CarbFix Report

PHREEQC mineral dissolution kinetics database

Date: 30.11.2014

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<td>151</td>
<td>Tremolite</td>
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<td>155</td>
<td>Variscite</td>
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<td>Witherite</td>
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<td>162</td>
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Prediction is hard - Especially about the Future (Attributed to Yogi Berra)

Introduction

The kinetic database is established from a thorough literature review of the existing data on precipitation and dissolution rates. These datasets are assessed and the values for the reaction rate constant (A), the activation energy (Ea), the exponent on the activity of element i (n_i) for H, Mg, Si, Na and K, selected when applicable and introduced into the database file. This file is organized by mineral, glass phase and the pH spaces where the experimental measurements were collected, as can be seen in Table 1 where an excerpt from the current database is shown. The reference from which the data was collected is specified in the last column.

After the initial development and definition of the minerals and glass phases the database is planned to become a collaborative effort, with each team able to input their results into the database. Therefore it has been organized in an excel spreadsheet to increase the accessibility and modifiability of its content.

The software unit will be a stand-alone program coupled with PHREEQC, using the data from the geochemical software and the kinetic database to produce dissolution and precipitation rates. Currently under development using it allows the user to determine dissolution or precipitation rates of the phases defined in the databases. The input from the user is a two staged process where first the conditions and phase dissolved or precipitated are defined in PHREEQC, then the kinetic software is accessed and generates the dissolution or precipitation rates of the specified phase in the previously defined conditions.
References


Mineral Description

Albite

Feldspars are amongst the most studied silicate minerals from the point of view of mineral dissolution. Dissolution rates of albite (NaAlSi$_3$O$_8$) from 25 °C and up to 225 °C, and various pH have been published by Lagache (1965), Holdren and Berner (1979), Chou and Wollast (1984, 1985), Holdren and Spayer (1987), Knauss and Wolery (1987), Hellmann et al. (1990), Burch et al. (1991, 1993), Casey et al. (1991), Rose (1991) Hellmann (1994, 1995), Chen and Brantley (1997) and Hellmann and Tisserand (2006). The rates are presented for the different temperatures as a function of pH in Figure A1. Oelkers et al. (1994) reported steady state dissolution rates of albite as a function of chemical affinity and thus are not included here. The rates gathered by Lagache at 200 °C were removed from the dataset because stoichiometric metal release was not observed in this experiment.

Based on the theoretical framework laid by Oelkers (2001) for the dissolution of multi-oxides minerals steady-state albite dissolution rates were fitted to:

\[ r = A_A \exp \left( - \frac{E_A}{RT} \right) \left( \frac{a_{H^+}}{a_{Al^{3+}}} \right)^{1/8} \]  

(A-1)

where \( r \) signifies the albite steady state dissolution rate, \( A_A \) refers to a pre-exponential factor equal to 2.38x10$^{-4}$ mol/cm$^2$/sec$^1$, \( E_A \) designates an activation energy equal to 66.5 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. \( A_A \) and \( E_A \) were determined from an Arrhenius plot of the data obtained at pH 4 by Chen and Brantley (1997), Chou and Wollast (1985), and Hellmann (1994). The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure A1. Rates calculated using Equation A-1 described 75 of 95 reported rates within 0.8 log units with an average uncertainty of 0.2 log units.
References


Table A1: Summary of experimentally measured albite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cleavelandite</td>
<td>80</td>
<td>3, 8.8</td>
<td>Burch et al. (1991)</td>
</tr>
<tr>
<td>Cleavelandite</td>
<td>80</td>
<td>8.8</td>
<td>Burch et al. (1993)</td>
</tr>
<tr>
<td>Albite</td>
<td>25</td>
<td>2</td>
<td>Casey (1991)</td>
</tr>
<tr>
<td>Cleavelandite</td>
<td>25</td>
<td>1–12.5</td>
<td>Chou and Wollast (1985)</td>
</tr>
<tr>
<td>Albite An2</td>
<td>5-90</td>
<td>1, 4.5</td>
<td>Chen and Brantley (1997)</td>
</tr>
<tr>
<td>Albite</td>
<td>225</td>
<td>0.8-8</td>
<td>Hellmann et al. (1990)</td>
</tr>
<tr>
<td>Albite</td>
<td>100-300</td>
<td>2-10</td>
<td>Hellmann (1994)</td>
</tr>
<tr>
<td>Albite</td>
<td>150</td>
<td>8.8</td>
<td>Hellmann and Tisserand (2006)</td>
</tr>
<tr>
<td>Albite, Ab 99.07</td>
<td>n.c.</td>
<td>n.c.</td>
<td>Holdren and Berner (1978)</td>
</tr>
<tr>
<td>Cleavelandite</td>
<td>n.c.</td>
<td>3</td>
<td>Holdren and Speyer (1987)</td>
</tr>
<tr>
<td>Albite, Ab 99.07</td>
<td>25, 70</td>
<td>1.39-11.75</td>
<td>Knauss and Wolery (1986)</td>
</tr>
<tr>
<td>Albite</td>
<td>25</td>
<td>1.4</td>
<td>Rose (1991)</td>
</tr>
<tr>
<td>Albite An2</td>
<td>25</td>
<td>3.32</td>
<td>Stillings et al. (1996)</td>
</tr>
<tr>
<td>Albite</td>
<td>22</td>
<td></td>
<td>Welch and Ullman (1996)</td>
</tr>
<tr>
<td>Albite</td>
<td>25</td>
<td>1-12</td>
<td>Wollast and Chou (1985)</td>
</tr>
</tbody>
</table>
Figure A1: Summary of experimentally measured albite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the various dashed lines.
Almandine

The dissolution rates of the iron alumina garnet almandine \((\text{Fe}_3\text{Al}_2\text{Si}_3\text{O}_{12})\) was reported by Nickel (1973) and Schott and Petit (1987) at 25 °C from pH 0.2 to 10.5. Sverdrup (1990) alludes to the existence of additional data at 8 °C but does not list these. In the absence of additional data to improve existing data fits, these we have adopted the empirical pH power function fit of Palandri and Kharaka (2004) in accord with:

\[
\begin{align*}
  r_{H^+} &= A_A \cdot a_{H^+}^{1} \cdot \exp\left(\frac{-E_A}{R \cdot T}\right) \\
  r_{H_2O} &= A_B \cdot \exp\left(\frac{-E_B}{R \cdot T}\right) \\
  r_{OH^-} &= A_C \cdot a_{OH^-}^{0.35} \cdot \exp\left(\frac{-E_C}{R \cdot T}\right) \\
  r_{tt} &= r_{H^+} + r_{H_2O} + r_{OH^-}
\end{align*}
\]

where \(r_{H^+}\), \(r_{H_2O}\) and \(r_{OH^-}\) signifies the almandine forward dissolution rate in acidic, neutral and basic solutions and \(r_{tt}\) the total dissolution rate, \(A_A\), \(A_B\) and \(A_C\) refer to pre-exponential factors equal to \(2.19 \times 10^7\), \(3.02 \times 10^5\) and \(8.13 \times 10^{-12}\) mol/cm²/s respectively, \(E_A\), \(E_B\), \(E_C\) designates an activation energy equal to 94.4, 103.8 and 37.8 kJ/mol, \(R\) represents the gas constant, and \(T\) denotes absolute temperature. As was the case for andradite in the absence of additional and more extensive data the Palandri and Kharaka fit has been adopted in this study. It should be noted that these activation energies are based on the fit of rates obtained at 8 and 25 °C, and the rate data at 8 °C were never directly reported. As the rate data reported by Sverdrup (1990) for andradite and almandine are identical the activation energies are the same. As such the degree to which these activation energies provide accurate rate estimates at substantially higher temperatures is questionable. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Alm1. Rates calculated using Equation Alm-1 described 8 of 9 reported rates within 0.8 log units.
References


Table Alm1: Summary of experimentally measured almandine steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Almandine</td>
<td>25</td>
<td>0.2-11</td>
<td>Nickel (1973)</td>
</tr>
<tr>
<td>Almandine</td>
<td>25</td>
<td>3-7</td>
<td>Schott and Petit (1987)</td>
</tr>
</tbody>
</table>

Figure Alm1: Summary of experimentally measured almandine steady-state dissolution rates reported in the literature as a function of pH. The dotted line represents the results of the model.
**Andesine / Labradorite**

Andesine \((\text{Na}_0.7\text{Ca}_0.\text{Al}_1.3\text{Si}_2.7\text{O}_8)\) and labradorite are two feldspars of the plagioclase solid solution, they are defined as having an anorthite percentage \((\frac{\text{Ca}}{\text{Ca}+\text{Na}})\) between 30 and 50% for andesine and 50 and 70% for labradorite. Dissolution rates of plagioclase at 25 °C and pH 3 have been published first by Tsuzuki and Suzuki in 1980 who investigated the dissolution kinetics at 230 to 245 C in acidic solutions. Holdren and Spayer followed in 1987 at 25 °C and pH 3, then Casey et al. (1991) investigated the dissolution kinetics of andesine at pH 2. They were followed by a kinetic study by Oxburgh et al. who dissolved andesine between pH 3 and 7, then Stillings and Brantley (1995) dissolved labradorite at pH 3 ± 0.1. In 1996 Stillings et al. studied the dissolution kinetics of andesine between pH 3 and 7.5 and finally Taylor et al. in 2000 dissolved labradorite at pH 3.1. The rates are presented for the different temperatures as a function of pH in Figure And1.

Based on the theoretical framework laid by Oelkers (2001) for the dissolution of multi-oxides minerals steady-state andesine dissolution rates were fitted to:

\[
r = A_A \exp \left( - \frac{E_A}{RT} \right) \left( \frac{a_{\text{Al}^3+}}{a_{\text{Al}^4+}} \right)^{1/8}
\]

(And-1)

where \(r\) signifies the andesine/labradorite steady-state dissolution rate, \(A_A\) refers to a pre-exponential factor equal to 2.9x10⁻⁵ mol/cm²/sec², \(E_A\) designates an activation energy equal to 56 kJ/mol, \(R\) represents the gas constant, and \(T\) denotes absolute temperature. \(E_A\) was assumed to be of similar values as the other feldspar and therefore \(A_A\) was recalculated to fit the data. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure And1. Rates calculated using Equation And-1 described 42 of 46 reported rates within 0.8 log units.
References


Table And1: Summary of experimentally measured andesine steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
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<tr>
<td>Andesine</td>
<td>25</td>
<td>3, 8.8</td>
<td>Casey et al. (1991)</td>
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<td>Andesine</td>
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<td>3</td>
<td>Holdren and Speyer (1987)</td>
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<td>Andesine, An46</td>
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<td>3.1 – 7.3</td>
<td>Oxburgh et al. (1994)</td>
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<td>Siegel and Pfannkuch (1984)</td>
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<td>Andesine, An30</td>
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<td>3.08</td>
<td>Swoboda-Colberg and Drever (1993)</td>
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<td>Labradorite</td>
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<td>Taylor et al. (2000)</td>
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<td>0.4 – 1.7</td>
<td>Tzuzuki and Suzuki (1980)</td>
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<td>Andesine, An49</td>
<td>22</td>
<td>3.1 – 9</td>
<td>Welch and Ullman (1996)</td>
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</table>

Figure And1: Summary of experimentally measured andesine steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the various dashed lines.
**Andradite**

The iron calcium-iron garnet andradite \((\text{Ca}_3\text{Fe}_2\text{Si}_3\text{O}_{12})\) dissolution was studied by Sverdrup (1990) at 25 °C from pH 4 to 4.6. The data was fitted by Palandri and Kharaka (2004) to the following equation:

\[
r_{H^+} = A_A \cdot a_{H^+}^1 \cdot \exp \left( \frac{-E_A}{R \cdot T} \right)
\]

\[
r_{H_2O} = A_B \cdot \exp \left( \frac{-E_B}{R \cdot T} \right)
\]

\[
r_{tt} = r_{H^+} + r_{H_2O}
\]

where \(r_{H^+}\) and \(r_{H_2O}\) signifies the almandine steady-state dissolution rate in acidic, neutral and basic solutions and \(r_{tt}\) the total dissolution rate, \(A_A\) and \(A_B\) refer to pre-exponential factors equal to 2.19x10^7 and 3.02x10^5 mol/cm²/s respectively, \(E_A\) and \(E_B\) designates an activation energy equal to 94.4 and 103.8 kJ/mol, \(R\) represents the gas constant, and \(T\) denotes absolute temperature. In the absence of additional and more extensive data the Palandri and Kharaka fit has been adopted in this study. It should be noted that the activation energies of this rate equation are based on the fit of rates obtained at 8 and 25 °C, and the rate data at 8 °C were never directly reported. As such the degree to which these activation energies provide accurate rate estimates at substantially higher temperatures is questionable. Note that as the rate data reported by Sverdrup (1990) for andradite and almandine are identical the activation energies are the same. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Adr1. Rates calculated using Equation Adr-1 described 2 of 2 reported rates within 0.8 log units.

**References**


Table Adr1: Summary of experimentally measured andradite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Andradite</td>
<td>25</td>
<td>4, 4.6</td>
<td>Sverdrup (1990)</td>
</tr>
</tbody>
</table>

Figure Adr1: Summary of experimentally measured andradite steady-state dissolution rates reported in the literature as a function of pH. The dotted line represents the results of the model.

![Graph showing the relationship between pH and log r+ (mol/cm²/s)](image-url)
Anglesite

The lead sulfate anglesite (PbSO₄) was studied by Dove and Czank (1995) at temperatures ranging from 25 to 61 °C and pH ranging from 1.96 to 12.05. Anglesite dissolution kinetics were also quantified by De Giudici et al. (2005) at 25 °C and pH 1.2 and 2.87. In the absence of sufficient data to characterize its dissolution mechanism we have adopted the fit of Palandri and Kharaka (2004) given by:

\[ r_{H^+} = A_A \cdot a_{H^+}^{0.298} \cdot \exp \left( \frac{-E_A}{RT} \right) \]

\[ r_{H_2O} = A_A \cdot \exp \left( \frac{-E_B}{RT} \right) \]

\[ r_+ = r_{H^+} + r_{H_2O} \]

where \( r_{H^+} \) and \( r_{H_2O} \) signifies the anglesite steady-state dissolution rate in acidic, neutral and basic solutions and \( r_+ \) the total dissolution rate, \( A_A \) and \( A_B \) refer to pre-exponential factors equal to 3.95x10⁻⁵ and 4.55x10⁻⁶ mol/cm²/s respectively, \( E_A \) designates an activation energy equal to 31.3 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Ang1. Rates calculated using Equation Ang-1 describes all reported rates within 0.8 log units with an average uncertainty of 0.2 log units.

References


Table Ang1: Summary of experimentally measured anglesite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anglesite</td>
<td>25</td>
<td>1.2 – 2.87</td>
<td>De Giudici et al. (2005)</td>
</tr>
<tr>
<td>Anglesite</td>
<td>25-61</td>
<td>1.96 – 12.0.5</td>
<td>Dove and Czank (1995)</td>
</tr>
</tbody>
</table>

Figure Ang1: Summary of experimentally measured anglesite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the various dashed lines.

De Giudici et al. (2005), 25 C
Dove and Czank (1995), 25 C
Dove and Czank (1995), 50 C
Model 25 C (Palandri and Kharaka, 2004)
Model 50 C (Palandri and Kharaka, 2004)
Anhydrite

There is relatively little dissolution rate data available for the anhydrous calcium sulphate anhydrite (CaSO₄). Barton and Wilde (1971) reported rates at 25 °C and pH 6, Dove and Czank (1995) reported rates at 50 °C and pH 5.7 and Jeschke and Dreybrodt (2001, 2002) reported rates at 10 °C and pH 6. The rates are presented for the different temperatures as a function of pH in Figure Anh1. Because rates have been reported for one pH at each temperature these data were fit to the pH independent Arrhenius equation:

\[ r_\text{a} = A_\text{A} \cdot \exp \left( -\frac{E_A}{R \cdot T} \right) \]

where \( r_\text{a} \) signifies the anhydrite steady-state dissolution rate, \( A_\text{A} \) refers to a pre-exponential factor equal to 24.4 mol/cm²/s, \( E_A \) designates an activation energy equal to 52.0 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. The \( E_A \) and \( A_\text{A} \) were recalculated with an Arrhenius fit of the existing data at pH 6±0.3. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Anh1. Rates calculated using Equation Anh-1 described 4 of 4 reported rates within 0.8 log units with an average uncertainty of 0.2 log units.

References


Table Anh1: Summary of experimentally measured anhydrite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anhydrite</td>
<td>25</td>
<td>6</td>
<td>Barton and Wilde (1971)</td>
</tr>
<tr>
<td>Anhydrite</td>
<td>50</td>
<td>5.7</td>
<td>Dove and Czank (1995)</td>
</tr>
<tr>
<td>Anhydrite</td>
<td>10</td>
<td>6</td>
<td>Jeschke and Dreybrodt (2002)</td>
</tr>
</tbody>
</table>

Figure Ang1: Summary of experimentally measured anhydrite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the various dashed lines.
Anorthite

The pure calcium endmember of the plagioclase feldspar solid solution, anorthite, was studied at 25 °C and pH of 2 and 5.6 and Fleer in 1982, then Holdren and Spayer as part of their series on feldspar measured the dissolution rate of anorthite at pH 3. Casey et al. (1991) measured the dissolution of anorthite at pH 2 and following suite Amrhein and Suarez studied the anorthite dissolution kinetics at pH ranging from 5 to 8. Oelkers and Schott (1995) investigated the feldspar dissolution mechanism by measuring the dissolution rate of anorthite at temperatures ranging from 45 to 95 and pH range of 2.4 to 3.2. Finally Hodson et al. (2003) measured the dissolution of Anorthite at 25 C and pH 2.6. The rates are presented for the different temperatures as a function of pH in Figure Ano1.

Those results were fitted to the dissolution equation of feldspar determined by Oelkers and Schott (1995):

\[
\begin{align*}
  r &= A_A (1 - \exp(-E_A / 3RT)) \left( \frac{a_{H^+}^{1/3}}{a_{H^+}} \right)^{1/3} \\
  \text{(Ano-1)}
\end{align*}
\]

where \( r \) signifies the andesine/labradorite steady-state dissolution rate, \( A_A \) refers to a pre-exponential factor equal to 2.77x10^{-13} \text{ mol/cm}^2/\text{sec}, \( E_A \) designates an activation energy equal to 18 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. \( E_A \) was assumed to be of similar values as the other feldspar and therefore \( A_A \) was recalculated to fit the data. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Ano1. Rates calculated using Equation Ano-1 described 39 of 46 reported rates at 25 C within 0.8 log units with an average uncertainty of 0.2 log units. The data reported by Fleer (1982) is on par with values measured at 95 °C and thus were excluded from the model.
References


Table Ano1: Summary of experimentally measured anorthite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anorthite, An 93</td>
<td>25</td>
<td>4-8</td>
<td>Amrhein and Suarez (1992)</td>
</tr>
<tr>
<td>Anorthite</td>
<td>25</td>
<td>2-9</td>
<td>Berg and Banwart (2000)</td>
</tr>
<tr>
<td>Anorthite</td>
<td>25</td>
<td>3, 8.8</td>
<td>Casey et al. (1991)</td>
</tr>
<tr>
<td>Anorthite</td>
<td>25</td>
<td>2, 5.5</td>
<td>Fleer (1982)</td>
</tr>
<tr>
<td>Anorthite</td>
<td>25</td>
<td>2.6</td>
<td>Hodson et al. (2003)</td>
</tr>
<tr>
<td>Anorthite</td>
<td>25</td>
<td>3</td>
<td>Holdren and Speyer (1987)</td>
</tr>
<tr>
<td>Microcline</td>
<td>25</td>
<td>5</td>
<td>Lasaga (1998)</td>
</tr>
<tr>
<td>Anorthite, An96</td>
<td>45, 95</td>
<td>2.4-3.2</td>
<td>Oelkers and Schott (1995)</td>
</tr>
<tr>
<td>Anorthite</td>
<td>25</td>
<td>2.96 – 7.65</td>
<td>Stillings et al. (1996)</td>
</tr>
<tr>
<td>Anorthite</td>
<td>25</td>
<td>3.08</td>
<td>Taylor et al. (2000)</td>
</tr>
</tbody>
</table>
Figure Ano1: Summary of experimentally measured anorthite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the various dashed lines.
**Anthophyllite**

Dissolution rates of the amphibole anthophyllite (Mg$_7$Si$_8$O$_{22}$(OH)$_2$) from 25 to 90 °C and pH ranging from 1 to 3.7 have been reported by Chen and Brantley (1998) and at 22 °C with pH ranging from 2 to 9 by Mast and Drever (1987). The values reported by Mast and Drever (1987) are significantly higher than corresponding values presented by Chen and Brantley (1998). As discussed by Chen and Brantley, the difference in rates can be attributed to a non-stoichiometric dissolution. The rate data from Mast and Drever (1987) was, therefore, not included in the fit. The rates are presented as a function of pH in Figure An1.

Following the theoretical framework of Oelkers (2001) for the dissolution of multi-oxides minerals steady-state anthophyllite dissolution rates were fit to:

$$ r_* = A_A \cdot \exp \left( \frac{-E_A}{RT} \right) \left( \frac{a_{H^+}^2}{a_{Mg^{2+}}} \right)^n $$

where $r_*$ signifies the anthophyllite steady state dissolution rate, $A_A$ refers to a pre-exponential factor equal to 7.54x10$^{-8}$ mol/cm²/s, $E_A$ designates an activation energy equal to 55.8 kJ/mol, $R$ represents the gas constant, $T$ denotes absolute temperature and $n$ signifies a stoichiometric coefficient equal to 0.125. $A_A$ and $E_A$ were determined from an Arrhenius plot of the data obtained at pH 2 by Chen and Brentley (1998). $n$ was obtained from a fit of the reported anthophyllite rate data and those of tremolite; tremolite has an almost identical structure to anthophyllite with 2 of its Mg replaced by Ca. As such, it is anticipated that these two minerals would have similar dissolution mechanisms. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure An1. Rates calculated using Equation An-1 described 8 of 8 reported rates within 0.8 log units.

**References**


**Table and Figure**

**Table An1**: Summary of experimentally measured anthophyllite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anthophyllite</td>
<td>25-90</td>
<td>1-3.7</td>
<td>Chen and Brantley (1998)</td>
</tr>
<tr>
<td>Anthophyllite</td>
<td>22</td>
<td>2-9</td>
<td>Mast and Drever (1987)</td>
</tr>
</tbody>
</table>

**Figure An1**: Summary of experimentally measured anthophyllite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the various dashed lines.
Apatite (fluorapatite)

The phosphate mineral apatite has been studied at 25 °C in a limited set of studies. Three studies were reported in Brantley et al. (2008), Guidry and Mackenzy (2003) looked at the dissolution of apatite at pH ranging from 2 to 8.5, Valsami-Jones et al. (1998) studied the dissolution kinetics of apatite between pH 4.2 and 7 and finally Welch and al. (2002) between pH 2.4 and 7.5. In addition Kohler et al. (2005) performed dissolution experiments on apatite at 25 °C from pH 3.01 to 8.9, Chairat et al. studied the dissolution kinetics of apatite at 25 °C and pH between 3 and 11.6 and Harouiya measured the dissolution rate of apatite at 25 °C in acidic solutions ranging from 1.08 to 5.6. Fig Ap1 summarized the published rates at 25 °C and Table Ap1 sum up the conditions under which those rates were obtained.

