



# **POROSITY CHANGES IN THE JANGGI BASIN**

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**05/01/2025 CHEM 428**



# AGENDA

Introduction

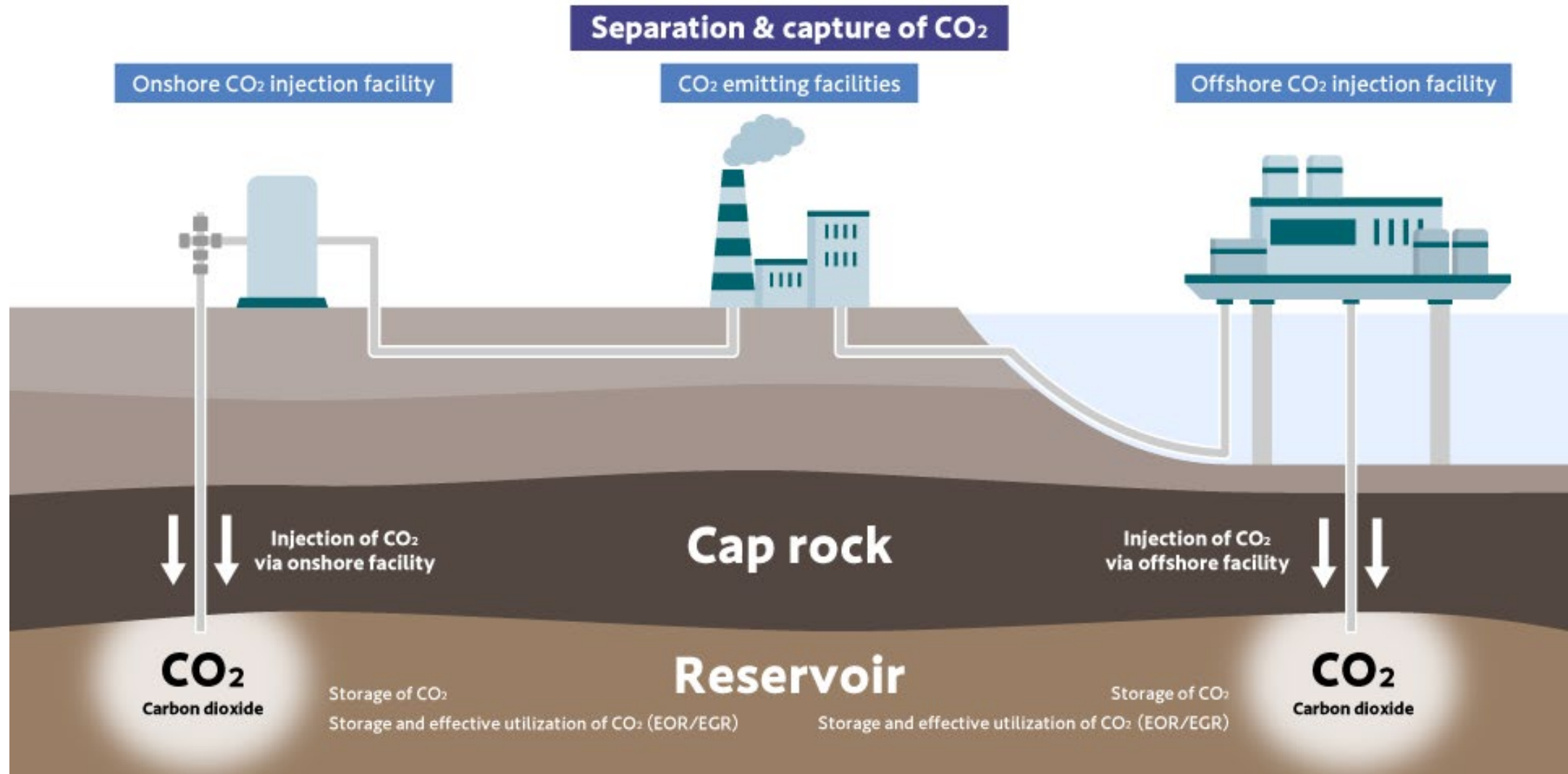
Background

Methods

Geochemical Modeling

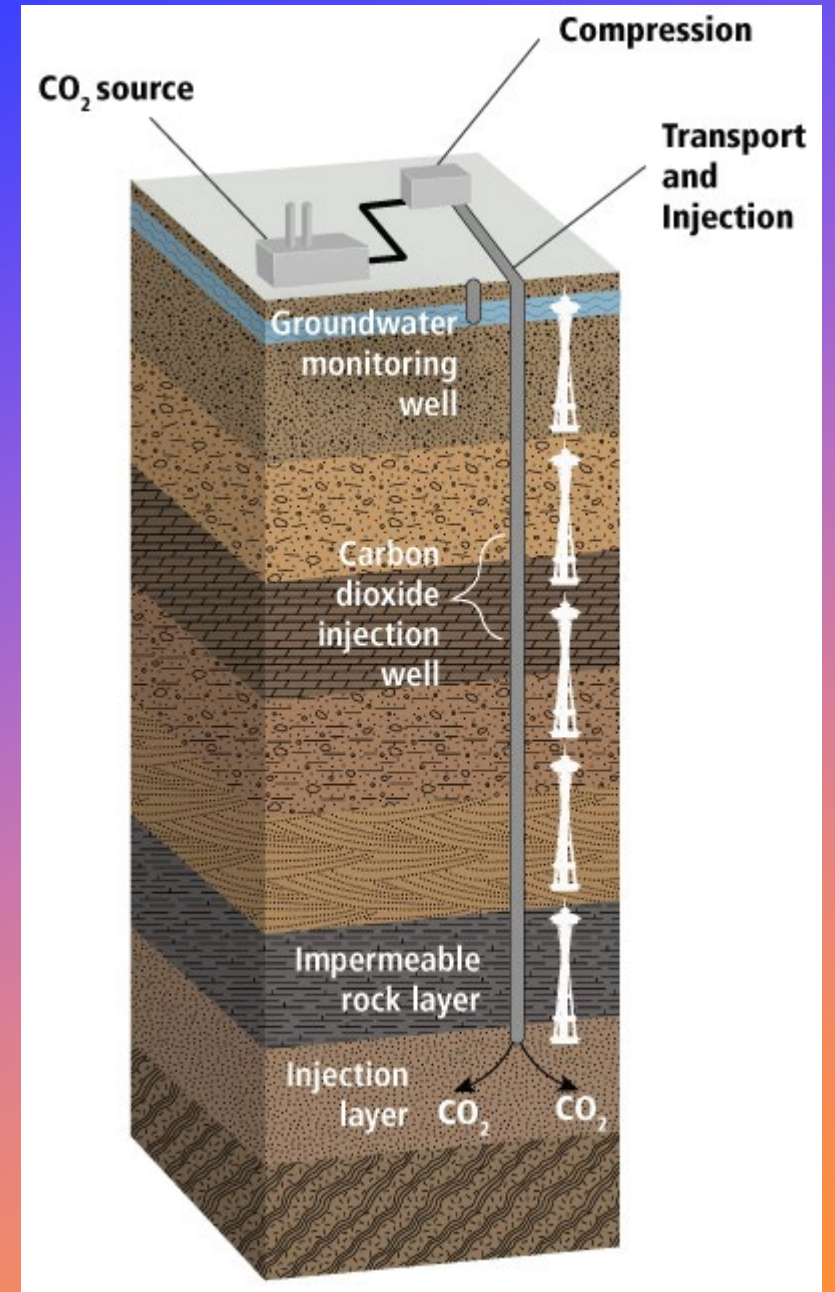
Summary

# CO<sub>2</sub> Capture



# CO2 Reservoirs

- Places that can securely contain CO2 over long periods of time
  - Depleted Oil and gas reservoirs
  - Layers of porous rock
- Basalt Formations
  - Solidified lava: potential to convert injected CO2 to solid mineral
- Shale Basins
  - Sedimentary rock characterized by thin horizontal layers with very low permeability in the vertical direction: Good adsorption for CO2 storage





# CAP ROCKS

Form over the top of reservoirs and create a layer that is impervious to fluid flow (fluids prevented from escaping the reservoir)

- When a rock that is harder to break down (resists weathering) overlies an easier to break down rock (less resistant to weathering) the rock on top is the cap rock
- Develop from solution of salt from the top of a salt core; leaves a residue of insoluble anhydrite that is later altered to gypsum, calcite and sulfur.

Often formed from shale, anhydrite or salts

- Ranges from 0 to 300 meters thick

Can protect the more delicate layers from weather and erosion OR cap rock can remain while the more delicate lower layers are weathered and eroded under it.

