Identifying Physical Descriptors for Metal-Support Interaction in Catalysis with Statistical Learning by Chun-Yen Liu

Abstract: The charge transfer between metals and oxide supports, known as electronic metal-support interaction (EMSI), determines the stability and reactivity of a wide range of heterogeneous catalysts. Common approaches to manipulate EMSI are placing the dopants in the oxide or adsorbates binding on the support, which alters the electronic structure of the oxide support. Given the fact that multiple charge transfers at play, the physical descriptors cannot be derived by chemical intuition alone. In this work, we use a chemically inactive oxide, MgO(100), as the support to study the impact of dopants and adsorbates on the metal binding energy. We collected the fundamental properties of each species, such as electronegativity and ionization energy, used mathematical operations to mix and combine them, and generate a pool of candidate descriptors (~ one million descriptors). Statistical learning (SL) methods, such as LASSO, Horseshoe prior, and Dirichlet-Laplace prior, are then trained against the energy computed by density functional theory (DFT) and select the physical descriptors. Among the SL algorithms, we found that the predictive model derived from Dirichlet-Laplace prior gives the lowest testing error, while these descriptors are also transferable to chemically similar oxides, such as CaO, BaO, and ZnO.