The acidic dissolution mechanism was defined by Harouiya et al. (2007) as:

\[ r = A_A \cdot a_{H^+}^n \cdot \exp\left(-\frac{E_A}{\sigma RT}\right) \]

where \( A_A \) stands for a rate constant equal to 4x10^-3 mol/cm²/s, \( a_{H^+} \) denotes the activity of the aqueous \( H^+ \), \( n \) designates a reaction order 0.6, \( E_A \) symbolize an activation energy equal to 46 kJ/mol, \( \sigma \) represents the Temkin’s coefficient equal to 5, \( R \) stands for the gas constant and \( T \) represent the absolute temperature.

The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Apa1. Rates calculated using Equation Apa-1 described 66 of 74 reported rates within 0.8 log units with an average uncertainty of 0.2 log units. The data acquired at high pH indicates a flattening of the rates not accounted for in Harouiya’s equation.

References


### Table and Figure

Table Ap1: Summary of experimental conditions for the various apatite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Apatite</td>
<td>25</td>
<td>3-11.6</td>
<td>Chaïrat et al. (2007)</td>
</tr>
<tr>
<td>Apatite</td>
<td>5-50</td>
<td>1.08-5.6</td>
<td>Harouiya et al. (2007)</td>
</tr>
<tr>
<td>Apatite</td>
<td>25</td>
<td>3.01-8.9</td>
<td>Kohler et al. (2005)</td>
</tr>
<tr>
<td>Apatite</td>
<td>25</td>
<td>4.6-7</td>
<td>Valsami-Jones et al. (1998)</td>
</tr>
<tr>
<td>Apatite</td>
<td>25</td>
<td>2.4-7.5</td>
<td>Welch et al. (2002)</td>
</tr>
</tbody>
</table>

Figure Ap1: Summary of experimentally measured apatite steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dashed line.
Apatite (hydroxylapatite)

Guidry and Mackenzy (2003)
Valsami-Jones et al. (1998)
Welch et al. (2002)
Harouiya et al. (2007)
Chaïrat et al. (2007)
Kohler et al. (2005)
Model

$25 ^\circ C$

$\log r_+ \text{ (mol/cm}^2\text{s)}$ vs pH

Guidry and Mackenzy (2003)
Valsami-Jones et al. (1998)
Welch et al. (2002)
Harouiya et al. (2007)
Chaïrat et al. (2007)
Kohler et al. (2005)
Model

$25 ^\circ C$

$\log r_+ \text{ (mol/cm}^2\text{s)}$ vs pH

Guidry and Mackenzy (2003)
Valsami-Jones et al. (1998)
Welch et al. (2002)
Harouiya et al. (2007)
Chaïrat et al. (2007)
Kohler et al. (2005)
Model

$25 ^\circ C$

$\log r_+ \text{ (mol/cm}^2\text{s)}$ vs pH

Guidry and Mackenzy (2003)
Valsami-Jones et al. (1998)
Welch et al. (2002)
Harouiya et al. (2007)
Chaïrat et al. (2007)
Kohler et al. (2005)
Model
Aragonite

Morse et al. (1979) studied the dissolution rate of aragonite at 25 °C and pH 7 as a function of the saturation state. Chou et al. (1989) measured the dissolution rate of aragonite at 25 °C and pH 4-10. In 1995 Gutjahr et al. studied the dissolution kinetics of aragonite as a function of temperature, from 20 to 70 °C and for pH values of 7.81 and 9.09. Finally Cubillas et al. (2005) investigated the dissolution rates of mussels, clam, cockles and pure aragonite as a function of pH. The dissolution rate of seashells is 3 orders of magnitudes lower than pure aragonite. A summary of the published measurements conditions are presented in Table Ar1 and the existing rates are depicted in Fig. Ar1.

The data was fitted with the equation presented by Busenberg and Plummer (1982) and adapted by Chou et al. (1989), modified to include an activation energy term:

\[ r_\text{v} = A_A \cdot \exp\left(\frac{-E_A}{R \cdot T}\right) \cdot (a_{H^+})^n + A_B \cdot \exp\left(\frac{-E_B}{R \cdot T}\right) \cdot (a_{H_2CO_3})^m + A_C \cdot \exp\left(\frac{-E_C}{R \cdot T}\right) \cdot (a_{H_2O})^n \]

where \( r \) signifies the aragonite steady state dissolution rate, \( A_A, A_B \) and \( A_C \) refers to pre-exponential factors equal to 0.12, 4×10^{-6} and 1×10^{-8} mol/cm²/sec respectively, \( E_A, E_B \) and \( E_C \) designate activation energies equal to 16.0, 46.0 and 46.0 kJ/mol respectively, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Ar1. Rates calculated using Equation Ar-1 described 43 of 45 reported rates within 0.8 log units with an average uncertainty of 0.2 log units. The data points by Cubillas on shells differ from the main aragonite values by 3 orders of magnitude despite following a similar mechanism; they were therefore not included in the model.

References


Table Ar1: Summary of experimental conditions for the various aragonite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aragonite, ground clam, cockle, mussel shells</td>
<td>4.5-9.8</td>
<td>25</td>
<td>Chou et al. (1989)</td>
</tr>
<tr>
<td>Aragonite</td>
<td>7.81-9.09</td>
<td>20-70</td>
<td>Gutjahr et al. (1995)</td>
</tr>
<tr>
<td>Aragonite, Pteropod tests</td>
<td>25</td>
<td>7.25-7.53</td>
<td>Morse et al. (1979)</td>
</tr>
</tbody>
</table>

Figure Ar1: Summary of experimentally measured aragonite steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dashed line.
**As$_2$S$_3$ – Orpiment**

The monoclinic arsenic sulfide orpiment and As$_2$S$_3$ the amorphous version dissolution rate were studied by Lengke and Tempel (2001, 2002, 2003 and 2005) at pH from 2.3 to 8 and temperature of 25 and 30 °C. The data produced was fitted by Palandri and Kharaka (2004) to:

$$r_{H^+} = A_A \cdot a_{H^+}^{1.208} \cdot \exp\left(\frac{-E_A}{R \cdot T}\right)$$

$$r_{H_2O} = A_B \cdot \exp\left(\frac{-E_B}{R \cdot T}\right)$$

$$r_{tt} = r_{H^+} + r_{H_2O}$$

where $r_{H^+}$ and $r_{H_2O}$ signifies the almandine steady-state dissolution rate in acidic, neutral and basic solutions and $r_{tt}$ the total dissolution rate, $A_A$ and $A_B$ refer to pre-exponential factors equal to 4.9x10$^{-13}$ and 1.35x10$^{-20}$ mol/cm²/s respectively, $E_A$ designates an activation energy equal to 87.0 kJ/mol, R represents the gas constant, and T denotes absolute temperature. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Orp1. Rates calculated using Equation Orp-1 described 4 of 4 reported rates within 0.8 log units with an average uncertainty of 0.2 log units.

**References**


Table Orp1: Summary of experimental conditions for the various As$_2$S$_3$ steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>As$_2$S$_3$</td>
<td>20-72</td>
<td>6</td>
<td>Lengke and Tempel (2001)</td>
</tr>
<tr>
<td>As$_2$S$_3$</td>
<td>22</td>
<td>4.1</td>
<td>Lengke and Tempel (2002)</td>
</tr>
<tr>
<td>As$_2$S$_3$</td>
<td>25</td>
<td>2.5-6.8</td>
<td>Lengke and Tempel (2003)</td>
</tr>
<tr>
<td>As$_2$S$_3$</td>
<td>25</td>
<td>2.5-6.8</td>
<td>Lengke and Tempel (2005)</td>
</tr>
</tbody>
</table>

Figure Orp1: Summary of experimentally measured As$_2$S$_3$ steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dashed line.
Augite

The dissolution kinetics of the pyroxene augite were investigated at 22 °C and a pH of 4.1 by Siegel and Pfannkuch (1984), from 20 to 72 °C at pH 6 by Schott and Berner (1985), and by Sverdrup (1990) at 25 °C from pH 2.5 to 6.8. A summary of these published rates are presented in Table Au1 and the existing rates are depicted as a function of pH in Fig. Au1.

Substantial scatter is apparent among the reported rates at low temperatures and near to neutral pH. Such scatter may be due to a combination of factors including precipitation of secondary phases, and variation in measured surface areas.

The fitting of augite dissolution rate data is confounded by the dearth of data and its evident scatter. By analogy with diopside, augite dissolution rates were fitted to:

$$r_* = \left( A_A \left( \frac{a_{H^+}}{Mg^{2+}} \right)^{1/4} + A_B \right) \cdot \exp \left( -E_A/RT \right)$$

(Au-1)

where $r_*$ signifies the augite steady state dissolution rate, $A_A$ and $A_B$ stands for rate constants, $E_A$ designates an activation energy, $R$ represents the gas constant, and $T$ denotes absolute temperature. This fit is strongly dependent on the selection of the low temperature neutral pH data. As the structure and chemical formula of augite is similar to diopside, it seems reasonable to assume their dissolution rates would be similar. 25 °C neutral pH rates of diopside appear to be most consistent with the relatively fast pH 6, 22 °C rate of Schott and Berner (1985). As such, the fit was forced to pass close to this measured rate to the detriment of the other slower values. Taking this into account yielded values of $A_A$ and $A_B$ of 4.2 mol.cm$^{-2}$.s$^{-1}$ and 0.4 mol.cm$^{-2}$.s$^{-1}$. A corresponding $E_A$ of 83 kJ/mol was determined from an Arrhenius plot of the data obtained at pH 6 by Schott and Berner (1985). The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Au1. Rates calculated using Equation Au-1 described 9 of 13 reported rates within 0.8 log units.
References


Table Au1: Summary of experimental conditions for the various augite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Augite</td>
<td>20-72</td>
<td>6</td>
<td>Schott and Berner (1985)</td>
</tr>
<tr>
<td>Augite</td>
<td>22</td>
<td>4.1</td>
<td>Siegel and Pfannkuch (1984)</td>
</tr>
<tr>
<td>Augite</td>
<td>25</td>
<td>2.5-6.8</td>
<td>Sverdrup (1990)</td>
</tr>
</tbody>
</table>

Figure Au1: Summary of experimentally measured augite steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dashed line.
**Barite**

The barium sulphate barite has been studied by Dove and Czank (1995) at temperature ranging from 40 to 90 °C and pH 2 to 11. The available data is plotted on fig. Bar1. Palandri and Kharaka (2004) fitted those values to following equations:

\[
r_{H^+} = A_A \cdot a_{H^+}^{0.298} \cdot \exp \left( \frac{-E_A}{R \cdot T} \right)
\]

\[
r_{H_2O} = A_B \cdot \exp \left( \frac{-E_B}{R \cdot T} \right)
\]

\[
r_{tt} = r_{H^+} + r_{H_2O}
\]

where \(r_{H^+}\) and \(r_{H_2O}\) signifies the barite steady-state dissolution rate in acidic, neutral and basic solutions and \(r_{tt}\) the total dissolution rate, \(A_A\) and \(A_B\) refer to pre-exponential factors equal to 3.16x10^{-6} and 3.16x10^{-7} mol/cm²/s respectively, \(E_A\) designates an activation energy equal to 30.8 kJ/mol, \(R\) represents the gas constant, and \(T\) denotes absolute temperature. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Bar1. Rates calculated using Equation Bar-1 described 4 of 4 reported rates within 0.8 log units with an average uncertainty of 0.2 log units.

**References**


Table Bar1: Summary of experimental conditions for the various barite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Barite</td>
<td>20-72</td>
<td>6</td>
<td>Dove and Czank (1995)</td>
</tr>
</tbody>
</table>

Figure Bar1: Summary of experimentally measured barite steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 50 °C are plotted in dashed line.
Bayerite

The aluminum oxide bayerite ($\beta$-AlOH$_3$) has been studied by Pulfer et al. (1984) at 25°C, pH ranging from 3 to 6.4 and as a function of HNO$_3$ and HF concentration. Nordin et al. (1993) measured the bayerite dissolution kinetics at 25°C and pH 3. The available data is plotted on fig. Bay1. The existing values were fitted to the following equation:

$$r_+ = A_A \left( \frac{a_{H^+}}{a_{Al^{3+}}} \right)^{0.2} \exp \left( \frac{-E_A}{R \cdot T} \right)$$

where $r_+$ signifies the bayerite steady-state dissolution rate in acidic, neutral and basic solutions, $A_A$ refers to pre-exponential factors equal to $34.27 \times 10^{-7}$ mol/cm$^2$/s, $E_A$ designates an activation energy equal to 55.0 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Bay1. Rates containing HF were excluded from the calculation. Rates calculated using Equation Bay-1 described 6 of 7 reported rates within 0.8 log units with an average uncertainty of 0.2 log units.

References


Table Bay1: Summary of experimental conditions for the various barite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
</table>

Figure Bar1: Summary of experimentally measured bayerite steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dashed line.
**Biotite**

Dissolution kinetics of the common phyllosilicate mica biotite \((\text{KMg}_{2.5}\text{Fe}_{0.5}\text{AlSi}_3\text{O}_{10}(\text{OH})_{1.75}\text{F}_{0.25})\) were investigated experimentally by Acker and Bricker (1992), Turpault and Trotignon (1994), Kalinowski and Schweda (1996), Malmström et al. (1996), Malmström and Banwart (1997), Taylor et al. (2000), Murakami et al. (2004), Samson et al. (2005) and Hu and Jun (2012). These authors used a variety of different experimental methods and procedures. Acker and Bricker (1992) used fluidized-bed reactors and flow through columns at 25 °C at pH 3 to 7 under different oxidizing conditions. Turpault and Trotignon (1994) studied the dissolution of single biotite crystals in batch experiments at 24 °C in 0.1 N HNO₃. Kalinowski and Schweda (1996) determined the dissolution kinetics of biotite at pH 1 to 4 and temperature of 22 °C. Malmström and coworkers (1997) performed biotite dissolution experiments in a fluidized-bed reactor at 25 °C over the pH range 2-10. The study of Taylor et al. (2000) aimed to quantify both the dissolution kinetics of biotite and the release of Sr and Sr isotopes at pH near 3 and 25 °C. Murakami et al. (2004) performed biotite dissolution experiments in a batch reactor at 1 bar pCO₂, pH 4.6 and 100 °C. Samson et al. (2005) investigated the transient and quasi steady state dissolution of biotite at 22 - 25 °C at and pH 8 - 14. Hu and Jun (2012) investigated the dissolution kinetics of biotite in batch reactors in conditions relevant for the geological storage of CO₂. A summary of these experimental conditions are presented in Table Bi1 and these rates are depicted in Fig. Bi1.

Assuming a similar dissolution mechanism for the micas, biotite and phlogopite, the reported dissolution rates were fitted to:

\[
  r_+ = A_A \cdot \exp \left( \frac{-E_A}{RT} \right) \left( \frac{a_{H^+}}{a_{Al^{3+}}} \right)^n
\]

(Bi-1)

where \(r_+\) signifies the biotite steady state dissolution rate, \(A_A\) refers to a pre-exponential factor equal to \(1\times10^{-7}\) mol/cm²/s, \(E_A\) designates an activation energy equal to 49.0 kJ/mol, \(R\) represents the gas constant, and \(T\) denotes absolute temperature and \(n\) signifies a stoichiometric coefficient equal to 0.125. This activation energy was obtained by McMaster et al. (2008) by direct observation of retreating steps on a biotite surface using AFM, a similar value to the 52±5 kJ/mol determined by Hu and Jun (2012); \(A_A\), was obtained by the
regression of the data shown in Fig Bi1. The degree to which this equation describes the
dissolution rates reported in the literature can be assessed in Fig Bi1. Rates calculated using
Equation Bi-1 described 38 of 55 reported rates within 0.8 log unit.

References


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201.

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Kalinowski, B.E. and Schweda, P. (1996) Kinetics of muscovite, phlogopite, and biotite dissolution and


resolution imaging of biotite dissolution and measurements of activation energy. Miner. Mag. 72, 115-120.


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Sci. Lett. 224, 117-129.

Acta 69, 399-413.

Swoboda-Colberg, N.G. and Drever, J.J. (1993) Mineral dissolution rates in plot-scale field and


Table Bi1: Summary of experimental conditions for the various biotite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Biotite</td>
<td>25</td>
<td>3-7</td>
<td>Acker and Bricker (1992)</td>
</tr>
<tr>
<td>Biotite</td>
<td>25</td>
<td>1.5</td>
<td>Boyle and Voigt (1973)</td>
</tr>
<tr>
<td>Biotite</td>
<td>25</td>
<td>1-4</td>
<td>Kalinowski and Schweda (1996)</td>
</tr>
<tr>
<td>Biotite</td>
<td>25</td>
<td>2.5-5</td>
<td>Lin and Clemency (1981)</td>
</tr>
<tr>
<td>Biotite</td>
<td>25</td>
<td>2-10</td>
<td>Malmström et al. (2006, 2007)</td>
</tr>
<tr>
<td>Biotite</td>
<td>25</td>
<td>4.2-5.6</td>
<td>Mehmel (1938)</td>
</tr>
<tr>
<td>Biotite</td>
<td>100</td>
<td>4.6</td>
<td>Murakami et al. (2004)</td>
</tr>
<tr>
<td>Biotite</td>
<td>22-25</td>
<td>8-14</td>
<td>Samson et al. (2005)</td>
</tr>
<tr>
<td>Biotite</td>
<td>25</td>
<td>4, 4.5</td>
<td>Swoboda-Colberg and Drever (1993)</td>
</tr>
<tr>
<td>Biotite</td>
<td>25</td>
<td>-3</td>
<td>Taylor et al. (2000a)</td>
</tr>
<tr>
<td>Biotite</td>
<td>24</td>
<td>1</td>
<td>Turpault and Trotignon (1994)</td>
</tr>
</tbody>
</table>
Figure Bi1: Summary of experimentally measured biotite steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dashed line.

![Graph showing biotite dissolution rates vs pH](image)

- Acker and Bricker (1992)
- Boyle and Voigt (1973)
- Kalinowski and Schweda (1996)
- Lin and Clemency (1981)
- Malmstrom et al. (1997)
- Mehmel (1938)
- Nickel (1973), 70°C
- Samson et al. (2005)
- Swoboda-Colberg and Drever (1993)
- Taylor et al. (2000)
- Turpault and Trotignon (1994)

- Model
Boehmite

References

Bronzite

Dissolution rates of bronzite from 25 °C at pH ranging from 1 to 6.1 was measured by Grandstaff (1977) and Schott and Berner (1983). These rates are summarized in Table Br1 and presented at 25 °C as a function of pH in Figure Br1.

As bronzite is identical in structure and close in composition it seems reasonable to expect its rates to closely follow that of enstatite. In accord with Oelkers and Schott (2001) enstatite dissolution rates are consistent with

$$ r_+ = A_A \cdot \exp \left( \frac{-E_A}{RT} \right) \cdot \left( \frac{a_{M^{2+}}^2}{a_{H^+}} \right)^{1/8} \quad (\text{Br-1}) $$

where $r_+$ signifies the steady state forward dissolution rate, $A_A$ refers to a pre-exponential factor equal to $4.9 \times 10^{-7}$ mol/cm²/sec, $E_A$ designates an activation energy equal to 48.5 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. $E_A$ was assumed to be equal to that of enstatite and $A_A$ was estimated in the present study as to provide a fit with the experimental values. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Br1. Rates calculated using Equation Br-1 described 9 of 9 reported rates within 0.8 log units.

References


Table Br1: Summary of experimentally measured bronzite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bronzite</td>
<td>25</td>
<td>2 – 6.1</td>
<td>Grandstaff (1977)</td>
</tr>
<tr>
<td>Bronzite</td>
<td>25</td>
<td>1, 1.5, 6</td>
<td>Schott and Berner (1983)</td>
</tr>
</tbody>
</table>

Figure Br1: Summary of experimentally measured bronzite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the various dashed lines.
Brucite

Disolution rates of brucite from 25 °C and up to 90 °C at pH ranging from 1 to 3.7 have been published by Pokrovsky (2004) and at 22 °C with pH ranging from 2 to 9 by Mast and Drever (1987). The rates are presented for the different temperatures as a function of pH in Figure An1.

Following the theoretical framework laid by Oelkers (2001) for the dissolution of multi-oxides minerals steady-state anthophyllite dissolution rates were fitted to:

\[ r = A_A \exp \left( \frac{E_A}{RT} \right) \frac{a_{H^+}^{1/8}}{a_{Mg^{2+}}^{1/8}} \]  

(Bru-1)

where \( r \) signifies the brucite steady state dissolution rate, \( A_A \) refers to a pre-exponential factor equal to 1.7x10^{-3} \text{ mol/cm}^2/\text{sec}^3, E_A designates an activation energy equal to 46 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. \( A_A \) was provided by Marini (2007) and \( E_A \) was determined to be intermediate between the values proposed by Jordan (1996) and Vermilyea (1969). The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure A1. Rates calculated using Equation Bru-1 described 12 of 14 reported rates within 0.8 log units with an average uncertainty of 0.2 log units.

References


1. Ea was recalculated and falls between the values determined by Jordan (1996) on brucite and Vermilyea (1969) on MgO


Table B1: Summary of experimentally measured brucite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Brucite</td>
<td>25</td>
<td>2.7</td>
<td>Jordan and Rammensee (1996)</td>
</tr>
<tr>
<td>Brucite</td>
<td>25</td>
<td></td>
<td>Pokrovsky et al. (2005a)</td>
</tr>
<tr>
<td>Brucite</td>
<td></td>
<td></td>
<td>Vermilyea (1969)</td>
</tr>
</tbody>
</table>

Figure B1: Summary of experimentally measured brucite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the various dashed lines.
**Bytownite**

Bytownite, a calcium-rich member of the plagioclase solid solution, usually falls between An70 and An90. Holdren and Speyer (1987) measured the dissolution rate of bytownite at 25°C and pH 3. Casey et al. (1991) studied bytownite dissolution kinetics at 25°C and pH 2. Oxburgh et al. (1993) reported dissolution rates at 25°C and for a pH ranging from 3.1 to 7.2 and Stilling and Brantley (1995) investigated the dissolution kinetics of bytownite at 25°C and 3.1<pH<4.7. The experimental conditions for those studies are summarized in Table Byt1 and the rates are presented at 25°C as a function of pH in Figure Byt1.

Following the theoretical framework laid by Oelkers (2001) for the dissolution of multi-oxides minerals steady-state bronzite dissolution rates were fitted to:

$$ r = A_A \left( \frac{a_{H^+}}{a_{Al^{3+}}} \right)^n \cdot \left( 1 - \exp \left( \frac{-E_A}{3.5RT} \right) \right) $$

where $r$ signifies the bytownite steady state dissolution rate, $A_A$ refers to a pre-exponential factor equal to $2.7 \times 10^{-13}$ mol/cm²/s, $n$ stands for the reaction order of 0.33, $E_A$ designates an activation energy equal to 18 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. The degree to which the model fits the data can be assessed with the help of Figure Byt1. Rates calculated using Equation Byt-1 described 20 of 23 reported rates within 0.8 log units with an average uncertainty of 0.2 log units.