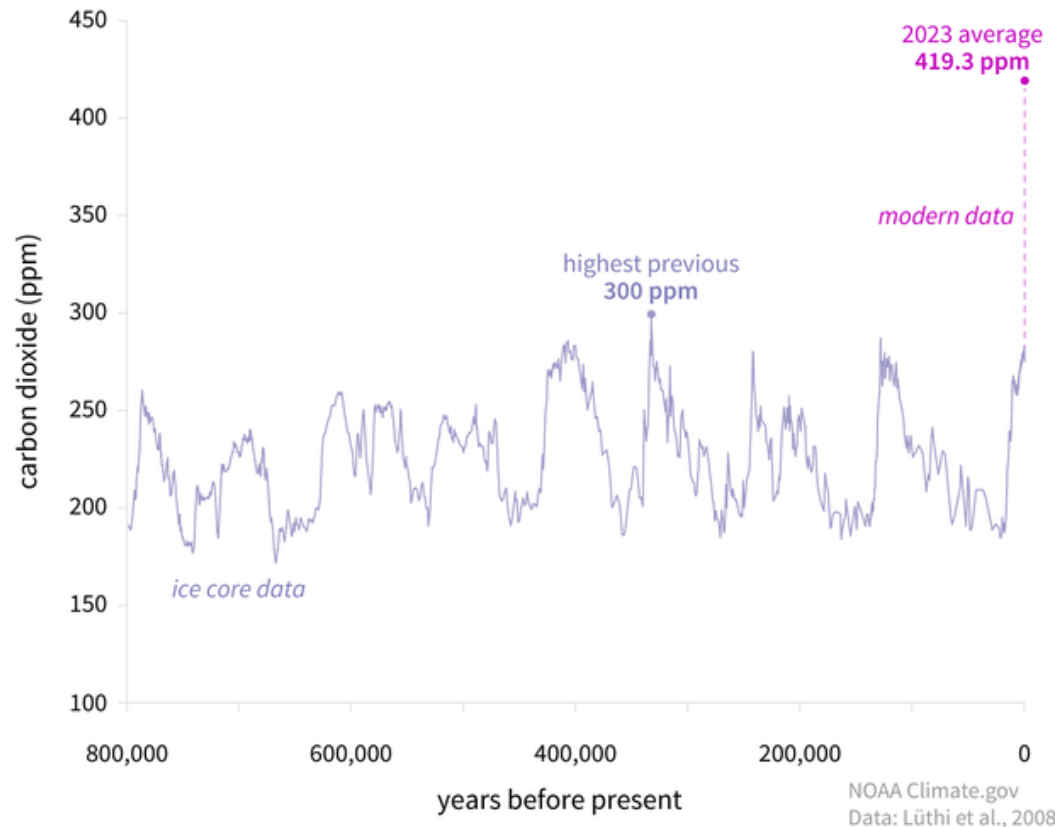




# THE ARTICLE

# RESEARCH PROBLEM

## CARBON DIOXIDE OVER 800,000 YEARS

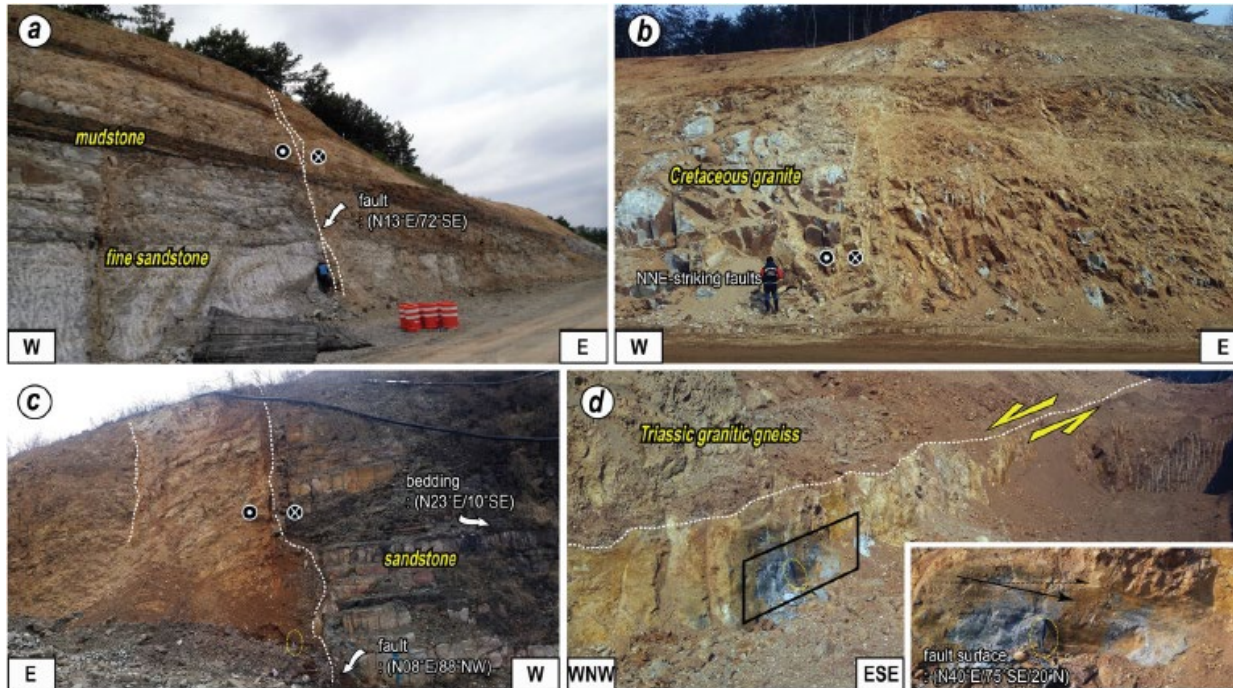


CO<sub>2</sub> accounts for 2/3 of Earth warming

- Atmospheric CO<sub>2</sub> 419.3ppm in 2023

CO<sub>2</sub> storage

- Uncertainties of geochemical storage
  - The CO<sub>2</sub> injected into reservoirs migrates upward due to its buoyancy, resulting in contact with cap rocks. Acidic conditions promote CO<sub>2</sub>–water–cap rock interactions.
  - Dissolution of carbonate rocks through acidic CO<sub>2</sub>-water mixtures
  - These geochemical reactions can affect pore water quality but also shape the pore and pore throat morphologies.



