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Research paper

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

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ABSTRACT

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 and 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 (XRD) 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.

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1. Introduction

The smectite-to-illite (S-I) reaction is an ubiquitous process in siliciclastic sedimentary environments, for example the Gulf Coast basin (Eberl and Hower, 1976; Hower et al., 1976; Ahn and Peacor, 1989; Freed and Peacor, 1989, 1992; Lynch et al., 1997; Ho et al., 1999), the San Joaquin basin (Ramseyer and Boles, 1986), the Denver basin (Elliot et al., 1991), and the Nankai Trough (Masuda et al., 2001). The S-I reaction is of great interest to mineralogists and engineers because it is strongly linked with petrophysical property changes in soil (Tolhurst et al., 2002), hydrocarbon maturation (Weaver, 1960; Burst, 1969; Perry and Hower, 1970), decomposition of mineral (Perry and Hower, 1970; Weaver et al., 1971), permeability and porosity changes in shale (Boles and Franks, 1979; Bjørkum and Nadeau, 1998; Brown et al., 2001), and illite formation as a fault lubricant (Van der Pluijm, 2011). Current researches exploring microbial diversity in the Nankai Trough fault (Inagaki et al., 2015) and the influence of illitization on fault behavior (Van der Pluijm, 2011) emphasize the significance of the biotic S-I reaction.

The role of microbes in the S-I reaction at ambient conditions is widely studied in current researches (Kostka et al., 1996, 1999; Dong et al., 2003; Kim et al., 2003, 2004; Zhang et al., 2007a, 2007b; Dong et al., 2009; Jaisi et al., 2011; Stucki, 2011; Koo et al., 2014) since the capability of microbial Fe-reduction has been reported to result in the dissolution of smectite (Dong et al., 2003) and the collapse of layer spacing with K-fixation termed "illitization" (Kim et al., 2004). The consequences of microbial S-I reaction often results in Fe-cycling (Koo et al., 2014), changes in cation exchange capacity and swelling property (Stucki et al., 1984; Gates et al., 1993; Kostka et al., 1999; Stucki and Kostka, 2006), and modification in the mobility of Tc (Jaisi et al., 2008; Yang et al., 2012) and U (Komlos et al., 2008) coupling with a redox reaction.

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Evidence of biotic illite formation in mesophilic (Kim et al., 2004; Koo et al., 2014) and thermophilic (Zhang et al., 2007b; Kashefi et al., 2008; Jaisi et al., 2011) conditions has been reported through direct observation of collapsed spacings (10 Å) in lattice fringes (Kim et al., 2004; Zhang et al., 2007b), euhedral lath shaped illite (Dong et al., 2003), chemical shift of THz profiles (Koo et al., 2014), and progressive modification in the elemental composition of Al/Si for the collapsed packets (Koo et al., 2014). The secondary phase mineral precipitation of vivianite ($Fe_3P_2O_8 \cdot 8H_2O$; Dong et al., 2003, 2009; Koo et al., 2014), siderite ($FeCO_3$; Kim et al., 2004; O'Reilly et al., 2005; Zhang et al., 2007b), and amorphous silica globules (Dong et al., 2003; Li et al., 2004; O'Reilly et al., 2005; Furukawa and O'Reilly, 2007; Zhang et al., 2007b) provide evidence of biotic reductive dissolution of nontronite. The release of Fe and Si affects elemental cycles at various redox conditions. For example, Fe- and Si-liberation was facilitated by microbial Fe-respiration in deep-sea marine sediments (Vorhies and Gaines, 2009) and is a major source of Fe-fertilization of phytoplankton in the Antarctic Ocean (Martínez-García et al., 2014). Furthermore, the structural and chemical reversibility of microbially mediated collapsed layers upon redox cycles indicates that the amount of permanently fixed K in the interlayer and residual structural Fe(II) increases consequently inducing the transformation of smectite to illite (Stucki, 2011). However, there is little attention of monitoring the progress of illitization and chemical modifications in aqueous and solid phases with increasing incubation time. The present study demonstrates evidence of biotic illitization during the reductive dissolution of nontronite (NAu-1) associated with microbial Fe-respiration, by the microscopic/spectroscopic measurements of progressive modification in morphology, structure, and elemental composition of bio-reduced nontronite as well as aqueous chemistry in the supernatant.

