Comparison between experimentally measured calcite dissolution rates and 1D and 2D reactive transport simulations: Importance of fluid flow and temporal surface evolution

PHILIPPE ACKERER[1], ARNAUD BOUISSONNIE[1], MARIANNA MARINONI[1], DAMIEN DAVAL[1]

[11] Laboratoire d'Hydrologie et de Géochimie de Strasbourg, Université de Strasbourg /EOST-CNRS UMR 7517, 1 Rue Blessig, 67084 Strasbourg, France
ackerer@unistra.fr

Fast dissolving minerals are often considered as important contributors of river chemistry in catchments. Knowledge of their dissolution and precipitation rates are therefore crucial when it comes to model or to understand the geochemical behaviors of such areas. Hence, for decades, laboratory experiments have been performed in order to measure dissolution/precipitation rates. However, it has been shown that a discrepancy exists between dissolution rates measured in the field and those measured in laboratory. Several factors may be involved to explain this discrepancy such as the nucleation and growth of etch pits at the mineral surface, which influence the long term reactivity of minerals, or the effect of fluid saturation at the solution/minerals interface. In our study [1], classical mixed flow reactor experiments conducted at different saturation indices show that transition state theory (TST) cannot account for the dependence of calcite dissolution rate on fluid saturation. Instead, the rate data are in good agreement with the StepWave model (SWM). In a second experiment conducted in a column filled with a chemically inert porous medium, the reactivity of disseminated cm-sized calcite crystals was compared with outputs of 1D and 2D reactive transport simulations using either TST or SWM. We show that in this specific configuration, the best agreement between experiments and models is obtained when the fluid flow path is correctly modeled, whereas the choice of the rate law (TST or SWM) remains of second order. Finally, we will discuss how the comparison between the model and the experiment question the implementation of the reactive surface area parameter and its evolution through time.

[1] Bouissonnié, A., Daval, D., Marinoni, M., Ackerer, P., 2018. From mixed flow reactor to column experiments and modeling: Upscaling of calcite dissolution rate. Chem Geol 487, 63-75.