

SPECIATION OF PLUTONIUM IN WATER

JEAN-PHILIPPE BLAUDEAU, *Department of Chemistry, The Ohio State University, Columbus, OH 43210*;
STAN A. ZYGMUNT, *Chemical Technology Division/Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439*; BRUCE E. BURSTEN, *Department of Chemistry, The Ohio State University, Columbus, OH 43210*.

The speciation of plutonium in water was modeled using both ab initio quantum chemistry and density functional theory (DFT) methods. Aqueous plutonium exists in four oxidation states: +3, +4, +5, and +6. The first two forms exist as the bare ions, but the +5 and +6 states exist as $(\text{PuO}_2)^+$ and $(\text{PuO}_2)^{2+}$ species, respectively. The coordination number for the number of water molecules around each of these species is determined and compared to experiment - which uses XANES (X-ray Absorption Near-Edge Spectroscopy) and eXAFS (extended X-ray Absorption Fine Structure) spectroscopy. The DFT calculations include generalized gradient corrections and the quantum chemistry calculations are at the spin-orbit configuration interaction level.