Biogeochemical dissolution of nontronite by Shewanella Oneidensis MR-1: Evidence of biotic illite formation

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Microbial Fe-reduction in smectite structure plays a significant role in illitization accompanying with the structural/chemical modification of smectite, closely linked to the physico-chemical properties of clays, Fe-liberation, water chemistry, elemental cycles, and fault behavior. Biotic dissolution of smectite is a major process that promotes illitization, however direct evidence of illite formation is not clearly understood. In the present study, a combination of spectroscopic, microscopic, and chemical analyses revealed evidence of illitization in bio-reduced smectite, with elemental composition measured on a nanoscale as incubation time increased. Fe-reducing bacteria (FeRB), *Shewanella Oneidensis* MR-1 was inoculated in M1 medium with nontronite (NAu-1) less than 0.2 μm as an electron acceptor and Na-lactate as a sole electron donor at 30°C in the anaerobic chamber for up to 120 days. The alkalinity was maintained at pH 8.0 in the whole experiment to enhance illite formation. The extent of Fe(III) reduction measured by 1,10-phenanthroline assay reached up to 10.6% in the experiment while less than ~1% of reduction was measured in no-bacteria control. In biotic and abiotic control, increases of elemental concentrations (Si, Al, and Fe) in the supernatant indicated the dissolution of nontronite. The progress of bio-reduced nontronite reaction can be explained as follows: altered nontronite (AN) with a scouring surface texture → K-nontronite (KN) with frayed edges → euhedral lath shaped illite. A progressive morphology change in bio-reduced nontronite corresponded to an increase in Al/Si and K/(K+2Ca) that ranged between 0.13 to 0.28 and 0.16 to 1.0, suggesting the biotic reductive dissolution of nontronite and neoformation of illite. The precipitation of biotic amorphous silica supported the reductive dissolution of nontronite. In contrast, there was no clear evidence of mineral precipitation in no-bacteria control. Following treatment with Li and ethylene glycol for the long-term incubation (70 and 120 days), the X-ray diffraction profiles confirmed illitization by displaying a 10-Å peak shoulder at around 8.9° 2θ in the bio-reduced nontronite. Indeed, a direct microscopic observation of distinct illite packets of 16 nm in thickness with d_{001} = 1.0 nm in the wavy nontronite matrix with various spacings (d_{001} = 1.2 – 1.3 nm) strongly suggested biotic illite formation.