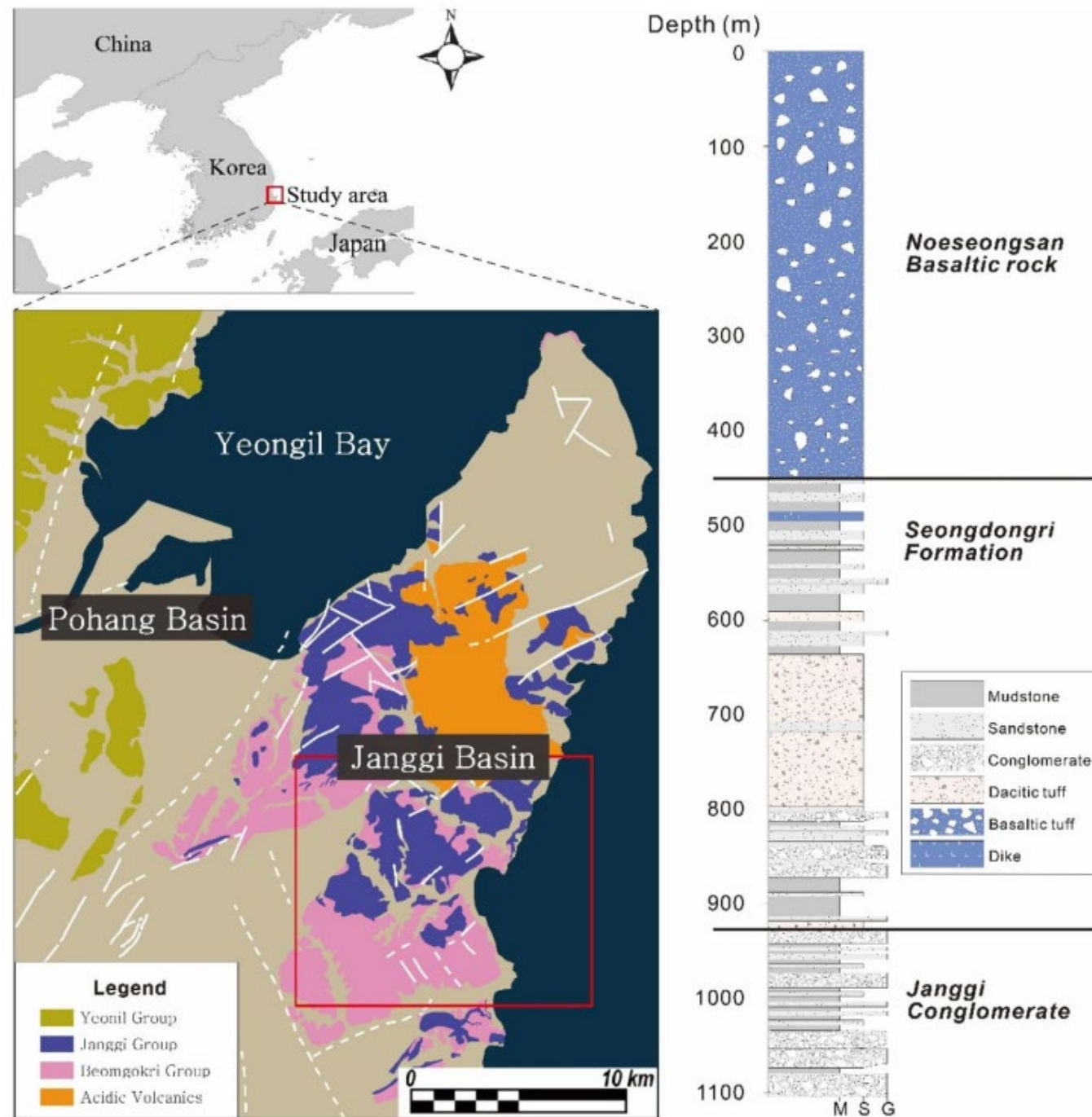
# JANGGI BASIN

- Miocene sedimentary basin in southeastern South Korea
- Formed by the opening of the East Sea associated with (NNW)-trending dextral strike-slip faults and (NNE)- to (NE)-trending normal faults
- Mainly filled with volcanic and nonmarine sedimentary deposits
- Basaltic Rock (mainly basaltic tuff) distributed at various depths depending on the basin structure and was observed to exist at depth of approximately 450 meters below the surface from a borehole
- Identify the geochemical reactivity of basaltic tuff as a cap rock and its effect on porosity changes by CO<sub>2</sub>-water-rock interactions

