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In-Tandem Microcalorimetry-2D Perturbation Correlation Spectroscopy for Resolving Catalytic and Non-catalytic Reactions at the Oxide-Water Interface

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Interactions at the mineral-water interface can be catalytic or non-catalytic. Our presentation will cover the integration of high-temporal resolution energy measurements (from flow adsorption microcalorimetery) with in-tandem 2D fluorescence and UV-vis spectroscopic analysis to resolve energetics and kinetics of catalytic and non-catalytic interactions between organic solutes and geomaterials. Specific focus will be placed on interactions of natural organic matter and amino-based biomolecules at the mineral-water interface of Fe-bearing oxides. Energy measurement indicate that while non-catalytic reactions at the oxide-water interface has energy dynamics that are directionally opposite in the forward and reverse stages; catalytic reactions have energetics that are directionally the same or only partially opposite. For example, while simple non-catalytic sorption and desorption of natural organic matter to and from an iron oxide is exothermic and endothermic respectively; catalytic organic-oxide interactions are exothermic or mixed exothermic/endothermic in both the sorption and desorption phase. Coupling these measurements with 2D perturbation correlation fluorescence and/or UV-vis spectroscopy reveal that while temporal changes in fluorescence or UV-vis spectra for non-catalytic interactions were intensity-driven that for catalytic reactions were complexity-based – typically increasing in complexity. Discussions will look at the catalytic versus non-catalytic interactions for a suite of 1) natural organic matter solutes from different sources and 2) non-hydrolyzable (glycine and alanine) versus hydrolyzable (ATP) amino-based biomolecule on a range of Fe-bearing oxides.