2. Materials and methods

2.1. Bio-reduction of nontronite

Fe-reducing bacterium, *Shewanella oneidensis* (*S. oneidensis*) MR-1 was cultured aerobically in a Luria-Bertani (LB) liquid medium at pH 7 for 24 h. *S. oneidensis* MR-1 was then subcultured in a LB liquid medium at pH 8 for 24 h in order to increase cell concentration and microbial activity. The bulk sample of NAu-1 ($M^+_{1.05}[Si_{6.98}Al_{1.02}][A1_{0.29}Fe_{3.68}Mg_{0.04}]O_{20}(OH)_4$, where M is interlayer cation Ca, Na or K (Keeling et al., 2000)) was purchased from the Source Clays Repository of the Clay Mineral Society. NAu-1 with a size fraction less than 0.2 μm was collected from a water column by Stokes' Law and then freeze-dried following centrifugation. Finally, the 3×10^8 CFU/ml of microbes were inoculated in the degassed M1 medium with 20 mM of Na-lactate as a sole electron donor with Fe-rich NAu-1 as an electron acceptor (final clay concentration = 3 mg/ml) up to 120 days of incubation time. The final composition of the M1 medium contained the following elements: 9 mM $(NH_4)_2SO_4$, 5.7 mM K_2HPO_4, 3.3 mM KH_2PO_4, 2 mM $NaHCO_3$, 1.01 mM $MgSO_4$, 0.485 mM $CaCl_2$, 67.2 μM Na_2EDTA, 56.6 μM H_3BO_3, 10 μM NaCl, 5.4 μM $FeSO_4$, 5 μM $CoSO_4$, 5 μM $Ni(NH_4)_2(SO_4)_2$, 3.87 μM Na_2MoO_4, 1.5 μM Na_2SeO_4, 1.26 μM $MnSO_4$, 1.04 μM $ZnSO_4$, 0.2 μM $CuSO_4$, 20 mg/L arginine, 20 mg/L glutamate, and 20 mg/L serine (Myers and Nealson, 1988). Control samples were prepared identical to the experiments except bacterial inoculation. Samples were then gently shaken by hand twice a day and incubated in the dark box in anaerobic chamber at 30 °C. The reaction was stopped at 0, 2, 7, 70, and 120 days of the incubation by freezing the samples in liquid nitrogen and then the samples were stored at −80 °C in a deep freezer to prevent further microbial reactions.

2.2. Extent of Fe(III) reduction

The extent of Fe(III) reduction in nontronite was measured with 1,10-phenanthroline assay (Stucki, 1981) by HF and H_2SO_4 extraction method. Briefly, ~5 mg of nontronite was re-suspended in the degassed distilled water and then the solutions of 3.6 N H_2SO_4, 48% HF, and 10% 1,10-phenanthroline-ethanol were applied in Teflon jar. The solution was heated in boiling water for 30 min to extract structural Fe. Following the boiling process, 5% boric acid was added to stabilize the residual acid after the extraction (Anastácio et al., 2008). The prepared sample was placed in a black paper box to prevent photochemical reduction of the 1,10-phenanthroline-Fe(III) complex (Stucki, 1981). Two aliquots from the prepared sample were used to measure Fe(II) and total Fe. The Fe(II) concentration was calculated using Beer's law with measured absorbance of the Fe(II)-1,10-phenanthroline complex at $\lambda = 510$ nm using UV–Visible spectroscopy (HARC DR4000UV). Total amount of Fe was determined by measuring total Fe(II) after all Fe in the solution was reduced completely using hydroxylamine. The extent of Fe(III) reduction was then calculated (Fe(II)/total Fe).

2.3. Water chemistry analysis

A 2-ml aliquot of the incubated sample was centrifuged at 14,000 rpm for 5 min to separate the solution. A nontronite pellet was also washed with the degassed deionized water (2 ml) and dispersed in a sonicator to collect the absorbed cations on the nontronite particles. The solution was then collected by centrifugation. A total concentration of Fe, Al, and Si was measured using inductively coupled plasma-atomic emission spectroscopy (ICP-AES) at Korea Basic Science Institute. The concentration of elemental composition was normalized with mass of NAu-1.

2.4. X-ray diffraction (XRD)

XRD profiles were obtained using a Rigaku MiniFlex(II) automated diffractometer utilizing Cu-Kα radiation for oriented air-dried, ethylene-glycolated and Li-saturated samples. The Li-saturation was achieved by washing the samples with a degassed solution of 1 M LiCl 3 times and was used to confirm the permanent fixation of K. The samples were washed again with the degassed distilled water to remove the residual ions (Koo et al., 2014). The washing process was repeated until the supernatant became clear when $AgNO_3$ was added to the solution. The sample preparation process was performed in the anaerobic chamber to minimize the possible re-oxidation. The XRD data were recorded at a scan speed of 1.5°/minute with 0.02° 2θ steps from 2 to 11° 2θ.

2.5. Scanning and transmission electron microscopy (SEM and TEM)

The samples were re-suspended in the degassed distilled water and then 0.5 ml of the solution was pipetted onto a slide glass. The surface of a slide glass was rinsed with the degassed distilled water until particles were separated (Dong et al., 2003). Samples were air-dried in the anaerobic chamber and coated with Au in a vacuum coater for 100 s prior to observation to prevent re-oxidation. A JEOL JSM-5610LV equipped with the OXFORD INCAx-act model 51-ADD0021 was operated at an accelerating voltage of 20 kV and a working distance of 19 mm. The TEM samples were impregnated with LR White resin in order to prevent layer collapse of the clay structure due to the high-energy electron beam during TEM analysis (Kim et al., 1995). The resin cured sample at 60°C for 48 h, was sliced to a thickness of 70 nm using an ultramicrotome (ULTRACUT UCT, Leica, installed at Eulji University, Korea) and then placed on a holey-carbon TEM grid. The bright-filed TEM images, selected area electron diffraction (SAED) patterns and energy dispersive X-ray spectroscopy (EDS) data were measured using a JEOL JEM-ARM200F equipped with EDS (INCA Energy TEM for JEM-ARM200F) operated at a voltage of 200 kV at Yonsei University, Song-do, Korea.

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