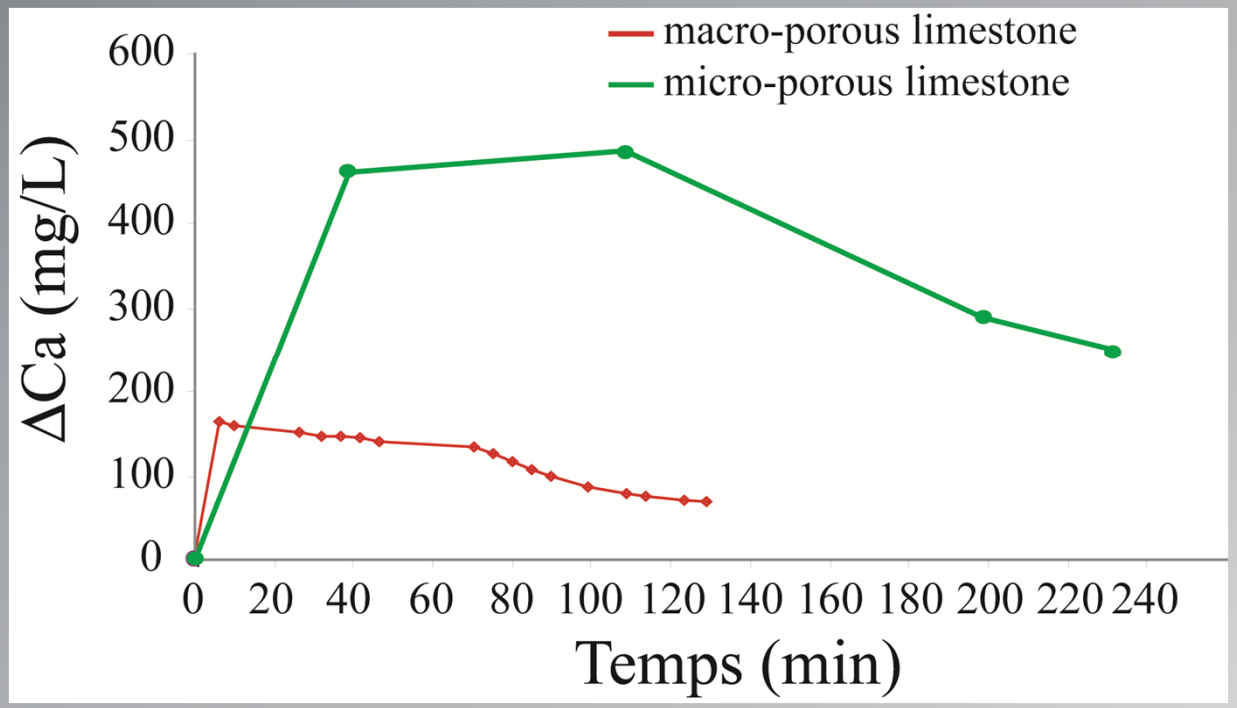


Luquot et al., 2014 Transp. Porous Media

Effect of structure heterogeneities on dissolution processes

In both cases, calcite dissolution is observed: $\Delta C_{Ca}(t) > 0 \rightarrow$ Dissolution

