

***Amplifying Reactivity of Metal Hydrides:
New Approaches for the Design of Multimetallic Complexes in Catalysis***

Abstract: Metal hydride complexes are common intermediates in a plethora of catalytic applications. The nature of the metal hydride within a well-defined complex is often dictated by the identity of the metal or ancillary ligand, both of which can be used to tune the properties of the hydride. Identifying new methodologies to modulate the reactivity of hydride complexes has the potential to uncover new catalytic applications. Cooperative methods such as usage of multimetallic species bearing hydride ligands has shown promise in accessing highly reactive moieties. However, access to heterometallic hydride-bearing complexes is a greater synthetic challenge. Given the challenges in accessing such systems, the reactivity and electronic structure of heterometallic hydrides are seldom studied. Our group reported a NiAl_2 heterotrimetallic hydride complex demonstrating enhanced reactivity in comparison to monometallic precursors in the hydrofunctionalization of *N*-heterocycles. Our work features three-center bonding within an Al-H-Ni subunit featuring accessible Lewis-acidic Al centers. To aid in our understanding of the electronic structure and enhanced reactivity of this system, the synthesis of related heterometallic complexes, bearing a Ni-H-Al subunit, with electronically differentiating ancillary ligands, were undertaken. Frontiers with heterometallic hydride complexes into new challenging reactivity will be discussed.