SUPPORTING INFORMATION SECTION

CO_2 SEQUESTRATION THROUGH MINERAL CARBONATION OF IRON OXYHYDROXIDES

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Twelve pages with 3 figures and 2 tables.

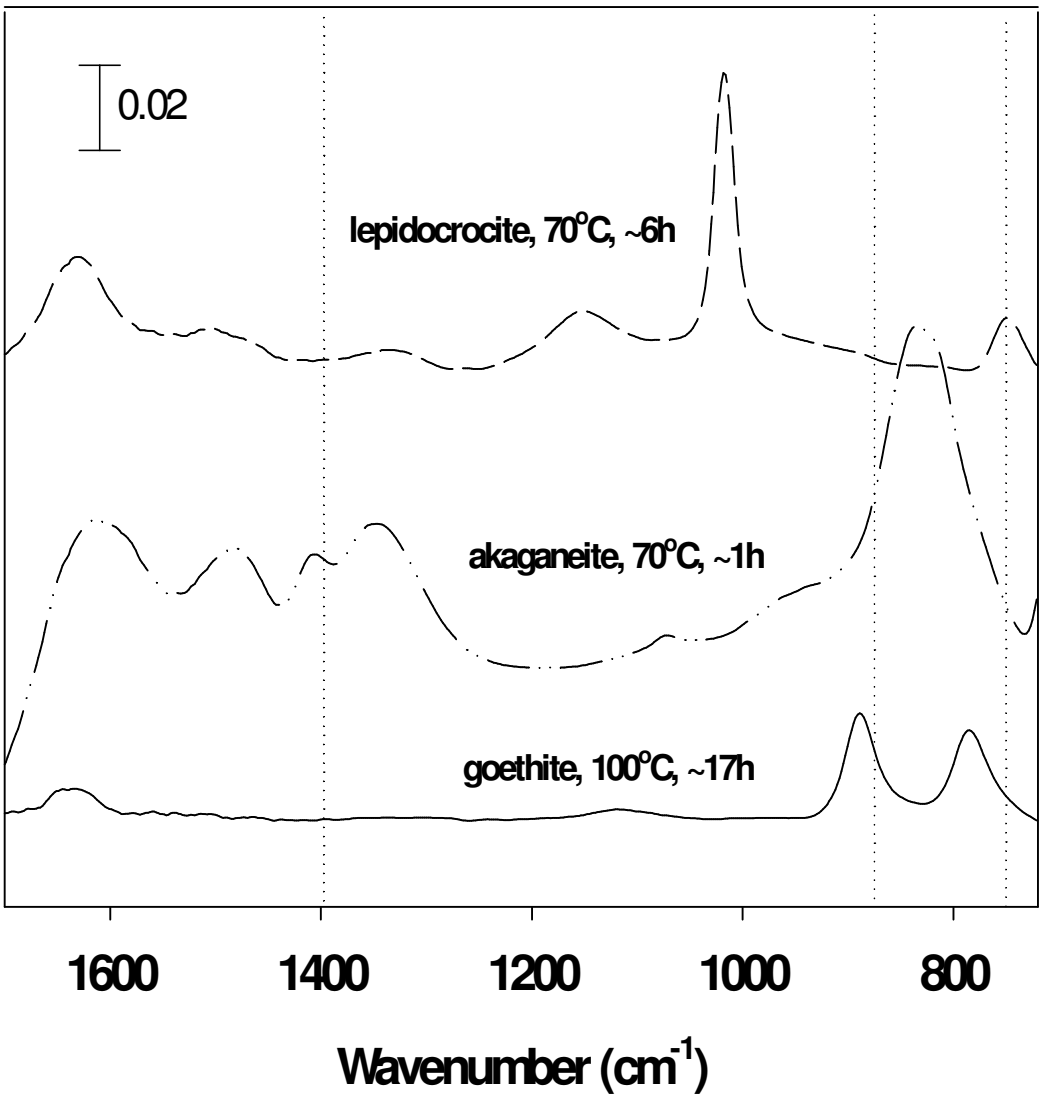

Figure S1. ATR-FTIR control spectra obtained in situ of goethite, akaganeite and lepidocrocite at the respective temperatures under $scCO_2$ in the absence of aqueous sodium sulfide. Vibrational modes associated with siderite modes (shown with dotted lines) were not experimentally observed during reaction time.

