

# Effective Extraction of Cobalt from Carbonates *via* Sulfidation and Subsequent Oxidation

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Carbonate minerals are ubiquitous in natural environments that form through authigenic or diagenetic processes, and they are important in technologies that capture carbon through mineralization. While calcium, magnesium, and iron can serve as the primary elements for carbon mineralization, critical elements such as cobalt will often be encapsulated in their structures. Developing critical element extraction techniques that can retain carbonate structure will enable us to design processes for simultaneous critical element recovery and carbon capture. Our novel approach combines sulfidation and subsequent oxidation by hydrogen peroxide to efficiently extract cobalt while preserving the structure of  $\text{MgCO}_3$ . In this study, we used carbonate samples containing Co and Mg, with Co primarily entrapped or incorporated into the  $\text{MgCO}_3$  structure, in an Mg:Co atomic ratio of 49:1. We reacted the samples with sulfide concentrations of 5, 10, and 20 mM, at pH values ranging from 6.5 to 8.5. The treatment transformed almost all the cobalt within the carbonate matrices into amorphous sulfides, with a conversion efficiency exceeding 99%, while over 92% of the  $\text{MgCO}_3$  structure remained stable. X-ray diffraction (XRD) suggested that during the transformation from crystalline  $\text{CoCO}_3$  to cobalt sulfide, the cobalt carbonate peak broadened due to the structural change induced by sulfide. The absence of peaks corresponding to cobalt sulfide indicated that the cobalt sulfide formed was amorphous. Scanning electron microscopy (SEM)-energy dispersive X-ray spectroscopy (EDX) analysis revealed that the starting carbonate samples had a porous and hole-ridden structure with a  $\text{MgCO}_3$  shell and a  $\text{CoCO}_3$  core in mixed Mg- $\text{CoCO}_3$ . Sulfidation at pH 6.5 caused significant  $\text{MgCO}_3$  dissolution, but at pH 7.5 and 8 dissolution of  $\text{MgCO}_3$  was rarely observed. The primary extraction mechanism at higher pH condition involves sulfide diffusion into the carbonate matrix. Cobalt sulfide separation from  $\text{MgCO}_3$  was achieved using hydrogen peroxide to oxidatively dissolve cobalt sulfide into soluble sulfate. Given the low solubility of  $\text{MgCO}_3$ , this process minimally contaminated the final cobalt product, which had a purity of over 99%. Our study provides an underpinning scientific foundation for the effective extraction of cobalt from mixed phases, and it supports simultaneous critical element recovery and carbon storage.