ANL-24/11

2023 LDRD Annual Report

Laboratory Directed Research and Development Program Activities

Laboratory Directed Research and Development

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Office of the Director, Argonne National Laboratory

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LDRD Program Overview

Argonne National Laboratory's Laboratory Directed Research and Development (LDRD) program encourages the development of novel technical concepts, enhances the Laboratory's research and development (R&D) capabilities, and enables pursuit of strategic laboratory goals.

Argonne's LDRD projects are proposal based and peer reviewed, supporting ideas that require advanced exploration so they can be sufficiently developed to pursue support through normal programmatic channels. Among the aims of the projects supported by the LDRD program are the establishment of engineering proofs of principle, assessment of design feasibility for prospective facilities, development of instrumentation or computational methods or systems, and discoveries in fundamental science and exploratory development.

All LDRD projects have demonstrable ties to one or more of the science, energy, environment, and national security missions of the U.S. Department of Energy (DOE) and its National Nuclear Security Administration (NNSA), and many are also relevant to the missions of other federal agencies that sponsor work at Argonne. A natural consequence of the more "applied" type projects is their concurrent relevance to industry.

The LDRD program is managed in overarching portfolios, each containing multiple projects each fiscal year. The LDRD Prime portfolio is further divided into strategic focus areas aligned with Argonne's strategic plan.

FY 2023 LDRD Program Components

LDRD Prime

The largest component of Argonne's program is LDRD Prime, which emphasizes R&D explicitly aligned with Laboratory major initiatives in support of Argonne's strategic plan. The choice of Focus Areas under the LDRD Prime component reflects the major initiatives; the state of development of relevant technical fields; the potential value of advancing those fields to DOE/NNSA and the nation; and the compatibility of the fields with existing facilities, capabilities, and staff expertise at Argonne.

Focus Areas with projects that ended in FY23 are:

AI + Science
Biological and Environmental Science
Clean Energy and Sustainability
Climate and Energy Action
Energy Security and Sustainability
Energy Storage
Hard X-ray Sciences
Imaging and Detection of Signatures
Microelectronics
Physical Sciences and Engineering
Quantum Information Sciences
The Universe as Our Laboratory (ULab)

Director’s Collaborations

The Director’s Collaborations LDRD projects support research that is paired with coordinating research efforts at a partner institution. These are generally small projects selected through a collaborative process.

Named Fellows

Argonne’s LDRD Named Fellows program aims to support the scientific or engineering research of exceptional early career scientists and engineers. Working with an Argonne sponsor (a senior member of research staff), LDRD Named Fellows carry out work that is either at the forefront of new research areas or is synergistic with current research efforts.

Innovate

The Innovate component of the LDRD program invests in a full spectrum of investigator-initiated proposals across the Laboratory in DOE-mission-related science and engineering areas. This provides an avenue for R&D staff to propose highly innovative projects in research areas outside the purview of the Prime Focus Areas. Within the Innovate LDRD portfolio is the Seed program, open to postdoctoral appointees and early career researchers to apply for a small project to perform independent research and explore their own ideas.

Swift

The LDRD Swift component provides an avenue for R&D staff to conduct short-term research with a targeted funding opportunity in mind, as well as a means for researchers to explore ideas before developing a full proposal. Projects funded through this component area have a maximum one-year duration. As with the Innovate component, the Swift component invests in a full spectrum of proposals across all mission-related science and engineering areas.

FY 2023 LDRD Summary Report

This summary report provides an overview of all LDRD projects at Argonne that concluded in Fiscal Year 2023. Many projects are funded for multiple years, the initial fiscal year for each project is indicated by the first four digits of the LDRD project number.

AI + Science

Biological and Environmental Science

Clean Energy and Sustainability

Climate and Energy Action

Energy Security and Sustainability

Energy Storage

Hard X-ray Sciences

Imaging and Detection of Signatures

Microelectronics

Physical Sciences and Engineering

Quantum Information Sciences

Universe as Our Laboratory (ULab)

Director’s Collaborations

Named Fellows

Innovate

Swift

AI + Science

LDRD # 2021-0133	AI + Science
Project Title:	Nuclear Quantum Monte Carlo with Machine Learning Techniques
Investigators:	Alessandro Lovato Corey J. Adams, Prasanna Balaprakash, Krishnan Raghavan

Abstract

Grand challenges in nuclear physics require a paradigm shift in nuclear many-body theory that can be accomplished by introducing artificial intelligence (AI) techniques to accurately solve the structure of large nuclei and their interactions with neutrinos. We introduced an AI representation of the imaginary-time propagator that enables quantum Monte Carlo studies of medium-mass nuclei being experimentally investigated at both the Argonne Tandem Linac Accelerator System and the Facility for Rare Isotope Beams. Concurrently, we developed novel deep-learning methods to accurately reconstruct the nuclear electroweak response functions of atomic nuclei, a critical ingredient for the success of the Deep Underground Neutrino Experiment, DOE's flagship neutrino-oscillation experiment in which Argonne aims to play a growing role.

