

The impacts of intragranular porosity on mineral dissolution rates

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Investigations of mineral reactions in porous media are typically focused on the dynamics at water-rock interfaces, i.e., the interplay between advection, diffusion, and kinetics at grain boundaries. This interplay, however, depends on whether the solid phase is permeable. To quantify the impacts of pervious interfaces, as would be the case for grains with intragranular porosity, we conducted systematic numerical experiments of a dissolving single mineral grain with different initial internal porosities under a range of reactive transport regimes. The simulation results highlighted that the evolution of a porous grain is shaped by reactions at the interface of and within the grain. The contribution of internal reactions, depending on the rate at which reactive fluids are depleted and transported into the grain, can account for up to 92% of the overall dissolution. While the overall reaction rate is enhanced by up to 67% due to the presence of internal reactions, the inward development of altered zones within the porous grain significantly mitigates the migration of the grain boundary. These effects are particularly evident in highly porous grains and under diffusion-controlled conditions. Insights gained from these numerical experiments were integrated into a hybrid conversion model, which was formulated to account for changes in grain properties (e.g., circularity, equivalent diameter, and average density) as the reaction progresses in a porous grain and is able to predict the overall reaction successfully. Furthermore, the impacts of intragranular porosity on dissolution rates and the hydraulic properties were investigated in a larger domain with grain packings. The presence of intragranular porosity enhances the

bulk dissolution rate by more than one order of magnitude; however, it limits the permeability increase, which is lower than that of the impervious grain packing by up to two orders of magnitude. Unlike the impermeable case, in which the permeability-porosity relationship remains consistent across reactive transport regimes, preferential surface reactions under advection-controlled conditions in packings with intragranular porosity lead to a less convex porosity-permeability relationship compared to diffusion-controlled regimes. Therefore, accounting for effects of intragranular porosity and dissolution regimes is necessary to accurately predict the long-term evolution of porous media.