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Sorption of U(VI) onto a decarbonated calcareous soil

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摘要

Abstract

Sorption of U(VI) from aqueous solution to decarbonated calcareous soil (DCS) was studied under ambient conditions using batch technique. Soil samples were characterized by XRD, FT-IR and SEM in detail and the effects of pH, solid-to-liquid ratio (m/V), temperature, contact time, fulvic acid (FA), CO₂ and carbonates on U(VI) sorption to calcareous soil were also studied in detail using batch technique. The results from experimental techniques showed that sorption of U(VI) on DCS was significantly influenced by pH values of the aqueous phase, indicating a formation of inner-sphere complexes at solid-liquid interface, and increased with increasing temperature, suggesting the sorption process was endothermic and spontaneous. Compared to Freundlich model, sorption of U(VI) to DCS was simulated better with Langmuir model. The sorption equilibrium could be quickly achieved within 5 h, and sorption results fitted pseudo-second-order model well. The presence of FA in sorption system enhanced U(VI) sorption at low pH and reduced U(VI) sorption at high pH values. In absence of FA, the sorption of U(VI) onto DCS was an irreversible process, while the presence of FA reinforced the U(VI) desorption process reversible. The presence of CO₂ decreased U(VI) sorption largely at pH >8, which might due to a weakly adsorbable formation of Ca₂UO₂(CO₃)₃ complex in aqueous phase.

Keywords

Soil, U(VI), Sorption, Desorption, FA, Carbonate

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Sorption of U(VI) onto a decarbonated calcareous soil

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Abstract Sorption of U(VI) from aqueous solution to decarbonated calcareous soil (DCS) was studied under ambient conditions using batch technique. Soil samples were characterized by XRD, FT-IR and SEM in detail and the effects of pH, solid-to-liquid ratio (*w/w*), temperature, contact time, fulvic acid (FA), CO₂ and carbonates on U(VI) sorption to calcareous soil were also studied in detail using batch technique. The results from experimental techniques showed that sorption of U(VI) on DCS was significantly influenced by pH values of the aqueous phase, indicating a formation of inner-sphere complexes at solid-liquid interface, and increased with increasing temperature, suggesting the sorption process was endothermic and spontaneous. Compared to Freundlich model, sorption of U(VI) to DCS was simulated better with Langmuir model. The sorption equilibrium could be quickly achieved within 5 h, and sorption results fitted pseudo-second-order model well. The presence of FA in sorption system enhanced U(VI) sorption at low pH and reduced U(VI) sorption at high pH values. In absence of FA, the sorption of U(VI) onto DCS was an irreversible process, while the presence of FA reinforced the U(VI) desorption process reversible. The presence of CO₂

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Keywords Soil · U(VI) · Sorption · Desorption · FA · Carbonate

Introduction

The transport processes of radionuclides in geological materials have been widely studied for better understanding of their physicochemical behavior in natural environment [1–5]. Uranium, as one of main radionuclides in nuclear industry, is an important contaminant in the place of uranium mining and milling, and nuclear waste disposal. Under natural conditions, uranium typically exists in hexavalent form as the mobile aqueous uranyl ion (U(VI)) [6]. Its mobility in environment is determined by several complexes and coupled processes, including oxidation-reduction, precipitation-dissolution, and sorption-desorption etc. Among these processes, sorption and desorption reactions play a critical part. In the last decade, U(VI) sorption on clay minerals, oxides and natural soils has been extensively studied. XIE et al. [7] investigated U(VI) sorption onto hematite as a function of solution pH, contact time, initial concentration, temperature, calcium and magnesium ions. Influence of calcite and dissolved calcium on uranium(VI) sorption to a Hanford subsurface sediment has been examined by DONG et al. [8], and results were obtained to indicate that formation of Ca₂UO₂(CO₃)₂(aq) has suppressed uranium sorption at pH <8.4, whereas UO₂(CO₃)₂²⁻ has been found as the dominated speciation in solution at pH >8.4. Charge distribution (CD) model was also used to evaluate uranyl sorption on ferrihydrite, which found that a

Zhang Yuying, Zhao Haogui The same contribution to this paper.

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