Pacific Northwest
NATIONAL LABORATORY

PNNL-36692

Developing Capabilities in Physical and Computational Sciences

September 2024

Wendy J Shaw

U.S. DEPARTMENT OF ENERGY

Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

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Pacific Northwest National Laboratory
Richland, Washington 99354

Abstract

The Physical and Computational Sciences Directorate (PCSD) performs fundamental research in support of the science missions of Offices of Basic Energy Sciences (BES), Advanced Scientific Computing Research (ASCR), High Energy Physics (HEP), Nuclear Physics (NP), and Fusion Energy Sciences (FES), and others within the domains of the chemical, materials, computational sciences, mathematics, and physics. This LDRD project aims to provide funding to develop/demonstrate research capabilities for proposals and publications to support these science missions. Staff will propose small research tasks/projects to be performed under this overall project.

Summary

Here are short summaries of selected tasks supported.

Project number: 80132
FY: 2022
Title: Selective Rare Earth Capture
PI: John Loring

This project was in collaboration with a team at Sandia National Lab (SNL) that was synthesizing functionalized metal organic frameworks (MOFs) for selectively absorbing rare earth elements. An infrared spectroscopic capability was developed at PNNL for investigating at a molecular level the complexation of lanthanide cations to the MOFs in aqueous suspension. This capability was tested on SNL synthesized MOFs, but the cation uptake concentration was much lower than the SNL project team intended and unfortunately below the detection limit of the infrared method.

Project number: 80457
FY: 2022
Title: Intercalation of Lithium into Gibbsite in Complex Brines
PI: Trent Graham

The project focused on the recovery of lithium through the intercalation of lithium into gibbsite, forming lithium aluminate layered double hydroxides. It supported research utilizing nuclear magnetic resonance (NMR) spectroscopy to study lithium sorption from a multicomponent brine, representative of Salton Sea water, into gibbsite. The study involved solution-state ^{7}Li NMR spectroscopy and solid-state characterization using X-ray diffraction (XRD). Results confirm that lithium intercalation can be tracked over time with solution-state ^{7}Li NMR spectroscopy and, in tandem with XRD, indicated that lithium sorption leads to the formation of lithium aluminate layered double hydroxides even in multicomponent brines containing magnesium, which potentially competes with lithium for sorption sites. This provides valuable insights into means to experimentally monitor mechanisms of lithium recovery from brine solutions.

Project number: 80922
FY: 2023
Title: Development of an Electrochemical Hydroformylation Catalyst
PI: Eric Wiedner

This project focused on the computational design of an electrochemical hydroformylation catalyst and the development of a high-pressure spectroelectrochemical cell that could be used to investigate such catalysts. Density Functional Theory (DFT) was used to establish a reaction network comprised of 58 potential elementary steps for electrochemical reaction of a model rhodium catalyst with ethylene, carbon monoxide, and an organic acid. These studies revealed that a competent catalyst could be feasible, and would require tailored ligands stereochemical control of the active site and the ability to finely tune the potential, pressure, and pH of the reaction medium. The high-pressure cell was designed and successfully built, laying the foundation for further study in this area.

Project number: 81137
FY: 2023
Title: Efficient and Selective Magnetoelectrochemical Synthesis and Separations
Team: Venkateshkumar Prabhakaran, Grant E. Johnson, Shuai Tan, and Lyndi Strange

This project established a new magneto-electrochemical spectroscopy capability in the Energy Sciences Center, which includes a combined *operando* spectroelectrochemical cell, an electromagnet power supply, and a Tesla meter that detects magnetic fields. This platform enables the study of electrochemical reactions under the influence of magnetic fields. For example, we recently demonstrated a 20-fold decrease in the charge transfer resistance of common redox molecules, such as Ferrocene, on electrode surfaces in an applied 700 Gauss magnetic field. Following this initial demonstration, preliminary data was collected to determine the effect of magnetic fields on the electron-mediated electrochemical deposition of platinum, a critical precious metal widely used in catalytic applications. The results obtained from this project will be a part of a forthcoming publication, which is expected to be submitted in November 2024. An invention disclosure report is also being submitted for applying the magnetoelectrochemical capability to recycle platinum group metals. The data generated from this project was also employed in the development of two proposals submitted in 2024: (1) co-PI in the Advanced Research Projects Agency–Energy Revolutionizing Ore to Steel to Impact Emissions proposal entitled “Negative Emission Ironmaking *via* Redox-Mediating Red Mud Electrowinning (NEIRME),” PI Xin Zhang (invited a full proposal but not funded) $1M/3years, (2) Lead PI in Office of Science Early Career Research Program proposal entitled “Selective and Sustainable Magneto-Electrochemical Reactive Separation of Platinum Group Metals” (invited a proposal but not funded). We continue exploring further funding opportunities, including using the new magnetoelectrochemical capability in the Basic Energy Sciences funded core program in Separation Science and the Non-Equilibrium Transport Driven Separations Initiative.