LDRD # 2021-0138	AI + Science
Project Title:	AI Patterns for Executable End-to-End Biological Programming
Investigators:	Arvind Ramanathan Gyorgy Babnigg, Benjamin J. Blaiszik, Qingteng Zhang

Abstract

Our project encompassed four principal areas:

(1) Cloning, protein expression and purification, and protein characterization of elastin-like proteins (ELPs). The purified ELPs were tested for phase transition. (2) Automation of experimental protocols with Opentrons-2 (OT-2) and Hudson robotics: Our ongoing efforts have resulted in an extensible software library in Python that can be used to implement a variety of common experimental workflows. Developed in coordination across several ongoing efforts, our extensible library automates several common experimental workflows. We have also implemented the cherry-picking protocol, which is not native to the Hudson robotics instruments. Another effort was to integrate the microfluidics lab confocal microscope with the Opentrons OT-2 liquid handler. We have developed methods to move microtiter plates between the OT-2 and the microscope, which is equipped with a specialized stage adaptor.
(3) Characterizing LLPS with small-angle X-ray photon correlation spectroscopy (SA-XPCS) at Advanced Photon Source: Studies of thermally induced LLPS with SA-XPCS have been carried out on a wide spectrum of samples at beamline 8-ID-I, APS. 8 different ELPs were investigated: VPAVG with 2, 4, 5 and 6 repeats, VPGVAG with 2, 4, 5 repeats, and APGVG with 4 repeats. Distinct phase separation behaviors have been observed consistently in all 8 conditions upon ramping up the temperature. The fact that coherent x-ray scattering (SA-XPCS) is much more sensitive than optical absorption coefficient towards

phase-separation allows LLPS in ELP to be studied at much lower temperature and much more nascent stages, which prompted us to probe the conditions that may lead to reversible LLPS in ELPs. We have observed that, for ELPs with VPAVG monomer, as the number of repeats increases, the phase separation temperature decreases, and the reversibility improves. This conclusion is supported by systematic SA-XPCS results from ELPs with different repeats of VPAVG monomers. (4) Simulations, AI/ML and data integration: We have also simultaneously carried out and built up our simulation and data integration framework. Several of the peptides that have been chosen for experiments have been simulated using our AI-driven simulation toolkit. The data generated from our simulations and experiments was being integrated into the data integration framework for our AI/ML methods. In particular, the data is being organized into a searchable matrix, where the metadata can be used to capture experimental conditions, simulation data (trajectories, inference of conformational states, etc.) and AI/ML methods.

LDRD # 2021-0158	AI + Science
Project Title:	Automated Model Inference for Cosmological Structure Formation
Investigators:	Salman Habib Prasanna Balaprakash, Xiaofeng Dong, Arindam Fadikar, Katrin Heitmann, Sandeep R. Madireddy, Nesar S. Ramachandra

Abstract

This project focused on the development of effective generative models for emergent behavior, where the fundamental theory is known, but a direct pathway to predictions for experiments or observations is prohibitively complex. In this project, we developed AI/ML methods to construct an effective theory for cosmological structure formation, based on data from large-scale simulations. Our focus was on the physics of matter clustering, which underlies the analysis and understanding of the majority of next-generation DOE, NASA, and NSF-supported cosmology missions. We have made a number of advances including the construction of fast 3-d generative models based on numerical simulations and developed multiple metrics for model accuracy, developed a simple diffusion-reaction model as a proxy for halo formation, which can be used as a basis for an AI/ML approach to universal behavior, and finally, constructed an AI/ML-based method for optimal fitting of observational spectral data that determines the best choices for modeling parameters of simulation subgrid models. In all of these efforts, the aim was to produce schemes that speed up computations by orders of magnitude in ways that also provide scientific insight.

