Carbon Dioxide Reaction Processes in a Model Brine Aquifer at 200°C and 200 bars: Implications for Subsurface Carbon Sequestration

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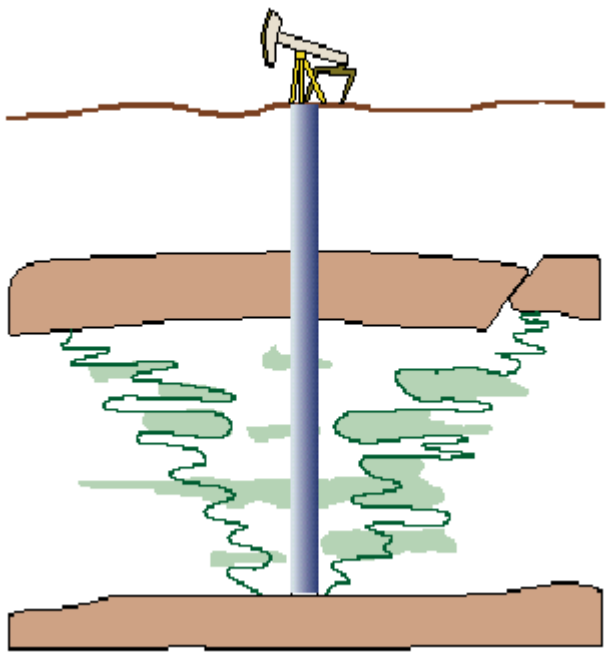
Background - Geologic Sequestration of CO_2



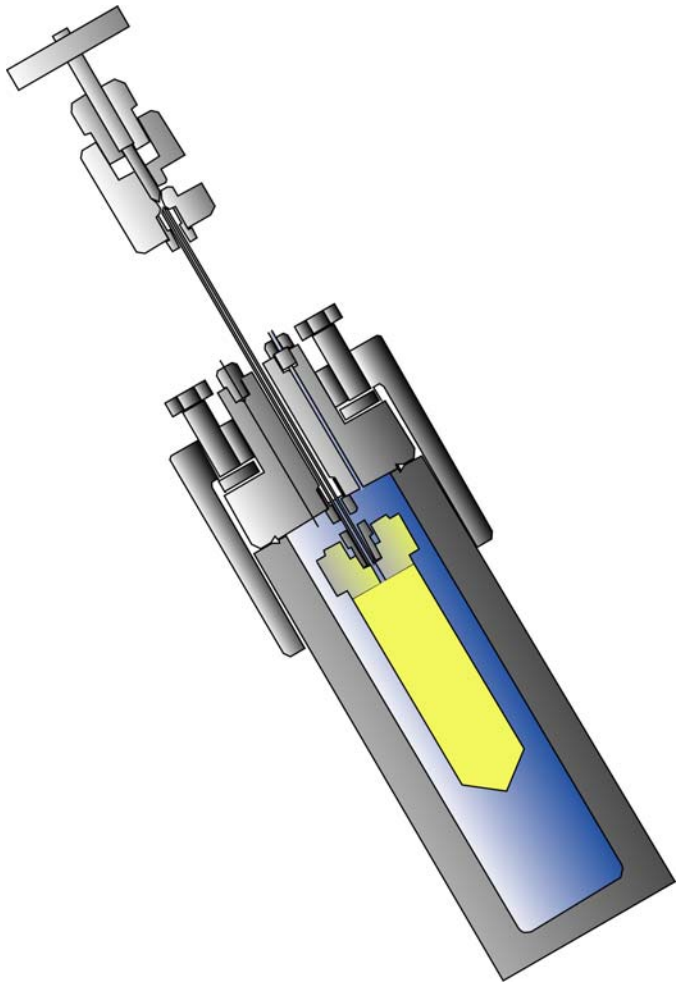
from Carbon Sequestration R&D, US DOE (1999)

- Carbon management in response to global warming
- Targets
  - Saline aquifers
  - Hydrocarbon reservoirs
  - Coal beds
- "Trapping" mechanisms
  - Stratigraphic/structural
  - Hydrodynamic
  - Mineral
- What is the geochemistry?

