

EGU21-11079

<https://doi.org/10.5194/egusphere-egu21-11079>

EGU General Assembly 2021

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Impact of Surface Reactivity on the Simulation of Mineral Dissolution Rates

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The kinetics of mineral dissolution plays a key role in many environmental and technical fields, e.g., weathering, building materials, as well as host rock characterization for potential nuclear waste repositories. Mineral dissolution rates are controlled by two parameters: (1) transport of dissolved species over and from the interface determined by advective fluid flow and diffusion (transport control) and (2) availability and distribution of reactive sites on the crystal surface (surface reactivity control). Reactive transport models (RTM) simulating species transport commonly calculate mineral dissolution by using rate laws [1]. However, the applied rate laws solely depend on species concentration in the fluid. While the effect of transport-controlled processes is addressed in current RTM approaches, the intrinsic variability of surface reactivity is neglected. Experimental studies under surface-controlled dissolution conditions have shown that surface reactivity is heterogeneously distributed over the surface [e.g., 2]. This heterogeneity in reactivity is largely caused by nanotopographical structures on the crystal surface, such as steps and etch pits. These structures are generated through defects in the crystal lattice. At these structures, the high density of reactive kink sites is leading to a local increase in surface reactivity observable through high dissolution rates.

In this study, we test whether the current rate calculation approach applied in RTMs is sufficient to reproduce experimentally observed rate heterogeneities. We apply a standard RTM approach combined with the measured surface topography of a calcite single crystal [2]. Calcite is an important mineral component in the sandy facies of the Opalinus clay formation, that is under investigation for nuclear waste storage. The modeled surface dissolution rate maps are compared to experimentally derived rate maps. Results show that the current RTM is not able to reproduce the measured rate heterogeneities on the calcite surface. To improve the predictive capabilities of RTMs over the large time scales required for the safety assessment of nuclear waste repositories, the surface reactivity that is intrinsic to the mineral needs to be implemented into future rate calculations. Investigating calcite surface reactivity in the context of dissolution can also yield information about other kinetic surface processes such as the adsorption of radionuclides during transport. We show the integration of surface reactivity into rate calculation by using a proxy parameter. The slope of the crystal surface at the nm scale is applied. We show that by adding a factor based on the slope to the rate law the RTM is able to approximate experimental rate maps. Other proxy parameters such as surface roughness could yield similar results as well. The

implementation of surface reactivity proxy parameters will allow for a more precise prediction of host rock-fluid interaction over large time scales in RTMs, relevant for safety assessment of nuclear waste repositories.

[1] Agrawal, P., Raoof, A., Iliev, O. and Wolthers, M. (2020), *Advances in Water Resources*, 136, 103480. [2] Bibi, I., Arvidson, R.S., Fischer, C. and Lüttge, A. (2018), *Minerals*, 8, 256.