Precipitation of Aldrich Humic Acid in Dependence of pH

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Humic substances (HS) are the largest fraction of natural organic matter in environmental water sources. HS are naturally occurring materials and play an important role in the migration of radionuclides due to their complex formation. According to their solubility three fractions of HS are described: humic acid, fulvic acid, and humin. Humic acid (HA) which is soluble at pH-values ≥ 3 [1] is normally the major part of humic substances.

To predict the migration of plutonium, it is important to investigate the complexation constants log($ß_{LC}$) of HA with plutonium. This was tried for Pu(IV) with Aldrich humic acid at pH 1.8 to 3.0 [2]. Due to the precipitation of the humic acid at these pH-values, the complexation constants may be influenced by the co-precipitation of Pu with humic acid. Therefore, a detailed understanding of the kinetics of precipitation of humic acid at low pH is necessary.

For this, Aldrich humic acid (AHA) was used at concentrations of 1, 10, and 25 mg/L. The influence of the pH-value on the precipitation of AHA at 1.8, 2.5, and 3.0 was studied by UV/VIS spectrometry (Carry50) by measuring the change of concentration of AHA in solution. The pH of the solution was checked regularly (Beckmann model-310 pH meter).

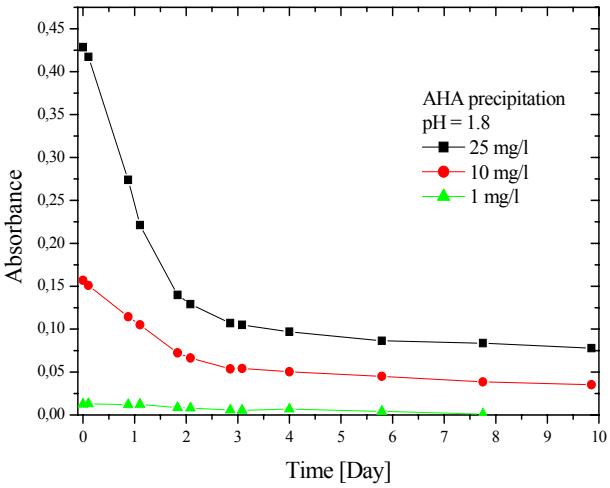

Figure 1: Precipitation of Aldrich humic acid for different concentrations (1, 10, and 25 mg/L) at pH 1.8 as a function of time

Figure 1 shows the precipitation of AHA at pH = 1.8 for three different humic acid concentrations (1, 10, and 25 mg/L). The concentration of AHA in solution decreases significantly with time. A precipitate of HA at the bottom of the cuvette is already observable by eye. A reduction of the AHA concentration in solution (original concentration 25 mg/L) of 80% at pH 1.8, 30% at pH 2.5 and almost 0% at pH 3.0 was found for the Aldrich humic acid after 240 h (see Figure 2).

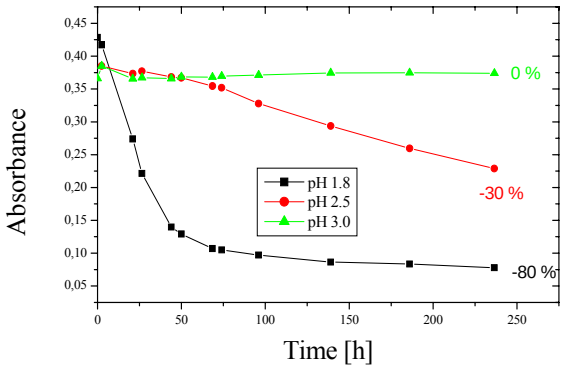

Figure 2: Decrease of AHA concentration in solution at different pH-values (1.8, 2.5, and 3.0) as a function of time (values in percent define the decrease after 10 d) for a starting AHA solution of 25 mg/L.

For the determination of the complexation constants log($ß_{LC}$) of HA with plutonium(IV), a contact time of several days is necessary to achieve equilibrium [2]. At such a long contact time the Aldrich humic acid precipitates in a significant amount and co-precipitation of Pu(IV) with HA cannot be excluded, especially for pH = 1.8 and 2.5. Further studies have to be done at pH > 3 to exclude co-precipitation, but adsorption of Pu(IV) on vessel materials or filters, depending on the method, with increasing pH must be considered.

References:

[1] G. R. Choppin, Radiochimica Acta, 44/45, 23 (1988)
[2] N. L. Banik et al., Institut für Kernchemie, Universität Mainz, Annual Report, C10 (2003)