**References**


Table Byt1: Summary of experimentally measured bytownite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bytownite</td>
<td>25</td>
<td>5.65</td>
<td>Busenberg and Clemency (1976)</td>
</tr>
<tr>
<td>Bytownite</td>
<td>25</td>
<td>2</td>
<td>Casey et al. (1991)</td>
</tr>
<tr>
<td>Bytownite</td>
<td>25</td>
<td>3</td>
<td>Holdren and Spayer (1987)</td>
</tr>
<tr>
<td>Bytownite</td>
<td>25</td>
<td>3.1-7.2</td>
<td>Oxburgh et al. (1993)</td>
</tr>
<tr>
<td>Bytownite</td>
<td>25</td>
<td>3.1-4.7</td>
<td>Stillings and Brantley (1995)</td>
</tr>
<tr>
<td>Bytownite</td>
<td>22</td>
<td>3.2 – 7.2</td>
<td>Welch and Ullman (1993)</td>
</tr>
<tr>
<td>Bytownite</td>
<td>5-35</td>
<td>3.2, 5.65</td>
<td>Welch and Ullman (1993)</td>
</tr>
</tbody>
</table>

Figure By1: Summary of experimentally measured bytownite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
Calcite

Calcite (CaCO$_3$) dissolution kinetics has been extensively studied in the past (Table Ca1). A summary of the published measurements conditions are presented in Table Ca1 and the existing rates are depicted in Fig. Ca1.

The data was fitted with the equation presented by Busenberg and Plummer (1982) and adapted by Chou et al. (1989), modified to include an activation energy term:

$$r_\text{p} = A_A \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \cdot (a_{H^+})^n + A_B \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \cdot (a_{H_2CO_3})^n + A_C \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \cdot (a_{H_2O})^n$$

where $r$ signifies the calcite steady state dissolution rate, $A_A$, $A_B$ and $A_C$ refers to pre-exponential factors equal to 2.13x10$^{-2}$, 3.47x10$^{-3}$ and 1.17x10$^{-2}$ mol/cm$^2$/s respectively, $E_A$, $E_B$ and $E_C$ designate activation energies equal to 16.0, 46.0 and 46.0 kJ/mol respectively, $R$ represents the gas constant, $T$ denotes absolute temperature and $n$ a reaction order equal to 0.33. Activation energies were determined by Sjöberg and Rickard (1984). The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Ca1. Rates calculated using Equation Ca-1 described 43 of 45 reported rates within 0.8 log units.

References


<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>25</td>
<td>4.7-5.9</td>
<td>Araki and Mucci (1995)</td>
</tr>
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<td></td>
<td></td>
<td></td>
<td>Arvidson et al. (2003)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Chou et al. (1989)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Cubillas et al. (2005)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Davis et al. (2007)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Gutjahr et al. (1995)</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td></td>
<td>Hales and Emerson (1996)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Jordan and Rammensee (1998)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>McInnis and Brantley (1992)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Oelkers et al. (2011)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Plummer et al. (1978)</td>
</tr>
</tbody>
</table>
Figure Ca1: Summary of experimentally measured celestite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
**Celestite**

The strontium sulphate celestite (SrSO$_4$) has been studied by Dove and Czank (1995) at temperature ranging from 32 to 140 °C and pH 3.8 to 12.0. The available data is plotted on fig. Cel1. Palandri and Kharaka (2004) fitted those values to following equations:

$$r_{H^+} = A_A \cdot d_H^{0.109} \cdot \exp \left( \frac{-E_A}{R \cdot T} \right)$$

where $r_{H^+}$ signifies the barite steady-state dissolution rate in acidic solutions, $A_A$ refers to pre-exponential factors equal to 5.4e$^{-9}$ mol/cm$^2$/s, $E_A$ designates an activation energy equal to 6.08 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Cel1. Rates calculated using Equation Cel-1 described all reported rates within 0.8 log units.

**References**


Table Cel1: Summary of experimentally measured celestite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Celestite</td>
<td>25</td>
<td>2.7</td>
<td>Dove and Czank (1995)</td>
</tr>
</tbody>
</table>

Figure Cel1: Summary of experimentally measured celestite steady-state dissolution rates reported in the literature at 50 °C as a function of pH and at pH 5.7 as a function of temperature. The results of the model are plotted as the dotted line.
Chlorite

A limited amount of studies have investigated the dissolution kinetics of chlorite at 25 °C, Ross (1967) published chlorite dissolution rate at pH 0, Kodama and Schnitzer (1973) reported rates gathered near pH 3. Sverdrup (1990) investigated the dissolution of chlorite at pH 2.5 to 5, May (1995) studied the dissolution rate at pH 5 and Malmström et al. (1996) at pH 8.2. Rochelle et al. (1996) reported rates measured at pH 7.7 to 12.5 and Salmon and Malmström (2001) at pH 3.5. In 2003 Hamer et al. measured rates from 3 to 4.5, Brandt et al. (2003) from pH 1.89 to 5.3 and Gustafsson and Puigdomenech (2003) from 2 to 12. Finally, Lowson et al. (2005, 2007) investigated the dissolution of Fe-rich chlorite under far from equilibrium conditions from pH of 3 to 10.5 at 25 °C. The experimental conditions for those studies are summarized in Table Ch1 and the rates are presented at 25 °C as a function of pH in Figure Ch1.

We have fitted the published data on chlorite on the equation developed by Lowson et al. (2005):

\[
    r = A_A \left( \frac{a_{\text{H}^+}^3}{a_{\text{Al}^{3+}}} \right)^{1/3.7}
\]  

(Ch-1)

where \( r \) signifies the chlorite steady state dissolution rate, \( A_A \) refers to a pre-exponential factor equal to \( 10^{-14.896} \) mol/cm²/sec, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Ch1. Rates calculated using Equation Ch-1 described 88 of 94 reported rates within 0.8 log units with an average uncertainty of 0.2 log units.

References


Table Ch1: Summary of experimentally measured chlorite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chlorite</td>
<td>25</td>
<td>1.89-5.3</td>
<td>Brandt et al. (2003)</td>
</tr>
<tr>
<td>Chlorite</td>
<td>25</td>
<td>2-12</td>
<td>Gustafsson and Puigdomenech (2003)</td>
</tr>
<tr>
<td>Ripidolite</td>
<td>25</td>
<td>2-4.5</td>
<td>Hamer et al. (2003)</td>
</tr>
<tr>
<td>Chlorite</td>
<td>25</td>
<td>3</td>
<td>Kodama and Schnitzler (1973)</td>
</tr>
<tr>
<td>Chlorite</td>
<td>25</td>
<td>3.1-10.44</td>
<td>Lowson et al. (2005)</td>
</tr>
<tr>
<td>Chlorite</td>
<td>25-45</td>
<td>3.1-10.34</td>
<td>Lowson et al. (2007)</td>
</tr>
<tr>
<td>Chlorite</td>
<td>25</td>
<td>8.2</td>
<td>Malmström et al. (1996)</td>
</tr>
<tr>
<td>Chlorite</td>
<td>25</td>
<td>5</td>
<td>May et al. (1995)</td>
</tr>
<tr>
<td>Chlorite</td>
<td>25</td>
<td>7.7-12.5</td>
<td>Rochelle et al. (1996)</td>
</tr>
<tr>
<td>Orthochlorite</td>
<td>25</td>
<td>-0.3</td>
<td>Ross (1967)</td>
</tr>
<tr>
<td>Chlorite</td>
<td>25</td>
<td>3.5</td>
<td>Salmon and Malmström (2001)</td>
</tr>
<tr>
<td>Chlorite</td>
<td>25</td>
<td>2.5-5</td>
<td>Sverdrup (1990)</td>
</tr>
<tr>
<td>Chlorite</td>
<td>25</td>
<td>4, 4.5</td>
<td>Swoboda-Colberg and Drever (1993)</td>
</tr>
</tbody>
</table>
Figure Ch1: Summary of experimentally measured chlorite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.

- Brandt et al. (2003)
- Gustafsson and Puigdomenech (2003)
- Hamer et al. (2003)
- Kodama and Schnitzler (1973)
- Lowson (2007)
- Lowson (2005)
- Malmström et al. (1996)
- May (1995)
- Rochelle et al. (1996)
- Ross (1967)
- Salmon and Malmström (2001)
- Sverdrup (1990)
- Swoboda-Colberg and Drever (1993)
- Model
1199  *Christobalite*

1200

1201  *Chrysotile*

1202

1203
Cordierite

The dissolution rates of the magnesium iron aluminum cyclosilicate cordierite were studied by Sverdrup (1990) and these data are plotted in fig. Cor1. In the absence of a mechanistic model Palandri and Kharaka (2004) fit these rates assuming they are controlled by two parallel reactions, one proton promoted, and one water promoted in accord with:

\[ r_{H^+} = A_A \cdot a_{H^+}^1 \cdot \exp\left(\frac{-E_A}{R \cdot T}\right) \]

\[ r_{H_2O} = A_B \cdot \exp\left(\frac{-E_B}{R \cdot T}\right) \]

which can be summed to yield the overall forward rate using

\[ r_+ = (r_{H^+} + r_{H_2O}) \]

where \( r_{H^+} \) and \( r_{H_2O} \) signifies the cordierite steady-state dissolution rate in response to the proton and water promoted mechanisms, respectively, and \( r_+ \), the total forward dissolution rate, \( A_A \) and \( A_B \) refer to pre-exponential factors equal to \( 1.12 \times 10^{12} \) and \( 5.75 \times 10^{-11} \) mol/cm²/s respectively, \( E_A \) designates an activation energy equal to 113.3 and 28.3 kJ/mol respectively, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. The degree to which these equations describes the measured can be assessed in Figure Cor1. Rates calculated using Equation Cor-1 described all 8 of the reported rates within 0.8 log units.

References


Table Cor1: Summary of experimental conditions for the various cordierite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cordierite</td>
<td>25</td>
<td>2 - 6</td>
<td>Sverdrup (1990)</td>
</tr>
</tbody>
</table>

Figure Cor1: Summary of experimentally measured barite steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dotted line.
**Corundum**

Corundum (Al2O3) is a crystalline form of aluminum oxide. Caroll-Webb and Walther (1988) studied the dissolution kinetics of corundum at 25 °C as a function of pH (0.99 – 11.21). The experimental conditions for this study are summarized in Table Co1 and the rates are presented at 25 °C as a function of pH in Figure Co1.

The data was fitted using the dissolution mechanisms defined by Palandri and Kharaka (2004):

\[ r_{H^+} = A_A \cdot a_{H^+}^{0.15} \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \]

\[ r_{H_2O} = A_B \cdot \exp \left( \frac{-E_B}{R \cdot T} \right) \]

\[ r_{OH^-} = A_C \cdot a_{OH^-}^{0.55} \cdot \exp \left( \frac{-E_C}{R \cdot T} \right) \]

\[ r_{tt} = r_{H^+} + r_{H_2O} + r_{OH^-} \]

where \( r_i \) represents the corundum dissolution rate in acidic, neutral and basic conditions while \( r_{tt} \) stands for the overall dissolution rate. \( A_A, A_B \) and \( A_C \) refer to pre-exponential factors equal to 6.45x10^{-9}, 4.36x10^{-12} and 1.94 mol/cm²/s respectively, \( E_A, E_B \) and \( E_C \) designate activation energies equal to 47, 61 and 79 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature.

The degree to which this model describes the dissolution rates reported in the literature can be assessed with the help of Figure Co1.

**References**


Table Co1: Summary of experimentally measured corundum steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Corundum</td>
<td>25</td>
<td>0.99-11.21</td>
<td>Caroll-Webb and Walter (1988)</td>
</tr>
</tbody>
</table>

Figure Co1: Summary of experimentally measured corundum steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
**Dawsonite**

The mineral dawsonite (NaAlCO$_3$(OH)$_2$) has been suggested to play a significant role as a CO$_2$ storage host in saline alkaline aqueous solutions. Following the work of Ferrante et al. (1976) and Bénézeth et al. (2007) the thermodynamic stability of dawsonite is well known. Dissolution rates of natural dawsonite have been studied by Hellevang et al. (2005) at 80 °C and pH ranging from 3 to 10. Those findings were subsequently refined by Hellevang et al. (2009) who measured dawsonite reactivity at 22, 60 and 77 °C and within the pH range of 0.5<pH<5.

The rate data obtained in Hellevang et al. (2009) suggests a 77 °C pH 4 rate of approximately 10$^{-10.7}$ moles/cm$^2$/s. This is more than two orders of magnitude higher than the 10$^{-12.8}$ moles/cm$^2$/s reported by Hellevang et al. (2005). One likely explanation for the large difference in estimated rates between the 2009 study and the 2005 study by Hellevang et al. (2005) lays in an overestimation of the BET surface area used in the surface area normalization in Hellevang et al. (2005) and therefore the rates were underestimated. Other reasons such as differences in the reactivity of natural dawsonite (Hellevang et al., 2005) compared to synthetic (Hellevang et al., 2009) cannot be excluded. The data reported by Hellevang et al. (2005) is inconsistent with the later study and was therefore not included in our fit.

The experimental conditions for those studies are summarized in Table Da1 and the rates are presented at 25 °C as a function of pH in Figure Da1. The experimental data was fitted by Declercq (2010) to:

\[
\begin{align*}
  r_{H^+} &= A_A \cdot \exp\left(\frac{-E_A}{RT}\right) \cdot a_{H^+}^{0.941} \\
  r_{H_2O} &= A_B \cdot \exp\left(\frac{-E_B}{RT}\right) \\
  r_t &= r_{H^+} + r_{H_2O}
\end{align*}
\]

(Daw1)

where $r_{H^+}$ and $r_{H_2O}$ signifies the dawsonite steady-state dissolution rate in acidic and neutral solutions and $r_t$ the total dissolution rate, $A_A$ and $A_B$ refer to pre-exponential factors equal to 2.7x10$^2$and 4.2x10$^{-5}$ mol/cm$^2$/s respectively, $E_A$ and $E_B$ designates an activation energy
equal to 63.8 and 49.4 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute
temperature. The degree to which this equation describes the dissolution rates reported in
the literature can be assessed in Figure Da1. Rates calculated using Equation Da-1 described
18 of 18 reported rates within 0.8 log units.

References

of its thermodynamic properties from solubility measurements: implications for mineral

Declercq, J. (2010) Dissolution rates and Mechanisms; Experiments on Alteration and Reactivity of

dawsonite. US Bureau of Miners Report Investigation 8129, Washington, DC.


pH 0.9 to 5 and temperatures of 22, 60 and 77 °C. Applied Geochem. 25, 1575-1586.
Table Da1: Summary of experimentally measured dawsonite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dawsonite, natural</td>
<td>80</td>
<td>3-10</td>
<td>Hellevang et al. (2005)</td>
</tr>
<tr>
<td>Dawsonite, Synthetic</td>
<td>22, 60, 77</td>
<td>0.9 - 5</td>
<td>Hellevang et al. (2009)</td>
</tr>
</tbody>
</table>

Figure Da1: Summary of experimentally measured dawsonite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
**Diaspore**

The dissolution rate of the aluminum hydroxide diaspore \((\text{AlO(OH)})\) has been studied by Chang et al. (1979). In the absence of additional data to improve existing data fits, these we have adopted the empirical pH power function fit of Palandri and Kharaka (2004) in accord with:

\[
\begin{align*}
    r_{H_{2}O} &= A_{B} \cdot e^{\left(\frac{-E_{A}}{R \cdot T}\right)} \\
    r_{OH^{-}} &= A_{C} \cdot a_{OH}^{-1.503} \cdot e^{\left(\frac{-E_{A}}{R \cdot T}\right)} \\
    r_{tt} &= r_{H_{2}O} + r_{OH^{-}}
\end{align*}
\]

where \(r_{H_{2}O}\) and \(r_{OH^{-}}\) signifies the almandine forward dissolution rate in neutral and basic solutions and \(r_{tt}\) the total dissolution rate, \(A_{B}\) and \(A_{C}\) refer to pre-exponential factors equal to 5.25x10\(^{-20}\) and 9.77x10\(^{-10}\) mol/cm\(^2\)/s respectively, \(E_{A}\) designates an activation energy equal to 47.5 kJ/mol, \(R\) represents the gas constant, and \(T\) denotes absolute temperature.
References:


Table Dia1: Summary of experimentally measured diaspore steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diaspore</td>
<td>25</td>
<td></td>
<td>Chang et al. (1979)</td>
</tr>
</tbody>
</table>

Figure Dia1: Summary of experimentally measured diaspore steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
**Diopside**

Dissolution rates of the clinopyroxene diopside were reported at 22 °C and with pH ranging from 1 to 6 by Schott et al. (1981) then Eggleston et al. (1989) published kinetic data on diopside dissolution at 25 °C and pH of 1. Knauss et al. (1993) measured dissolution rates at 25°, 50 ° and 70 °C. Brantley and Chen (1995) reported dissolution rates at 25° and 90 °C with pH of 1 to 3.8. More recently Golubev et al. (2005) investigated the effect of CO₂ on the dissolution kinetics of diopside dissolution at 25 °C and pH from 1 to 11.7. Stockmann et al. investigated the dissolution rates at 25° and 70 °C with pH ranging from 4.3 to 8.5. Daval et al. (2010) reported rates acquired at 90 °C and pH close to 5. The rates acquired at 25 °C are presented as a function of pH in Figure Di1. The values reported by Schott et al. (1981) are several orders of magnitude faster than the data produced by the other authors; it was not thus included in the model. The experimental conditions for those studies are summarized in Table Di1 and the rates are presented at 25 °C as a function of pH in Figure Di1.

Following the definition of an equation describing the dissolution mechanism of pyroxenes by Oelkers (2001) diopside dissolution rates were fitted to:

\[ r_+ = \left( A_A \left( \frac{a_{\text{Mg}^{2+}}}{\text{Mg}^{2+}} \right)^{1/4} + A_B \right) \exp\left(-\frac{E_A}{RT}\right) \]  

where \( r_+ \) signifies the diopside steady state dissolution rate, \( A_A \) and \( A_B \) stands for rate constants, \( E_A \) designates an activation energy, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. \( A_A \) and \( A_B \) were determined to be 9.97x10⁻⁷ and 0.2x10⁻⁶ while an \( E_A \) of 48.14 kJ/mol was calculated from an Arrhenius plot of the data obtained by Knauss et al. (1993) at pH 2. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Di1. Rates calculated using Equation Di-1 described 62 of 72 reported rates within 0.8 log units with an average uncertainty of 0.2 log units.
References


Oelkers, E.H. (2001)?


Table Di1: Summary of experimentally measured diopside steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diopside</td>
<td>25, 90</td>
<td>1.2-3.6</td>
<td>Brantley and Chen (1995)</td>
</tr>
<tr>
<td>Diopside</td>
<td>90</td>
<td>5</td>
<td>Daval et al. (2010)</td>
</tr>
<tr>
<td>Diopside</td>
<td>25</td>
<td>1</td>
<td>Eggleston et al. (1989)</td>
</tr>
<tr>
<td>Diopside</td>
<td>25</td>
<td>1-11.75</td>
<td>Golubev et al. (2005)</td>
</tr>
<tr>
<td>Diopside</td>
<td>25-70</td>
<td>2-12.1</td>
<td>Knauss et al. (1993)</td>
</tr>
<tr>
<td>Diopside</td>
<td>22</td>
<td>1-6</td>
<td>Schott et al. (1981)</td>
</tr>
<tr>
<td>Diopside</td>
<td>25, 70</td>
<td>4.3-8.5</td>
<td>Stockmann et al. (2008)</td>
</tr>
</tbody>
</table>

Figure Di1: Summary of experimentally measured diopside steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted and dashed lines.
Dolomite

The study of dolomite (CaMg(CO$_3$)$_2$) dissolution started with Busenberg and Plummer (1982) at temperature ranging from 5 to 65 °C and pH from 1 to 10 and fixed pCO$_2$. Chou et al. (1989) used continuous flow through reactor to study dolomite dissolution at 25 °C and in the pH range 3 to 10. Orton and Unwin (1993) investigated the dissolution kinetics of dolomite and reported transport rates at 25 °C and pH 3-4 through a channel-flow method, Gautelier et al. (1999) reported rates at 25, 50 and 80 °C and pH ranging from -0.39 to 4.44. Lütte et al. (2002) used vertical scanning interferometry to study dolomite dissolution at pH 3 and 25 °C. Pokrovsky et al. (1999) and Pokrovsky and Schott (2001) performed measurements of the steady state of dolomite dissolution at 25 °C as a function of pH (5-12). Gautelier et al. (2007) reported rates at 80 °C as a function of chemical affinity. The experimental conditions for those studies are summarized in Table D1 and the rates are presented at 25 °C as a function of pH in Figure D1.

We based our rate law on the one defined by Busenberg and Plummer (1982) who followed the interpretative guidelines of Plummer et al. (1978) for calcite:

$$ r_{+,\text{calcite}} = k_1 \cdot a_{H^+}^{n_1} + k_2 \cdot a_{H_2CO_3}^{n_2} + k_3 \cdot a_{H_2O}^{n_3} $$  \hspace{1cm} (Do-1)

and added an activation energy term for each dissolution mechanism. The pre-exponential factors were recalculated to fit the data:

$$ r_+ = [A_A \cdot a_{H^+}^{n_1} \times \exp \left( \frac{-E_A}{R \cdot T} \right)] + [A_B \cdot a_{H_2CO_3}^{n_2} \times \exp \left( \frac{-E_B}{R \cdot T} \right)] + [A_C \cdot a_{H_2O}^{n_3} \times \exp \left( \frac{-E_C}{R \cdot T} \right)] $$  \hspace{1cm} (Do-2)

where $r_+$ signifies the dolomite steady state dissolution rate, $A_A$, $A_B$ and $A_C$ refer to pre-exponential factor equal to 1.49x10$^2$, 1x10$^2$ and 2.2x10$^{-7}$ mol/cm$^2$/s respectively, $E_A$, $E_B$ and $E_C$ designate an activation energy equal to 29.0, 34.8 and 52.2 kJ/mol respectively, $R$ represents the gas constant, $T$ denotes absolute temperature and $n$ is a reaction order equal to 0.75. The activation energies were determined from an Arrhenius plot of the data reported by Gautelier et al. (1999). The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure D1. Rates calculated using Equation Do-2 described 85 of 87 reported rates within 0.8 log units.
References


Pokrovsky, O.S. and Schott, J. (2001)

Table Do1: Summary of experimentally measured dolomite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dolomite</td>
<td>5-65</td>
<td>1-10</td>
<td>Busenberg and Plummer (1982)</td>
</tr>
<tr>
<td>Dolomite</td>
<td>25</td>
<td>2.5-9.5</td>
<td>Chou et al. (1989)</td>
</tr>
<tr>
<td>Dolomite</td>
<td>25</td>
<td>4</td>
<td>Deike (1990)</td>
</tr>
<tr>
<td>Dolomite</td>
<td>25-80</td>
<td>-0.4-4.2</td>
<td>Gautelier et al. (1999)</td>
</tr>
<tr>
<td>Dolomite</td>
<td>80</td>
<td>6.5-7.2</td>
<td>Gautelier et al. (2007)</td>
</tr>
<tr>
<td>Dolomite</td>
<td>25</td>
<td>3</td>
<td>Lüttge et al. (2002)</td>
</tr>
<tr>
<td>Dolomite</td>
<td>25</td>
<td>3-4</td>
<td>Orton and Unwin (1993)</td>
</tr>
<tr>
<td>Dolomite</td>
<td>25</td>
<td></td>
<td>Pokrovsky et al. (2001)</td>
</tr>
<tr>
<td>Dolomite</td>
<td></td>
<td></td>
<td>Pokovsky et al. (2003)</td>
</tr>
</tbody>
</table>

Figure Do1: Summary of experimentally measured dolomite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dashed line.
Busenberg and Plummer (1982)
Chou et al. (1989)
Deike (1990)
Herman and White (1985)
Gautelier et al. (1999), 25°C
Gautelier et al. (1999), 80°C
Gautelier et al. (2006), 80°C
Luttge et al. (2002)
Pokrovsky et al. (2005)
Pokrovsky et al. (2003)
Model, 25°C
Model, 80°C
**Enstatite**

Dissolution rates of enstatite (MgSiO$_3$) at 25 °C and various pH have been published by Luce et al., (1972), Schott et al., (1981), Ferruzzi (1993), Siegel and Pfannkuch (1984), and Halder and Walther (2011). The rates are presented as a function of pH in Figure E1, other than the pH 4.1 rate reported by Siegel and Pfannkuch (1984). This rate was removed from the dataset because stoichiometric metal release was not observed in this experiment.

