

Biogeochemical modification of Nontronite by *Shewanella oneidensis* MR-1: Evidence of Microbially induced Smectite-to-Illite reaction

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The biogeochemical modification of chemistry/structure of smectite associated with microbial Fe(III) respiration is a major process of promoting smectite-to-illite reaction (S-I reaction). Direct evidence of illitization including K-fixation and changes in Al/Si, formation of K-nontronite/illite-like structure has not been suggested systematically.

Nontronite (NAu-1) was inoculated with Fe-reducing bacteria (FeRB), *Shewanella oneidensis* MR-1 at 30 °C with pH buffered (7.0 and 8.0) M1 medium in the anaerobic chamber, and the evidence of illitization was suggested by microscopic/spectroscopic measurements as well as aqueous chemistry in the supernatant with various incubation time.

A progressive morphological change in bio-reduced nontronite (altered nontronite → K-nontronite → illite) corresponded to chemical modification in solid phase (Al/Si 0.16 to 0.28). Fe and Al contents in the supernatant increased continuously up to 70 days of incubation (3.4 to 20 and 1.7 to 13.20 μmol/mg of NAu-1, respectively) then decreased in 120 days of incubation (20 to 8 and 13 to 3 μmol/mg of NAu-1, respectively) indicating new mineral phase precipitated. Si contents showed slightly decreased in 7 days (133 to 100 μmol/mg of NAu-1) then showed fluctuated pattern (increased to 183 μmol/mg of NAu-1 in 70 days, then decreased to 102 μmol/mg of NAu-1 in 120 days of incubation). Formation of biotic silica globule within 120-day incubation supported the dissolution of bio-reduced nontronite. Indeed, modification in structure (appearance of 10-Å shoulder in X-ray diffraction profile) and formation of discrete illite-like packet ($d_{001}=1.0$ nm) in the wavy bio-reduced nontronite matrix ($d_{001}=1.2-1.3$ nm) strongly suggest that bio-reduced nontronite underwent the reductive dissolution and precipitated the newly formed illite.