

Development of steady-state surface topography and the determination of dolomite dissolution rates

G.D. SALDI^{1*}, D. DAVAL¹, M. XU², S.R. HIGGINS²
AND K.G. KNAUSS¹

¹ESD, LBNL, Berkeley CA 94720, USA

(*correspondence: gdsaldi@lbl.gov)

²Department of Chemistry, WSU, Dayton, OH 45435, USA

The knowledge of the elementary mechanisms controlling the reactivity of mineral surfaces which undergo dissolution is critical to the development of rate-models that can accurately describe the processes occurring at the mineral/water interface. The correct interpretation of experimentally measured steady-state dissolution rates is affected by the duration of the experiments and by the development of a steady-state surface morphology, particularly at conditions close to equilibrium [1, 2].

Dolomite dissolution has been studied by hydrothermal atomic force microscopy (HAFM) on fresh-cleaved crystal surfaces as a function of the chemical affinity from 60 to 100 C, while a parallel series of experiments has been performed on a mineral powder of the same origin, under the same conditions, using a mixed-flow reactor.

The microscopic observations show the development of rectangular-shaped etch pits at mildly acidic pH's and a progressive decrease of the step velocities accompanied by a concurrent rounding of the acute step morphology when a carbonate-rich solution is introduced in the AFM cell. This change required several hours to be clearly manifested and effectively slow the rates of dissolution. Depending on the previous history of the sample, the same morphological steady-state may even require a few days to be attained. The results obtained from bulk experiments confirm the inhibiting effect of both carbonate species and aqueous Ca on dissolution and indicate that this drastic decrease of the rates is the consequence of a lower etch pit density.

[1] Bose S. *et al.* (2008) *Geochim. Cosmochim. Acta* **72**, 759–770. [2] Arvidson R.S. & Lutge A. (2010) *Chem. Geo.* **269**, 79–88.

Biologically enhanced silicate mineral dissolution for CO₂ sequestration

S.S.S. SALEK¹, R. KLEEREBEZEM¹, H.M. JONKERS²
AND M.C.M. VAN LOOSDRECHT¹

¹Delft University of Technology, 2628BC Delft, The Netherlands

²Delft University of Technology, 2628CN Delft, The Netherlands

During the past two centuries release rate of CO₂ to atmospheric reservoir has been increased by combustion of fossil fuels [1]. The consequence of CO₂ accumulation in the atmosphere is reflected as global climate change which is one of the main challenges facing humanity today [2]. CO₂ fixation as carbonates has been introduced as one the most sustainable and promising mitigation methods [3]. The efficiency of this method however is mainly limited by the raw material availability (Ca or similar divalent cations) [4]. Naturally occurring alkaline silicates are rich in divalent cations and have large reservoirs on earth; however with low dissolution rate. In the present study we describe dissolution enhancement of an alkaline silicate (wollastonite) during microbial anaerobic fermentation producing organic acids. Fermentation is an intermediate step in the anaerobic digestion process, a common process used for industrial or domestic purposes to manage waste and/or to release energy. Integration of wollastonite in an anaerobic fermentation process can result in release of Ca to the solution and neutralization of the process. ICP-OES and HPLC measurements showed an increase of dissolution rate by both proton release resulted from dissociated organic acids and complexation of Ca with these organic-products. Upon subsequent degradation of the organic acids to biogas, inorganic CO₂ will be sequestered as carbonate mineral. This work could provide an alternative route to reduce climate impacts from waste treatment plants.

[1] Falkowski, P. *et al.* (2000) *Science* **290**(5490), 291–296.
[2] Rockstrom, J. *et al.* (2009) *Nature* **461**(7263), 472–475.
[3] Lackner, K. S. *et al.* (1995) *Energy* **20**(11), 1153–1170.
[4] Renforth, P. *et al.* (2011) *Environmental Science & Technology* **45**(6), 2035–2041.