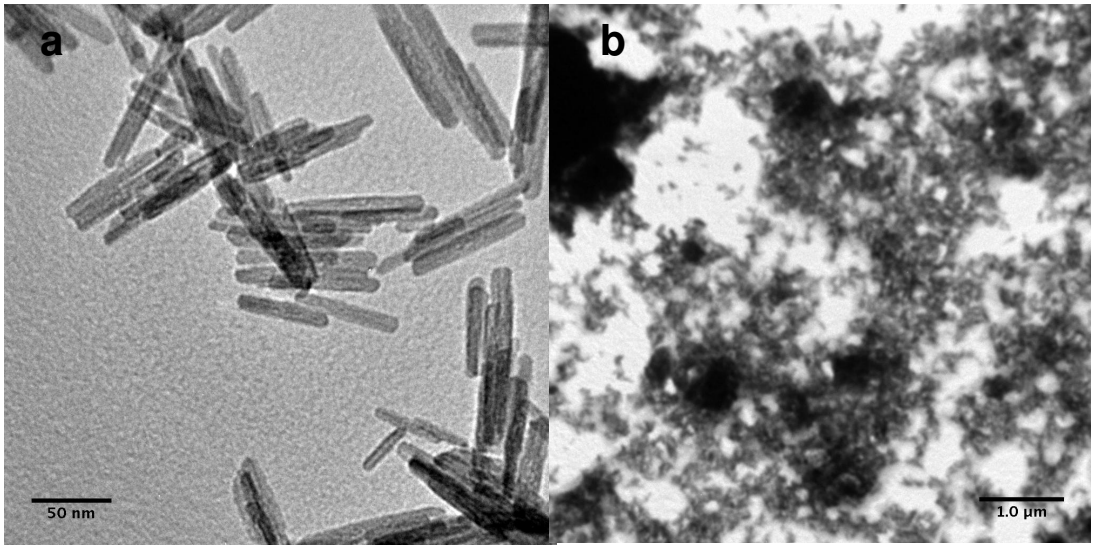

Figure S2. TEM of (a) akaganeite and (b) akaganeite exposed in situ to 83 bar $scCO_2$ and aqueous sulfide (14.5 mM) at 70˚C. Micrograph shows change in particle size and morphology.

