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A natural analogue approach to uranium-water-rock interactions: Insights from column experiments and reactive transport modeling

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Understanding uranium transport resulting from water-rock interactions is crucial for assessing groundwater contamination risks in the deep geological disposal of high-level radioactive waste (HLW). This study conducted column experiments (NA-PJ1, NA-PJ2, and NA-M) using coaly slate and fracture-filling minerals (0.025–2 mm grain size) containing uranium minerals from the Okcheon Metamorphic Belt, a natural analogue study site in Korea. NA-PJ1 contained coaly slate, while NA-PJ2 included coaly slate with 10 wt.% limestone for pH buffering. NA-M was packed with fracture-filling minerals. Artificial groundwater, manufactured to simulate that from an observation well at the study site, was purged with Ar gas to create reducing conditions. Profile sampling and tracer tests were performed to analyze uranium transport characteristics. The multi-component reactive transport model MIN3P (Mayer et al., 2002) was used to quantify groundwater flow, uranium dissolution, and various geochemical reactions. The experimental results showed low dissolved oxygen (DO) levels across all columns. The pH dropped below 4.5 in NA-PJ1 but remained around 8 in NA-PJ2. Uranium concentrations increased with distance in NA-PJ1 and NA-PJ2, with a maximum value of 67.2 μg/L due to mineral dissolution (e.g., ekanite and uraninite). Reactive transport modeling results suggested that the increase in uranium concentration with distance was primarily due to uranium complexation with carbonate and sulfate (SO_4^{2-}) in the coaly slate, forming aqueous complexes (e.g. $Ca_2UO_2(CO_3)_3$, UO_2CO_3, and UO_2SO_4). In contrast, uranium concentrations in NA-M remained at 0.1–1.1 μg/L, likely due to limited adsorption or dissolution in soil. MIN3P simulations confirmed gradual uranium increases via UO_2(s) dissolution, aligning with experimental results. These findings suggest that uranium exhibits distinct transport behaviors depending on the characteristics of the geological medium. The study provides valuable insights into uranium mobility and reactions within groundwater flow systems, contributing to the long-term safety assessment and management strategies for deep geological disposal of HLW.

[1] Multicomponent reactive transport modeling in variably saturated porous media using a generalized formulation for kinetically controlled reactions, Mayer, K. U., Frind, E. O., Blowes, D. W.(2002), Water Resources Research, 38(9).