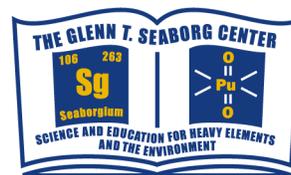




# Glenn T. Seaborg Center Seminar



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## **Relativistic Quantum Studies of Molecules Containing Actinides to Superheavy Elements**

**Wednesday, October 15, 2003**

**4 pm**

**Building 70A-3377**

We are carrying out relativistic computations that vary from complete active space multi-configuration interaction (CAS-MCSCF) followed by multi-reference configuration interaction (MRCI) computations that include up to 50 million configurations of molecules containing actinides to superheavy elements. The results of these computations have provided valuable insight into the structure, bonding and spectroscopy of actinide complexes and have revealed some unusual trends compared to their lighter analogs in the periodic table. My talk will emphasize these unusual features and trends concerning structure and properties of superheavy molecules. Investigations of actinide complexes are not only fundamentally important but are also central to the understanding of the mechanism of migration of radionuclides in high level radioactive wastes from the natural ground water to soil and forms of the actinides under aqueous environmental conditions. It seems that the migrating actinide in the natural ground water can be sorbed to geological interfaces, plants or bacteria. Sorption reactions of the actinide on these interfaces play a major role in the distribution and migration of actinides in the environment. The nature of chemical reactions, coordination chemistry, and the geometries of the sorbed actinide complexes at the mineral interfaces are poorly understood at present, and there are several conflicting interpretations and models of the observed data. Our work focuses on the U(VI) and plutonyl complexes with carbonates and silicates. An interesting case is that our computed ionization energy of uranyl conflicted with the previous experiment while a subsequent experiment by Haven and coworkers confirmed our computed result. Our computations are not only the ground electronic states, but also concern with several excited electronic states. We have computed the vibrational frequencies of many actinide complexes and also their properties in aqueous solution. We have carried out relativistic computations for the electronic states of the newly discovered superheavy elements and yet to be discovered elements such as 113 (eka-thallium) 114 (eka-lead) and  $114^+$ . Many unusual periodic trends in the energy separations of the electronic states of the elements 114, 113,  $114^+$ , (113)H, 114H, etc., and unusual features in electronic state compositions are found due to relativistic effects. It is shown that the potential energy surfaces of (114)H<sub>2</sub> exhibit unusual trends compared to PbH<sub>2</sub> due to relativity and break-down of singlet and triplet features of the electronic states due to spin-orbit coupling. Similar unusual features are noted for Lawrencium and Nobelium compounds.

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