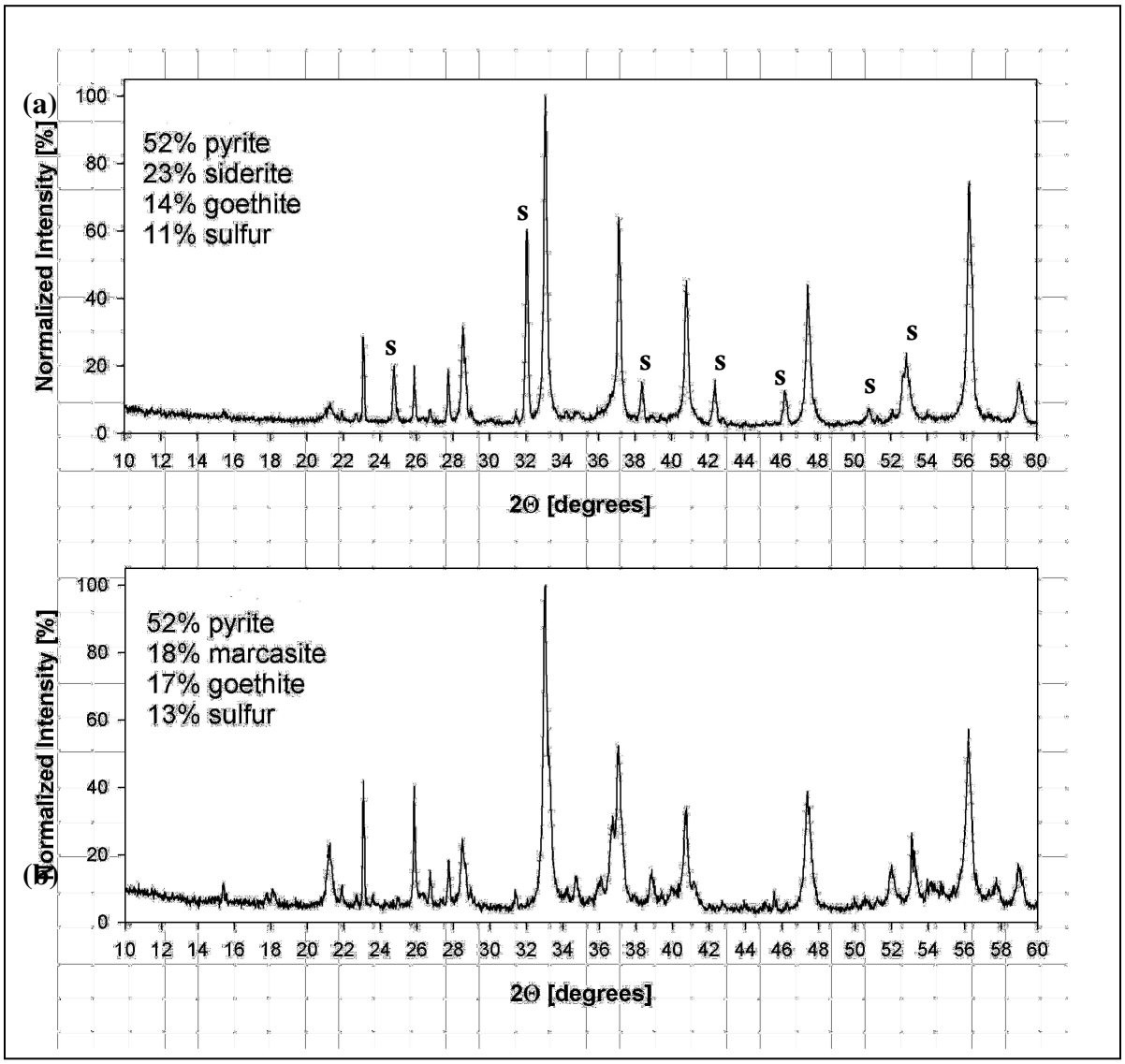

Figure S3. XRD patterns associated with the product formed via the reaction of (a) lepidocrocite with $scCO_2$ and 100 mM aqueous sulfide (3.1% H_2S) at 70˚C and (b) goethite with $scCO_2$ 100 mM aqueous sulfide (6% H_2S) at 100˚C. Quantitative analysis based on Rietveld refinement of the powder diffractogram using Topas Academic (See Table 1 in manuscript). **s**=siderite.

Table S1. PHREEQC output data for a system exposed to 14.5 mM aqueous sodium sulfide at 70°C and 84 bar.

```
Input file: C:\DOCUME~1\Owner\LOCALS~1\Temp\phrq0000.tmp
  Output file: D:\GC\Phreeqc for Na2S\input Na2S.out
Database file: C:\Program Files\Phreeqc\Databases\Phreeqc.dat

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Reading data base.
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      SOLUTION_MASTER_SPECIES
      SOLUTION_SPECIES
      PHASES
      EXCHANGE_MASTER_SPECIES
      EXCHANGE_SPECIES
      SURFACE_MASTER_SPECIES
      SURFACE_SPECIES
      RATES
      END
-----------------------------------
Reading input data for simulation 1.
-----------------------------------

      EQUILIBRIUM_PHASES 1
            co2(g) 1.92 10
                       SOLUTION 1
                       temp      70
                       pH        7 charge
                       pe        4
                       redox     pe
                       units     mmol/kgw
                         Na  29.2
                    S(-2) 14.6
                             density   1
                             water    .01 # kg
-------------------------------------------
Beginning of initial solution calculations.
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Initial solution 1.

-----------------------------Solution composition------------------------------

      Elements           Molality       Moles

      Na                2.920e-02   2.920e-04
      S(-2)             1.460e-02   1.460e-04

----------------------------Description of solution----------------------------

                                       pH  =  10.818      Charge balance
                                       pe  =   4.000
       Specific Conductance (uS/cm, 70 oC) = 9635
                            Density (g/cm3)  =   0.97957
```

```
                       Activity of water  =   0.999
                          Ionic strength  =   3.155e-02
                       Mass of water (kg)  =   1.000e-02
                 Total alkalinity (eq/kg)  =   2.920e-02
                    Total carbon (mol/kg)  =   0.000e+00
                       Total CO2 (mol/kg)  =   0.000e+00
                     Temperature (deg C)  =  70.000
                   Electrical balance (eq)  =  -2.501e-13
 Percent error, 100*(Cat-|An|)/(Cat+|An|)  =  -0.00
                                Iterations  =   9
                                   Total H  = 1.110369e+00
                                   Total O  = 5.551845e-01
```

