Natural analogue study on the stability of smectite under hyper-alkaline conditions - Exploration of active sites at Narra in Palawan Island, Philippines -

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Smectite will play an important role in engineered barriers for the geological disposal of radioactive wastes due to its low permeability that contributes to the isolation of radioactive nuclides, and its high cation exchange capacity that retards the migration of radioactive nuclides. However, it is predicted that hyper-alkaline fluids from cementitious components of the barrier could affect the smectite, resulting in the loss of its favorable properties as a barrier component. Although many experiments have been conducted to assess the stability of smectite under hyper-alkaline conditions, there is a significant disparity between time scales involved in laboratory experiments and actual disposal environments. Natural analogue studies to assess the stability of smectite under hyper-alkaline conditions may be used to bridge these differences in time-scales. In particular, active sites, where the reaction between smectite and hyper-alkaline fluids can be presently observed, may lead to the better understanding of the geological time-scales of the reaction and to the more precise assessment of the long-term stability of smectite under hyper-alkaline conditions. However, natural analogue studies at active sites are a few. Thus, the objectives of this study are to search for suitable active sites, and to understand the interaction between smectite and hyper-alkaline fluids.

We excavated trenches at Narra in Palawan, Philippines on the contact between ultramafic rocks and recent sediments. Fluids analysis of groundwater from trench sites revealed that hyper-alkaline water (pH>11) generated by serpentinization flows in the sites. XRD analysis of solid samples from trenches revealed the presence of trioctahedral Fe-smectite. SEM observation and thermodynamic modeling suggest that these phases were generated as the result of the alteration of primary ultramafic minerals by hyper-alkaline fluids. SEM observation of samples from the layer with the Fe-smectite also showed the presence of CSH phases that precipitate from hyper-alkaline fluids. These provide the evidence that hyper-alkaline fluids actively interact with the sediments overlying the ultramafic rocks. Therefore, Narra is surely the site where the interaction between smectite and hyper-alkaline fluids can be actively observed in nature. The reactions observed in this site can thus be considered as the analogues of the reactions that might occur at actual disposal sites where Fe could be supplied from the engineered barrier, wastes or microbial activities, and the Fe-smectite might be produced by hyper-alkaline fluids. Radiocarbon dating of organic material, from the layer of calcite which is the precipitation from surface water overlying the the Fe-smectite-bearing layer suggest these organic materials are formed about 4500 years ago. This implies that hyper-alkaline fluids circulation in the site may have begun approximately 4500 years ago. Therefore, this study implies that the Fe-smectite could be formed at actual disposal sites and it requires a relatively short time.

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Keywords: Natural analogue study, Fe-smectite, geological disposal, active site, hyper-alkaline fluids