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A model surface for weathered spent nuclear fuel

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Hydrated uranium(VI) oxyhydroxides will be an important mineral group within the alteration phases formed during the corrosion of spent nuclear fuel, in the case where groundwater has entered a breach in the fuel cladding. By controlling the addition of base to an aqueous uranyl solution, we have shown that crystalline films of layer lattice uranium oxyhydroxides can be grown on silicon [1] or conductive glass under nonhydrothermal conditions. The structures of these minerals are based on sheets of uranyl polyhedra with cations distributed between the sheets or, where the sheets are electrostatically neutral, solely water exemplified here by the mineral metaschoepite.

The crystallographic textures of two films, metaschoepite and a lanthanum-intercalated phase, were studied by grazing incidence wide-angle X-ray scattering (GIWAXS) up to 200 ℃. The X-ray data are supported by SEM-EDS measurements before and after thermal evacuation, as well as thermogravimetric analysis of powders in air up to 800 ℃. The higher desorption temperatures required to remove molecular water, and the enhanced stability of the lanthanum-intercalated phase against collapse compared with metaschoepite are consistent with La(III)-water interactions and La(III)-hydrate layers that kept apart the uranyl polyhedral sheets.

With the new Radioactive Materials Beamline available at the Shanghai Synchrotron Radiation Facility, we intend to complement our X-ray diffraction data on uranium oxyhydroxide films to explore the local interlayer site structure and composition for incorporated fission products, in the frame of heat-generating high-level radioactive waste for an engineered deep geological repository.

[1] X. Gong, Q. Tian, J. Li, J. Wang, M. Yan, and M.J. Henderson, Appl. Surf. Sci. 615 (2023) 156307.

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