2023-2024 Grand Challenge Award Final Report

Awardee: **Graeme Henkelman, Professor**
Chemistry

Research Award Title: **Exascale Computing Methods for the Design of New Battery Materials and Catalysts**

Research Summary

The opportunity provided to me through the Grand Challenge Faculty Award was extremely valuable for my research program. With the freedom to focus on research, I published fifteen papers,[i-xv] gave four invited talks, and perhaps most importantly, established new research directions for the future. I was able to spend an extended period of time at the Institute for Pure and Applied Mathematics (IPAM) on the campus of UCLA during my leave in the Spring of 2023. Here, I will briefly describe a few of the research highlights from the award period and the opportunities that it has allowed.

The focus of my research has been the computational design of materials, with application to the development of new energy sources. The research involves development of the computational methods required to model the function of materials, as well as strategies to invert the problem and predict new materials with a desired function. Key to this effort is a new area of research for my group in exascale computing methods. To learn the state-of-the-art in exascale computing for materials design, I was a core participant in the Long Program at IPAM entitled "New Mathematics for the Exascale: Applications to Materials Science" from March 13 - June 16, 2023.

A good model system for these studies is that of nanoparticle catalysts. Catalysts are important whenever energy is converted to or from a chemical form. A grand challenge, for example, is to find a catalytic system which will allow the production of fuel from sunlight; another is to replace platinum for the efficient electrochemical burning of fuel in a fuel cell. Nanoparticles are attractive because they can have properties which are different from bulk materials, and with on the order of 100 atoms, the problem of searching the composition space and predicting catalytic activity is tractable.

We have made several advances in this area in 2023. In the area of catalysts for water purification, we have worked with the group of Charlie Werth and Simon Humphrey to design Pd-Au-Cu nanoparticle catalysts for nitrite reduction, which is a common pollutant from excess fertilizer in water sources.[ix] For efficient hydrogen evolution from seawater, we have developed a novel Ni-CO@Fe-Co core@shell nanobox of Prussian blue analoges.[xv] Using metal-support interactions to tune catalytic activity, we have shown that classical scaling relations can be broken to facilitate efficient oxygen reduction.[xii] Perhaps the most scientifically interesting aspect of our work was to show that Cu dendrites are highly active for electrocatalytic hydrogen evolution when exposed to a plasma - a very interesting possibility in the catalytic field.[iv] This advance is being followed up with collaborators here at UT to see if we can activate methane with a plasma-assisted catalyst.

In the area of battery materials, we collaborated with the Mullins group to show how intermolecular interaction in electrolytes can be tuned to facilitate the development of rechargeable Zn-air batteries.[ii] A large push was made in the area of Na-ion batteries, where we also tuned electrolyte composition to enhance the performance of Na-S batteries with the Manthiram group,[vi] and developed intermetallic to prevent dendimer formation at the Na anode with the Mitlin group.[vi] Finally, we used tools from machine learning to help discover a novel solid electrolyte with a zeolite framework to maintain the structure and a liquid host to provide a high cation conductivity.[v]

Perhaps the largest effort while my group was at IPAM was to develop a machine learning (ML) package called PyAMFF.[vii] While we are just at the early stages of this project, we are continuing to develop methods for ML acceleration of electronic structure calculations, determining saddle points, and discovering the function of new materials.

The Moncrief Award allowed me to help organize a future three-month long program at IPAM entitled "Bridging the Gap: Transitioning from Deterministic to Stochastic Interaction Modeling in Electrochemistry", which will take place from Sept 3 - Dec 12, 2025. This program includes four week-long workshops focusing on computational methods to model electrochemical interfaces. While acting in an advisory role, I was able to interact closely with the diverse group of researchers. As a core participant, this program will be of tremendous value for developing novel methods associated with the very challenging aspect of electrochemistry at interfaces including modeling the double layer, charge transfer, constant potential simulations, and the role of solvent in batteries and electrocatalysts.

The time provided by this program has given me the chance to explore new research directions to keep my research program alive and exciting. I am grateful to the Oden Institute and the Moncrief Grand Challenge Faculty Award Program for giving me this opportunity.

Papers Published

i. B. Kim, K. Shin, G. Henkelman, and W.-H. Ryu, "CO_2-mediated Porphyrin Catalysis in Reversible Li-CO_2 Cells" *Chem.Eng. J.* **477**, 14741 (2023).

ii. Y. Wang, N. Katyal, Y. Tang, H. Li, K. Shin, W. Liu, R. He, M. Xu, G. Henkelman, and S.-J. Bao, "One-Step Pyrolysis Construction of Bimetallic Atom-Cluster Sites for Boosting Bifunctional Catalytic Activity in Zn-air Batteries" *Small* 2306504 (2023).

iii. Z. Wang, J. Diao, J.N. Burrow, K.K. Reimund, N.Katyal, G. Henkelman, and C.B. Mullins, "Urea-modified Ternary Aqueous Electrolyte with Tuned Intermolecular Interactions and Confined Water Activity for High-Stability and High-Voltage Zinc-Ion Batteries" *Adv. Func. Mater.* 2311271 (2023).