**Table 1** Mineral components of the basaltic tuff determined using powder X-ray diffraction (XRD)

Mineral	Weight %
Plagioclase	29.9
Smectite	23.4
Clinoptilolite	22.9
Analcime	6.3
Pyroxene	9.5
Stilbite	6.2
Quartz	1.8





# EXPERIMENTAL METHODS

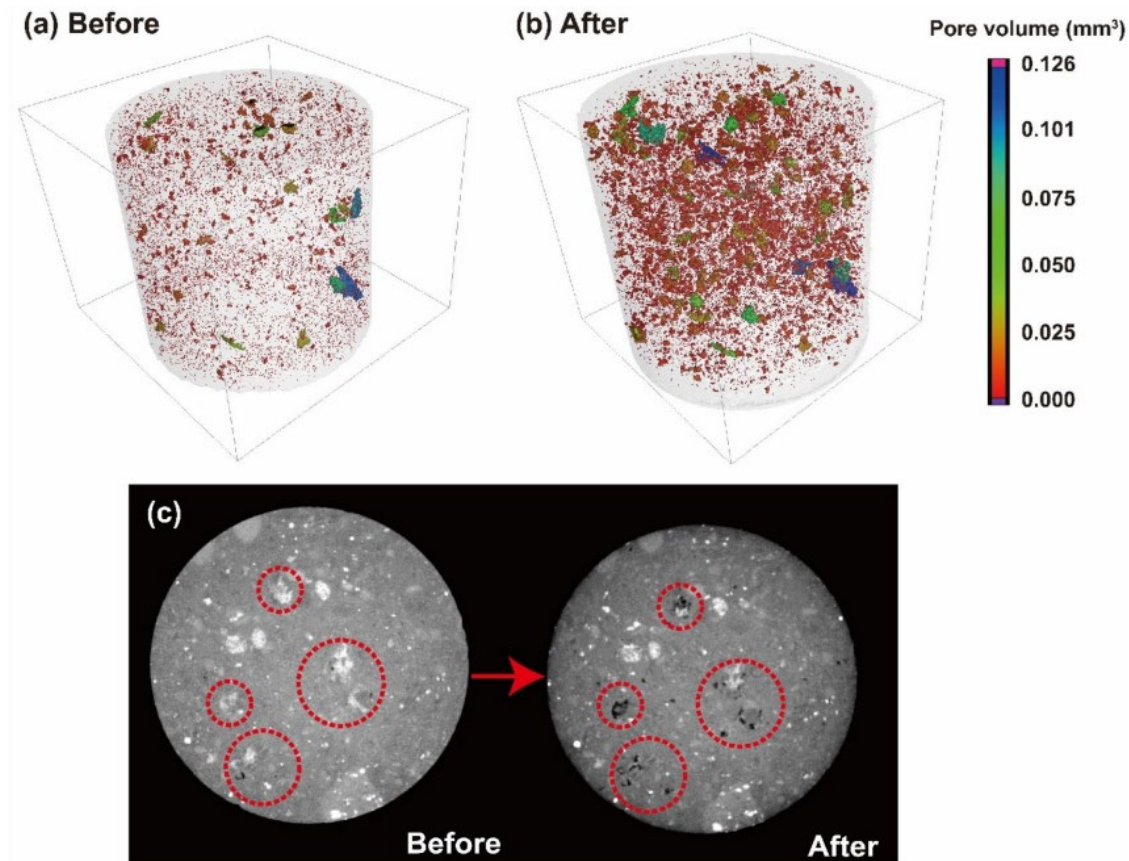
## Solid Sampling

1. Mineralogical analysis with X-ray diffraction
2. Changes in surface area by N<sub>2</sub> adsorption
3. Porosity changes by 3D x-ray CT scans

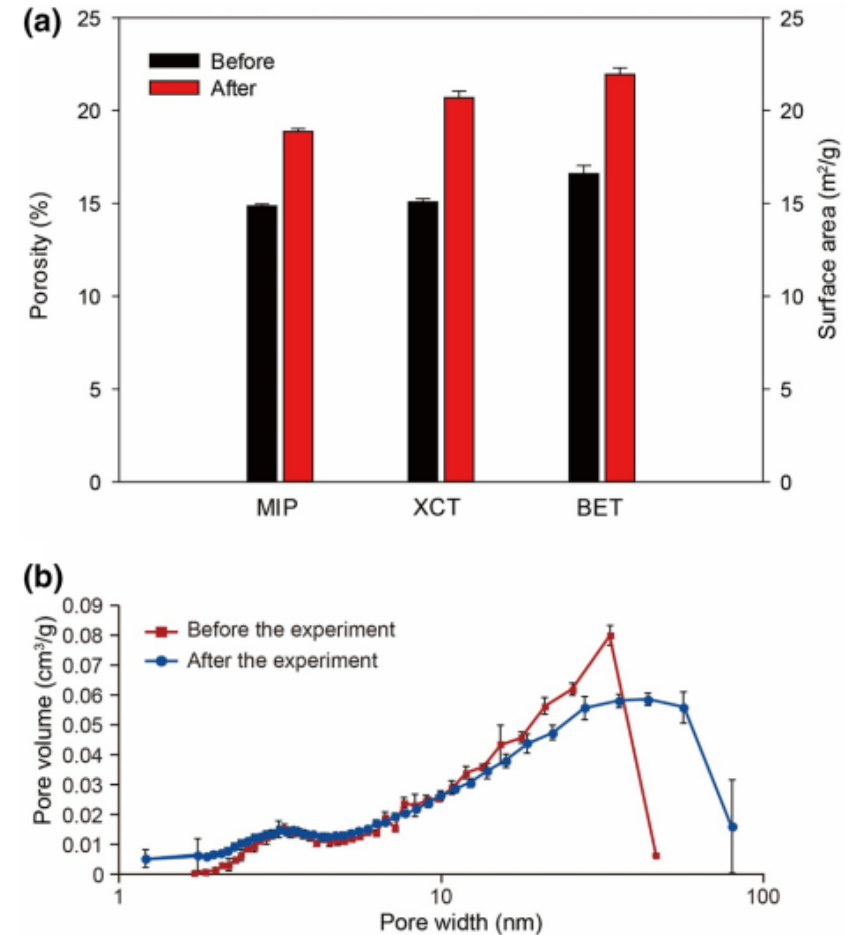
## PREPARATION

1. 13.77 g of the total rock samples and 140 ml of distilled water were filled in a reactor.
2. The reactor was purged with gaseous CO<sub>2</sub> to remove oxygen from the water and then pressurized to 100 bar of CO<sub>2</sub> gas using a syringe pump
3. The experiment was run at 50 °C and 100 bar for 84 days without a stirrer.