Project number: 82060
FY: 2023
Title: Development of Atmospheric Pressure Infrared Spectroscopic Titration Cell
PI: John Loring

Progress was made in the development of an infrared spectroscopic cell for following the reactivity of minerals that are covered in Angstrom to nanometer thin water films in humidified gases.

Project number: 82252
FY: 2023
Title: Designing Electrolytes for Accelerated Plating of Functional Alloys with Morphological Control
Team: Vijayakumar Murugesan, Dan Thien Nguyen

The project identified an initial electrolyte system for the electroplating of cobalt metal and established electrochemical parameters for accelerated cobalt deposition, achieving a high deposition rate of over 3 mA/mm² and a Coulombic efficiency greater than 90% under static mode operation of the electrochemical cell. We completed the design of the first 3D printing prototype, capable of printing objects with diameters of approximately 0.5 mm. Additionally, we

trained a summer intern in electroplating technology as part of the project. The preliminary data generated has laid the foundation for pursuing new funding from PNNL's LDRD program for FY25.

Project #: 83034
Title: Machine Learning for Improved Atomic Force Microscopy
FY: 2024
PI: Benjamin Legg

This project used machine-learning (ML) tools to understand the impact of instrument settings on AFM image quality and explore the ability of ML methods to quantify image quality and/or remove imaging artifacts. The project was led by Benjamin Legg and Shuai Zhang, with important contributions from five students (Lejla Biberic, Thom Snoeren, Christina Yang, Greg Moore, and Haoqing Zhang) from data-analysis course at University of Washington led by David Beck. A database of over 200 multi-channel AFM images and ideal ground-truth images was produced using systematically varied instrument settings. Python code was written to extract image metadata and generate coaligned image/ground-truth pairs. The labeled pairs were used to quantify image-quality, and ML-algorithms were developed to predict image quality. New insights into AFM image-structure were obtained that may be exploited by future algorithms. Regression algorithms for image artifact removal showed that improvements could be achieved under ideal conditions, but also revealed a high probability of introducing major new artifacts if the training-data is insufficient. These insights will help develop robust image-acquisition routines that exploit the potential of ML while minimizing the dangers of producing biased data.

Project number: 83219
FY: 2024
Title: Retrieval Augmented Language Model for Facilitating Catalysis Understanding
PIs: Sai Munikoti, Sameera Horawalavithana

A wealth of theoretical and experimental scientific knowledge is contained within research/scientific papers. However, analyzing these papers and accurately retrieving key information requires domain expertise and can be very time-consuming. Retrieval augmented generation (RAG) is a popular technique to adapt language models toward domain-specific tasks including information retrieval. In this project, we first develop the standard RAG-based chat (research) assistant capability (ChemAIst) by leveraging a text-based vector database from 370 catalysis-related scientific documents. ChemAIst can be used for various tasks, including question answering, summarization, search, extraction and synthesis of crucial insights, where along with the response, it also assures trustworthiness by yielding evidence in terms of text chunks from the scientific articles in the corpus. Then, we enrich the vector database by incorporating structurization via a knowledge graph (KG). The KG is created from scientific corpus in a semi-automatic fashion leveraging both Human (subject matter (catalysis) expert) and AI (GPT-4). ChemAIst with Text+KG vector database could enhance the response as well as trustworthiness by providing both text chunks and KG triplets in evidence. We believe ChemAIst with AI-driven information retrieval/synthesis capability could assist scientific discoveries in catalysis. This work along with systematic evaluation would be part of a forthcoming paper to be submitted in the coming months.

Project number: 83229

FY: 2024
Title: Generative Models for Bacterial Self-Organization in Spatial Environments
PI: Mahantesh Halapanavar

The project focused on developing and optimizing high-performance agent-based simulations to study spatial patterns in microbial ecology. Initially, the algorithm was created using a high-level interpreted programming language (Python), then optimized and parallelized in an object-oriented language (C++ and Open MP). The simulations consist of agents performing random walks in a continuous 2D space, interacting with nearby agents, which updates their states, eventually leading to reproduction or death. An agent represents a microbial entity, which interacts with other agents in its locality. The computation is enabled by an optimized algorithm for the building a dynamical structure called Random Geometric Graph (RGG) to efficiently compute the agent interactions in a local neighborhood. The simulations have now reached realistic scales -- tens to hundreds of millions -- and model certain experimental findings in microbiomes. The model was further adapted to simulate publicly available mouse gut microbiome data. Additionally, discussing and exchanging ideas with teams from PNNL's Biological Sciences Division also enriched our understanding and provided valuable insights into the field.

