

Carbonation of sulfate wastes containing Pb: Examining the possibility of simultaneous heavy metal and carbon sequestration

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Sulfate wastes such as flue gas desulfurization (FGD) residues and phosphogypsum contain gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), which is able to capture carbon dioxide (CO_2) via carbonate mineral precipitation. Conversion of calcium sulfate minerals to calcium carbonate minerals is thermodynamically favorable due to the lower solubility of calcium carbonate minerals. However, sulfate waste streams commonly contain toxic heavy metals such as lead (Pb). This study investigated the fate of Pb during the carbonation of sulfate precipitates, providing information about the potential for dual-purpose sequestration of both CO_2 and Pb. High-resolution powder X-ray diffraction (XRD) indicated the presence of $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ and anglesite (PbSO_4) in sulfates that had been co-precipitated with Pb^{2+} . After carbonation at pH 11.5, calcite (CaCO_3) and cerussite (PbCO_3) were detected via XRD. Scanning electron microscopy (SEM) indicated a transformation in the morphology due to carbonation, from needle-like solids to cubic-like solids (Figure 1), which is consistent with calcite formation. Solids were analyzed at the tender x-ray beamline (8-BM TES) at the National Synchrotron Light Source II (NSLS-II). Micro-X-ray fluorescence (μXRF) elemental mapping revealed the spatial distributions of Ca, Pb and sulfur (S) in the experimental precipitates (Figure 2). Co-location of Ca and Pb in the precipitates from carbonation indicated the co-precipitation of Pb and Ca. This suggests that precipitation of CaCO_3 served as a host mineral to sequester Pb, possibly as a solid solution. Due to the relatively low solubility of calcium carbonate, this can effectively immobilize Pb and keep it out of the aqueous environment. Another important contribution of this research is the advancement of tender X-ray spectroscopy for analysis of Pb, which is challenging given the significant overlap in fluorescence energies for the S K-edge and Pb M-edge. We employed a unique approach of Pb M3-Edge X-ray absorption near edge structure (XANES) to tentatively identify Pb mineral phases. Overall, our study showcased the capture of Pb in an engineered CO_2 mineralization process, demonstrating that sulfate wastes contaminated with heavy metals can be safely used for the dual purpose of carbon and heavy metal sequestration.

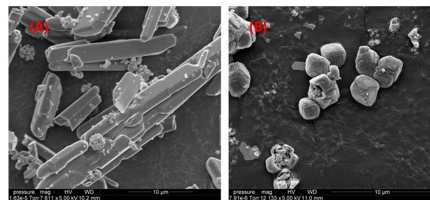


Figure 1. Scanning electron microscopy (SEM) images of (A) Pb-containing sulfate precipitates; (B) precipitates from the carbonation of sulfate precipitates.

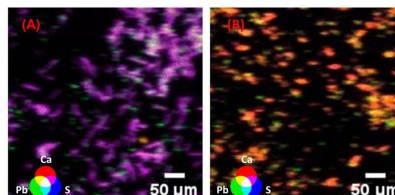


Figure 2. Micro-X-ray fluorescence (μXRF) elemental mapping of (A) Pb-containing sulfate precipitates; (B) precipitates from the carbonation of sulfate precipitates.