# PHYSICAL RESULTS



Three-dimensional images of pore volume in basaltic tuff after experiment using XCT scan.



Comparison of porosities and surface areas. Error bars are standard deviation of the mean.

# GEOCHEMICAL MODELING

## Concentration

- Kinetic reaction modeling for CO<sub>2</sub>–water–basaltic tuff interactions was conducted using PHREEQC.
- Lawrence Livermore National Laboratory (LLNL) database.
- Plagioclase was represented as albite and anorthite in a ratio of 1:2.13 based on the molar ratio of Ca and Na.
- Pyroxene was represented as enstatite and hedenbergite in a ratio of 1:1.15 from the molar ratio of Fe, Mg, and Ca.
- The possible secondary minerals considered were calcite, dolomite, illite, kaolinite, magnesite, magnetite, montmorillonite, siderite, and SiO<sub>2</sub> (am).

## Kinetics

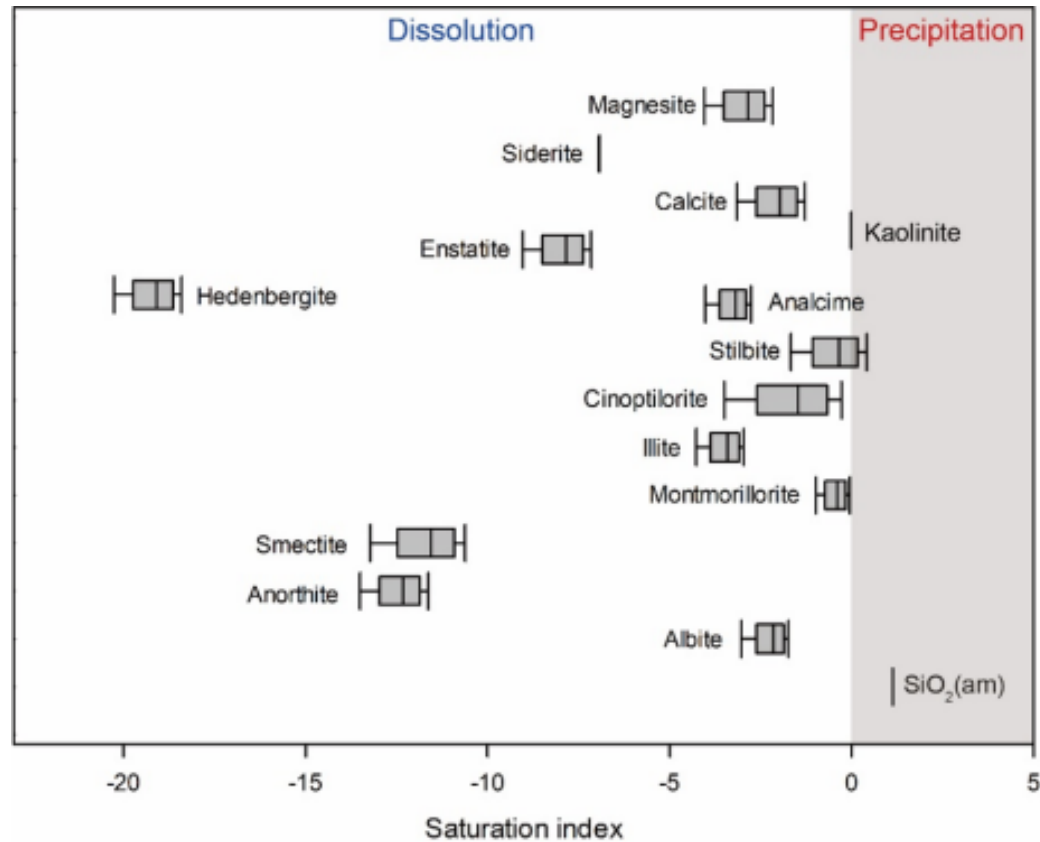
- Rate calculated by rate law equation

$$\begin{aligned} &= A \left[ k_a \exp \left[ \frac{-E_a}{R} \left( \frac{1}{T} - \frac{1}{298.15} \right) \right] a_{H^+}^{na} \left( 1 - \frac{Q}{k} \right) \right] \\ &+ k_{nu} \exp \left[ \frac{-E_n}{R} \left( \frac{1}{T} - \frac{1}{298.15} \right) \right] \left( 1 - \frac{Q}{k} \right) \\ &+ k_b \exp \left[ \frac{-E_b}{R} \left( \frac{1}{T} - \frac{1}{298.15} \right) \right] a_{H^+}^{nb} \left( 1 - \frac{Q}{k} \right) \end{aligned}$$

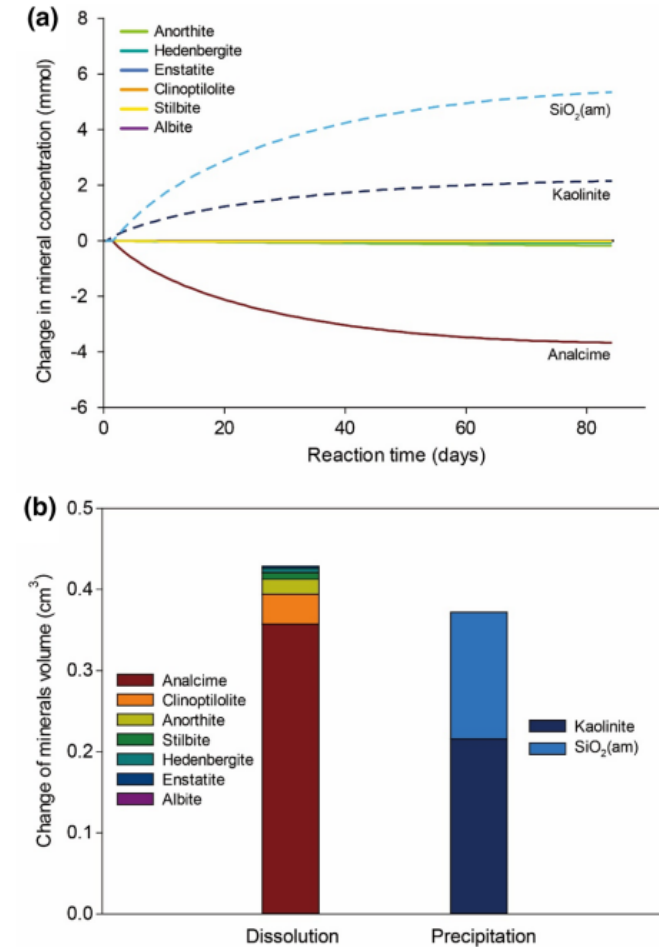
- $E$  = activation energy |  $Q$  = reaction product |  $k$  = equilibrium constant |  $A$  = specific reactive surface area |  $a$  = activity |  $n$  = power constant |  $R$  = gas constant
- Calculated for all minerals and then modeled



# MODELING RESULTS



Saturation indices of primary and secondary minerals.



