Machine-learning-assisted prediction of catalytic activity of alloy spherical nanoparticles for the hydrogen evolution reaction

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Alloy nanoparticles are an important class of materials used for catalyzing the hydrogen evolution reaction (HER). However, the complexity of predicting the catalytic behavior of alloy nanoparticles at experimental sizes remains a major challenge in the search for effective catalysts. To accelerate the design of effective catalysts and evaluate their HER activity, we developed an approach to predict the binding free energies of active sites on alloy spherical nanoparticles (SNPs) by integrating machine learning moment tensor potentials (MTPs) with active learning. We successfully evaluated HER activities across various morphologies, compositions and sizes including PtAu (core-shell), PtNi (solid solution), and PtCo (size/composition-dependent morphology). We highlight the critical role of local environments in optimizing HER performance across nanoparticle sizes and compositions based on the correlation between the generalized coordination number (GCN) and HER overpotential (η_{HER}). Notably, we propose the "crown-jewel" structure as the optimal morphology for Pt-based alloy nanoparticles, where the synergistic arrangement leverages the advantages of Pt and heteroatoms to achieve superior HER activity. In addition, we extended our method to predict the turnover frequency (TOF) of SNPs at various applied electrode potentials, providing insights into the kinetics of the HER. The results show TOF also has the same trend as the GCN-η_{HER} correlation according to SNP composition, suggesting the optimal composition and morphology for HER activity. Pt@Au core-shell SNPs exhibited the highest HER catalytic activity at $Pt_{0.92}Au_{0.08}$, which has the highest Pt concentration among stable SNPs. Similarly, Co@Pt core-shell SNPs exhibited superior HER performance at $Pt_{0.08}Co_{0.92}$ compared to solid solution SNPs, suggesting their potential as efficient catalysts with reduced Pt dependency. For PtNi SNPs, which prefers a solid solution morphology, intermetallic $Pt_{0.75}Ni_{0.25}$ demonstrates the highest HER activity across all sizes considering the order–disorder effect. The study findings provide valuable insights for optimizing catalysts and guiding experimental processes, highlighting the potential of computational methods in catalyst design.