Lenfest Center for Sustainable Energy
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COLUMBIA UNIVERSITY
IN THE CITY OF NEW YORK

Geo-Chemo-Mechanical Studies for Permanent Storage of CO_2 in Geologic Formations
DE-FE0002386

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U.S. Department of Energy

National Energy Technology Laboratory
Carbon Storage R&D Project Review Meeting
Developing the Technologies and Building the Infrastructure for CO_2 Storage

August 21-23, 2012

Park Group

Presentation Outline

- **Benefit and Overview**

- **Results and Accomplishments**
  - Mineral Characterization
  - Effect of Temperature, Pressure and Chemical Additives on Mineral Carbonation
  - Changes in Pore Structure and Morphology due to Carbonation
  - Effect of Particle Size and Grinding on Mineral Carbonation
  - Mineral Dissolution Studies
  - Comparison of Mineral Morphology due to Mineral Dissolution and Carbonation
  - Reactive Cracking

- **Summary**

Benefit of the Program

- Identify the Program goals being addressed.

  Develop technologies to demonstrate that 99 percent of injected CO_2 remains in the injection zones surface area.

- Project Benefits

  The project is to identify the effect of in-situ carbonation on the stability of geologic formations injected with CO_2. The technology, when successfully demonstrated, will provide valuable information on the stability of the CO_2 geological storage. This technology contributes to the Carbon Storage Program's effort of ensuring 99 percent CO_2 storage permanence in the injection zone(s).

Project Overview: Goals and Objectives

(i) Determine and compare the effect of temperature, partial pressure of CO_2 and chemical additives on carbonation of various minerals such as olivine, anorthite and basalt

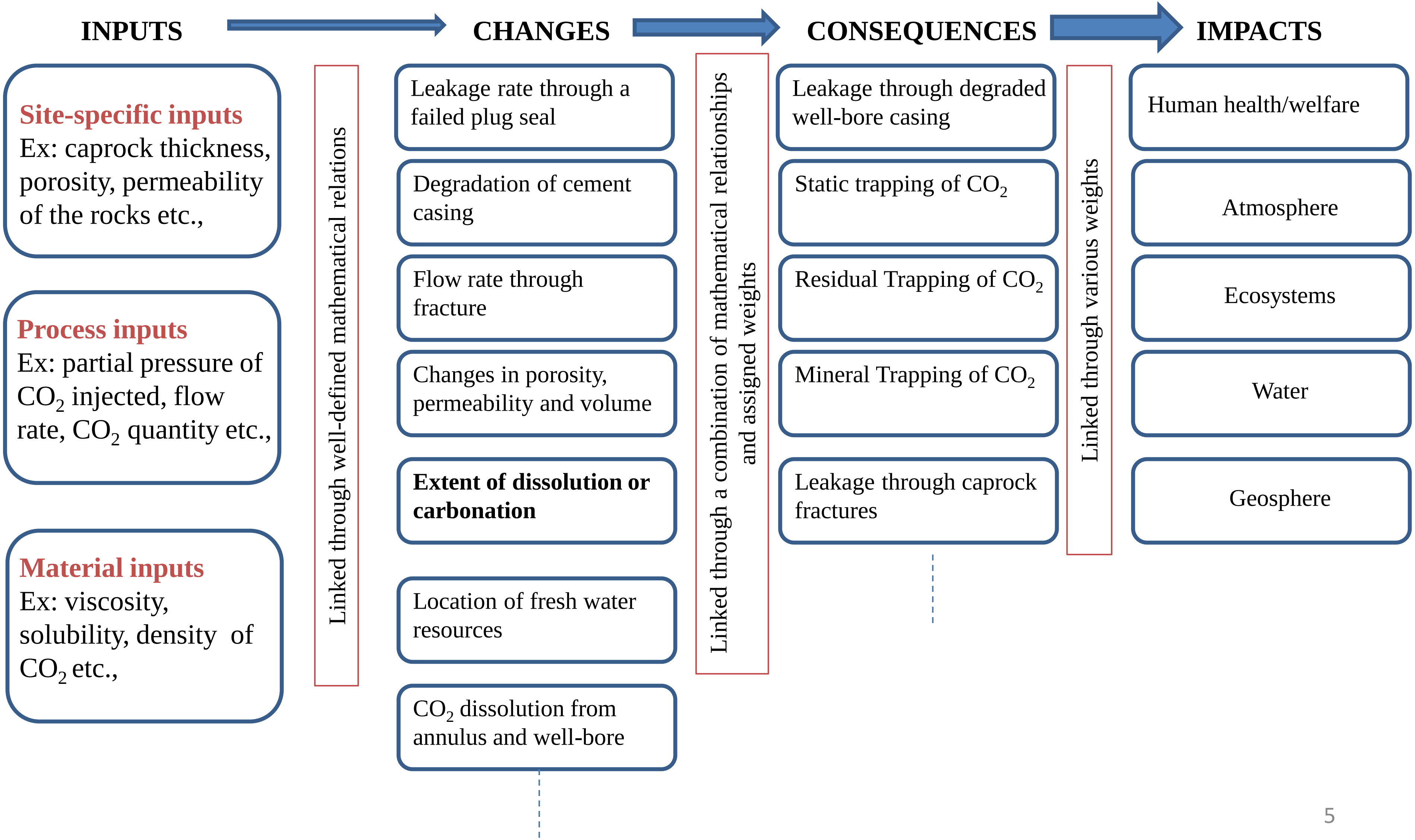
(ii) Quantify changes in pore structure and particle size before and after carbonation and analyze changes in morphological structure of the mineral due to carbonation

(iii) Determine fast and slow kinetics of mineral dissolution

(iv) Compare and contrast changes in mineral morphology due to carbonation and dissolution

(v) Determine the effect of pore fluid chemistry on mechanical behavior of rocks such as changes in hydrostatic compaction and strain on thermally cracked dunite saturated with CO_2-saturated brines

Risk Framework for Geological Storage of CO_2



INPUTS → **CHANGES** → **CONSEQUENCES** → **IMPACTS**

INPUTS

Site-specific inputs
Ex: caprock thickness, porosity, permeability of the rocks etc.,

Process inputs
Ex: partial pressure of CO_2 injected, flow rate, CO_2 quantity etc.,

Material inputs
Ex: viscosity, solubility, density of CO_2 etc.,

Linked through well-defined mathematical relations

CHANGES

- Leakage rate through a failed plug seal
- Degradation of cement casing
- Flow rate through fracture
- Changes in porosity, permeability and volume
- **Extent of dissolution or carbonation**
- Location of fresh water resources
- CO_2 dissolution from annulus and well-bore

Linked through a combination of mathematical relationships and assigned weights

CONSEQUENCES

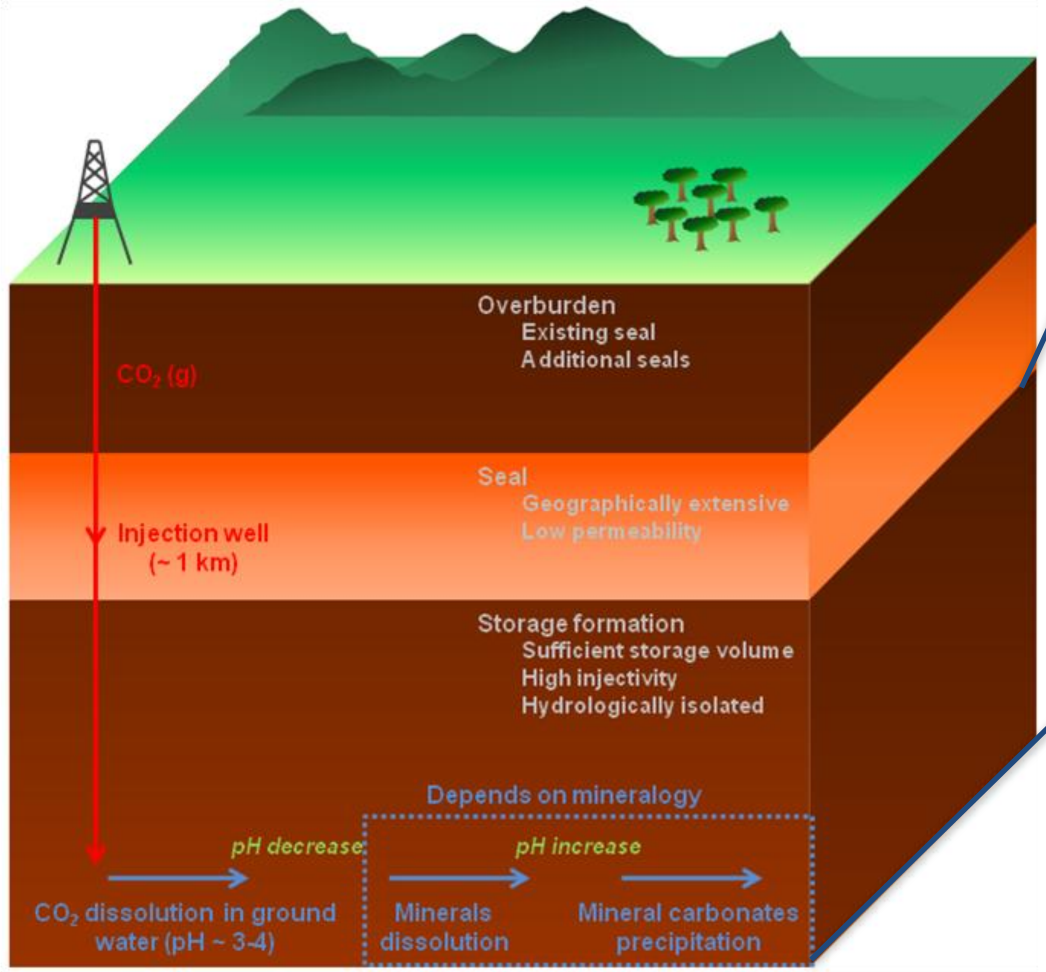
- Leakage through degraded well-bore casing
- Static trapping of CO_2
- Residual Trapping of CO_2
- Mineral Trapping of CO_2
- Leakage through caprock fractures

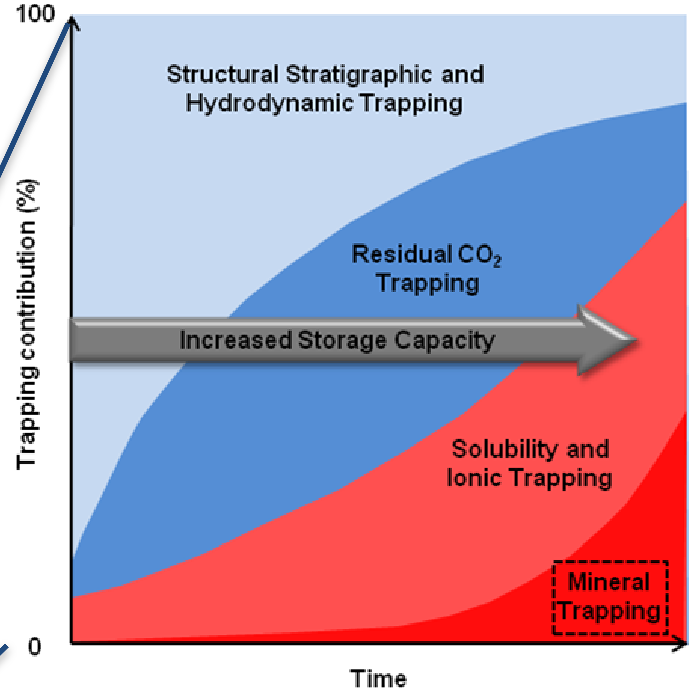
Linked through various weights

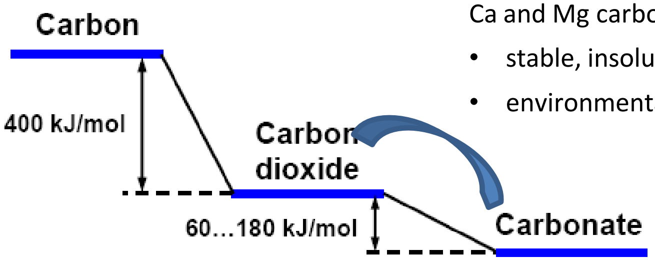
IMPACTS

- Human health/welfare
- Atmosphere
- Ecosystems
- Water
- Geosphere

Carbon Storage in Geologic Formations



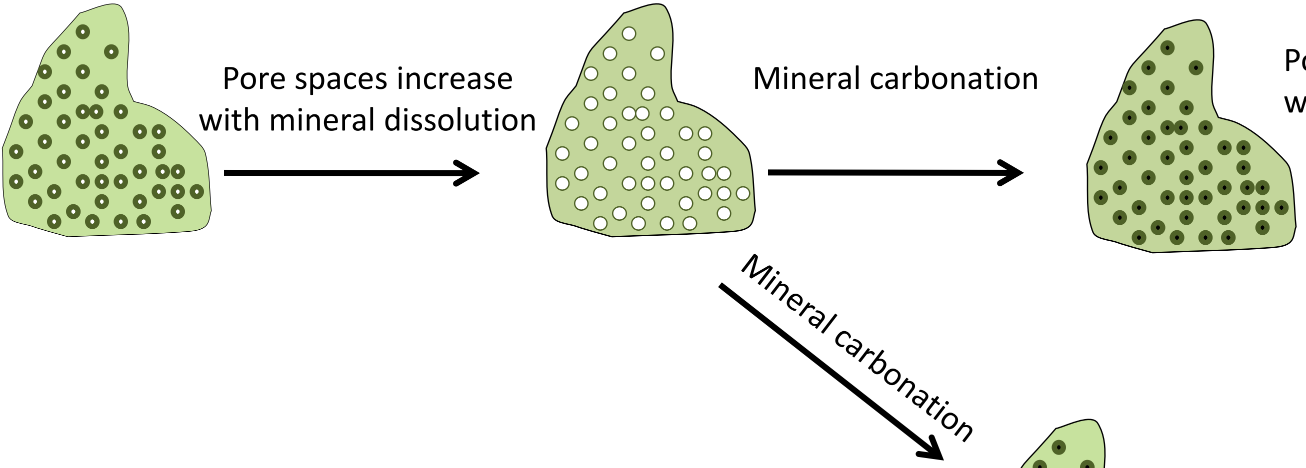




Ca and Mg carbonates are

- stable, insoluble in water
- environmentally benign

Mineral Carbonation and Reactive Cracking



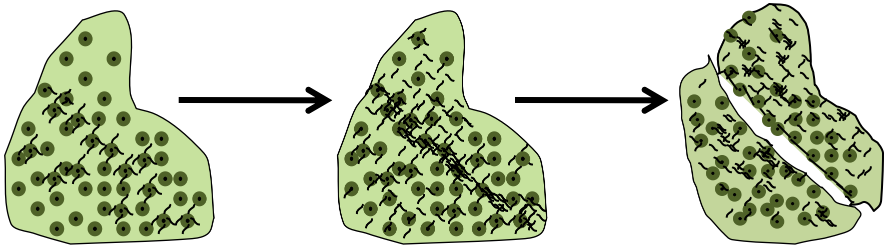
Pore spaces increase with mineral dissolution

