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A general, microkinetic model for dissolution of silicate and aluminosilicate minerals and glasses as a function of pH and temperature

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We present a microkinetic model for dissolution rates for a range of silicate materials as a function of pH and temperature. The goal was to predict stone wool dissolution rates as a function of material and solution composition, in order to evaluate key aspects of biosolubility and material durability. Our model is applicable to minerals as well as amorphous (alumino)silicate materials, with variable Al:Si composition. Comparisons between the model and experimental dissolution rates for quartz, anorthite, amorphous silica and calcium aluminosilicate glass are shown in Figure 1.

The model assumes that the dissolution rate is the sum of all hydrolysis rates for the neutral and charged silanol and aluminol surface groups. The enthalpy barriers for hydrolysis of the various surface groups were fitted to obtain agreement with experimental dissolution rates, for pH 2-12, for quartz, anorthite, amorphous silica and amorphous calcium aluminosilicate glass (CAS). We found that the fitted barriers for hydrolysis agreed with density functional theory (DFT) calculations on CAS model systems. In particular, hydrolysis barriers of charged surface groups were significantly lower than for neutral groups, which was also consistent with our DFT calculations.

Because of the central importance of charged surface groups to the pH dependence of the dissolution rate, the degree of (de)protonation was therefore critical. We determined the degree of (de)protonation theoretically, using a combination of computational chemistry, for prediction of the pKa, and calculation of the screened surface electrostatic repulsion between (de)protonated silanol and aluminol groups. By taking into account the electrostatic repulsion at the surface, the dissolution rate slope with pH agrees with experiments for all materials and pH, without resorting to empirical rate laws, which supports our developed theory. By also including the entropy loss of the hydrolysis reaction, the absolute agreement with experiments is very good, including total dissolution rates as well as activation energies for quartz at pH 4 and 6. All parameters required for the model are physically meaningful and based on simple thermodynamic quantities, making the model generalizable to other silicate materials beyond the ones we compare to in this paper.

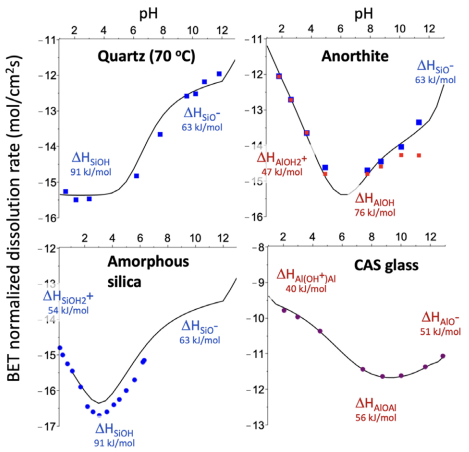

Figure 1. Comparison of dissolution rates determined using the microkinetic model and experiments for quartz, anorthite, amorphous silica and calcium aluminosilicate glass.