From atomic simulations to larger system sizes - New approaches to upscaling surface reactivity

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In the context of enhanced reactive transport modeling in reservoir rocks, we examine ways to connect atomic simulation methods like kinetic Monte Carlo (kMC, see e.g., [1]) with reactive transport models at the pore scale and above (see e.g., [2]). Specifically, we are focusing on the relationship between atomic-scale reaction kinetics and the evolution of porosity and permeability of reservoir rocks, and, ultimately, to develop a reactive transport model that spans all the scales from nano to drill core. We examine a calcite-cemented Rotliegend sandstone, and in particular, the calcite dissolution kinetics.

One of the major challenges modellers face is that while there are atomic-scale simulations of calcite-water interaction available [3], these are limited in system size by computing power. There is a gap between these simulations and reactive transport models that necessarily operate at continuum scale. Also, the connection of kMC simulation results with experimental data is difficult due to this issue of scale.

To get a handle on this problem, we developed a new, fast geometrical simulation method based on the concept of Voronoi diagrams that has been successfully tested on the example system of a Kossel crystal [4]. First results of this strategy of upscaling kMC with geometrical methods are promising and additional applications in entirely different contexts [5] underscore the general applicability of the concept. We applied our new method to the calcite structure. First quantitative results show how to generate plausible models of the surface morphology and reaction rate maps of a dissolving calcite crystal while we manage to significantly reduce the required computational power. We conclude that methods of computational geometry are a viable strategy of upscaling atomic simulations.

[1] Meakin & Rosso (2008) *J. Chem. Phys.* **129**, 204106. [2] Yoon *et al.* (2012) *Water Resour. Res.* **48**, W02524. [3] Kurganskaya *et al.* (2016) *J. Phys. Chem. C* **120**, 6482-6492. [4] Rohlfs *et al.* (2018) *Minerals* **8**, 133. [5] Kaiser *et al.* (2018) *Algorithms* **11**, 37.