Objectives for this Study

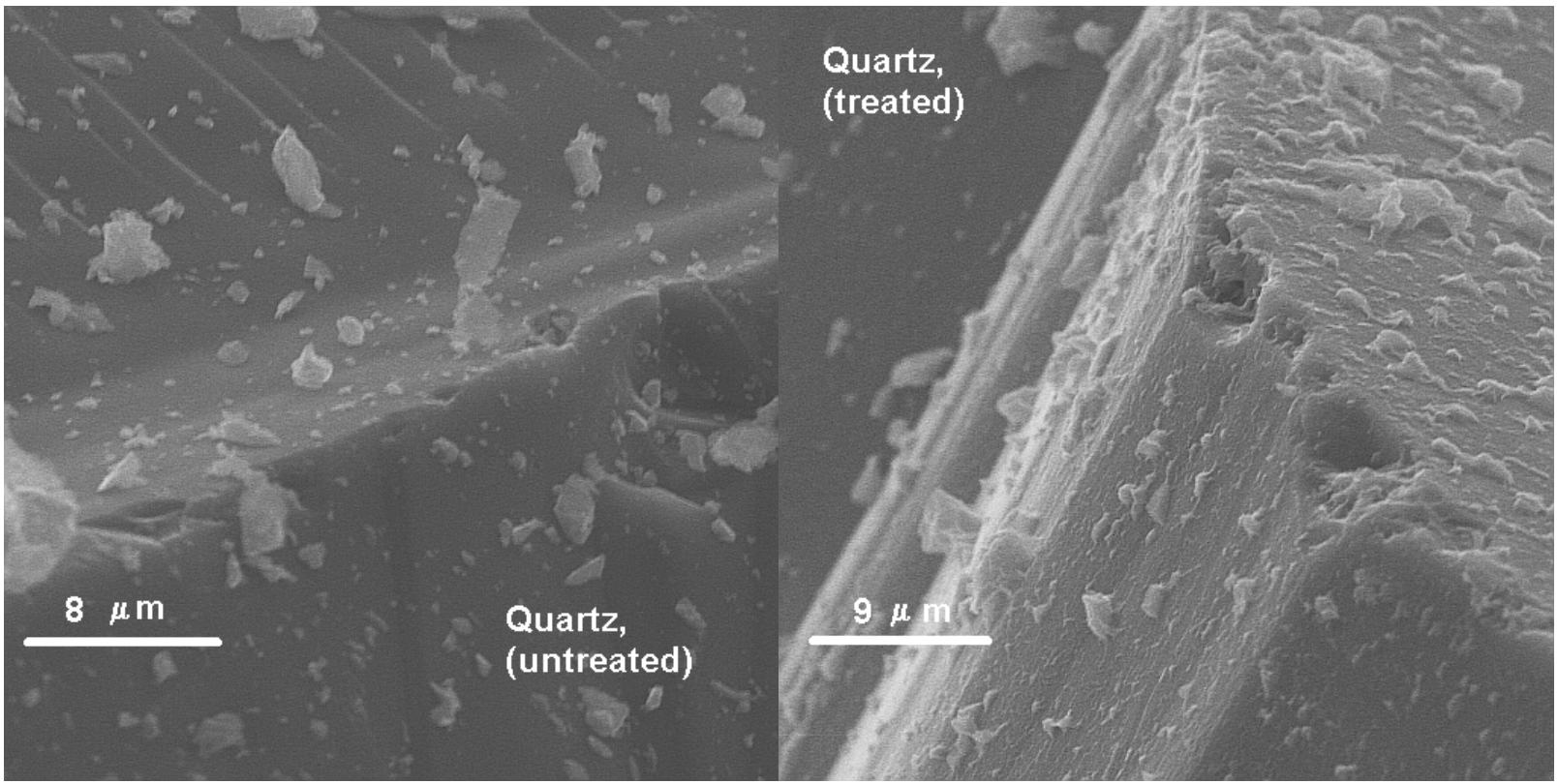
- Experimentally examine CO_2 liquid for geochemical reaction processes
- Investigate model aquifer-aquitard-brine system using hydrothermal apparatus

