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The role of nano-porosity during mineral armoring

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During interface-coupled dissolution and precipitation (ICDP), secondary mineral rims may either sustain reaction progress or induce passivation, yet the physical mechanisms underlying this contrasting behavior remain poorly understood. The aim of this work was to investigate how the porosity of secondary precipitates controls the transport of charged ions and mineral armoring. We analyzed samples from the study of Poonoosamy et al.[1], in which passivation was observed during barite overgrowth on celestine, and characterized the porosity of secondary barite. High-resolution electron microscopy (FIB-SEM and STEM) revealed dual-scale porosity in the barite rim, consisting of micrometer-scale pores and a pervasive network of 5-20 nm pores. Using pore-scale simulations on the resolved geometry, we show that nano-porosity alone substantially reduces diffusive transport through the barite rim. Finally, we conducted one-dimensional reactive transport simulations incorporating pore size-controlled solubility effects and assessed the roles of nanoconfinement and of the electrical double layer (EDL) in the mineral armoring process. Reactive transport simulations incorporating electro-diffusion through the Nernst-Planck model demonstrate that passivation emerges from the interplay between nanoporosity and electrostatic ion exclusion at charged pore surfaces, jointly reducing sulfate flux through the rim and constraining sulfate supply to the reaction front. This integrated approach provides a mechanistic description of rim formation and passivation and offers a framework for interpreting transport-limited behavior in ICDP systems relevant, e.g., to mineral carbonation, serpentinization, and subsurface fluid-rock interaction.

[1]: Poonoosamy J, et al. Effects of solution supersaturation on barite precipitation in porous media and consequences on permeability: Experiments and modelling. Geochim Cosmochim Acta 270, 43-60 (2020).