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First principles NEXAFS simulations of N-donor Uranyl complexes C.D. PEMMARAJU, Molecular Foundry, LBNL, R. DUAN, Peking University, China, R. COPPING, Chemical Sciences Division, The Glenn T. Seaborg Center, LBNL, B. JEON, Computational Research Division, LBNL, S.J. TEAT, Advanced Light Source, LBNL, M. JANOUSCH, Laboratory for Synchrotron Radiation, Paul Scherrer Institute, Switzerland, T. TYLISZCZAK, Advanced Light Source, LBNL, A. CANNING, N. GRØNBECH-JENSEN, Computational Research Division, LBNL, D.K. SHUH, Chemical Sciences Division, The Glenn T. Seaborg Center, LBNL, D. PRENDERGAST, Molecular Foundry, LBNL — The synthesis and study of soft-donor uranyl complexes can provide new insights into the coordination chemistry of non-aqueous $[\text{UO}]2^+$. Recently, the tunable N-donor ligand 2,6-Bis(2-benzimidazolyl)pyridine (BBP) was employed to produce novel uranyl complexes in which the $[\text{UO}]2^+$ cation is ligated by anionic and covalent groups with discrete chemical differences. In this work we investigate the electronic structure of the three such uranyl-BBP complexes via near-edge X-ray absorption fine structure (NEXAFS) experiments and simulations using the eXcited electron and Core-Hole (XCH) approach [1]. The evolution of the structural as well as electronic properties across the three complexes is studied systematically. Computed N K-edge and O K-edge NEXAFS spectra are compared with experiment and spectral features assigned to specific electronic transitions in these complexes. Studying the variations in spectral features arising from N K-edge absorption provides a clear picture of ligand-uranyl bonding in these systems. References: [1] D. Prendergast and G. Galli, X-ray absorption spectra of water from first-principles calculations, Phys. Rev. Lett., 215502 (2006).

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