Mineral carbonation

Porosity decreases with carbonation

Safe for CO_2 storage as mechanical strength increases

Mineral carbonation



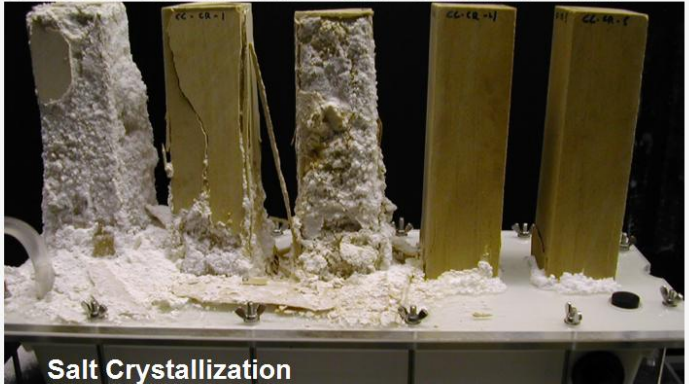
Increased porosity results in microfractures

Alignment of pores

Cracking of rocks

If overburden and caprock seal is good, then cracking is ok

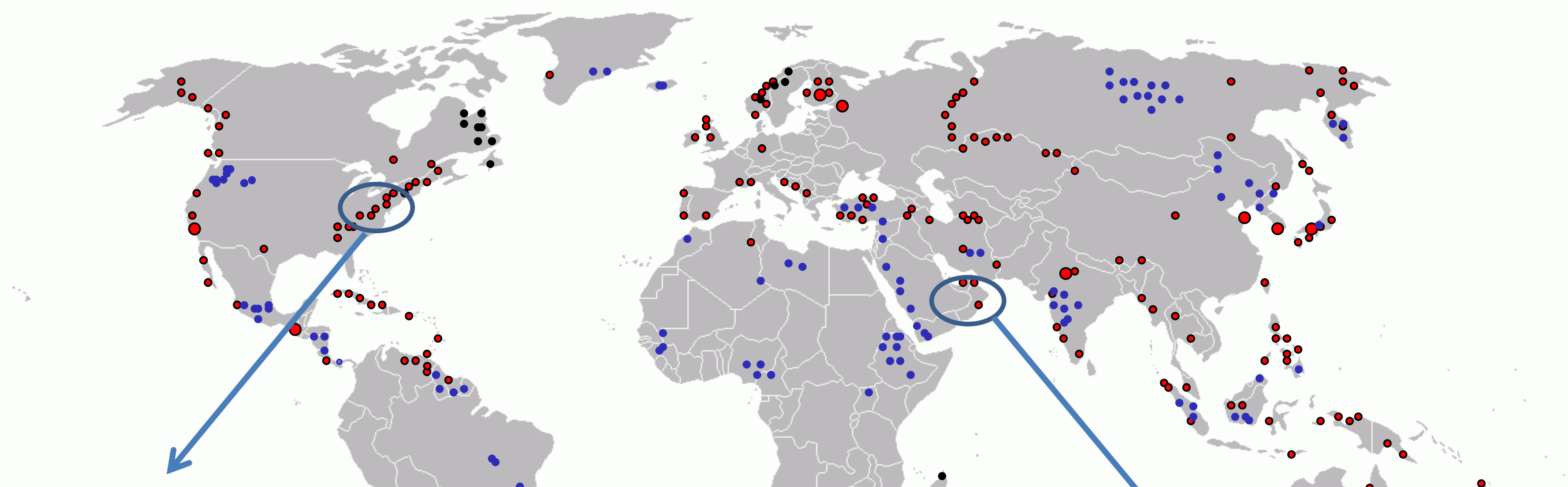
If overburden and caprock seal is not good, is there a problem?



Salt Crystallization

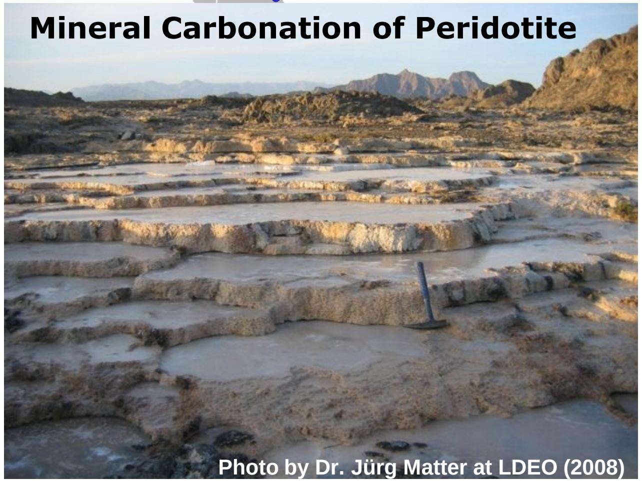
http://gwsgroup.princeton.edu/SchererGroup/Salt_Crystallization.html

Availability of Minerals



Belvidere Mountain, Vermont
Serpentine Tailings

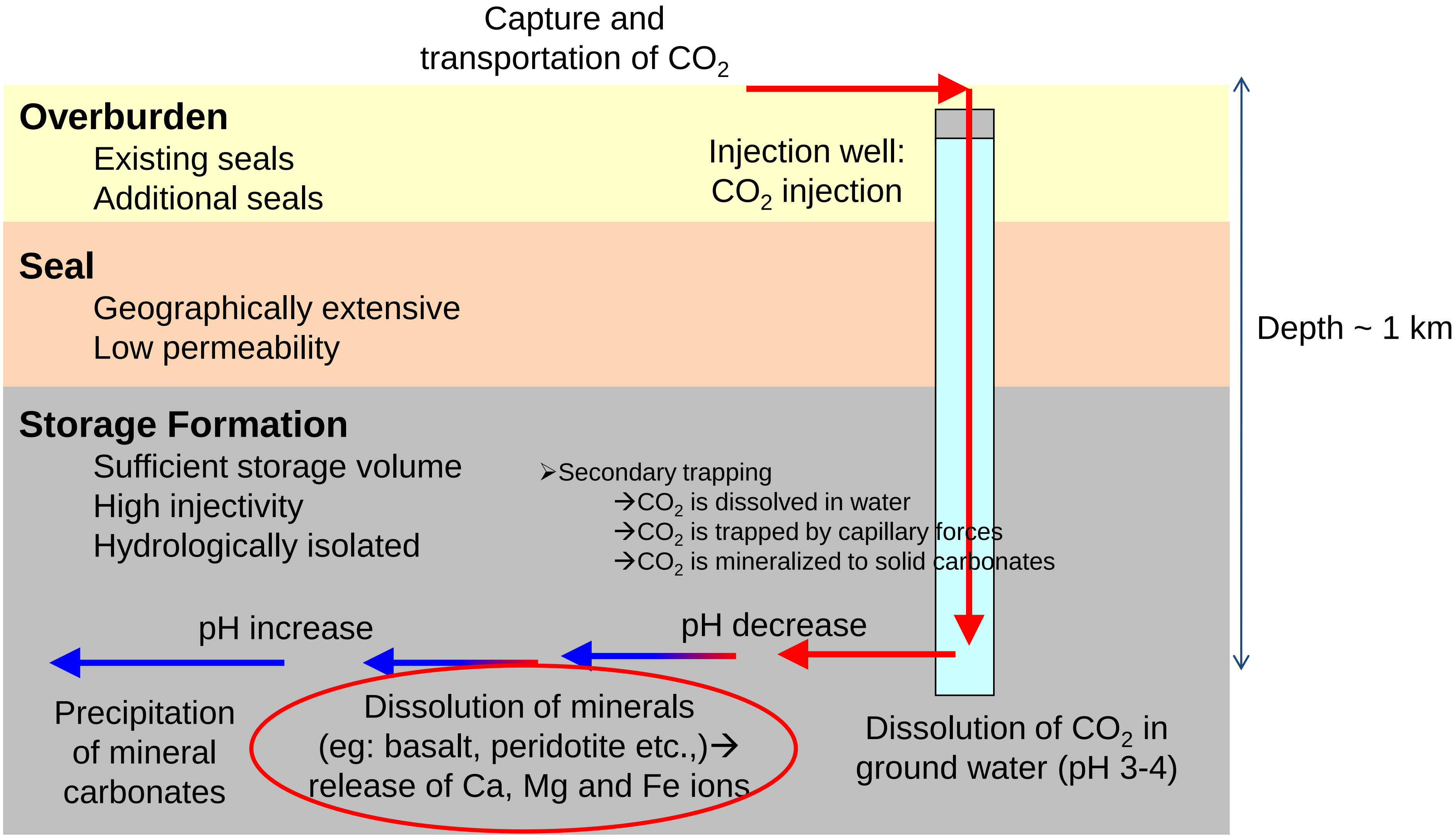
- ● Basalt
- ● Labradorite
- ● Magnesium-based Ultramafic Rocks (Serpentine, Olivine)



Mineral Carbonation of Peridotite

Photo by Dr. Jürg Matter at LDEO (2008)

CO_2 Storage via In-situ Mineralization



Capture and transportation of CO_2

Overburden

Existing seals

Additional seals

Injection well: CO_2 injection

Seal

Geographically extensive

Low permeability

Depth ~ 1 km

Storage Formation

Sufficient storage volume

High injectivity

Hydrologically isolated

- Secondary trapping
  - →CO_2 is dissolved in water
  - →CO_2 is trapped by capillary forces
  - →CO_2 is mineralized to solid carbonates

pH increase

pH decrease

Precipitation of mineral carbonates

Dissolution of minerals (eg: basalt, peridotite etc.,)→ release of Ca, Mg and Fe ions

Dissolution of CO_2 in ground water (pH 3-4)

Minerals of Interest

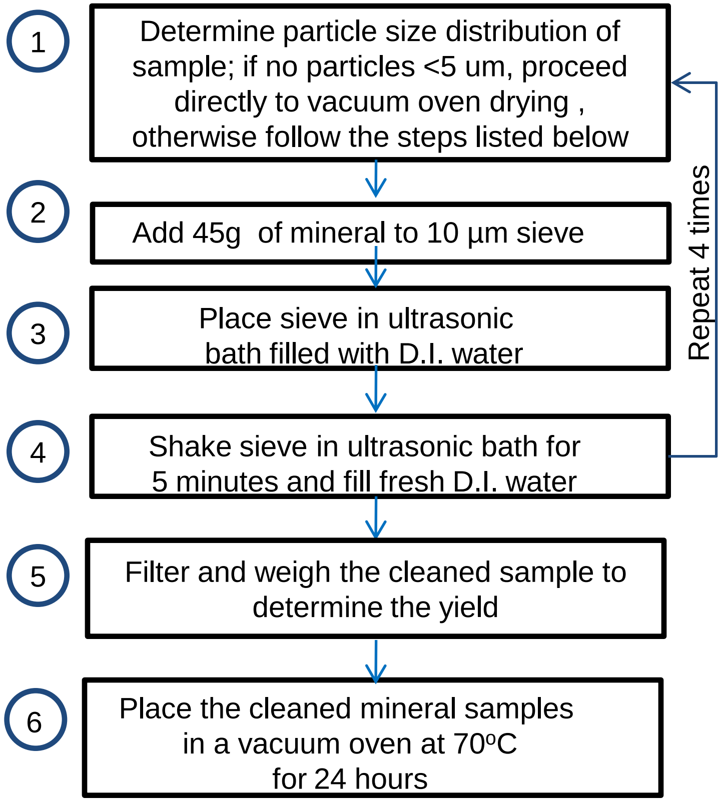
Mineral	MgO	CaO	Fe_2O_3	SiO_2	Al_2O_3	Na_2O	K_2O	TiO_2	MnO	Cr_2O_3	V_2O_5	LOI%	Sum %	Ni %
Olivine	47.3	0.16	13.9	39.7	0.2	0.01	<0.01	<0.01	0.15	0.78	< 0.01	-0.7	101.5	0.27
Anorthite	8.74	14.1	10.6	41.8	24.2	0.59	0.03	0.04	0.13	0.08	< 0.01	0.12	100.4	0.02
Basalt	4.82	8.15	14.6	51.9	13.4	2.91	1.09	1.74	0.21	0.10	0.06	0.27	99.6	0.04

$$Olivine:\ Mg_2SiO_4 + 2CO_2 \rightarrow 2MgCO_3 + SiO_2$$

$$Anorthite:\ CaAl_2Si_2O_8 + CO_2 \rightarrow CaCO_3 + Al_2\,Si_2\,O_7$$

Basalt is a mixture of plagioclase (anorthite), pyroxene and olivine

Mineral Cleaning Protocol



① Determine particle size distribution of sample; if no particles <5 um, proceed directly to vacuum oven drying , otherwise follow the steps listed below

