

Geochemical controls on schwertmannite transformation upon aging

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Weathering of hard-rock sulfide deposits from mineral resource extraction or natural exposure creates conditions that favor the formation acidic drainage with elevated concentrations of metals and metalloids that mix with surface waters, potentially impairing large tracts of mountain streams, fisheries, and drinking water sources. The oxidative dissolution of sulfide minerals generates high concentrations of iron(II), sulfate, acidity, and when combined with microbial Fe(II) oxidation, favor the precipitation of poorly crystalline schwertmannite (ideal composition $\text{Fe}_8\text{O}_8(\text{OH})_6\text{SO}_4$). Schwertmannite, like ferrihydrite (ideal composition $\text{Fe}_2\text{O}_3 \times n\text{H}_2\text{O}$), has a large surface area and sorptive capacity for metal(loids). Although schwertmannite is predicted to transform to other, more stable phases such as goethite (FeOOH) or jarosite ($\text{KFe}_3(\text{OH})_6(\text{SO}_4)_2$), the transformation rate varies widely depending upon the aqueous conditions and the ions and metals adsorbed onto the surface. When mineral transformations occur, it is typically to a phase with a greater crystallinity and smaller surface area, which can release metals back into solution. Conversely, certain conditions can stabilize the phases with high capacity for sequestering metals. Although specific conditions have been documented in the literature, the fundamental geochemical controls on the rate of schwertmannite transformation and the associated fate of adsorbed metals are not well constrained. Using laboratory-synthesized phases and field samples from California and Colorado, the geochemical controls on stability of schwertmannite have been evaluated by studying the effects of aqueous sulfate, potassium, Fe(II), pH, and ionic strength. Aqueous composition, pH, conductivity, and mineralogy were monitored for 3-5 months, depending on the rate of transformation. Mineralogical changes were monitored using quantitative X-ray diffraction. Fundamental controls on the rate of transformation of schwertmannite reflect an intricate interplay among pH, ionic strength, sulfate, K, and Fe(II) concentrations. In all cases, the transformation of laboratory-synthesized schwertmannite was faster than has been observed in the field. This information is important for the development of robust conceptual and quantitative models of metal mobility. In addition, the accumulation of metals in iron oxides may make these phases potential targets for metal recovery efforts which will benefit from a fundamental understanding of the metal sorption, co-precipitation, and iron oxide stability.