Experimental Approach

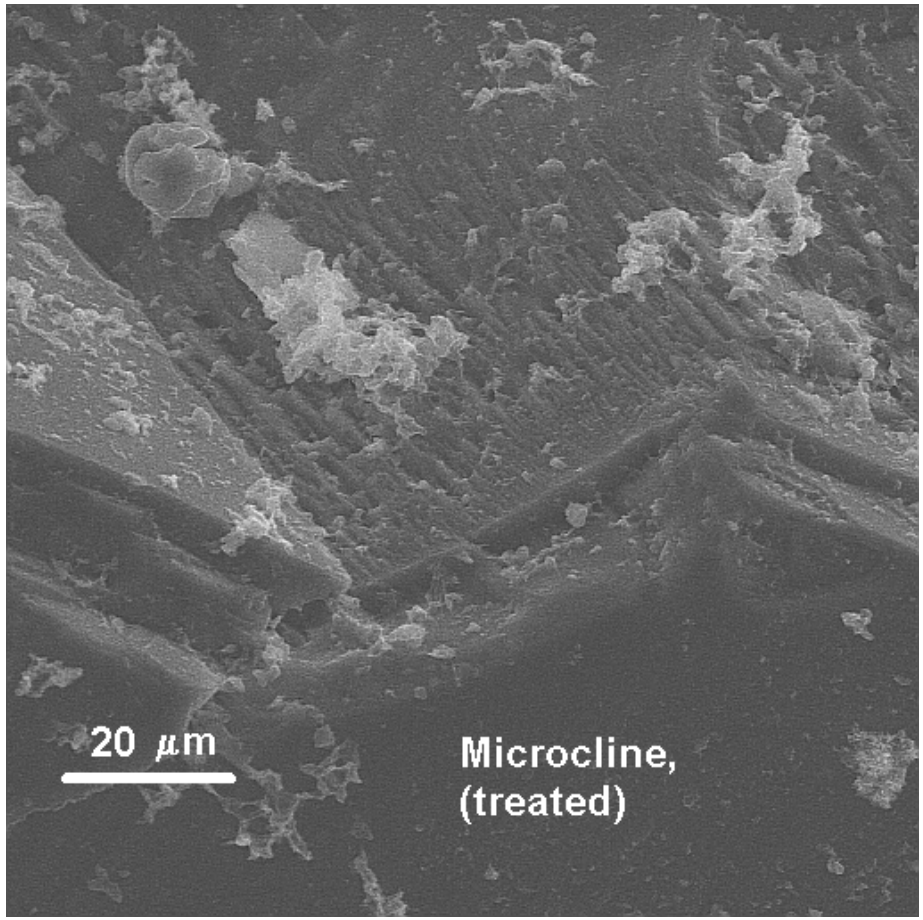


- Flexible cell hydrothermal apparatus
- 200°C and 200 bars
- 5.5 molal brine
- Aquifer = model arkose
- Aquitard = argillaceous shale
- Equilibrate 60 days
- Inject CO_2 into ongoing reaction, 80 days