$$CaCO_3 + H_20 + CO_2 \longleftrightarrow Ca^{2+} + 2HCO_3^-$$

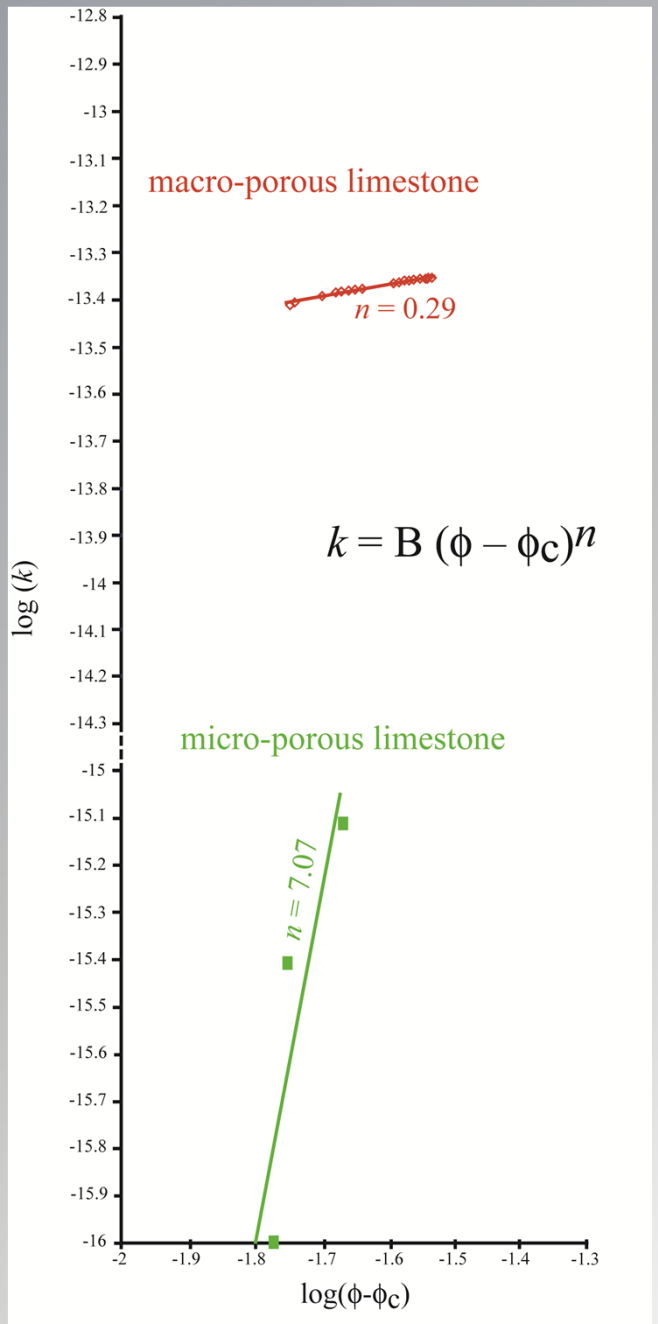


Luquot and Gouze, 2009. Chem. Geol.
Gouze and Luquot, 2011. J. Cont. Hydro.
Luquot et al., 2014 Transp. Porous Media

Effect of structure heterogeneities on dissolution processes

- Higher permeability increase for the micro-porous limestone for a less porosity increase.

