Synchrotron X-ray Studies of Super-critical Carbon Dioxide / Reservoir Rock Interfaces

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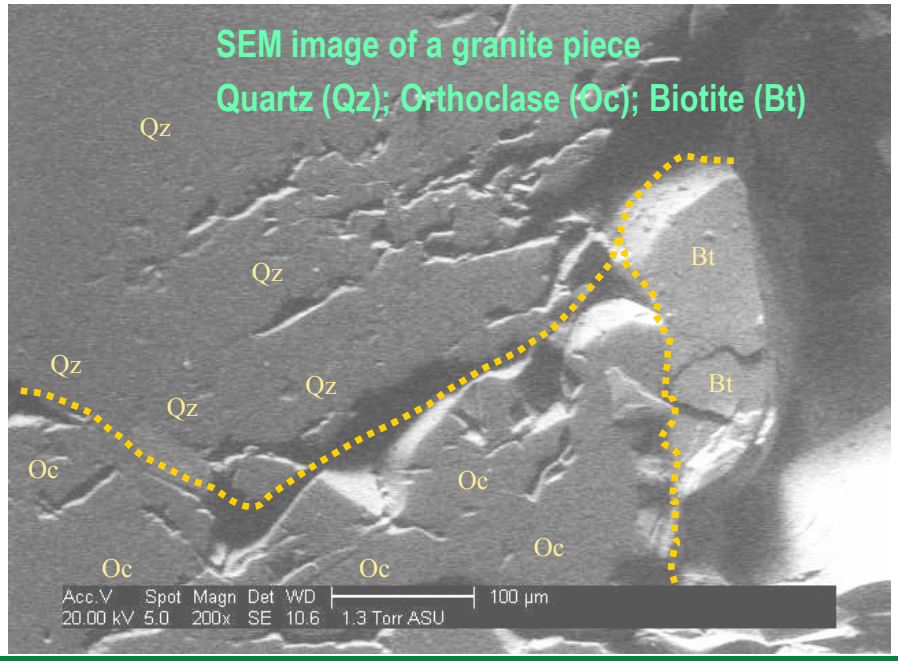
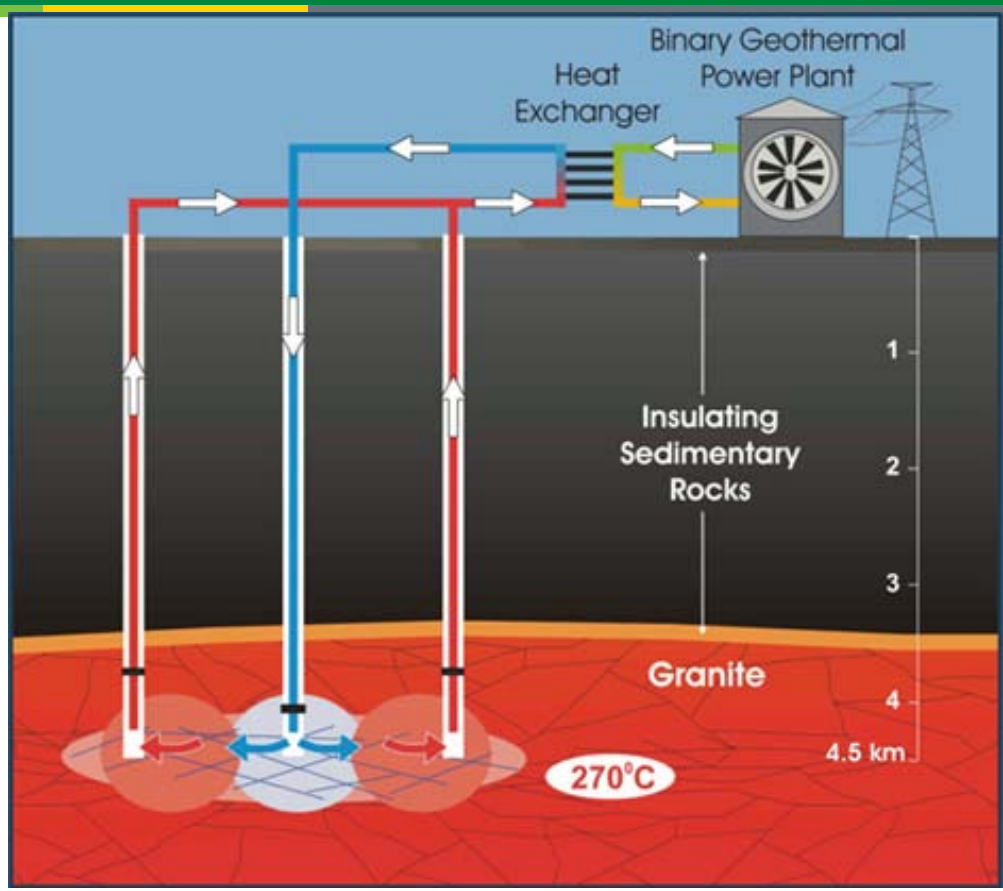
Track Name

