

Hydrogenation and oxidation over palladium clusters and surfaces

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One main application of novel cluster properties is heterogeneous catalysis. In fact, technical catalysts are generally designed by dispersion of nano and sub-nanometer sized metal particles on oxide supports. Catalytic reactions have, within the surface science paradigm often been modelled by extended low-index surfaces. One such example is the hydrogenation of ethylene which is an important model reaction for selective upgrading of biofuels and conversion of alkenes to saturated alkanes.

In the present contribution we have used density functional theory to investigate ethylene hydrogenation over palladium clusters [1] and compared the results with recent results for flat and stepped surfaces [1-3]. The comparison exemplify several novel aspects of clusters. We find that the adsorption energies are always higher for the clusters, even if the comparison is made to adsorption on stepped-surfaces in the low coverage regime. Clusters are found to accommodate considerable higher adsorbate coverages where, for example, the ethylene coverage on Pd₁₃ is one, whereas the saturation coverage on Pd(111) is 0.33. The activation energies for hydrogenation varies in a non-monotonic manner, where, for examples the barrier for the first hydrogenation step is higher on Pd₃₈ than on Pd(211).

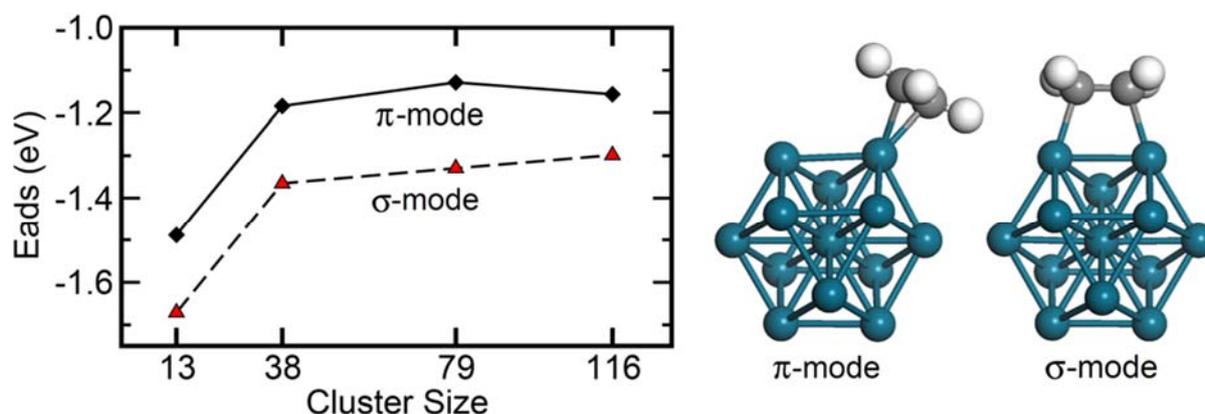


Figure 1: Ethylene adsorption energy as a function of clusters size. The two possible modes are exemplified for Pd₁₃. Atomic color code: Pd(blue), C(gray), H (white).

We have also investigated oxidation reactions over palladium, namely methane and carbon monoxide oxidation. By construction ab initio based micro-kinetic models we are able to investigate differences in reaction kinetics for different surfaces and and types of steps [4,5].

- [1] A. Posada-Borbón, C.J. Heard, H. Grönbeck, In preparation.
- [2] C.J. Heard, S. Siahrostami and H. Grönbeck, *J. Phys. Chem. C* **120**, 995 (2016).
- [3] C.J. Heard, C. Hu, M. Skoglundh, D. Creaser and H. Grönbeck, *ACS Catalysis* **6**, 3277 (2016).
- [4] M. Jorgensen and H. Grönbeck, submitted.
- [5] S. Blomberg *et al.*, submitted.