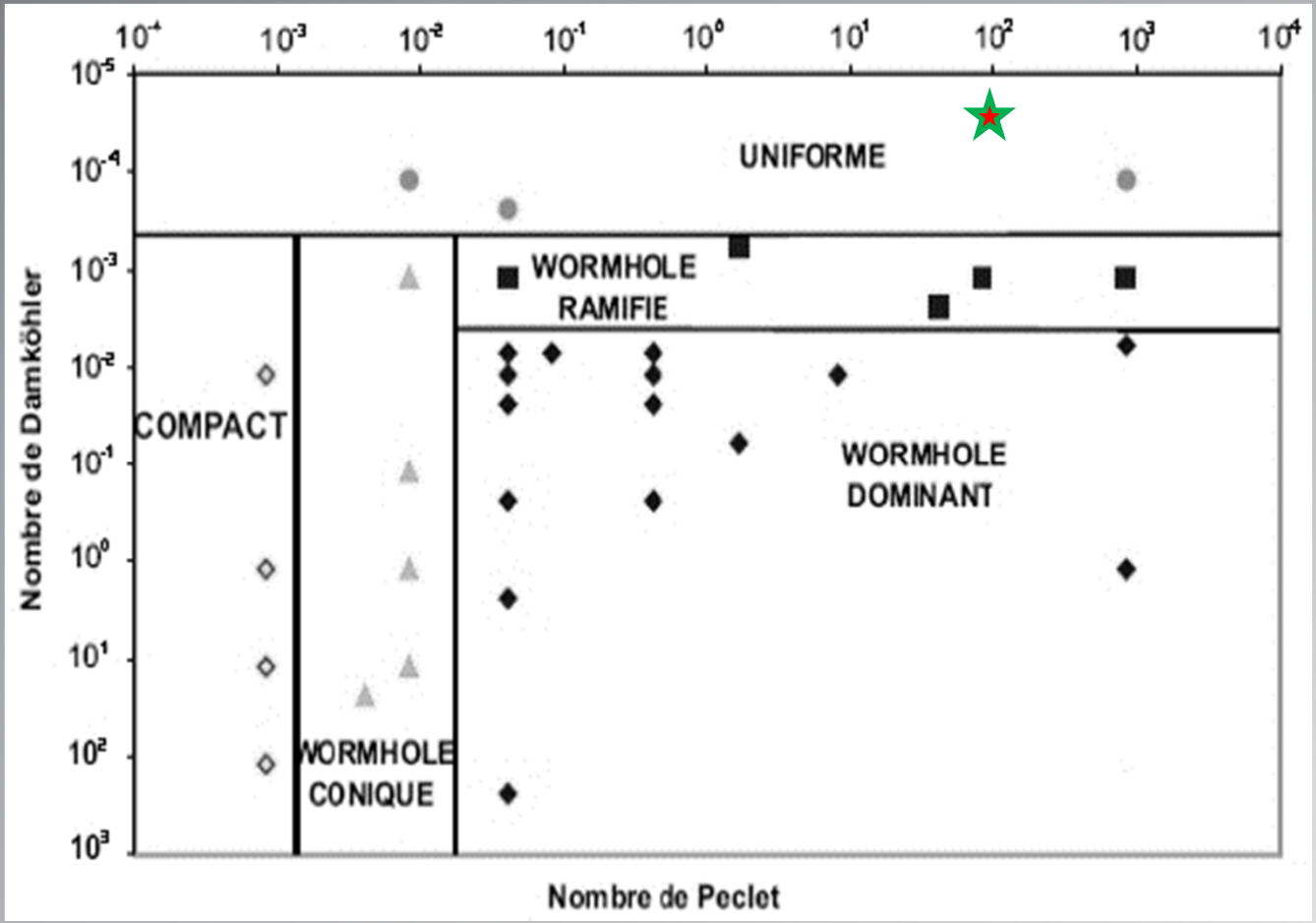
Different dissolution features ?



Luquot and Gouze, 2009. Chem. Geol.
Gouze and Luquot, 2011. J. Cont. Hydro.
Luquot et al., 2014 Transp. Porous Media

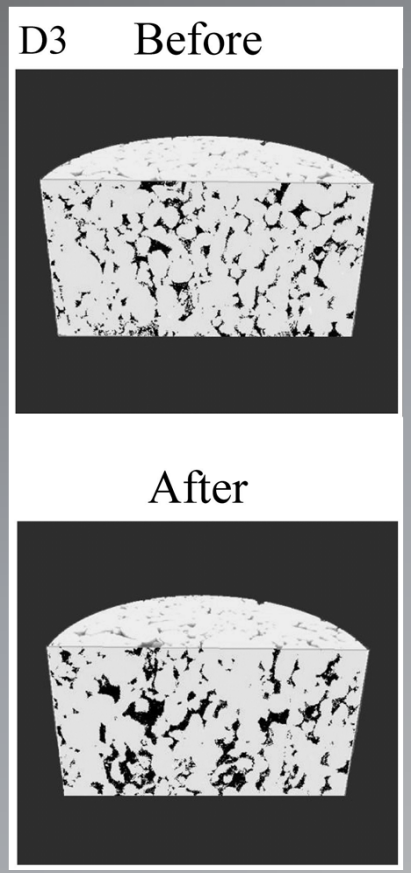
Effect of structure heterogeneities on dissolution processes

