Prediction of Pd, Pt, and Rh adsorption energies on graphene quantum dots using graph neural networks

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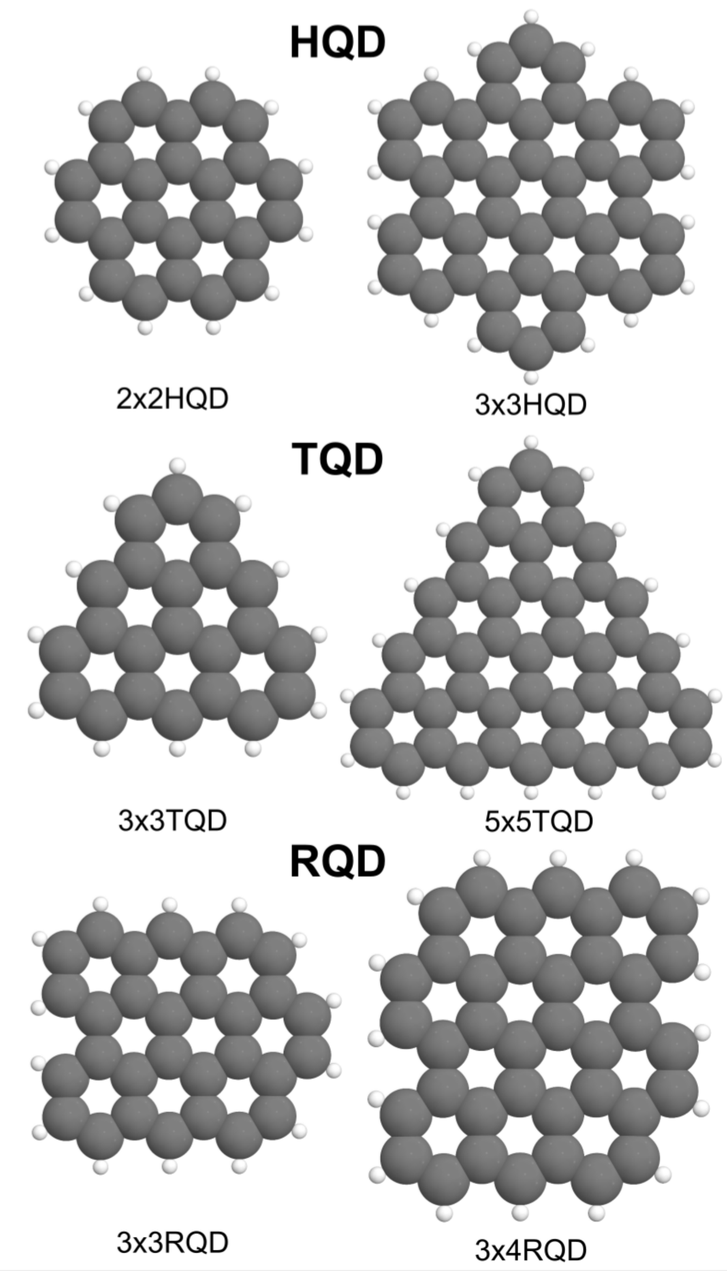
INTRODUCTION

Transition metals such as Pd, Pt, and Rh exhibit excellent electrocatalytic activity but suffer from the disadvantages of being expensive and scarce. An effective strategy to optimize their use is to employ them as adatoms on suitable supports, as the graphene quantum dots (GQDs) [1].

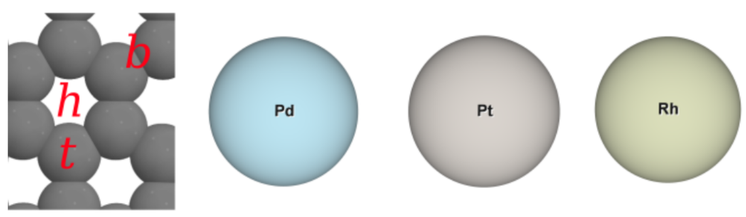
At the same time, artificial intelligence (AI) and machine learning (ML) are transforming the way materials are designed by enabling the efficient exploration of vast chemical and structural spaces, with computation times significantly shorter than conventional methods [2]. Within this context, graph neural networks (GNNs) have emerged as particularly suitable tools for modeling non-periodic atomic systems, as they explicitly capture connectivity, local interactions, and spatial symmetries without relying on periodic cells. Their application in materials science allows for property prediction at a substantially lower computational cost compared to conventional approaches [3].

METHOD AND MODEL

Ab initio computational methods were used to model the adsorption of a metal atom on the surface of GQDs with different geometries, with all system edges saturated by hydrogen. Density functional theory (DFT) calculations were conducted using the VASP package.



