THEORETICAL CALCULATIONS FOR THE HIGH-LYING EXCITED LEVELS AT THE OXYGEN K-EDGE FOR $Cs_2UO_2Cl_4$

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The Soft X-ray spectrum of Denning and co-workers for the high-lying excited levels at the oxygen K-edge for $Cs_2UO_2Cl_4$ was studied theoretically. The absorption data consists of the X-ray absorption spectrum at the oxygen K-edge for $Cs_2UO_2Cl_4$ with the X-ray polarization vector aligned both parallel and perpendicular to the uranyl bond axis. We performed a detailed theoretical analysis of the K-edge absorption spectrum for both the uranyl ion and the uranyl ion in the crystalline environment of $Cs_2UO_2Cl_4$. In addition to getting insights into the wave function character of the excited state, we were also interested in studying the nature of the 1s hole in the final state. In the uranyl ion, the excitation energy was lowered by 11eV when symmetry restrictions were released on the localization of the 1s hole state. Our analysis shows that the localization of the hole is favored due to the large charge transfer between the two oxygen atoms. All calculations were performed using the COLUMBUS suite of programs. We carried out self-consistent field (SCF) calculations (without spin-orbit) and single-excitation configuration interaction (CI) calculations (with spin-orbit). Correlation-consistent polarized double-zeta basis sets were used. The treatment of complexes containing heavy atoms like uranium implies the inclusion of relativistic effects. These relativistic effects are treated through an implementation of the 68-electron relativistic effective core potentials (RECPs).