Relevance/Impact of Research

Hypotheses to be tested

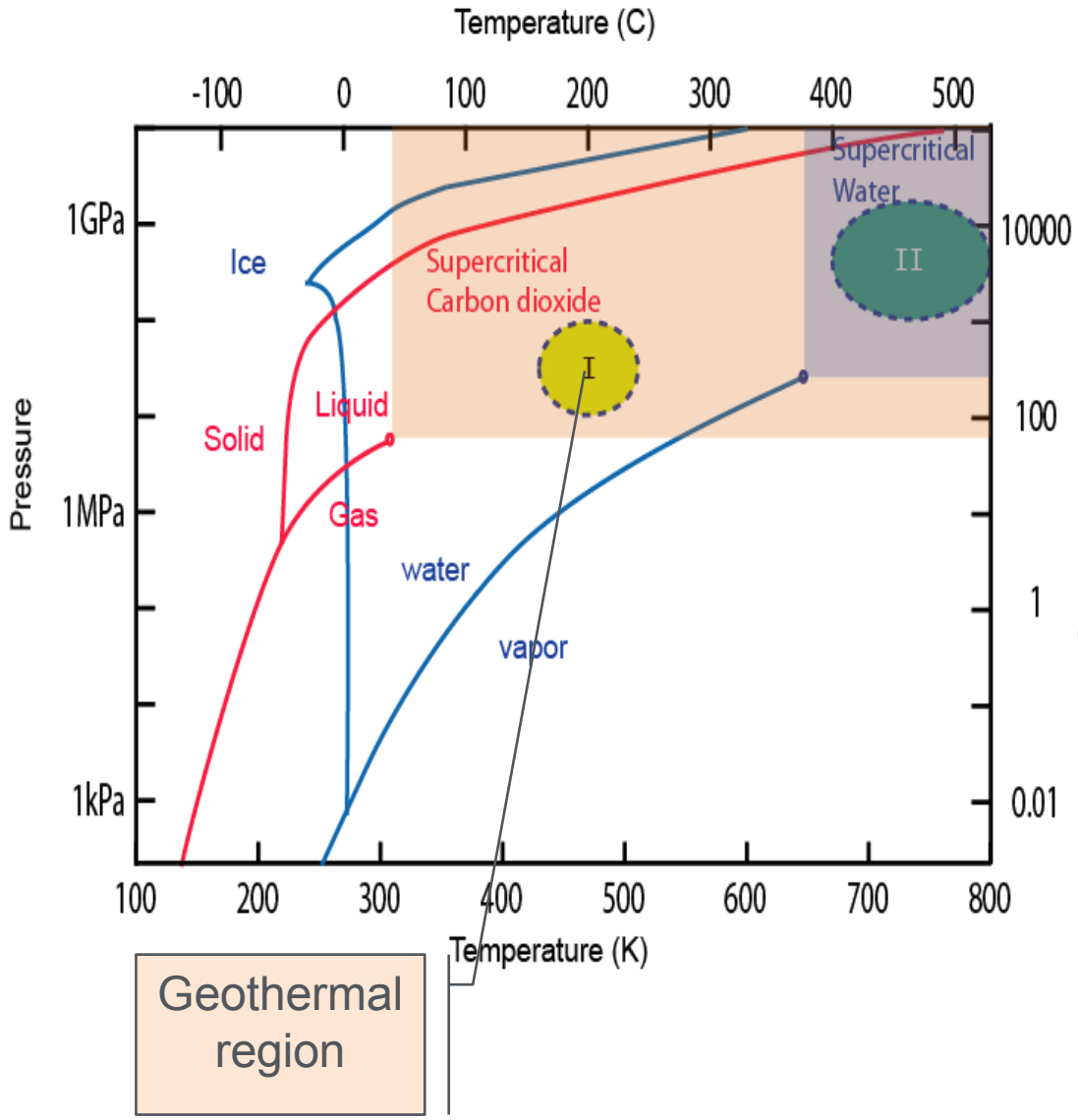
- Supercritical CO_2 will not dissolve and transport minerals
- CO_2 will be partially sequestered
  - Calcite, magnesite formation
  - Granite reaction with $scCO_2$





- Granite composition (Example)
  - Quartz
  - Feldspar (Orthoclase)
  - Mica (Biotite)