Steady-state enstatite dissolution rates were reported as function of aqueous magnesium and silica activity, pH from 1 to 11, and temperature from 28° to 168° C by Oelkers and Schott (2001). These authors fit their data to

$$r_+ = A_A \exp \left( \frac{-E_A}{RT} \right) \left( \frac{a_{M^{2+}}}{a_{Mg^{2+}}} \right)^{1/8}$$

(E1)

where $r_+$ signifies the enstatite steady state dissolution rate, $A_A$ refers to a pre-exponential factor equal to 4.\(\times10^{-6}\) mol/cm$^2$/sec, $E_A$ designates an activation energy equal to 48.5 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure E1. Rates calculated using Equation E-1 described 12 of 14 reported 25 C rates within 0.8 log units with an average uncertainty of 0.2 log units.

**References**


Table E1: Summary of experimentally measured enstatite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Enstatite</td>
<td>25</td>
<td>1,6</td>
<td>Luce et al. (1972)</td>
</tr>
<tr>
<td>Bramble Enstatite</td>
<td>25</td>
<td>1-6</td>
<td>Schott et al. (1981)</td>
</tr>
<tr>
<td>Enstatite</td>
<td>25</td>
<td>4,1</td>
<td>Siegel and Plannkuch (1984),</td>
</tr>
<tr>
<td>Enstatite</td>
<td>25</td>
<td>2-7</td>
<td>Ferruzzi (1993)</td>
</tr>
<tr>
<td>Bramble Enstatite</td>
<td>28-160</td>
<td>2-11</td>
<td>Oelkers and Schott (2001)</td>
</tr>
</tbody>
</table>

Figure E1: Summary of experimentally measured enstatite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted and dashed lines.
Epidote

The dissolution kinetics of epidote (Ca$_2$FeAl$_2$Si$_3$O$_{12}$OH) was investigated at high
temperature (250-350 °C) and high-pressure (0.5-12 kbar) conditions by several authors
including Blanchard (1994) and Brandon et al. (1996). Experimental studies at ambient
temperature and atmospheric pressure were conducted by Nickel (1973) at pH of 0.2 to
10.6, Sverdrup (1990) at pH ranging from 3 to 10.5, Rose (1991) at pH of 1.3 to 5.5 and
Kalinowski et al. (1998) at pH ranging from 1 to 6. The experimental conditions where those
experiments were conducted are reported in Table Ep1 and the available experimental rates
at 25 °C are plotted in Fig. Ep1

The variation of epidote dissolution rates on pH and the presence of substantial
aluminum in the epidote structure suggests that its rates may depend on aqueous aluminum
concentration in a manner similar to aluminosilicates, e.g. albite. Nevertheless owing to the
dearth of direct measurements of the effect of aqueous aluminum on epidote dissolution
rates the fit of Palandri and Kharaka (2004) has been adopted in this study. This fit is
expressed by:

\[
\begin{align*}
    r_{H^+} &= A_A \cdot a_{H^+}^{0.338} \cdot \exp \left(\frac{-E_A}{R \cdot T}\right) \\
    r_{H_2O} &= A_B \cdot \exp \left(\frac{-E_B}{R \cdot T}\right) \\
    r_{OH^-} &= A_C \cdot a_{OH^-}^{0.556} \cdot \exp \left(\frac{-E_C}{R \cdot T}\right) \\
    r^+ &= r_{H^+} + r_{H_2O} + r_{OH^-}
\end{align*}
\]

where $r_{H^+}$, $r_{H_2O}$ and $r_{OH^-}$ signify the almandine forward dissolution rate in acidic, neutral
and basic solutions and $r^+$ the total dissolution rate, $A_A$, $A_B$ and $A_C$ refer to pre-exponential
factors equal to 7.244x10$^{-3}$, 2.51x10$^{-4}$ and 3.38x10$^{-8}$ mol/cm$^2$/s respectively, $E_A$, $E_B$ and $E_C$
designate activation energies equal to 71.1, 70.7, and 79.1 kJ/mol, $R$ represents the gas
constant, and $T$ denotes absolute temperature. The degree to which this equation describes
the dissolution rates reported in the literature can be assessed in Figure Ep1. Rates
calculated using Equation Ep-1 described 37 of 40 reported rates within 0.8 log units.
References


Table Ep1: Summary of experimentally measured epidote steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Epidote</td>
<td>250</td>
<td>6.4 - 12</td>
<td>Blanchard (1994)</td>
</tr>
<tr>
<td>Epidote</td>
<td>250-350</td>
<td>xxxxx</td>
<td>Brandon et al. (1996)</td>
</tr>
<tr>
<td>Epidote</td>
<td>25</td>
<td>2-6.5</td>
<td>Kalinowski et al. (1998)</td>
</tr>
<tr>
<td>Epidote</td>
<td>25</td>
<td>0.2-10.6</td>
<td>Nickel (1973)</td>
</tr>
<tr>
<td>Epidote</td>
<td>25</td>
<td>1.3-5.5</td>
<td>Rose (1991),</td>
</tr>
<tr>
<td>Epidote</td>
<td>25</td>
<td>3-10.5</td>
<td>Sverdrup (1990).</td>
</tr>
</tbody>
</table>

Figure Ep1: Summary of experimentally measured epidote steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted and dashed lines.
**Fayalite and other non-forsteritic olivines**

Owing to their relative rarity, relatively few studies have characterized the dissolution rates of the non-forsteritic olivines. The most common of these is fayalite (Fe$_2$SiO$_4$) the iron end member of this mineral family. The study of fayalite dissolution kinetics is confounded by the oxidation of aqueous divalent iron at neutral to basic conditions. Fayalite dissolution kinetics were studied by Siever and Woodford (1979), Schott and Berner (1983), Siegel and Pfannkuch (1984), and Westrich et al. (1993). Of these studies only Siever and Woodford (1979) and Westrich et al. (1993) reported rates and only at pH 2 and 4.5 at 25 °C. This limited dataset precludes a quantitative fit of the rate data for this mineral.

Westrich et al. (1993) also reported dissolution rates for a variety of other divalent metal olivines at acidic pH and temperatures ranging from 25 to 45 °C. The dissolution rates of each of these minerals exhibit similar behaviors over the condition studied. Each had activation energy of 56± 6 kJ/mol and each had 25 °C dissolution rates that decreased by ~0.5 orders of magnitude with each pH unit increase in acid conditions. These behaviors are identical within uncertainty to those of forsterite (see below). The observation of Westrich et al (1993), suggests that the dissolution rates of all olivines as a function of temperature and pH can, at least provisionally, be considered proportional to those of forsterite. This observation was thus adopted in this study to estimate the dissolution rates of the non-forsteritic olivines.

Non-forsteritic olivines dissolution rates were calculated from

\[ r_+ = r_a + r_{H_2O} \]  \hspace{1cm} \text{(Fay1)}

where \( r_a \) and \( r_{H_2O} \) refer to rates of two distinct dissolution pathways, a pH dependent pathway that dominates at pH<9 and a pH independent pathway that dominates at higher pH. These two rate equations are given by

\[ r_a = A_a s \exp\left( -\frac{E_{A,a}}{RT} \right) a_{H^+}^{0.5} \]  \hspace{1cm} \text{(Fay2)}

and

\[ r_{H_2O} = A_{H_2O}s \exp\left( -\frac{E_{A,H_2O}}{RT} \right) \]  \hspace{1cm} \text{(Fay3)}
where $A_{A,a}$ and $A_{A,H_2O}$ represent pre-exponential factors listed in Table Fay2, and $E_{A',a}$ and $E_{A',H_2O}$ designate activation energies equal to 55.38 and 71.20 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. These activation energies are those of generated by a fit of the forsterite rate data (see below), whereas the pre-exponential factors were calculated such that dissolution rates of each olivine were proportional to that of forsterite and consistent with the rates reported by Westrich et al (1993). Examples of the results of these fits are shown in Fig Fay1 and Fay2.

References


Table Fay1: Summary of experimentally measured fayalite steady-state dissolution rates reported in the literature. N/R refers to not reported.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fayalite</td>
<td>25</td>
<td>N/R</td>
<td>Schott and Berner (1983)</td>
</tr>
<tr>
<td>Fayalite</td>
<td>22</td>
<td>N/R</td>
<td>Siegel and Pfannkuch (1984)</td>
</tr>
<tr>
<td>Fayalite</td>
<td>25</td>
<td>4.5</td>
<td>Siever and Berner (1979)</td>
</tr>
</tbody>
</table>

Table Fay2: Summary of experimentally measured fayalite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>Pre-exponential factor $A_A$ (mol/cm$^2$/s)</th>
<th>Pre-exponential factor $A_{H2O}$ (mol/cm$^2$/s)</th>
<th>Composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Calcio-olivine</td>
<td>3.51x10$^{-2}$</td>
<td>1.01x10$^5$</td>
<td>Ca$_2$SiO$_4$</td>
</tr>
<tr>
<td>Co-olivine</td>
<td>1.11x10$^{-6}$</td>
<td>3.19x10$^2$</td>
<td>Co$_2$SiO$_4$</td>
</tr>
<tr>
<td>Fayalite</td>
<td>4.42x10$^{-4}$</td>
<td>1.27x10$^3$</td>
<td>Fe$_2$SiO$_4$</td>
</tr>
<tr>
<td>Kirschsteinite</td>
<td>1.40x10$^{-2}$</td>
<td>4.02x10$^4$</td>
<td>CaFeSiO$_4$</td>
</tr>
<tr>
<td>Liebenbergite</td>
<td>1.76x10$^{-7}$</td>
<td>5.06x10$^{-1}$</td>
<td>Ni$_2$SiO$_4$</td>
</tr>
<tr>
<td>Monticellite</td>
<td>4.42x10$^{-4}$</td>
<td>1.27x10$^3$</td>
<td>CaMgSiO$_4$</td>
</tr>
<tr>
<td>Phenacite</td>
<td>2.21x10$^{-8}$</td>
<td>6.36x10$^{-2}$</td>
<td>BeSiO$_4$</td>
</tr>
<tr>
<td>Tephroite</td>
<td>1.73x10$^{-3}$</td>
<td>3.7x10$^3$</td>
<td>Mn$_2$SiO$_4$</td>
</tr>
<tr>
<td>Willemite</td>
<td>1.11x10$^{-6}$</td>
<td>3.19x10$^2$</td>
<td>ZnSiO$_4$</td>
</tr>
</tbody>
</table>
Figure Fay1: Summary of experimentally measured non forsteritic olivine steady-state dissolution rates reported in the literature at 25 °C as a function of pH. The results of the model are plotted as the dotted line. All data were from Westrich et al. (1993) unless otherwise specified. The dissolution rate of Co-forsterite at pH 6, 25 C, shown in the Fig was taken from Fig 4 of Westrich et al. (1993) rather than from their table in annex 1. The later value is inconsistent with the trend of the data and appears to be a typographical error.
Figure Fay2: Summary of experimentally measured non forsteritic olivine steady-state dissolution rates at 25 °C and pH 2. The results of the models are plotted as various line, forsterite, the solid line is given as a reference.
Fluorite and other divalent metal fluorides.

The halide mineral fluorite (CaF$_2$) dissolution has been studied by Gardner and Nancollas (1976), Hamza et al. (1987), Christoffersen et al. (1988), Zang and Nancollas (1990), Hamza and Hamdona (1991), Bosbach et al. (1995), De Giudici et al. (2004), Zhang et al. (2005) and Cama et al. (2010). Zang and Nancollas (1990) reported rate parameters at 25 °C, including an activation energy, Hamza and Hamdona reported rates at 25 °C in NaCl solutions and Bosbach et al. (1995) reported direct AFM observation of fluorite dissolution but did not report rates. De Giudici measured the fluorite dissolution kinetics at 25 °C and from pH 2, Zhang et al. (2005) studied the dissolution rate of fluorite in flow through reactor at 25 °C and pH from 3.2 to 3.6 and Cama et al. (2010) reported rates measured with flow through systems, VSI and AFM at 25 °C and pH ranging from 1.1 to 3.0. The experimental conditions where those experiments were conducted are reported in Table Flu1 and the available experimental rates at 25 °C are plotted in Fig. Flu1. The reported data was fitted to

$$r_{H^+} = A_A e^{-0.1} \exp \left( \frac{-E_A}{R \cdot T} \right)$$

Flu1

where $r_*$ signifies the fluorite steady-state dissolution rate in acidic solutions, $A_A$ refers to pre-exponential factors equal to 1.51x10$^3$ mol/cm$^2$/s, $E_A$ designates an activation energy equal to 9.87 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. An $E_A$ of 73.0 kJ/mol was reported by Zhang and Nancollas (1990) based on Gardner and Nancollas (1976) but we recalculated the $E_A$ to be 9.87 kJ/mol based on the data provided by Zhang et al. (2005). The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Flu1. Rates calculated using Equation Flu1 described 20 of 26 reported rates within 0.8 log units.
References


Table Flu1: Summary of experimentally measured fluorite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fluorite</td>
<td>25</td>
<td>1 - 3</td>
<td>Cama et al. (2010)</td>
</tr>
<tr>
<td>Fluorite</td>
<td>25</td>
<td>2</td>
<td>De Giudici et al. (2004)</td>
</tr>
<tr>
<td>Fluorite</td>
<td>25</td>
<td>6.5</td>
<td>Hamzam and Hamdona (1991)</td>
</tr>
<tr>
<td>Fluorite</td>
<td>25</td>
<td>3.2-3.6</td>
<td>Zhang et al. (2005)</td>
</tr>
</tbody>
</table>
Table Flu2: Summary of activation energies and pre-exponential parameters for earth fluorides reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>$A_A$(mol/cm$^2$/s)</th>
<th>$E_A$(kJ/mol)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sellaite (MgF$_2$)</td>
<td>86.0</td>
<td>9.87</td>
<td>Abdul-Rahman (1988)</td>
</tr>
<tr>
<td>Fluorite (CaF$_2$)</td>
<td>9.8</td>
<td>72.0</td>
<td>This study</td>
</tr>
<tr>
<td>SrF$_2$</td>
<td>72.0</td>
<td>9.87</td>
<td>Abdul-Rahman (1988)</td>
</tr>
</tbody>
</table>

Figure Flu1: Summary of experimentally measured fluorite steady-state dissolution rates reported in the literature at 25 °C as a function of pH. The results of the model are plotted as the dotted line.
A larger number of studies have focused on characterizing the dissolution rates of forsterite ($\text{Mg}_2\text{SiO}_4$) – see table F1. The variation of 25 °C forsterite dissolution rates are depicted as a function of pH in Fig F1. There is a general consistency of the rates reported in most of the published studies. The two notable exceptions are Grandstaff (1978, 1986) which are approximately two orders of magnitude lower than the corresponding values reported in more recent studies, and those of Blum and Lasaga (1988) which in contrast to all of the other studies suggest that forsterite dissolution rates increase with increasing pH at basic conditions. In contrast to numerous other divalent metal silicates, the dissolution rates of forsterite appear to be independent of aqueous Mg concentration at far-from-equilibrium conditions. This observation was attributed to the fact that there are no Si-O-Si linkages in its structure (Oelkers, 2001). Several different approaches have been proposed to fit forsterite dissolution rates as a function of temperature and solution composition. Pokrovsky and Schott (2000) proposed a surface speciation model to describe these rates. In contrast, Olsen and Rimstidt (2008) and Hänchen et al. (2006) used an empirical model assuming constant temperature rates are solely a function of pH in the absence of organic species. This later assumption was validated by Declercq et al. (2013) who observed little effect of the presence of 13 aqueous organic ligands on forsterite dissolution rates.

The lack of parameters describing the surface speciation of forsterite surfaces at elevated temperatures precludes the general application of the Pokrovsky and Schott (2000) model to describe forsterite dissolution at present. As such an empirical approach has been adopted to describe these rates. Forsterite dissolution rates have been assumed to follow two parallel reaction paths in accord with

\[ r_+ = r_a + r_{H_2O} \quad (F1) \]

where \( r_a \) and \( r_{H_2O} \) refer to rates of two distinct forsterite dissolution pathways, a pH dependent pathway that dominates at pH<9 and a pH independent pathway that dominates at higher pH. These two rate equations are given by

\[ r_a = A_a s \exp\left(-\frac{E_{A,a}}{RT}\right)a_{H^+}^{0.5} \quad (F2) \]
and

\[ r_{H2O} = A_{H2O}^a \exp\left(- \frac{E_{A,H2O}}{RT}\right) \]

where \( A_{A,a} \) and \( A_{A,H2O} \) represent to a pre-exponential factors equal to 1.11x10^5 and 31.88 mol/cm²/s, respectively, and \( E_{A,a} \) and \( E_{A,H2O} \) designate activation energies equal to 55.38 and 71.20 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature.

These parameters were obtained using a least squares fit of the data illustrated in Fig. F1-3. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in these figures. Rates calculated using Equation F-1 to F3 described 54 of 60 reported rates within 0.8 log units.

References


Westrich et al. (1993)


Table F1: Summary of experimentally measured forsterite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Forsterite</td>
<td>25</td>
<td>2</td>
<td>Awad et al. (2000)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>25</td>
<td>1.8-11.35</td>
<td>Blum and Lasaga (1988)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>65</td>
<td>2-5</td>
<td>Chen and Brantley (2000)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>25</td>
<td>1.95-11.85</td>
<td>Golubev et al. (2005)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>1-46</td>
<td>3-5</td>
<td>Grandstaff (1978)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>25</td>
<td>2</td>
<td>Grandstaff (1986)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>90-150</td>
<td>2-10.59</td>
<td>Hänchen et al. (2006)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>25</td>
<td>2</td>
<td>Jonckbloedt et al. (1997)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>25</td>
<td>1.66-9.58</td>
<td>Luce et al. (1972)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>25-65</td>
<td>2</td>
<td>Oelkers et al. (2000)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>25</td>
<td>0.6-6.5</td>
<td>Olsen and Rimstidt (2008)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>120</td>
<td></td>
<td>Prigibbe et al. (2009)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>25</td>
<td>1.03-12.06</td>
<td>Pokrovsky and Schott (2000)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>25-45</td>
<td>1.8-3.8</td>
<td>Rosso and Rimstidt (2000)</td>
</tr>
<tr>
<td>Dunnite</td>
<td>25</td>
<td>4</td>
<td>Siegel and Pfannkuch (1984)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>40-70</td>
<td>1-3</td>
<td>Van Herk et al. (1989)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>25</td>
<td>1.9-6</td>
<td>Westrich et al. (1993)</td>
</tr>
<tr>
<td>Forsterite</td>
<td>25-65</td>
<td>1.8-9.8</td>
<td>Wogelius and Walther (1991)</td>
</tr>
</tbody>
</table>
Figure F1: Summary of experimentally measured forsterite steady-state dissolution rates reported in the literature at 25 °C as a function of pH. The results of the model are plotted as the dashed line.

Figure F2: Summary of experimentally measured forsterite steady-state dissolution rates reported in the literature at 65 °C as a function of pH. The results of the model are plotted as the dashed line.
Figure F3: Arrhenius plot of the data reported at pH 2 and used to calculate $E_A$ and $A_A$. 

\[ y = -3718.6x + 0.5035 \]

- Chen and Brantley (2000)
- Hänechen et al. (2006)
- Oelkers et al. (2006)
- Rosso and Rimstidt (2000)
- Awad et al. (1999)
- Blum and Lasaga (1988)
- Jonckbloedt (1997)
- Pokrovsky and Schott (2000)

Linear (fit)
Glauconite

Glauconite \((K_{0.8}Na_{0.05}Fe_{1.3}Mg_{0.4}Fe_{0.2}Si_{3.8}O_{10}(OH)_2))\) is an hydrated iron-rich micaceous clay mineral of the mica group. Glauconite dissolution was studied by Cloos et al. (1960). A summary of the published measurements conditions are presented in Table Gla1 and the existing rates are depicted in Fig. Gla1.

Assuming a similar dissolution mechanism for micas the dissolution rate data obtained were fitted to:

\[
r_* = A_A \exp \left( -\frac{E_A}{RT} \right) \left( \frac{a_{H^+}}{a_{A^{3+}}} \right)^n
\]

\hspace{\textwidth} (Gla-1)

where \(r_*\) signifies the biotite steady state dissolution rate, \(A_A\) refers to a pre-exponential factor equal to \(1x10^{-7}\) mol/cm²/s, \(E_A\) designates an activation energy equal to \(49.0\pm2\) kJ/mol, \(R\) represents the gas constant, and \(T\) denotes absolute temperature and \(n\) signifies a stoichiometric coefficient equal to 0.125. In the absence of enough available data the different parameters of Eqn. Gla-1 were taken from the glauconite equation Gla-1. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Gla1. Rates calculated using Equation Gla-1 described xx of xx reported rates within 0.8 log unit.

References


Table Gla1: Summary of experimental conditions for the various glauconite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glauconite</td>
<td>25</td>
<td></td>
<td>Cloos et al. (1960)</td>
</tr>
</tbody>
</table>
Figure Gla1: Summary of experimentally measured glauconite steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dashed line.
Glaucophane

Sverdrup (1990) reported the dissolution rates of glaucophane \((\text{Na}_2\text{Mg}_3\text{Al}_2\text{Si}_8\text{O}_{22} \cdot \text{OH})_2\), a Na bearing amphibole at 25 °C and pH ranging from 3.1 to 6.8. These experiments are summarized in Table Gl1 and plotted on Fig. Gl1.

In the absence of extensive rate data, the dissolution rates of glaucophane were assumed to follow a similar behavior to that of hornblende, in accord with:

\[
r_+ = A_A \cdot \exp \left( \frac{-E_A}{RT} \right) \left( \frac{a_{\text{Na}^+}}{a_{\text{Al}^{3+}}} \right)^n \quad \text{Gl-1}
\]

where \(r_+\) signifies the glaucophane steady state dissolution rate, \(A_A\) refers to a pre-exponential factor equal to \(9.8 \times 10^{-2} \text{ mol/cm}^2\text{/s}\), \(E_A\) designates an activation energy equal to 55.8 kJ/mol, \(R\) represents the gas constant, \(T\) denotes absolute temperature, \(n\) signifies a stoichiometric coefficient equal to 0.3. By structural analogy, the \(E_A\) was set to be equal to that of anthophyllite, the only amphibole for which there is sufficient temperature dependent data to constrain an Arrhenius plot. Owing to the dearth of rate data for glaucophane this rate equation is preliminary at best, and should be only used with caution. \(A_A\) was recalculated to fit the Sverdrup (1990) glaucophane data. The degree to which this equation describes these dissolution rates can be assessed in Figure Gl1. Rates calculated using Equation Gl-1 described 3 of 4 reported rates to within 0.2 log units.

