Accelerating Catalyst Design via AI: High-Throughput Screening and Machine Learning Reveal Defect-Enhanced Activity in Pt-Au Nanoclusters

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The catalytic properties of bimetallic Pt-Au nanoclusters are governed by a complex interplay of size, composition, atomic ordering, and support interactions, making traditional trial-and-error exploration inefficient and motivating the use of advanced computational methods for accelerated discovery. In this work, we employ a multi-stage AI-driven framework combining evolutionary structure prediction, high-throughput DFT screening, and machine learning potentials to comprehensively investigate Pt-Au nanoclusters (up to 24 atoms) supported on graphene. Using the USPEX evolutionary algorithm in variable-composition mode, we performed an unbiased global search for stable cluster configurations, identifying twelve particularly stable structures where Pt atoms preferentially occupy central, highly-coordinated positions while Au segregates to the surface.

Subsequent high-throughput screening of over 12,000 DFT calculations across all non-equivalent active sites revealed that adsorption strength is primarily determined by the local atomic composition, with Pt-rich sites binding O and CO more strongly. Critically, we demonstrate that the graphene support plays an active and tunable role: while clusters are highly mobile on pristine graphene, a single carbon monovacancy serves as a potent anchoring site, profoundly modifying the cluster's electronic structure through significant charge redistribution. This defect engineering activates traditionally inert Au atoms, enhancing electron transfer to adsorbed O_2 from $0.27e^-$ to $0.4e^-$ and populating its antibonding orbitals, which leads to stronger adsorption and significantly reduced energy barriers for CO oxidation across all active sites. Furthermore, large-scale molecular dynamics simulations using a Moment Tensor Potential (MTP) trained on actively learned DFT data revealed an adsorption-induced structural transition under CO saturation: Pt atoms migrate from the core to the surface, becoming the preferred adsorption sites and dynamically optimizing the cluster's catalytic function in a temperature- and concentration-dependent manner.

Our integrated computational framework demonstrates that the rational design of high-performance bimetallic catalysts must explicitly account for the intrinsic, composition-dependent atomic structure, the active electronic synergy with support defects, and the dynamic restructuring of nanoparticles under operating conditions, providing new mechanistic insights and design principles for low-temperature CO oxidation catalysts.