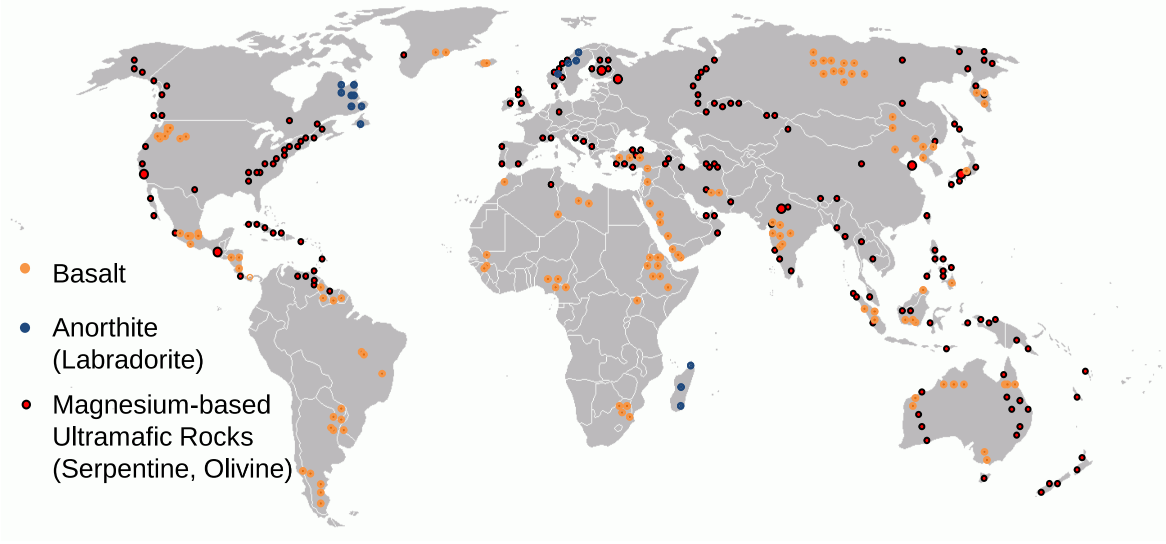
② Add 45g of mineral to 10 µm sieve

③ Place sieve in ultrasonic bath filled with D.I. water

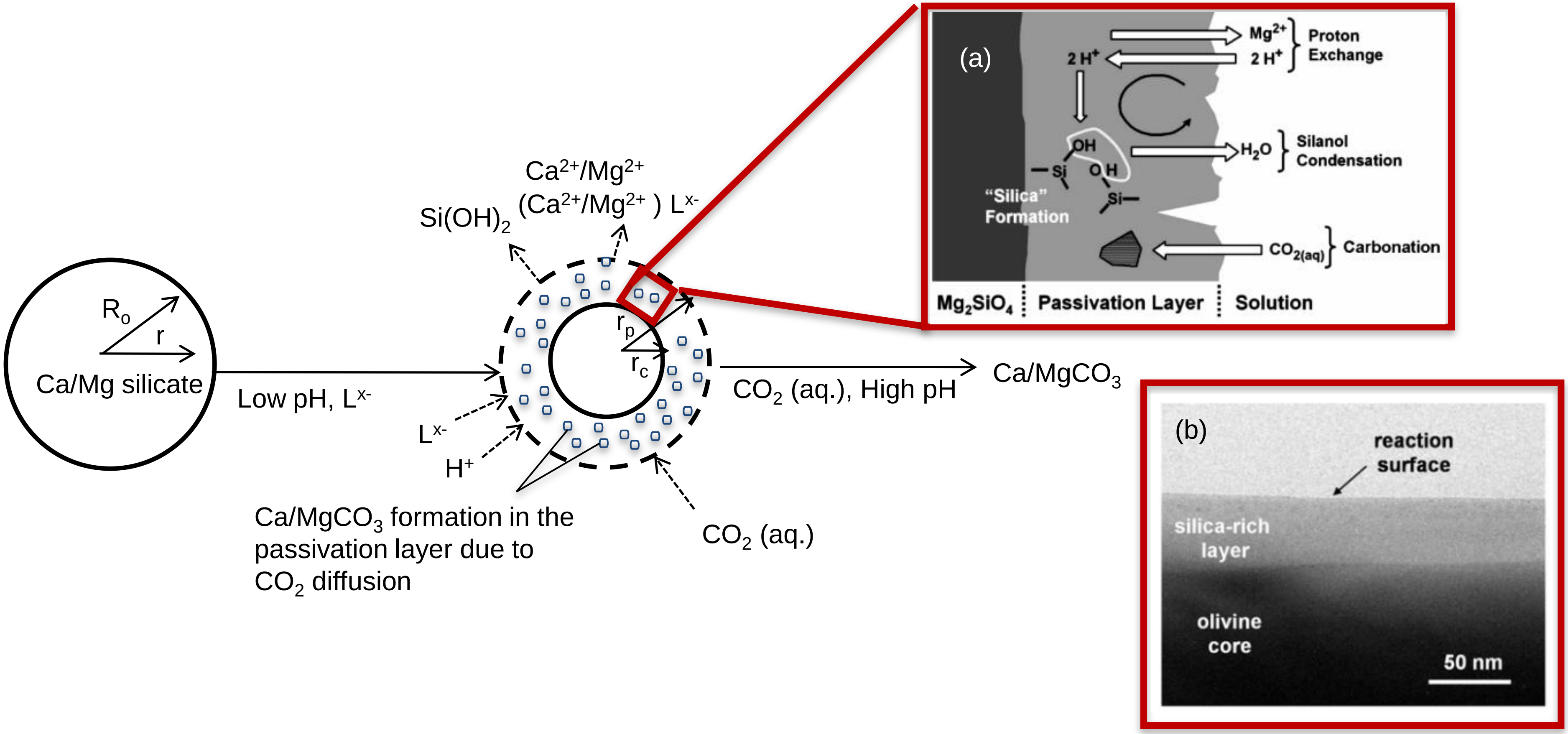
④ Shake sieve in ultrasonic bath for 5 minutes and fill fresh D.I. water

⑤ Filter and weigh the cleaned sample to determine the yield

⑥ Place the cleaned mineral samples in a vacuum oven at 70ºC for 24 hours



Reaction Mechanisms

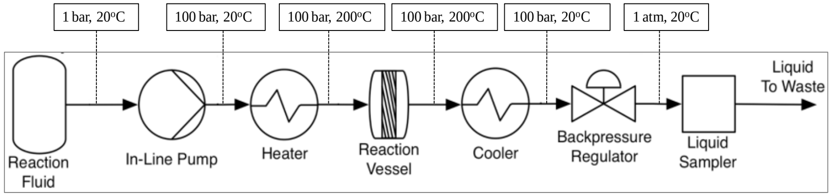


Formation of silica passivation layer

Figures (a), (b) from Bearat et al., Environ Sci Technol (2006) vol. 40 (15) pp. 4802-4808

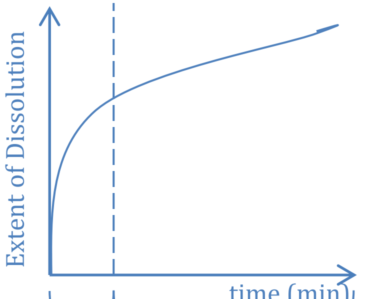
Research Scheme

Mineral Dissolution



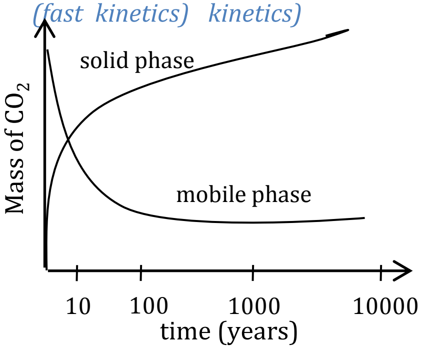
Differential Bed Reactor

Mineral Dissolution Rates and Conversion

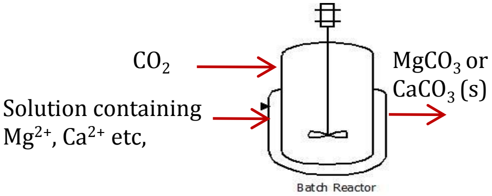


Surface rxn. limited regime (fast kinetics)

Mass transfer limited regime (long-term kinetics)

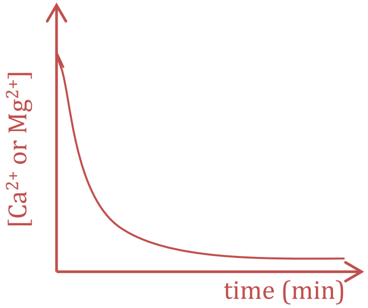


Mineral Carbonation



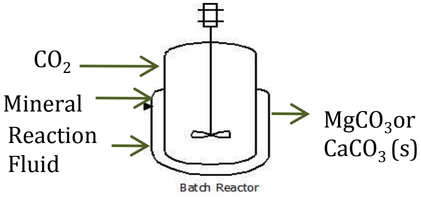
Batch Reactor

Mineral Carbonation Rates and Conversion



Effects of Chemical and Physical Properties (e.g. chemical composition, pore size distribution, surface area, etc.,)

Coupled Reaction Studies



Batch Reactor

Effect of Coupled Reactions on Physical and Chemical Properties

Coupled Geo-Chemo-Physical Simulations such as ToughReact to determine CO$_2$ plume distribution

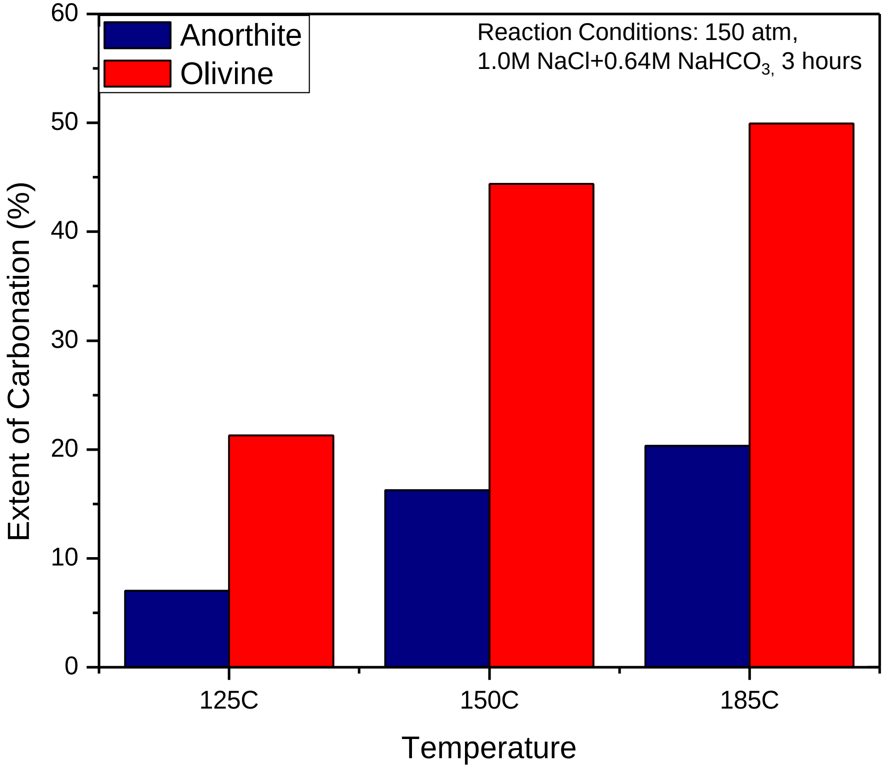
HAZOP Analysis of Geological Storage of CO$_2$

Impacts

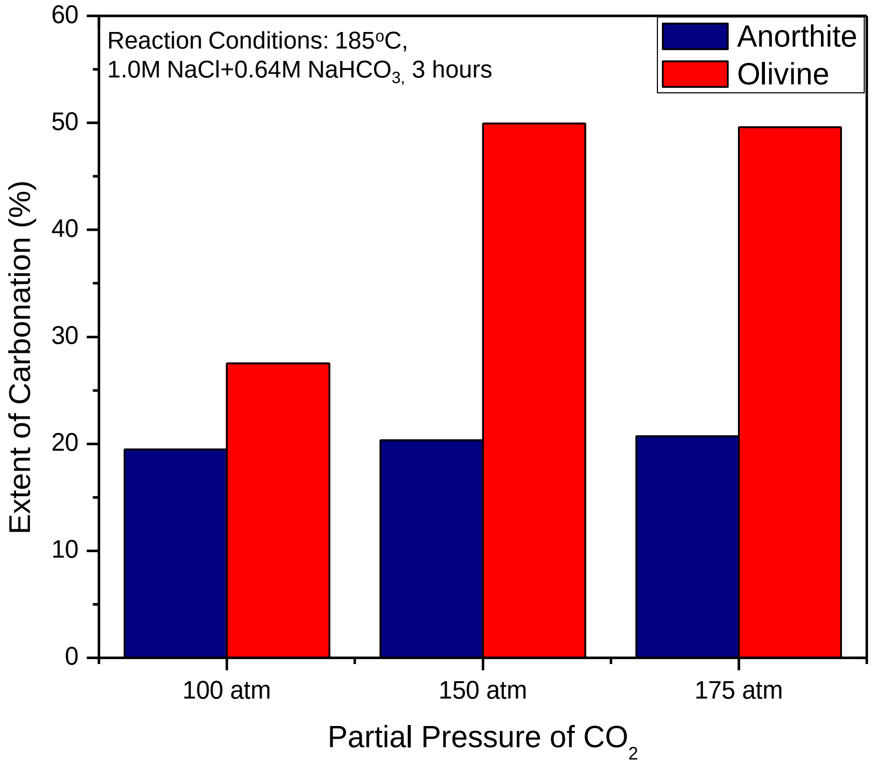
Human Welfare Water Ecosystems Atmosphere Geosphere

Effect of Temperature and Pressure on Mineral Carbonation

(a)

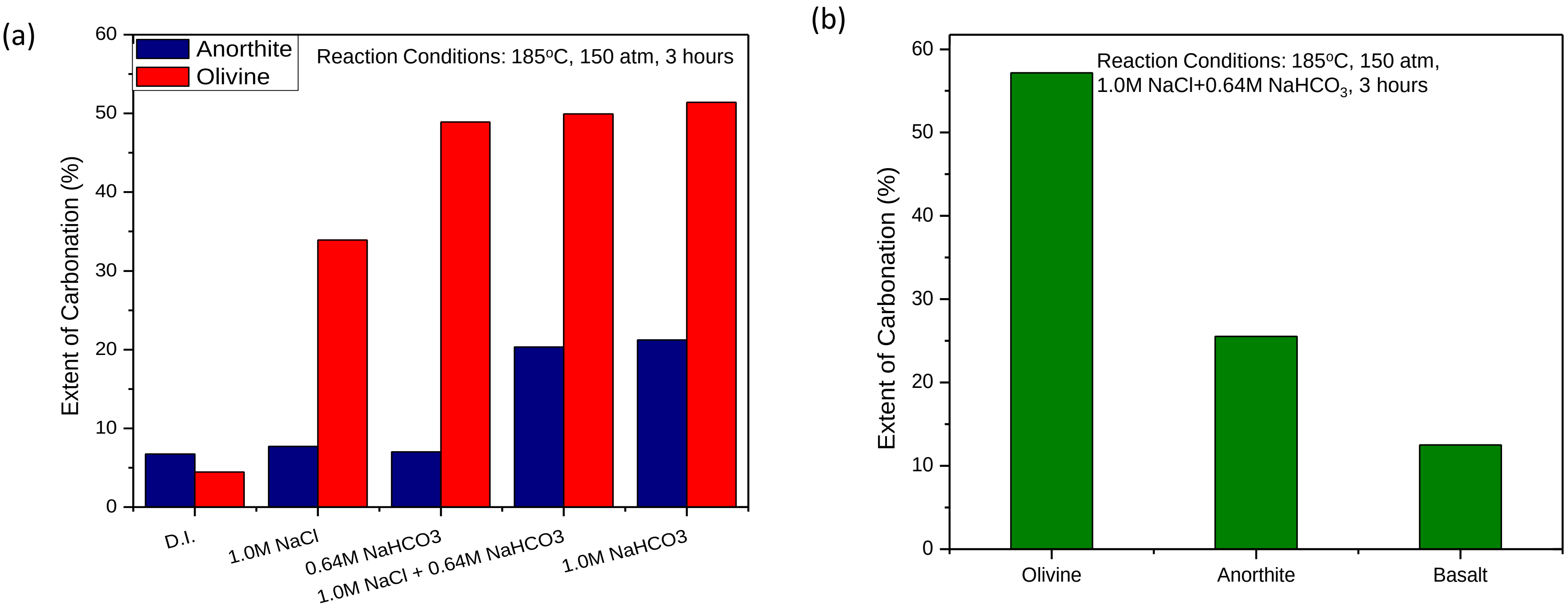


(b)



