OXYGEN INFLUENCE ON AUN STRUCTURES: DFT STUDY

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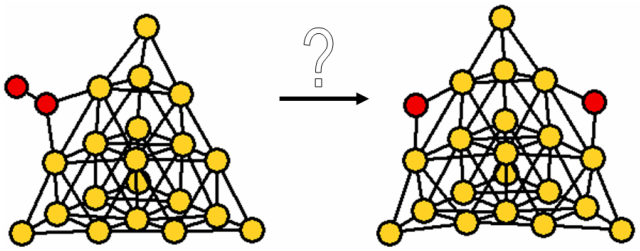
Gold nanoparticles supported on metal oxides can efficiently catalyze a variety of reactions at low temperatures associated with environmental problems: CO oxidation, NO reduction, propylene partial oxidation, the direct production of H_2O_2 from H_2 and O_2, and others [1]. Although a lot of extensive investigations focused previously on the mechanism governing Au reaction, the details of the mechanism and the origin of the catalytic activity of Au_n is not well understood so far and under debates.

The first step of any catalytical reaction is the interaction of the isolated reactants with the cluster. It is important to understand how the adsorbed molecule influences on gold cluster structure and catalytical properties of last one. Thus, the following questions arise: whether oxygen molecules can chemisorb on Au_n, and whether the chemisorption is dissociative or molecular in nature.



Au_{20}–O_2 O–Au_{20}–O

The interaction of O and O_2 with symmetrical tetrahedral Au20 cluster has been studied using the density functional theory with PBE functional and gold pseudopotential with relativistic corrections included. Oxygen atom may be bonded with apex or facet Au atom (33.7 and 15.6 kcal/mol), and with two Au atoms in edge (46.6 and 62.7 kcal/mol). It was found that the dissociative adsorption is more favorable (two Au-O-Au bonds, 34.9 kcal/mol) than molecular adsorption (Au-O-O bond, 19.3 kcal/mol) for O_2 chemisorption.

The possibility of dissociative chemisorption found is important for the catalytic reactivity of the cluster. The results of this study could provide the impetus for further experimental studied on structure of gold nanoparticles, the interaction of oxygen and hydrogen with gold clusters and their catalytic reactivity

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References

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