Accordingly with the Pe and Da numbers homogeneous dissolution is expected for both experiments

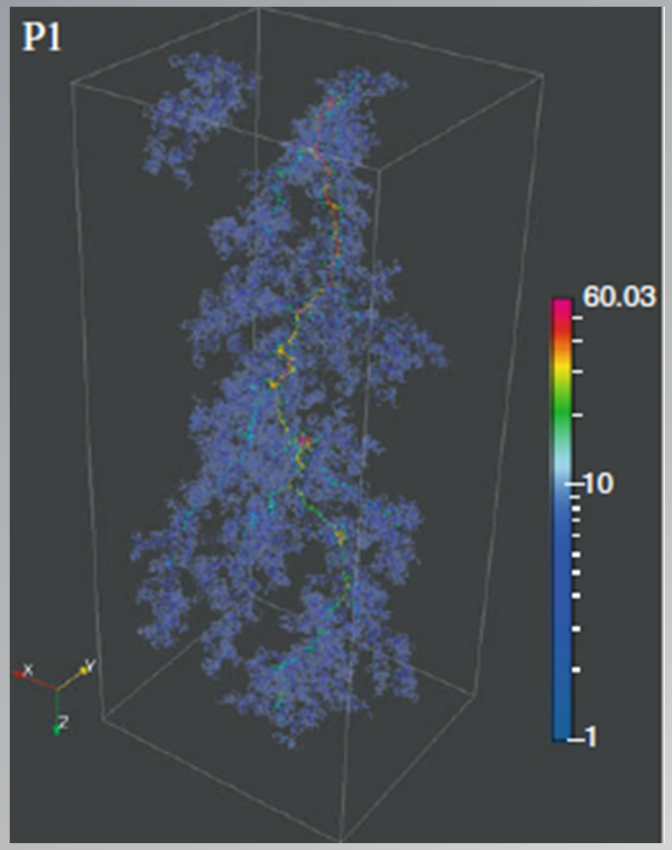


Golfier et al., 2002 J. Fluid Mech.

