Accurate electron density distributions can be obtained from high-resolution X-ray diffraction data measured at low temperatures, even in the case of systems containing very heavy elements such as uranium. Modeling of the experimental electron density distribution with an augmented Hansen-Coppens multipolar formalism and subsequent topological analysis of the model density provides insight into the nature of U-X bonding in the electronic ground-state. The results of charge density studies measured at 20 K for systems containing uranium-halogen bonds are presented and compared to complementary results obtained from gas-phase theoretical calculations.