----------Distribution of species----------

Species	Molality	Activity	Log Molality	Log Activity	Log Gamma
OH-	1.223e-02	1.012e-02	-1.913	-1.995	-0.082
H+	1.765e-11	1.522e-11	-10.753	-10.818	-0.065
H2O	5.551e+01	9.990e-01	1.744	-0.000	0.000
H(0)	2.965e-33				
H2	1.483e-33	1.493e-33	-32.829	-32.826	0.003
Na	2.920e-02				
Na+	2.919e-02	2.441e-02	-1.535	-1.612	-0.078
NaOH	1.051e-05	1.059e-05	-4.978	-4.975	0.003
O(0)	2.781e-14				
O2	1.390e-14	1.400e-14	-13.857	-13.854	0.003
S(-2)	1.460e-02				
HS-	1.223e-02	1.013e-02	-1.912	-1.994	-0.082
S-2	2.365e-03	1.170e-03	-2.626	-2.932	-0.305
H2S	5.726e-07	5.768e-07	-6.242	-6.239	0.003

----------Saturation indices----------

Phase	SI	log IAP	log KT	
H2(g)	-29.51	-32.83	-3.32	H2
H2O(g)	-0.50	-0.00	0.50	H2O
H2S(g)	-4.80	-6.24	-1.44	H2S
O2(g)	-10.76	-13.85	-3.09	O2
Sulfur	19.43	23.40	3.97	S

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Beginning of batch-reaction calculations.
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Reaction step 1.

Using solution 1.
Using pure phase assemblage 1.
```

----------Phase assemblage----------

```
                                                    Moles in assemblage
```

Phase	SI	log IAP	log KT	Initial	Final	Delta
CO2(g)	1.92	0.07	-1.85	1.000e+01	9.988e+00	-1.206e-02

----------Solution composition----------

Elements	Molality	Moles
C	1.206e+00	1.206e-02
Na	2.921e-02	2.920e-04
S	1.460e-02	1.460e-04

----------Description of solution----------

pH	=	4.626	Charge balance
pe	=	-1.777	Adjusted to redox equilibrium
Specific Conductance (uS/cm, 70 oC)	=	5453	
Density (g/cm3)	=	0.98065	
Activity of water	=	0.979	
Ionic strength	=	2.900e-02	
Mass of water (kg)	=	9.997e-03	
Total alkalinity (eq/kg)	=	2.898e-02	
Total CO2 (mol/kg)	=	1.206e+00	
Temperature (deg C)	=	70.000	
Electrical balance (eq)	=	-2.501e-13	
Percent error, 100*(Cat-|An|)/(Cat+|An|)	=	-0.00	
Iterations	=	33	
Total H	=	1.110369e+00	
Total O	=	5.793053e-01	

----------Distribution of species----------

Species	Molality	Activity	Log Molality	Log Activity	Log Gamma
H+	2.734e-05	2.367e-05	-4.563	-4.626	-0.063
OH-	7.651e-09	6.375e-09	-8.116	-8.196	-0.079
H2O	5.551e+01	9.788e-01	1.744	-0.009	0.000
C(-4)	1.154e-04				
CH4	1.154e-04	1.162e-04	-3.938	-3.935	0.003
C(4)	1.206e+00				
CO2	1.177e+00	1.185e+00	0.071	0.074	0.003
HCO3-	2.848e-02	2.410e-02	-1.545	-1.618	-0.073
NaHCO3	3.268e-04	3.290e-04	-3.486	-3.483	0.003
NaCO3-	2.937e-07	2.463e-07	-6.532	-6.609	-0.076
CO3-2	1.480e-07	7.582e-08	-6.830	-7.120	-0.291
H(0)	2.565e-09				
H2	1.282e-09	1.291e-09	-8.892	-8.889	0.003
Na	2.921e-02				
Na+	2.887e-02	2.428e-02	-1.540	-1.615	-0.075
NaHCO3	3.268e-04	3.290e-04	-3.486	-3.483	0.003
NaSO4-	9.865e-06	8.274e-06	-5.006	-5.082	-0.076
NaCO3-	2.937e-07	2.463e-07	-6.532	-6.609	-0.076
NaOH	6.591e-12	6.635e-12	-11.181	-11.178	0.003

O(0)	0.000e+00					
O2		0.000e+00	0.000e+00	-61.748	-61.745	0.003
S(-2)	1.449e-02					
H2S		1.429e-02	1.439e-02	-1.845	-1.842	0.003
HS-		1.950e-04	1.625e-04	-3.710	-3.789	-0.079
S-2		2.385e-11	1.207e-11	-10.623	-10.918	-0.296
S(6)	1.154e-04					
SO4-2		1.051e-04	5.306e-05	-3.978	-4.275	-0.297
NaSO4-		9.865e-06	8.274e-06	-5.006	-5.082	-0.076
HSO4-		4.500e-07	3.774e-07	-6.347	-6.423	-0.076

-------------------------------Saturation indices--------------------------------

Phase	SI	log IAP	log KT	
CH4(g)	-0.75	-3.93	-3.18	CH4
CO2(g)	1.92	0.07	-1.85	CO2
H2(g)	-5.57	-8.89	-3.32	H2
H2O(g)	-0.51	-0.01	0.50	H2O
H2S(g)	-0.41	-1.84	-1.44	H2S
O2(g)	-58.66	-61.75	-3.09	O2
Sulfur	-0.11	3.86	3.97	S

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End of simulation.
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Reading input data for simulation 2.
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End of run.
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Table S2. PHREEQC output data for a system exposed to 100 mM aqueous sodium sulfide at 100˚C and 84 bar.