Effect of structure heterogeneities on dissolution processes

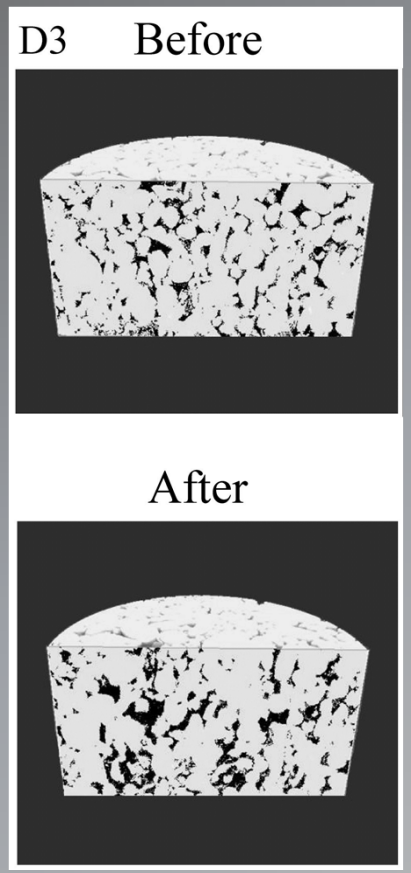


Uniform dissolution for the macro-porous limestone

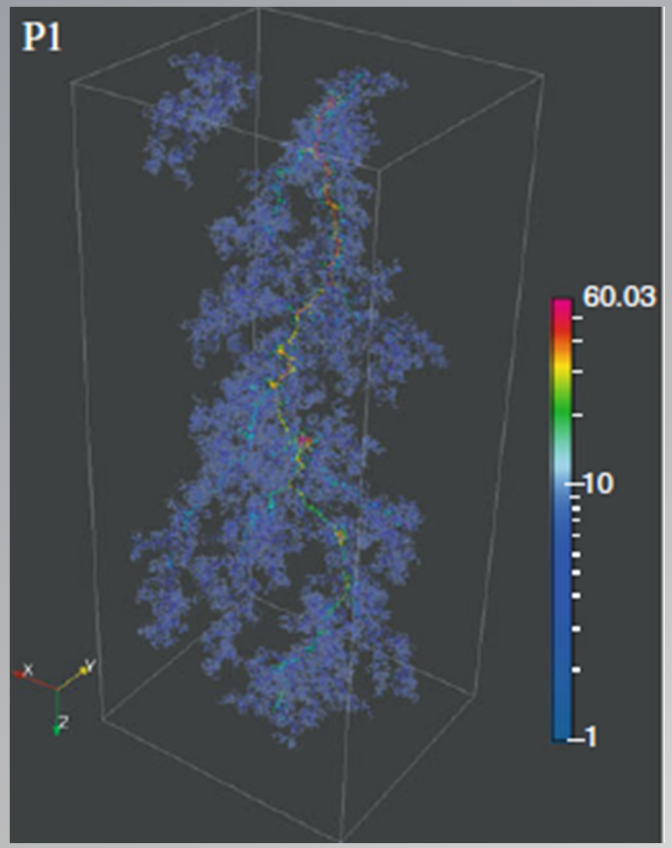


Wormhole formation for the micro-porous limestone

Effect of structure heterogeneities on dissolution processes



Homogeneous dissolution for the macro-porous limestone



Wormhole formation for the micro-porous limestone

Initial **pore structure** (connectivity, reactive surface /pore volume ratio, local transport control) seems to be an important factor **controlling the dissolution features and localization.**

Contents

- Effect of structure heterogeneities
  - on dissolution processes (localization and rate)
  - on reaction type

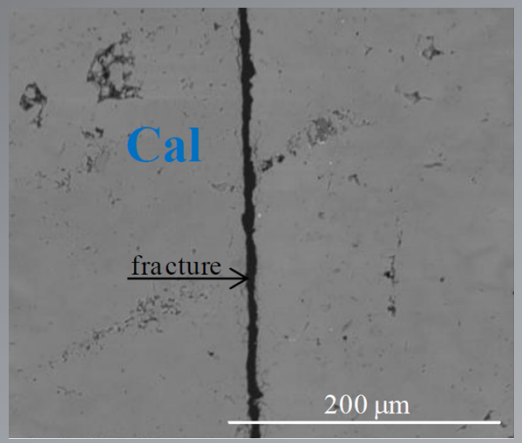
- Effect of mineral heterogeneities
  - on dissolution processes (localization and rate)
  - on reaction type

Effect of mineral heterogeneities on dissolution processes

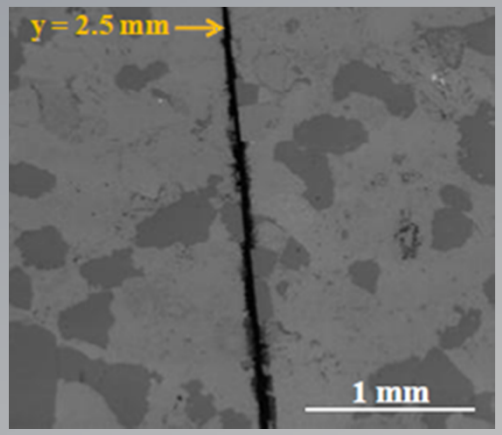
Injection of CO_2-rich brine through fracture core samples

T = 60°C, P = 15 MPa, P_{CO2} = 6 Mpa, Q = 60 mL/h

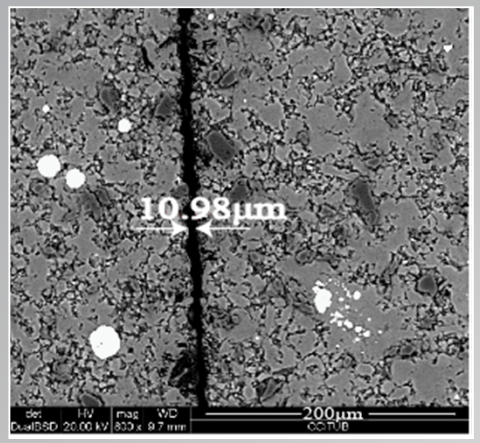
$$CaCO_3 + H_20 + CO_2 \longleftrightarrow Ca^{2+} + 2HCO_3^-$$



