Designing Clusters for Efficient Catalytic Activity ata Realistic Condition from First-Principles Simulation

The ultimate goal of research in heterogeneous catalysis is to engineer the efficient and optimized catalyst for wide range of catalytic processes. The meaningful strategy to find suitable catalysts is to think what really limits the utility of existing catalysts. Basically, the development and rational design of catalytic materials largely depend on the ability to grasp the knowledge of targeted functionality at atomistic level. Usually, under realistic conditions catalytic materials come into contact with reactive molecules of surrounding phase. This induces the changes in local structure, composition and morphology of the catalyst. The newly formed configurations can account for the observed activity of catalyst. Moreover, it is often considered that an operating catalyst is a static entity or in equilibrium state with surrounding, though it is in dynamic nature thus situation becomes more elusive under operating conditions. Hence, complementary in situ modeling is an essential prerequisite to provide the novel insights for designing promising catalyst. The first principles methods such as density-functional theory (DFT) combined with concepts from thermodynamics have become stand tools for the accurate description of underlying factors that drive the activity of catalyst in operating conditions. Therefore, the aim of our work is to design the metal/metal-oxides nanoclusters for catalytic applications (e. g. C–H bond activation, overall water splitting, reduction and hydrogenation) and thoroughly explore their electronic and catalytic properties at finite temperature and pressure. Further, the performance of clusters are enhanced by mixing/doping different add-atoms, charge defect, changing the shape and tunning the morphology of support. The significant efforts have been dedicated for efficient designing of meta stable structures and their role in catalysts' performance. The calculations that I have carried out for designing efficient catalytic materials are: (a) clusters designing via property based cascade genetic algorithm (b) ground state geometry, electronic structure (c) stability using ab initio atomistic thermodynamics, fundamental gap using GW approximation (d) transition state and reaction pathways using the Nudge Elastic Band method.