

# SORPTION OF URANIUM(VI) ONTO KAOLINITE

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The sorption of uranium(VI) on kaolinite was studied as function of pH and various uranium(VI) concentrations in a system open to the atmosphere. At starting uranium(VI) concentration of  $1 \cdot 10^{-6}$  mol/l, the sorption had its sorption maximum in the pH range of about 6.0 to 8.0. At pH 7.0 and uranium(VI) concentration between  $1 \cdot 10^{-6}$  to  $5 \cdot 10^{-5}$  mol/l total uranium sorbed on kaolinite increases linearly with increasing uranium concentration.

For the safety assessment and design of nuclear waste repositories, detailed studies of the migration behavior of radionuclides are necessary. Adsorption of uranium onto clay is one of the significant reactions affecting the transport of uranium in the environment. Therefore, an atomic level understanding of sorption mechanisms of contaminants on mineral surfaces is of fundamental importance for maintaining environmental quality and assessing the long-term stability of waste repositories.

In this study kaolinite KGa-1b (Source Clays Repository) was chosen as adsorbent. The uranium adsorption on kaolinite was investigated as a function of pH. Our aim was to combine macroscopic studies (batch experiments) of uranium(VI) sorption on kaolinite with spectroscopic techniques such as extended x-ray absorption fine structure (EXAFS) spectroscopy and X-ray photoelectron spectroscopy (XPS). Such sorption studies are very useful for determining the surface complex formation constants of uranium. These formation constants are required for the geochemical speciation modeling.

Before the sorption investigations kaolinite was characterized with different analytical methods such as X-ray diffraction (XRD), infrared spectroscopy (IR), X-ray fluorescence analysis (XRF), and neutron activation analysis (NAA). The obtained results agree with the Baseline Studies [1]. Sorption measurements were carried out by batch experiments under ambient conditions. The pH range of 3.0 – 9.0 was studied with an uranium concentration of  $1 \cdot 10^{-6}$  mol/l and a solid to solution ratio of 4 g/L, using the < 100  $\mu$ m size fraction of kaolinite.

The results of the experiments (Fig. 1) show that the uptake of uranium(VI) by kaolinite increases from pH 3.0 to 5.0 and reaches its sorption maximum between pH 6.0 and 8.0. Similar results were found by Sekine, Redden and coworkers [2,3].

Uranium sorption on kaolinite was examined for total uranium from  $1 \cdot 10^{-6}$  to  $5 \cdot 10^{-5}$  mol/l at pH 7.0. In this range the uranium sorption was still significant and total uranium sorbed on kaolinite increases linearly with increasing uranium concentration. This agrees with the results of Sekine et al. [2].

After the validation of our batch experiments, we will identify the dominating uranium species at the kaolinite surface in the presence and absence of CO<sub>2</sub> as a function of pH and uranium concentration. The structure parameters of the near-neighbor surrounding of U(VI) sorbed onto kaolinite will be determined using EXAFS spectroscopy.

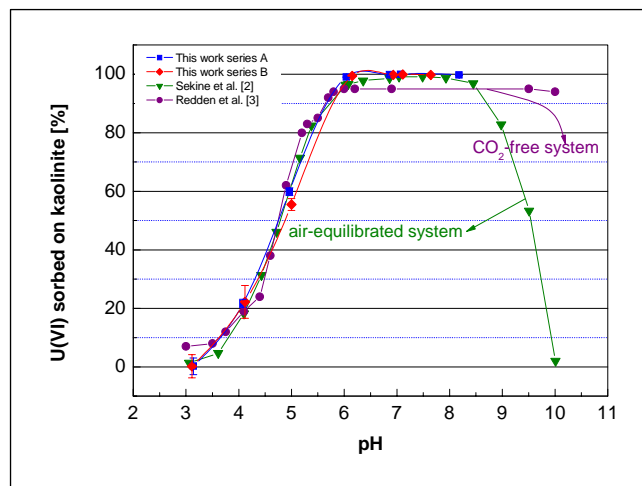


Fig. 1: Uranium(VI) adsorption on kaolinite ([U] =  $1 \cdot 10^{-6}$  mol/l, 4 g/L kaolinite, 0.1 mol/L NaClO<sub>4</sub>) compared with literature data [2,3]

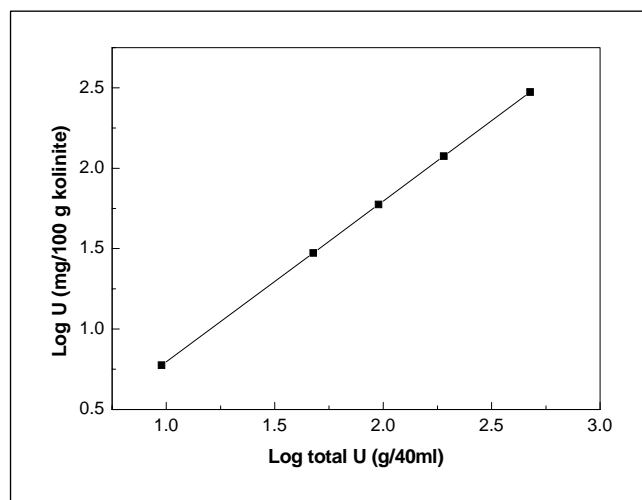


Fig. 2: Effect of total uranium(VI) adsorption on kaolinite at pH 7.0

## References

- [1] P.A. Costanzo and S. Guggenheim; Journal of clays and clay minerals, Vol. 49, No. 5, 371-452 (2001)
- [2] K. Sekine, T.E. Payne, T.D. Waite, J.A. Davis; International Alligator Rivers analogue project (18): JAERI-memo 03-036, 1-14 (1991)
- [3] G.D. Redden, J.Li, J. Leckie; In Adsorption of Metals by Geomedia (Ed. E. A. Jenne) 1998, Academic Press, p. 291-313