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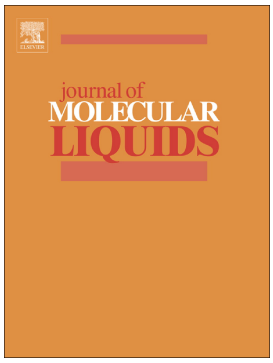

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ACCEPTED MANUSCRIPT

Sorption mechanism of Th(IV) at iron oxyhydroxide (IOHO)/water interface: Batch, model and spectroscopic studies

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Abstract

In this study, sorption mechanism of Th(IV) at iron oxyhydroxide (IOHO)/water interface was studied under ambient conditions to explore the retardation effects of IOHO on actinides in the engineering barrier system of the high-level waste radioactive waste geological repository. The sorption of Th(IV) on IOHO was strongly dependent on pH, and the intra-particle diffusion was the rate-controlling step. The presence of humic acid can enhance the sorption of Th(IV) on IOHO below pH ~3.5, whilst slightly inhibit Th(IV) sorption above pH 3.5 due to the competing effect of humic acid remaining in aqueous phase. The surface complexation model (SCM) confirmed that the bi-dentate complex of $(\equiv FeO)_2Th^{2+}$ and mono-dentate complexes of $\equiv FeOThOH^{2+}$, $\equiv FeOTh(OH)_2^{+}$ and $\equiv FeOTh(OH)_3^{0}$ are controlling the sorption process of Th(IV) on IOHO. Th(IV) sorption on IOHO is favorable to high temperature with respect to the hydrolysis effect of Th(IV). XPS analysis also confirmed the above mono- and bi-dentate surface complexes of Th(IV) on IOHO, which are very sensitive to environmental conditions. Mössbauer spectroscopy showed a decreasing geometry of each components of IOHO after Th(IV) sorption, indicating the chemisorption of Th(IV) on IOHO. Moreover, the contributions from Fe_3O_4(A) and Fe_3O_4(B) components to Th(IV) sorption were higher under high

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