

Interaction at the interface between compacted bentonite and carbon steel plate in artificial seawater

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Geological disposal of high-level radioactive waste is definitely based on multi-barrier system. The barrier system will be composed many kinds of materials such as borosilicate glass, stainless steel, carbon steel, bentonite, rock and so on. Whereas previous studies have aimed to understand respective longevity and alteration of materials, understanding of the interaction between the constituent materials in multi-barrier system is crucial for long-term safety assessments of the disposal. In this context, iron-bentonite interaction has been studied to understand alteration of bentonite by interaction with iron materials. According to some of the previous studies, montmorillonite, main constituent mineral of bentonite was changed to non-swelling clay minerals such as berthierine, iron-serpentine. Of course, this mineralogical change was clearly one of results due to the iron-bentonite interaction. However, it is not available to be assumed berthierinization for mineralogical evolution at the interface because this alteration was observed in the experiment in the system by using deionized water, powder bentonite and powder or fibrous iron to accelerate reaction, which is completely different from real situation at the interface in multi-barrier system.

This study focuses on interaction at the interface between compacted bentonite and carbon steel plate in artificial seawater to observe the alteration of minerals in the bentonite and the extent of alteration at the interface. Interface of the sample immersed in artificial seawater for three years at 80°C under anaerobic condition was target for analyses by XRD, SEM-EDS, TEM-EDS and EPMA.

Color of the bentonite contacted with carbon steel plate was change from gray to black during the immersion in artificial seawater. Therefore, it can be easily recognized the area occurring some chemical reactions between two materials. In this area, migration of constituent element in the iron plate such as Fe and Mn into the bentonite was observed by SEM-EDS and EPMA. However, non-swelling clay minerals such as serpentine, kaolinite and chlorite were not observed based on XRD analysis. Montmorillonite was still survived in the altered area, even though small change of 060 reflection in the XRD pattern was observed, indicating change in octahedral chemistry of the montmorillonite. On the contrary, several metal carbonates such as siderite, calcite and rhodochrosite were observed in the altered area. From SEM-EDS analysis, dissolved iron reached 700 mm away from the carbon steel plate. According to iron depth profile from the line analysis, the apparent diffusion coefficient of Fe was estimated to be $3.59 \times 10^{-16} \text{ m}^2/\text{s}$, which was clearly smaller than that obtained in previous studies for compacted bentonite (Arima et al., 2004 and Kozaki et al., 1995). Observations by electron microscopes showed formations of solid solution carbonate phases consisting Fe, Mn, Mg, Ca and Cr in the area around 100 mm away from the carbon steel plate and Fe-rich montmorillonite. The smaller apparent diffusion coefficient may be attributed to the secondary Fe-carbonate formations at the interface. Observations by electron microscopes also showed Cl ion concentrated area just at the interface. Thus, the reaction and mineralogical evolution at the interface between compacted bentonite and carbon steel plate in artificial sea water were completely different from the previous studies. Further studies including geochemical reaction and transport modeling are necessary to reproduce the above observed phenomena and to understand why there is difference in different experimental systems.

References

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