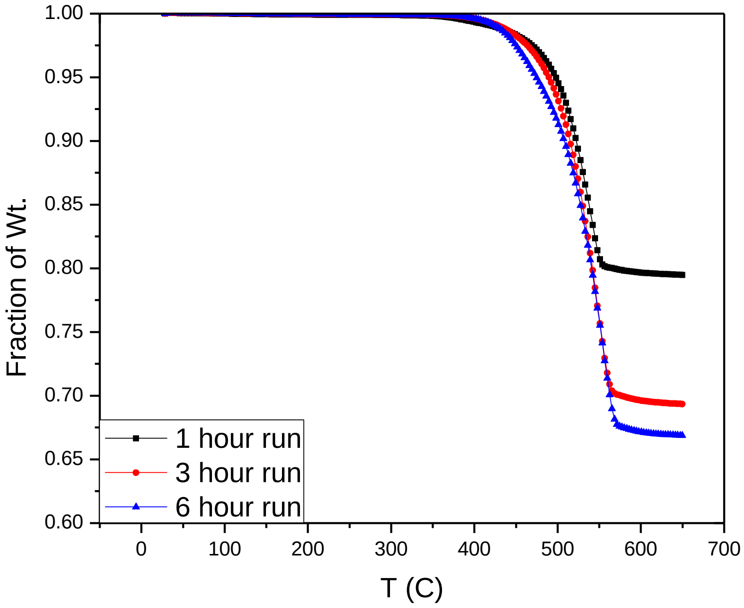
- Increasing temperature results in higher extents of carbonation in olivine and anorthite
- Increasing partial pressure of CO_2 from 100 atm to 150 atm results in much higher carbonation of olivine
- Anorthite carbonation does not vary significantly with increasing pressure

Effect of Chemical Additives and Comparison of Carbonation in Minerals

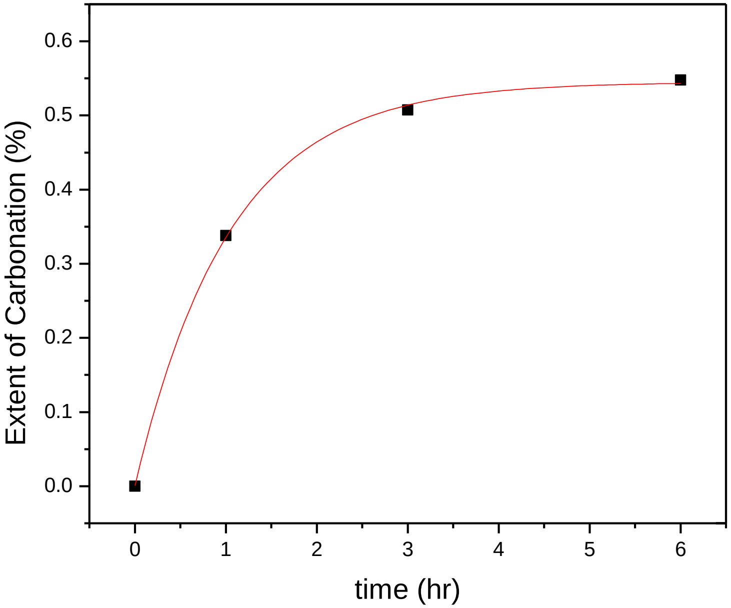


- Addition of additives such as NaCl and $NaHCO_3$ increases extent of anorthite and olivine carbonation
- Addition of NaHCO3 is effective in enhancing mineral carbonation
- Extent of carbonation achieved with olivine> anorthite> basalt

Effect of Reaction Time on Olivine Carbonation Rates



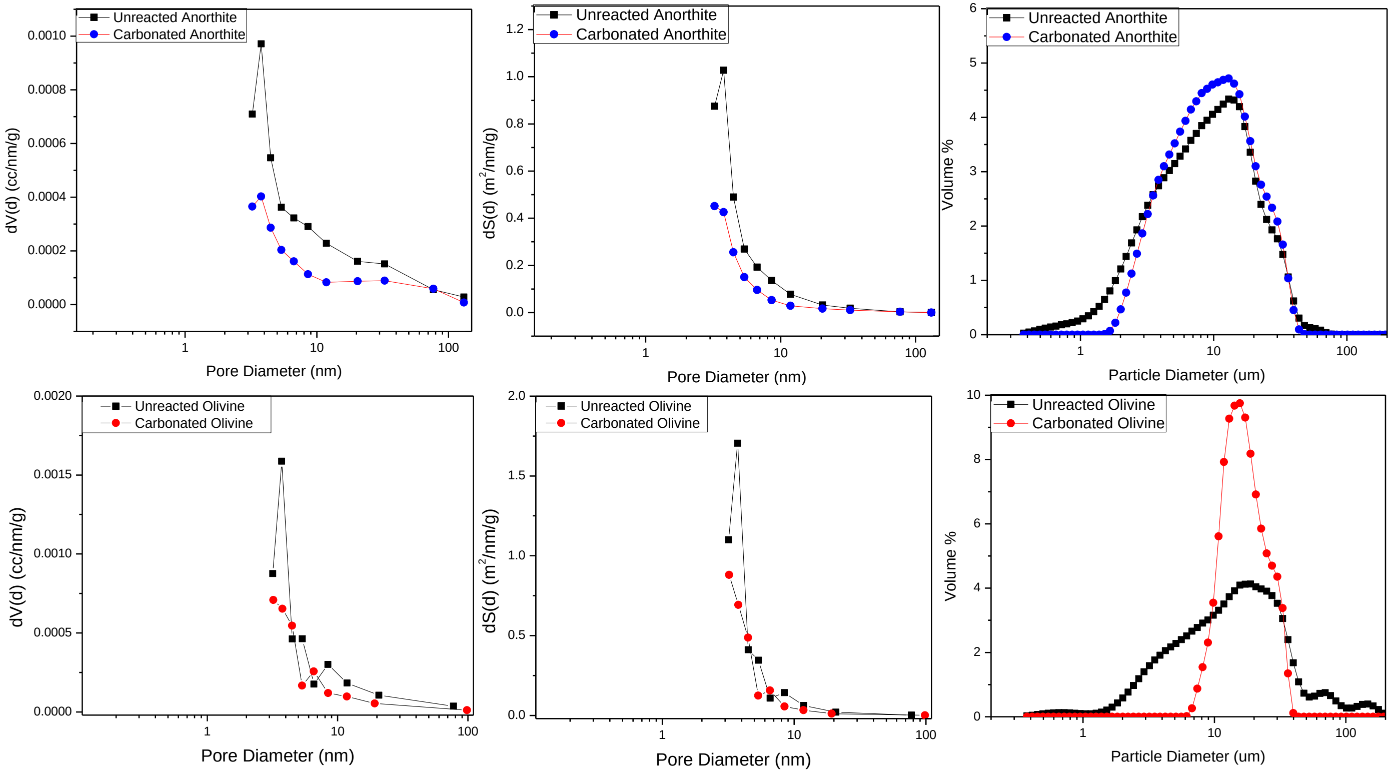
Estimation of Extent of Carbonation using TGA



Effect of Reaction Time on Olivine Carbonation

Experimental Conditions: Olivine, 185°C, P_{CO2} = 150 atm, 15 wt.% mineral, 1.0M NaCl + 0.64M $NaHCO_3$ in high pressure, high temperature batch reactor

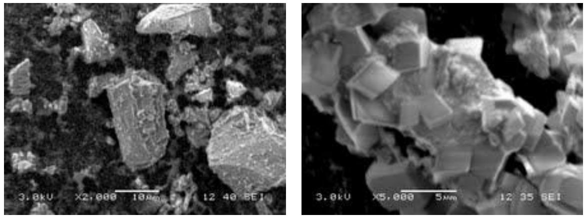
Changes in Mineral Morphology Due to Carbonation



Carbonated mineral is obtained after the mineral is reacted with 150 atm of CO_2 at 185ºC in 1.0M NaCl+0.64M $NaHCO_3$, 15 wt. % solid, 800 rpm for 3 hours

- Significant reduction in the pore volume, pore surface area after carbonation
- Increase in the mean particle diameter and narrower particle size distribution after carbonation due to carbonate crystal growth
- Surface area decreases up to 54% and 40% after anorthite and olivine carbonation respectively
- Significant decreases in porosity and surface area have implications on long-term in-situ storage of CO_2 in mineral formations

	Unreacted Anorthite	Carbonated Anorthite	Unreacted Olivine	Carbonated Olivine
Mean Particle Diameter ± Std. Dev.	11.94 ± 9.52	12.42 ± 8.52	21.41 ± 25.46	27.71 ± 11.64
Surface Area (m^2/g)	2.95	1.60	3.89	1.55
Pore Volume (cc/g)	0.013	0.008	0.009	0.005



Unreacted vs. Carbonated Olivine

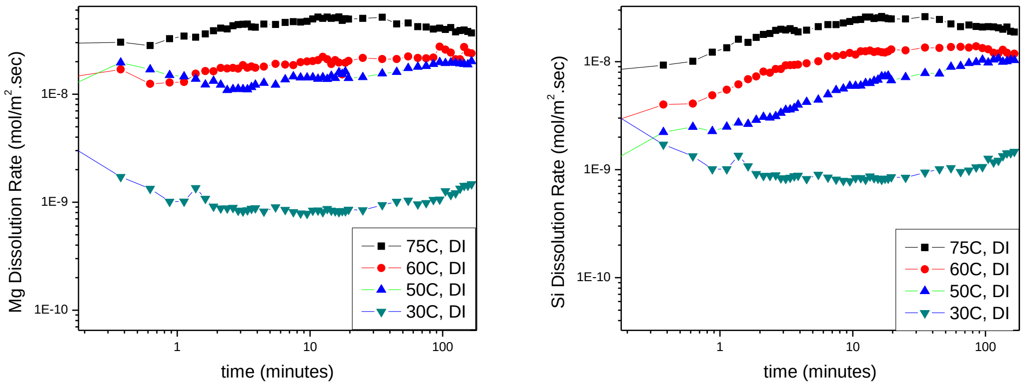
Effect of Grain Size and Grinding on Olivine Carbonation

Olivine Sample Type	**Slurry of freshly ground old olivine sample (2012)**	**Freshly ground old olivine sample (2012)**	**Slurry of old olivine sample from 2010**	**Slurry of old olivine sample from 2010**	**Cleaned old olivine sample from 2010**
Rxn Conditions	1.0M NaCl+0.64M NaHCO3, 185C, 150 atm, 800 rpm, 3 hours	1.0M NaCl+0.64M NaHCO3, 185C, 150 atm, 800 rpm, 3 hours	1.0M NaCl+0.64M $NaHCO_3$, 185C, 150 atm, 800 rpm, 3 hours	DI, 185C, 150 atm, 800 rpm, 3 hours	1.0M NaCl+0.64M $NaHCO_3$, 185C, 150 atm, 800 rpm, 3 hours
Particle Size Distribution	<20 µm	0.3-92 µm (95% < 37 µm)	<20 µm (for the sieved sample)	<20 µm (for the sieved sample)	10 – 90 µm
Extent of Carbonation	50.17%	47.09%	13.2%	1-2%	1-2%

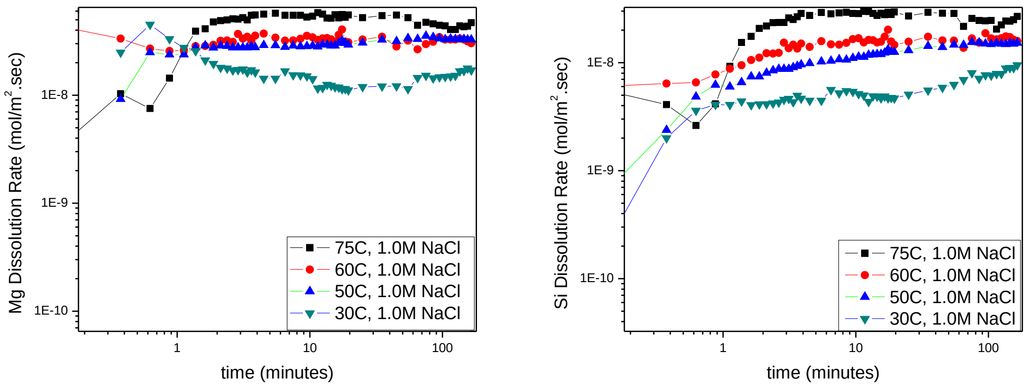
- Fresh grinding of olivine results in a much higher extent of carbonation compared to old samples
- Particle size governs extent of carbonation - Smaller the particle size, greater the extent of carbonation

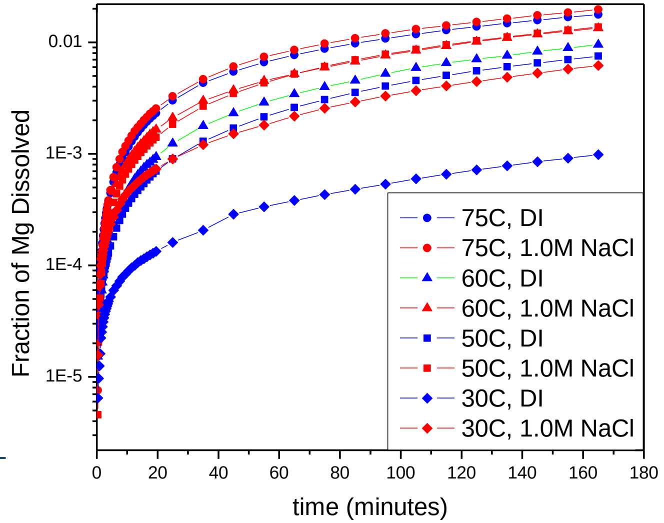
Olivine Dissolution Kinetics – Fast vs. Slow Kinetics

