

Direct observation of the oxygen atoms in UO_{2+x}

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UO_2 is the principal fuel in nuclear reactors. The isometric structure also accommodates oxygen interstitials, which have important effects on chemical and physical properties. Computational studies have shown that excess oxygen atoms in UO_{2+x} occur as interstitial defects and are partially ordered [1]. However, the exact distribution of the defects has remained controversial. We present the atomic structures and bonding characteristics of a series of UO_{2+x} compositions using high-resolution (HR) aberration-corrected transmission electron microscopy (TEM) negative spherical aberration imaging (NCSI), which enables direct observation of light elements (oxygen atoms in UO_{2+x}) as compared with conventional TEM images. Stoichiometric UO_2 was synthesized from an uranyl peroxide precursor and oxidized to UO_{2+x} under a controlled Ar/ O_2 atmosphere. Subsequently, x of UO_{2+x} was determined *via* Thermogravimetric Analysis (TGA). Oxygen atoms are firstly observed at the octahedral anion sites along [100], which is consistent with the TEM image simulation results by JEMS. The role of stoichiometry (x) factor in structure of UO_{2+x} and the excess oxygen atoms' locations will be discussed.

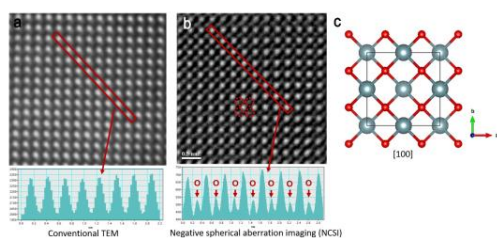


Figure 1. (a) Conventional high-resolution TEM image of a grain in the $\text{UO}_{2.00}$ sample. (b) Negative spherical aberration image obtained from the same grain in (a). The contrast profile along the red squares in (a, b) are indicated by the red arrows. The dotted red circles show the oxygen atoms located at the anion sites. (c) Structural model of $\text{UO}_{2.00}$.

[1] J. Wang, R. C. Ewing, and U. Becker, Average structure and local configuration of excess oxygen in UO_{2+x} , *Sci. Rep.* 4, 4216 (2014).