References

Table Gl.1: Summary of experimentally measured glaucophane steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glaucophane</td>
<td>25</td>
<td>3 – 6.8</td>
<td>Sverdrup (1990)</td>
</tr>
</tbody>
</table>

Figure Gl.1: Summary of experimentally measured glaucophane steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
**Gibbsite**

Gibbsite is one of the mineral forms of aluminum hydroxide and one of the three minerals making up the rock bauxite. Due to its importance for the industry as one of the main aluminum ore the dissolution kinetics of gibbsite have been investigated by many authors since the 1930s. Early dissolution experiments carried were carried out by Clay and Thomas (1938) showed that the dissolution rate of hydrous alumina in 0.2 M HCl is strongly increased in moderate concentration of some anions (i.e. fluoride, phosphate, phosphite, oxalate and sulphate). They recognized that those effects are due to the formation of a rate-determining complex on the surface of the mineral. Experimental studies by Scoford and Glastonbury (1971, 1972), Packter and Dhillon (1973) and Peric’ et al. (1985) demonstrated that the dissolution kinetics of gibbsite has a first order dependence on NaOH concentration in the strongly alkaline solutions (pH>13.25) relevant to the Al-mining industry.

Packter and Dhillon (1969) also carried out dissolution experiments in acidic solutions at constant molar concentrations and showed that the dissolution rates depends on the type of acid, with faster rates in H$_2$SO$_4$, intermediate rates in HCl and slower rates un HClO$_4$. Bloom (1983) measured the dissolution rate of two synthetic gibbsite samples at 25 °C in HNO$_3$ solutions with pH varying from 1.5 to 3.2. This work was then updated by Bloom and Erich (1987) measuring the dissolution kinetics at 25 °C in the pH interval 1.7-3.9. Nagy and Lasaga (1992) studied dissolution and precipitation rates of gibbsite at 80 °C and pH 3 as a function of chemical affinity. More recently Mogollón et al. (1996) measured the dissolution rate of natural gibbsitic bauxite in HClO$_4$ solutions at 25 °C using column reactors in the pH range 3.3 to 3.9. Mogollón et al. (1999) build upon their previous study by analyzing the dissolution kinetics of gibbsite at 25 °C and 3.5 < pH < 4.5. Ridley et al. (1997) carried out far-from-equilibrium dissolution experiments of gibbsite at 5 °C and near constant pH (=2). They showed that the dissolution kinetic of gibbsite in H$_2$SO$_4$-NaCl solutions are nearly 10 times faster than in HCl-NaCl solutions. Ganor et al. (1999) performed far from equilibrium dissolution experiments of synthetic gibbsite both in a stirred flow through reactor and in a column reactor at 25 °C and outlet pH values from 2.5 to 4.1. Dietzel and Böhme (2005) measured the gibbsite dissolution rates at 20 °C under both far-from-equilibrium and close-to-equilibrium conditions, by using both batch and flow-through reactors from pH 2 to 6.

The data was fitted to the following equation:

$$ r = A_A \exp\left(-\frac{E_A}{RT}\right) \left(\frac{a_{H^+}^2}{a_{Mg^{2+}}^2}\right)^{1/3} $$

where $r$ signifies the glaucophane steady state dissolution rate, $A_A$ refers to a pre-exponential factor equal to 3.9x10$^{-3}$ mol/cm$^2$/sec, $E_A$ designates an activation energy equal to 56 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. $A_A$ and


$E_A$ were estimated from the value of other amphibole. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Ep1. Rates calculated using Equation Gl-1 described 4 of 4 reported rates within 0.1 log units with an average uncertainty of 0.1 log units.

References

**Goethite**

The iron bearing oxide goethite (FeO(OH)) dissolution rate has been studied by Cornell et al. (1976) who reported rate at 20 °C and pH 0.4, by Sidhu et al. (1981) at 25 °C and pH 0.4, by Suter et al. (1991) at 25 °C in the presence of $10^{-3}$ M oxalate. Ruan and Gilke (1995) reported rates as a function of temperature (30-50 °C) and at pH 0.1 and Nowack and Sigg (1997) reported rates at 25 °C in the presence of $1.2 \times 10^{-5}$ M EDTA from pH 3.5 to 10. It can be noted that the values reported by Nowack and Sigg (1997) are inconsistent with the rest of the data, and thus was not included in the model. This inconsistency might stem from the use of EDTA in the experiment. The available data was fitted to:

$$r_+ = A_A \cdot a_{H^+}^{0.4} \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \quad (\text{Goe-1})$$

where $r_+$ the total dissolution rate, $A_A$ refers to a pre-exponential factors equal to $1.95 \times 10^{-6}$ and $3.02 \times 10^{-9}$ mol/cm²/s respectively, $E_A$ designates an activation energy equal to 85.0 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. $E_A$ was calculated by Palandri and Kharaka (2004) based on the values of Ruan and Gilke (1995). The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Goe1. Rates calculated using Equation Goe-1 described 12 of 16 reported rates within 0.8 log.

**References**


Table Goe1: Summary of experimental conditions occurring during goethite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Goethite</td>
<td>20</td>
<td>0.4</td>
<td>Cornell et al. (1976)</td>
</tr>
<tr>
<td>Goethite</td>
<td>25</td>
<td>2.5-10</td>
<td>Nowack and Sigg (1997)</td>
</tr>
<tr>
<td>Goethite</td>
<td>30-50</td>
<td>0.1</td>
<td>Ruan and Gilkes (1995)</td>
</tr>
<tr>
<td>Goethite</td>
<td>25</td>
<td>0.4</td>
<td>Sidhu et al. (1981)</td>
</tr>
<tr>
<td>Goethite</td>
<td>25</td>
<td>2.2-5.8</td>
<td>Suter et al. (1991)</td>
</tr>
</tbody>
</table>

Figure Goe1: Summary of experimentally measured goethite steady-state dissolution rates reported in the literature at 25 °C as a function of pH. The results of the model are plotted as the dotted line.
**Gypsum**

Similar anhydrous analog, anhydrite, there is little quantitative dissolution rate data available for the hydrated calcium sulphate gypsum (CaSO\(_4\).2H\(_2\)O). Also as was the case for anhydrite data are available for only a single pH at any given temperature. Dissolution rates were reported by Barton and Wilde (1971) at 25 °C and pH 6, by Raines and Dewers (1997) at 25 °C and pH 6, Jeschke and Dreybrodt (2001) at 20 °C and pH 6 and Colombani and Bert at 20 °C and pH 5.7. These rates are presented as a function of pH in Figure Gyp1. Owing to the scatter evident in these data any fit would be ambiguous, as such we have adopted the simple Arrhenius fit of anhydrite to describe gypsum dissolution (see above). Although this approach is preliminary at best, the rapid rates of gypsum dissolution likely lead this mineral to be close to equilibrium in most geologic systems of interest. As such the uncertainties associated with this preliminary fit will likely not impact significantly most geological calculations.

The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Gyp1. Rates calculated using Equation Gyp-1 described 9 of 10 reported rates within 0.8 log units.

**References**


Table Gyp1: Summary of experimentally measured gypsum steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gypsum</td>
<td>25</td>
<td>6</td>
<td>Barton and Wilde (1971)</td>
</tr>
<tr>
<td>Gypsum</td>
<td>20</td>
<td>5.7</td>
<td>Colombani and Bert (2007)</td>
</tr>
<tr>
<td>Gypsum</td>
<td>20</td>
<td>6</td>
<td>Jeschke and Dreybrodt (2002)</td>
</tr>
<tr>
<td>Gypsum</td>
<td>25</td>
<td>6</td>
<td>Raines and Dewers (1997)</td>
</tr>
</tbody>
</table>

Figure Gyp1: Summary of experimentally measured gypsum steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the various dashed lines.
The mechanism and rates of the dissolution of hydroxydes of trivalent ions and oxides of trivalent iron or partly containing it (e.g. magnetite and hematite) have been the subject of several studies. Cornell et al. (1976) reports hematite dissolution rates at 20 °C and pH 0.4, Sidhu et al. (1981) reports rates at 25 °C and pH 4. Banwart et al. (1989) report the results of an investigation of the effect of oxalate, acting as complex-former, and L-ascorbic acid, acting as reductant on the dissolution of hematite at pH 3, 25 °C and ionic strength of 0.01 M, in a NaNO3-(HNO3) aqueous medium. The slowest dissolution mechanism was the proton promoted dissolution. The reductive dissolution, assisted by 10 µM ascorbates proceeded faster than previous runs. The fastest dissolution mechanism was achieved by adding 100 µM ascorbate along with 50 µM oxalate. Suter et al. (1991) investigated the reductive dissolution of hematite assisted by ascorbate (10-4 M) in the pH range 2.5-6. Bruno et al. (1992) reported dissolution rates at 25 °C in bicarbonate solutions. The dissolution of Fe(III) hydroxide in 4.8 x 10^{-2} M ascorbate was also investigated by Deng (1997) as a function of both pH, from 4.3 to 7.6, and temperature, from 20 to 60 °C. He found that at 20 C the reductive dissolution rates are pH independent from pH 4.3 to 6. In contrast the rates decrease substantially with increasing pH in the pH interval 6-7.6. In agreement with what was observed by Suter et al. (1991). As shown by Deng (1997) these pH effects are consistent with the pH dependence of ascorbate adsorption onto Fe(III) hydroxide. The apparent activation energy resulted to be 27 kJ/mol at pH 6 and 47 kJ/mol at pH 7.5. Dos Santos and Stumm (1992) studied the reductive dissolution of hematite governed by H2S under constant P_{H2S} of 10^{-4}, 10^{-3} and 10^{-2} atm at 25 °C and pH 3-7, below saturation with iron sulfide. Hematite dissolution proceeds very fast under these conditions.

These experimental conditions are reported in Table Hem1 and the data points plotted on Fig. Hem1. The data was fitted by Palandri and Kharaka (2004) to the following equation:

\[ r_{H^+} = A_A \cdot a_{H^+}^{0.421} \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \]

\[ r_{H_2O} = A_A \cdot \exp \left( \frac{-E_B}{R \cdot T} \right) \]
2014 \[
    r_{tt} = r_{H^+} + r_{H_2O}
\]

where \( r_{H^+} \) and \( r_{H_2O} \) signifies the hematite steady-state dissolution rate in acidic, neutral and basic solutions and \( r_{tt} \) the total dissolution rate, \( A_A \) and \( A_B \) refer to pre-exponential factors equal to 1.95x10\(^{-6}\) and 3.02x10\(^{-9}\) mol/cm\(^2\)/s respectively, \( E_A \) designates an activation energy equal to 37.9 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. In the absence of additional and more extensive data the Palandri and Kharaka fit has been adopted in this study. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Hem1. Rates calculated using Equation Hem-1 described 4 of 36 reported rates within 0.8 log units.

References


Table He1: Summary of experimental conditions for the various hornblende steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hematite</td>
<td>25</td>
<td>3</td>
<td>Banwart (1989)</td>
</tr>
<tr>
<td>Hematite</td>
<td>20</td>
<td>0.4</td>
<td>Cornell et al. (1976)</td>
</tr>
<tr>
<td>Hematite</td>
<td>25</td>
<td>4.5 – 7.5</td>
<td>Deng (1997)</td>
</tr>
<tr>
<td>Hematite</td>
<td>25</td>
<td>3 - 11.5</td>
<td>Dos Santos and Stumm (1992)</td>
</tr>
<tr>
<td>Hematite</td>
<td>25</td>
<td>3</td>
<td>Larsen and Postma (2001)</td>
</tr>
<tr>
<td>Hematite</td>
<td>25-50</td>
<td>0.1</td>
<td>Ruan and Gilke (1995)</td>
</tr>
<tr>
<td>Hematite</td>
<td>25</td>
<td>0.4</td>
<td>Sidhu et al. (1981)</td>
</tr>
<tr>
<td>Hematite</td>
<td>25</td>
<td>2.5 - 6</td>
<td>Sutter et al (1991)</td>
</tr>
</tbody>
</table>

Figure He1: Summary of experimentally measured hematite steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 ºC are plotted in dashed line.
**Grossular**

The dissolution rates of the calcium-aluminum garnet grossular $\text{(Ca}_3\text{Al}_2\text{(SiO}_4)_3}$ were studied by Sverdrup (1990) at 25 °C from pH 2.5 to 6.4. The data was fitted by Palandri and Kharaka (2004) to the following equation:

$$r_{H^+} = A_A \cdot a_{H^+}^1 \cdot \exp\left(\frac{-E_A}{R \cdot T}\right)$$

$$r_{H_2O} = A_B \cdot \exp\left(\frac{-E_B}{R \cdot T}\right)$$

$$r_{tt} = r_{H^+} + r_{H_2O}$$

where $r_{H^+}$ and $r_{H_2O}$ signifies the grossular steady-state dissolution rate in acidic and neutral solutions and $r_{tt}$ the total dissolution rate, $A_A$ and $A_B$ refer to pre-exponential factors equal to 6.17x10^5 and 3.02x10^3 mol/cm²/s respectively, $E_A$ and $E_B$ designates an activation energy equal to 85.0 and 103.8 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. As was the case for the garnets andradite and almandine, these activation energies are based on the fit of rates obtained at 8 and 25 °C, and the rate data at 8 °C were never directly reported. As such the degree to which these activation energies provide accurate rate estimates at substantially higher temperatures is questionable. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Gro1. Rates calculated using Equation Gro-1 described 6 of 6 reported rates within 0.8 log units.

**References**


Table Gro1: Summary of experimentally measured grossular steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Grossular</td>
<td>25</td>
<td>2.5-6.4</td>
<td>Sverdrup (1990)</td>
</tr>
</tbody>
</table>

Figure Gro1: Summary of experimentally measured grossular steady-state dissolution rates reported in the literature as a function of pH. The dotted line represents the results of the model.
**Halite**

The mineral form of NaCl, halite (NaCl) or rock salt has been studied by Alkattan et al. (1997a, 1997b) in rotating disk reactors at temperature ranging from 25 to 80 °C as a function of rotation speed and trace metal concentration. The available data is plotted on fig. Hal1 and fit to:

\[ r_s = A_A \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \]  

(Hal1)

where \( r_s \) signifies the halite forward dissolution rate, \( A_A \) refers to pre-exponential factors equal to 3.11x10^{-8} mol/cm²/s, \( E_A \) designates an activation energy equal to -22.5kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. This fit differs from that reported by Palandri and Kharaka (2004), these authors apparently mistakenly fit NaCl diffusion rates reported by Alkattan et al. (1997a, b) rather than dissolution rates. Note, the activation energy calculated from the Alkattan et al (1997b) data is negative suggesting halite dissolution rates decrease somewhat with increasing temperature. Furthermore there is no pH dependence implied by our fit, the degree to which halite dissolution rates depend on pH cannot be assessed at present due to the lack of relevant data. The ability of equation Hal-1 equation to describe the dissolution rates reported in the literature can be assessed in Figure Hal1. Rates calculated using Equation Hal-1 described all of the reported rates within 0.1 log units.

**References**


Table Hal1: Summary of experimentally measured halite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Halite</td>
<td>25 - 80</td>
<td>7</td>
<td>Alkattan et al. (1997a)</td>
</tr>
<tr>
<td>Halite</td>
<td>25</td>
<td>7</td>
<td>Alkattan et al. (1997b)</td>
</tr>
</tbody>
</table>

Figure Hal1: Summary of experimentally measured halite steady-state dissolution rates reported in the literature at 50 °C as a function of pH and at pH 5.7 as a function of temperature. The results of the model are plotted as the dotted line.
**Hornblende**

Hornblende \((\text{Ca}_2\text{Mg}_4\text{Al}_{1.75}\text{Fe}_{0.25}\text{Si}_7\text{O}_{22}(\text{OH})_2)\) is an amphibole variable composition typical of igneous rocks. It commonly has a chemical formula similar to that of tremolite, with substantial Fe and Al substituted for Mg and Si in its structure. There have been several studies of the dissolution rates of the amphibole hornblende. Nickel (1973) reported hornblende dissolution rates at 25 °C as a function of pH from 1 to 5, and Cygan et al. (1989) reported rates at pH 4 and 25 °C. Sverdrup (1990) reported hornblende dissolution at 25 °C and pH from 1.6 to 7. Zhang et al. (1993) and Swoboda-Colberg and Drever (1993) reported rates at 25 °C and pH from 3.6 to 4 and 4.5 respectively. Frogner and Schweda (1998) reported rates at 25 °C and pH 1 to 5, Golubev et al. (2005) experimentally determined the rates at 25 °C as a function of pH \((2.1 – 11.1)\) and experiment duration. Brantley (2008) reported rates attributed to Givens and Brantley at 25 °C and pH from 1.1 to 6.0. These experimental conditions are reviewed in Table Ho1 and the data points plotted on Fig. Ho1.

Large inconsistencies are evident amongst these rates; at pH of ~3 there is nearly 4 orders of magnitude difference between the fastest and slowest rates, some of this large discrepancy may be attributable to the extensive amount of time required to achieve steady state hornblende dissolution. Golubev et al. (2005) observed that dissolution rates obtained from flow-reactor experiments running 1000 hours were approximately one order of magnitude slower than those ran for 100 hours. Moreover, of the earlier studies, only Frogner and Schweda (1998) conducted experiments which were demonstrated to exhibit stoichiometric metal release. An additional source of scatter may be the result of differences in the compositions of the hornblendes used in the various studies.

The attempts to accurately fit hornblende forward dissolution rates are greatly confounded by the scatter of the data. This scatter also obscures the choice of rate equation. In the absence of definitive data, it is assumed that the hornblende dissolution is similar in rate behavior to tremolite. As such we have described hornblende rates using the tremolite rate equation:

\[
r_+ = A_0 \exp \left( \frac{-E_A}{RT} \right) \left( \frac{a_{H^{+}}}{a_{Mg^{2+}}} \right)^n \tag{Ho-1}
\]

together with values of \(E_A\) and \(n\) deduced from the fit of tremolite dissolution rate data.
55.8 kJ/mol, and 0.125, respectively). To best describe the scattered hornblende rate data, $A_A$ was set to be 0.5 log units higher than that determined for tremolite. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Ho1. Rates calculated using Equation Ho-1 described 18 of 42 reported rates within 0.9 log units.

**References**


Table Ho1: Summary of experimental conditions for the various hornblende steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hornblende</td>
<td>25</td>
<td>4</td>
<td>Cygan et al. (1989)</td>
</tr>
<tr>
<td>Hornblende</td>
<td>25</td>
<td>1 - 5</td>
<td>Frogner and Schweda (1998)</td>
</tr>
<tr>
<td>Hornblende</td>
<td>25</td>
<td>1.15-5.67</td>
<td>Givens and Brantley</td>
</tr>
<tr>
<td>Hornblende</td>
<td>25</td>
<td>2.1 – 11.1</td>
<td>Golubev et al. (2005)</td>
</tr>
<tr>
<td>Hornblende</td>
<td>25</td>
<td>3.6 – 10.6</td>
<td>Nickel (1973)</td>
</tr>
<tr>
<td>Hornblende</td>
<td>25</td>
<td>1.6 - 7</td>
<td>Sverdrup (1990)</td>
</tr>
<tr>
<td>Hornblende</td>
<td>25</td>
<td>4, 4.5</td>
<td>Swobodan-Colberg and Drever (1993)</td>
</tr>
<tr>
<td>Hornblende</td>
<td>25</td>
<td>3.6 - 4</td>
<td>Zhang (1993)</td>
</tr>
</tbody>
</table>

Figure Ho1: Summary of experimentally measured hornblende steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dotted line.
Illite

Relatively few studies have been performed to determine the rates of illite ($K_{0.6}Mg_{0.25}Al_{1.8}Al_{1.5}Si_{3.5}O_{10}(OH)_2$) dissolution. Early studies were carried out by Heydeman (1966) and Feigenbaum and Shainberg (1975) while more recent work was carried out by Köhler et al. (2003). Heydeman (1966) measured illite dissolution rates in batch reactors at pH 2.5 to 10 and 25 °C. Feigenbaum and Shainberg (1975) determined dissolution rates of the same illite as Heydeman (1966) at 5 and 25 °C and a pH close to 2.9 and 7.5. Nagy (1995) recalculated both the rates of Heydemann (1966) and Feigenbaum and Shainberg (1975) based on Si release. Holmqvist et al. (2001) reported dissolution rate from 6 to 23 °C and pH ranging from 1 to 6. Köhler et al. (2003) investigated the dissolution rates of illite in batch reactors as a function of both pH, from 1.4 to 12.4, and temperature from 5 to 50 °C.

Köhler et al. (2003) provided the following empirical equation describing illite dissolution as a function of pH and temperature:

$$r_+ = 2.2 \times 10^{-8} \cdot a_H^{0.6} \cdot \exp\left(\frac{E_A}{RT}\right) + 2.5 \times 10^{-17} \cdot \exp\left(\frac{E_B}{RT}\right) + 2.7 \times 10^{-5} \cdot a_{OH}^{-0.6} \cdot \exp\left(\frac{E_C}{RT}\right)$$

where $r_+$ signifies the illite steady state dissolution rate, $E_A$, $E_B$ and $E_C$ designates activation energies, $R$ represents the gas constant, and $T$ denotes absolute temperature. The rate constants and activation energies were calculated by Köhler et al. (2003) with $E_A$, $E_B$ and $E_C$ equal to 46, 14 and 67 kJ/mol respectively. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure II-1. Rates calculated using Equation II-1 described 26 of 28 reported rates within 0.8 log. It should be kept in mind that the structure and formula of illite approximates that of muscovite. As such it might be anticipated that illite far-from-equilibrium dissolution rates depend on the aqueous aluminum and to some extent the aqueous silica concentration in the reactive fluid. Such a dependence of illite dissolution rates on these species is precluded at present by the dearth of relevant rate data.
References


Table II1: Summary of experimental conditions for the various illite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Illite</td>
<td>5-25</td>
<td>2.9-7.5</td>
<td>Feigenbaum and Shainberg (1975)</td>
</tr>
<tr>
<td>Illite</td>
<td>25</td>
<td>2.5-10</td>
<td>Heydeman (1966)</td>
</tr>
<tr>
<td>Kungsängen illite</td>
<td>6-23</td>
<td>1-6</td>
<td>Holmqvist et al. (2001)</td>
</tr>
<tr>
<td>Illite du Puy</td>
<td>5-50</td>
<td>1.4-12.4</td>
<td>Köhler et al. (2003)</td>
</tr>
</tbody>
</table>

Figure II1: Summary of experimentally measured illite steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 5, 25 and 50 °C are plotted in dotted, dashed and dotted-dashed lines respectively.
Ilmenite

The weakly magnetic titanium oxide mineral ilmenite dissolution has been studied by White et al. (1994) who reported rates at 25 C and pH 1 to 6. The available rate data was fitted by Palandri and Kharaka (2004) to:

\[ r_{H^+} = A_A \cdot a_{H^+}^{0.421} \cdot e^{(-E_A / RT)} \]

\[ r_{H_2O} = A_A \cdot e^{(-E_B / RT)} \]

\[ r_{tt} = r_{H^+} + r_{H_2O} \]

where \( r_{H^+} \) and \( r_{H_2O} \) signifies the ilmenite steady-state dissolution rate in acidic, neutral and basic solutions and \( r_{tt} \) the total dissolution rate, \( A_A \) and \( A_B \) refer to pre-exponential factors equal to 1.95x10^-6 and 3.02x10^-9 mol/cm²/s respectively, \( E_A \) designates an activation energy equal to 37.9 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. In the absence of additional and more extensive data the Palandri and Kharaka fit has been adopted in this study. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Ilm1. Rates calculated using Equation Ilm-1 described all reported rates within 0.8 log.