**a.** Changes in the mineral concentrations in the basaltic tuff sample by CO<sub>2</sub>-water-rock interaction. **b.** Changes in mineral volumes. Left bar represents dissolved volume of primary minerals. Right bar represents the precipitated volume of secondary minerals.

# MY GEOCHEMICAL MODELING

My goals:

- Track SI's of the primary and secondary minerals
- Calculate concentration of ions
- Model pH of solution after interactions
- Model kinetics of the system

```
SOLUTION_MASTER_SPECIES
Ca(+2)      Ca+2  1  Ca

Al(+3)      Al+3  1  Al

Fe(+2)      Fe+2  1  Fe

Fe(+3)      Fe+3  -2  Fe

K(+ )       K+    1  K

Na(+ )      Na+    1  Na

Mg(+2)      Mg+2  1  Mg

Si(0)       SiO2   1  SiO2
```

```
SOLUTION 1
temp 50
-pressure 98.6923
pH 7
units mol/l
density 1
-water 1 #kg
```

```
EQUILIBRIUM_PHASES 1
```

```
Albite
Anorthite
Calcite
Quartz
Enstatite
Hedenbergite
Analcime
Clinoptilolite-Ca
Stilbite
CO2(g) -2.0
dolomite
kaolinite
magnesite
magnetite
siderite
```

```
SOLUTION_MASTER_SPECIES
Ca(+2)      Ca+2    2.513e-06  Ca

Al(+3)      Al+3    5.062e-07  Al

Fe(+2)      Fe+2    1.485e-09  Fe

Fe(+3)      Fe+3    1.485e-09  Fe

K(+ )       K+      1  K

Na(+ )      Na+    1.876e-08  Na

Mg(+2)      Mg+2    2.798e-08  Mg

Si(0)       SiO2    2.777e-06  SiO2
```

```
SOLUTION 1
temp 50
-pressure 98.6923
pH 7
units mol/l
density 1
-water 1 #kg
```

```
SOLUTION 1
EQUILIBRIUM_PHASES 1
```

```
Albite 0.0
Anorthite 0.0
Calcite 0.0
Quartz 0.0
Enstatite 0.0
Hedenbergite 0.0
Analcime 0.0
Clinoptilolite-Ca 0.0
Stilbite 0.0
CO2(g) -2.0
dolomite 0.0
kaolinite 0.0
magnesite 0.0
magnetite 0.0
siderite 0.0
```

# CONCENTRATION AND SI'S

-----Phase assemblage-----						
Phase	SI	log IAP	log K(T, P)	Moles in assemblage		
				Initial	Final	Delta
Albite	-14.88	-13.00	1.89	1.000e+01	1.960e+01	9.600e+00
Analcime	-12.34	-7.35	4.99	1.000e+01	0	-1.000e+01
Anorthite	-13.50	8.82	22.32	1.000e+01	3.725e+01	2.725e+01
CO2 (g)	-9.20	-17.19	-7.99	1.000e+01	0	-1.000e+01
Calcite	-9.90	-8.44	1.46	1.000e+01	8.497e-02	-9.915e+00
Clinoptilolite-Ca	-43.52	-52.40	-8.88	1.000e+01	0	-1.000e+01
Enstatite	-9.46	0.69	10.15	1.000e+01	1.000e+01	-1.415e-05
Hedenbergite	-15.74	2.06	17.80	1.000e+01	0	-1.000e+01
Quartz	-2.50	-6.10	-3.61	1.000e+01	1.324e+02	1.224e+02
Stilbite	-22.00	-22.47	-0.47	1.000e+01	1.000e+01	1.667e-08
Dolomite	-20.47	-18.84	1.63	1.000e+01	2.000e+01	1.000e+01
Kaolinite	-4.55	0.07	4.62	1.000e+01	0	-1.000e+01
Magnesite	-12.05	-10.40	1.66	1.000e+01	0	-1.000e+01
Magnetite	-5.53	1.95	7.47	1.000e+01	1.009e+01	8.500e-02
Siderite	-11.00	-11.68	-0.67	1.000e+01	1.991e+01	9.915e+00

-----Solution composition-----		
Elements	Molality	Moles
Al	1.443e-07	5.062e-07
C	1.933e-10	6.779e-10
Ca	7.165e-07	2.513e-06
Fe	4.234e-10	1.485e-09
K	0.000e+00	0.000e+00
Mg	7.978e-09	2.798e-08
Na	5.350e-09	1.876e-08
Si	7.916e-07	2.777e-06

-----Phase assemblage-----						
Phase	SI	log IAP	log K(T, P)	Moles in assemblage		
				Initial	Final	Delta
Albite	-14.55	-11.89	2.66	1.000e+01	1.960e+01	9.600e+00
Analcime	-12.05	-5.99	6.06	1.000e+01	0	-1.000e+01
Anorthite	-13.17	13.31	26.48	1.000e+01	1.725e+01	7.250e+00
CO2 (g)	-11.24	-19.07	-7.83	1.000e+01	0	-1.000e+01
Calcite	-10.77	-8.95	1.82	1.000e+01	4.009e+01	3.009e+01
Clinoptilolite-Ca	-41.04	-48.56	-7.52	1.000e+01	0	-1.000e+01
Enstatite	-10.69	0.60	11.29	1.000e+01	3.000e+01	2.000e+01
Hedenbergite	-15.55	3.98	19.53	1.000e+01	0	-1.000e+01
Quartz	-2.43	-6.46	-4.03	1.000e+01	1.124e+02	1.024e+02
Stilbite	-20.07	-19.26	0.81	1.000e+01	1.000e+01	1.667e-08
Dolomite	-23.43	-20.96	2.47	1.000e+01	0	-1.000e+01
Kaolinite	-3.53	3.18	6.72	1.000e+01	2.000e+01	1.000e+01
Magnesite	-14.28	-12.01	2.27	1.000e+01	0	-1.000e+01
Magnetite	-7.28	3.14	10.42	1.000e+01	1.009e+01	8.500e-02
Siderite	-12.08	-12.30	-0.22	1.000e+01	1.991e+01	9.915e+00