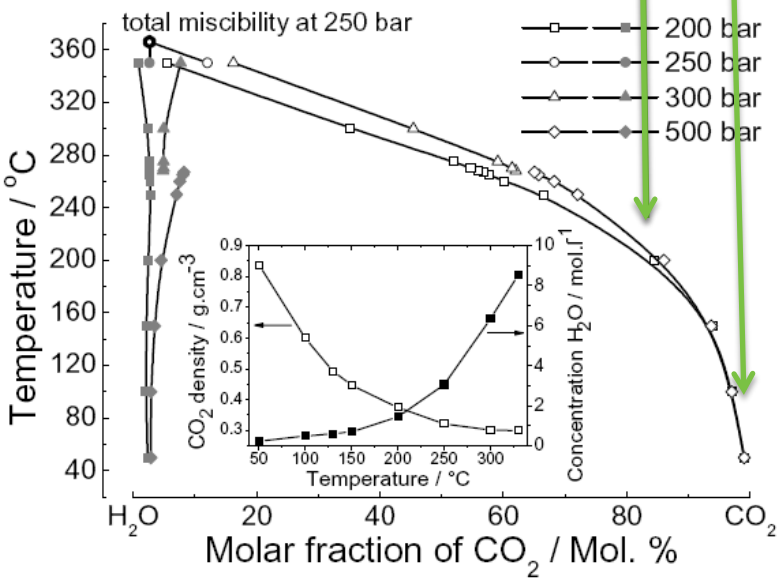
Relevance/Impact of Research
Supercritical CO_2/Water Binary Fluids