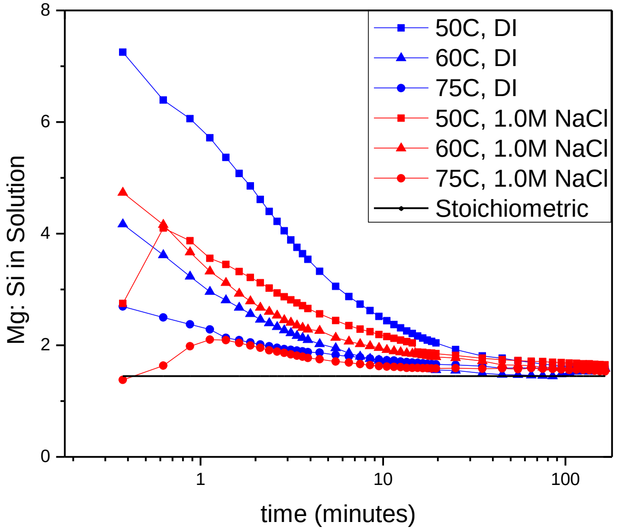
(a) DI



(b) 1.0M NaCl



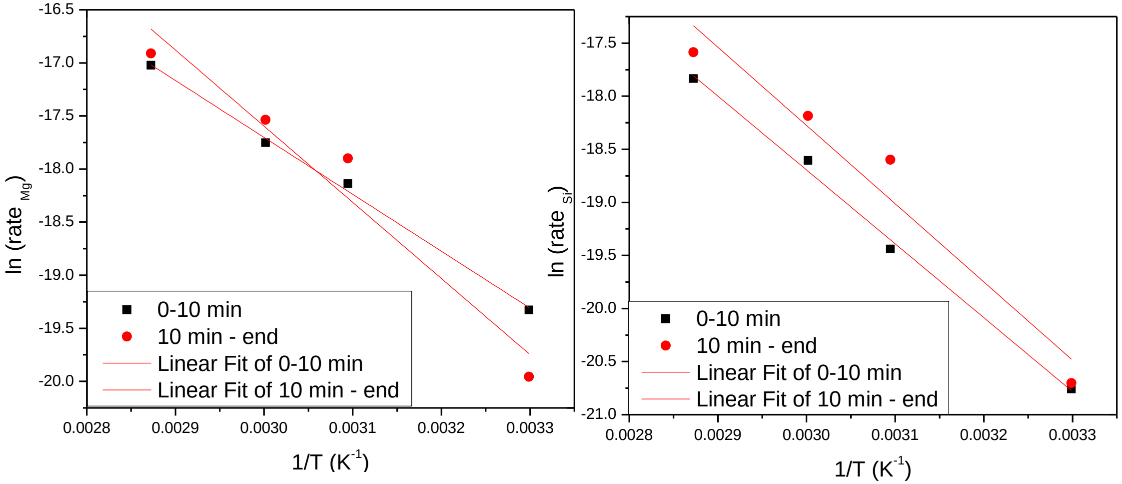




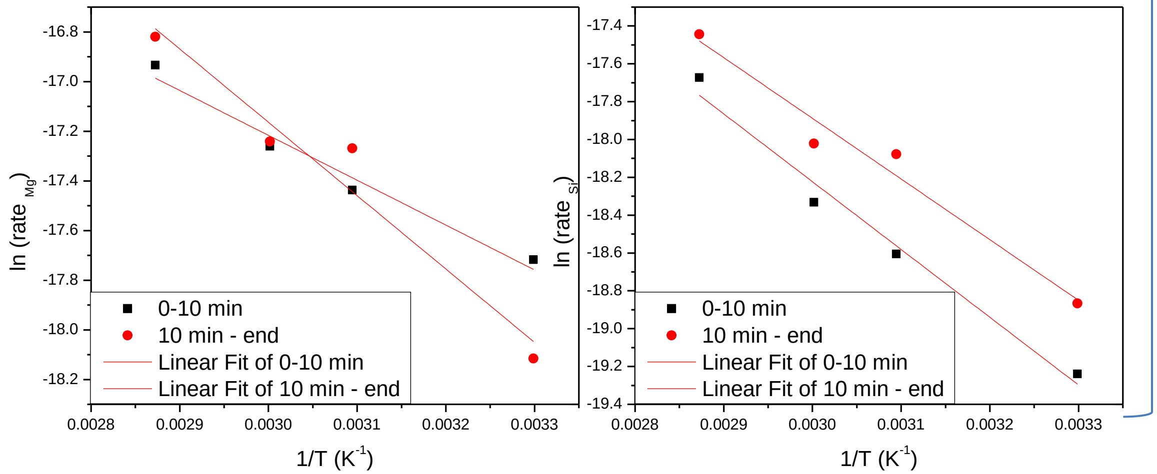
- Much higher initial extent of dissolution
- Initial incongruent dissolution with high Mg: Si ratio
- Increasing temperature results in higher dissolution
- Effect of NaCl on enhancing olivine dissolution is more significant at lower temperatures

Extent of Olivine Dissolution and Activation Energy Determination

(a) DI



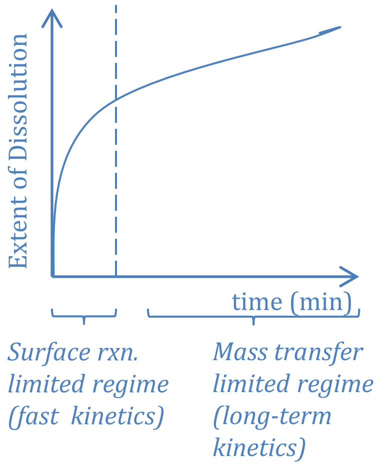
(b) 1.0M NaCl



	E_a- DI (kJ/mol)	E_a- 1.0M NaCl (kJ/mol)	ΔE_a (kJ/mol)
Mg: 0-10 min	44.6	15.0	29.6
Mg: 10-180 min	59.7	24.6	35.1
Si: 0-10 min	57.9	29.8	28.1
Si: 10-180 min	61.3	26.7	34.6

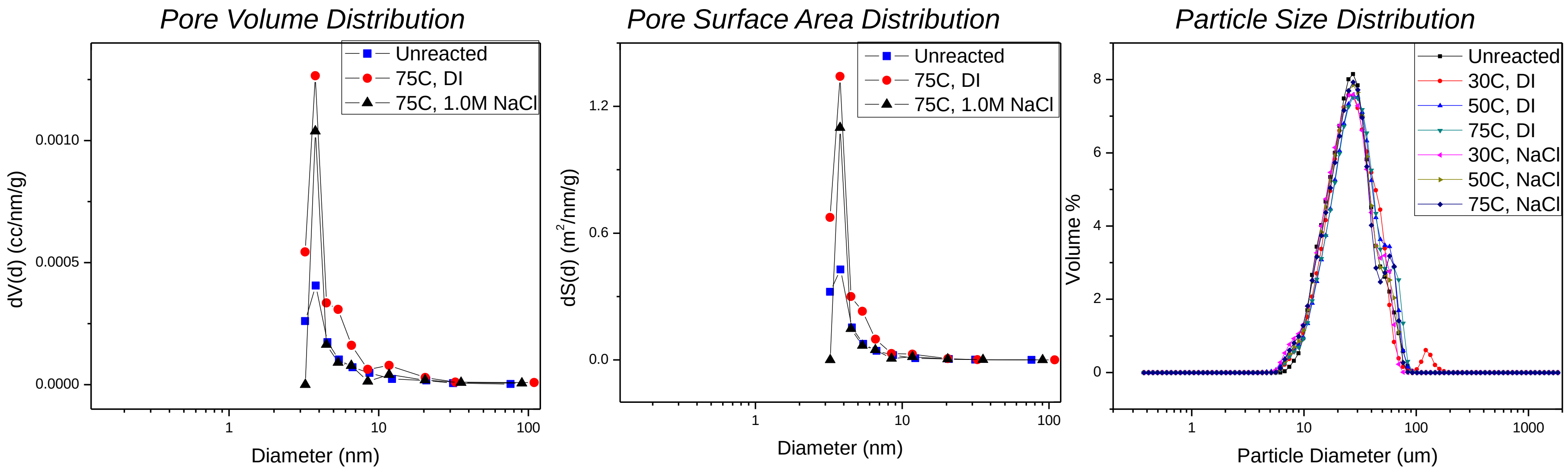
Activation energy for the first 10 minutes of dissolution is considerably lower than the activation energy from 10-180 minutes.

NaCl has a significant effect on reducing the activation energy of olivine dissolution

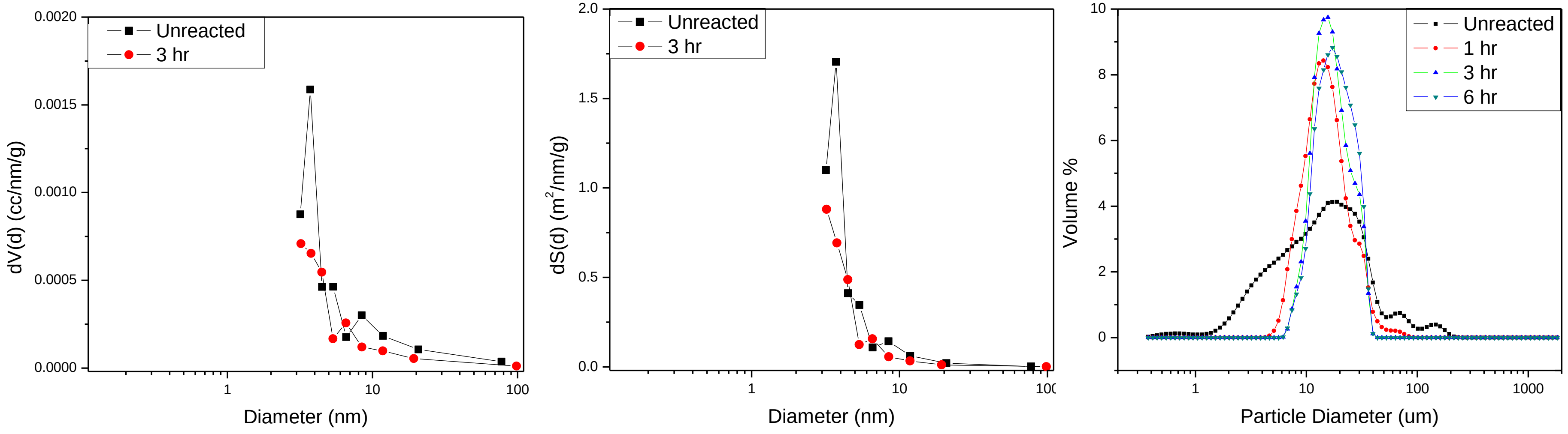


Changes in Pore and Particle Size after Mineral Dissolution vs. Carbonation

(a) Mineral Dissolution

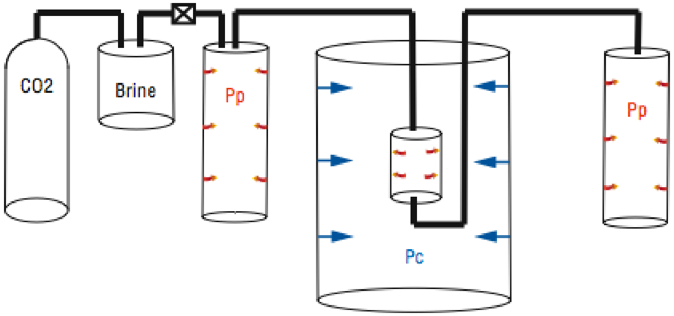


(b) Mineral Carbonation



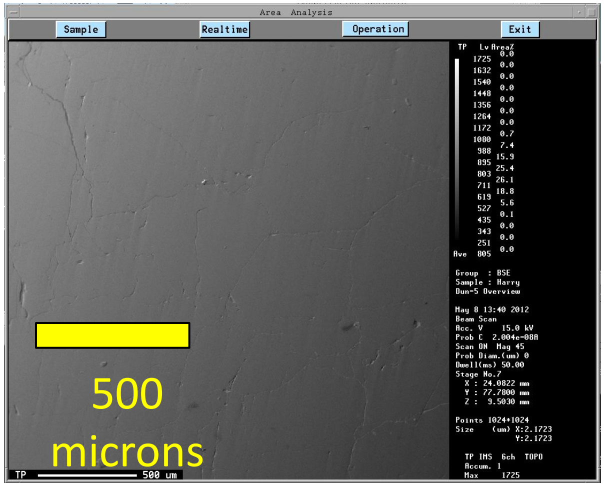
Reactive Cracking

Objective: Assess the effect of high CO_2 fluids on the behavior of ultramafic rocks such as <u>hydrostatic compaction, constant strain rate and constant displacement</u> creep experiments on thermally cracked dunite saturated with CO_2-saturated brines



- Autolab 1500 triaxial deformation apparatus from New England Research (NER)
- Retrofitted fluid mixing system
- Independent T, P_{CO2} control
- 15 MPa confining pressure
- 10 MPa pore pressure
- 150ºC Temperature
- Thermally cracked dunite with ~ 1 mm grain size

Deformation of Rocks due to Reactive Cracking



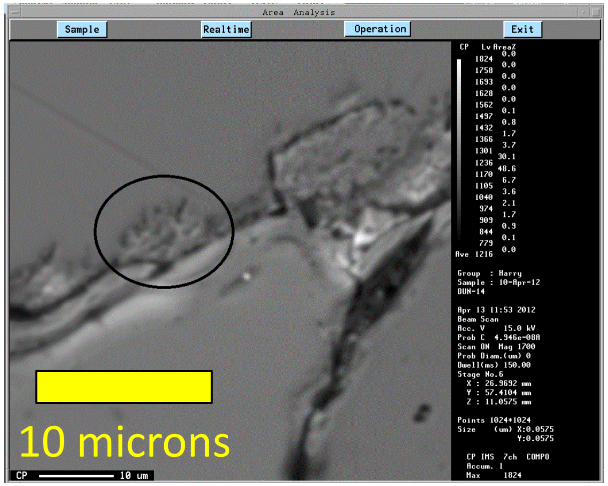
```
TP   Lv Area%
          0.0
   1725
          0.0
   1632
          0.0
   1540
          0.0
   1448
          0.0
   1356
          0.0
   1264
          0.0
   1172
          0.7
   1080
          7.4
    988
         15.9
    895
         25.4
    803
         26.1
    711
         18.8
    619
          5.6
    527
          0.1
    435
          0.0
    343
          0.0
    251
          0.0
Ave  805

Group  : BSE
Sample : Harry
Dun-5 Overview

May 8 13:40 2012
Beam Scan
Acc. V    15.0 kV
Prob C  2.004e-08A
Scan ON  Mag 45
Prob Diam.(um) 0
Dwell(ms) 50.00
Stage No.7
  X : 24.0822 mm
  Y : 77.7800 mm
  Z :  9.5030 mm

Points 1024*1024
Size    (um) X:2.1723
             Y:2.1723

  TP IMS  6ch  TOPO
  Accum. 1
  Max     1725
```



```
CP   Lv Area%
          0.0
   1824
          0.0
   1758
          0.0
   1693
          0.0
   1628
          0.0
   1562
          0.1
   1497
          0.8
   1432
          1.7
   1366
          3.7
   1301
         30.1
   1236
         48.6
   1170
          6.7
   1105
          3.6
   1040
          2.1
    974
          1.7
    909
          0.9
    844
          0.1
    779
          0.0
Ave 1216

Group  : Harry
Sample : 10-Apr-12
DUN-14

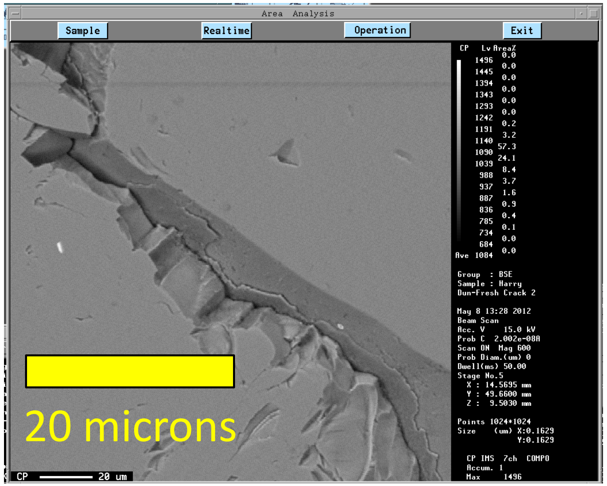
Apr 13 11:53 2012
Beam Scan
Acc. V    15.0 kV
Prob C  4.946e-08A
Scan ON  Mag 1700
Prob Diam.(um) 0
Dwell(ms) 150.00
Stage No.6
  X : 26.9692 mm
  Y : 57.4104 mm
  Z : 11.0575 mm

Points 1024*1024
Size    (um) X:0.0575
             Y:0.0575

  CP IMS  7ch  COMPO
  Accum. 1
  Max     1824
```

