Harnessing the nominally terminal oxo atoms of the linear uranyl (UO$_2^{2+}$) cation represents both a significant challenge and opportunity within the field of f-element hybrid materials. We have developed multiple approaches for promoting oxo atom participation in halogen-oxo and cation-cation interactions via synthetic strategies based on the judicious selection of halogen atoms and selected transition metal (TM) cations. These synthesis efforts have yielded a diverse suite of hybrid materials including uranyl molecular complexes, coordination polymers, and heterometallic hybrid materials, which have all been characterized via single crystal X-ray diffraction, Raman, Infrared (IR), and luminescence spectroscopy. Raman and IR spectroscopy results are used to generate stretching force and interaction force constants, which indicate that both halogen-oxo and cation-cation interactions weaken the U=O bond. Presented will be an overview of the routes that yield oxo participation in non-covalent interactions along with the relevant spectroscopy data that highlight our ongoing efforts to enhance structure-property delineations in this area at the frontier of uranyl crystal chemistry.