Geothermal region

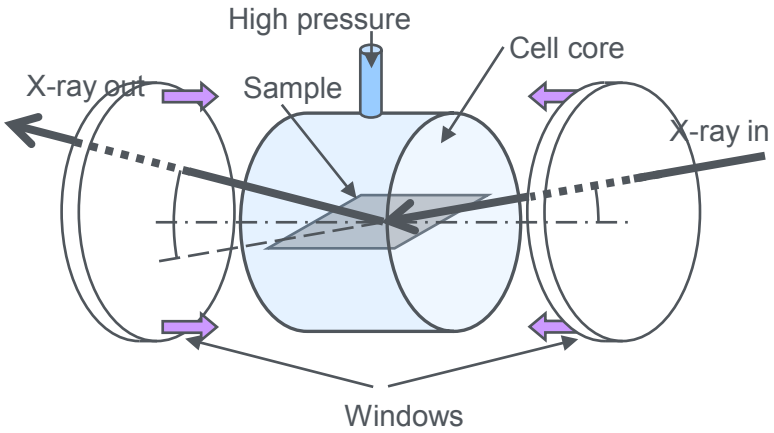
$scCO_2 + H_2O$

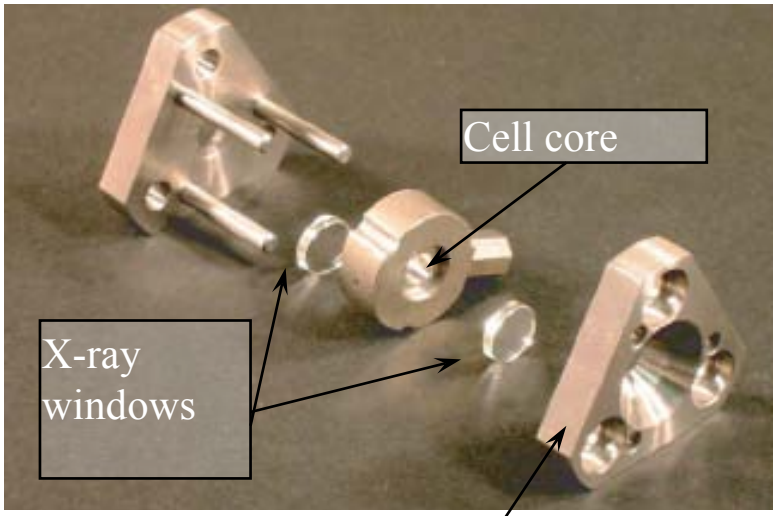
Explore CO_2 rich phases

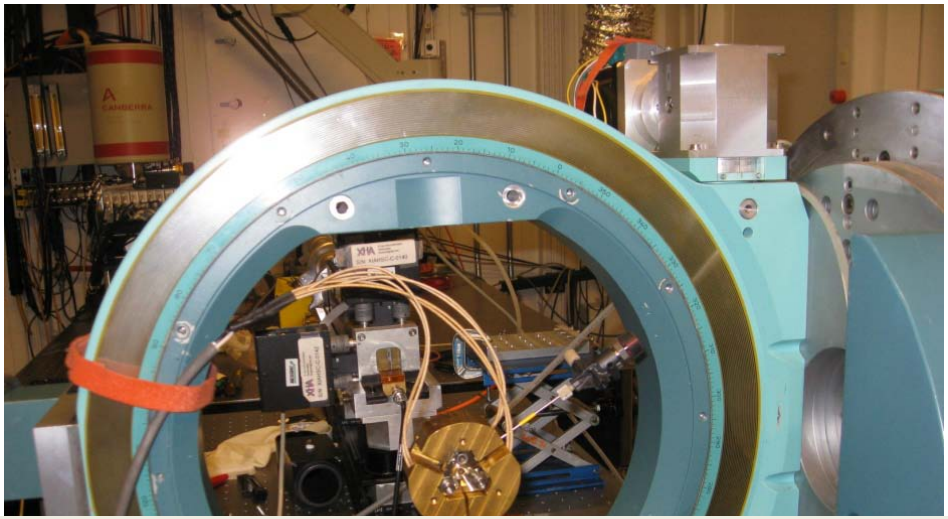


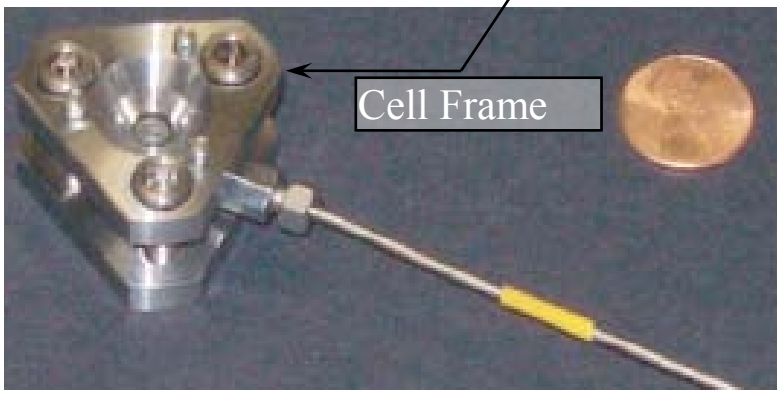
T. Tassaing et al., proc. 14th Int'l Conf. on properties of Water and Steam in Kyoto (04)

Scientific/Technical Approach



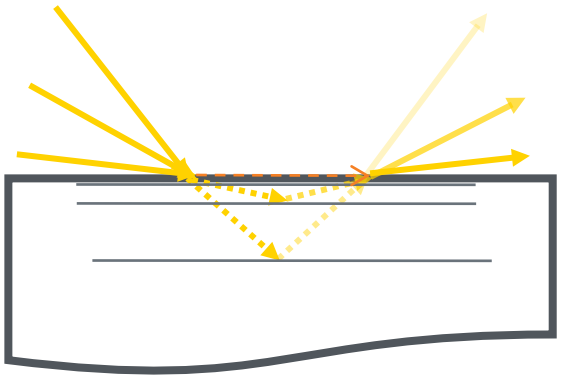




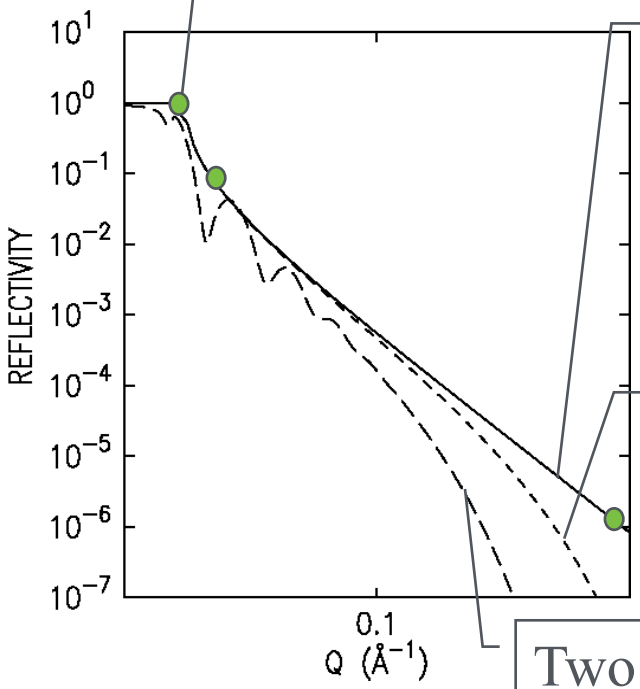


- Window materials: moissanites 1 mm.
- Will explore thinner and more transparent diamond (0.5 mm) windows
- Flow cell design is under way

