

H₂S and CO₂ sequestration in geothermal systems

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Geothermal fluids contain considerable quantities of volatile gases including H₂S and CO₂. At present, these gases are emitted to the atmosphere from most geothermal power plants causing potential environmental problems. To prevent this, injection of waster waters containing H₂S and CO₂ into the geothermal reservoirs are being considered in Iceland, were CO₂ and H₂S may mineralize into carbonates and sulfides, respectively.

Geochemical modeling, batch type reaction experiments and field studies have been conducted on the detailed geochemistry of CO₂ and H₂S under geothermal conditions (100-300°C). The purpose was twofold, to characterize the geochemistry of H₂S and CO₂ in geothermal systems characterized by basaltic rock formations and to evaluate the fluid-rock interaction in such systems including the effects of acid supply (CO₂ and H₂S), extend of reaction (mass fluxes) and temperature on the secondary mineral formation and fluid composition.

The results indicate that under natural conditions the aquifer geothermal CO₂ and H₂S concentrations are controlled by equilibrium with common secondary minerals. The buffer reactions are mostly dependent on temperature and relatively independent of extend of reaction or water-rock ratio. However, there is a very fine balance of supply of matter, composition of secondary minerals and quantity of individual secondary minerals formed. The addition of CO₂ and H₂S to the natural geothermal fluids does not change the main types of secondary minerals formed but rather the composition and quantities of various minerals at $t > 200^\circ\text{C}$. In addition, oxidation state of Fe and the geochemistry and competition of the main divalent cations, Mg²⁺, Ca²⁺ and Fe²⁺, between aluminum silicates (chlorites, zeolites, epidotes etc) and sulfides (pyrite) and carbonates (Mg, Ca and Fe carbonates) were found to play a major role in CO₂ and H₂S mineralization reactions under sub-boiling conditions.

Spectroscopic studies of Zn(II) and Cu(II) sorption to Fe-oxyhydroxide nanoparticle aggregates

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Due to their ubiquity and capacity to sequester metals, iron oxyhydroxides play a critical role in the fate and transport of potentially toxic metals in natural systems. Iron oxyhydroxides often initially precipitate as nanoparticles which quickly aggregate due to natural geochemical conditions. In order to simulate a natural environment, laboratory-synthesized nanoparticles were subjected to various geochemical conditions (increased pH, increased ionic strength, temperature aging) in order to induce aggregation prior to Zn or Cu adsorption. A desorption step to release bound metals was then introduced to mimic an acid mine drainage event and allow analysis of the relative binding strength of metal sorption sites on the iron oxyhydroxide aggregates. The specific locations of the metal binding sites were investigated using a combination of macroscopic and spectroscopic techniques including extended X-ray absorption fine structure (EXAFS) spectroscopy.

Results indicate that zinc and copper form multiple surface bound complexes of varying strengths and that binding environment is influenced by the degree and mode of aggregation. Interatomic Zn-O and Zn-Fe distances on pH- and ionic strength-aggregated iron oxyhydroxides correspond with binding as a bidentate corner-sharing complex onto 2-line ferrihydrite and a bidentate edge-sharing complex onto the [110] face of goethite. Cu-O and Cu-Fe distances suggest bidentate binding to the [110] and [021] faces of goethite under pH and ionic strength-based aggregation. After aggregation by temperature, both zinc and copper display all three sorption complexes associated with 2-line ferrihydrite and the [110] and [021] faces of goethite.

EXAFS results also indicate systematic increases in interatomic Zn-Fe distances when comparing adsorbed to retained zinc. Geometric modeling suggests straining of the zinc bidentate sorption complexes due to the distortion of the angles between the edge-sharing surface iron octahedra. This distortion may be characteristic of increased disorder and surface curvature at the nanoparticle size regime.