

Modification of hematite surfaces during colonization by a dissimilatory Fe(III) reducing bacterium under controlled hydrodynamic conditions

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The influence of biofilm development by Fe(III)-reducing bacteria on the structure and chemistry of specular hematite was evaluated under conditions that simulated saturated subsurface conditions to obtain a better understanding of biological transformations of minerals in subsurface environments. An open, hydrodynamically-controlled reactor described previously (1) was used to follow non-destructively in real time hematite surface colonization by cells of the dissimilatory iron reducing bacterium (DIRB) *Shewanella oneidensis* MR-1 when the hematite was the sole electron acceptor in the system.

Accumulation of cells on the hematite surface was accompanied by the release of soluble Fe(II) into the flowing aqueous phase when no precautions were taken to remove amorphous Fe(III) from the mineral surface before inoculation. No soluble Fe(II) was detectable in the aqueous phase during surface colonization when the mineral surface was treated with citrate-bicarbonate-dithionite (CBD) to remove amorphous Fe(III) before inoculation of the surface with cells. Hematite reduction by the surface-associated bacteria led to localized surface pitting and localized discrete areas where Fe(II) precipitation occurred. The cleavage plane of hematite left behind after bacterial Fe(III) reduction suggests that heterogeneous energetics of the mineral surface play a strong role in this bioprocess.

Dissolution, precipitation, and Fe(III) reduction in experimental systems with nontronite (NAu-1) and *Shewanella oneidensis* MR-1

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We investigated interactions between nontronite (NAu-1) and facultative dissimilatory iron reducing bacteria (DIRB), *Shewanella oneidensis*, in aqueous environments using flow-through and batch experimental systems. The evolution of major structural components of NAu-1 (i.e., Fe, Al, and Si) were monitored along with an in-depth series of control experiments. Solid phase analyses included solid Fe speciation, morphology and crystal structure using atomic force microscopy (AFM) and transmission electron microscopy (TEM), as well as elemental compositions using TEM energy dispersive X-ray spectroscopy. The effluent solution analyses indicated that $\approx 9.35\%$ of Si and 0.18% of Fe present in the original NAu-1 were transferred to and remained in solution during the seven-day flow experimental run, whereas the Al concentrations in aqueous solution remained below detection limits and batch experiments showed similar results. Up to 7 mol % of Fe in the original NAu-1 was reduced to Fe(II) and remained in solid. Control experiments showed that Si was found in solution regardless of the presence or absence of microbial Fe reduction activities. Ratios of Si, Al, and Fe in solution, released from the clay, were not stoichiometric to the clay, whereas crystal morphology observed with AFM suggested stoichiometric dissolution. Solid analysis using TEM showed the precipitation of siderite (FeCO_3) in anaerobic, reduced systems, amorphous silica associated with bacteria cells (regardless of activity) and extracellular polymeric substances, amorphous aluminosilicates in both aerobic and Fe(III)-respiring systems, and Al and Si associated with bacterial cells. Bulk thermodynamic calculations confirmed that siderite precipitation is controlling the reduced, anaerobic system. The systems were undersaturated with respect to amorphous silica, amorphous aluminum hydroxide, and halloysite except for aerobic experiments.