

## Using siderite to remove lead from contaminated acidic (sub)surface waste waters

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Remediation of toxic heavy metals such as lead from acidic (sub)surface waste waters is a global challenge. Contaminated sites can be very complex and treatment often requires a combination of neutralization agents and metal-binding materials that immobilize hazardous aqueous species under both oxygenated and reduced conditions. Considering both these requirements, siderite ( $\text{FeCO}_3$ ) has emerged as a great potential candidate phase for the simultaneous remediation of toxic metals and acidity: the dissolution of this highly soluble carbonate in acidic environments produces alkalinity and provides highly reactive, redox sensitive ferrous iron ( $\text{Fe}^{2+}$ ) that acts as a precursor for iron (hydr)oxides, which are effective sorbents of heavy metals.

To verify this hypothesis, fine-grained natural siderite ( $<63 \mu\text{m}$ ) was dissolved in batch experiments in the presence of 100 mg/L lead ( $\text{Pb}^{2+}$ ) at pH 3.1 under oxic and anoxic conditions for up to 1008 h. As a result, 90% and 100% of the initial lead was removed from solution and the solution pH increased to 5.3 (oxic) and 6.9 (anoxic), respectively. The mechanisms driving the metal uptake differed between the two redox environments. Under oxic conditions,  $\text{Fe}^{2+}$  released during siderite dissolution oxidized to  $\text{Fe}^{3+}$  and precipitated as nanoparticulate ferric iron (hydr)oxides as soon as the pH reached  $> 4.5$ . The aqueous lead adsorbed onto these newly formed iron phases already after 2 h of reaction. Under anoxic conditions, however,  $\text{Fe}^{2+}$  oxidation was suppressed and the co-precipitation of the lead carbonate cerussite ( $\text{PbCO}_3$ ) was the mechanism responsible for the rapid and complete lead consumption after 96 h of reaction. These results and the natural abundance of siderite strongly suggest its application for *in situ* remediation purposes can be a low-cost and effective option in either oxygenated or reduced environments.