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Why sulfide precipitates are elusive in the field: colloidal stability and aqueous Cd–S complexation upon calcium polysulfide injection

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High concentrations of heavy metals released to groundwater from mining and smelting facilities can expand plumes via advective transport, requiring effective in situ control. Calcium polysulfide (CPS) supplies sulfide (S^{2-}) that can immobilize dissolved metals by forming low-solubility metal sulfides (low K_{sp}). Yet, field evidence remains limited on the speciation of CPS-derived metal-bearing phases under realistic aquifer conditions and on their mobility under changing geochemistry. Here, we investigated the formation, stability, and colloid transport of CPS–groundwater reaction products at a CPS-injected site contaminated with cadmium and zinc, combining field monitoring with batch and column experiments.

Perforated plastic soil tubes packed with pristine aquifer material were installed in monitoring wells to allow in situ formation of CPS-derived solids and were retrieved periodically following injections. Sequential extraction and X-ray–based analyses showed that, contrary to expectations, metal hydroxide phases dominated field samples, whereas sulfide precipitates were comparatively inconspicuous. To explain this discrepancy, batch tests mimicking injection-well conditions were performed. Abundant suspended solids formed in the supernatant; sequential extraction indicated these particulates were predominantly metal sulfides, and their mass increased with CPS dose. Zeta-potential measurements of the suspended fraction (1:1 v/v CPS:groundwater) were consistently negative across pH 2–12, with increasing magnitude at higher pH, indicating a negative surface charge and strong electrostatic repulsion. This supports a DLVO-based mechanism in which repulsive interactions suppress homoaggregation and enhance colloidal stability. In addition, a separate PHREEQC calculation indicated the formation of aqueous Cd–S complexes ($CdHS^+$, $Cd(HS)_{2(aq)}$, $Cd(HS)_3^-$, $Cd(HS)_4^{2-}$, etc.) in the presence of an excessive amount of sulfide, which could suppress $CdS_{(s)}$ precipitation and thereby compromise in situ immobilization.

Mobility was further evaluated in a column continuously fed with contaminated groundwater while injecting 5% CPS at a controlled seepage velocity. Upon injection of CPS, fine sulfide particulates accumulated toward the column effluent end, consistent with advective colloid transport followed by localized retention. Together, results indicate that CPS can generate coexisting hydroxide and sulfide phases and that their stability and transport are governed by pH-dependent surface charge, DLVO-controlled aggregation, and colloid transport. Future work will numerically simulate these coupled processes under groundwater flow dynamics.