References


Table Ilm1: Summary of experimental conditions occurring during ilmenite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ilmenite</td>
<td>25</td>
<td>1.1-6.0</td>
<td>White et al. (1993)</td>
</tr>
</tbody>
</table>

Figure Ilm1: Summary of experimentally measured ilmenite steady-state dissolution rates reported in the literature at 25 °C as a function of pH. The results of the model are plotted as the dotted line.
Quartz

The quartz is one of the most studied mineral of all time. The pH dependence of the dissolution rates of silicates has been the subject of numerous studies, cf. Dove (1995). Knauss and Wolery (1988) studied the dissolution of quartz at pH from 1.4 to 11.8 at 70 °C. Tester et al. (1994) investigated the dissolution kinetics of quartz in pure water and also reported the rates obtained by 10 previous studies by Kitahara (1960), Van Lier et al. (1960), Siebert et al (1963), Weil and Fyfe (1964), Rimstidt and Barnes (1980), Blum et al. (1990), Brady and Walter (1990), Dove and Crerar (1990), Bennet (1991) and Berger et al. (1994a). Tester obtained two series of dissolution data on temperature ranging from 25 to 625 °C. Dove and Elston (1992) described the kinetics of quartz at 25 °C, pH 2 to 12 and NaCl concentration from 0 to 0.5 M. Dove (1994) extended those results to include temperature up to 300 °C. Dove and Crerar (1990) investigated the dissolution of quartz at circumneutral pH in the temperature interval 100-300 °C. Gislason et al. (1997) also measured at pH 3.5, temperature of 25 to 200 °C and far from equilibrium conditions the dissolution of quartz.

References


Casey


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Table Qu1: Summary of experimental conditions for the various quartz steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Quartz</td>
<td>25</td>
<td>4</td>
<td>Bennet et al. (1988)</td>
</tr>
<tr>
<td>Quartz</td>
<td>25</td>
<td>1 - 5</td>
<td>Bennet (1991)</td>
</tr>
<tr>
<td>Quartz</td>
<td>25</td>
<td>2.1 – 11.1</td>
<td>Berger et al. (1994)</td>
</tr>
<tr>
<td>Quartz</td>
<td>25</td>
<td>3.6 – 10.6</td>
<td>Brady and Walther (1990)</td>
</tr>
<tr>
<td>Quartz</td>
<td>25</td>
<td>1.6 - 7</td>
<td>Casey et al. (1990)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Dove and Crerar (1990)</td>
</tr>
<tr>
<td>Quartz</td>
<td>25</td>
<td>4, 4.5</td>
<td>Dove (1994)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Dove (1999)</td>
</tr>
<tr>
<td>Quartz</td>
<td>70</td>
<td></td>
<td>House and Orr (1992)</td>
</tr>
<tr>
<td>Quartz</td>
<td>25</td>
<td>3.6 - 4</td>
<td>Wollast and Chou (1988)</td>
</tr>
<tr>
<td>Quartz</td>
<td></td>
<td></td>
<td>Knauss and Copenhaver (1995)</td>
</tr>
<tr>
<td>Quartz</td>
<td></td>
<td></td>
<td>Tester et al. (1994)</td>
</tr>
</tbody>
</table>
25 C

-19 -18 -17 -16 -15 -14
log r+ (mol/cm^2/s)

0 2 4 6 8 10 12
pH

- Dove (1994)
- Brady and Walther (1990)
- Bennett et al. (1988)
- Bennet (1991)
- Berger et al. (1994)
- House and Orr (1992)
+ Wollast and Chou (1988)
- Model Bandtra et al 2005
**Jadeite**

Jadeite is an inosilicate with a clinopyroxene structure, where Si-O chains are held together with Na and Al bridges. Its dissolution kinetics were only reported by Sverdrup (1990) at 25 °C and pH ranging from 3 to 6. These 3 data points are plotted on Fig. Ja1 and listed in Table Ja1.

Owing to its structure, and by analogy with Al-silicate minerals it seems reasonable to assume that its dissolution mechanism consists of the sequential removal of Na then Al via proton exchange reactions prior to the final removal of Si-O tetrahedra from the mineral. In such cases dissolution rates are commonly consistent with:

\[
\frac{r_{+}}{A_{\text{A}}} = \exp \left( \frac{-E_{\text{A}}}{RT} \right) \left( \frac{a_{n+}^{3}}{a_{\text{Al}^{3+}}} \right)^{n}
\]

where \( r_{+} \) signifies the jadeite steady state dissolution rate, \( A_{\text{A}} \) refers to a pre-exponential factor equal to 1.5x10\(^{-3}\) mol/cm\(^2\)/sec, \( E_{\text{A}} \) designates an activation energy equal to 48 kJ/mol, \( n \) refers to a constant equal to 0.25, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. In the absence of any measured dependence of rates of temperature, \( E_{\text{A}} \) was set to be equal to 48 kJ/mol by analogy with the amphiboles (reference), chain silicates also held together by bridging Al-octahedrals. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Ja1. Rates calculated using Equation Ja-1 described 3 of 3 reported rates within 0.1 log units with an average uncertainty of 0.1 log units.

**References**

Table Ja1: Summary of experimentally measured jadeite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Jadeite</td>
<td>25</td>
<td>3, 4.5, 6</td>
<td>Sverdrup (1990)</td>
</tr>
</tbody>
</table>

Figure Ja1: Summary of experimentally measured jadeite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
Kaolinite

The dissolution rate of kaolinite under controlled temperature and pH conditions has been the subject of many investigations. Kaolinite dissolution kinetics was studied by Turner (1964) at 25 °C and pH ranging from -1 to 7. Polzer and Hem (1965) reported rate data at 25 °C and pH 4 and Jörgensen and Mitchell (1970) reported rates at 25 °C and pH of 3.6 and 6. The dissolution kinetics was investigated at pH 1 to 12 by Caroll and Walther (1990) at temperatures of 25 to 80 °C. Wieland and Stumm (1992) measured kaolinite dissolution between pH 2 and 6.5 at 25 °C while Ganor et al. (1994) reported rates measured at pH 3 to 4.2 and temperature ranging from 25 to 80 °C. Devidal et al. (1997) observed kaolinite dissolution rates at 150 °C, 40 bars of CO₂ and at pH from 2 to 7.8. Kaolinite dissolution kinetics were investigated in 0.1-4 M KOH solutions at 35 and 80 °C by Bauer and Berger (1998). Metz and Ganor (2001) reported rates from 25 to 80 °C and pH ranging from 3.0 to 4.2, Huertas et al. (1999) studied kaolinite dissolution at 25 °C and at pH from 1 to 13 and Cama et al. (2002) reported rates taken from pH 1 to 5 and temperature ranging from 25 to 70 °C. Yang and Steefel (2008) reported kaolinite dissolution rates at 22 °C and pH of 4. The different conditions where the data was acquired are summarized in table Ka1. The rates are presented for the different temperatures as a function of pH in Figure Ka1, except for the rates obtained at 150 °C by Devidal et al. (1997).

The published data behavior presented in Fig. Ka1 was obtained by summing the contributions of the acidic, neutral and basic mechanisms reported by Palandri and Kharaka (2004):

\[ r_{H^+} = A_A \cdot \exp \left( - \frac{E_A}{RT} \right) (a_{H^+}^{n_A}) \]  

Ka-1

\[ r_w = A_B \cdot \exp \left( - \frac{E_B}{RT} \right) \]  

Ka-2

\[ r_{OH^-} = A_C \cdot \exp \left( - \frac{E_C}{RT} \right) (a_{OH^-}^{0.55}) \]  

Ka-3

\[ r_v = r_{H^+} + r_w + r_{OH^-} \]  

Ka-4
where $r_i$ signifies the kaolinite forward dissolution rate, $A_A$, $A_B$ and $A_C$ refer to pre-exponential factors equal to respectively $6.45 \times 10^{-9}$, $4.36 \times 10^{-17}$ and $1.94 \times 10^{-4}$ mol/cm²/sec, $E_A$, $E_B$ and $E_C$ designate the activation energies of their respective mechanisms equal to 46, 14 and 67 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. The pre-exponential factors and activation energies were determined from an Arrhenius plot of the available data. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Ka1. Rates calculated using Equation Ka-4 described 50 of 205 reported rates within 0.8 log units.

References


Table Ka1: Summary of experimental conditions occurring during kaolinite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>kaolinite</td>
<td>35, 80</td>
<td>11.5 – 13.9</td>
<td>Bauer and Berger (1998)</td>
</tr>
<tr>
<td>kaolinite</td>
<td>25</td>
<td>1 - 5</td>
<td>Cama et al. (2002)</td>
</tr>
<tr>
<td>kaolinite</td>
<td>25, 60, 80</td>
<td>1 - 12</td>
<td>Caroll and Walter (1990)</td>
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<td>kaolinite</td>
<td>150</td>
<td>2, 6.8, 7.8</td>
<td>Devidal et al. (1997)</td>
</tr>
<tr>
<td>kaolinite</td>
<td>25, 50, 80</td>
<td>3 - 4.2</td>
<td>Ganor et al. (1995)</td>
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<td>kaolinite</td>
<td>25</td>
<td>1 - 13</td>
<td>Huertas et al. (1999)</td>
</tr>
<tr>
<td>kaolinite</td>
<td>25</td>
<td>3.6, 6</td>
<td>Jørgensen and Mitchell (1970)</td>
</tr>
<tr>
<td>Kaolinite</td>
<td>25-70</td>
<td>2-3</td>
<td>Metz and Ganor (2001)</td>
</tr>
<tr>
<td>Kaolinite</td>
<td>25</td>
<td>4</td>
<td>Polzer and Hem (1965)</td>
</tr>
<tr>
<td>Kaolinite</td>
<td>25</td>
<td>-1-7</td>
<td>Turner (1964)</td>
</tr>
<tr>
<td>kaolinite</td>
<td>25</td>
<td>2 - 6.5</td>
<td>Wieland and Stumm (1992)</td>
</tr>
<tr>
<td>kaolinite</td>
<td>22</td>
<td>4</td>
<td>Yang and Steefel (2008)</td>
</tr>
</tbody>
</table>
Figure Ka1: Summary of experimentally measured K-feldspar steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.

\[ y = 0.058x^2 - 0.7314x - 15.97 \]
K-Feldspar

Dissolution rates of K-feldspar have been published by Bevan and Savage (1989) at temperature ranging from 25 to 95°C and pH of 1 to 9, Busenberg and Clemency (1976) studied the K-feldspar dissolution at pH 5 and 24°C while Gautier et al. (1994) measured the rates at pH 9 and 150°C as a function of ΔG. Knauss and Copenhaver (1994) reported dissolution rates at 70°C and between pH 4 and 10. Siegel and Pfannkuch (1984) published dissolution rates at 25°C and pH 4 while Stillings et al. (1996) studied the dissolution kinetics of K-feldspar at 25°C and pH ranging from 3 to 7. The different conditions where the data was acquired are summarized in Table Kf1. The rates are presented for the different temperatures as a function of pH in Figure Kf1.

The dissolution rates of K-feldspar were fitted by Gautier et al. (1994) to the following equation:

\[
r = A_A \cdot (1 - \exp\left(-\frac{E_A}{3RT}\right)) \left(\frac{a_{H^+}}{a_{Al^{3+}}}\right)^{1/3}
\]

(Kf-1)

where \(r\) signifies the K-feldspar steady state dissolution rate, \(A_A\) refers to a pre-exponential factor equal to \(2.77 \times 10^{-13}\) mol/cm²/sec, \(E_A\) designates an activation energy equal to 57.34 kJ/mol, \(R\) represents the gas constant, and \(T\) denotes absolute temperature. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Kf1. Rates calculated using Equation Kf-1 described 12 of 14 reported rates within 0.8 log units with an average uncertainty of 0.2 log units.

References


Table Kf1: Summary of experimentally measured forsterite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Orthoclase</td>
<td>70, 90</td>
<td>1, 4, 9</td>
<td>Bevan and Savage (1989)</td>
</tr>
<tr>
<td>Orthoclase, Microcline</td>
<td>25, 70</td>
<td>5</td>
<td>Busenberg and Clemency (1976)</td>
</tr>
<tr>
<td>K-feldspar</td>
<td>150</td>
<td>9</td>
<td>Gautier et al. (1994)</td>
</tr>
<tr>
<td>K-feldspar</td>
<td>70</td>
<td>4-10</td>
<td>Knauss and Copenhaver (1995)</td>
</tr>
<tr>
<td>K-Feldspar</td>
<td>25</td>
<td>3.82-5.65</td>
<td>Manley and Evans (1986)</td>
</tr>
<tr>
<td>Microcline</td>
<td>25</td>
<td>4</td>
<td>Siegel and Pfannkuch (1984)</td>
</tr>
<tr>
<td>K-feldspar</td>
<td>25</td>
<td>3-7</td>
<td>Stillings et al. (1996)</td>
</tr>
<tr>
<td>K-feldspar</td>
<td>25</td>
<td>4, 4.5</td>
<td>Swoboda-Colberg and Drever (1993)</td>
</tr>
<tr>
<td>K-feldspar</td>
<td>25</td>
<td>7</td>
<td>Tan (1980)</td>
</tr>
</tbody>
</table>

Figure Kf1: Summary of experimentally measured K-feldspar steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
Busenberg and Clemency (1976), 25 °C
Bevan and Savage (1989), 25 °C
Bevan and Savage (1989), 70 °C
Gautier et al. (1994), 150 °C
Knauss and Copenhaver (1995), 70 °C
Siegel and Pfannkuch (1984), 25 °C
Stillings et al. (1996), 25 °C

Model (25 °C)
Model (70 °C)
Model (150 °C)
Kyanite steady state dissolution rates were reported by Nickel (1973) at 25 °C and pH from 0.2 to 11 and measured in mixed flow reactors as a function of aqueous aluminum and silica concentration at pH 1.6 to 2.2 and at temperatures from 108 to 194°C by Oelkers and Schott (1999). These experimental conditions are summarized in table Ky1. The rates obtained at pH 2 are presented as a function of the inverse of temperature in Figure Ky1.

All measured rates, which were obtained at far from equilibrium conditions where the contribution of reverse reaction is negligible, are consistent with Oelkers and Schott (1999):

\[
    r_\oplus = A_A \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \left( \frac{a_{H^+}^{3+}}{a_{Al^{3+}}} \right)^{0.5}
\]  

(Ky-1)

where \( r_\oplus \) signifies the overall kyanite steady state dissolution rate normalized to a 1 cm² surface, \( A_A \) refers to a pre-exponential factor equal to 2x10⁻⁵ mol/cm²/sec, \( E_A \) designates an apparent activation energy equal to 75 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Ky1. Rates calculated using Equation Ky-1 described all of the reported rates by Oelkers and Schott (1999) within 0.8 log units.

References


Table Ky1: Summary of experimentally measured kyanite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kyanite</td>
<td>108-194</td>
<td>1.6-2.2</td>
<td>Oelkers and Schott (1999)</td>
</tr>
<tr>
<td>Kyanite</td>
<td>25</td>
<td>0.2 - 11</td>
<td>Nickel (1973)</td>
</tr>
</tbody>
</table>

Figure Ky1: Summary of experimentally measured kyanite steady-state dissolution rates reported in the literature as a function of $1/T$ and pH. The results of the model are plotted as the dotted line.

- $r_+$ (mol/cm$^2$/s)
- $1/T$ (K)
- pH
**Leucite**

The potassium aluminum feldspathoid leucite (KAlSi$_2$O$_6$) dissolution has been studied by Krueger (1939) and reported by Sverdrup (1990) at 25 °C and from pH 0 to 11. Although Sverdrup (1990) suggested that the dissolution mechanism of leucite would be similar to that of the pyroxene, especially jadeite, its structure most closely approaches that of the feldspathoid nepheline. In the absence of sufficient data to deduce a more comprehensive rate description and to be consistent with the rates adopted in this study for nepheline we fit the available leucite dissolution rate data to the equations reported by Palandri and Kharaka (2004) for:

\[
\begin{align*}
    r_{H^+} &= A_A \cdot a_{H^+}^{0.7} \cdot \exp\left(\frac{-E_A}{R \cdot T}\right) \\
    r_{H_2O} &= A_B \cdot \exp\left(\frac{-E_B}{R \cdot T}\right) \\
    r_{OH^-} &= A_C \cdot a_{OH^-}^{0.2} \cdot \exp\left(\frac{-E_C}{R \cdot T}\right) \\
    r_t &= r_{H^+} + r_{H_2O} + r_{OH^-}
\end{align*}
\]

where $r_{H^+}$, $r_{H_2O}$ and $r_{OH^-}$ signify the leucite forward dissolution rate in acidic, neutral and basic solutions and $r_t$ the total dissolution rate, $A_A$, $A_B$ and $A_C$ refer to pre-exponential factors equal to $1.21 \times 10^{13}$, 1.08 and $8.69 \times 10^{-6}$ mol/cm$^2$/s respectively, $E_A$, $E_B$ and $E_C$ designates an activation energy equal to 132.2, 75.5 and 56.6 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. The origin of these activation energies is, however, unclear as we have been unable to locate the source of leucite rate data at temperatures other then 25 °C. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Leu1. Rates calculated using Equation Leu-1 described all reported rates within 0.8 log units.
References


Table Leu1: Summary of experimentally measured leucite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Leucite</td>
<td>25</td>
<td>0 - 11</td>
<td>Krueger (1939) <em>in</em> Sverdrup (1990)</td>
</tr>
</tbody>
</table>

Figure Leu1: Summary of experimentally measured leucite steady-state dissolution rates reported in the literature as a function of pH. The dotted line represents the results of the model.

![Figure Leu1](image-url)
Magnesite

Few kinetics studies have been carried out on magnesite (MgCO$_3$), with respect to the work done on calcite dissolution. At 25 °C Chou et al. (1989) reported a linear dependence of magnesite dissolution rate as a function of proton activity in the 2<pH<5 range while Pokrovsky and Schott (1999) report nearly pH-independent rates in the range 0<pH<3 and rates proportional to the proton activity in the range 3<pH<5. Higgins et al. (2002) reported dissolution rates measured with bulk techniques and AFM at 60 °C and pH from 2 to 4.5., Pokrovsky et al. (2003) reported rates at 25 °C and pH ~3.5 as a function of pCO$_2$, Jordan et al. (2007) reported rates at 100 °C at pH ranging from 5.55 to 9.7. The conditions where the data was acquired are summarized in table Mag1 and the available rate data are presented in Fig. Mag1.

The available data was fitted to an equation from Busenberg and Plummer (1982) adapted by Chou et al. (1989):

$$
\begin{align*}
 r_{H^+} &= A_A \cdot \exp\left(\frac{-E_A}{R \cdot T}\right) \cdot (a_{H^+})^n \\
 r_{H_2CO_3} &= A_B \cdot \exp\left(\frac{-E_B}{R \cdot T}\right) \cdot (a_{H_2CO_3})^n \\
 r_{H_2O} &= A_C \cdot \exp\left(\frac{-E_C}{R \cdot T}\right) \cdot (a_{H_2O})^n \\
 r_{tt} &= r_{H^+} + r_{H_2CO_3} + r_{H_2O}
\end{align*}
$$

where $r_{H^+}$, $r_{H_2CO_3}$ and $r_{H_2O}$ signifies the magnesite forward dissolution rate in acidic, neutral and basic solutions and $r_{tt}$ the total dissolution rate, $A_A$, $A_B$ and $A_C$ refer to pre-exponential factors equal to 8.86, 1x10$^{-3}$ and 2.2x10$^{-8}$ mol/cm$^2$/s respectively, $E_A$, $E_B$ and $E_C$ designate activation energies equal to 34.0, 21.0 and 16.0 kJ/mol, $R$ represents the gas constant, $T$ denotes absolute temperature and $n$ signifies a stoichiometric coefficient equal to 0.75. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Mag1. Rates calculated using Equation Mag-1 described xx of xx reported rates within 0.8 log units.
References


Table Mag1: Summary of experimentally measured magnesite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Magnesite</td>
<td>25</td>
<td>2.1 - 8</td>
<td>Chou et al. (1989)</td>
</tr>
<tr>
<td>Magnesite</td>
<td>60</td>
<td>2 - 4.5</td>
<td>Higgins et al. (2002)</td>
</tr>
<tr>
<td>Magnesite</td>
<td>100</td>
<td>5.55 – 9.7</td>
<td>Jordan et al. (2007)</td>
</tr>
<tr>
<td>Magnesite</td>
<td>25</td>
<td>0.2 – 11.4</td>
<td>Pokrovsky and Schott (1999)</td>
</tr>
<tr>
<td>Magnesite</td>
<td>25</td>
<td>3.2 – 4.5</td>
<td>Pokrovsky et al. (2003)</td>
</tr>
</tbody>
</table>
Figure Mag1: Summary of experimentally measured magnesite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
**Magnetite**

The iron oxide mineral magnetite (Fe$_3$O$_4$) dissolution has been studied by White et al. (1994) who reported rates at 25 °C and pH 1 to 6. The available rate data was fitted by Palandri and Kharaka (2004) to:

\[
\begin{align*}
  r_{H^+} &= A_A \cdot a_{H^+}^{0.279} \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \\
  r_{H_2O} &= A_A \cdot \exp \left( \frac{-E_B}{R \cdot T} \right) \\
  r_{tt} &= r_{H^+} + r_{H_2O}
\end{align*}
\]

where $r_{H^+}$ and $r_{H_2O}$ signifies the magnetite steady-state dissolution rate in acidic, neutral and basic solutions and $r_{tt}$ the total dissolution rate. $A_A$ and $A_B$ refer to pre-exponential factors equal to $4.68 \times 10^{-10}$ and $3.02 \times 10^{-12}$ mol/cm$^2$/s respectively, $E_A$ designates an activation energy equal to 18.6 kJ/mol (Palandri and Kharaka, 2004), $R$ represents the gas constant, and $T$ denotes absolute temperature. In the absence of additional and more extensive data the Palandri and Kharaka fit has been adopted in this study. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Mag1.

Rates calculated using Equation Ilm-1 described all reported rates within 0.8 log.

**References**


Table Mag1: Summary of experimental conditions occurring during magnetite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Magnetite</td>
<td>25</td>
<td>1.1-6.0</td>
<td>White et al. (1993)</td>
</tr>
</tbody>
</table>

Figure Mag1: Summary of experimentally measured magnetite steady-state dissolution rates reported in the literature at 25 °C as a function of pH. The results of the model are plotted as the dotted line.
Muscovite

The mica muscovite (KAl$_3$Si$_3$O$_{10}$(OH)$_2$) dissolution rates were measured by Böhmeke (1946) at 25 °C and pH ranging from 5 to 11. Nickel (1973) reported the dissolution rates at 25 °C and pH = 0.2 to 10 measured in stirred tank reactors. Cabrera and Talibudeen (1978) studied the muscovite dissolution kinetics at 25 °C and pH 3, Hurd et al. (1979) reported kinetic data at 1 °C and pH 8. Lin and Clemency (1981) measured the dissolution rates of muscovite at 25 °C in a CO$_2$ rich solution at pH 5 in closed system reactors. Knauss and Wolery (1989) reported muscovite dissolution rates at 70 °C as a function of pH in flow-through reactors. Kalinowski and Schweda (1996) reported muscovite dissolution rates at 25 °C and pH = 1.1, 2.0, 3.0 and 4.1 measured in dialysis-cell reactors.

