

Sorption of Pu(III) onto Kaolinite

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The sorption of Pu(III) and Pu(IV) onto kaolinite as a model clay mineral has been studied separately [1].

The sorption of $^{239}\text{Pu(III)}$, obtained by electrolysis and stabilised with NH_2OHHCl [2], onto kaolinite [Ka-1 Gb] was investigated in batch experiments under aerobic and anaerobic conditions as a function of pH. A pH range between 0.7 and 11 was covered.

A suspension of 4 g/L kaolinite in 0.1 M NaClO_4 was prepared and the pH was adjusted using 0.1 M HClO_4 and 0.1 M NaOH , respectively. The solution was maintained under stirring for 48 hours under the conditions later used for the sorption experiments in polyethylene vials. Meanwhile, the pH was constantly readjusted. Pu(III) was added in different concentrations ($C_{\text{Pu}} = 1 \cdot 10^{-6} \text{ M} - 1 \cdot 10^{-8} \text{ M}$). After a contact time of five days, in which the solutions were continuously stirred, the suspension was separated by means of centrifugation.

The plutonium not sorbed onto the kaolinite was quantified by means of Liquid Scintillation Counting (LSC) of the supernatant.

No significant differences were found for the sorption of Pu(III) onto kaolinite, between the various concentrations of the plutonium solutions. Also, it was observed that there are no considerable discrepancies between the sorption of Pu(III) onto kaolinite in the presence of air, and under argon atmosphere as illustrated in Fig. 1.

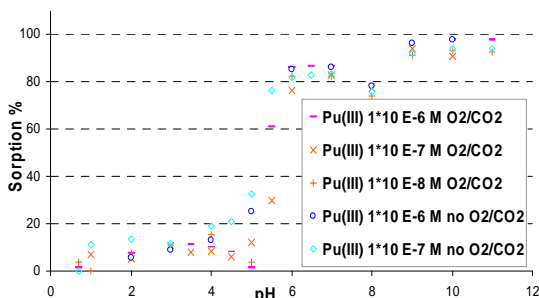


Figure 1: Sorption of Pu(III) onto kaolinite as a function of pH, under normal atmosphere and under Ar atmosphere, after the correction of the wall adsorption

Besides the sorption on kaolinite, there was observed a competitive wall adsorption. In

order to determine the wall adsorption, the vials were emptied, rinsed with Millipore water, and then a mixture of 0.36 M $\text{HF}/0.025 \text{ M HCl}$ was used to desorb the plutonium from the walls. The plutonium was then quantified with LSC. It was found that the wall adsorption was pH dependent, varying between 1 %, at pH = 1, and approximately 18 % at pH \approx 8.

The reversibility of the sorption onto kaolinite was also investigated. It could be shown that up to 10 % of the plutonium are found in a freshly prepared solution, at the same pH value, desorbed from the kaolinite. Furthermore, after an acidification of the solutions (down to pH \approx 1), almost all the plutonium is desorbed from kaolinite.

The speciation of the plutonium (^{244}Pu) sorbed on kaolinite at pH = 6 has been performed by means of Extended X-ray Absorption Fine Structure (EXAFS) [3].

For comparison, the sorption of Am(III), as an analog, onto kaolinite was investigated under similar conditions. There was a good agreement between the results for the sorption of Pu(III) and Am(III) onto kaolinite and also with the published data for the sorption of Am(III) onto mineral surfaces [4].

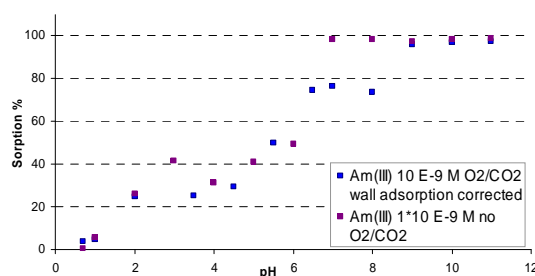


Figure 2: Sorption of Am(III) onto kaolinite

References:

- [1] N. L. Banik et al.: Annual Report, Institut für Kernchemie, Universität Mainz, **this issue** (2005)
- [2] R. A Buda et al.: Annual Report, Institut für Kernchemie, Universität Mainz, **this issue** (2005)
- [3] R. A Buda et al.: Annual Report, Institut für Kernchemie, Universität Mainz, **this issue** (2005)
- [4] L. Righetto et al, Environ. Sci. Technol., **25**, 1913, (1991)