Scientific/Technical Approach
X-ray Reflectivity --- Specular

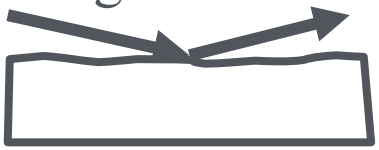


critical angle ~ √density change

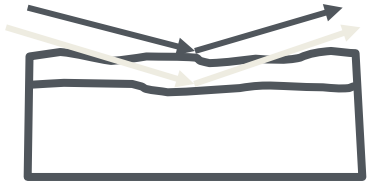


Ideal Surface
~$1/Q^4$

Rough Surface



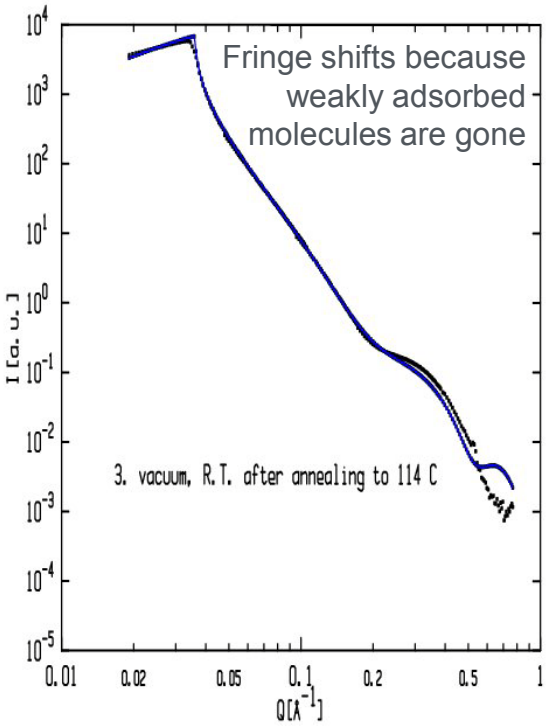
Two Rough Interfaces

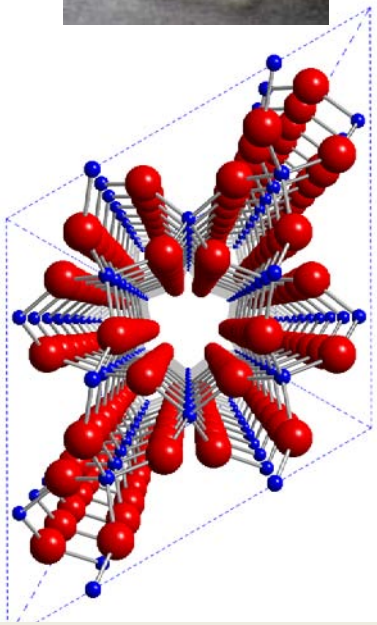


Accomplishments
Preliminary results

- The X-ray/pressure cell
- Silica surfaces
- Mica
- Orthoclase

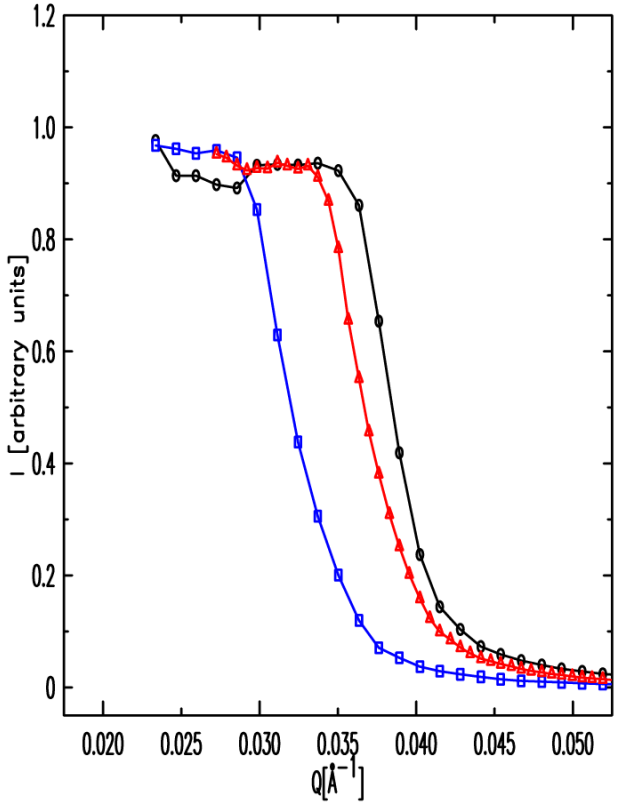
Proof of Principle Experiment
Quartz (0001)