```
Input file: C:\DOCUME~1\STRONG~1\LOCALS~1\Temp\phrq0000.tmp
  Output file: C:\Documents and Settings\Stronginlab\Desktop\input Na2S.out
Database file: C:\Documents and
Settings\Stronginlab\Desktop\Phreeqc\Databases\Phreeqc.dat

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Reading data base.
------------------

      SOLUTION_MASTER_SPECIES
      SOLUTION_SPECIES
      PHASES
      EXCHANGE_MASTER_SPECIES
      EXCHANGE_SPECIES
      SURFACE_MASTER_SPECIES
      SURFACE_SPECIES
      RATES
      END
-----------------------------------
Reading input data for simulation 1.
-----------------------------------

      EQUILIBRIUM_PHASES 1
            co2(g) 1.90 10
                       SOLUTION 1
                       temp      100
                       pH        7 charge
                       pe        4
                       redox     pe
                       units     mmol/kgw
                         Na  200
                   S(-2) 100
                             density   1
                             water    .01 # kg
-------------------------------------------
Beginning of initial solution calculations.
-------------------------------------------

Initial solution 1.

-----------------------------Solution composition------------------------------

      Elements           Molality       Moles

      Na                2.000e-01   2.000e-03
      S(-2)             1.000e-01   1.000e-03

----------------------------Description of solution----------------------------

                                       pH  =  10.742      Charge balance
                                       pe  =   4.000
       Specific Conductance (uS/cm, 100 oC) = 64503
```

```
                        Density (g/cm3)  =   0.97630
                      Activity of water  =   0.994
                         Ionic strength  =   2.539e-01
                      Mass of water (kg)  =   1.000e-02
                Total alkalinity (eq/kg)  =   2.000e-01
                   Total carbon (mol/kg)  =   0.000e+00
                      Total CO2 (mol/kg)  =   0.000e+00
                     Temperature (deg C)  = 100.000
                  Electrical balance (eq)  =   2.757e-14
 Percent error, 100*(Cat-|An|)/(Cat+|An|)  =   0.00
                              Iterations  =   8
                                 Total H  = 1.111046e+00
                                 Total O  = 5.555228e-01
```

----------Distribution of species----------

Species	Molality	Activity	Log Molality	Log Activity	Log Gamma
OH-	4.602e-02	2.977e-02	-1.337	-1.526	-0.189
H+	2.383e-11	1.813e-11	-10.623	-10.742	-0.119
H2O	5.551e+01	9.941e-01	1.744	-0.003	0.000
H(0) 3.806e-33					
H2	1.903e-33	2.017e-33	-32.721	-32.695	0.025
Na 2.000e-01					
Na+	2.000e-01	1.382e-01	-0.699	-0.860	-0.160
NaOH	4.722e-05	5.007e-05	-4.326	-4.300	0.025
O(0) 1.035e-07					
O2	5.174e-08	5.485e-08	-7.286	-7.261	0.025
S(-2) 1.000e-01					
S-2	5.394e-02	1.203e-02	-1.268	-1.920	-0.652
HS-	4.606e-02	2.980e-02	-1.337	-1.526	-0.189
H2S	1.690e-06	1.791e-06	-5.772	-5.747	0.025

----------Saturation indices----------

Phase	SI	log IAP	log KT	
H2(g)	-29.29	-32.70	-3.41	H2
H2O(g)	0.04	-0.00	-0.04	H2O
H2S(g)	-4.08	-5.75	-1.67	H2S
O2(g)	-4.15	-7.26	-3.11	O2
Sulfur	20.25	23.74	3.48	S

```
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Beginning of batch-reaction calculations.
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Reaction step 1.

