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Charge-Lattice Coupling and the Dynamic Structure of the U-O Distribution in UO 2+x

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The different structures and behaviors of UO_{2+x} observed in crystallographic and local structure measurements are examined by Extended X-ray Absorbance Fine Structure (EXAFS) measurements of pristine UO_{2.0}, p + and He₂₊ irradiated UO_{2.0}, and at multiple temperatures bulk U₄O₉ and U₃O₇ and thin film U₄O_{9-δ} on an epitaxial substrate. The disorder caused by irradiation is mostly limited to increased widths of the existing U-O/U pair distributions with any new neighbor shells being minor. As has been previously reported, the disorder caused by oxidative addition to U₄O₉

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bulk and thin film U₄O₉. This includes the significant spectral feature near R=1.2 Å for all of the U₄O₉ and U₃O₇ samples that is fit with a U-oxo type moiety with a U-O distance around 1.7 Å. In addition to indicating that these anomalies only occur in the mixed valence materials, this work confirms the continuous rearrangement of the U-O distributions from 10-250 K. Although these variations of the structure are not observed in crystallography, their prominence in the EXAFS indicates that the dynamic structure underlying these effects is an essential factor of these materials.

Keywords: UO_{2+x}, disorder, EXAFS, local structure, Radiation Effects, epitaxial films, Dynamic structure, internal tunneling polarons

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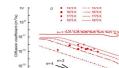
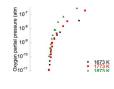
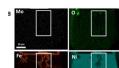
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