

Tailoring strong and weak electron correlation in actinide-containing compounds

Electronic structures of the actinide-containing molecules are significantly influenced by relativity and electron correlation effects. We elucidate the activation of the uranyl oxo bond in uranium-containing complexes¹ using relativistic Hamiltonian combined with the state-of-the-art correlated wave function models based on geminal theory and coupled-cluster theory.² Moreover, we use quantum information theory³ to dissect the process of reducing the strong U–O interactions while preserving the weak nitride uranyl bond.

In this work, we use a tailored coupled-cluster approach to account for strong and weak electron correlation effects. The external electronic structure model provides the subset of cluster amplitudes which ensures an accurate description of the multireference nature of the molecular system. Our work focuses on coupled-cluster models tailored by orbital-optimized pair coupled cluster doubles (pCCD).^{4,5} These methods scale polynomially with the system size and provide a robust way to include strong electron correlation effects.

References

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