



Katz et al., 2012 : Electron small polarons and their mobility in iron (oxyhydr)oxide nanoparticles

Geochemical Reaction Intermediates in Metal Redox Cycling

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Redox-active metals in the environment, including iron and manganese, undergo cycles of oxidation state change driven by photochemical, geochemical and biological processes. In low-temperature aqueous settings, including surface waters, soils and sediments, metal redox cycling frequently involves the formation or dissolution of minerals, including nanoparticles. Such redox reactions control the speciation and mobility of these metals as well as contaminants and nutrients that can sorb to the surfaces of mineral precipitates or participate in electron-transfer processes. A robust understanding of metal redox cycling is required to anticipate how near-surface settings evolve in response to environmental change.

Metal redox reactions proceed through a combination of chemical steps that can include electron and proton transfer, the breaking or formation of bonds, and mineral phase transformation. The rate and yield of the overall reaction depends upon the nature and lifetime of reaction intermediates, information that is typically not accessible by conventional kinetics studies. For example, it has been proposed that structural iron(II) sites are key intermediates in (photo)reduction reactions of iron(III) oxides, but direct observation of such species has been impossible until very recently. This presentation will give an introduction to time-resolved experimental methods that provide direct characterization of geochemical reaction intermediates. We are using these methods to identify crystal and solution chemical controls on the rates and yields of environmental metal cycling.

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