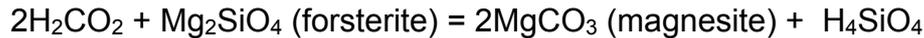


Aqueous Geochemistry Practical 5

Geochemical Modelling of CO₂ sequestration

There is great interest in developing strategies for the geological sequestration of CO₂. The dissolution of CO₂ in high temperature/pressure fluids and the reaction of those fluids with rock might be a way to bury CO₂.

CO₂ can react with primary silicates in reactions such as



However, as CO₂ reacts with primary silicates, a variety of secondary phases (e.g., clay minerals) are also expected to form.

We have two candidate repositories to investigate. One is a granite with the following mineralogy:

Microcline 20 mole %
Quartz 20 mole %
Anorthite 20 mole %
Albite 30 mole %
Biotite (50:50 Annite-Phlogopite) 10%

Assume the granite has a porosity such that there will be 1 kg of porewater for every 10 moles of exposed microcline and that the initial pore fluid (into which the CO₂ is injected) is pure water. The pressure of the pore-fluid will be 300 bar and the temperature will be 150 °C.

The second is a basalt with the following mineralogy:

Forsterite 30 mole %
Fayalite 10 mole %
Anorthite 10 mole %
Albite 5 mole %
Enstatite 45 mole %

Assume the basalt has a porosity such that there will be 1 kg of porewater for every 10 moles of exposed forsterite and that the initial pore fluid (into which the CO₂ is injected) is pure water. The pressure of the pore-fluid will be 300 bar and the temperature will be 150 °C.

Which rock type will make the most effective CO₂ repository? To determine this, we need to simulate how CO₂ will react with the rocks and what alteration phases (e.g., clay minerals, magnesite etc.) will form. The capacity of each reservoir will be determined by how much CO₂ we can add before we get a free CO₂ gas phase. This means that we need to run a reaction progress simulations using a REACTION block in which we add CO₂. We need to include our starting phases and any possible secondary phases in an EQUILIBRIUM_PHASES block.

Here is an example input file that I made up. You will need to edit it for the rock type you are simulating. I'm using the latest version of the PHREEQC database so that I have molar volume data (-vm) from which I can calculate the pressure dependence. However, there are some minerals that are absent in the phreeqc database; I have included a PHASES block to in the input file to augment the database used in the simulation. If you think another mineral might be needed, let me know.

```

DATABASE database/phreeqc.dat
SOLUTION
-temperature 150
EQUILIBRIUM_PHASES
Calcite 0.0 0.0
Magnesite 0.0 0.0
Forsterite 0.0 10.0
Anorthite 0.0 10.0
Talc 0.0 0.0
Pyrophyllite 0.0 0.0
CO2(g) 2.47 0.0 #set target CO2 to be the log Ptot
REACTION_PRESSURE
300
REACTION
    CO2(g)
    50.000 moles in 50 steps

SELECTED_OUTPUT
    -file co2seq.txt
    -reset false
    -pH true
USER_PUNCH
-start
-heading Forsterite CO2 Magnesite Calcite Talc Pyrophyllite
10 punch, EQUI("Forsterite"), EQUI("CO2(g)"),
EQUI("Magnesite"), EQUI("Calcite"), EQUI("Talc"),
EQUI("Pyrophyllite")
-end
PHASES
Forsterite
    Mg2SiO4 +4.0000 H+ = + 1.0000 H4SiO4 + 2.0000 Mg++
    log_k          27.8626
    -delta_H       -205.614          kJ/mol # Calculated
enthalpy of reaction      Forsterite
#   Enthalpy of formation: -520 kcal/mol
    -analytic -7.6195e+001 -1.4013e-002 1.4763e+004
2.5090e+001 -3.0379e+005
#   -Range: 0-300
    -vm 95.76
Magnesite
    MgCO3 +1.0000 H+ = + 1.0000 HCO3- + 1.0000 Mg++
    log_k          2.2936
    -delta_H       -44.4968          kJ/mol # Calculated
enthalpy of reaction      Magnesite
#   Enthalpy of formation: -265.63 kcal/mol

```

```

        -analytic -1.6665e+002 -4.9469e-002 6.4344e+003
6.5506e+001 1.0045e+002
#       -Range: 0-300
        -vm 30.56
Pyrophyllite
Al2Si4O10(OH)2 +6.0000 H+ + 4.000 H2O = + 2.0000
Al+++ + 4.0000 H4SiO4
        log_k           0.4397
        -delta_H        -102.161          kJ/mol # Calculated
enthalpy of reaction      Pyrophyllite
#       Enthalpy of formation: -1345.31 kcal/mol
        -analytic 1.1066e+001 1.2707e-002 1.6417e+004 -
1.9596e+001 -1.8791e+006
#       -Range: 0-300
        -vm 66.63
Illite
K0.6Mg0.25Al1.8Al0.5Si3.5O10(OH)2 +8.0000 H+ + 2.00
H2O = + 0.2500 Mg++ + 0.6000 K+ + 2.3000 Al+++ + 3.5000
H4SiO4
        log_k           9.0260
        -delta_H        -171.764          kJ/mol # Calculated
enthalpy of reaction      Illite
#       Enthalpy of formation: -1394.71 kcal/mol
        -analytic 2.6069e+001 -1.2553e-003 1.3670e+004 -
2.0232e+001 -1.1204e+006
#       -Range: 0-300
        -vm 141.48
Annite
KFe3AlSi3O10(OH)2 +10.0000 H+ = + 1.0000 Al+++ +
1.0000 K+ + 3.0000 Fe++ + 3.0000 H4SiO4
        log_k           29.4693
        -delta_H        -259.964          kJ/mol # Calculated
enthalpy of reaction      Annite
#       Enthalpy of formation: -1232.19 kcal/mol
        -analytic -4.0186e+001 -1.4238e-002 1.8929e+004
7.9859e+000 -8.4343e+005
#       -Range: 0-300
        -vm 152.63
Phlogopite
KAlMg3Si3O10(OH)2 +10.0000 H+ = + 1.0000 Al+++ +
1.0000 K+ + 3.0000 Mg++ + 3.0000 H4SiO4
        log_k           37.4400
        -delta_H        -310.503          kJ/mol # Calculated
enthalpy of reaction      Phlogopite
#       Enthalpy of formation: -1488.07 kcal/mol
        -analytic -8.7730e+001 -1.7253e-002 2.3748e+004
2.4465e+001 -8.9045e+005
#       -Range: 0-300
        -vm 152.33

```

END