Limestone
(100% calcite)



Sandstone
(67% calcite, 33% quartz)



Marl
(70% calcite, 10% quartz,
18% clays, 2% oxides)

Garcia-Rios etal., ChemGeol, 2015
Garcia-Rios etal., AppGeochem, 2017
Davila etal., IJGGC, 2016

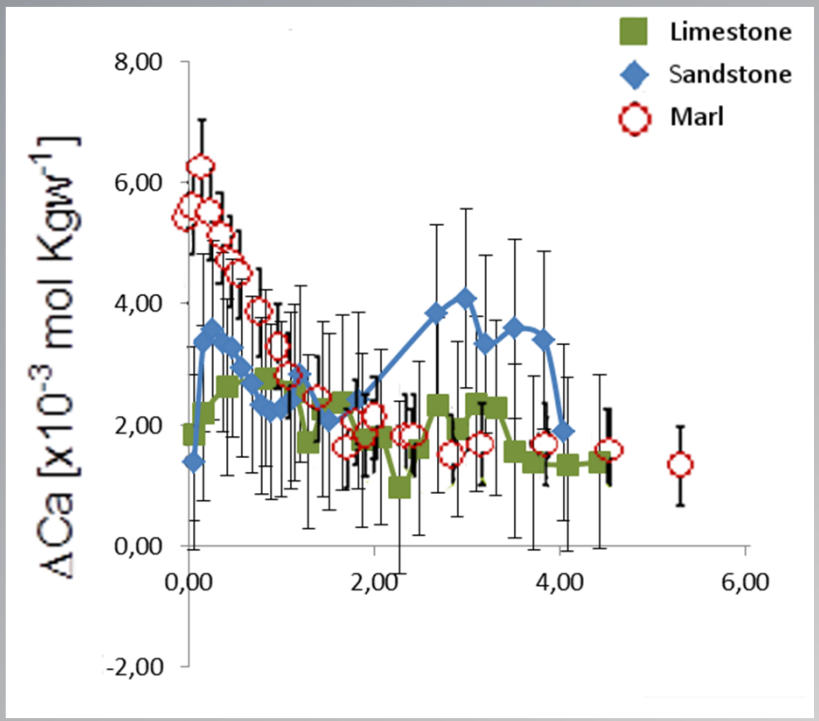
Effect of mineral heterogeneities on dissolution processes

Calcite dissolution for the 3 samples but at different rate:

