

Electronic structure of Pt cluster and its surface chemistry

Qingguo Meng, Dmitri Kilin

Department of Chemistry, University of South Dakota, 414E Clark, CL115, Vermillion, SD, 57069, USA

Abstract

There have been lots of investigations on the Pt surface due to its importance in catalysis. The impact of H₂ pressure on the morphology and electronic properties of Pt nanoparticle surface is of great importance in the catalytic field of hydrogenation, fuel-cell technologies, and water splitting photo-catalytically. In this work, a Pt₁₃ cluster system (*O_h*), which has one Pt atom in the center and coordinated by other twelve Pt atoms, is investigated by using DFT calculation. After computing geometry optimization on the Pt₁₃ cluster [1], two nonequilibrium modifications, e.g. addition or removal of electrons and heat treatment were applied on the cluster, with the aim to simulate the experimental reaction of hydrogen on the Pt cluster surface. The computation results reveal that negative ions of Pt cluster facilitates desorption of hydrogen from the surface in a molecular form, which would be a fuel potentially [2]. The higher temperature is also beneficial to the H₂ desorption. A elementary hydrogen evolution mechanism [3] on the Pt surface can be established as $H_{abs} + H_{abs} (or H^+ + e^-) + Pt_{abs} \rightarrow H_2 + Pt$. A rate (mol/h) of hydrogen desorption and turnover (mol H₂/ mol Pt) from the system is determined, together with the hydrogen dissociative chemisorption energy. The photoactivities of the catalysts with various sized of clusters are also studied. The results computed in the current work can be compared to the experiment on Thermal Desorption Spectra (TDS) [4].

Reference

- [1]. Kresse, G.; Furthmuller, J. *Phys. Rev. B* **1996**, *54*, 11169.
- [2]. Link to YouTube video <http://youtu.be/crz8andwK40>.
- [3]. Skúlason, E.; Tripkovic, V.; Björketun, M.E.; Gudmundsdóttir, S.; Karlberg, G.; Rossmeisl, J.; Bligaard, T.; Jónsson, H.; Nørskov, J.K. *J. Phys. Chem. C* **2010**, *114*, 18182.
- [4]. J.T. Miller, B.L. Meyers, F.S. Modica, G.S. Lane, M. Vaarkamp, D.C. Koningsberger, *J. Catal.* **1993**, *143*, 395.