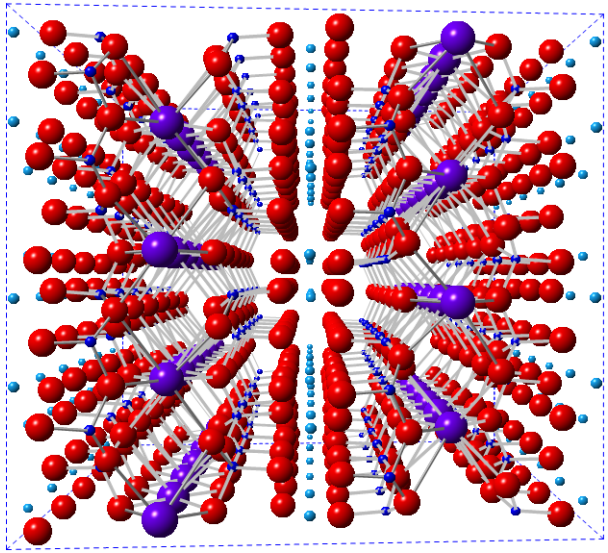
scCO$_2$ + heat
anneal

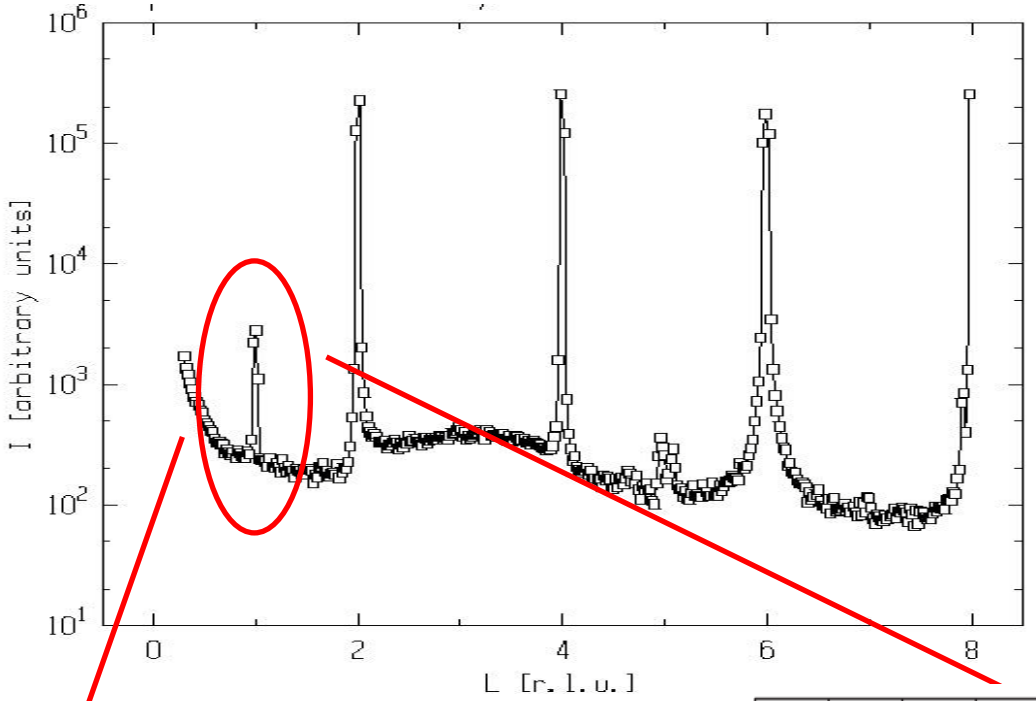
Critical angle changes

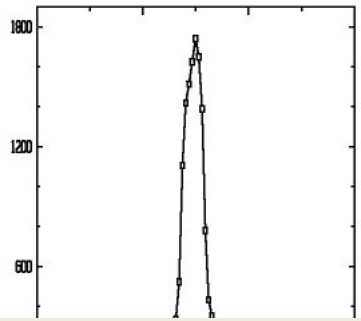


- The surface roughness <1 nm.
- No measurable dissolution or roughening under static scCO$_2$-H$_2$O

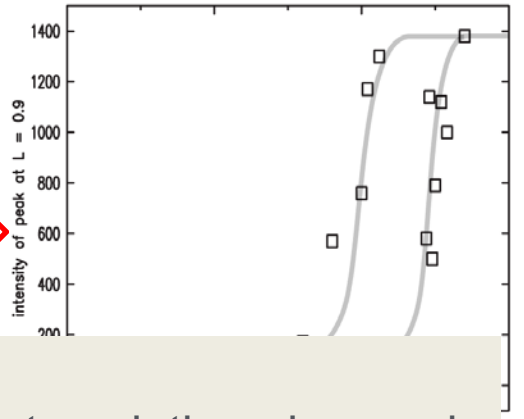
Intercalation of Water Into Muscovite Under $scCO_2$