- Deformed with reactive brine

- Pitting, signs of dissolution of olivine



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CP   Lv Area%
          0.0
   1496
          0.0
   1445
          0.0
   1394
          0.0
   1343
          0.0
   1293
          0.0
   1242
          0.2
   1191
          3.2
   1140
         57.3
   1090
         24.1
   1039
          8.4
    988
          3.7
    937
          1.6
    887
          0.9
    836
          0.4
    785
          0.1
    734
          0.0
    684
          0.0
Ave 1084

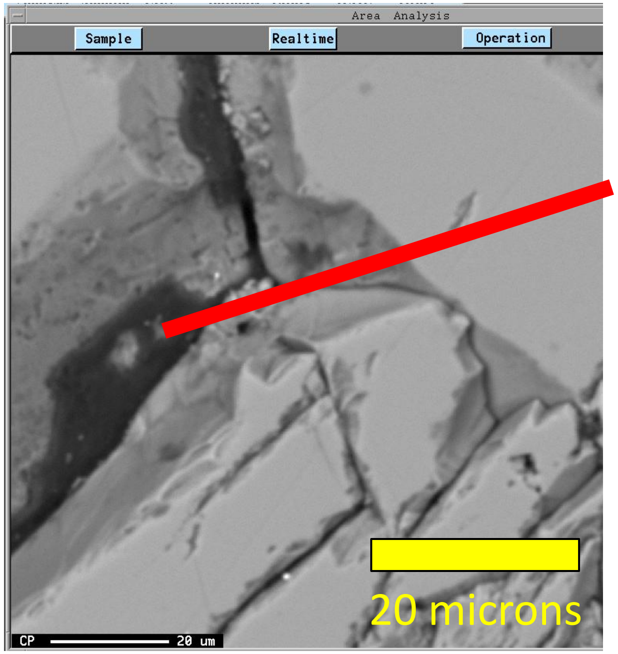
Group  : BSE
Sample : Harry
Dun-Fresh Crack 2

May 8 13:28 2012
Beam Scan
Acc. V    15.0 kV
Prob C  2.002e-08A
Scan ON  Mag 600
Prob Diam.(um) 0
Dwell(ms) 50.00
Stage No.5
  X : 14.5695 mm
  Y : 49.6600 mm
  Z :  9.5030 mm