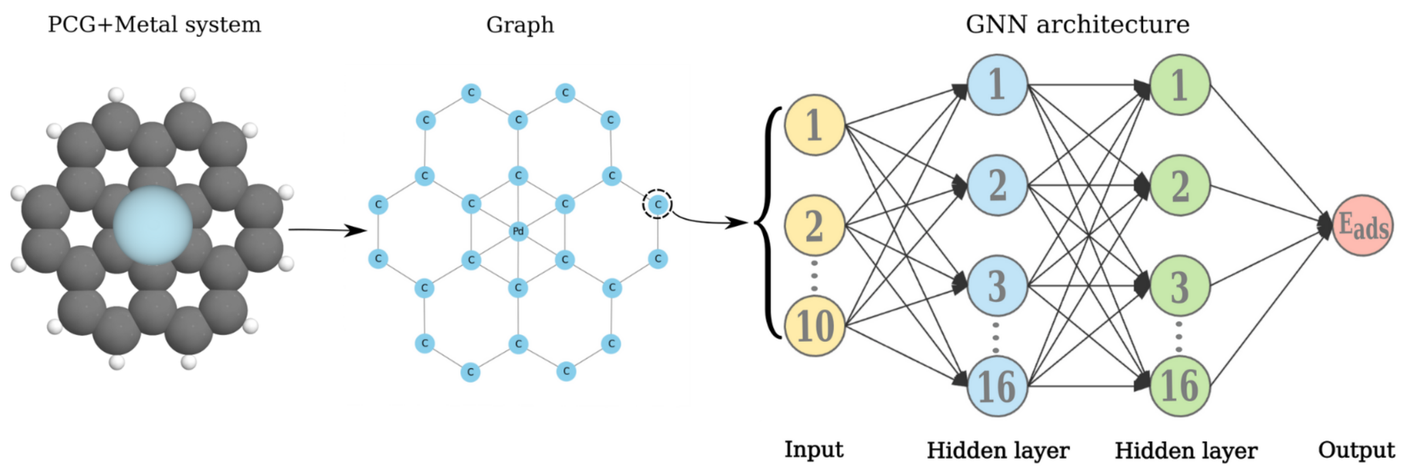
The adsorption of Pd, Pt, and Rh metals was evaluated at the high-symmetry sites of each quantum dot (*h: hollow, b: bridge, t: top*)



PyTorch and its specialized library were used for graph construction and GNN implementation

GNN ARCHITECTURE

Each PCQ–Metal system is represented as a graph with $N^o_{nodes} = N^o_C + 1$ (n° carbon atoms + metal atom). The graph edges represent the chemical bonds formed within each system. As an example, HQD+Pd adsorbed a *hollow* site is shown.

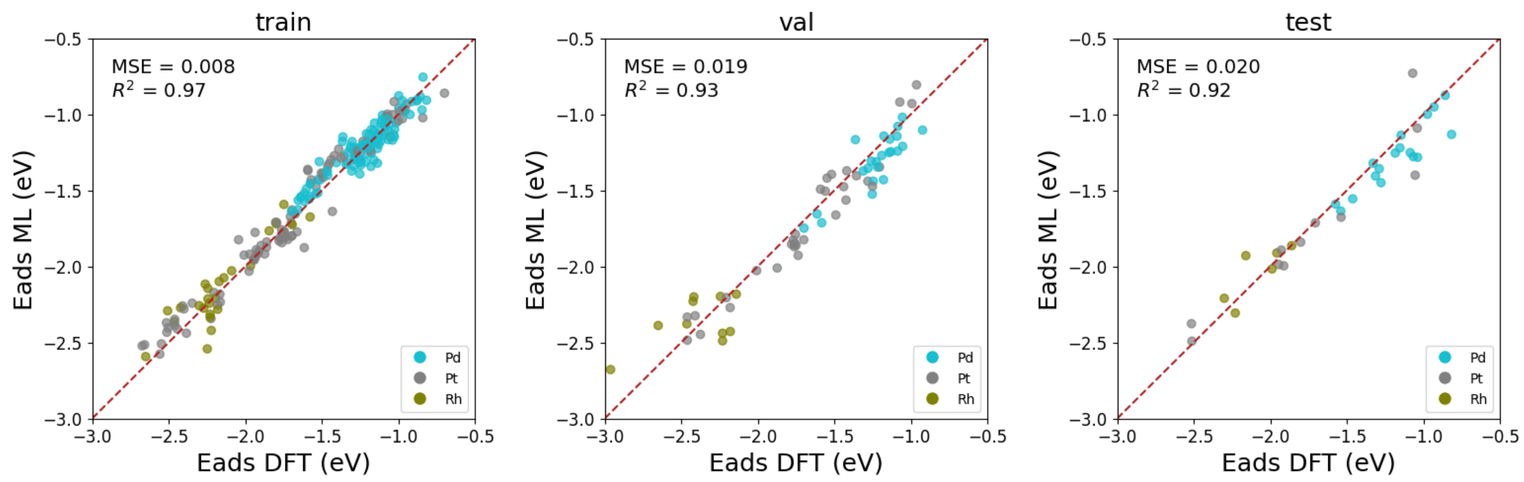


Parameters	Optimization
Hidden layers	2
Neurons per hidden layer	16
Node features	10
Output	Adsorption energy
Optimizer	Adam
Loss function	MSE
Regularization	Early stopping

Features	
Group	Period
Charge	Magnetization
Distance to metal	Bond order
Electron affinity	Hydrogen connection
Atomic number	Atom location

RESULTS

The dataset used consists of 328 graphs. The training was performed with a 70/20/10 split for train/validation/test. Predicted adsorption energies from the GNN were compared with the DFT-calculated values, and the mean squared error (MSE) and coefficient of determination (R^2) were computed for each subset.



CONCLUSION

The GNN showed consistent performance (R^2 = 0.97 train, 0.93 val, 0.92 test), accurately predicting adsorption energies. The graph representations captured the structural information of the systems, enabling strong predictive power. These findings highlight the potential of GNNs to accelerate for property prediction and support the design of electrocatalysts alongside DFT-based approaches.

ACKNOWLEDGEMENTS

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