

First principles studies of nanoscale effects on catalytic activity and selectivity

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Computational studies can be carried out on the effect of size, composition, and support on the catalytic properties of a nanoscale clusters using density functional theory. They provide insight into the special reactivity properties exhibited by supported metal and metal oxide clusters that are not observed in their bulk analogs, which can make them attractive as new materials for catalysis.^{1,2,3} Understanding the properties of these clusters is instrumental for addressing fundamental aspects of catalysis that may lead to the development of new classes of catalytic materials with control of their bond breaking and making capabilities. Our effort, which is integrated with experimental work on synthesis, characterization, and catalytic testing, is resulting in the discovery of structure-property relationships of supported nanoscale metal clusters. While the computational studies can provide information on the structural and electronic properties of the intermediates involved in the reaction mechanisms, it is difficult to experimentally obtain information on these intermediates and, thus confirm the computational modeling. Several examples will be given of catalytic reaction mechanisms and the complexities involved.

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(1) Vajda, S.; Pellin, M. J.; Greeley, J. P.; Marshall, C. L.; Curtiss, L. A.; Ballentine, G. A.; Elam, J. W.; Catillon-Mucherie, S.; Redfern, P. C.; Mehmood, F.; Zapol, P. *Nature Materials* 2009, 8, 213-216.

(2) Kim, H.-S.; Zygmunt, S. A.; Stair, P. C.; Zapol, P.; Curtiss, L. A. *J. Phys. Chem C*, 2009, 113, 8836.

(3) Lei, Y.; Mehmood, F.; Lee, S.; Greeley, J. P.; Lee, B.; Seifert, S.; Winans, R. E.; Elam, J. W.; Meyer, R. J.; Redfern, P. C.; Teschner, D.; Schlögl, R.; Pellin, M. J.; Curtiss, L. A.; Vajda, S. *Science* 2010, 328, 224.