iv. H. Zhang, J. Diao, Y. Liu, H. Zhao, B.K.Y. Ng, Z. Ding, Z. Guo, H. Li, J. Jia, C. Yu, F. Xie, G. Henkelman, M.-M. Titirici, J. Robertson, P. Nellist, C. Duan, Y. Guo, D.J. Riley, and J. Qiu, "In-Situ Grown Cu Dendrites Plasmonically Enhance Electrocatalytic Hydrogen Evolution on Facet-Engineered Cu_2O" *Adv. Mater.* **35**, 2305742 (2023).

v. X. Chi, M. Li, X. Chen, J. Xu, X. Yin, S. Li, Z. Jin, Z. Luo, X. Wang, D. Kong, M. Han, J.-J. Xu, Z. Liu, D. Mei, J. Wang, G. Henkelman, and J. Yu, "Enabling

High-Performance All-Solid-State Batteries via Guest Wrench in Zeolite Strategy" *J. Am. Chem. Soc.* **145**, 24116-24125 (2023).

vi. W. Guo, S. Wang, Y. Xie, C. Fang, L. Liu, Q. Lou, X. Lian, and G. Henkelman, "Hydrogen Peroxide Synthesis via Electrocatalytic Water Oxidation on sp^3 and sp^2 Carbon Materials Mediated by Carbonates and Bicarbonates" *ACS Sustain. Chem. Eng.* **11**, 12114-12122 (2023).

vii. L. Li, R.A. Ciufo, J. Lee, C. Zhou, B. Lin, and G. Henkelman, "Atom-Centered Machine-Learning Force Field Package" *Comp. Phys. Comm.* **292**, 10883 (2023).

viii. H. Ha, C. Lee, J.S. Park, C.-H. Chung, S. Lee, G. Henkelman, H.Y. Kim, and K. Shin, "Genetically Evolved Graphene Encapsulated Random Alloy Nanoparticles for Li-Air Battery" *Catal. Today* **424**, 114303 (2023).

ix. P. Kunal, C. Yan, H. Guo, H. Li, C. Brady, M. Duncan, X. Zhan, C. Werth, G. Henkelman, and S. Humphrey, "Pd-Au-Cu Ternary Alloy Nanoparticles: Highly Tunable and Economical Nitrite Reduction Catalysts" *ACS Catal.* **13**, 11945-11953 (2023).

x. C. Du, S. Lu, J. Wang, X. Wang, M. Wang, H. Fruehwald, L. Wang, B. Zhang, T. Guo, J. Mills, W. Wei, Z. Chen, Y. Teng, J. Zhang, C. Sun, H. Zhou, R. Smith, B. Kendall, G. Henkelman, and Y. Wu, "Selectively Reducing Nitrite into NH_3 in Neutral Media by PdCu Single-Atom Alloy Electrocatalysis" *ACS Catal.* **13**, 10560-10569 (2023).

xi. Y. Wang, H. Dong, N. Katyal, B.S. Vishnugopi, M.K. Singh, H. Yao, Y. Liu, P. Liu, P.P. Mukherjee, G. Henkelman, J. Watt, and D. Mitlin, "Intermetallics Based on Sodium Chalcogens Promote Stable Electrodepostion - Electrodissolution of Sodium Metal Anodes" *Adv. Energy. Mater.* **13**, 2204402 (2023).

xii. C. Lee, K. Shin, Y. Park, Y.H. Yoon, G. Doo, G.H. Jung, M. Kim, W.C. Cho, C.-H. Kim, H.M. Lee, H.Y. Kim, S. Lee, G. Henkelman, and H.-S. Cho, "Catalyst-Support Interactions in Zr_2ON_2-Supported IrO_x Electrocatalysts to Break the Trade-Off Relationship Between the Activity and Stability in the Acidic Oxygen Evolution Reaction" *Adv. Func. Mater.* **33**, 2301557 (2023).

xiii. J. Eichler, J. Burrow, N. Katyal, G. Henkelman, and C.B. Mullins, "Modulation of CO_2 Absorption Thermodynamics and Selectivity in Alkali-Carbonate Activated N-Rich Porous Carbons" *J. Mater. Chem. A* **11**, 12811-12826 (2023).

xiv. K. Liu. J. Wang, C. Lou, Z. Zhou, N. Zhang, Y. Yu, Q. Zhang, G. Henkelman, M. Tang, and J. Sun, "Simple Construction and Reversible Sequential Evolution Mechanism of Nitrogen-Doped Mesoporous Carbon/SnS_2 Nanosheets in Lithium-Ion Batteries" *Appl. Surf. Sci.***618**, 156673 (2023).

xv. H. Zhang, J. Diao, M. Ouyang, H. Yadegari, M. Mao, M. Wang, G. Henkelman, F. Xie, and D.J. Riley, "Heterostructured Core-Shell Ni-Co@ Fe-Co Nanoboxes of Prussian Blue Analogues for Efficient Electrocatalytic Hydrogen Evolution from Alkaline Seawater" *ACS Catal.* **13**, 1349-1358 (2023).

Invited Talk

1. GetCO2, Brisbane, Australia, "Correlating Structure and Function for Nanoparticle Catalysts", 12/2023.

2. Global Conference on Innovation Materials, Jeju Island, Korea, "Correlating Structure and Function for Nanoparticle Catalysts", 06/2023.
3. Society for Industrial and Applied Mathematics, Amsterdam, NL. "From Saddle Point Finding Methods to Long Time Scale Dynamics", 02/2023.
4. Alan Turing Institute, London, UK, "Correlating Structure and Function for Nanoparticle Catalysts", 01/2023.