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Selenium repartitioning to pyrite during sulfide-driven Se-ferrihydrite transformation

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Se oxyanions strongly interact with substrates like ferric (hydr)oxides, retarding their mobility and mitigating their environmental impacts. While Se sorption on such phases is well studied, subsequent behaviors in response to post-sorption phase transformations are poorly understood. This represents a gap in the current knowledge of long-term Se retention because substrate transformations could potentially mobilize adsorbed Se. Understanding how Se partitions during secondary reactions could enhance our knowledge of Se mobility in dynamic environments.

This project investigates the behavior of Se(IV) adsorbed on ferrihydrite during sulfide-driven reductive transformation. This process likely contributes to trace element cycling in redox-intermediate environments (e.g. shallow subsurface, etc.), where ferric minerals could react with microbially generated sulfide species. Se(IV)-ferrihydrite was reacted with sulfide under anoxic conditions at Fe/S ratios of 0.1, 0.5 and 1, and at pH 7 and 9. Over 5 weeks, aliquots were extracted for solution and solid characterization.

Solution analyses results showed partial Se release over the course of the experiments. High-resolution PXRD and Mössbauer spectroscopy showed that ferrihydrite was transformed in the presence of sulfide ions to a mixture of goethite, lepidocrocite, magnetite, pyrite and elemental sulfur at pH 7. Se *K*-edge XAS revealed that adsorbed Se(IV) was first reduced to Se(0), before gradually becoming incorporated into the pyrite lattice, which was confirmed by SEM-EDXS imaging. Reactions at pH 9, in contrast, yielded a mostly amorphous iron sulfide and elemental sulfur. Se(IV) was reduced to Se(0) with no further transformation or incorporation into a secondary phase.

The results showed that while reaction with sulfide drives partial Se remobilization, secondary reaction products offer effective pathways for partial retention, limiting the extent of remobilization. This work presents the first report of Se incorporation into pyrite via the reductive transformation of a ferric oxide substrate. This is of particular interest because while pyrite has been documented as stable host for Se in sedimentary environments, the pathways leading to the lattice incorporation of Se into pyrite is not well-understood.

