

## **Molecular mechanisms of carbonate dissolution: surface charge and ionic structure of mineral-water interface**

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Dissolution of carbonate minerals is a fundamental process taking place on the Earth surface, from the biosphere to deeply buried sediments. Despite the seeming simplicity of the carbonate crystal structure, surface sites exhibit diverse reactivity defining kinematics of reactive surface features, e.g. etch pits, steps and stepwaves. Their spatio-temporal arrangement can be very complex and non-predictable, thus creating spatio-temporal rate fluctuations [1]. Composition of the reacting fluid affects surface reactivity via defining protonation/deprotonation reactions and adsorption of ions [2]. This many-fold increase of reactive system's complexity requires a step-wise systematic study of the influencing parameters. In order to achieve this goal, we are working on a stochastic modelling approach combining equilibrium Grand Canonical Monte Carlo (GCMC) and Kinetic Monte Carlo (KMC) simulations into a one modelling platform. GCMC models allow us to obtain detailed quantitative information about surface site charge and electric double layer structure. This information is used to parameterize KMC models [3] with respect to the reactive site populations and estimate dissolution rates. A direct comparison of data obtained in simulations and experiments allows us to test the hypotheses used to construct the models. In this way we reveal the key molecular scale mechanisms controlling mineral dissolution rate in experimental and natural fluid media.

[1] Lüttge A., Arvidson R. S. and Fischer C. (2013), *Elements* **9**, 183–188.; [2] Churakov S. V., Labbez C., Pegado L. and Sulpizi M. (2014), *J. Phys. Chem. C* **118**, 11752–11762.; [3] Kurganskaya I. and Lüttge A. (2016), *J. Phys. Chem. C* **120**, 6482–6492.