

Redox chemistry of the Fe mineral – water Interface

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We have been studying the redox chemistry of the Fe mineral – water interface for awhile now, yet I think a fundamental, molecular understanding of what happens still eludes us. Two of my electrochemistry heroes, Werner Stumm and John Westall, recognized decades ago how dang complicated it was, especially once we learned that microbes could respire on Fe(III) in these minerals! Building on the electrochemistry insights from Werner and John, we have used Mössbauer spectroscopy, isotopes, and electrochemistry to explore the wild, wild west of redox chemistry at the Fe-mineral water interface for the last few decades.

We confirmed several things that Werner and John suspected. We observed interfacial electron transfer (IET) between sorbed Fe(II) and structural Fe(III) in Fe oxides and Fe clay minerals. In some cases, a cool templating effect occurred that resulted in growth of the underlying oxides (e.g., homoepitaxy on goethite and hematite), whereas in other cases, it remains unclear what is happening (e.g., magnetite and ferrihydrite). We were also surprised by how difficult it was to shut down IET. IET happens in the presence of carbonate, phosphate, silicate, organic matter — you name it, we tried it. About the only thing that inhibited IET was removing defects by hydrothermal treatment or reaction with Fe(II). Even more surprisingly, we observed that the newly formed Fe(II), in some cases, mixes with the surrounding water. The extent of mixing varies, but in some cases, we observed near complete mixing of the Fe atoms. That blew our mind (and I think it would surprise them as well!). Today, I'd like to provide a recap and update on these findings and also share some of our more recent work exploring how magnetite stoichiometry and incorporation and sorption of metals (e.g., Al, Ni, Zn, Cu) influences the redox chemistry of the Fe-mineral water interface.