Supporting information

The partitioning of a trace element between a carbonate mineral and aqueous solution is generally expressed by the distribution coefficient, \( K_d \), which is defined as

\[
K_d = \frac{X_{\text{TrCO}_3}}{X_{\text{MCO}_3}} \left( \frac{m_{\text{Tr}}}{m_M} \right)
\]  \hspace{1cm} (A1)

McIntire (1963) calculated that, \( K_d \) is related to the quotient of the thermodynamic solubility products of major element carbonate (MCO\(_3\)) and trace element carbonate (TrCO\(_3\)) and \( K_d \) can be expressed:

\[
K_d = \left( \frac{K_{\text{MCO}_3}}{K_{\text{TrCO}_3}} \right)^{\frac{1}{T_3}} \gamma_{\text{Tr}}^T \left( \frac{C_{\text{Tr}}}{C_{\text{M}}^*} \right) \exp \left( \frac{-\Delta \mu}{\nu_d R T} \right)
\]  \hspace{1cm} (A2)

The amount of Ca and Tr precipitated per kilogram of water passing through the reactor is:

\[
n_{\text{Ca}} = (m_{\text{in}})_{\text{Ca}} - (m_{\text{out}})_{\text{Ca}} \text{(kg)}  \hspace{1cm} (A3)
\]

\[
n_{\text{Tr}} = (m_{\text{in}})_{\text{Tr}} - (m_{\text{out}})_{\text{Tr}} \text{(kg)}  \hspace{1cm} (A4)
\]

From the definition of mole fraction;

\[
X_{\text{TrCO}_3} = \frac{n_{\text{TrCO}_3}}{n_{\text{TrCO}_3} + n_{\text{CaCO}_3}}
\]  \hspace{1cm} (A5)

\[
X_{\text{CaCO}_3} = \frac{n_{\text{CaCO}_3}}{n_{\text{TrCO}_3} + n_{\text{CaCO}_3}}
\]  \hspace{1cm} (A6)

So, the Eq.A3, A4, A5 and A6 can be written as:

\[
X_{\text{TrCO}_3} = \frac{n_{\text{TrCO}_3}}{n_{\text{TrCO}_3} + n_{\text{CaCO}_3}} = \frac{(m_{\text{in}})_{\text{Tr}} - (m_{\text{out}})_{\text{Tr}}}{(m_{\text{in}})_{\text{Ca}} - (m_{\text{out}})_{\text{Ca}}}
\]  \hspace{1cm} (A7)

Also because, the effluent stream (out) is simply the contents of the reactor, and the concentration ratio of Tr to Ca in the reactor is;

\[
\frac{m_{\text{out}}}_{\text{Tr}}\frac{m_{\text{out}}}_{\text{Ca}}
\]

This ratio and Eq. A7 can be substituted into the definition of \( K_d \) to give;

\[
K_d = \left( \frac{X_{\text{TrCO}_3}}{X_{\text{CaCO}_3}} \right)^{\frac{1}{T_3}} \gamma_{\text{Tr}}^T \left( \frac{C_{\text{Tr}}}{C_{\text{M}}^*} \right) \exp \left( \frac{-\Delta \mu}{\nu_d R T} \right)
\]  \hspace{1cm} (A8)

After measuring the \( m_{\text{in}}_{\text{Tr}}, m_{\text{in}}_{\text{Ca}}, m_{\text{out}}_{\text{Tr}}, m_{\text{out}}_{\text{Ca}} \) and each sampling time, the \( K_d \) can be given. Eq. A8 can be rearranged to;

\[
\frac{m_{\text{out}}_{\text{Tr}}}{m_{\text{in}}_{\text{Tr}}} = \frac{1}{K_d \left( \frac{m_{\text{in}}}{m_{\text{in}}_{\text{Ca}}} \right) - K_d + 1}
\]  \hspace{1cm} (A9)

\( f_i \) indicates the fraction of species \( i \) remaining in solution, as:

\[
f_i = \frac{m_{\text{out}}_{i}}{m_{\text{in}}_{i}}
\]  \hspace{1cm} (A10)

So, the Eq. A10 becomes:

\[
f_{\text{Tr}} = \frac{1}{(K_d/f_{\text{Ca}}) - K_d + 1}
\]  \hspace{1cm} (A11)

And \( F_i \) defines the fraction of species \( i \) precipitated from solution, as

\[
F_i = 1 - f_i
\]  \hspace{1cm} (A12)

So that, the Eq. A12 becomes:
\[ f_{Tr} = \frac{1}{K_d \left( \frac{1}{1 - F_{Ca}} \right) - K_d + 1} \]  \hspace{1cm} (A13)

This can be rearranged to:

\[ f_{Tr} = \frac{1}{K_d \left( \frac{1}{1 - F_{Ca}} \right) - K_d + 1} \]  \hspace{1cm} (A14)

**Notation**

- **M**: major element
- **Tr**: trace element
- **m_{Ca}**: concentration of Ca element in aqueous solution, mol/kg
- **m_{Tr}**: concentration of a trace element in aqueous solution, mol/kg
- **n_{Ca}**: number of moles of Ca element
- **n_{Tr}**: number of moles of the trace element
- **R**: the gas constant, 8.314 J/mol K
- **T**: temperature, K
- **\nu_a**: ratio of the number of anions in Tr_{CO_3} to the number of anions in M_{n}CO_3
- **X_{CaCO_3}**: mole fraction of calcite component in the solid solution
- **X_{TrCO_3}**: mole fraction of the trace element carbonate component in the solid solution
- **\Delta \mu**: difference between the chemical potential of Tr_{CO_3} in a pure crystal of Tr_{CO_3} and its chemical potential as a solid solution component in MCO_3
- **\gamma**: activity coefficient of element in aqueous solution
- **Fe_{w}-OH**: weak-sites of Fe(OH)_3
- **Fe_{s}-OH**: strong-sites of Fe(OH)_3
- **Al_{w}-OH**: weak-sites of Al(OH)_3
- **Al_{s}-OH**: strong-sites of Al(OH)_3