Goldschmidt2017 Abstract

Defect control of Ca_3SiO_5 dissolution in early cement hydration reactions

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A significant theoretical framework [1–3] allows dissolution kinetics to be understood as the link between material properties (surface free energy, elastic moduli) and defect centers (screw dislocations, hollow cores, surficial etch pits), and is the formal, mechanistic basis for the step wave model [4]. We compare the ΔG_r dependence of well-studied silicate and carbonate dissolution rates, show how differences in this dependence reflect the role of ΔG_{crit}, and examine published data for monoclinic Ca_3SiO_5 (C3Sm) dissolution in this context. Although small-scale etch pits on C3Sm surfaces have been documented, their mechanistic role in terms of overall dissolution and hydration remains enigmatic. Comparison with stepwave model results suggest that C3Sm dissolution may be unique, i.e., observed rates do not reflect simple dissolution of the anhydrous mineral, regardless of the maintenance of its surface far from equilibrium.

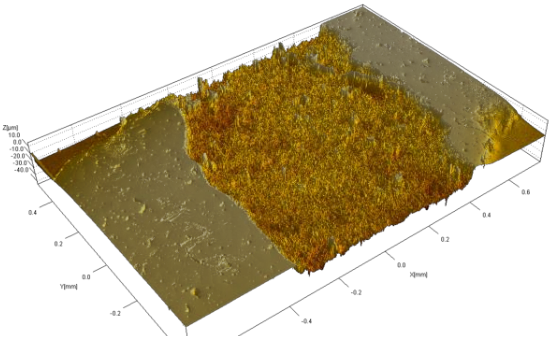

Fig. 1. Pure water dissolution of Ca_3SiO_5 (VSI data)

We discuss new C3Sm dissolution data highlighting the very early attack of the surface and its subsequent alteration. These results underscore the general difficulty in using free energy terms to parameterize dissolution kinetics; in cementitious materials, such approaches are of dubious merit.

[1] F.C. Frank, *Acta Crystallographica* **4**, 497 (1951). [2] N. Cabrera & M.M. Levine, *Philosophical Magazine* **1**, 450 (1956). [3] B. van Der Hoek, J.P. van der Eerden, & P. Bennema, *Journal of Crystal Growth* **56**, 108 (1982). [4] A.C. Lasaga and A. Luttge, *Science* **291**, 2400 (2001).