

Abstract Submitted
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Influence of Structure and Charge State on the Mechanism of CO Oxidation on Gold Clusters GRANT JOHNSON, Penn State University, CHRISTIAN BURGEL, Humboldt Universitat zu Berlin, NELLY REILLY, ROLAND MITRIC, Humboldt Universitat zu Berlin, MICHELE KIMBLE, ERIC TYO, A.W. CASTLEMAN, Penn State University, VLASTA BONACIC-KOUTECKY, Humboldt Univesitat zu Berlin, HUMBOLDT UNIVERSITAT ZU BERLIN COLLABORATION — Gas-phase reactivity experiments and high level theoretical calculations have been employed to study the interaction of both positively and negatively charged gold oxide clusters with carbon monoxide (CO). We demonstrate that for negatively charged clusters CO is oxidized to CO₂ by an Eley-Rideal-like (ER-) mechanism involving the attack of CO on oxygen rather than gold. In contrast, for positively charged clusters, the oxidation reaction may also occur by a Langmuir-Hinshelwood-like (LH-) mechanism involving the initial binding of CO to a gold atom followed by subsequent migration to an oxygen site. The LH mechanism is made possible through the large energy gain associated with the adsorption of two CO molecules onto cationic gold clusters. Structure-reactivity relationships are also established which demonstrate that terminally bound oxygen atoms are the most active sites for CO oxidation. Bridge bonded oxygen atoms and molecularly bound O₂ units are shown to be inert. We also establish an inverse relationship between the binding energy of CO to gold clusters and the energy of the clusters lowest unoccupied molecular orbital (LUMO).

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