SPECIATION OF THE PLUTONYL FORMS OF PLUTONIUM

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Plutonium ions in the 5+ and 6+ oxidation states are found in the plutonyl forms, $(PuO_2)^{+}$ and $(PuO_2)^{2+}$, respectively. The speciation of these two complexes was examined using density functional theory (DFT) methods. More specifically, we use the Amsterdam Density Functional (ADF) code, which includes relativistic effects and generalized gradient corrections. The coordination number for the number of water molecules around each of these species, as well as the bond length between the plutonium ion and the water-based oxygen atoms, are determined and compared to experiment - which uses XANES (X-ray Absorption Near-Edge Spectroscopy) and eXAFS (extended X-ray Absorption Fine Structure) spectroscopy. Furthermore, we have examined different geometrical arrangements for the water molecules in these complexes. Optimized structures with real frequencies are obtained and provide us insight into the chemical interactions involved in the solvation of plutonium species.