Design of materials for energy conversion from first principles: Metallic nanoparticles of targeted shapes as highly selective catalysts and photo-catalysts

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Materials for almost all commercial heterogeneous catalytic, electro-catalytic, and photo-catalytic processes have been designed through trial and error experimental approaches. This approach to discovery has led to many commercial processes which are environmentally unfriendly, have high overall activation barriers (rendering them less energy efficient), and are limited by low selectivity, i.e., they lead to the production of undesired and environmentally harmful byproducts.

In our research group we have been developing strategies for the ‘rational’, bottom-up design of solid materials for energy-efficient and environmentally friendly chemical transformations. This approach relies on the design of materials based on understanding of underlying molecular phenomena that govern the outcome of a process rather than on empirical trial and error approaches. We are motivated by a realization that recent scientific advancements, mainly in the area of molecular science, are bringing a revolutionary transformation to the field of discovery in heterogeneous catalysts, electro-catalysis, and photo-catalysis. The landscape-changing advances driving the transformation are:

(i) Development of powerful spectroscopy and microscopy techniques, allowing us to study chemical transformations on catalytic particles with high spatial and temporal resolutions and at relevant conditions,

(ii) Development of quantum computational methodologies (for example, Density Functional Theory (DFT)), which can be utilized to study chemical transformations at the elementary step level with reasonable accuracy and efficiency. These tools are allowing us for the first time to make reasonable quantitative predictions about the outcome of elementary chemical surface reactions,

(iii) Development of novel synthetic chemistry approaches designed to synthesize targeted nano-structured materials with almost atomic precision and with a high degree of uniformity.

I will show a few examples where we used the above-mentioned advancements to design, synthesize, and test targeted nano-structures for energy-efficient and environmentally friendly catalytic and photo-catalytic chemical transformations. I will discuss partial oxidation of olefins to form epoxides over well-defined silver nano-particles of targeted shapes (spheres, wires, and cubes). Epoxides are critical gateway chemicals used in the synthesis of ethylene glycol (antifreeze), ethanolamines, and detergents. I will show how we used DFT quantum chemical calculations to identify optimal catalytic sites (these are intimately related to the shape of Ag particles). Furthermore, I will illustrate our approach to the controlled synthesis of these catalytic sites. I will also compare the performance of the novel catalysts to industrial standards. In the second example, I will show that composite photo-catalysts combing shaped metallic nano-particles of noble metals (Au or Ag) and semiconductor nanostructures (for example TiO2) exhibit significantly improved photo-chemical activity compared to conventional photo-catalytic materials. The critical feature of these composite photo-catalyst is that they couple excellent optical absorption properties of shaped metallic nanostructures (Au or Ag), manifested in the formation of surface plasmons in response to a photon flux, and photo-catalytic potential of semiconductors, therefore enabling more efficient conversion of solar flux into electron/hole pairs. By changing the size and shape of metallic nano-particles, it is possible to enhance the photo-chemical activity across the entire UV-vis region of solar spectrum. These composite systems represent a new family of materials for efficient photo-catalytic conversion of solar energy into chemical energy of solar fuels.