Geoelectrical monitoring of dissolution and precipitation reactions in a saturated calcareous porous medium

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Experimental setup
Geoelectrical monitoring of dissolution and precipitation reactions in a saturated calcareous porous medium

Background

Experimental results

Conclusions

Experimental setup
Carbonate reservoirs are **essentials**

- They are worldwide distributed

**Background**

The main issues of this study

**Karst Aquifer Maps from Chen et al. (2017)**

- Carbonate rocks (continuous / discontinuous)
- Large freshwater lake
Carbonate reservoirs are **essentials**

- They are worldwide distributed
- They cover many needs and are related to many problematics

*Carbonate issues (from the French Geological Survey, BRGM, 2009)*
In a near-surface context carbonate reservoirs form complex karst-systems.

This karstic shape is driven by chemical reactions caused by the water flux.

Which methods should we use to study dissolution and precipitation processes?
Spectral induced polarization

This electrical method is conducted through the injection of an electrical current and the measure of the resulting voltage with unpolarizable electrodes.

The alternating current is injected at several frequencies. The measured voltage is sinusoidal: it oscillates at the same frequency but is temporally shifted.

The resulting resistivity is a complex number which possesses an amplitude and a phase.
Spectral induced polarization

The SIP method allows to get the spectra of these two values over the explored frequency range.

While the amplitude is linked to the microstructure of the porous medium, the phase gives also insights about the mineral surface state.

This information is crucial for the study of rock-water chemical processes.

\[
\frac{dV(\omega)}{I(\omega)} \propto \rho^*(\omega) = |\rho(\omega)| e^{i\varphi(\omega)}
\]
Spectral induced polarization

The study of Wu et al. (2010) highlights that during precipitation of calcite grains in a glass beads packed cell:
- The phase peak grows
- And is shifted towards low frequencies.

Leroy et al. (2017) developed a mechanistic model to interpret this evolution as the growth of the grains’ size during the experiment.
Self potential

This passive method measures the electrical current generated by natural sources.

These sources can be:
- Electrokinetic
- Electrochemical

Cherubini et al. (2019) measured SP signal of brine flowing through limestone.
- The electrokinetic source is observed at the beginning of the experiment
- Then the electrochemical source is added with the injection of CO$_2$ with the brine.

SP reaches a plateau after CO$_2$ release
Experimental results

**Experimental setup**

Calcite grains $125 \mu m < d < 250 \mu m$

Pump

Upstream solution

Column

$P_1$ $P_2$ $P_3$ $P_4$

Tightening structure

25 cm

Downstream solution

Sampling

Pore water chemistry monitoring

Alkalinity

Concentrations

pH

Saturation state

$\Omega = \text{ion activity product} / \text{Solubility constant}$

Experimental results

Alkalinity

Concentrations

pH

Saturation state

$\Omega = \text{ion activity product} / \text{Solubility constant}$

Calcite grains $125 \mu m < d < 250 \mu m$

Air-conditioned room

$T = 21,8 \pm 1,3 ^{\circ}C$
Experimental setup

Pump

Upstream solution

σ_{up}

Column
\( P_1, P_2, P_3, P_4 \)

Tightening structure

Downstream solution

Sampling

Alkalinity

Concentrations

pH

Saturation state
\[ \Omega = \frac{\text{ion activity product}}{\text{Solubility constant}} \]

Experimental results

Pore water chemistry monitoring

Pore water conductivity

Geo-electrical monitoring
Experimental setup

Pore water chemistry monitoring

Saturation state
\[ \Omega = \frac{\text{ion activity product}}{\text{Solubility constant}} \]

Alkalinity
Concentrations
pH

Experimental results

Pump
Upstream solution

σ_{up}

P_1 P_2 P_3 P_4

Column

σ_{down}

Downstream solution

Sampling

Tightening structure

Geo-electrical monitoring

Self potential (SP)

Pore water conductivity

Experimental results

Pore water conductivity

Self potential (SP)
Experimental setup

- Upstream solution
- Pump
- Column $P_1, P_2, P_3, P_4$
- Downstream solution
- Sampling
- Tightening structure
- Pore water chemistry monitoring
- Alkalinity
- Concentrations
- pH
- Saturation state
  \[ \Omega = \frac{\text{ion activity product}}{\text{Solubility constant}} \]

- Experimental results
  - Pore water conductivity
  - Self potential (SP)
  - Spectral induced polarization (SIP)

- Geo-electrical monitoring

PSIP

5 mHz → 10 kHz
I - Initialization
Reach a stationary state
Fluid injection and measurements
II - Dissolution
III - Precipitation

Experimental results
Pore water monitoring

*Upstream and downstream pore water electrical conductivity*

Saturation state calculated from the measurements of water samples collected downstream.

Stabilization until the saturation state.
Pore water monitoring

*Upstream and downstream pore water electrical conductivity*

Saturation state calculated from the measurements of water samples collected downstream

**Experimental results**

Decrease then stabilize

Reach the saturation state

Upstream solution

Downstream solution

σ<sub>up</sub>

σ<sub>down</sub>
Experimental results

Pore water monitoring

*Upstream and downstream pore water electrical conductivity*

Upstream solution

Saturation state calculated from the measurements of water samples collected downstream

Reach the saturation state

New plateau

Experimental results

Pore water monitoring

*Upstream and downstream pore water electrical conductivity*

Upstream solution

Saturation state calculated from the measurements of water samples collected downstream

Reach the saturation state

New plateau
During all the experiment, we observed no polarization. Why?
We succeed to predict $[\text{Ca}^{2+}]$ with this 1D approach during both dissolution and precipitation.

The adjusted models show that the porosity change due to chemical processes on the sample is small and occurs at the entrance of the column.

Dissolution and precipitation occur in the first centimeter of the sample, which generates no SIP signal.
Self potential results

- Evolution of the intrinsic potential of the electrodes
- Transient variations after the injection of a new fluid
Self potential results

• Evolution of the intrinsic potential of the electrodes
• Transient variations after the injection of a new fluid

Variations begin staggered in time and end simultaneously.
Self potential results

• Evolution of the intrinsic potential of the electrodes
• Transient variations after the injection of a new fluid
• These SP signals come from an electrochemical coupling due to ionic concentration gradients

Variations begin staggered in time and end simultaneously.

Experiment of NaCl diffusion in a sand matrix from Maineult (2004) with SP monitoring on 4 channels:

• Variations are related to NaCl diffusion
• The solute front speed can be quantified
Self potential results

- Evolution of the intrinsic potential of the electrodes
- Transient variations after the injection of a new fluid
- These SP signals come from an electrochemical coupling due to ionic concentration gradients
- There is a good correlation between $\sigma_w$ and SP variations
In a new dissolution experiment, acid injection was shifted to locate the reactive zone between electrodes P₁ and P₂.

We observe that the measured voltage for channel P₁-P₄ is different from the others because of dissolution process. ⇒ The injection shift allows to identify the reactive zone.

However, the phase spectrum obtained with SIP remained flat.
⇒ Dissolution generates no phase signal
Conclusions

- Successful monitoring of dissolution and precipitation processes with the self potential method
- No SIP response in dissolution
- Good agreement and complementarity between geoelectrical methods and geochemical monitoring and modeling

Next steps

- Quantitative interpretation of self potential measure using the junction potential theory
- New acquisition during precipitation process with a better control of the injection location
Thank you for watching.
I look forward for your questions.

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