-----Solution composition-----		
Elements	Molality	Moles
Al	1.999e-07	5.572e-07
C	1.172e-11	3.267e-11
Ca	7.443e-07	2.074e-06
Fe	3.418e-10	9.525e-10
K	0.000e+00	0.000e+00
Mg	6.421e-10	1.790e-09
Na	2.025e-09	5.643e-09
Si	3.538e-07	9.861e-07

# CONCENTRATION AND SI'S CONT.

-----Phase assemblage-----						
Phase	SI	log IAP	log K(T, P)	Moles in assemblage		
				Initial	Final	Delta
Albite	-14.55	-11.89	2.66	1.000e+01	1.960e+01	9.600e+00
Analcmite	-12.05	-5.99	6.06	1.000e+01	0	-1.000e+01
Anorthite	-13.17	13.31	26.48	1.000e+01	1.725e+01	7.250e+00
CO2(g)	-11.24	-19.07	-7.83	1.000e+01	0	-1.000e+01
Calcite	-10.77	-8.95	1.82	1.000e+01	4.009e+01	3.009e+01
Clinoptilolite-Ca	-41.04	-48.56	-7.52	1.000e+01	0	-1.000e+01
Enstatite	-10.69	0.60	11.29	1.000e+01	3.000e+01	2.000e+01
Hedenbergite	-15.55	3.98	19.53	1.000e+01	0	-1.000e+01
Quartz	-2.43	-6.46	-4.03	1.000e+01	1.124e+02	1.024e+02
Stilbite	-20.07	-19.26	0.81	1.000e+01	1.000e+01	1.667e-08
Dolomite	-23.43	-20.96	2.47	1.000e+01	0	-1.000e+01
Kaolinite	-3.53	3.18	6.72	1.000e+01	2.000e+01	1.000e+01
Magnesite	-14.28	-12.01	2.27	1.000e+01	0	-1.000e+01
Magnetite	-7.28	3.14	10.42	1.000e+01	1.009e+01	8.500e-02
Siderite	-12.08	-12.30	-0.22	1.000e+01	1.991e+01	9.915e+00
-----Solution composition-----						
Elements	Molality		Moles			
Al	1.999e-07		5.572e-07			
C	1.172e-11		3.267e-11			
Ca	7.443e-07		2.074e-06			
Fe	3.418e-10		9.525e-10			
K	0.000e+00		0.000e+00			
Mg	6.421e-10		1.790e-09			
Na	2.025e-09		5.643e-09			
Si	3.538e-07		9.861e-07			

-----Phase assemblage-----						
Phase	SI	log IAP	log K(T, P)	Moles in assemblage		
				Initial	Final	Delta
Albite	-23.70	-21.04	2.66	1.960e+01	0	-1.960e+01
Analcmite	-17.63	-11.57	6.06	1.124e+02	1.328e+02	2.042e+01
Anorthite	-19.05	7.42	26.48	1.725e+01	0	-1.725e+01
CO2(g)	-7.56	-15.39	-7.83	1.000e+01	0	-1.000e+01
Calcite	-15.66	-13.84	1.82	4.009e+01	2.650e+02	2.249e+02
Clinoptilolite-Ca	-75.56	-83.08	-7.52	1.000e+01	1.991e+01	9.911e+00
Enstatite	-18.96	-7.68	11.29	3.000e+01	2.548e+02	2.248e+02
Hedenbergite	-37.69	-18.16	19.53	1.124e+02	0	-1.124e+02
Quartz	-5.36	-9.39	-4.03	1.124e+02	0	-1.124e+02
Stilbite	-33.11	-32.30	0.81	1.000e+01	1.000e+01	1.667e-08
Dolomite	-29.99	-27.51	2.47	1.124e+02	0	-1.124e+02
Kaolinite	0.64	7.35	6.72	2.000e+01	3.000e+01	1.000e+01
Magnesite	-15.95	-13.68	2.27	1.124e+02	0	-1.124e+02
Magnetite	-2.54	7.88	10.42	1.009e+01	1.001e+01	-8.362e-02
Siderite	-16.10	-16.32	-0.22	1.991e+01	1.324e+02	1.125e+02
-----Solution composition-----						
Elements	Molality		Moles			
Al	8.264e+01		1.127e-08			
C	4.463e+01		6.087e-09			
Ca	1.496e-21		2.041e-31			
Fe	5.885e-04		8.027e-14			
K	0.000e+00		0.000e+00			
Mg	9.862e-22		1.345e-31			
Na	3.440e-17		4.693e-27			
Si	8.479e-05		1.156e-14			



# CONT.

-----Phase assemblage-----						
Phase	SI	log IAP	log K(T, P)	Moles in assemblage		
				Initial	Final	Delta
Albite	-23.32	-20.67	2.66	1.960e+01	0	-1.960e+01
Analcime	-18.62	-12.57	6.06	1.328e+02	1.532e+02	2.042e+01
Anorthite	-14.69	11.78	26.48	1.725e+01	0	-1.725e+01
CO2(g)	-9.74	-17.57	-7.83	1.000e+01	0	-1.000e+01
Calcite	-14.23	-12.41	1.82	4.009e+01	0	-4.009e+01
Clinoptilolite-Ca	-59.85	-67.37	-7.52	1.000e+01	2.304e+01	1.304e+01
Enstatite	-16.26	-4.97	11.29	3.000e+01	0	-3.000e+01
Hedenbergite	-33.02	-13.49	19.53	1.124e+02	1.198e+02	7.410e+00
Quartz	-4.23	-8.25	-4.03	1.124e+02	0	-1.124e+02
Stilbite	-26.76	-25.95	0.81	1.000e+01	1.000e+01	1.667e-08
Dolomite	-29.17	-26.69	2.47	1.124e+02	1.397e+02	2.733e+01
Kaolinite	0.58	7.30	6.72	2.000e+01	1.487e+01	-5.130e+00
Magnesite	-16.56	-14.29	2.27	1.124e+02	1.151e+02	2.673e+00
Magnetite	-10.51	-0.09	10.42	1.009e+01	9.979e+00	-1.108e-01
Siderite	-19.50	-19.72	-0.22	1.991e+01	1.261e+01	-7.299e+00
-----Solution composition-----						
Elements	Molality		Moles			
Al	4.744e+01		2.202e-08			
C	3.509e-02		1.629e-11			
Ca	6.300e-20		2.925e-29			
Fe	1.299e-07		6.029e-17			
K	0.000e+00		0.000e+00			
Mg	3.915e-22		1.817e-31			
Na	3.873e-20		1.798e-29			
Si	5.545e-05		2.574e-14			

pH	
Iteration 1	7.449
Iteration 2	6.128
Iteration 3	6.128
Iteration 4	5.651
Iteration 5	5.636

