

## **Redox and Sorption Behaviour of Arsenic During Co-precipitation with Fe(II,III) (hydr)oxides**

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The contamination of water supplies by toxic levels arsenic (As), a potent carcinogen, is a serious threat to public health and environmental quality. The co-precipitation of As with Fe(II,III) (hydr)oxides, such as magnetite (Mag) and green rust (GR), is a promising As remediation strategy because of the minerals' unique properties, e.g., large reactive surface area and potential for distinct As uptake mechanisms. The formation conditions of Mag and GR by Fe(0) electrolysis have been established recently. However, the redox and sorption behaviour of As during co-precipitation with Fe(II,III) (hydr)oxides must be understood before implementing their formation in remediation strategies.

In this work, we investigated the As removal efficiency and changes in As speciation during the formation of Mag, GR, and lepidocrocite (Lp) by Fe(0) electrolysis in the presence of O<sub>2</sub>. Batch As removal experiments at initial As(III) or As(V) levels from 100 to 10,000 µg/L and at pH 7.5 and 9 (3 mM total Fe) were combined with chemical extractions of the solids using PO<sub>4</sub><sup>3-</sup> and NaOH. Aqueous As in the removal experiments and extractions was measured by hydride generation-AAS and the speciation of As was determined using As K-edge X-ray absorption spectroscopy.

We found that As(III) removal ranged from 66 to 88% for GR, but was >95% for Mag and Lp. The removal of As(V) was >95% for Mag, GR and Lp in all experiments. However, Mag outperformed the other minerals, consistently leading to residual As concentrations <10 µg/L. The outstanding As(III) and As(V) removal by co-precipitated Mag generated by Fe(0) electrolysis coincided with the detection of As substituting for tetrahedral Fe in the structure of Mag by EXAFS spectroscopy, whereas As formed only binuclear surface complexes in GR and Lp samples. The extractions mobilized the least amount of As from the co-precipitated Mag samples, with preferential release of As(III) in the PO<sub>4</sub><sup>3-</sup> extraction and As(V) in the NaOH extraction. We conclude that co-precipitation with Mag is the optimum choice for Fe-based As remediation strategies, such as Fe(0) electrolysis.