

Influence of Humic Acid on the Sorption of Pu(III) onto Kaolinite

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The interaction of plutonium with humic substances and with kaolinite [Ka-1 Gb] as a model clay mineral was studied in the corresponding binary systems [1, 2, 3] including the sorption of Aldrich humic acid onto kaolinite as a function of pH [4].

In order to get closer to natural conditions, the ternary system Aldrich humic acid (AHA), kaolinite (K), and $^{239}\text{Pu(III)}$ was investigated..

The experiments were conducted using a fixed concentration for each component ([AHA] = 25 mg/L, [K] = 4 g/L, [Pu(III)] = 1×10^{-6} M, at room temperature, under inert gas atmosphere, and at an ionic strength of 0.1 M NaClO_4 . All experiments were performed in the presence of 0.025 M NH_2OHHCl in order to prevent the oxidation of Pu(III) to higher oxidation states. The question whether the sequence of metal ion and humic acid addition to the solid phase influences the sorption of plutonium is discussed controversially [5, 6].

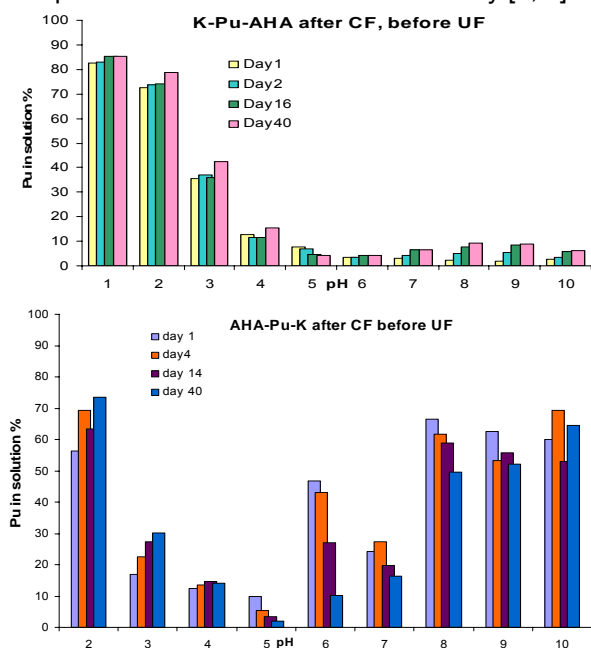


Figure 1: Sorption of Pu(III) onto kaolinite in the presence of AHA, as a function of pH and sequence of addition (A) – top, (C) – bottom (see text)

Therefore, the following four different approaches have been investigated: (A) – K mixed with Pu(III) and AHA added after 48 h, (B) – K + AHA and Pu(III) added after 48 h, (C) – AHA + Pu(III) and K after 28 days, and (D) all reactants added at the same time. The mixtures were continuously agitated, and after certain periods were centrifuged (CF) for 45

minutes at 2500 rpm. The concentration of Pu and/or AHA in the supernatant was determined by LSC and UV/VIS spectroscopy, respectively, and afterwards this fraction was ultrafiltrated (UF) through filters with pore size 1 kDalton. The content of free plutonium in the filtrate was also measured. In this way, it was possible to determine the amount of plutonium fixed on kaolinite, complexed with AHA, and of free plutonium. It was also possible to find out the amount of AHA sorbed onto kaolinite.

A significant difference was observed between the cases (A) and (C) as illustrated in Fig. 1. It is obvious that e.g., at $\text{pH} \geq 6$, the amount of Pu(III) in solution increases as a function of time in A, while it decreases in C. Within 40 days, equilibrium is obviously not attained. Furthermore, the influence of the humic acid on the sorption constant K_d of Pu(III) onto kaolinite as a function of pH and sequence of addition was studied. As illustrated in Fig.2, AHA enhances the sorption of Pu(III) onto kaolinite below pH 6 and decreases it above pH 7, depending in detail on the sequence in which the components are added.

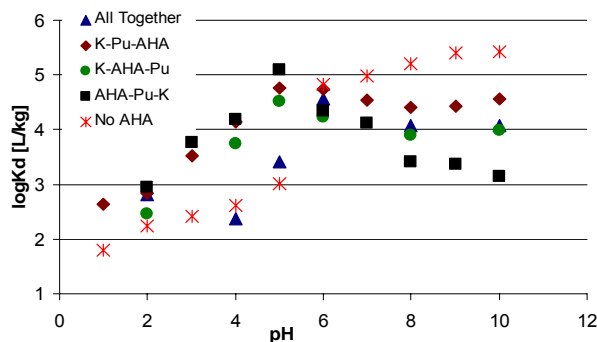


Figure 2: K_d values for the sorption of plutonium (III) onto kaolinite as a function of pH; comparison between different sequences of addition of humic acid (AHA), kaolinite (K), and plutonium

References:

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