UNDERSTANDING Pa ($5f^1 6d^1$)O^+ ($^3H_4$) IN TERMS OF THE ELECTRONIC STRUCTURE OF $Pa^{n+(n=2,3,4)}$

MICHAEL K. MROZIK, RUSSELL M. PITZER, *The Ohio State University, Columbus, OH 43210*; JOHN K. GIBSON, *Oak Ridge National Lab, Oak Ridge, TN 37831*; BRUCE E. BURSTEN, *University of Tennessee, Knoxville, TN 37996*.

Previous work by Gibson et al.[a] of Fourier transform/ion cyclotron radiation techniques/mass-spectrometry (FTICR-MS) has attempted to elucidate the nature of the bonding properties of the actinide-oxide cations, AnO^+ (An = Th, Pa, U, Np, Pu, Am, Cm). Results of these studies identified the kinetics of small alkane/alkene activation (C_1-C_4) by the AnO^+ species in the early actinides to be substantially more reactive than that of the later series counterparts, in particular the reactivity of PaO^+ is unexpectedly larger than that of the Th or U analogs. Subsequent determinations in collaboration with theoretical efforts for PaO^+ identified continued occupation of the 5f orbitals for any of the alkane/alkene activations. To further investigate the components of the seemingly simple diatomic system, the nature of the Pa^{3+} atom in a range of oxidation states ($Pa^{2+/4+}$) should help explain the perturbation presented by the oxygen atom. As in PaO^+ the determination of ground state and low-lying excited states are being completed using spin-orbit configuration interaction with single and double excitations (SO-CISD) level of theory with a *68-electron* relativistic effective core potential (RECP) and an RECP correlation consistent basis set (cc-pVDZ). Initial findings for ground state electron configurations and excitation energies are in agreement with experimentally determined values for all three $Pa^{n+(n=2,3,4)}$ atoms, and seem to indicate the large error bars associated with the atomic data reference tables could be reduced substantially. Greater understanding of the system's components should give support to previously hypothesized reasons for the drastic increase in reactivity for PaO^+.

[a] Gibson,J.K. et al. *In Press*