

Pore scale visualization of multiphase reactions in the unsaturated zone

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Mineral-fluid reactions are important controls on element and carbon cycling, yet predictions of large-scale mass transport are hindered by the scale-dependence of reaction rates and the use of the continuum approach in numerical models that represent porous media with volume-averaged properties [1] [2]. In particular, the unsaturated zone exhibits highly heterogeneous microenvironments that are subject to significant physical change during evaporation, imbibition, and drainage [3]. We hypothesize that pore scale reaction rates will be highly dynamic in these environments, but few experiments to date have allowed direct visualization of reaction progress in unsaturated material [4]. Here, we use microfluidics experiments to examine mineral dissolution-precipitation reactions under evaporative conditions and identify pore-scale processes that control reaction rate. The reaction of brucite [Mg(OH)₂] with gaseous CO₂ to form carbonate minerals was directly visualized using a glass micromodel. The entrainment of mineral particles by a mobile gas-water interface during evaporation dramatically altered the relative abundance of reactive mineral surface area and fluid reservoir volume. This ratio, which directly influences reaction rate and reaction progress, was observed to vary by orders of magnitude during a single experiment. Force balance analysis revealed that silt to sand-sized particles could be mobilized by the retreating gas-water interface due to surface tension forces, a size fraction likely to include the reactive primary minerals in soils (e.g., feldspars). In addition, the heterogeneous distribution of brucite within the micromodel was observed to dictate the extent and distribution of secondary carbonate precipitates. The potentially dynamic nature of the reactive primary phase distribution, suggests that spatial and temporal variability of pore scale reaction rate may be significant during evaporation, imbibition, or drainage in the unsaturated zone.

[1] Molins *et al.* (2012) *Water Resour. Res.* **48**, W03527. [2] Li *et al.* (2006) *Adv. Water Resour.* **29**, 1351-1370. [3] Bresson & Moran (1995) *Eur. J. Soil Sci.* **46**, 205-214. [4] Kim *et al.* (2013) *Lab Chip* **13**, 2508-2518.