Supplementary Material

Surrogate modelling-assisted comparison of reactor schemes for carbon dioxide removal by enhanced weathering of minerals using seawater

Jinyuan Zhang[1], Aidong Yang[1], Richard Darton[1], Lei Xing[2], Adam Vaughan[1]

[1]Department of Engineering Science, University of Oxford, OX1 3PJ, Oxford, UK

[2]Department of Chemical Process Engineering, University of Surrey, GU2 7XH, Guildford, UK

Section S1. Derivation of Equation 5

Consider a single spherical particle with a mass m_p, volume V_p, diameter d_p and density ρ_p. The change of m_p in time (due to dissolution) can be related to the change of d_p:

$$\frac{dm_p}{dt} = \rho_p \frac{dV_p}{dt} = \frac{\pi}{2} \rho_p d_p^2 \frac{dd_p}{dt} \quad \text{(S1)}$$

However, the dissolution process with a specific surface dissolution rate R_d means

$$\frac{dm_p}{dt} = \pi d_p^2 R_d M_p \quad \text{(S2)}$$

where M_p is the molecular weight of the mineral. Combining Equations S1 and S2 gives:

$$\frac{dd_p}{dt} = \frac{2}{\rho_p} R_d M_p \quad \text{(S3)}$$

On the other hand, the particle surface area per unit bed volume, a_s, relates to d_p by:

$$a_s = \frac{6}{d_p}(1 - \epsilon) \quad \text{(S4)}$$

where ϵ is the bed porosity and remains approximately constant (~0.39) for the range of particle size and bed diameter considered in this work. The range of change in $a_s$due to dissolution, R_a can thus be derived by time-differentiating the right-hand-side of equation S4:

$$R_a = -6(1 - \epsilon) d_p^{-2} \frac{dd_p}{dt} \quad \text{(S5)}$$

Substituting Equation S3 into S5 yields Equation 5 in the main text:

$$R_a = -12(1 - \epsilon) \rho_p^{-1} d_p^{-2} M_p R_d$$

Section S2. Additional data for modelling forsterite dissolution kinetics

The activity of species *i* in the liquid phase is calculated by:

$$a_i = \gamma_i \, c_i \quad \text{(S6)}$$

where γ_i is the activity coefficient of species *i* and c_i is the aqueous concentration (M) of species *i*.

Activity coefficients for all species except H^+ are calculated using the Truesdell-Jones equation [1]:

$$log_{10} \gamma_i = -A \, {z_i}^2 \left(\frac{\sqrt{I}}{1 + B \, a_i^0 \sqrt{I}} \right) + b_i \, I \quad \text{(S7)}$$

where *A* and *B* are general constants found from aqion hydrochemistry software [1], a_i^0 and b_i are species specific constants found from WATEQ4F thermodynamic database [2], z_i is the charge of species *i* and *I* is the ionic charge of the solution (M) found from equation S8:

$$I = \frac{1}{2}\sum_{i=1}^{n} c_i\, {z_i}^2 \tag{S8}$$

The activity coefficient of H^+ is found according to the extended Debye-Hückel equation [1] (equation S9), following the convention used in WATEQ4F:

$$log_{10}\, \gamma_i = -A\, {z_i}^2 \left(\frac{\sqrt{I}}{1 + B\, a_i^0\, \sqrt{I}}\right) \tag{S9}$$

The parameters are from the same source stated above.

Table S1: Parameters for forsterite dissolution kinetics and equilibrium

parameter	value	source
A_1	83800 [mol/m^2/s]	[3]
E_1	6.72E+04 [J/mol]	
n	0.47	
A_2	1.58E+03 [mol/m^2/s]	
E_2	7.90E+04 [J/mol]	
A	-1098.376618	[4]
B	-0.153857	
C	73215.029991	
D	391.599004	
E	-3706160.894881	

References

[1] aqion, “Activity Coefficients (Activity Models).” [Online]. Available: https://www.aqion.de/site/101. [Accessed: 09-Apr-2021].

[2] Ball J. W. and Nordstrom D. K., “User’s manual for WATEQ4F, with revised theromodynamic data base and test cases for calculating speciation of major, trace and redox elements in natural waters”, 2001.

[3] James L. Palandri and Yousif K. Kharaka, A COMPILATION OF RATE PARAMETERS OF WATER-MINERAL INTERACTION KINETICS FOR APPLICATION TO GEOCHEMICAL MODELING. U.S. GEOLOGICAL SURVEY PEN FILE REPORT 2004-1068, 2004

[4] Thermoddem geochemical database (https://thermoddem.brgm.fr/).