Introducing a new technique to distinguish between surface and transport controlled mineral dissolution

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Laboratory experiments can provide insight in rate-determining dissolution regimes, whether mineral dissolution is controlled by surface reactions or by transport of ions away from the surface. To date, a good understanding of mass transport to and from the mineral surface has been achieved by (a) the use of a rotating disk[1], or (b) locating a calcite crystal in part of one wall of a rectangular duct, generating laminar conditions[2]. However, these techniques depend on complex custom equipment to ensure hydrodynamic requirements are met. Geochemical kineticists often attribute differences in mineral dissolution kinetics to differences in experimental design, affecting the dissolution regime[3].

Here, we are introducing a purpose-built flow-through time-resolved analysis (FT-TRA) module, which can be employed to empirically assess which dissolution regime is dominating the forward reaction. A mineral sample, loaded in a flow-through cell, is exposed to a continuous incoming stream of eluent at a given flow-rate using an automated gradient pump. By varying the flow rate and assuming well mixed conditions in the flow-through cell (25 μl), a range in eluent residence time is established. If an analyte steady state concentration increases proportionally to the eluent residence time through the origin, we conclude the forward dissolution reaction to be surface controlled. If this increase is non-linear or does not go through the origin, transport control is affecting the forward reaction under the hydrodynamic conditions used.

Using our findings from two minerals (calcite and forsterite) we ground truth our hypothesis. Even using very short eluent residence times, calcite dissolves under a transport controlled regime. In contrast, forsterite dissolves under surface control across a wide range of residence times.

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