

Measuring and interpreting the dissolution rates of cementitious silicate and aluminate minerals

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Chemical interactions among silicate, aluminate, and aluminosilicate minerals in high-pH aqueous solutions are responsible for every important change in the microstructure and properties of cementitious construction materials. Characterization of the reaction kinetics is needed to enable better predictive modeling and for better stewardship of civil infrastructure. It is made difficult by the fact that reactions in these multiphase systems are coupled in ways that are not well understood and, like natural geochemical processes, appear to be greatly influenced by defects and impurities. Consequently, progress toward better understanding of these reactions has been slow. Measuring the dissolution rates of isolated cementitious minerals in aqueous solutions is a logical point to begin building a repository for cement reaction kinetics data and is a prerequisite for understanding multiphase interactions.

Digital holographic microscopy (DHM) has emerged as a promising method for assessing rapid *in situ* changes to mineral surface topography in static or flowing solutions. It has been used recently to measure dissolution rate spectra of gypsum [1] and calcite [2] in flowing water. Applying the same methods to tricalcium aluminate and dicalcium silicate is challenging because of their rapid reaction and severe roughening upon contact with water. We address those challenges for DHM by modifying the water activity [3], which both retards the dissolution and enables the formulation of a rate law as a function of water activity. The form of the resulting rate law for dicalcium silicate can be analyzed in terms of molecular detachment rates, and the large local variation in rates can be partially understood in terms of compositional heterogeneities, observed by X-ray microprobe analysis, which induce lattice strain and thereby enhance local dissolution rates.

[1] Feng *et al.* (2017) *Chem Geol*, **460**, 25-36. [2] Brand, Feng, & Bullard (2017) *Geochim Cosmochim Acta*, **213**, 317-329. [3] Brand & Bullard (2017) *Langmuir*, **33**, 9645-9656.