# KINETICS

```
KINETICS 1
  Analcime
    -m0 0.174          # 6.3% Analcime, 0.1 mm cylinders
    -m 0.174           # Moles per L pore space
    -parms 6.04 0.1     # m^2/mol Albite, fraction adjusts lab rate to field rate
    -time 84 days

  Analcime
  -start
  1 REM Sverdrup and Warfvinge, 1995, mol m^-2 s^-1
  2 REM PARM(1) = Specific area of Albite m^2/mol Albite
  3 REM PARM(2) = Adjusts lab rate to field rate
  4 REM temp corr: from A&P, p. 162 E (kJ/mol) / R / 2.303 = H in H*(1/T-1/281)
  5 REM Albite parameters
  10 DATA 11.5, 0.5, 4e-6, 0.4, 500e-6, 0.2, 13.7, 0.14, 0.15, 11.8, 0.3
  20 RESTORE 10
  30 READ pk_H, n_H, lim_Al, x_Al, lim_BC, x_BC, pk_H2O, z_Al, z_BC, pk_OH, o_OH
  40 DATA 3500, 2000, 2500, 2000
  50 RESTORE 40
  60 READ e_H, e_H2O, e_OH, e_CO2
  70 pk_CO2 = 13
  80 n_CO2 = 0.6
  100 REM Generic rate follows
  110 dif_temp = 1/TK - 1/281
  120 BC = ACT("Na+") + ACT("K+") + ACT("Mg+2") + ACT("Ca+2")
  130 REM rate by H+
  140 pk_H = pk_H + e_H * dif_temp
  150 rate_H = 10^-pk_H * ACT("H+")^n_H / ((1 + ACT("Al+3") / lim_Al)^x_Al * (1 + BC / lim_BC)^x_BC)
  160 REM rate by hydrolysis
  170 pk_H2O = pk_H2O + e_H2O * dif_temp
  180 rate_H2O = 10^-pk_H2O / ((1 + ACT("Al+3") / lim_Al)^z_Al * (1 + BC / lim_BC)^z_BC)
  190 REM rate by OH-
  200 pk_OH = pk_OH + e_OH * dif_temp
  210 rate_OH = 10^-pk_OH * ACT("OH-")^o_OH
  220 REM rate by CO2
  230 pk_CO2 = pk_CO2 + e_CO2 * dif_temp
  240 rate_CO2 = 10^-pk_CO2 * (SR("CO2(g)"))^n_CO2
  250 rate = rate_H + rate_H2O + rate_OH + rate_CO2
  260 area = PARM(1) * M0 * (M/M0)^0.67
  270 rate = PARM(2) * area * rate * (1-SR("Albite"))
  280 moles = rate * TIME
  290 SAVE moles
  -end
END
```

```
KINETICS 1
  Analcime
    -m0 0.174          # 6.3% Analcime, 0.1 mm cylinders
    -m 0.174           # Moles per L pore space
    -parms 6.04 0.1     # m^2/mol Albite, fraction adjusts lab rate to field rate
    -time 84 days

  Analcime
  -start
  1 REM Sverdrup and Warfvinge, 1995, mol m^-2 s^-1
  2 REM PARM(1) = Specific area of Albite m^2/mol Albite
  3 REM PARM(2) = Adjusts lab rate to field rate
  4 REM temp corr: from A&P, p. 162 E (kJ/mol) / R / 2.303 = H in H*(1/T-1/281)
  5 REM Albite parameters
  10 DATA 11.5, 0.5, 4e-6, 0.4, 500e-6, 0.2, 13.7, 0.14, 0.15, 11.8, 0.3
  20 RESTORE 10
  30 READ pk_H, n_H, lim_Al, x_Al, lim_BC, x_BC, pk_H2O, z_Al, z_BC, pk_OH, o_OH
  40 DATA 3500, 2000, 2500, 2000
  50 RESTORE 40
  60 READ e_H, e_H2O, e_OH, e_CO2
  70 pk_CO2 = 13
  80 n_CO2 = 0.6
  100 REM Generic rate follows
  110 dif_temp = 1/TK - 1/281
  120 BC = ACT("Na+") + ACT("K+") + ACT("Mg+2") + ACT("Ca+2")
  130 REM rate by H+
  140 pk_H = pk_H + e_H * dif_temp
  150 rate_H = 10^-pk_H * ACT("H+")^n_H / ((1 + ACT("Al+3") / lim_Al)^x_Al * (1 + BC / lim_BC)^x_BC)
  160 REM rate by hydrolysis
  170 pk_H2O = pk_H2O + e_H2O * dif_temp
  180 rate_H2O = 10^-pk_H2O / ((1 + ACT("Al+3") / lim_Al)^z_Al * (1 + BC / lim_BC)^z_BC)
  190 REM rate by OH-
  200 pk_OH = pk_OH + e_OH * dif_temp
  210 rate_OH = 10^-pk_OH * ACT("OH-")^o_OH
  220 REM rate by CO2
  230 pk_CO2 = pk_CO2 + e_CO2 * dif_temp
  240 rate_CO2 = 10^-pk_CO2 * (SR("CO2(g)"))^n_CO2
  250 rate = rate_H + rate_H2O + rate_OH + rate_CO2
  260 area = PARM(1) * M0 * (M/M0)^0.67
  270 rate = PARM(2) * area * rate * (1-SR("Albite"))
  280 moles = rate * TIME
  290 SAVE moles
  -end
END
```

# SUMMARY

- Porosity of the basaltic tuff increased predominantly because of the dissolution of primary minerals
  - Primary mineral that contributed most was porosity
  - Continued CO<sub>2</sub>-water-rock interactions will lead to an increasing acidification which will always influence surface area and porosity
  - More research is needed to properly determine sealing capacity of cap rock because analcime was only 6.3% by weight of the basaltic tuff
- pH will decrease and create a more acidic environment with time in the set parameters
- SI's were majority primary minerals
- Kinetics is hard to model 😊



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# THANK YOU

