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Investigating interface coupled mineral dissolution and precipitation processes using advanced analytical and modelling tools

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Interface-coupled dissolution and precipitation (ICDP) [1] occur in various subsurface applications, e.g., serpentine carbonation in the context of CO₂ sequestration, the carbonation of concrete causing degradation, anoxic steel corrosion in geological disposal facilities for nuclear waste, or during groundwater remediation using permeable reactive barriers. ICDP is characterized by the dissolution of a primary mineral and the precipitation of a secondary mineral on its surface (rim formation). The precipitated secondary minerals can either develop porosity and fractures that enable fluid exchange and thereby completely replace the primary minerals or shield/passivate them. Therefore, understanding the factors that regulate the development of the rim is critical for predicting the geochemical reactions and the resulting impacts on various geological and environmental systems over geological time scales. To this end, we developed a column experiment where we investigated the dissolution of celestine followed by the precipitation of barite [2]. This is a well-controlled chemical system that is neither pH- nor redox-sensitive. The theoretical models to describe passivation processes in our experiment (e.g., Daval et al., [2]) were simplified and did capture the underlying processes [3]. Micro-continuum modelling work showed that the porosity of the precipitates could play a secondary yet significant role in shaping the evolution of the co-dissolution and precipitation interface [5]. Currently, we investigate the micro and nano porosity of the secondary mineral using 3D FIB-SEM and evaluate the effective diffusivity in the precipitates using pore-scale modeling [6]. In addition, we conduct complementary microfluidic experiments combined with in-situ Raman 2D/3D and modelling to evaluate the effect of fluid velocities and solution saturation ratio (Peclet number and Damköhler number) with respect to the barite overgrowth thickness. These investigations will enable the identification of parameters that control or limit passivation and its associated potential effects on further mineral reactivity.

[1] Renard et al., Timescales of interface-coupled dissolution-precipitation reactions on carbonates, <https://doi.org/10.1016/j.gsf.2018.02.011>

[2] Poonoosamy et al., Combination of MRI and SEM to Assess Changes in the Chemical Properties and Permeability of Porous Media due to Barite Precipitation, <https://doi.org/10.3390/min10030226>

[3] Daval et al., Carbonation of Ca-bearing silicates, the case of wollastonite: Experimental investigations and kinetic modelling, <https://doi.org/10.1016/j.chemgeo.2009.01.022>

[4] Poonoosamy et al., Effects of solution supersaturation on barite precipitation in porous media and consequences on permeability: Experiments and modelling, <https://doi.org/10.1016/j.gca.2019.11.018>

[5] Deng et al., A reactive transport modeling perspective on the dynamics of interface-coupled dissolution-precipitation <https://doi.org/10.1016/j.apgeochem.2022.105207>

[6] Yang and Wang, Pore-scale modeling of chloride ion diffusion in cement microstructures, <https://doi.org/10.1016/j.cemconcomp.2018.11.011>

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