Quartz

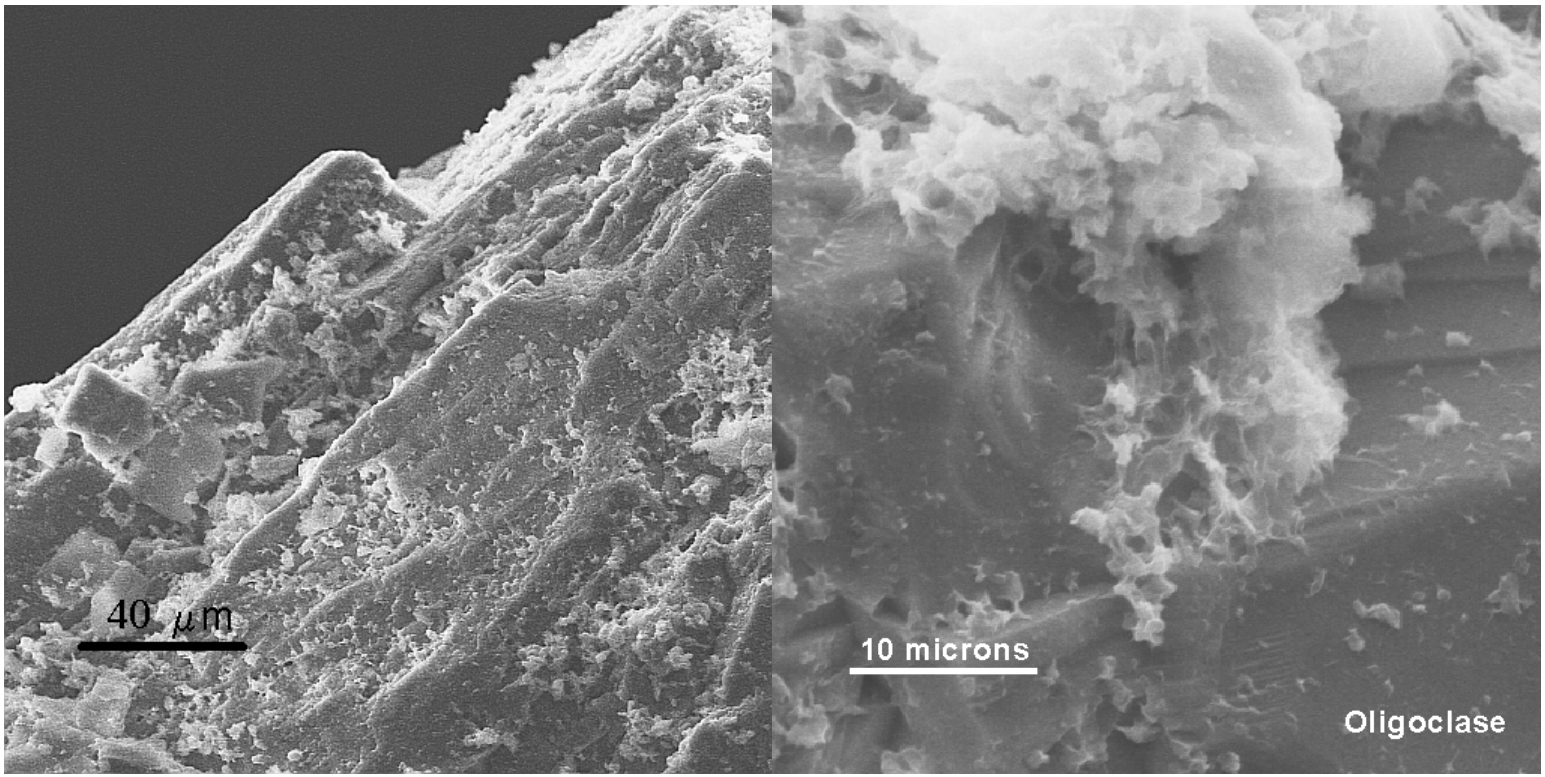


Perthitic Microcline

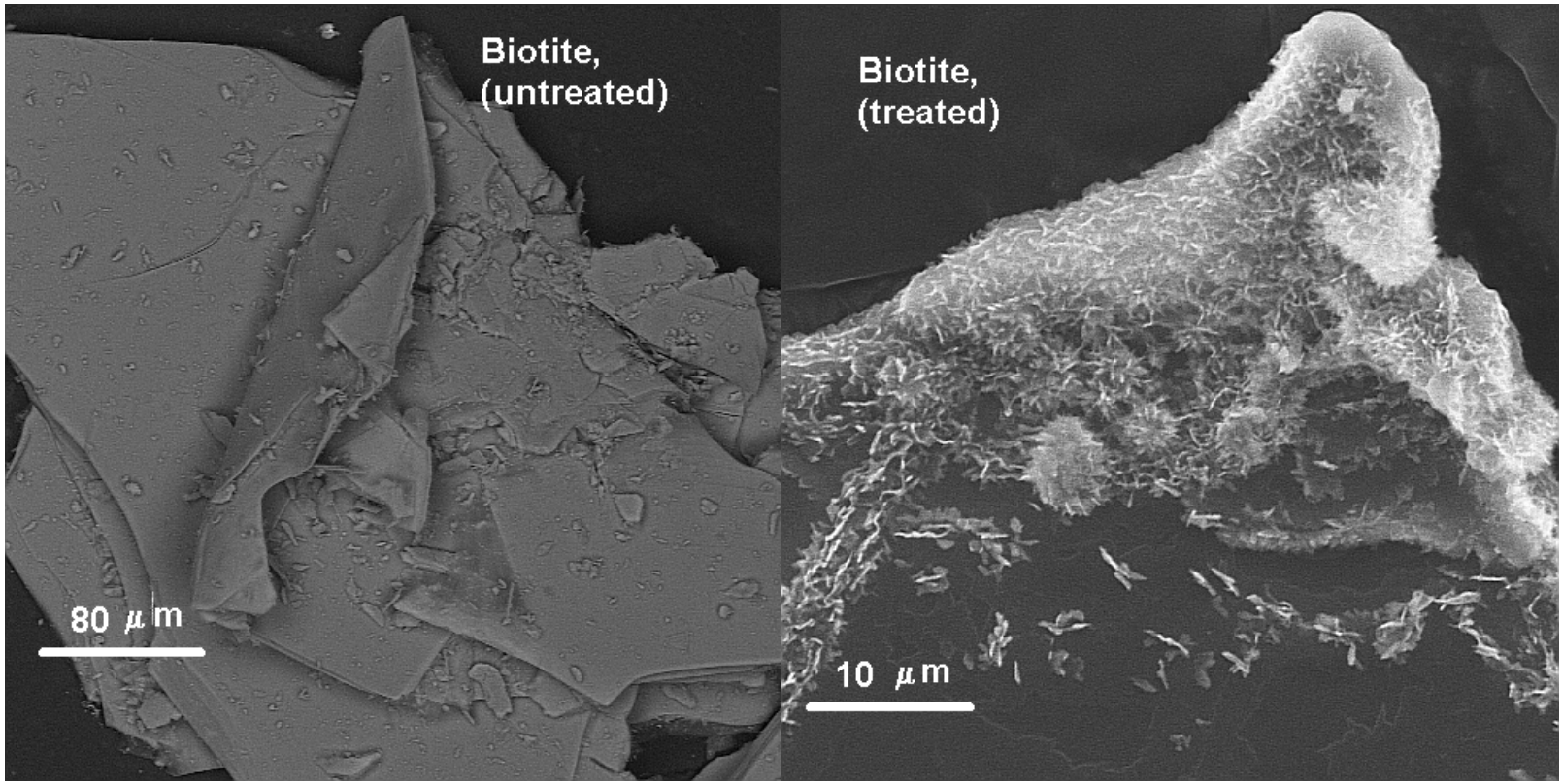


- **Etching of perthite lamellae**

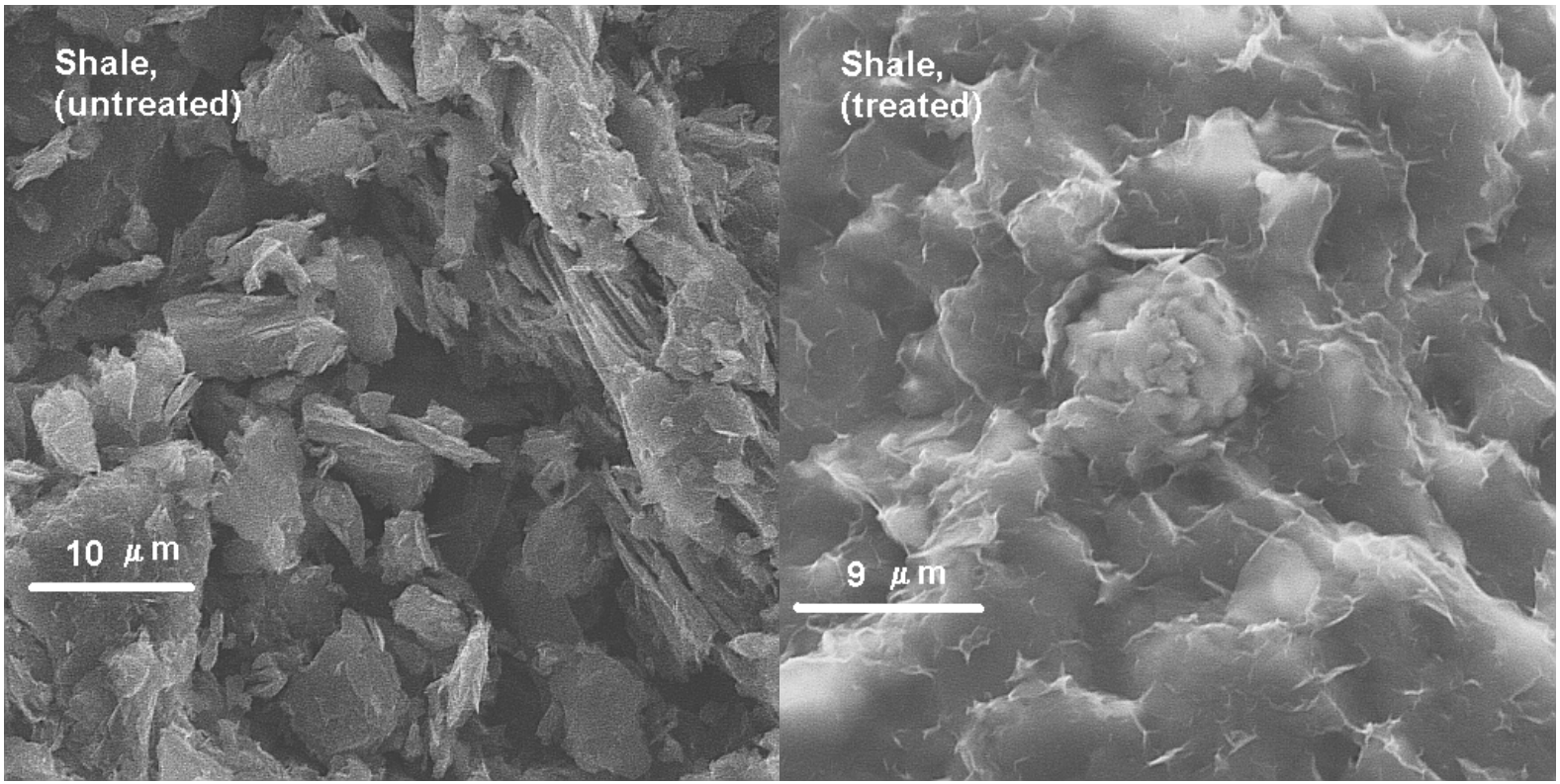
Oligoclase



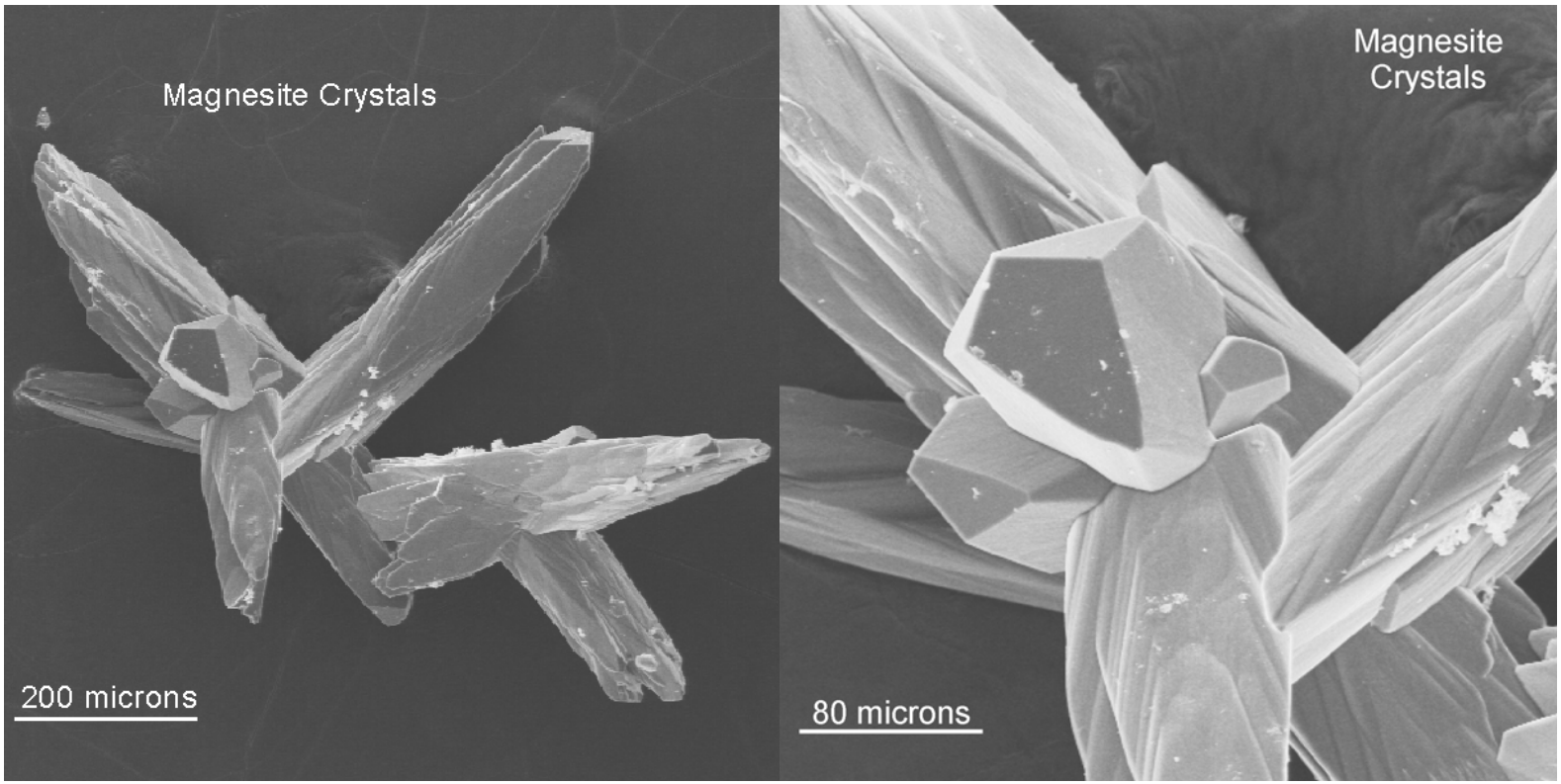
Biotite



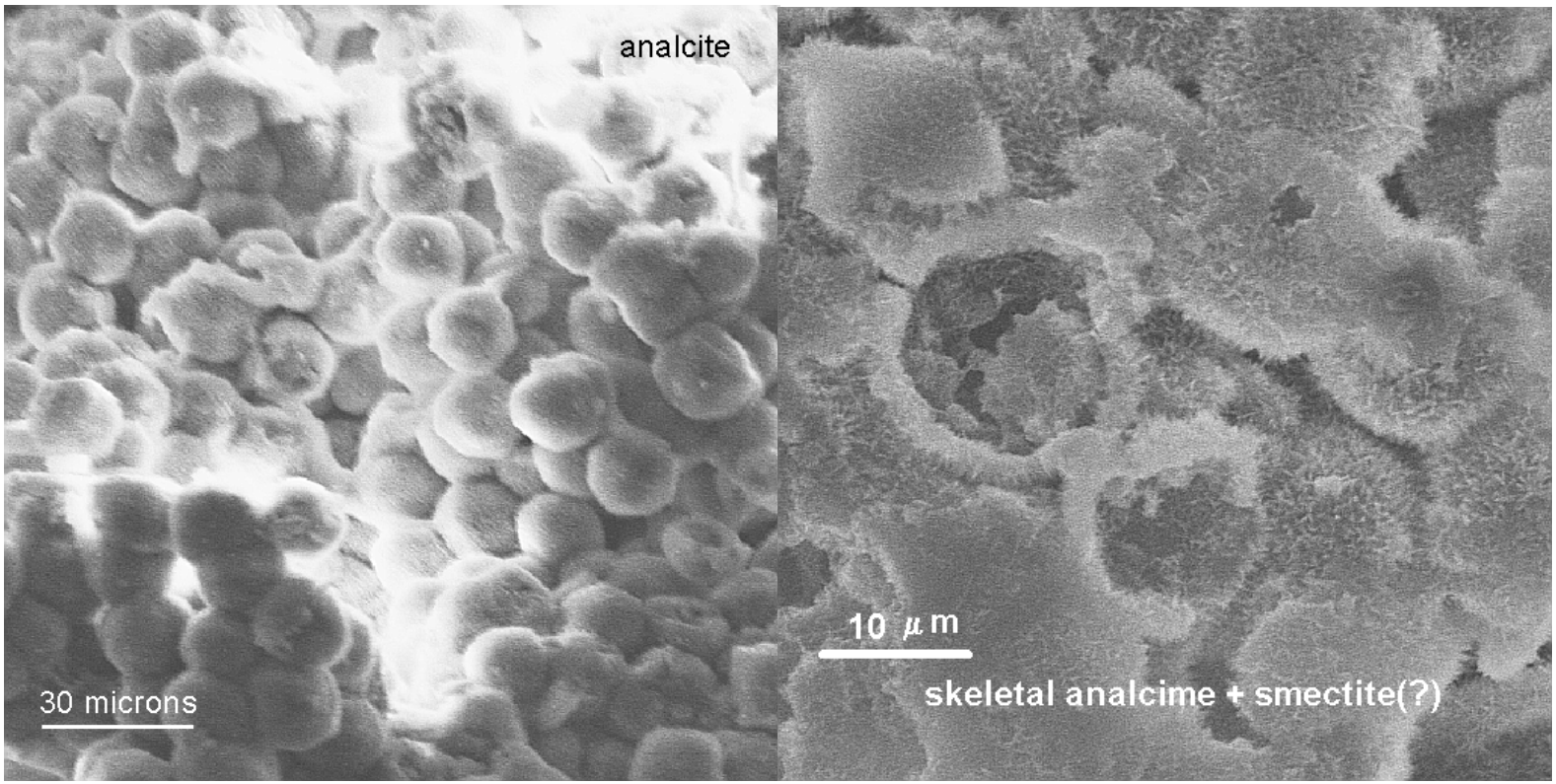
Shale



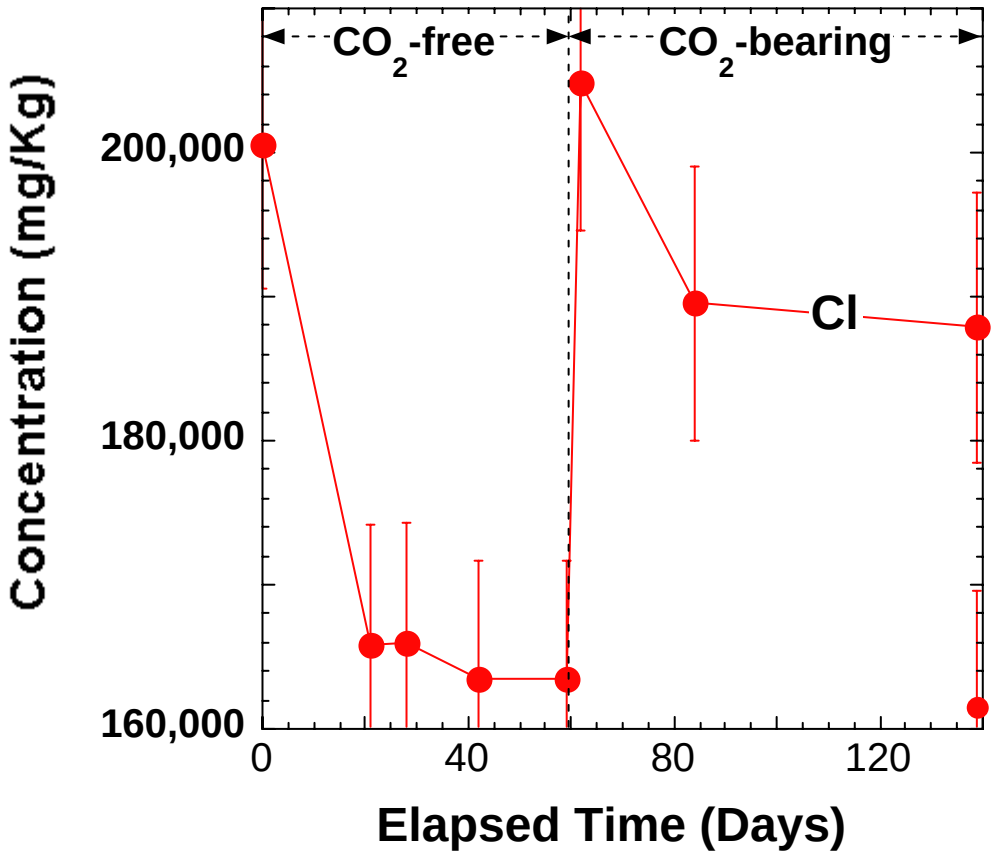
New Phase - Magnesite



New Phases - Analcime Trapezohedrons & Smectite

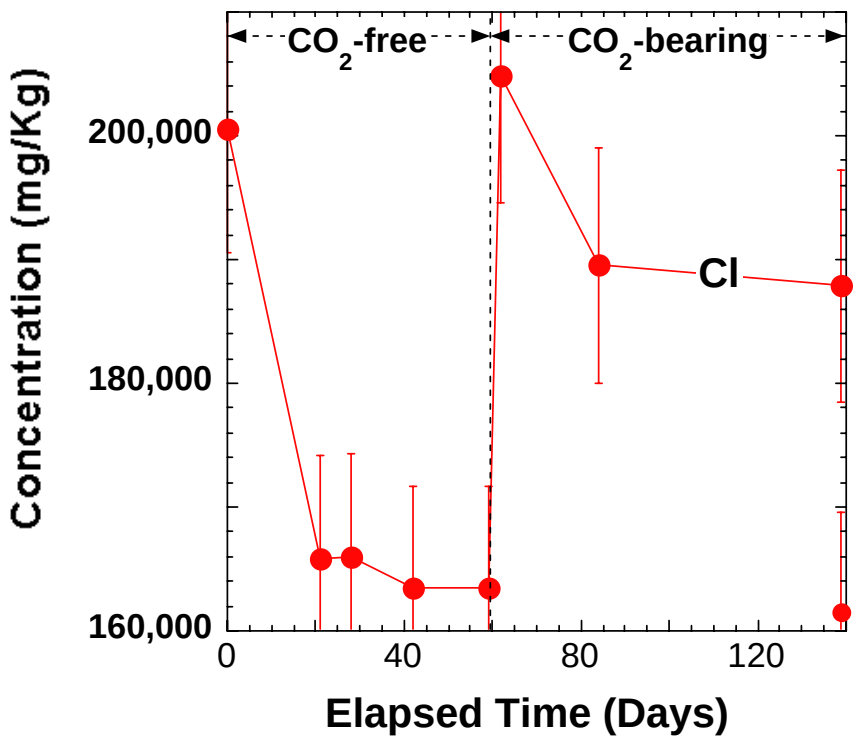


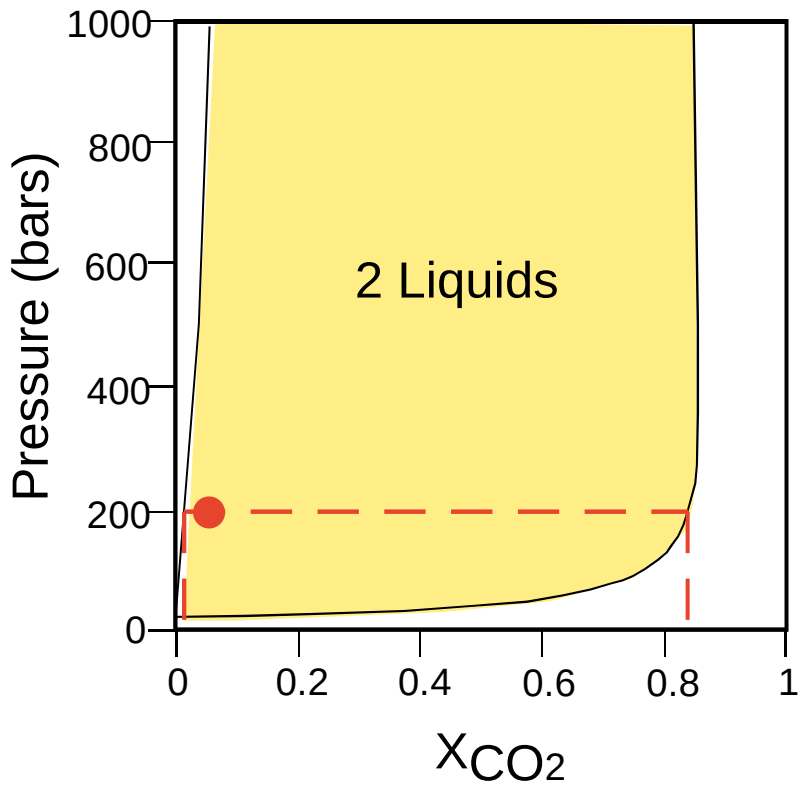
Brine Chemistry - Chloride



- Steady state CO_2-free system
- Increases after CO_2 injected
- Steady state CO_2-bearing system
- Decrease on quench

Pressure-X_{CO2} Phase Diagram for H_2O + CO_2 at 200°C (Shyu *et al*., 1997)





