Abstract: Surface Organometallic Chemistry is an approach to synthesizing heterogeneous catalysts with molecular precision and relies on knowledge of homogeneous organometallic reactions. In our lab, we aim to install active sites on surfaces using oxidative addition of low-valent metal centers to surface functional groups. This strategy is a complementary route to the more common approaches (e.g., protonolysis of metal-ligand bonds by acidic surface sites) taken in surface organometallic chemistry. Using our approach, we developed a heterogeneous nickel-hydride catalyst that is highly active and selective for alkene isomerization. This reaction development was enabled by our work on homogeneous N-heterocyclic carbene-nickel catalysts for both alkene isomerization and hydrosilylation, which both utilize a nickel-hydride complex as the active catalyst.