Using solution 1.
Using pure phase assemblage 1.
```

----------Phase assemblage----------

Phase	SI	log IAP	log KT	Moles in assemblage: Initial	Final	Delta
CO2(g)	1.90	-0.08	-1.98	1.000e+01	9.990e+00	-9.767e-03

----------Solution composition----------

Elements	Molality	Moles
C	9.792e-01	9.767e-03
Na	2.005e-01	2.000e-03
S	1.002e-01	1.000e-03

----------Description of solution----------

pH = 5.600 Charge balance
pe = -3.356 Adjusted to redox equilibrium
Specific Conductance (uS/cm, 100 oC) = 47020
Density (g/cm3) = 0.98388
Activity of water = 0.978
Ionic strength = 1.929e-01
Mass of water (kg) = 9.975e-03
Total alkalinity (eq/kg) = 1.946e-01
Total CO2 (mol/kg) = 9.762e-01
Temperature (deg C) = 100.000
Electrical balance (eq) = 2.759e-14
Percent error, 100*(Cat-|An|)/(Cat+|An|) = 0.00
Iterations = 27
Total H = 1.111046e+00
Total O = 5.750570e-01

----------Distribution of species----------

Species	Molality	Activity	Log Molality	Log Activity	Log Gamma
H+	3.256e-06	2.514e-06	-5.487	-5.600	-0.112
OH-	3.148e-07	2.113e-07	-6.502	-6.675	-0.173
H2O	5.551e+01	9.784e-01	1.744	-0.009	0.000
C(-4)	2.951e-03				
CH4	2.951e-03	3.085e-03	-2.530	-2.511	0.019
C(4)	9.762e-01				
CO2	7.972e-01	8.334e-01	-0.098	-0.079	0.019
HCO3-	1.700e-01	1.215e-01	-0.770	-0.916	-0.146
NaHCO3	8.806e-03	9.206e-03	-2.055	-2.036	0.019
NaCO3-	2.455e-04	1.743e-04	-3.610	-3.759	-0.149
CO3-2	1.296e-05	3.379e-06	-4.887	-5.471	-0.584
H(0)	3.828e-08				
H2	1.914e-08	2.001e-08	-7.718	-7.699	0.019
Na	2.005e-01				
Na+	1.907e-01	1.348e-01	-0.720	-0.870	-0.151
NaHCO3	8.806e-03	9.206e-03	-2.055	-2.036	0.019
NaSO4-	7.492e-04	5.317e-04	-3.125	-3.274	-0.149
NaCO3-	2.455e-04	1.743e-04	-3.610	-3.759	-0.149

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   NaOH                 3.316e-10   3.466e-10    -9.479    -9.460     0.019
O(0)            0.000e+00
   O2                   0.000e+00   0.000e+00   -57.287   -57.268     0.019
S(-2)           9.730e-02
   H2S                  8.198e-02   8.570e-02    -1.086    -1.067     0.019
   HS-                  1.532e-02   1.028e-02    -1.815    -1.988    -0.173
   S-2                  1.202e-07   2.994e-08    -6.920    -7.524    -0.604
S(6)            2.951e-03
   SO4-2                2.200e-03   5.382e-04    -2.658    -3.269    -0.612
   NaSO4-               7.492e-04   5.317e-04    -3.125    -3.274    -0.149
   HSO4-                1.366e-06   9.696e-07    -5.864    -6.013    -0.149

------------------------------Saturation indices-------------------------------

      Phase               SI log IAP  log KT

      CH4(g)            0.85   -2.51   -3.36  CH4
      CO2(g)            1.90   -0.08   -1.98  CO2
      H2(g)            -4.29   -7.70   -3.41  H2
      H2O(g)            0.03   -0.01   -0.04  H2O
      H2S(g)            0.60   -1.07   -1.67  H2S
      O2(g)           -54.15  -57.27   -3.11  O2
      Sulfur           -0.06    3.42    3.48  S

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End of simulation.
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Reading input data for simulation 2.
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End of run.
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