Bridging the Materials and Pressure Gaps in Rh-Catalyzed Syngas Conversion

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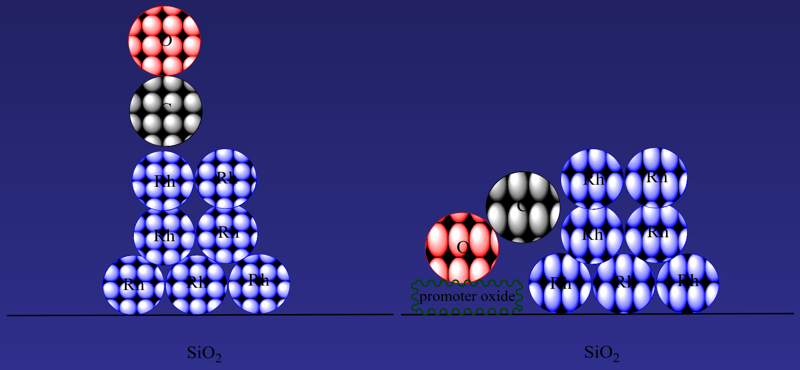
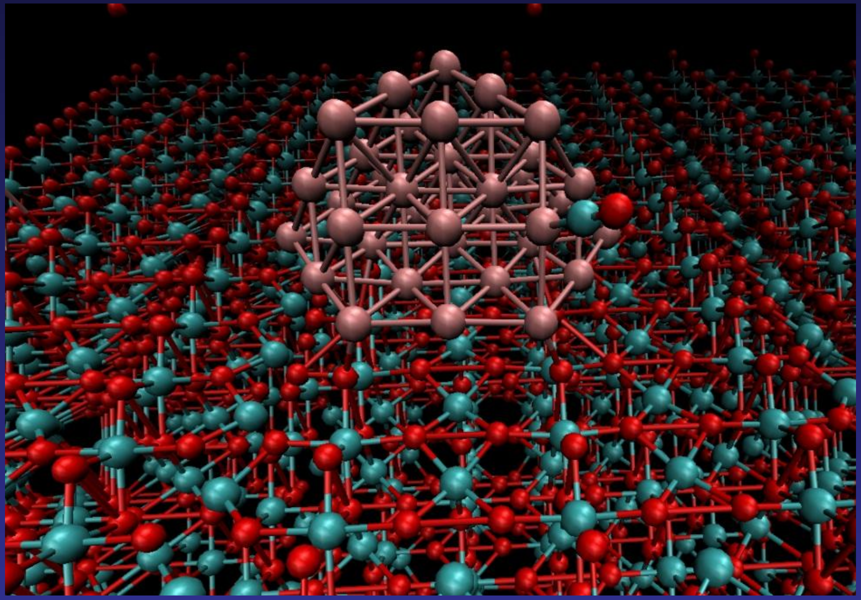
The principal objective of our work is to understand the role of high surface area oxide catalyst supports in modulating the reactivity of common metallic catalysts. These oxide supports are ubiquitous in heterogeneous catalysis, although their detailed role in modulating catalyst activity and selectivity are poorly understood. We focus specifically on the conversion of syngas (CO + H_2) to ethanol over rhodium, a reaction which exhibits strong support effects. Using computational modeling, we have developed realistic computational models for metal nanoparticles in the presence of both silica (SiO_2) and titania (TiO_2) supports (see below). We are using plane-wave density functional theory to evaluate the influence of the support in terms of

- indirect effects, modifying the geometry of the metal nanoparticle
- indirect effects, modifying the electronic structure of the supported nanoparticle
- and direct effects, catalyzing reactions on the support and/or at the metal-oxide interface

In all cases, we are contrasting the role of the support in modifying the reaction *thermodynamics* (e.g. changing adsorption energies, etc.) vs. kinetics (modifying reaction barriers). We have thus far focused on the former, and find considerable changes in adsorption energies when going from unsupported to supported nanoparticles, and between SiO_2 and TiO_2 supports.



Conceptual model of supported Rh nanoparticle with adsorbed CO



Computational model