Oelkers et al. (2008) determined that the variation of rates with reactive solution composition depends on the solution pH. At pH 6, 7 rates were found to decrease significantly with increasing reactive fluid Al activity but be independent of aqueous SiO$_2$ activity. pH < 7 rates measured from 60 to 175 °C are consistent with:

$$r_\star = A_\star \exp \left( \frac{-E_A}{RT} \right) \left( \frac{a_{H^+}}{a_{Al^{3+}}} \right)^n$$

(Mu-1)

where $r_\star$ refers to the far-from-equilibrium muscovite dissolution rate, $R$ designates the gas constant, $T$ signifies absolute temperature and $a_i$ represents the activity of the subscripted aqueous species and $n$ signifies a stoichiometric coefficient equal to 0.2. $E_A$ depicting the activation energy of the reaction was determined from an Arrhenius plot of the data obtained at pH 2 by Oelkers et al. (2008) to be 46.03 kJ/mol and $A_\star$ a pre-exponential factor was calculated to be 5.32x10$^{-9}$ mol/cm$^2$/s.

The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Mu1. Rates calculated using Equation Mu-1 described 63 of 70 reported rates within 0.8 log units.
References


Table Mu1: Summary of experimentally measured forsterite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Muscovite</td>
<td>25</td>
<td>5-11</td>
<td>Böhmeke (1946)</td>
</tr>
<tr>
<td>Muscovite</td>
<td>25</td>
<td>3</td>
<td>Cabrera and Talibudeen (1978)</td>
</tr>
<tr>
<td>Muscovite</td>
<td>1</td>
<td>8</td>
<td>Hurd et al. (1979)</td>
</tr>
<tr>
<td>Muscovite</td>
<td>25</td>
<td>1-4</td>
<td>Kalinowski and Schweda (1996)</td>
</tr>
<tr>
<td>Muscovite</td>
<td>70</td>
<td>2-10</td>
<td>Knauss and Wolery (1989)</td>
</tr>
<tr>
<td>Muscovite</td>
<td>25</td>
<td>5</td>
<td>Lin and Clemency (1981)</td>
</tr>
<tr>
<td>Muscovite</td>
<td>25</td>
<td>0.2-10.6</td>
<td>Nickel (1973)</td>
</tr>
<tr>
<td>Muscovite</td>
<td>60-175</td>
<td>2</td>
<td>Oelkers et al. (2008)</td>
</tr>
<tr>
<td>Muscovite</td>
<td>25</td>
<td>3, 5</td>
<td>Stumm et al. (1987)</td>
</tr>
<tr>
<td>Muscovite</td>
<td>25</td>
<td>4, 4.5</td>
<td>Swoboda-Colberg and Drever (1993)</td>
</tr>
</tbody>
</table>
Figure Mu1: Summary of experimentally measured muscovite steady-state dissolution rates reported in the literature at 25 °C as a function of pH. The results of the model are plotted as the dotted line.

Figure Mu2: Summary of experimentally measured muscovite steady-state dissolution rates reported by Oelkers et al. (2008) at 150 °C as a function of pH. The results of the model are plotted as the dotted line.
Nepheline

The dissolution rates of the feldspathoid nepheline (NaAlSiO₄) were reported by Tole et al. (1986) at 25 to 80 °C and pH ranging from 3 to 11, Brady and Walther (1989) reported values at 25 °C and pH 8, Lasaga (1998) reported rate at 25 °C and pH 5 and Hamilton et al. (2001) reported rates at 25 °C and pH from 1.3 to 12.4. In the absence of additional data to improve existing data fits, these we have adopted the empirical pH power function fit of Palandri and Kharaka (2004) in accord with:

\[ r_{H^+} = A_A \cdot a_{H^+}^{1.13} \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \]
\[ r_{H_2O} = A_B \cdot \exp \left( \frac{-E_B}{R \cdot T} \right) \]
\[ r_{OH^-} = A_C \cdot a_{OH^-}^{0.2} \cdot \exp \left( \frac{-E_C}{R \cdot T} \right) \]
\[ r_+ = r_{H^+} + r_{H_2O} + r_{OH^-} \]

where \( r_{H^+}, r_{H_2O} \) and \( r_{OH^-} \) signifies the nepheline forward dissolution rate in acidic, neutral and basic solutions and \( r_+ \) the total dissolution rate, \( A_A, A_B \) and \( A_C \) refer to pre-exponential factors equal to \( 1.95 \times 10^4 \), \( 7.94 \times 10^{-2} \) and \( 7.24 \times 10^{-9} \) mol/cm²/s respectively, \( E_A, E_B \) and \( E_C \) designate an activation energy equal to 62.9, 65.4 and 37.8 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. In the absence of additional and more extensive data the Palandri and Kharaka fit has been adopted in this study. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Nep1. Rates calculated using Equation Nep-1 described all of the rates reported by Tole et al. (1986) within uncertainty. In contrast, it can be seen in this figure that the rates provided by Hamilton et al. (2001) are systematically ~1 order of magnitude lower than the corresponding Tole et al. (1986) values, and are thus approximately one order of magnitude lower than those calculated with the fit equation listed above.
References


Table Nep1: Summary of experimentally measured nepheline steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nepheline</td>
<td>25</td>
<td>8</td>
<td>Brady and Walther (1989)</td>
</tr>
<tr>
<td>Nepheline</td>
<td>25</td>
<td>1.3 – 12.4</td>
<td>Hamilton et al. (2001)</td>
</tr>
<tr>
<td>Nepheline</td>
<td>25</td>
<td>5</td>
<td>Lasaga (1998)</td>
</tr>
<tr>
<td>Nepheline</td>
<td>25 - 80</td>
<td>3 - 11</td>
<td>Tole et al. (1986)</td>
</tr>
</tbody>
</table>

Figure Nep1: Summary of experimentally measured nepheline steady-state dissolution rates reported in the literature as a function of pH and temperature. The dotted and dashed lines represent the results of the model.
Oligoclase

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure.png}
\caption{Oligoclase dissolution rate at 25°C.}
\end{figure}

- Casey et al. (1991)
- Cygan et al. (1989)
- Holdren and Speyer (1987)
- Oxburgh et al. (1993)
- Stillings and Brantley (1995)
- Stillings et al. (1996)
- Swoboda-Colberg and Drever (1993)

\begin{align*}
\text{log } r^+ & (\text{mol/cm}^2/\text{s}) \\
\text{pH} &
\end{align*}
Paragonite

The phyllosilicate of the mica group paragonite (NaAl$_3$Si$_3$O$_{10}$(OH)$_2$) was studied by Lin and Clemency (1981) at 25°C and pH of 5.5 and reported by Nagy (1995). The conditions where the data was acquired are summarized in Table Par1 and the available rate data are presented in Fig. Par1.

Paragonite is a mica with a structure similar to muscovite where the K is replaced by Na, therefore owing to this similarity the dissolution rate data were fitted to:

$$r_\ast = A_\ast \exp \left( \frac{-E_A}{R \cdot T} \right) \left( \frac{a_{H^+}}{a_{Al^{3+}}} \right)^n$$

(Par-1)

where $r_\ast$ refers to the far-from-equilibrium muscovite dissolution rate, $R$ designates the gas constant, $T$ signifies absolute temperature and $a_i$ represents the activity of the subscripted aqueous species and $n$ signifies a stoichiometric coefficient equal to 0.2. $E_A$ depicting the activation energy of the reaction was assumed to be equal to muscovite at 46.03 kJ/mol and $A_\ast$ the pre-exponential factor was calculated to fit the data reported by Nagy (1995) at 1.32x10$^{-7}$ mol/cm$^2$/s.

The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Par1. Rates calculated using Equation Par-1 described all the reported rates within 0.8 log units.

References


Table Par1: Summary of experimental conditions occurring during paragonite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Paragonite</td>
<td>25</td>
<td>5.5</td>
<td>Lin and Clemency (1981)</td>
</tr>
</tbody>
</table>
**Pyrite**

The dissolution kinetics of the iron sulfide pyrite (FeS$_2$) have been investigated by Gibbs et al., 1997a; Gibbs et al., 1997b; Holmes and Crundwell, 2000; Kamei and Ohmoto, 2000; Lennie and Vaughan, 1992; McKibben and Barnes, 1986; Moses and Herman, 1991; Moses et al., 1987; Nicholson et al., 1988; Nicholson et al., 1990; Rickard, 1997; Rickard and Luther, 1997; Rickard, 1975; Rimstidt and Vaughan, 2003; Schoonen and Barnes, 1991a; Schoonen and Barnes, 1991b; Schoonen and Barnes, 1991c; Wiersma and Rimstidt, 1984; Williamson and Rimstidt, 1994. The different conditions where the data was acquired are summarized in table Ph1. The rates are presented for the different temperatures as a function of pH in Figure Ph1.
**Phlogopite**

The dissolution kinetics of phlogopite (KAlMg_3Si_3O_10(OH)_2), an iron free mica, was studied by Lin and Clemency (1981a,b) at pH 5.34 and 25 °C, Clemency and Lin (1981) reported dissolution rates at pH 3.05 and 25 °C, McClelland (1950), Pitault et al. (1981), Schnitzler and Kodama (1976), Rausell-Colom et al. (1965), Razzaghie and Robert (1979), Marshall (1962). Kuwahara and Aoki (1995) studied phlogopite dissolution at pH 2 from 50° to 120 °C but did not generate dissolution rates. Kalinowski and Schweda (1996) reported phlogopite rates at pH ranging from 1.1 to 4.1 and 25 °C, Rufe and Hochella (1999) and Taylor et al. (2000). The different conditions where the data was acquired are summarized in table Ph1. The rates are presented for the different temperatures as a function of pH in Figure Ph1.

As phlogopite is a biotite the same equation was used:

\[ r_\text{+} = A_\text{A} \cdot \exp \left( \frac{-E_A}{RT} \right) \left( \frac{a_{\text{H}^+}}{a_{\text{Al}^{3+}}} \right)^n \]  

(Ph-1)

where \( r_\text{+} \) refers to the far-from-equilibrium phlogopite dissolution rate, \( A_\text{A} \) represents a rate constant, \( R \) designates the gas constant, \( T \) signifies absolute temperature and \( a_i \) represents the activity of the subscripted aqueous species, \( E_A \) defines the activation energy of the reaction and \( n \) designates a stoichiometric coefficient equal to 0.125. The \( E_A \) of 49kJ/mol calculated for biotite was used, and \( A_\text{A} \) was recalculated to 6.21.10^{-8} \text{ mol/cm}^2/\text{s} to fit the data by Kalinowski and Schweda (1996). The estimated \( E_A \) value used in Ph-1 is somewhat higher than the \( E_A \) calculated by Kuwahara and Aoki (29-42 kJ/mol).

The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Ph1. Rates calculated using Equation Ph-1 described 14 of 16 reported rates within 0.8 log units.

**References**


Table Ph1: Summary of experimental conditions occurring during smectite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>phlogopite</td>
<td>25</td>
<td>3.05</td>
<td>Clemency and Lin (1981)</td>
</tr>
<tr>
<td>phlogopite</td>
<td>25</td>
<td>1 - 4</td>
<td>Kalinowski and Schweda (1996)</td>
</tr>
<tr>
<td>phlogopite</td>
<td>50 - 120</td>
<td>2</td>
<td>Kuwahara and Aoki (1995)</td>
</tr>
<tr>
<td>phlogopite</td>
<td>25</td>
<td>5.34</td>
<td>Lin and Clemency (1981c)</td>
</tr>
<tr>
<td>Phlogopite</td>
<td>25</td>
<td>2, 5.7</td>
<td>Rufe and Hochella (1999)</td>
</tr>
<tr>
<td>phlogopite</td>
<td>25</td>
<td>3.3</td>
<td>Taylor et al. (2000)</td>
</tr>
</tbody>
</table>

Figure Ph1: Summary of experimentally measured phlogopite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
Prehnite

The calcium and aluminum phyllosilicate prehnite (Ca$_2$Al$_2$Si$_3$O$_{10}$(OH)$_2$) has been studied by Rose (1991) at temperatures of 25 to 90 °C and pH from 1.4 to 10.8. In the absence of additional data to improve existing data fits, these we have adopted the empirical pH power function fit of Palandri and Kharaka (2004) in accord with:

\[ r_{H^+} = A_A \cdot a_{H^+}^{0.256} \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \]

\[ r_{H_2O} = A_B \cdot \exp \left( \frac{-E_B}{R \cdot T} \right) \]

\[ r_{OH^-} = A_C \cdot a_{OH^-}^{0.2} \cdot \exp \left( \frac{-E_C}{R \cdot T} \right) \]

\[ r_{tt} = r_{H^+} + r_{H_2O} + r_{OH^-} \]

where \( r_{H^+}, r_{H_2O} \) and \( r_{OH^-} \) signifies the prehnite forward dissolution rate in acidic, neutral and basic solutions and \( r_{tt} \) the total dissolution rate, \( A_A, A_B \) and \( A_C \) refer to pre-exponential factors equal to 2.75x10$^{-1}$, 1.58x10$^{-1}$ and 3.16x10$^{-3}$ mol/cm²/s respectively, \( E_A, E_B, E_C \) designates an activation energy equal to 80.5, 93.4 and 93.4 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes the absolute temperature. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Pre1. Rates calculated using Equation Pre-1 described all the reported rates within 0.8 log units.

References


Table Pre1: Summary of experimental conditions occurring during prehnite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Prehnite</td>
<td>25-90</td>
<td>1.4 – 10.8</td>
<td>Rose (1991)</td>
</tr>
</tbody>
</table>

Figure Pre1: Summary of experimentally measured prehnite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted and dashed line for the 25 and 90 °C models, respectively.
**Pyrophyllite**

The phyllosilicate of the mica group pyrophyllite \((Al_2Si_4O_{10}(OH)_2)\) were reported by Kline and Fogler (1981) in Nagy (1995) and they reported dissolution rates at 25 °C and 1M HF solution. Scheidegger et al. (1996) also reported rates of pyrophyllite dissolution at 25 °C and pH 7.5 in batch reactors. The different conditions where the data was acquired are summarized in table Pyr1 and the available rate data are presented in Fig. Pyr1.

Pyrophyllite is a phyllosilicate with mica sheets. The dissolution rate data were fitted to:

\[
r_+ = A_A \exp \left( \frac{-E_A}{R \cdot T} \right) \left( \frac{a_{H^+}}{a_{Al^{3+}}} \right)^n
\]  

where \(r_+\) refers to the far-from-equilibrium muscovite dissolution rate, \(R\) designates the gas constant, \(T\) signifies absolute temperature and \(a_i\) represents the activity of the subscripted aqueous species and \(n\) signifies a stoichiometric coefficient equal to 0.125. \(E_A\) depicting the activation energy of the reaction was assumed to be equal to muscovite at 46.03 kJ/mol and \(A_A\) the pre-exponential factor was calculated to fit the data reported by Scheidegger et al. (1996) at \(2.2 \times 10^{-8}\) mol/cm²/s.

The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Pyr1. Rates calculated using Equation Pyr-1 described 1 of 2 reported rates within 0.8 log units.

**References**


Table Pyr1: Summary of experimental conditions occurring during pyrophyllite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pyrophyllite</td>
<td>25</td>
<td>0</td>
<td>Kline and Fogler (1981)</td>
</tr>
<tr>
<td>Pyrophyllite</td>
<td>25</td>
<td>7.5</td>
<td>Scheidegger et al. (1996)</td>
</tr>
</tbody>
</table>

Figure Pyr1: Summary of experimentally measured pyrophyllite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
**Rhodochrosite**

The dissolution kinetics of rhodochrosite (MnCO₃) were investigated by Jensen et al. (2002) who reported rates gathered in anaerobic aquatic environment at 25 °C and pH 7.5. Duckworth and Martin (2003) reported rates at 25 °C and pH from 1.5 to 11.

We based our rate law on the one defined by Busenberg and Plummer (1982) who followed the interpretative guidelines of Plummer et al. (1978) for calcite and added an activation energy term for each dissolution mechanism. The pre-exponential factors were recalculated to fit the data:

\[
r_+ = \left[ A_A \cdot a_{H+}^{n+} \times \exp \left( \frac{-E_A}{RT} \right) \right] + \left[ A_B \cdot a_{H_2CO_3}^{n} \times \exp \left( \frac{-E_B}{RT} \right) \right] + \left[ A_C \cdot a_{H_2}^{n} \times \exp \left( \frac{-E_C}{RT} \right) \right]
\]  

(Rho-1)

where \( r_+ \) signifies the siderite steady state dissolution rate, \( A_A, A_B \) and \( A_C \) refer to pre-exponential factor equal to 57.9, 1x10⁻², and 2.2x10⁻² mol/cm²/s respectively, \( E_A, E_B \) and \( E_C \) designate an activation energy equal to 61.0, 45.0 and 44.0 kJ/mol respectively, \( R \) represents the gas constant, \( T \) denotes absolute temperature and \( n \) is a reaction order equal to 0.7. The activation energies were assumed to be similar to siderite and the pre-exponential factors recalculated to fit Duckworth and Martin (2003) data. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Sid1. Rates calculated using Equation Rho-2 described 24 of 25 reported rates within 0.8 log units.
References


Table Rho1: Summary of experimental conditions occurring during rhodochrosite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rhodochrosite</td>
<td>25</td>
<td>1.5-11</td>
<td>Duckworth and Martin (2004)</td>
</tr>
<tr>
<td>Rhodochrosite</td>
<td>25</td>
<td>7.5</td>
<td>Jensen et al. (2002)</td>
</tr>
</tbody>
</table>

Figure Rho1: Summary of experimentally measured rhodochrosite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
**Serpentine minerals**

The dissolution kinetics of serpentine minerals (chrysotile, antigorite and lizardite) has been the subject of a rather limited number of experimental investigation, i.e. Luce et al. (1972) carried out batch experiments on lizardite at 25 °C from pH 1.5 to 9, Lin and Clemency (1981) measured the dissolution rate of antigorite at 25 °C and constant 1 bar CO2 pressure at pH 6 to 6.5, Bales and Morgan (1985) determined the kinetics of chrysotile dissolution in batch experiments at 25 C and in the pH range 7-10 and Hume and Rimstidt (1992) studied the dissolution rate of chrysotile in synthetic lung fluid at 37 °C. Thomassin et al. (1977) determined the activation energy for chrysotile dissolution to be 70 kJ/mol. These datasets were reviewed in 2004 by Cipolli et al., considerations upon which we based our kinetic equation:

\[
\log r + = -0.31x\text{pH} - 7.08
\]

where \( r \) refers to the far-from-equilibrium phlogopite dissolution rate, \( R \) designates the gas constant, \( T \) signifies absolute temperature and \( a_i \) represents the activity of the subscripted aqueous species. \( E_A \) depicting the activation energy of the reaction was determined to be 70 kJ/mol and \( A_A \) a pre-exponential factor was calculated -0.31 mol/cm²/s. The degree to which the Eqn. Serp1 defines the available data can be assessed with the help of Fig. Serp 1.

**References**


dissolution kinetics of chrysotile in 0.1 N oxalic acid at different temperatures. *Phys. Chem.
Miner.* **1**, 385-398.
**Siderite**

The dissolution kinetics of siderite (FeCO$_3$) were investigated by Greenberg and Tomson (1992), Duckworth and Martin (2003), Golubev et al. (2009) and Tang and Martin (2011). Greenberg and Tomson (1992) determined the dissolution kinetics at 26 and 60 °C at pH 4.88. Jensen et al. (2002) reported rates gathered in anaerobic aquatic environment at 25°C and pH 7.5. Duckworth and Martin (2003) reported rates at 25 °C and pH from 1.5 to 11, Golubev et al. (2009) reported rates at 25 to 100 °C in acidic conditions (1<pH<4.6) and Tang and Martin (2011) reported dissolution rates in the presence of chromate at 25 °C and pH from 1 to 11.

We based our rate law on the one defined by Busenberg and Plummer (1982) who followed the interpretative guidelines of Plummer et al. (1978) for calcite and added an activation energy term for each dissolution mechanism. The pre-exponential factors were recalculated to fit the data:

\[
 r = \left( A_A \cdot A_{H^+}^{n_A} \times \exp \left( \frac{-E_A}{RT} \right) \right) + \left( A_B \cdot A_{H_2CO_3}^{n_B} \times \exp \left( \frac{-E_B}{RT} \right) \right) + \left( A_C \cdot A_{H_2}^{n_C} \times \exp \left( \frac{-E_C}{RT} \right) \right) \]  

(Do-2)

where \( r \) signifies the siderite steady state dissolution rate, \( A_A, A_B \) and \( A_C \) refer to pre-exponential factor equal to 57.9, 1x10$^{-2}$ and 2.2x10$^{-7}$ mol/cm²/s respectively, \( E_A, E_B \) and \( E_C \) designate an activation energy equal to 61.0, 45.0 and 45.0 kJ/mol respectively, \( R \) represents the gas constant, \( T \) denotes absolute temperature and \( n \) is a reaction order equal to 0.7. The activation energies were determined from an Arrhenius plot of the data reported by Golubev et al. (1999) in acidic condition and calculated by Marini (2004) for neutral and basic conditions. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Sid1. Rates calculated using Equation Sid-2 described 85 of 87 reported rates within 0.8 log units.
References


Table Sid1: Summary of experimental conditions occurring during siderite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Siderite</td>
<td>25</td>
<td>1.5-11</td>
<td>Duckworth and Martin (2004)</td>
</tr>
<tr>
<td>Siderite</td>
<td>25-100</td>
<td>1-4.6</td>
<td>Golubev et al. (2009)</td>
</tr>
<tr>
<td>Siderite</td>
<td>26-60</td>
<td>4.88</td>
<td>Greenberg and Tomson (1992)</td>
</tr>
<tr>
<td>Siderite</td>
<td>25</td>
<td>7.5</td>
<td>Jensen et al. (2002)</td>
</tr>
<tr>
<td>Siderite</td>
<td>25</td>
<td>1-11</td>
<td>Tang and Martin (2011)</td>
</tr>
</tbody>
</table>

Figure Sid1: Summary of experimentally measured siderite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
This complex group of minerals has been the subject of comparatively limited studies aimed at determining their dissolution kinetics. Hayasi and Yamada (1990) reported rates for Wyoming montmorillonite at pH 11 and 80 °C. Zysset and Schindler (1996) determined the dissolution rate of Crook Country Wyoming montmorillonite at 25 °C and pH ranging from 1 to 5. Bauers and Berger (1998) carried out dissolution experiments on two smectites, Ceca and Ibeco, at 35 and 80 °C with pH ranging from 11.5 to 14. Huertas et al. (2001) reported smectite dissolution rates at pH ranging from 7.5 to 8.6 and temperatures of 20°, 40° and 60 °C, Cama et al. (2002) investigated the dissolution rate of smectite as a function of thermodynamic activity at 80 °C and pH of 8.8 and Rozalen et al. (2008) measured the dissolution kinetics of smectite at 25 °C with pHs ranging from 1.1 to 13.7. The different conditions where the data was acquired are summarized in table Sm1. The rates are presented for the different temperatures as a function of pH in Figure Sm1.

