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The role of mineral surface reactivity during mineral armoring

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Interface-coupled mineral dissolution and precipitation processes are ubiquitous in nature and describe the simultaneous dissolution of a primary mineral and the precipitation of a secondary mineral. The latter can either create porosity and thus promote fluid exchange, leading to the complete replacement of the primary mineral, or form a protective coating that passivates its dissolution. Understanding these processes is therefore essential in nuclear waste disposal systems, where coupled dissolution-precipitation reactions control the alteration of vitrified waste forms, the corrosion and passivation of steel or copper canisters and the mineralogical transformation of bentonite clay barriers.

Here, we investigate how microscale surface reactivity governs interface-coupled dissolution–precipitation (ICDP) dynamics and passivation using the celestine - barite ($SrSO_4$-$BaSO_4$) replacement system as a model [1]. Microfluidic lab-on-chip reactors with quasi-2D and 3D geometries were employed to resolve mineral transformation in situ under controlled flow conditions. Time-lapse confocal Raman imaging and AFM analysis reveal that barite nucleation preferentially initiates at etch pits, steps, and kink-rich surfaces of celestine, while specific crystallographic faces exhibit distinct resistance to dissolution. Increasing aqueous Ba^{2+} concentration counterintuitively suppresses celestine dissolution, consistent with adsorption-induced blocking of reactive step and kink sites in accordance with the Terrace-Ledge-Kink framework. Hybrid Density-functional theory-solvation models, which can be used to simulate the kinetics of complete ion uptake processes at mineral-water interfaces [2], show that barite preferentially nucleates on interfaces rich in kinks and steps as well as on pre-etched planes. The reduced SO_4^{2-} release limits further barite precipitation and illustrates the close coupling of dissolution and growth. Supplementary simulations investigated the influence of different crystal surface stabilities on the mineral-water interface reaction kinetics in the $(Ba,Sr)SO_4$-H_2O system bridging experimental observed heterogeneous passivation to processes on the atomic scale. These combined approaches provide mechanistic insight into surface-controlled ICDP processes relevant to mineral replacement observed in nuclear waste disposal environments, etc.

[1] Poonoosamy et al. (2020). GCA 270, 43-60. [2] Rudin et al. (2024). Cryst. Growth Des. 24(19), 7774-7788.