

Attenuation of Hg by pyrite and other Fe/Mn sulfide minerals

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Elemental mercury (Hg^0) used in innumerable industrial processes has resulted in subsurface contamination. We studied Hg(II) sorption onto pyrite, pyrrhotite, troilite, mackinawite, and alabandite, individually and in mixtures, to assess their potential for use in permeable reactive barriers. X-ray fluorescence microprobe mapping was performed at PNC-CAT BL20 ID at the Advanced Photon Source. At locations where Hg was concentrated, XAFS spectra were collected to ascertain its coordination environment. In mixtures, Fe- and Mn- K edge XAFS spectra and two-dimensional X-ray diffraction patterns were also collected to identify the phases associated with Hg. Hg sorption onto both Fe- and Mn-sulfides demonstrates that a variety of sulfide minerals may provide Hg sequestration mechanisms. In mixtures of pyrite, troilite and pyrrhotite aged 24 hours in $\text{Hg(NO}_3)_2$ solutions, pyrite was the major Hg(II) substrate. In a mixture of pyrite+mackinawite+alabandite, all retained Hg.

Further Hg(II) sorption studies focused on pyrite exposed to HgCl_2 solutions. Hg(II) removal by pyrite increases with both pH and time. In column studies packed with quartz sand, the transport of Hg(II) was significantly retarded by the presence of a thin pyrite+sand reactive barrier. Hg L(III) edge XAFS showed that a transformation in the coordination environment at low pH occurred during aging over two weeks (Fig. 1); an ordered monolayer of monodentate Hg-Cl complexes on pyrite is consistent with the data.

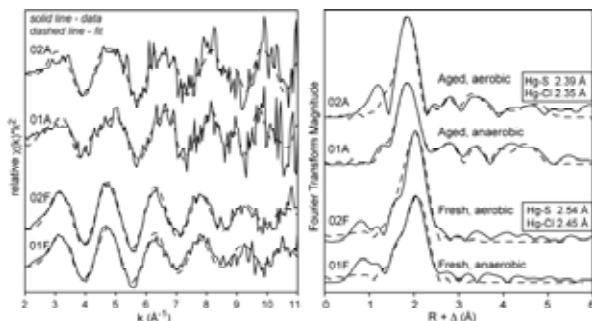


Figure 1: k^2 -weighted EXAFS spectra of Hg(II) on pyrite. Fourier transforms show shortened interatomic distances and additional backscatterers after two weeks of aging relative to samples analyzed within 24 hours.

Mobilization of Aluminum from major minerals with acidizing of surface waters

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High toxicity of dissolved aluminum is known today. The solubility of aluminum compounds has a minimum in neutral medium where the complex Al(OH)_3^0 predominates quantitatively. The solubility rises up in both alkaline ($\text{pH} > 9$) and acidic ($\text{pH} < 4$) media. With acidizing of surface waters due to either natural or anthropogenic agents the content of dissolved aluminum rises up, and consequently the negative effect increases on ecologic conditions of water objects. Rocks varying in mineral compositions are the main source of aluminum in surface waters. Therefore we have provided the experimental study of aluminum mobilization in acidic medium from major minerals and several rocks.

Six mineral phases of major alumina silicates were used in experiments (hornblende, biotite, mica, microcline, labradorite, albite), as well as the polymineral Gzhel clay and three rock samples (granite, travertine sandstone, schist). Samples were milled to powder and mixed with the water from the Moscow River in proportion 1 : 100. The pH values of solutions were controlled within the range 2.1–8.3 by addition of small amounts of 1 M sulfuric acid. Besides that the additional series of experiments was provided for lower proportions between solid phase and solution (1 : 300 to 1 : 3000). The exposure time lasted for 9 months, after that solutions were filtered through the 0.45 μm membrane filter, and equilibrium pH values and concentrations of dissolved aluminum and silica were determined.

The experiments indicate that alumina silicates dissolve incongruently in the whole range of pH values under study. This is evidenced by correlation of amounts of dissolved aluminum and silica to the corresponding amounts in solid phases. The ratios between equilibrium concentrations of aluminum and silica were lower in solutions compared to solid phases for the most of mineral phases (hornblende, biotite, mica, labradorite) and for the polymineral Gzhel clay within the whole range of pH values (2.1–8.3). Surplus removal of silica into solution under $\text{pH} > 4$, but surplus removal of aluminum into solution under $\text{pH} < 4$ was observed for microcline, albite, and rocks.

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