Comparative study of diffusion of HDO in compacted Na- and Cs-montmorillonites characterized by three-dimensional X-ray tomographic imaging technique

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Compacted bentnite is considered as a reference material to be used as an engineered barrier in deep geological repository of HLW. The swelling capacity of the clay materials, that is important key function for reterdation capability of the engineered barrier, would decrease during the change in geochemical conditions. The aims of this work is to characterize the influence of the change in geochemical conditions on the three-dimensional (3D) microstructure of compacted bentonites and the diffusion properties of HDO in these samples. In this study, a comparative analysis of diffusivities and microstructures in compacted Na- and Cs-montmorillonites (mon.) as crays with a different swelling capacity at a dry density of 0.8 Mg m^{-3} were carried out by using nano-frocus X-ray CT at 360 nm voxel resolution. The high-resolution 3D images of the structure are processed to estimate tortuosity.

The diffusion tests showed effective diffusion coefficient of HDO was a half value in the Na-mon. than in the Cs-mon. under IS=0.1 M. From the Cross-sectional images (Fig. 1), Na-mon. was swelled and 'lens-like' pores were separated by isolated clay minerals. On the other hand, Cs-mon. didn't swell and kept the layer structure similar to that in the dry condition. The 3D image processings are in progress.

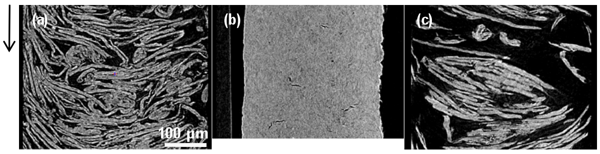

Figure 1: Cross-sectional images of compacted (a) dry Na-mon., (b) saturated Na- and (c) Cs-mon. by X-ray CT. The arrow shows the compression and diffusion direction.

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Enrichment of heavy rare earth elements on bacterial cell surfaces

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Among various microbial influences on the fate of metal ions, adsorption on bacterial cell surfaces is an initial step of interactions of metal ions and microbes. We have studied the dependence of adsorption on ionic radius of metal ions by examining all rare earth elements (except for Pm), which can be analogues of trivalent actinides (III) that are important radionuclides in radiactive wastes. In this presentation, we suggest that rare earth element (REE) pattern, often employed in geochemistry as a unique tracer, is quite useful to identify binding site at bacterial cell surfaces.

REE distribution pattern between bacteria and water exhibits anomalous enrichment in the heavy REE (HREE) part, which can act as a signature of bacteria-related materials in natural samples [1, 2]. REE binding site on the cell surface of a Gram-positive bacterium responsible for HREE enrichment has been identified using EXAFS coupled with a study of the variation in REE distribution patterns. The EXAFS data showed that the HREEs form complexes with multiple phosphate site (including phosphoester site) with a larger coordination number (CN) at lower REE-bacteria ratios ([REE]/[bac]), while light and middle REEs form complexes to the phosphate site with a lower CN. The average bond length between the REE and oxygen was compared for various REE sorbed on bacteria, showing that the bond length for HREE (Er to Lu) was much shorter than those extrapolated from the trend between La and Dy, because of the selective binding of the HREE as the multiple phosphate surface complexes. Our results are consistent with the selective enrichment of the HREE at the bacterial cell surfaces, considering that chemical species with a shorter bond length are more stable. Thus, it is clear that the HREE enrichment at the bacterial cell surfaces is caused by the formation of the multiple phosphate surface complexes. Based on these results, it is thought that materials having such phosphate sites can induce enrichment of HREE, or relatively smaller actinide (III) ions, in natural systems. Compared with other possible host phases of REEs and actnides (III), such as metal oxides and humic substances, phosphate sites are unique to bacteria and bacteria-related materials, such as biofilms and microbial mats, which can be importand adsorbents for HREE and smaller actnides (III).

[1] Y. Takahashi *et al.* (2005) *Chem. Geol.* **219**, 53–67. [2] Y. Takahashi *et al.* (2007) *Chem. Geol.* **244**, 569–583.