

Metastable Structures as Key Catalytic Sites: A Computational Perspective



Professor Philippe Sautet

Departments of Chemical and Biomolecular Engineering and Chemistry and Biochemistry

University of California, Los Angeles

Friday, September 21, 2018
3:00pm (Reception at 2:45pm)
66-110

Abstract: The determination of the structure of heterogeneous catalytic systems, under reaction conditions, is a key point for a detailed understanding of the nature of active sites and for rational design of efficient catalysts. The lecture will discuss two systems. The first one will focus on the modelling of small Pt cluster (Pt_{13}) under hydrogen pressure and on their reactivity for methane activation.¹ The approach combines Density Functional Theory, high-dimensional Neural Networks and evolutionary techniques. The bare Pt clusters show a large number of low energy isomers (> 60 in 0.5 eV). Hydrogenated clusters adopt different geometries and appear more rigid, with a smaller number of low energy isomers. These metastable isomers nevertheless play a major role for the catalytic reactivity of the hydrogen covered cluster, which cannot be described by considering the most stable structure alone. Fluxionality and accessible metastable structure are hence key characteristics for the catalytic properties of small Pt clusters. The second part will deal with single-atom catalysts, that are widely investigated heterogeneous catalysts. We will consider Rh single atoms on TiO_2 as a generic example, investigating optimal structure of the single atoms under H_2 reduction, CO adsorption and its reactivity for reverse water gas shift (RWGS) reaction. The combination of theoretical and experimental studies clearly demonstrates that Rh single atoms change their structure and adapt their catalytic site under reaction conditions.