

Benchmarking of a general speciation code for charge regulated solid-liquid interfaces interacting on the nanoscale

TEBA GIL-DÍAZ^{1,2}, DANIEL JARA-HEREDIA¹, FRANK HEBERLING³, JOHANNES LUETZENKIRCHEN⁴, JULIAN LINK⁵, THOMAS SOWOIDNICH⁶, HORST-MICHAEL LUDWIG⁶, MICHAEL HAIST⁵ AND THORSTEN SCHÄFER⁷

¹Friedrich-Schiller University Jena

²Karlsruhe Institute of Technology

³Karlsruhe Institute of Technology (KIT)

⁴KIT

⁵Leibniz University Hannover

⁶Bauhaus University Weimar

⁷Friedrich Schiller University Jena

Presenting Author: teba.gil-diaz@uni-jena.de

Surface chemistry of mineral phases in aqueous environments is a topic of broad interest concerning technological applications and environmental studies. Few electrostatic surface complexation models (SCMs) directly account for the influence on both chemical (i.e., surface speciation and changes in solution speciation) and mechanical properties (i.e., osmotic pressure and inter-particle forces) when diffuse layer potential profiles of adjacent surfaces interact on the nanoscale and affect the respective surface chemistries [1].

This work presents a new speciation code that couples state-of-the-art SCMs (i.e., up to the Four Layer Model – FLM) for both symmetric and asymmetric interacting surfaces with a numerical solution for the Poisson-Boltzmann distribution of electrostatic potential and ions in the inter-particle space, including mono- and bidentate sorption cases, in arbitrary electrolytes through charge regulation. The concept is implemented as a Python-based code and within COMSOL. The flexibility, robustness and precision of the approach is demonstrated here in several benchmark calculations.

The variety of benchmark calculations test the present code against published results or defined systems that have been simulated with established speciation codes [2,3]. Examples include aqueous speciation, surface complexation and various interaction force simulations. The developed code overcomes restrictions of previous codes (e.g., diffuse layer – DLM, Basic Stern – BSM, or a triple layer model – TLM, for monovalent electrolytes), broadening and facilitating future applications by considering charge regulation effects. This new development provides insights into the chemistry and speciation of solid-liquid interfaces in narrow pores. It offers mechanistic insight into coupled chemo-mechanical effects via precise calculations of osmotic pore pressure, most relevant for nanoporous systems.

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References:

[1] Zhao, Ebeling, Siretanu, van den Ende, & Mugele (2015).