

Multiscale Simulations of Gold Catalysis

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Au has attracted considerable interest in the last two decades because, while bulk gold is a chemically inert material¹, Haruta² showed an exceptional reactivity of Au NPs of diameters ranging from 2 to 5 nm, when supported on metal oxides. The low temperature CO oxidation is one of the most extensively studied reactions on Au NPs. Despite the ongoing debate on the possible mechanisms of the CO oxidation on Au, the consensus that emerges is that only low coordinated sites of NPs^{3, 4}, such as corners and edges, are catalytically active. However, experiments showed that small Au clusters (consisting of 3 to 20 atoms, < 1 nm diameter), when supported on defective MgO support, show magic numbers on their catalytic activity towards the CO oxidation reaction.

In the present work, we use first principle calculations to develop suitable relationships, capable of predicting the adsorbate (CO) binding strength on every site of Au nanoparticles^{5, 6}. Our results unravel how the size, shape, and symmetry of nanoparticles affect their electronic properties and, consequently, their interaction with the adsorbates. Most importantly, the effects of the support (i.e. presence of defects) on the electronic and structural characteristics of gold clusters along with their effect on the CO oxidation mechanism are investigated. With the use of Multi-Scale Modelling (combining first principle calculations with Kinetic Monte Carlo simulations) we give insights into the mechanisms out of which a catalyst can remain active towards CO oxidation or poisoned by either CO or by the formation of carbonates⁷.

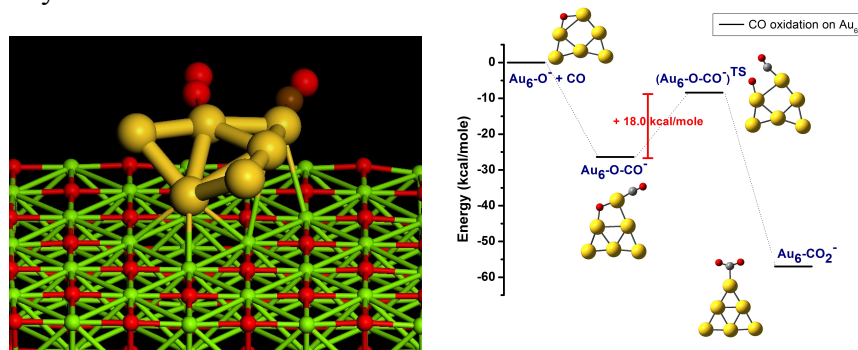


Fig. 1. MgO supported Au₆ catalyst and the second step of the CO oxidation reaction mechanism.

References

1. Hammer, B.; Norskov, J. K. *Nature* **1995**, 376, (6537), 238-240.
2. Haruta, M.; Kobayashi, T.; Sano, H.; Yamada, N. *Chemistry Letters* **1987**, (2), 405-408.
3. Hvolbaek, B.; Janssens, T. V. W.; Clausen, B. S.; Falsig, H.; Christensen, C. H.; Norskov, J. K. *Nano Today* **2007**, 2, (4), 14-18.
4. Remediakis, I. N.; Lopez, N.; Norskov, J. K. *Angewandte Chemie-International Edition* **2005**, 44, (12), 1824-1826.
5. Mpourmpakis, G.; Andriotis, A. N.; Vlachos, D. G. *Nano Letters* **2010**, 10, (3), 1041-1045.
6. Mpourmpakis, G.; Vlachos, D. G. *Journal of Physical Chemistry C* **2009**, 113, (17), 7329-7335.
7. Mpourmpakis, G.; Christiansen, M.; Vlachos, D. G. *in preparation* **2010**.

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