The published data presented in Fig. Sm1 was fitted with the help of Eqn. Sm-1 defined by Huertas et al. (2001):

$$r = A_e \exp\left(-\frac{E_A}{RT}\right) \left(\frac{a_{H^+}^{1.0}}{a_{H^+}^{0.1}}\right)^n$$

where $r_*$ signifies the kaolinite steady state dissolution rate, $A_e$ refers to a pre-exponential factor equal to 5.65x10^{-13} mol/cm²/s, $E_A$ designates an activation energy equal to 27 kJ/mol, $R$ represents the gas constant, $T$ denotes absolute temperature and $n$ signifies a stoichiometric coefficient equal to 0.1. $A_e$ and $E_A$ were determined from an Arrhenius plot of the data obtained at pH 8 by Huertas et al. (2001). The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Sm1. Rates calculated using Equation Sm-1 described xx of xx reported rates within 0.8 log units. The data provided by Zisset and Schindler (1996) is out of range for the serie containing 1M KCl which is probably due to the effect of the high concentration of the salt and the data published by Bauer and Berger (1998), Cama et al. (2000) and Hayasi and Yamada (1990).
References


Table Sm1: Summary of experimental conditions occurring during smectite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>smectite</td>
<td>35, 80</td>
<td>11.5 – 13.9</td>
<td>Bauer and Berger (1998)</td>
</tr>
<tr>
<td>montmorillonite</td>
<td>25</td>
<td>1 - 5</td>
<td>Cama et al. (2002)</td>
</tr>
<tr>
<td>montmorillonite</td>
<td>25, 60, 80</td>
<td>1 - 12</td>
<td>Hayasi and Yamada (1990)</td>
</tr>
<tr>
<td>montmorillonite</td>
<td>150</td>
<td>2, 6.8, 7.8</td>
<td>Huertas et al. (2001)</td>
</tr>
<tr>
<td>montmorillonite</td>
<td>25, 50, 80</td>
<td>3 - 4.2</td>
<td>Rozalén et al. (2008)</td>
</tr>
<tr>
<td>K-montmorillonite</td>
<td>25</td>
<td>2 - 6.5</td>
<td>Zysset and Schindler (1996)</td>
</tr>
</tbody>
</table>

Figure Sm1: Summary of experimentally measured smectite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
**Sphalerite**

The zinc sulfide sphalerite (Zn, Fe S) dissolution has been studied by Majima et al. (1981) at temperature from 25 to 55 C and pH of 0.5, Crundwell and Verbaan (1987) at 25 C and pH 1, Moses and Hernan (1990) at 25 C and pH 6 and 7, Rimstidt et al. (1994) at 40 C and pH 2, Domenech et al (2002) at 22 C and pH of 2.5 to 4.3, Weisener et al. (2003) at 25 to 85 C and pH of 1, Malmström and Collin (2004) at 22 C and pH 1, Acero et al. (2007) at temperature of 25-70 C and pH from 1 to 4.2 and Jordan et al (2011) at pH 2 and temperature ranging from 20 to 200 C.

**References**


Table Sph1: Summary of experimentally measured sphalerite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>sphalerite</td>
<td>25</td>
<td>5</td>
<td>Acero et al. (2007)</td>
</tr>
<tr>
<td>sphalerite</td>
<td>37</td>
<td>2-8</td>
<td>Crundwell and Verbaan (1987)</td>
</tr>
<tr>
<td>Sphalerite</td>
<td></td>
<td></td>
<td>Demènech et al. (2002)</td>
</tr>
<tr>
<td>Sphalerite</td>
<td></td>
<td></td>
<td>Jordan et al. (2011)</td>
</tr>
<tr>
<td>Sphalerite</td>
<td></td>
<td></td>
<td>Majima et al. (1981)</td>
</tr>
<tr>
<td>sphalerite</td>
<td>25-150</td>
<td>1.7-9</td>
<td>Moses and Herman (1990)</td>
</tr>
<tr>
<td>sphalerite</td>
<td></td>
<td></td>
<td>Rimstidt et al. (1994)</td>
</tr>
</tbody>
</table>

Figure Sph1: Summary of experimentally measured sphalerite steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dotted line.
Spodumene

Spodumene like jadeite (NaAl(SiO3)2) is an inosilicate with a clinopyroxene structure, where Si–O chains are held together with a monovalent cation and Al bridges; for the case of spodumene the monovalent cation is Li. Its dissolution kinetics was only reported by Sverdrup (1990) at 25 °C and pH ranging from 3 to 6.5. These data points are plotted on Fig. Sp1 and detailed in Table Sp1.

By analogy with jadeite it seems reasonable to assume that its dissolution mechanism consists of the sequential removal of Li then Al via proton exchange reactions prior to the final removal of Si–O tetrahedra from the mineral. In such cases dissolution rates are commonly consistent with:

\[
r_\ast = A \cdot \exp \left( \frac{-E_A}{RT} \right) \cdot \left( \frac{a_{Li^+}}{a_{Al^{3+}}} \right)^n
\]

where \( r \) signifies the spodumene steady state dissolution rate, \( A \) refers to a pre-exponential factor equal to 1.5x10^{-3} mol/cm²/sec, \( E_A \) designates an activation energy equal to 48 kJ/mol, \( n \) refers to a constant equal to 0.25, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. As was the case for jadeite, \( E_A \) was set to be equal to 48 kJ/mol by analogy with the amphiboles, chain silicates also held together by bridging Al-octahedrals. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Sp1. Rates calculated using Equation Sp-1 described 3 of 3 reported rates within 0.1 log units with an average uncertainty of 0.1 log units.

References

Table S1: Summary of experimental conditions occurring during spodumene steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spodumene</td>
<td>25</td>
<td>3, 4.5, 6.5</td>
<td>Sverdrup (1990)</td>
</tr>
</tbody>
</table>

Figure S1: Summary of experimentally measured spodumene steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dashed line.
**Staurolite**

The dissolution rates of the iron aluminum nesosilicate staurolite (Fe_{1.4}Li_{0.1}Mg_{0.1}Al_{8.7}Si_{3.9}O_{21.7}(OH)_{2.3}) was reported by Nickle (1973) at 25 °C and pH ranging from 0.2 to 11. These data were fit by Palandri and Kharaka (2004) using:

\[
\begin{align*}
    r_{H^+} &= A_A \cdot a_{H^+} \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \\
    r_{H_2O} &= A_B \cdot \exp \left( \frac{-E_B}{R \cdot T} \right) \\
    r_{OH^-} &= A_C \cdot a_{OH}^{-0.3} \cdot \exp \left( \frac{-E_C}{R \cdot T} \right) \\
    r_+ &= r_{H^+} + r_{H_2O} + r_{OH^-}
\end{align*}
\]

where \( r_{H^+} \), \( r_{H_2O} \) and \( r_{OH^-} \) signifies the staurolite forward dissolution rate in acidic, neutral and basic solutions and \( r_+ \), the overall dissolution rate, \( A_A \), \( A_B \) and \( A_C \) refer to pre-exponential factors equal to 2.57x10^{-8}, 5.25x10^{-5} and 2.34x10^{-11} mol/cm²/s respectively, \( E_A \), \( E_B \) and \( E_C \) designate an activation energies equal to 18.9, 56.6 and 47.2 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. As was the case for andradite in the absence of additional and more extensive data, the Palandri and Kharaka (2004) fit was adopted in this study. It should be noted that these activation energies are based on the fit of rates obtained at 8 and 25 °C, and the rate data at 8 °C were never directly reported. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure St1. Rates calculated using Equation St-1 described 8 of 9 reported rates within 0.8 log units.

**References**


Table St1: Summary of experimentally measured staurolite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Staurolite</td>
<td>25</td>
<td>0.2-11</td>
<td>Nickel (1973)</td>
</tr>
</tbody>
</table>

Figure St1: Summary of experimentally measured staurolite steady-state dissolution rates reported in the literature as a function of pH. The dotted line represents the results of the model.
**Struvite**

The magnesium ammonium phosphate struvite (NH₄MgPO₄·6H₂O) dissolution has been studied by Roncal-Herrero and Oelkers (2011) at 25 °C and pH 7.7 to 10.6.

**References**


Table Str1: Summary of experimental conditions for the struvite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tourmaline</td>
<td>25</td>
<td>0.2-10.5</td>
<td>Roncal-Herrero and Oelkers (2011)</td>
</tr>
</tbody>
</table>

Figure Str1: Summary of experimentally measured struvite steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dotted line.
**Talc**

Dissolution rates of talc at 25 to 150 °C and various pH have been studied by Lin and Clemency (1981), Jurinski and Rimstidt (2001) and Saldi et al., (2007). The rates are presented as a function of pH in Figure E1. Rates reported by Jurinski and Rimstidt (2001) at 37 °C exhibit a pH dependence inconsistent with those of Saldi et al. (2007); these former rates are mostly pH independent, whereas the later rates decrease continuously with increasing pH. This latter dependence is consistent with that of other Mg silicate minerals including enstatite and tremolite (e.g. Oelkers and Schott, 2001).

Steady-state talc dissolution rates were fit to (Saldi et al., 2007)

\[
 r = S A_A \left( \frac{a_{H^+}^2}{a_{Me^{2+}}} \right)^{1/8} + A_B \exp\left( - \frac{E_A}{\sigma RT} \right) 
\]

where \( r \) signifies the talc steady-state dissolution rate, \( A_A \) and \( A_B \) refer to pre-exponential factors equal to \( 5 \times 10^{-9} \) and \( 0.8 \times 10^{-9} \) mol/cm²/sec⁴, \( E_A \) designates an activation energy equal to 45 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. \( \sigma \) stands for Temkin’s average stoichiometric number equal to the ratio of the rate of destruction of the activated or precursor complex relative to the overall rate. For the case of talc, \( \sigma \) is taken to be equal to 4, the number of Si in the talc chemical formula. This choice is consistent with the assumption that the activated species for talc dissolution contains one Si atom. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure T1. Rates calculated using Equation T-1 described 46 of 50 reported rates within 0.8 log units with an average uncertainty of 0.3 log units.

**References**


\(^4\) Note this value is corrected from that in the original Oelkers and Schott (2001) that was reported as \( 2.4 \times 10^{-4} \) mol/cm²/sec.

Table T1: Summary of experimentally measured talc steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Talc</td>
<td>37</td>
<td>2-8</td>
<td>Jurinski and Rimstidt (2001)</td>
</tr>
<tr>
<td>Talc</td>
<td>25-150</td>
<td>1.7-9</td>
<td>Saldi et al. (2007)</td>
</tr>
</tbody>
</table>

Figure T1: Summary of experimentally measured talc steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dotted line.
Tourmaline

The dissolution rates of the boron containing cyclosilicate tourmaline were studied by Nickel (1973) and recalculated by Sverdrup (1990) and these data are plotted in fig. Tou1. In the absence of a mechanistic model Palandri and Kharaka (2004) fit these rates assuming they are controlled by two parallel reactions, one proton promoted, and one water promoted in accord with:

\[ r_{H^+} = A_A a_H^{1/2} \exp \left( \frac{-E_A}{R.T} \right) \]

\[ r_{H_2O} = A_B \exp \left( \frac{-E_B}{R.T} \right) \]

which can be summed to yield the overall forward rate using

\[ r_+ = (r_{H^+} + r_{H_2O}) \]

where \( r_{H^+} \) and \( r_{H_2O} \) signifies the tourmaline steady-state dissolution rate in response to the proton and water promoted mechanisms, respectively, and \( r_+ \) the total forward dissolution rate, \( A_A \) and \( A_B \) refer to pre-exponential factors equal to 5.37x10^2 and 4.9x10^{-1} mol/cm²/s respectively, \( E_A \) designates an activation energy equal to 75.5 and 85.0 kJ/mol respectively, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. The degree to which these equations describes the measured can be assessed in Figure Tou1. Rates calculated using Equation Tou-1 described all 3 of the reported rates within 0.8 log units.

References


Table Tou1: Summary of experimental conditions for the various tourmaline steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tourmaline</td>
<td>25</td>
<td>0.2-10.5</td>
<td>Nickel (1973)</td>
</tr>
</tbody>
</table>

Figure Tou1: Summary of experimentally measured tourmaline steady-state dissolution rates reported in the literature as a function of pH. The results of the model at 25 °C are plotted in dotted line.
**Tremolite**

The Ca bearing amphibole tremolite (Mg$_7$Si$_8$O$_{22}$(OH)$_2$) dissolution was studied by Tunn (1939) at 25 °C and pH ranging from 0.2 to 11; these data were recalculated and reported by Sverdrup (1990). Schott et al. (1981) reported dissolution rates at 20 °C and pH 1 and 6 at 25 °C. Recently Dietrich et al. (2012) reported tremolite rate data at temperatures from 25 to 150 °C and pH ranging from 1.9 to 6.9. These experimental conditions are reported in Table Tre1 and plotted on Fig. Tre1.

The data was fitted to:

$$r_\ast = A_A \exp \left( \frac{-E_A}{RT} \right) \left( \frac{a_{Mg}^{2+}}{a_{Mg}^{2+}} \right)^n$$

where $r_\ast$ signifies the tremolite steady state dissolution rate, $A_A$ refers to a pre-exponential factor equal to 1.54x10$^{-6}$ mol/cm²/s, $E_A$ designates an activation energy equal to 55.8 kJ/mol, $R$ represents the gas constant, $T$ denotes absolute temperature and $n$ signifies a stoichiometric coefficient equal to 0.125. In the absence of extensive data for tremolite dissolution rates as a function of the temperature $E_A$ was set equal to that of anthophyllite (55.8 kJ/mol). $A_A$ was calculated to fit the data shown in Fig Tre1. As such tremolite forward dissolution rates are equal to 0.9 times that of anthophyllite rates for all temperature and solution composition. The faster rates of tremolite dissolution can be attributed to the presence of rapidly removed Ca from its structure. The degree to which equation Tre-1 describes the dissolution rates reported in the literature can be assessed in Figure Tre1. Rates calculated using Equation Tre-1 described 30 of 35 reported rates within 0.8 log units.

**References**


Table Tre1: Summary of experimentally measured tremolite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tremolite</td>
<td>25-150</td>
<td>1.9-6.8</td>
<td>Dietrich et al. (2012)</td>
</tr>
<tr>
<td>Tremolite</td>
<td>22</td>
<td>2-9</td>
<td>Mast and Drever (1987)</td>
</tr>
<tr>
<td>Tremolite</td>
<td>20</td>
<td>1, 6</td>
<td>Schott et al. (2001)</td>
</tr>
<tr>
<td>Tremolite</td>
<td>25</td>
<td>3 – 6.8</td>
<td>Sverdrup (1990)</td>
</tr>
<tr>
<td>Tremolite</td>
<td>25</td>
<td>0.2-11</td>
<td>Tunn (1939)</td>
</tr>
</tbody>
</table>

Figure Tre1: Summary of experimentally measured tremolite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.
Uraninite

The oxide uraninite (UO$_2$) dissolution kinetics have been studied by Grandstaff (1976) and lately by Ivanov and Popov (2011) at 25°C from 5 to 8.5. The available data was fitted by Palandri and Kharaka (2004) to:

$$ r_+ = A_A \exp \left( \frac{-E_A}{R \cdot T} \right) $$  \hspace{1cm} \text{(Ur1)}

where $r_+$ signifies the uraninite total forward dissolution rate, $A_A$ refers to a pre-exponential factor equal to $2.26 \times 10^{-9}$ mol/cm$^2$/s, $E_A$ designates an activation energy equal to 75.5 kJ/mol, $R$ represents the gas constant, and $T$ denotes absolute temperature. The degree to which this equation describes the measured can be assessed in Figure Ur1. Rates calculated using Equation Ur1 described all the reported rates within 0.8 log units.

References


Table Ur1: Summary of experimental conditions occurring during variscite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uraninite</td>
<td>25</td>
<td>-</td>
<td>Granstaff (1976)</td>
</tr>
<tr>
<td>Uraninite</td>
<td>25</td>
<td>5-8.5</td>
<td>Ivanov and Popov (2011)</td>
</tr>
</tbody>
</table>

Figure Ur1: Summary of experimentally measured variscite steady-state dissolution rates reported in the literature at 25 °C as a function of pH. The results of the model are plotted as the dotted line.
**Variscite**

The hydrated aluminum phosphate variscite (AlPO$_4$·2H$_2$O) dissolution has been studied by Roncal-Herrero and Oelkers (2011) in mixed flow and batch reactors at 25 °C and pH ranging from 1.51 to 8.99. The different conditions where the data was acquired are summarized in table Var1. The rates are presented for the different temperatures as a function of pH in Fig. Var1.

Variscite dissolution rates were described by Roncal-Herrero and Oelkers (2011) by:

$$r_+ = 10^{-14.9} \cdot a_{H^+}^{0.25} + 10^{-19.0} \cdot a_{H^+}^{-0.4}$$  \[\text{Var1}\]

Where $r_+$ represent the forward variscite dissolution rates.

**References**


**Table Var1:** Summary of experimental conditions occurring during variscite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Variscite</td>
<td>25</td>
<td>1.5 - 9</td>
<td>Roncal-Herrero and Oelkers (2011)</td>
</tr>
</tbody>
</table>

**Figure Var1:** Summary of experimentally measured variscite steady-state dissolution rates reported in the literature at 25 °C as a function of pH. The results of the model are plotted as the dotted line.
Roncal-Herrero and Oelkers (2011)

Model
Witherite

The barium carbonate mineral witherite (BaCO$_3$) has been studied by Chou et al. (1989) at 25 °C and pH ranging from 4.5 to 9.8. The different conditions where the data was acquired are summarized in table Wit1. The rates are presented for the different temperatures as a function of pH in Fig. Wit1.

The available data was fitted to an equation from Chou et al. (1989) with the inclusion of activation energies to:

\[ r_{H^+} = A_A \cdot \exp \left( \frac{-E_A}{R \cdot T} \right) \cdot (a_{H^+})^n \]

\[ r_{H_2CO_3} = A_B \cdot \exp \left( \frac{-E_B}{R \cdot T} \right) \cdot (a_{H_2CO_3})^n \]

\[ r_{H_2O} = A_C \cdot \exp \left( \frac{-E_C}{R \cdot T} \right) \cdot (a_{H_2O})^n \]

\[ r_+ = r_{H^+} + r_{H_2CO_3} + r_{H_2O} \]

where $r_{H^+}$, $r_{H_2CO_3}$ and $r_{H_2O}$ signifies the witherite forward dissolution rate in acidic, neutral and basic solutions and $r_+$ the total dissolution rate, $A_A$, $A_B$ and $A_C$ refer to pre-exponential factors equal to 8.86, 10 and 2.2x10$^{-5}$ mol/cm²/s respectively, $E_A$, $E_B$ and $E_C$ designate activation energies equal to 34.0, 21.0 and 16.0 kl/mol, $R$ represents the gas constant, $T$ denotes absolute temperature and $n$ signifies a stoichiometric coefficient equal to 0.75. The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Wit1. Rates calculated using Equation Wit-1 described all of the reported rates at 25 °C within 0.8 log units.

References

Table Wit1: Summary of experimental conditions occurring during witherite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Witherite</td>
<td>25</td>
<td>4.5-9.8</td>
<td>Chou et al. (1989)</td>
</tr>
</tbody>
</table>

Figure Wit1: Summary of experimentally measured witherite steady-state dissolution rates reported in the literature at 25 °C as a function of pH. The results of the model are plotted as the dotted line.
Wollastonite dissolution rates were reported by Rimstidt and Dove (1986) at 23.5 °C and pH ranging from 4.5 to 7, Sverdrup (1990) at 25 °C and pH 3, 4 and 7, Feruzzi (1994) at 25 °C at 25 C and pH from 2 to 9.6, Xie (1994), Xie and Walther (1994), Weissbart and Rimstidt (2000) at 23.5 °C and pH 1.9 to 6.2, Golubev et al. (2005) at 25 C and pH ranging from 3.1 to 12.2, Pokrovsky et al. (2009), also at 25 °C and with pH from 6.9 to 11.0, and Schott et al. (2012) at 25 °C and pH ranging from 0.26 to 12.

The different conditions where the data was acquired are summarized in table Wo1. The rates are presented for the different temperatures as a function of pH in Figure Wo1. The published data presented in Fig. Wo1 was fitted using (c.f. Oelkers and Schott, 2001)

\[ r_+ = A_A \cdot \exp \left( -\frac{E_A}{R} \right) \cdot \left( \frac{a_{H^+}^2}{a_{Ca^{2+}}} \right)^{1/8} \]  

where \( r_+ \) signifies the wollastonite steady state dissolution rate, \( A_A \) refers to a pre-exponential factor equal to \( 2 \times 10^{-4} \) mol/cm²/s, \( E_A \) designates an activation energy equal to 45 kJ/mol, \( R \) represents the gas constant, and \( T \) denotes absolute temperature. \( E_A \) was taken from Palandri and Kharaka (2004). The degree to which this equation describes the dissolution rates reported in the literature can be assessed in Figure Wo1. Rates calculated using Equation Wo-1 described 95 of 108 reported rates at 25 °C within 0.8 log units.
References


Table Wo1: Summary of experimental conditions occurring during smectite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wollastonite</td>
<td>25</td>
<td>2 – 9.6</td>
<td>Feruzzi (1994)</td>
</tr>
<tr>
<td>Wollastonite</td>
<td>25</td>
<td>3.1 – 12.2</td>
<td>Golubev et al. (2005)</td>
</tr>
<tr>
<td>Wollastonite</td>
<td>25</td>
<td>6.9 – 11</td>
<td>Pokrovsky et al. (2009)</td>
</tr>
<tr>
<td>Wollastonite</td>
<td>23.5</td>
<td>4.5 – 7</td>
<td>Rimstidt and Dove (1986)</td>
</tr>
<tr>
<td>Wollastonite</td>
<td>25</td>
<td>0.9 – 12</td>
<td>Schott et al. (2012)</td>
</tr>
<tr>
<td>Wollastonite</td>
<td>25</td>
<td>2, 4, 7</td>
<td>Sverdrup (1990)</td>
</tr>
<tr>
<td>Wollastonite</td>
<td>25</td>
<td>1.9 – 6.2</td>
<td>Weissbart and Rimstidt (2000)</td>
</tr>
<tr>
<td>Wollastonite</td>
<td>25 – 80</td>
<td>1.6 – 12.4</td>
<td>Xie (1994)</td>
</tr>
<tr>
<td>Wollastonite</td>
<td>25</td>
<td>-</td>
<td>Xie and Walther (1994)</td>
</tr>
</tbody>
</table>

Figure Wo1: Summary of experimentally measured smectite steady-state dissolution rates reported in the literature at 25 °C as a function of pH. The results of the model are plotted as the dotted line.
Zoisite

Zoisite (Ca$_2$Al$_3$Si$_4$O$_{12}$OH) is a Ca endmember of the epidote group of minerals. To date, only a single study has reported zoisite dissolution rates (Sverdup, 1990). The dissolution rates of zoisite reported by Sverdup (1990) match closely corresponding values reported by the same author on epidote. As can be seen in Fig Ep1 the dissolution rates of epidote reported by Sverdup (1990) have a pH variation inconsistent with rates reported in the majority of other studies. As such we have adopted the epidote fit to describe the dissolution rates of zoisite in this study. This approach contrast to that of Palandri and Kharaka (2004) who fit these Sverdrup (1990) rates to a distinct equation. As such, rate calculations performed using the Palandri and Kharaka (2004) equations would yield 25 C dissolution rates of zoisite that differ by as much as 3 orders of magnitude from those of epidote depending on pH; which is likely unrealistic considering the similarity of their respective structures and compositions.
References


Table Z1: Summary of experimentally measured zoisite steady-state dissolution rates reported in the literature.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>T, °C</th>
<th>pH range</th>
<th>Reference</th>
</tr>
</thead>
</table>

Figure Z1: Summary of experimentally measured zoisite steady-state dissolution rates reported in the literature as a function of pH. The results of the model are plotted as the dotted line.