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Investigations on stability and structure of thorium colloids

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This work presents results about the formation of thorium(IV) colloids and precipitates in the presence of silicic acid. Three methods were used for the preparation of the thorium compounds:

- Experiment A: Silicic acid concentrations and ionic strengths were adjusted in the presence of preformed stable ThO₂ colloids at neutral pH (0.4 mM Th; 0.3 mM Si; I = 0.05 M NaClO₄)
- Experiment B: Precipitation of ThO₂ colloids in the presence of silicic acid by neutralization from the acidic side of the pH scale (1 mM Th; 0-4.2 mM Si; I = 0.1 M NaClO₄)
- Experiment C: Precipitation of Th(IV) colloids in the presence of silicic acid by neutralisation of a Th(IV) carbonate solution from the alkaline side of the pH scale (1 mM Th; 0-3 mM Si; I = 0.1 M NaClO₄)

For the characterization of the colloids, zeta potential measurements as well as NMR and extended X-ray absorption fine structure (EXAFS) spectroscopy were applied.

The zeta potentials of the thorium compounds in experiments A, B and C were determined by laser Doppler velocimetry at pH values of 2–10. These measurements allowed the estimation of the isoelectric points (IEPs) of the colloids and conclusions about the colloidal stability of the suspensions formed.

By the addition of silicic acid to preformed ThO₂ colloids (experiment A), a significant shift of the IEPs from pH ~ 8.5 to lower values (pH ~ 6.5) was observed. Similar results were found in experiment B. Here, the IEP shifted from pH ~ 7.3 to pH ~ 6.0 in the presence of silicic acid. For both experiments the shift of the IEP was a function of the silicic acid concentration. Obviously, the significant amounts of silicate in/on the Th(IV) particles shift the IEP toward the IEP of pure silicic acid (pH < 3[1]).

On the other hand, the results of the zeta potential measurements on the Th(IV) colloids from experiment C demonstrated that the IEP did not change in the presence of varying amounts of silicic acid but remained constantly at pH ~ 5.2. It was also significantly lower than the IEPs of the colloids from experiments A and B formed under acidic conditions. The IEP behaviour of the experiment C colloids is not yet fully understood.

The results of ²⁹Si solid state NMR and EXAFS spectroscopy of the thorium samples from experiment C showed that silicate was built into the solid structure of ThO₂. A possible mechanism is the partial replacement of the Th-O-Th bonds by bridging silicic acid and the formation of Th-O-Si-O-Th links. It is discussed whether the integration of silicic acid in the solid structure results in the formation of a still unknown thorium compound or a thorite (ThSiO₄) like material was formed.

[1] R. K. Iler, K. Ralph, *The Chemistry of Silica: Solubility, Polymerization, Colloid and Surface Properties and Biochemistry of Silica*, John Wiley & Sons, 1979.

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