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Mobility of uranium during weathering

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ABSTRACT

Mineralogical and geochemical mechanisms of U fixation under oxidizing conditions in the vicinity of the secondary U ore deposit at Koongarra, Australia, were examined using transmission and scanning electron microscopy and thermodynamic calculations. The formation of saléeite, $Mg(UO_2)_2(PO_4)_2 \cdot 10H_2O$, is the predominant mechanism for U fixation upstream from the deposit, where saléeite replaces sklodowskite and granular apatite. Within the deposit and further downstream, U is fixed in microcrystals (10–50 nm) of saléeite and (meta)torbernite scattered within veins of fine-grained (2–50 nm) Fe^{3+} minerals (primarily goethite and hematite). Thermodynamic calculations indicate the groundwater is undersaturated with respect to saléeite and metatorbernite and that these minerals should precipitate at higher U or P concentrations than observed. This suggests that the upstream saléeite precipitated at the reaction interfaces of dissolving sklodowskite and apatite under local saturation conditions. Observed textural relationships of saléeite and (meta)torbernite microcrystals with the Fe minerals, combined with thermodynamic calculations, suggest surface precipitation as the formation mechanism for saléeite and (meta)torbernite microcrystals within, and downstream from, the secondary ore deposit. Phosphorous released during the aging of ferrihydrite and U adsorbed onto Fe minerals are probably the sources of the major components of the microcrystals. Downstream, the microcrystals exist where groundwater U concentrations are as low as 10–30 μg/L. Once released from the ore deposit, U is fixed in uranyl phosphates even where measured groundwater is undersaturated with respect to uranyl phosphates. The surface precipitation is an important example of long-term post-adsorption U fixation in a natural system. The fully crystalline and radiation-damaged microstructures of saléeite indicate uranyl phosphates have formed continuously (or intermittently) for the last few million years.