Linking microscale to macroscale dissolution rates in carbonates with X-ray tomography imaging and stochastic modelling

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Reactive transport models at the continuum scale require reliable kinetic rate formulations to constrain the reactivity of minerals in rocks during dissolution/precipitation processes. Investigations of mineral surface reactivity have recently challenged the classical approach of determining dissolution rates from mineral powders in the lab as crystals or more generally mineral-fluid interfaces often exhibit heterogeneous and/or anisotropic reactivity. Here, we provide a detailed characterization of the dissolution kinetics of carbonate samples (calcite spar, aragonite, and micritic and microsparite rock fragments), time-lapse 3D X-ray microtomography with a resolution below 1 μm. The local dissolution rates determined at crystal faces, edges and corners from calcite dissolution experiments [1,2] serve to constrain a stochastic numerical model based on the probabilities of detachment of face, edge and corners elements obtained from kinetic Monte Carlo simulations. Based on the estimated detachment probability for each class of elements, of the size of the voxel, a selection of voxels is removed at each time step to mimic the dissolution process. The geometry of the carbonate samples measured experimentally using X-ray microtomography imaging are used as input for stochastic modelling. By tracking the topography and voxel detachments through time, we demonstrate that the model reproduces fairly well the observed topographic evolution of the fluid-mineral or fluid-rock interface during the experiments. In particular, the role of convex and concave interfaces is evidenced. This modelling approach provides a new tool to link dissolution rates at the microscale of fluid-mineral interfaces to macroscopic dissolution rates measured on bulk samples.

[1] Noiriel, C., Oursin, M., Saldi, G., and Haberthür, D. (2019). Direct determination of dissolution rates at crystal surfaces using 3D X-ray micro-tomography. ACS Earth and Space Chemistry, vol 3, no. 1, 101-108. DOI: https://doi.org/10.1021/acsearthspacechem.8b00143

[2] Noiriel, C., Oursin, M., and Daval, D. (2020). Examination of crystal dissolution in 3D: a way to reconcile dissolution rates in the laboratory? Geochimica et Cosmochimica Acta, 273, 1-25. DOI: https://doi.org/10.1016/j.gca.2020.01.003

Figure: View (as a cross-section) of a calcite spar geometry obtained with X-ray micro-tomogtaphy during 5 stages of dissolution and corresponding geometries from stochastic modelling