Technical University of Denmark

DTU

Impact of surface complexation and electrostatic interactions during pH fronts propagation in silica porous media: Experiments and model-based interpretation

Stolze, Lucien; Muniruzzaman, Muhammad; Rolle, Massimo

[Link back to DTU Orbit](#)

DTU Library
Technical Information Center of Denmark

Contribution ID : **452**   Type : **Oral 15 Minutes**

Impact of surface complexation and electrostatic interactions during pH fronts propagation in silica porous media: Experiments and model based interpretation

Wednesday, 16 May 2018 11:17 (15)

Propagation of proton fronts exerts a fundamental control on geochemical processes and contaminant transport in subsurface systems (Muniruzzaman & Rolle, 2015). Protons are of key importance in pore water solutions since they affect the sorption behavior of charged contaminants by setting the surface charge at the mineral-liquid interface. Therefore, it is of primary importance to enhance our understanding regarding proton transport and complexation on mineral surfaces in order to accurately describe and predict reactive transport processes in porous media. Although significant effort, based on experimental studies and model development, has been dedicated to investigate pH sorption and surface complexation behavior in batch systems, only a few studies addressed the impact of these processes under flow-through conditions (McNeece & Hesse, 2016).

In this work, we systematically investigate the transport and sorption behavior of proton under flow-through conditions by means of laboratory experiments and reactive transport modeling. We performed experiments both in batch (titration) and in flow-through setups packed with saturated silica media (i.e. quartz sand and glass beads) with different grain sizes as porous matrix. Batch experiments were focused on characterizing and obtaining insights on the pH sorption capacities of the porous matrix and on the electrostatic interactions at the mineral pore water interface. Successively, we performed flow-through experiments in laboratory columns by applying continuous injections of acidic solutions. Breakthrough curves of pH and all other major ions were measured at the outlet of the columns. In order to identify the importance of ionic strengths on charged species at the liquid-solid interface and on the multicomponent ionic transport, we performed parallel sets of analogous experiments (both in batch and column setups) with different background electrolyte solutions.

To quantitatively interpret the experimental results, we used a reactive transport model explicitly taking into account the triple-layer charge distribution multisite surface complexation (CD-MUSIC) model (Hiemstra & Van Riemsdijk, 1996) together with the Donnan approximation (Appelo & Wersin, 2007), and the cross-coupling of dispersive fluxes due to the Coulumbic interactions between aqueous charged species (Rolle, et al., 2013). The modeling approach was based on a coupling between the geochemical code PHREEQC with MATLAB using IPhreeqc module (Muniruzzaman & Rolle, 2016). The relative affinity constants of the surface complexation reactions were obtained through inverse modeling of the batch-titration and flow-through experiments. Experimental and modeling results suggest that the sorption behavior of protons differs in the considered silica porous media and are differently affected by the ionic strength of the background solutions.

References

Appelo, C. & Wersin, P., 2007. Multicomponent diffusion modeling in clay systems with application to the diffusion of tritium, iodide and sodium in opalinus clay. Environ. Sci. Technol., Volume 41, pp. 5002-5007.

Hiemstra, T. & Van Riemsdijk, W., 1996. A surface structural approach to ion adsorption: the charge distribution (CD) model.. J. Colloid Interf. Sci., Volume 179, pp. 488-508.

McNeece, C. & Hesse, M., 2016. Reactive transport of aqueous protons in porous media. Adv. in Water Resour., Volume 97, pp. 314-325.

Muniruzzaman, M. & Rolle, M., 2015. Impact of multicomponent ionic transport on pH fronts propagation in saturated porous media. Water Resour. Res., 51(8), pp. 6739-6755.

Muniruzzaman, M. & Rolle, M., 2016. Modeling multicomponent ionic transport in groundwater with IPhreeqc coupling: Electrostatic interactions and geochemical reactions in homogeneous and heterogeneous domains. Adv. Water Resour., Volume 98, pp. 1-15.

Rolle, M., Muniruzzaman, M., Haberer, C. & Grathwohl, P., 2013. Coulombic effects in advection-dominated transport of electrolytes in porous media: Multicomponent ionic dispersion. Geochim. Cosmochim. Acta, Volume 120, pp. 195-205.

Acceptance of Terms and Conditions

Click here to agree

Primary author(s) : STOLZE, Lucien

Co-author(s) : MUNIRUZZAMAN, Muhammad (Geological Survey of Finland, Neulaniementie 5, 70211 Kuopio, Finland); ROLLE, Massimo (Technical University of Denmark)

Presenter(s) : STOLZE, Lucien

Session Classification : Parallel 7-A

Track Classification : MS 1.11: Conservative and Reactive Transport of Charged Species in Permeable and Impermeable Porous Media