- Limestone 4,9 mm^3/h
- Sandstone 6,8 mm^3/h

} Constant along the entire experiments

- Marl from 10,3 to 4,6 mm^3/h



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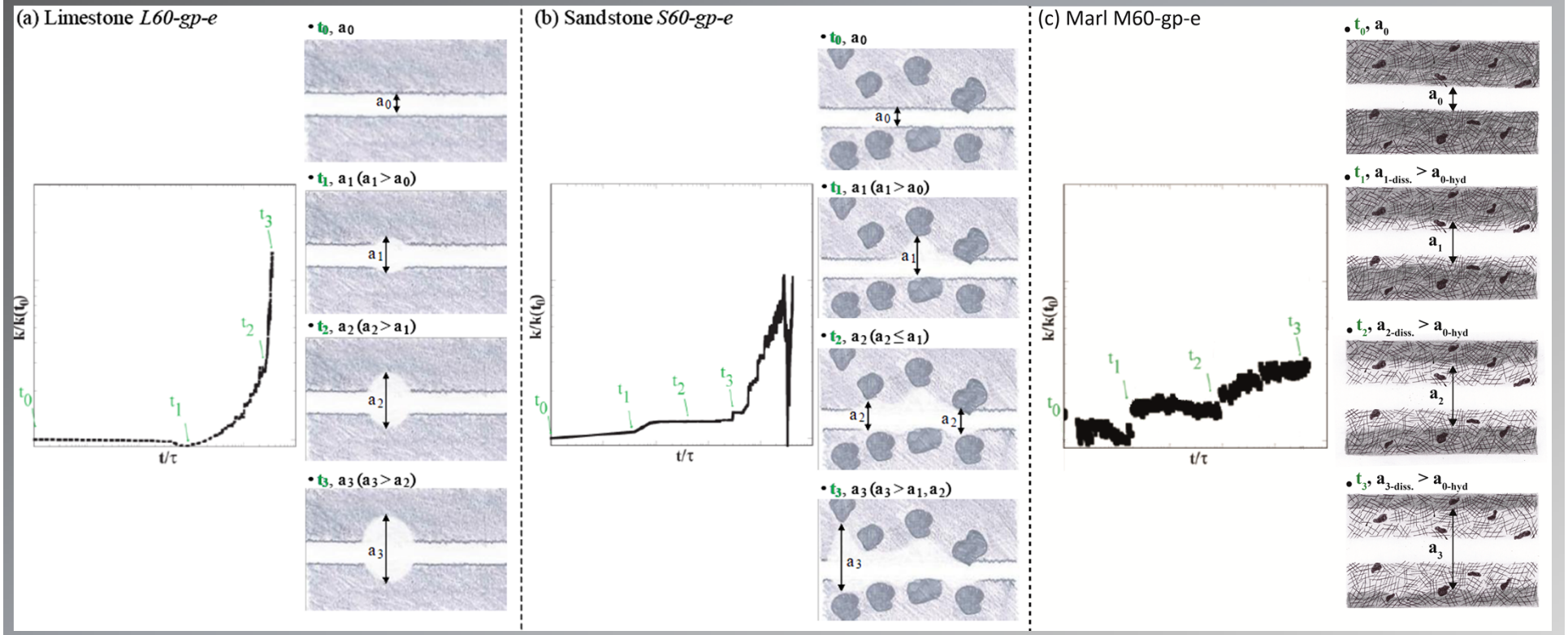
Effect of mineral heterogeneities on dissolution processes



- Inert silicate grains in the sandstone experiments favored more extended dissolution features
- Limestone dissolution tended to be localized (wormhole), whereas sandstone dissolution tended to be extended (uniform).
- Clay skeleton keep the initial fracture aperture and thus the hydraulic aperture (more homogeneous dissolution)

Garcia-Rios etal., ChemGeol, 2015
Garcia-Rios etal., AppGeochem, 2017
Davila etal., IJGGC, 2016

Effect of mineral heterogeneities on dissolution processes

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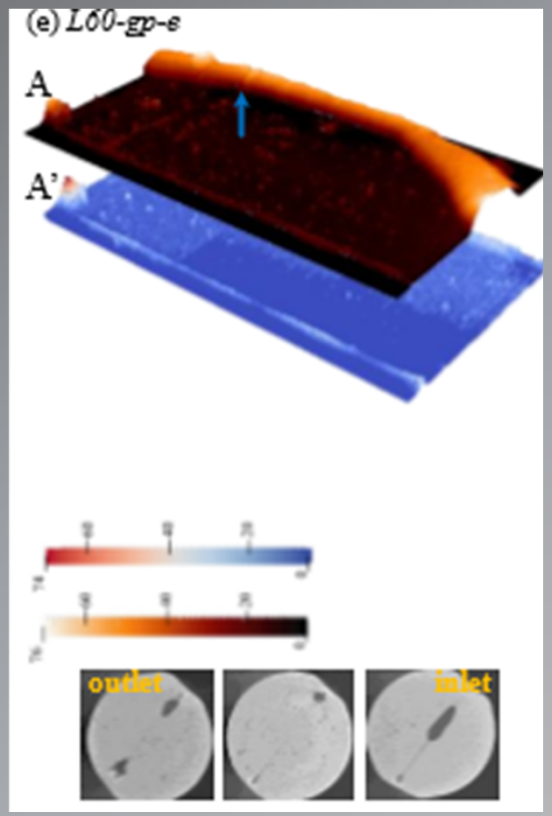