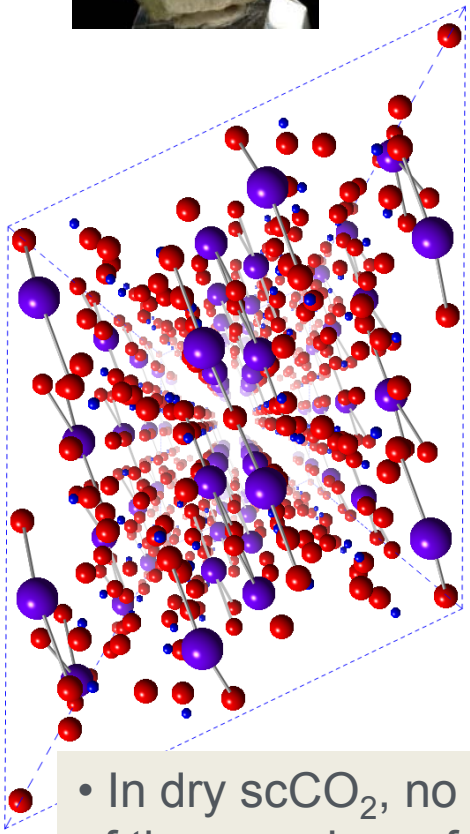


1022 psi, 140° C
$scCO_2 + H_2O$

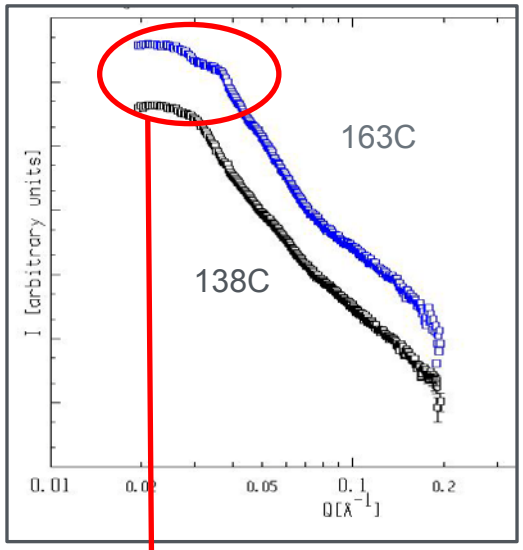


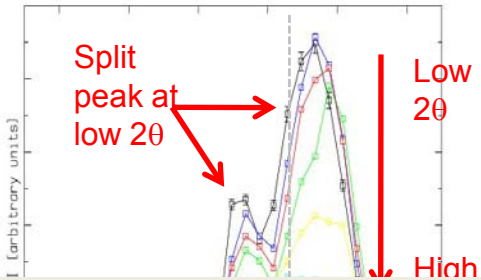
- In dry $scCO_2$, no intercalation.
- In $scCO_2$-H_2O, intercalation/de-intercalation observed.

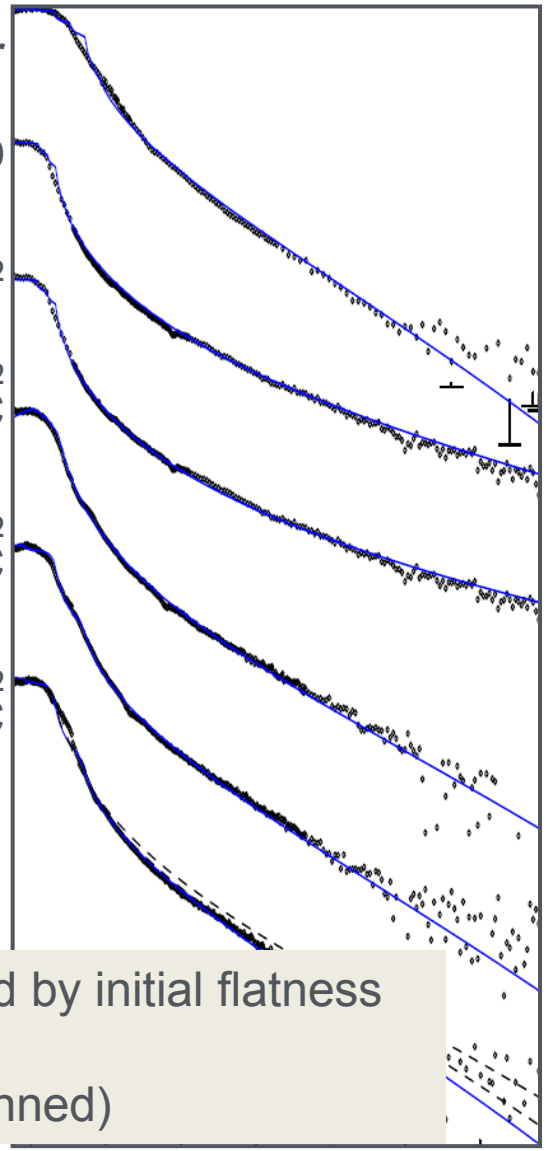
Dissolution of Orthoclase Surface



$KAlSi_3O_8$







- In dry $scCO_2$, no measurable dissolution (accuracy limited by initial flatness of the sample surface)
- In $scCO_2$-H_2O, rapid dissolution was observed. (AFM planned)