LDRD # 2021-0173	AI + Science
Project Title:	AI-Steer: AI-driven Online Steering of Light Source Experiments
Investigators:	Rajkumar Kettimuthu Jonathan D. Almer, Tekin Bicer, Francesco De Carlo, Barry Lai, Viktor Nikitin, Aniket Tekawade

Abstract

Over the course of this three year project, we have successfully developed impactful tools and frameworks to move the state-of-the-art in autonomous steering of light source experiments. We demonstrated their utility for several experiments at multiple beamlines. We developed a semi-feature detection tool, TomoEncoders, that intelligently reduces 3D image data by selecting and segmenting morphological descriptors of interest using latent spacing embeddings of 3D convolutional autoencoders. We built a digital twin for training RL/DL models, TomoTwin, for full-field X-ray tomography (attenuation and phase contrast) that produces realistic images from 3D phantoms emulating data-quality response to instrument parameters. Utilizing these tools, we demonstrated computation time measurements for searching and characterizing porosity within 100 gigabytes (acquired over 2 hours of data-acquisition) of raw data in under 7 minutes on a single high-performance workstation. We deployed our software for processing such raw data on an evolving stream from a CT detector. At the Bionanoprobe (BNP) beam line, we demonstrated automated acquisition with regions of interest containing biological cells first detected and located from coarse scans and then subsequent fine scans acquired to improve local resolution.

We also demonstrated feature-detection and optimization of experimental parameters such as beam filtering and scanning parameters in real-time at 7-BM for an experiment to understand the morphological evolution of Bi particles in Sn-58Bi solders during the solidification process. We created a rapid event/anomaly detection framework that incorporates image representation models and clustering algorithms to assist users in visualizing, understanding, and guiding experiments. We applied this framework to multiple experiment datasets and submitted a comprehensive journal paper detailing our findings. To amplify its impact, we collaborated with various researchers and teams, demonstrating how our framework could enhance their research.

LDRD # 2021-0280	AI + Science
Project Title:	Benchmarking and Evaluation of the AI Accelerator Testbed at ALCF
Investigators:	Rajeev Thakur Murali K. Emani, Venkatram Vishwanath

Abstract

With the increasing use of machine learning and artificial intelligence techniques in applications, a number of vendors are developing hardware accelerators to speed up these computationally demanding AI tasks. The Argonne Leadership Computing Facility (ALCF) is building a testbed of next-generation AI accelerator hardware, currently comprising systems from Cerebras, SambaNova, Graphcore, Groq, and Habana. However, there is a lack of information on the comparative performance of these systems on benchmarks and scientific applications of interest to Argonne and DOE. This project aimed to evaluate

and benchmark the performance of these systems on representative workloads, which will both enable application scientists understand the performance characteristics and differences among these systems and also help guide the procurement of future systems for the testbed.

We have evaluated the performance of these AI accelerators on several deep learning (DL) primitives, benchmark models, and scientific machine learning applications. Specifically, we benchmarked their performance on four key fine-grained DL primitives (GEMM, Convolution, ReLU, LSTM), standard benchmarks from the MLPerf suite (UNet, BERT, ResNet-50), two scientific machine learning applications—X-ray Bragg diffraction peak detection (BraggNN) and drug discovery (Uno), and scaling benchmarks for collective communications and data-parallel training. We also performed another study focused on the performance of large language models on these accelerators.

LDRD # 2022-0006	AI + Science
Project Title:	Artificial Intelligence-Driven Electrospun Nanofiber Fabrication Using a Multi-Fidelity Machine Learning Framework
Investigators:	Debolina Dasgupta Bethany A. Lusch, Yuepeng Zhang

Abstract

Electrospinning is a material fabrication method that can produce nanometer to micron diameter fibers of various compositions for a wide variety of applications including filtration, fuel cells, batteries, and medical membranes. Because of the lack of *in situ* characterization of nanofiber morphology, the control of electrospun materials has largely remained empirical and limited industry’s adoption and scaling of the electrospinning process. The main objectives of the project was to advance the state-of-the-art in understanding the governing physics in the electrospinning process and achieving precise control of nanostructure morphology using artificial intelligence. Two new Argonne capabilities have been developed. A physics-based computational fluid dynamics (CFD) model of the electrospinning process has been developed within OpenFOAM that can shed light on the near needle behavior and its impact on fiber morphology. A multi-fidelity machine learning (MF-ML) framework has been developed that fuses information from expert user knowledge with volumes of data from CFD predictions and experiments to relate process parameters to near-needle jet structure and fiber morphology.

LDRD # 2022-0154	AI + Science
Project Title:	Inverse Microstructure Design for Enhanced Thermal and Electrical Conductivity
Investigators:	Angel Yanguas-Gil Maria K. Chan, Pierre T. Darancet, Noah H. Paulson, Subramanian Sankaranarayanan

Abstract

This proposal targeted the development of novel computational and machine learning capabilities to accelerate the development of composite materials. As part of this work, the team in the Applied Materials Division focused on the development of novel simulation approaches of the thermal and electric transport of composite materials, as well as of fast surrogate models that can predict the transport properties from experimentally available parameters, while the team at the Center for Nanomaterials focused on the development of new models capable of predicting the microstructure of materials for various additive manufacturing techniques. One of the outcomes of this project was the development of a new software, cowalker, that can simulate the electrical and thermal properties of composite materials in a massively parallel fashion. We used Argonne's LCRC as the testbed to demonstrate the ability to run on high performance computing facilities. This software was used to generate datasets that were used to build surrogate models for the effective transport properties of composite materials including non-idealities present in real materials such as contact and interfacial thermal resistance. This model was used to demonstrate why metal-carbon composites underperform, which could be used to inform DOE programs such as CABLE, which aimed at developing better conductors to reduce transmission losses in the grid and improve the efficiency of electric vehicles.

