Abstract:
Control and tunability of the catalytic oxidn. of CO by gold clusters deposited on MgO surfaces grown on molybdenum, Mo(100), to various thicknesses are explored through temp.-programmed reaction measurements on mass-selected 20-atom gold clusters and via first-principles d. functional theory calcns. Au20 was chosen because in the gas phase it is characterized as an extraordinarily stable tetrahedral-pyramidal structure. Dependencies of the catalytic activities and microscopic reaction mechanisms on the thickness and stoichiometry of the MgO films and on the dimensionalities and structures of the adsorbed gold clusters are demonstrated and elucidated. Langmuir-Hinshelwood mechanisms and reaction barriers corresponding to obsd. low- and high-temp. CO oxidn. reactions are calcd. and analyzed. These reactions involve adsorbed O2 mols. that are activated to a superoxo- or peroxo-like state through partial occupation of the antibonding orbitals. In some cases, we find activated, dissociative adsorption of O2 mols., adsorbing at the cluster peripheral interface with the MgO surface. The reactant CO mols. either adsorb on the MgO surface in the cluster proximity or bind directly to the gold cluster. Along with the oxidn. reactions on stoichiometric ultrathin MgO films, we also study reactions catalyzed by Au20 nanoclusters adsorbed on relatively thick defect-poor MgO films supported on Mo and on defect-rich
thick MgO surfaces contg. oxygen vacancy defects. [on SciFinder(R)]

Stichworte:
Clusters Nanoparticles (nanoclusters, Au20 control and manipulation of gold nanocatalysis: effects of metal oxide support thickness and compn.) manipulation gold nanocluster nanocatalysis metal oxide support thickness compn carbon monoxide oxidn catalyst manipulation gold nanocluster support

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