Points 1024*1024
Size    (um) X:0.1629
             Y:0.1629

  CP IMS  7ch  COMPO
  Accum. 1
  Max     1496
```

smooth, uniform crack surfaces in thermally cracked dunite



probable Mg-carbonate, magnesite ($MgCO_3$) or nesquehonite, hydromagnesite

supported by DOE DE-FE0002386 & C11E10947

Accomplishments to Date

- Quantified extents of carbonation of the olivine and anorthite as a function of temperature, partial pressure of CO_2 and in the presence of various additives

- Demonstrated significant changes in pore structure, morphology and particle size occur after carbonation and dissolution

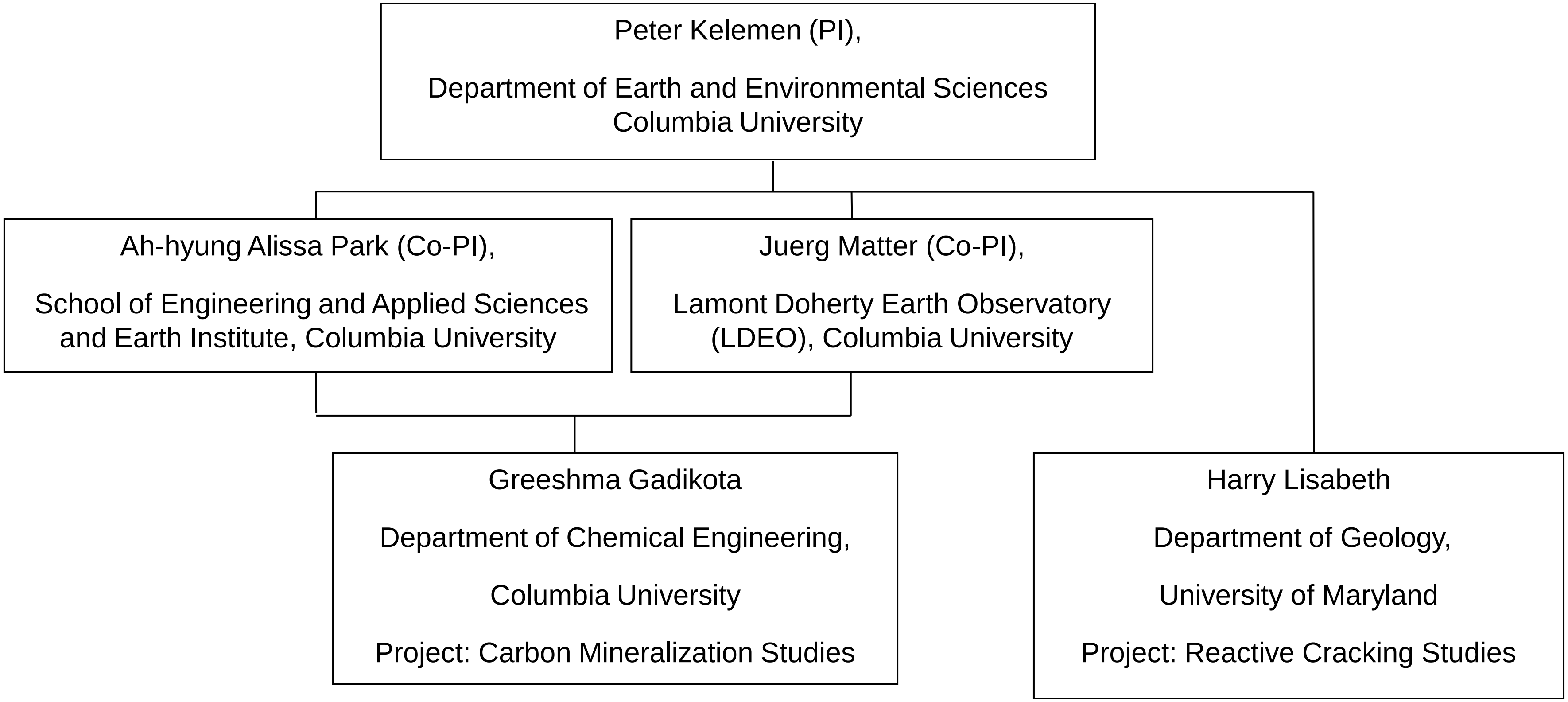
- Initial mineral dissolution rates are substantially higher than longer-term rates with preferential leaching of Mg which has implications for long-term storage of CO_2 in geologic formations

- Determined that reactive brines cause samples to deform more rapidly due to olivine dissolution

Summary

- Higher temperatures and presence of additives such as NaCl and $NaHCO_3$ have a significant impact on enhancing mineral carbonation
- Significant reduction in pore size and surface area after carbonation is evident
- In terms of reactivity with CO_2: olivine > anorthite > basalt
- Initial mineral dissolution rates are higher than longer-term dissolution rates with preferential leaching of Mg compared to Si . Therefore, characterizing initial weathering behavior is more important than previously anticipated
- Reactive brines cause samples to deform more rapidly due to olivine dissolution
- Rapid deformation is apparently due to olivine dissolution, reducing solid-solid contact area along fractures
- Permeability drops due to mechanical compaction are delayed; there is a sudden loss of connectivity, but not of porosity

Organization Chart



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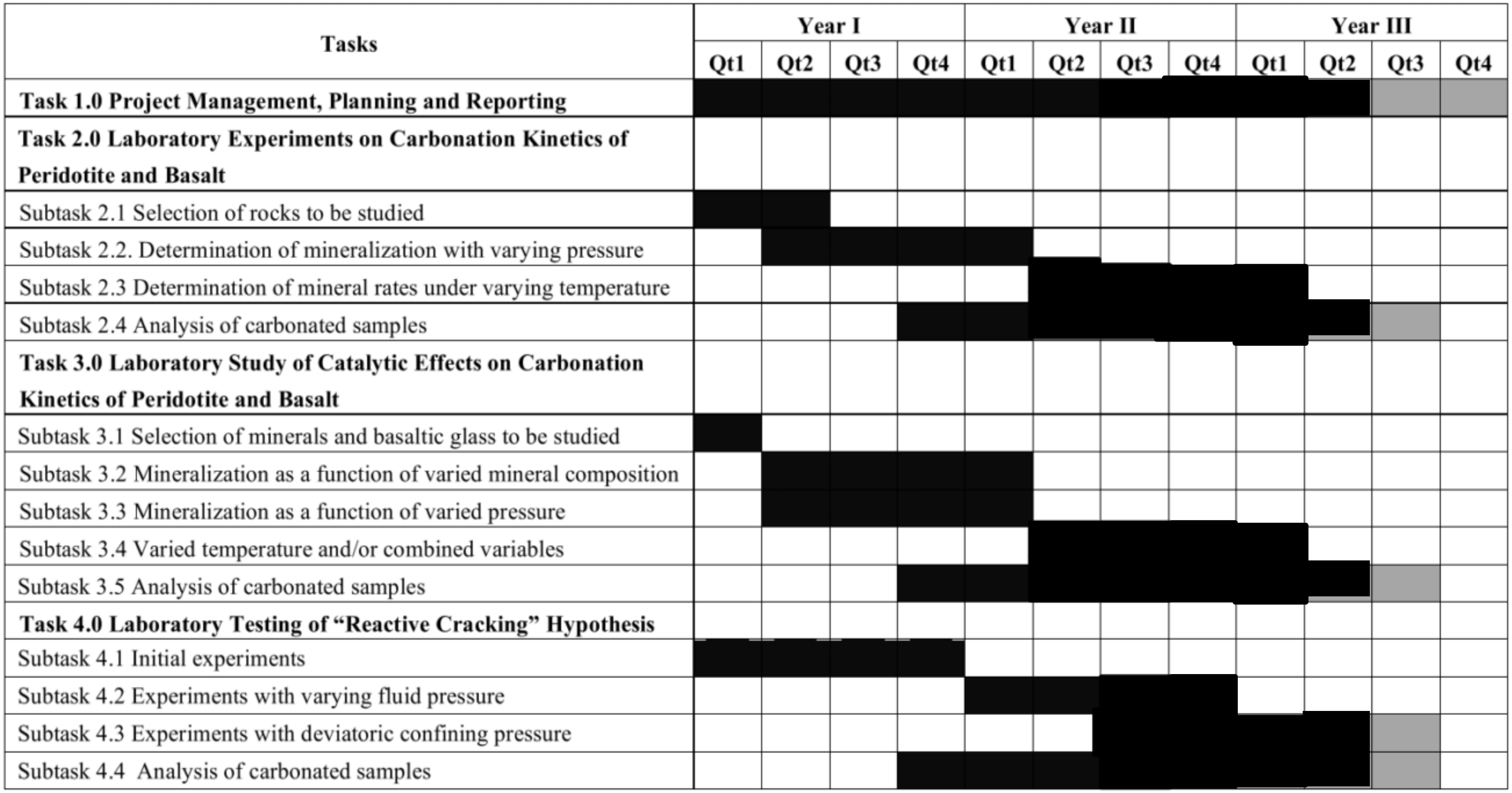
Project: Reactive Cracking Studies

Gantt Chart



Tasks	Year I				Year II				Year III			
	Qt1	Qt2	Qt3	Qt4	Qt1	Qt2	Qt3	Qt4	Qt1	Qt2	Qt3	Qt4
Task 1.0 Project Management, Planning and Reporting	■	■	■	■	■	■	■	■	■	■	▨	▨
Task 2.0 Laboratory Experiments on Carbonation Kinetics of Peridotite and Basalt												
Subtask 2.1 Selection of rocks to be studied	■	■										
Subtask 2.2. Determination of mineralization with varying pressure		■	■	■	■							
Subtask 2.3 Determination of mineral rates under varying temperature						■	■	■	■			
Subtask 2.4 Analysis of carbonated samples				■	■	■	■	■	■	■	▨	
Task 3.0 Laboratory Study of Catalytic Effects on Carbonation Kinetics of Peridotite and Basalt												
Subtask 3.1 Selection of minerals and basaltic glass to be studied	■											
Subtask 3.2 Mineralization as a function of varied mineral composition		■	■	■	■							
Subtask 3.3 Mineralization as a function of varied pressure		■	■	■	■							
Subtask 3.4 Varied temperature and/or combined variables						■	■	■	■			
Subtask 3.5 Analysis of carbonated samples				■	■	■	■	■	■	■	▨	
Task 4.0 Laboratory Testing of "Reactive Cracking" Hypothesis												
Subtask 4.1 Initial experiments	■	■	■	■								
Subtask 4.2 Experiments with varying fluid pressure					■	■	■	■				
Subtask 4.3 Experiments with deviatoric confining pressure							■	■	■	■	▨	
Subtask 4.4 Analysis of carbonated samples				■	■	■	■	■	■	■	▨	

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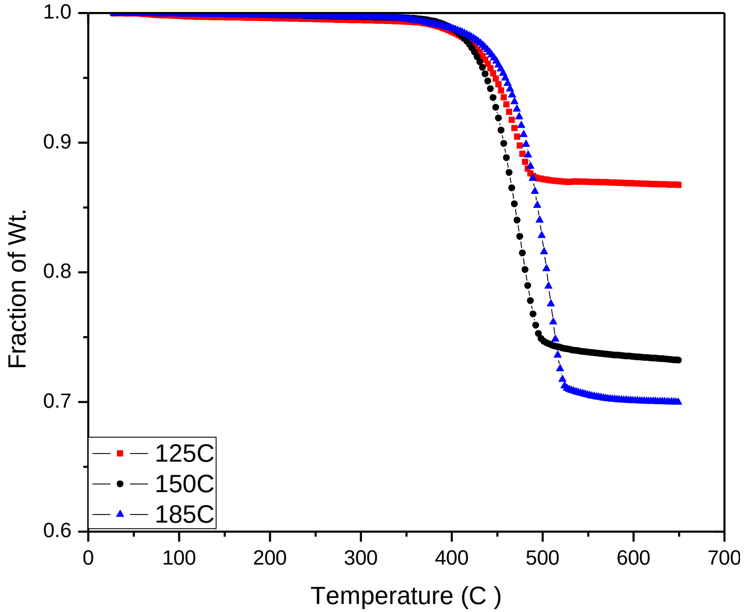
- Journal, multiple authors:
  - Gadikota, G., Matter J., Kelemen P.B., and Park, A-H.A, Mineral Cleaning Procedures and Effect of Grinding on Olivine Carbonation (In Preparation)
  - Kelemen, P.B. and G. Hirth, Reaction-driven cracking during retrograde metamorphism: Olivine hydration and carbonation, Earth Planet. Sci. Lett 345–348, 81–89 2012
  - Kelemen, P.B., J. Matter, E.E. Streit, J.F. Rudge, W.B. Curry, J. Blusztajn, Rates and mechanisms of mineral carbonation in peridotite: Natural processes and recipes for enhanced, in situ CO2 capture and storage, Ann. Rev. Earth Planet. Sci. 39, 545–76, 2011
  - Paukert, A.P., J.M. Matter, P.B. Kelemen, E.L. Shock and J.R. Havig, 2012, Reaction path modeling of enhanced in situ CO2 mineralization for carbon sequestration in the peridotite of the Samail Ophiolite, Sultanate of Oman: Chem. Geol., in press 2012
  - Streit, E., P.B. Kelemen, and J. Eiler, Coexisting serpentine and quartz from carbonate-bearing serpentinized peridotite in the Samail Ophiolite, Oman, Contrib. Mineral. Petrol., online, DOI 10.1007/s00410-012-0775-z, 2012, print publication in press.

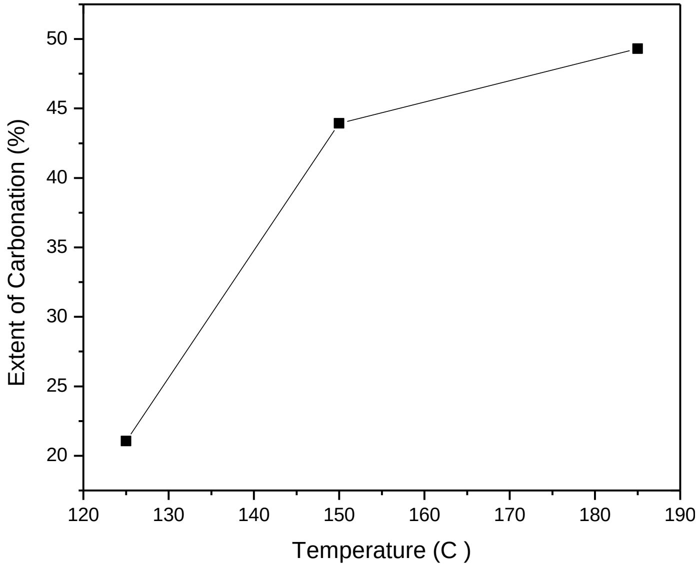
- Conference Proceedings:
