Interaction of Uranyl Ions and Zr-based Metal-Organic Framework by Density Functional Theory

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Abstract

Zr-based metal-organic frameworks (Zr-MOFs) have been widely used as ion absorbents for the removal or extraction of toxic and/or radionuclide species from aqueous solutions. The nature of the bonding the uranyl ion (UO_2^{2+})and Zr-MOF complexes is very important for understanding this adsorption behavior. We have optimized the structures of 19 different complexes using density functional theory; the results show that the uranyl ions can stably bond to Zr-MOF with the binding energies of the order of 2 eV. This strong adsorption is achieving by forming two U-O bonds in a configuration in which the uranyl ion attaches to one middle oxygen and one edge oxygen of Zr-MOF. In addition, we also found that the higher the degree of deprotonation of Zr-MOF, the higher binding energy between Zr-MOF and uranyl ions. This work was supported by the Center for Hierarchical Waste Form Materials (CHWM), an Energy Frontier Research Center (EFRC) funded by the United States Department of Energy Office of Basic Energy Sciences through Award DESC0016574.