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Catalytic Thermodynamics of Nanocluster Adsorbates from Informational Statistical Mechanics

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Abstract

This research presents a new approach for studying the catalytic thermodynamics of gold, platinum, and palladium cuboctahedral nanoclusters, using informational statistical mechanics. Nanoclusters and their adatoms are viewed as chemical graphs with a nearest neighbor adjacency matrix. Oxygen radical and molecular adsorbates are studied on platinum clusters, hydrogen on palladium clusters, and carbon monoxide on gold clusters. A scaled Morse potential is used to determine bond energies between cluster atoms in a coordination type calculation. For gold clusters we also use an electronic potential that calculates partial charges of surface atoms. The partial charge of low coordinated gold sites is negative, which may promote bonding with the positive oxygen end of the dipole on the CO molecule. Applied density functional theory (DFT) calculations demonstrate adatom effects on the thermodynamic quantities, which are derived from a Hamiltonian. Calculations of the entropy, free energy, and total energy show linear behavior, as the coverage of oxygen on platinum, and hydrogen on palladium, increases from bridge sites on the surface. The data exhibits size effects for the measured thermodynamic properties with cluster diameters between 2 and 7 nm. Entropy and enthalpy calculations of Pt- O_2 compare well with previous theoretical data for Pt(111)-O_2, and trends for Pd-H are similar to experimental measurements on Pd-H_2 nanoclusters. These techniques are applicable to a wide variety of cluster-adsorbate interactions, encouraging further research.