Redefining Catalysis Predictions Through Physics-Based Gaussian Model and Data-Driven Benchmarks: AuPd Alloy in ORR Catalysis for Fuel Cell Applications

Neeru Chaudhary[a], **Chuhong Lin**[a], **Ng Yan Ting**[b], **Kedar Hippalgaonkar**[b], **Tej S. Choksi**[a]

[a]*Nanyang Technological University, 62 Nanyang Drive, Singapore 637459* neeru.chaudhary@ntu.edu.sg, chuhong.lin@ntu.edu.sg, tej.choksi@ntu.edu.sg

[b]*Agency for Science, Technology and Research, 2 Fusionopolis Way, Innovis #08-01, Singapore 138634* Ng_Yan_Ting@imre.a-star.edu.sg, kedar@ntu.edu.sg

1. Introduction

The global push for sustainable energy and the critical need to mitigate climate change drive the pursuit of transformative innovations in renewable energy technologies. Among these technologies, fuel cells stand out for their potential to revolutionize the transportation and energy sectors.[1] Alloying Pd with Au modulates the electronic structure and local coordination environment, offering an optimal adsorption for reaction intermediates that significantly enhance oxygen reduction reaction (ORR) activity with improved H_2O selectivity and thus reducing overall cost.[2]

2. Results and Discussions

In this study, we present an end-to-end machine learning-guided computational framework that uniquely integrates density functional theory (DFT) with a physics-based Gaussian Process Regression (GPR) model, amalgamating stability and reactivity predictions through an alliance of our bespoke model and available foundational models from the OpenCatalyst (OC) Project by FAIRCHEM, Meta.[3] This novel approach enables the design and optimization of AuPd random alloy bimetallic catalysts for fuel cell applications, achieving a balance of high accuracy and computational efficiency. Our methodology initiates with the identification of stable nanoparticle (NP) compositions and morphologies through excess energy predictions derived from cohesive energy calculations within the bond-cutting model framework.[4] By analyzing over 1100 datasets across a spectrum of NP sizes (309 to 11000 atoms), shapes (cuboctahedron, decahedron, octahedron, and icosahedron), and configurations (10 random atomic arrangements), free energy evaluations, incorporating configurational entropy at 700 K, revealed cuboctahedron and decahedron shapes with smaller atom counts (N < 1100) and 60-80 % Pd compositions to exhibit potential for enhanced stability (Fig. 1a). The alignment of free and excess energy trends with established literature reinforces the robustness of our findings.5 To assess catalyst performance for fuel cell applications, we employed GPR models trained on small, high-quality DFT datasets, across diverse AuPd surface facets (111, 110, 100, 211, 221, 311) and compositions (5 different composition along 0 to 100% Pd). Our model achieved a strong predictive accuracy for ORR intermediates (O* (shown in Fig. 1b), OH*, and OOH*) adsorption energies, with a mean absolute error (MAE) of 0.22 eV (averaged across all ORR intermediates), by harnessing only three critical fingerprints—cohesive energy, electronegativity, and bond distance. This streamlined yet effective approach effectively captured the complexity of surface interactions, demonstrating robust performance even for intricate bimetallic surfaces. In evaluating foundational models from OC20 and OC22 dataset, we systematically compared different architectures including SCN, SchNet, GemNet, DimeNet++, and EquiFormerV2 from the OC20 and OC22 S2EF (including Total) model suite. Foundational models showed an MAE of 0.29 eV (averaged across all ORR intermediates) for selected model based on generalization capability, and alignment with DFT-calculated benchmarks.

The observed errors revealed a critical trade-off between specificity and adaptability, highlighting where bespoke models thrive in precision, foundational models offer resilience and insight into unexplored chemical environments. The integration of OC models enables high-throughput screening of ORR active sites, guiding targeted DFT calculations to generate compact and high-quality training data for bespoke GPR models. This integrated approach will be further utilized to conduct a comprehensive analysis of catalytic activity, selectivity, and reactivity metrics to establish a correlation with DFT results and existing literature and provide overall catalytic performance under operational conditions.

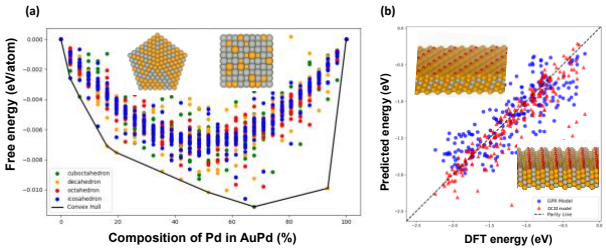

Fig. 1: Computational Design of AuPd alloy for enhanced ORR performance (a) Free energy of mixing varying along Pd compositions (%) in AuPd at temperature (T) of 700 K across different shapes and sizes (N). N varying between ~300 to 11000 atoms. Convex hull (solid black line) identifies the thermodynamically stable structures highlighted through the points on or close to the convex hull. Insets show two representative most stable structures of the nanoparticles. (b) Predictive accuracy of machine learning models –GPR and OC20 model against DFT-calculated energies for O* adsorbed on AuPd surfaces shown alongside the parity line (dashed). Insets depict surface structures used in the adsorption studies.

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