Modeling calcite growth and dissolution at nano and microscale

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Calcite is the most abundant carbonate mineral in the Earth system. The description of its growth and dissolution is essential to understand natural weathering processes and the modern techniques of carbon sequestration by injection of reactive fluids in rocks. This talk summarizes the recent work of my research group in modeling dissolution and growth of calcite at nano and microscale using kinetic Monte Carlo simulations and scaling methods. First we studied the dissolution of the mineral with initially rough surfaces and confirmed the smoothening and the expansion of surface valleys which were observed in limestone dissolution in the laboratory [1]. Next we determined the rates of molecule detachment from calcite surfaces in alkaline environment and far from equilibrium conditions which simultaneously matched velocities of monolayer step propagation, formation of shallow pits on defect-free surfaces, dissolution rates of macroscopic grains, and the corresponding activation energies (we also explained why nanoscale grains dissolve much faster) [2]. Third, we studied the dissolution of the walls of a pore with a concentration gradient and showed that the resulting shapes significantly differ from the predictions of a corresponding reaction-diffusion equation (continuous model) for different conditions of the pore boundaries [3]. The propagation of monolayer steps from points of faster dissolution to points of slower dissolution is shown to be the mechanism controlling the pore shape and this mechanism may be incorporated into hydrodynamic approaches. Finally, in a recent work, we studied the growth on rough fracture walls of the same mineral in contact with supersaturated solutions. Step propagation also controls the growth and this result may be used to predict the final configurations of walls of various shapes (possibly with fracture clogging) and the times to reach those configurations.

References

[1] Geochim. Cosmochim. Acta 228, 27 (2018).

[2] Geochim. Cosmochim. Acta 292, 271 (2021); ACS Earth Space Chem. 5, 2755 (2021).

[3] Geochim. Cosmochim. Acta 379, 219 (2024).

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