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A time-resolved X-ray diffraction study of Cs exchange into hexagonal H-birnessite

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ABSTRACT

To measure the uptake of radioactive Cs in soils that are rich in Mn oxides, we applied time-resolved synchrotron X-ray diffraction (TR-XRD), inductively coupled plasma-mass spectrometry (ICP-MS), and analytical scanning electron microscopy (SEM) to determine the kinetics and mechanisms of aqueous Cs substitution in the phyllomanganate hexagonal H-birnessite at pH values ranging from 3 to 10. We observed that the rate of Cs cation exchange into hexagonal H-birnessite exhibited only a weak dependence on pH, but the total amount of Cs loading in the interlayer region increased dramatically above pH 6.5. The increase in Cs content at higher pH may be attributed to the increasingly negative charge on the Mn-O octahedral sheets and perhaps to a structural change toward triclinic symmetry with high pH. Dissolution at low pH may have inhibited Cs sequestration. Our work supports delamination-reassembly as a mechanism of cation exchange.

Keywords: Hexagonal birnessite, cesium, cation exchange, time-resolved X-ray diffraction