American Mineralogist, Volume 110, pages 1318–1328, 2025

High-pressure single-crystal X-ray diffraction and Raman spectroscopy of boltwoodite, $K_{0.63}Na_{0.37}[(UO_2)(SiO_3OH)](H_2O)_{1.5}$

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Abstract

Uranyl silicates are common constituents of the oxidized portions of uranium ore deposits. They are also known to form on used nuclear fuel pellets under simulated geologic repository conditions. These phases can incorporate various cations, including higher actinides and fission products, which makes them relevant for studies that probe the stability of these phases in various contexts, including irradiation, high temperatures, and high pressures.

The changes in the single-crystal structure and Raman spectroscopy of the uranyl silicate mineral boltwoodite, $K_{0.63}Na_{0.37}[(UO_2)(SiO_3OH)](H_2O)_{1.5}$, have been investigated at 11 pressures from ambient to 16.8 GPa. The bulk modulus was determined to be 26.3(4) GPa from a third-order Birch-Murnaghan fit of the unit-cell volume vs. pressure. No obvious phase transition was observed between ambient pressure and 16.8 GPa. The structure is most easily compressed along the *a*-axis, which decreases by ca. 15% throughout the investigated pressure range, leading to distortions in the bonding geometries and higher coordination numbers of the interlayer cations. The *b*- and *c*-axes parallel to the uranyl silicate sheets are each compressed by ca. 4% compared to the ambient-pressure structure at 16.8 GPa, attributed to the uranyl silicate sheets becoming more highly corrugated with increasing pressure.

Previous experimental high-pressure work on uranium(VI) phases is limited due to the complexity of refining heavy atom crystal structures with disordered cations and/or water molecules, as well as the limitations imposed by the opening angle of the diamond-anvil cell used to achieve high pressures. A method for overcoming these challenges is presented here to facilitate further high-pressure studies of heavy element minerals and other phases.

Keywords: Boltwoodite, uranyl mineral, high-pressure X-ray diffraction, nuclear waste