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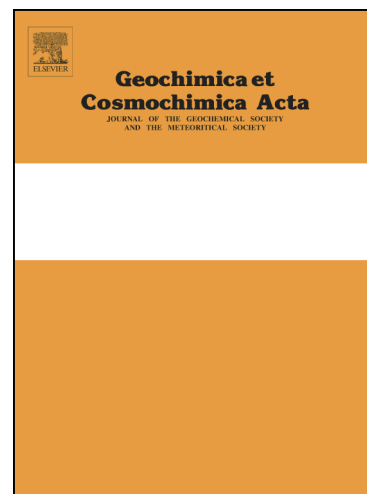
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Rates and mechanisms of uranyl oxyhydroxide mineral dissolution

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ABSTRACT

Uranyl oxyhydroxide minerals are important weathering products in uranium-contaminated surface and subsurface environments that regulate dissolved uranium concentrations. However, dissolution rates for this class of minerals and associated dissolution mechanisms have not been previously reported for circumneutral pH conditions, particularly for the case of flow through porous media. In this work, the dissolution rates of K- and Na-compreignacite ($K_2(UO_2)_6O_4(OH)_6 \cdot 8H_2O$ and $Na_2(UO_2)_6O_4(OH)_6 \cdot 8H_2O$ respectively) were measured using flow-through columns reacted with two simulated background porewater (BPW) solutions of low and high dissolved total carbonate content (ca. 0.2 and 2.8 mmol L⁻¹). Column materials were characterized before and after reaction with electron microscopy, bulk chemistry, and EXAFS to identify structural and chemical changes during dissolution and to obtain insight into molecular-scale processes. The reactive transport code CrunchFlow was used to calculate overall dissolution rates while accounting for fluid transport and changes in mineral volume and reactive surface area and results were compared to steady-state dissolution rate calculations. In low carbonate BPW systems, interlayer K and Na were initially leached from both minerals, and in Na-compreignacite, K and minor divalent cations from the input solution were incorporated into the mineral structure. Results of characterization analyses suggested that after reaction both K-

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