Goldschmidt2020 Abstract

Multiscale Investigation of Fluid-Silicate Interfaces and their Control on Dissolution Kinetics

BASTIEN WILD[1], CLAIRE WHITE[1,2] AND IAN BOURG[2,3]

[1] Andlinger Center for Energy and the Environment
[2] Department of Civil and Environmental Engineering
[3] Princeton Environmental Institute

Princeton University, Princeton, NJ 08544, USA
bwild@princeton.edu

Silicate-fluid interfaces are ubiquitous in naturally-occurring geochemical reactions and technological processes. Our understanding of the functioning of these interfaces, and in particular of their control on matter and energy exchanges through dissolution processes, determines, for example, our ability to reliably forecast mineral weathering and associated carbon fluxes or nutrient production, as well as the durability of urban infrastructure and nuclear waste depositories.

Navigating between fundamental processes occurring at the molecular scale and their macroscopic consequences in the field or in operational contexts has been a challenge for decades. One explanation might reside in the fact that processes at intermediate length and time scales are still incompletely probed and understood.

This study is aimed at unravelling some of the emerging nanometer to millimeter scale processes controlling the dissolution rate of silicate materials, encompassing rock-forming minerals, glasses and cements. In particular, we have combined advanced experimental techniques with high-performance atomistic simulations to evaluate the impact of the formation of interfacial amorphous silica layers on silicate dissolution kinetics. Textural and transport properties of such nanoporous media have been elucidated using in-situ and/or surface sensitive techniques including synchrotron-based grazing-incidence small-angle X-ray scattering (GISAXS), X-ray reflectivity (XRR), transmission electron microscopy (TEM) and vertical scanning interferometry (VSI). Furthermore, the mobility of water and alkali ions have been evaluated via molecular dynamics (MD) modeling of model nanoporous systems, where we have generated relevant water-silica-ions interactions by including the effect of negative surface charge. We evaluated the ability of single cylindrical silica nanopores to sustain concentration gradients by estimating their permeability and salt rejection capabilities. Our results support the analogy of such systems with semi-permeable membranes or nanofluidic diodes, which may ultimately explain the ability of nanoporous interfacial layers to modulate fluid-mineral interactions.