Project Management/Coordination Personal/Budget $650K/yr for 2 yrs

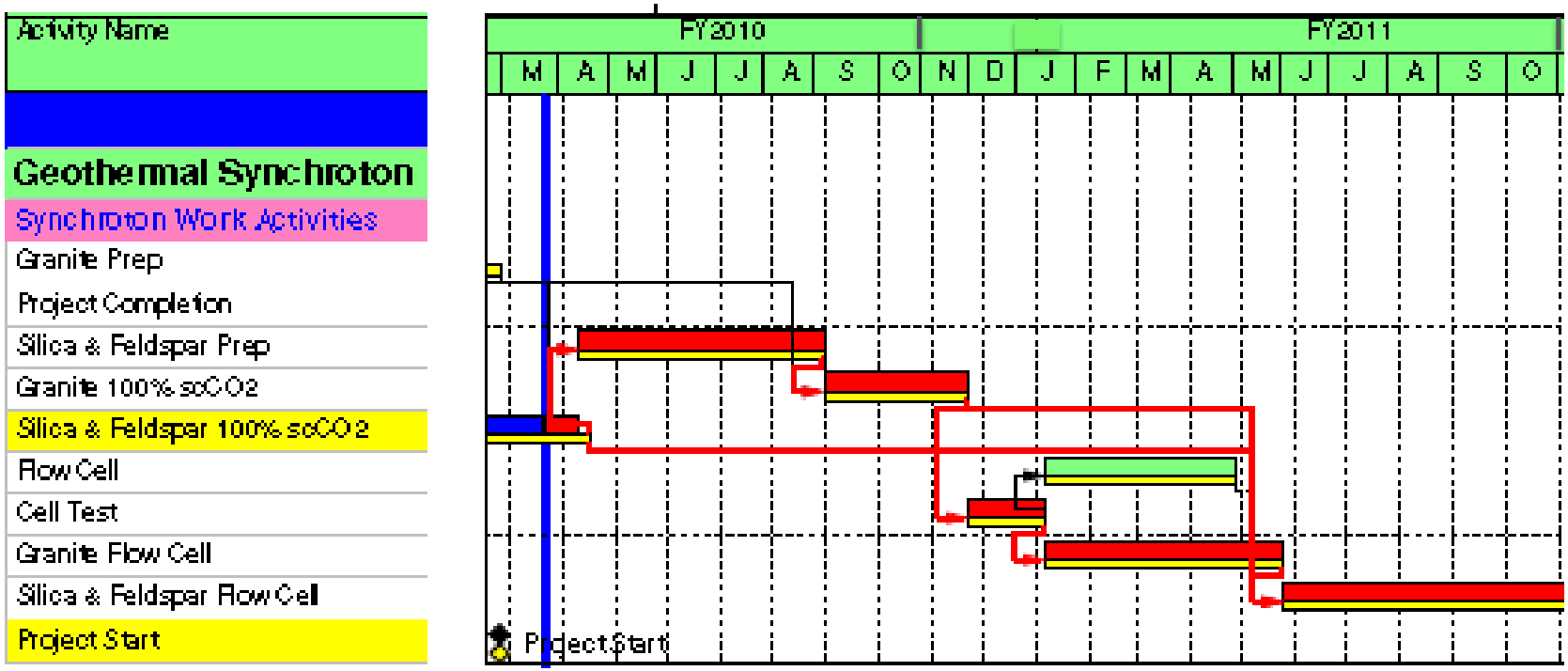
- ***Staff Effort*** ($330K Argonne National Laboratory, MSD)
  - ***Hoydoo You (25%)***
  - ***Kee-Chul Chang (10%)***
  - ***Daniel Hennessy (50%)***
  - ***Michael Pierce (15%)***
- ***Postdoc (to be hired) 100%*** ($100K ANL)
- ***Subcontract to (***$160K Arizona State University)
  - ***Hamdallah Béarat*** School of Mechanical, Aerospace, Chemical and Materials Engineering
- ***M&S and HB's trips to APS, ANL*** ($60K)

Project Management/Coordination

- *ANL MSD*
  - *In-house preparation of samples*
  - *In-house X-ray / AFM characterization*
- *ANL APS: Beamtime 2 or 3 weeks a year*
  - *Typically 1 week at a time*
  - *Beamline 12BM-B or 11ID-D*
  - *Completed two beam trips by April*
  - *One more trip is scheduled during Summer*
- *Arizona State University*
  - *Cell preparation and modification for beam trips*
  - *Develop a flow cell*
  - *HB comes to APS ANL for the beam trips*

Future Directions
Expected Outcomes and Progress

- One more synchrotron beamtime with the static cell
- Flow cell will be constructed
- Granite components will be examined in a similar manner as done in the static cell
- Ex situ AFM measurement are planned to complement X-ray work
- In situ search for calcite, magnesite, etc



Summary

- Dry $scCO_2$ will be an ideal circulating fluid based on our measurements on quartz and orthoclase
- Water, phase separated and carbonated, if present, accelerates orthoclase dissolution
- Intercalation/deintercalation into mica (muscovite)
- Calcite, magnesite, etc, may form but still need more work to measure them in situ
- Some ex situ AFM / SEM measurements are planned
- Flow cell construction will begin on schedule