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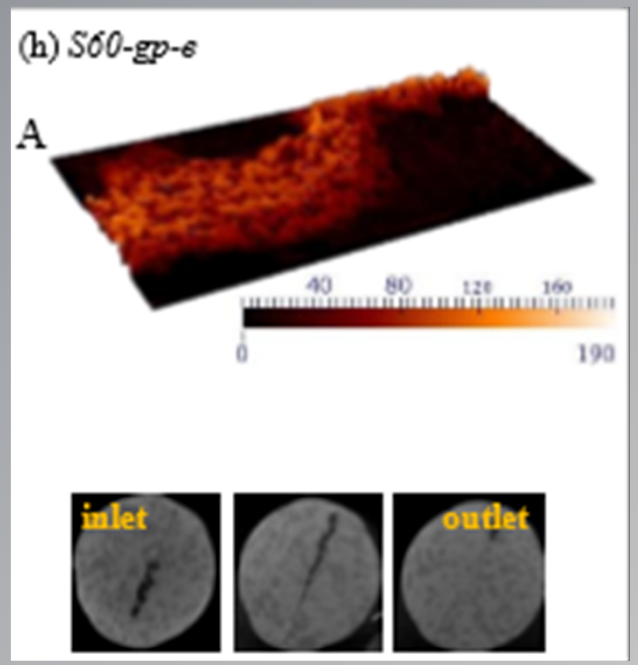

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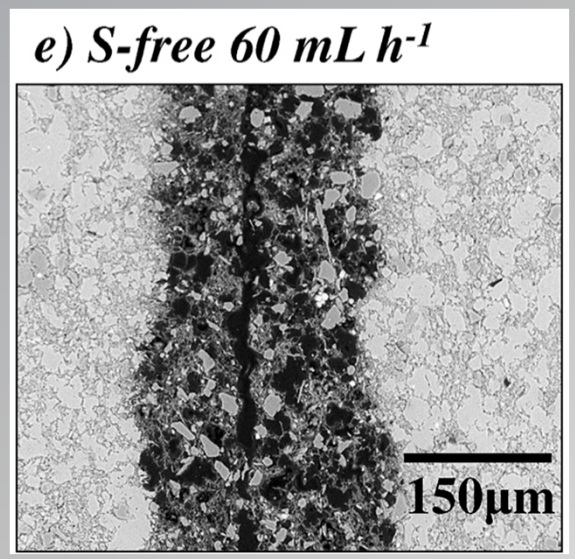

Garcia-Rios etal., ChemGeol, 2015
Garcia-Rios etal., AppGeochem, 2017
Davila etal., IJGGC, 2016

Conclusions

- Most of the processes of dissolution and precipitation are controlled by local heterogeneity (structure, flow, mineralogy)
  - ✓ Wormhole formation is controlled by the pore size diameter, local connectivity and tortuosity (role of the micro-porosity)
  - ✓ Role of the non-dissolved grains on the localization of the dissolution (homogenization, more uniform dissolution)

- Local processes affect macroscopic parameters
  - ✓ Relationship as k-ϕ, S_r-ϕ always depend on the rock structure and not only on the reactivity and the transport boundary conditions (Pe and Da numbers) (control of CO_2 injectivity)
  - ✓ Rate of dissolution and precipitation are controlled by mixing process and mineral distribution (volume of stored CO_2)

Take home message

To characterize and predict chemical processes (dissolution, precipitation, redox reactions, …), it is necessary to take into account the coupled reactive-transport processes and the local structural, hydrodynamic and mineralogical heterogeneities.

Thank you