Conclusions

- Injection of CO_2 lead to precipitation of carbonate mineral
- Other fluid-rock reactions take place
  - Brine chemistry changes, providing complex (non-intuitive) chemical reaction environment
  - Components of the arkose (aquifer) and the shale (aquitard) reacting
- Multiphase fluid (brine and CO_2) processes
  - Cl behavior reflects changes in phase equilibrium relationships between H_2O & supercritical CO_2
  - Dessication of brine

Applications and Acknowledgements

- Applications
  - Geologic CO_2 sequestration
  - Petroleum CO_2 flood recovery systems
  - Low-grade metamorphic processes
- Acknowledgements
  - US DOE BES Geoscience Program
  - Los Alamos National Laboratory

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Injection of carbon dioxide into saline aquifer systems is being evaluated as a basis for in-situ geologic storage and/or sequestration. While the technical aspects of collection, injection and formation of a relatively stable carbon dioxide fluid in the rock pore space are essential to storage, the stability of the containment interface is also critical to long-term sequestration. However, the reactive behavior of carbon dioxide under reservoir conditions is largely unknown. Key reaction conditions and components include temperature, pressure, brine and liquid composition, and minerals.

We are continuing to experimentally model the chemical components and reactive behavior of a reservoir-aquitard system. Our experiments employ a flexible cell hydrothermal apparatus consisting of a gold reaction cell within a rocking autoclave. The aquifer is a synthetic arkose (potassium feldspar+oligoclase+quartz+biotite), the aquitard is argillaceous shale, and the brine has an ionic strength of 5.5 molal. After reaction to steady state between brine and minerals at 200 C and 200 bars, carbon dioxide was injected into the system and the experiment continued for 80 days. Ca, Mg, Br, and SO4 decreased in the steady state CO2-brine-arkose-shale system relative to the CO2-free system, whereas K increased. A pressure decrease of 26 bars occurred in the experimental cell over a 3-day period following carbon dioxide injection. Pressure was stable afterwards. The pressure decrease is interpreted as consumption of supercritical carbon dioxide fluid by dissolution in brine and subsequent precipitation as carbonate mineral. SEM and XRD examination of the solids showed euhedral magnesite as a reaction product. Two distinctive morphologies of magnesite were present. SEM analysis also indicates the shale was being consumed by reaction. The experimental reactions provide initial constraints on mineral reactions that may impact the containment interface in moderate temperature brine aquifer systems with potential for carbon sequestration.

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