SPECIATION OF PLUTONIUM IN WATER

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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 $(PuO_2)^+$ and $(PuO_2)^{2+}$ 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 Spectroscpy) 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.