Project number: 83516
FY: 2024
Title: Novel Searches for Non-Classical Signatures in Axion Dark Matter
PI: Erik Lentz

The purpose of this project is to study how novel searches for non-classical signatures of axion dark matter could significantly enhance their discovery potential over traditional searches. The project has constructed the cavity QED formalism to analyze signatures in single and arrayed cavity haloscope detectors such as those implemented by multiple axion DM search collaborations (ADMX, CAPP, RADES, etc.), and has found multiple example novel signatures in those detectors from prescribed axion states including squeezed, NOON-type, and pair-wise spatially entangled condensates. This project is currently writing a white paper motivating the presence of these example non-classical signatures. This white paper will be followed in the next FY by an expanded study of possible non-classical signatures, design principles for targeted searches, and the resulting sensitivity enhancement over baseline search techniques.

Project number: 83700
FY: 2024
Title: Neural Network Enhanced Force Fields from AIMD Data
PI: Mal-Soon Lee

We have developed a neural network potential within the framework of DeePMD for water in zeolite. The developed force field demonstrates a comparable accuracy with ab initio molecular dynamics (R^2>0.95, Energy RMSE<10^{-3} eV), but allows for a more efficient (>2 orders of magnitude faster) molecular dynamics simulation of water in zeolite. This model provides the foundational parameters for further development to include organic species such as cyclohexanol. A preliminary reactive force field for solvated cyclohexanol in zeolite has been parameterized via transfer learning. The current model maintains a high accuracy for zeolite and water (R^2>0.95), but less satisfying accuracy for cyclohexanol (R^2<0.65), which requires further fine-tuning via other methods such as active learning or other frameworks such as MACE.

Project number: 83840
FY: 2024
Title: The Kinetics and Thermodynamics of Energy Material Nucleation Monitored by In Situ SAXS
PI: Jinhui Tao

We have measured the nucleation of zinc oxide on SAMs with methyl (CH3), hydroxyl (OH) and carboxylate (COOH) functional groups by in situ microscopy. The highest number density was observed for the case of COOH groups, then bare gold and CH3, with the lowest number density on the surface of OH group. The COOH and OH show the best control of the face with (001) and (100) faces. This study suggests the binding affinity are strongest for COOH/ZnO and weakest for OH/ZnO. This research is going to be complemented by the nucleation pathways and interfacial energy from our next stage of homogeneous nucleation monitored by in situ SAXS in our division. This work would be part of our publication on the plan to be submitted by this November.

Project number: 84500
FY: 2024
Title: Developing Peptoid-Based Crystalline Ultrathin Nanosheets for Next-Generation Quantum Information Platform
PI: Ying Chen

This project has supported pulsed electron paramagnetic resonance (EPR) experiments conducted at temperatures ranging from 5 K to 40 K on a series of peptoid-based crystalline nanomembranes with 2D arrays of molecular qubits. The results reveal that the phase memory time of these qubits are modulated by variations in the peptoid sequences, the types of molecular qubits (such as rare earth elements vs organic radicals), and the spatial arrangement between qubits. Notably, peptoid-based ultrathin nanosheets featuring organic radical TEMPO as side chains exhibit unique quantum properties, achieving coherences in the microsecond range at 40K. These sequence-defined peptoid-based nanomembranes represent a promising platform for the development of quantum information materials, enabling precise manipulation of spin qubits and qubit local environment to achieve sequence-programmed quantum properties. Additional EPR experiments are planned for the final week of this fiscal year, with a forthcoming paper to be submitted in the coming months, laying the groundwork for future proposals in Quantum Information Science (QIS).

Acknowledgments

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Acronyms and Abbreviations

ADMX - Axion Dark Matter eXperiment
AFM - atomic force microscopy
ASCR - Advanced Scientific Computing Research
AIMD - ab initio molecular dynamics
BES - Basic Energy Sciences
CAPP - Center for Axion and Precision Physics Research
DFT - Density Functional Theory
EPR - electron paramagnetic resonance
FES - Fusion Energy Sciences
HEP - High Energy Physics
KG - knowledge graph
LDRD - Laboratory Directed Research and Development
ML – machine learning
MOFs - metal organic frameworks
NMR - nuclear magnetic resonance
NP - Nuclear Physics
PCSD - Physical and Computational Sciences Directorate
QED - quantum electrodynamics
QIS - quantum information science
RADES - Relic Axion Dark-Matter Exploratory Setup
RAG - retrieval augmented generation
RGG - random geometric graph
RMSE - root mean square error
SAMs - self-assembled monolayers
SAXS - small-angle x-ray scattering
SNL - Sandia National Laboratory
XRD - x-ray diffraction

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