  - Gadikota, G., Zhao H., Kelemen P.K., & A.-H. A. Park, “Carbon Mineralization via Carbonation of Calcium and Magnesium-bearing Minerals as Permanent Storage of Anthropogenic CO_2,” The 28th International Pittsburgh Coal Conference, Pittsburgh, PA, Sept., 12-15, 2011.

  - Gadikota, G. & A.-H. A. Park, “Thermodynamic and Kinetic Studies of Mineral Trapping of Carbon in Geologic Formations,” AIChE annual meeting, Salt Lake City, UT, November 7-12, 2010.

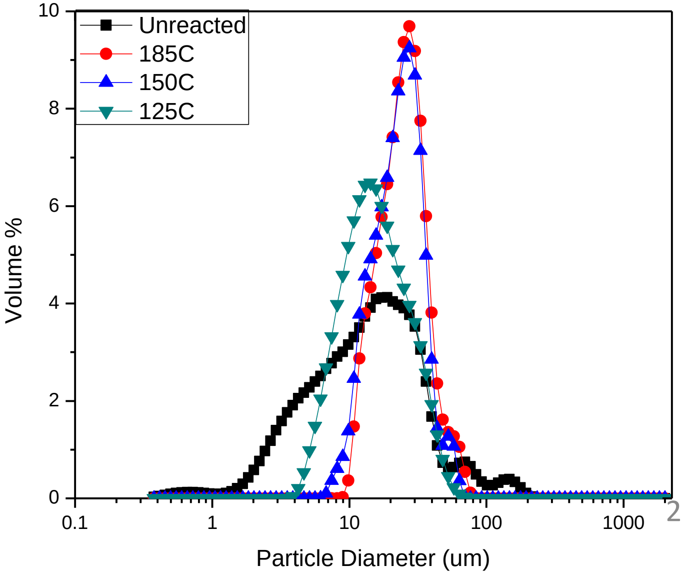
Additional Slides

Effect of Temperature on Olivine Carbonation



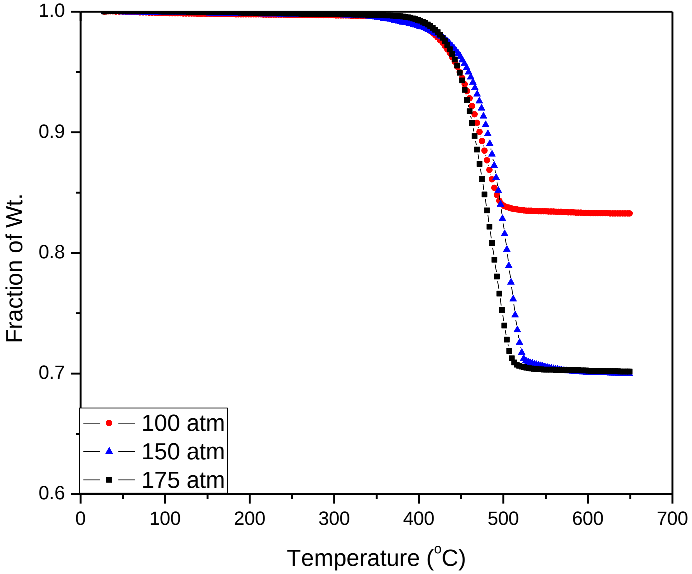


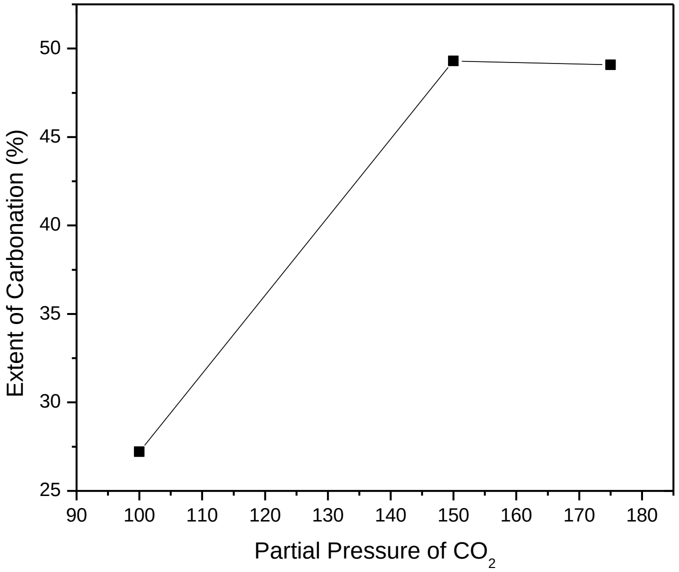
Temperature (°C)	Mean Particle Diameter ± Std. Deviation	Extent of Carbonation (%)
Unreacted	21.41 ± 25.46 µm	-
125°C	18.51 ± 10.43 µm	24.35%
150°C	25.40 ± 10.59 µm	50.93%
185°C	27.71 ± 11.64 µm	57.17%



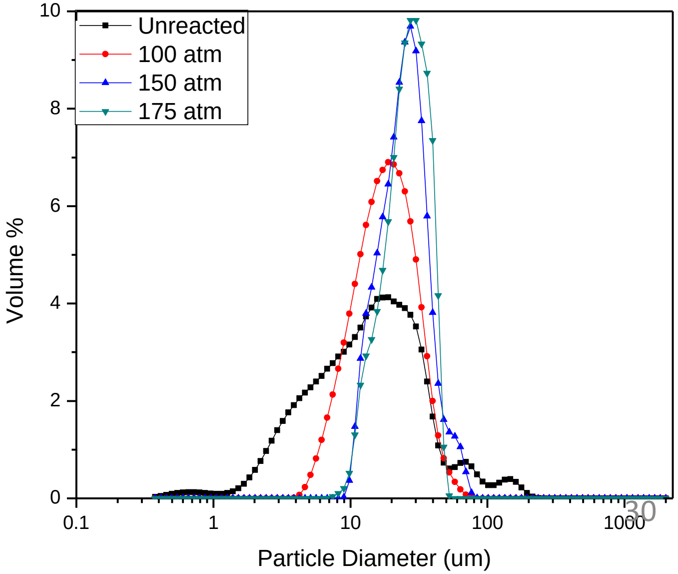
Experimental Conditions: Olivine, 1.0M NaCl + 0.64M NaHCO$_3$,P$_{CO2}$ = 150 atm, 15 wt.% mineral, 800 rpm in high pressure, high temperature batch reactor

Effect of Pressure on Olivine Carbonation



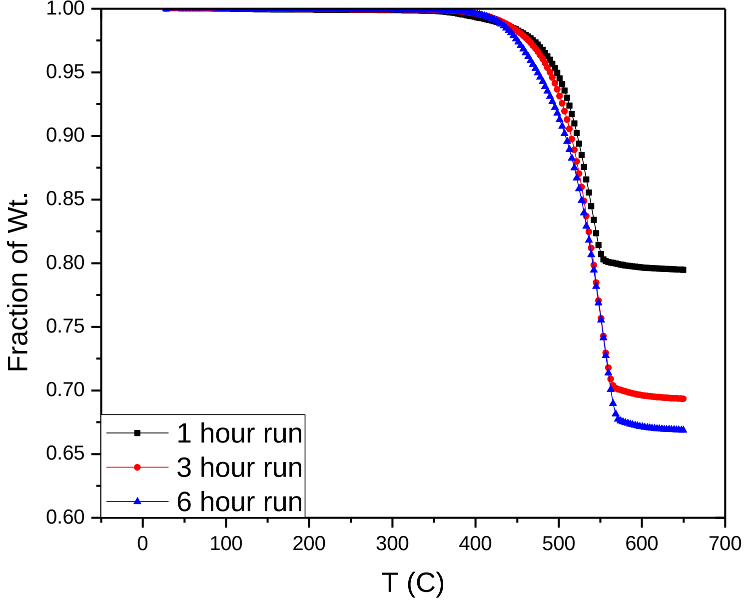


Pressure	Mean Particle Diameter ± Std. Deviation	Extent of Carbonation (%)
Unreacted	21.41 ± 25.46 µm	-
100 atm	20.67 ± 10.45 µm	31.55%
150 atm	27.71 ± 11.64 µm	57.17%
175 atm	27.96 ± 9.42 µm	56.91%

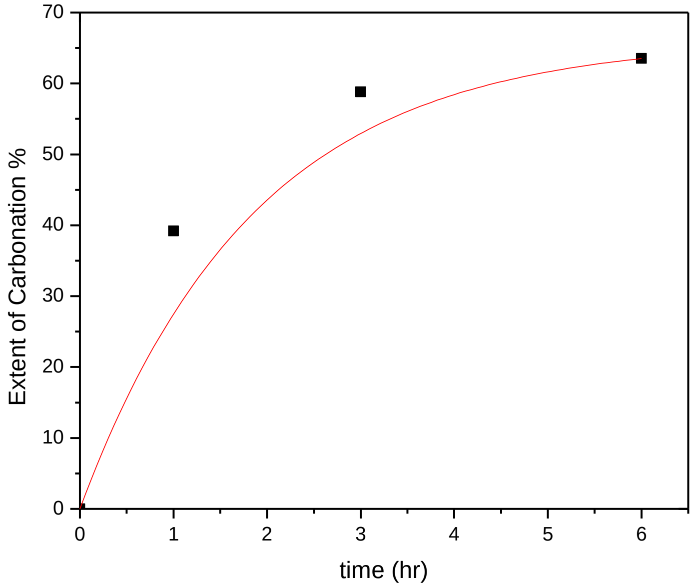


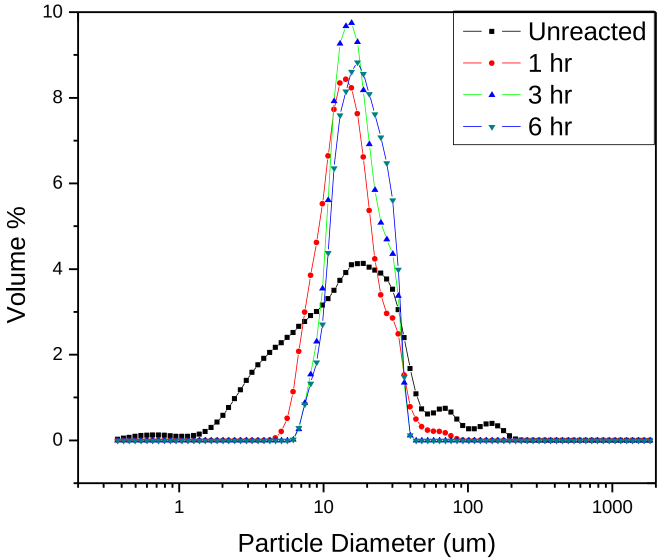
Experimental Conditions: Olivine, 185°C, 15 wt.% mineral, 800 rpm in high pressure, high temperature batch reactor

Effect of Reaction Time on Olivine Carbonation



Time (hours)	Mean Particle Diameter ± Std. Deviation	Extent of Carbonation (%)
Unreacted	21.41 ± 25.46 µm	-
1 hour	17.57 ± 9.35 µm	39.20%
3 h ours	18.68 ± 7.01 µm	58.82%
6 hours	19.93 ± 7.21 µm	63.51%



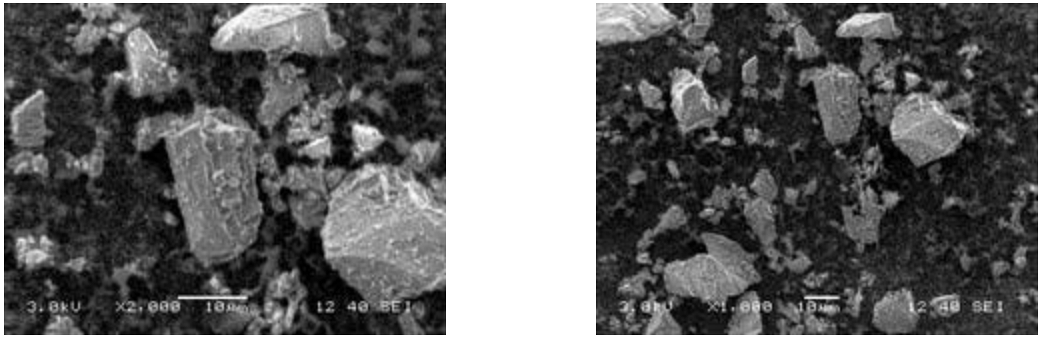


Changes in Composition and Morphology Before and After Olivine Carbonation

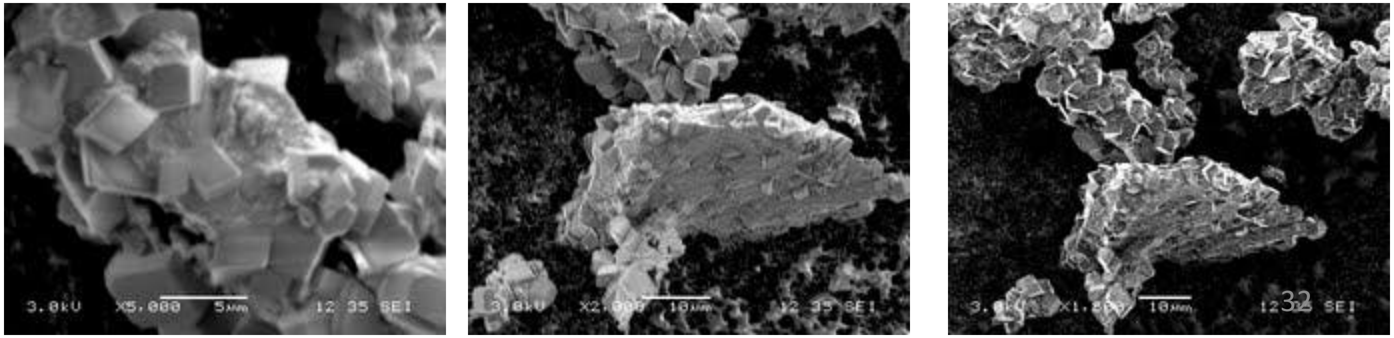
Mineral Composition using XRF Analysis (%)

Mineral	MgO	CaO	Fe_2O_3	SiO_2	Al_2O_3	Na_2O	K_2O	TiO_2	P_2O_5	MnO	Cr_2O_3	V_2O_5	LOI	Sum	Ni
Unreacted Olivine	47.3	0.16	13.9	39.7	0.2	0.01	<0.01	<0.01	< 0.01	0.15	0.78	< 0.01	-0.7	101.5	0.27
Reacted Olivine	32.2	0.11	9.25	26.9	0.14	0.28	<0.01	<0.01	< 0.01	0.10	0.53	< 0.01	30.4	100.0	0.19

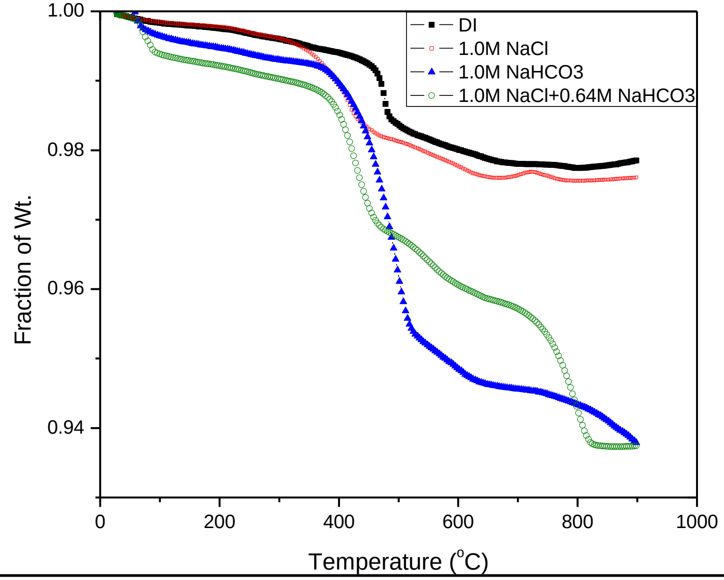
Unreacted Olivine

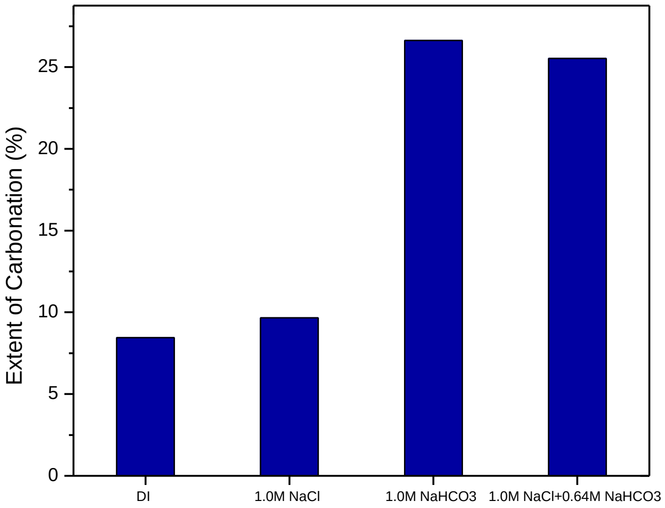


Reacted Olivine at 185°C, 150 atm, 1.0M NaCl, 0.64M $NaHCO_3$, 800 rpm, 15 wt. % solid

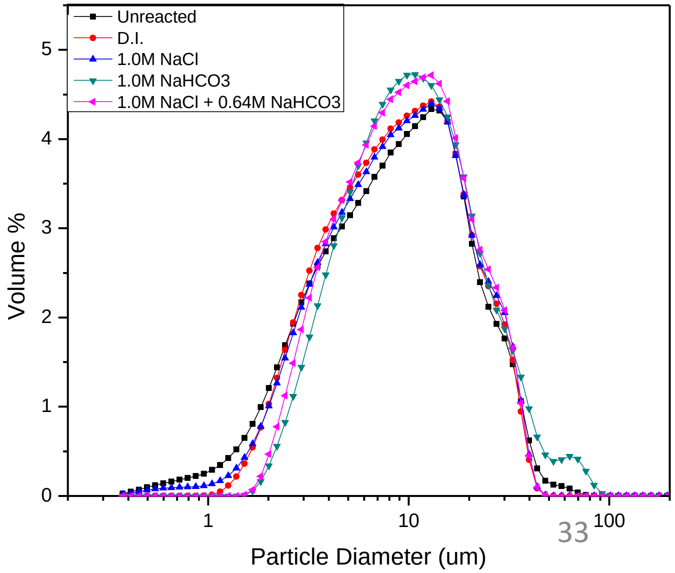


Effect of Chemical Additives on Anorthite Carbonation





Additives	Mean Particle Diameter $\pm$ Std. Deviation	Extent of Carbonation (%)
Unreacted	11.94 ± 9.52 μm	-
DI	11.78 ± 8.54 μm	8.45%
1.0M NaCl	11.89 ± 9.49 μm	9.66%
1.0M $NaHCO_3$	14.34 ± 12.26 μm	26.63%
1.0M NaCl+0.64M $NaHCO_3$	12.42 ± 8.52 μm	25.53%



Experimental Conditions: Olivine, 185ºC, P_{CO2} = 150 atm, 15 wt.% mineral, 800 rpm in high pressure, high temperature batch reactor

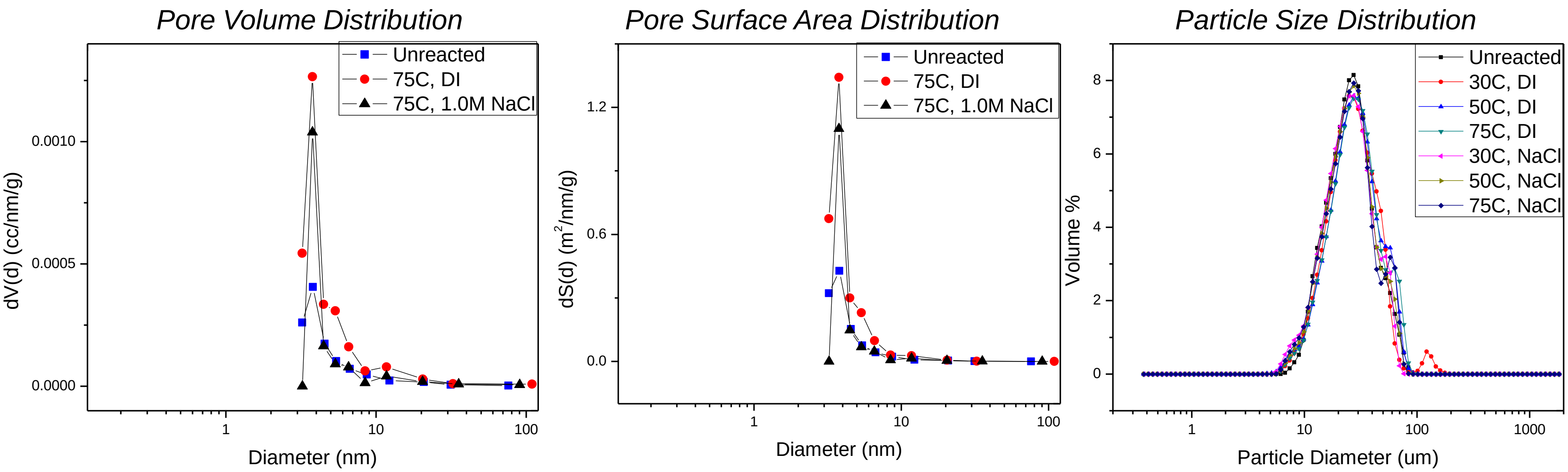
Results Summary

	Reaction Conditions	Mean Particle Diameter ± Std. Deviation	Extent of Carbonation (%)
Olivine	Unreacted	21.41 ± 25.46 µm	-
	DI, 185^{o}C, 150 atm	16.43 ± 11.85 µm	4.40%
	1.0M NaCl, 185^{o}C, 150 atm	26.54 ± 12.77 µm	37.44%
	1.0M NaHCO$_3$, 185^{o}C, 150 atm	32.76 ± 14.39 µm	50.60%
	1.0M NaCl+0.64M NaHCO$_3$, 185^{o}C, 150 atm	27.71 ± 11.64 µm	49.30%
	1.0M NaCl+0.64M NaHCO$_3$, 150^{o}C, 150 atm	25.40 ± 10.59 µm	43.93%
	1.0M NaCl+0.64M NaHCO$_3$, 125^{o}C, 150 atm	18.51 ± 10.43 µm	21.05%
	1.0M NaCl+0.64M NaHCO$_3$, 185^{o}C, 100 atm	20.67 ± 10.45 µm	27.21%
	1.0M NaCl+0.64M NaHCO$_3$, 185^{o}C, 175 atm	27.96 ± 9.42 µm	49.08%
Anorthite	Unreacted	11.94 ± 9.52 µm	-
	DI, 185^{o}C, 150 atm	11.78 ± 8.54 µm	8.45%
	1.0M NaCl, 185^{o}C, 150 atm	11.89 ± 9.49 µm	9.66%
	1.0M NaHCO$_3$, 185^{o}C, 150 atm	14.34 ± 12.26 µm	26.63%
	1.0M NaCl+0.64M NaHCO$_3$, 185^{o}C, 150 atm	12.42 ± 8.52 µm	25.53%

All experiments performed at 15 wt. % solid and 800 rpm, reaction time of 3 hours (+ 1 hour for heating, 1.25 hours for cooling)

Changes in Pore and Particle Size Distribution due to Mineral Dissolution and Coupled Reactions

(a) Mineral Dissolution

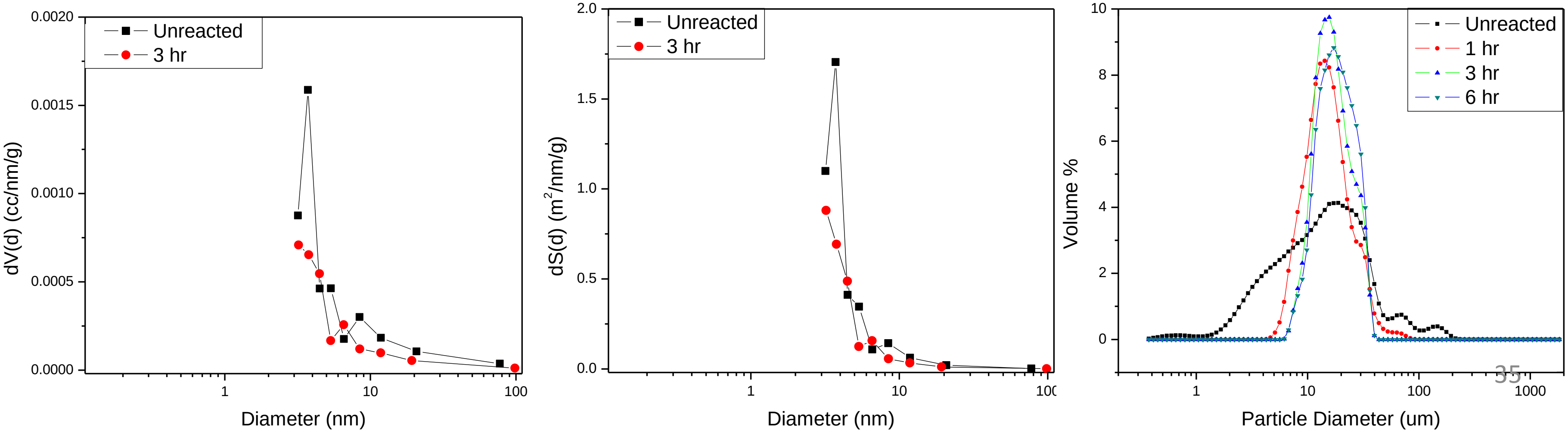


Pore Volume Distribution

Pore Surface Area Distribution

Particle Size Distribution

(b) Coupled Mineral Dissolution and Carbonation



Conclusion

- Additives such as NaCl and $NaHCO_3$ enhance mineral carbonation significantly
- Determined that increasing $[NaHCO_3]$ increases olivine and anorthite carbonation
- Average particle diameter increases and particle size distribution becomes narrower with higher carbonation
- Pore size decreases with increasing carbonation – this has implications for in-situ mineral carbon storage

Questions?