LDRD # 2023-0376	AI + Science
Project Title:	Advanced Computing Expedition 2023
Investigators:	Michael Papka Ian T. Foster, Salman Habib, Valerie E. Taylor

Abstract

This Expedition supports investigator-led efforts targeted at Argonne-related artificial intelligence/machine learning (AI/ML) or computational science problem areas, focus on the use of Argonne Leadership Computing Facility’s (ALCF’s) AI Testbed, and, ideally, promote collaboration across Argonne. Nine expedition research projects were supported for FY 2023.

Biological and Environmental Science

LDRD # 2022-0047	Biological and Environmental Science
Project Title:	Using Metabolic Modeling to Develop Stable Synthetic Microbial Communities
Investigators:	Pamela B. Weisenhorn Christopher S. Henry

Abstract

This project compared machine learning (ML) models and metabolic models (MM) based on multiple genomes and carbon sources. These models were generated with both publicly available and private datasets and were validated through the creation of a self-driving laboratory (SDL) to study carbon-source utilization. Validation was performed with E.coli strains. The team has successfully applied the SDL pipeline to measure 29 carbon utilization phenotypes in 48 selected E.coli environmental isolates and predicted these same phenotypes using ML and MM methods. Results showed that ML models consistently display better performance than MM models and the addition of new SDL data improved overall ML accuracy. The team plans to explore using combined classifiers,

hybrid classifiers, fitting MM to experimental data, and identifying the next set of isolates to test in the upgraded SDL pipeline.

LDRD # 2023-0103	Biological and Environmental Science
Project Title:	Evaluating Climate Intervention Strategies through Cloud Albedo Modification
Investigators:	Yan Feng Dimitrios Fytanidis, Virendra P. Ghate

Abstract

As the race between climate change and the required mitigation steps will be a close one, there is an emergent need of predictive tools for making rational considerations on feasibility, effectiveness, and impacts of climate intervention. Progress has been made in developing high-resolution Earth System modeling (ESM) frameworks such as at convection-permitting scales, but we are still far from realizing the prediction objectives, especially related to the aerosol and cloud processes. More efforts are needed in accurate representation of the unresolved aerosol-cloud interactions in current Earth System modeling (ESMs). We addressed the fundamental challenges with high-fidelity numerical models in evaluating the consequences of aerosol-induced cloud albedo change by tackeling cloud physics uncertainty through studying aerosol-cloud interactions and micro-processes with high-fidelity models (nek5000/nekRS) at fundamental scales (100 to10-3 m), and improving process-scale understanding of aerosol-cloud interactions under turbulent conditions that would advance the predictability of Earth System models. We have completed the Nek5000 development in cloud droplet condensation, built on the model physics design and construction from previous project. Using the developed model, we have performed a series of simulations to explore the parameter space and sensitivities of the droplet-droplet and particle-particle interactions.

LDRD # 2023-0344	Biological and Environmental Science
Project Title:	Pilot Platform for Earth System Science
Investigators:	Pamela B. Weisenhorn

Abstract

The goal of this project was to develop a pilot infrastructure that supports the Environmental Systems Science's model-experiment (ModEx) framework and the development of artificial intelligence and machine learning approaches by increasing access and interoperability of data and modeling and analysis tools spanning Biological and Environmental Resarch (BER) science. This project successfully developed back-end services including a job runner and user account support. Additionally, we completed a JupyterHub web-based graphical user interface with web-based account access and widgets to allow for site selection to input pre-processed atmospheric data, web-based parameter modification, and point-based Earth Land Model (a component of DOE's E3SM model) simulations.

Clean Energy and Sustainability

LDRD # 2022-0017	Clean Energy and Sustainability
Project Title:	Decarbonizing Transportation using Hydrogen Fuel for Combustion Engines
Investigators:	Riccardo Scarcelli Muhsin Ameen, Christopher Powell

Abstract

On the path to decarbonized transportation, the only viable solution for combustion engines to compete against electrification consists of using low- or zero-carbon fuels. Hydrogen (H_2) is the most attractive of these fuels, nevertheless predictive computational tools that can speed up the design of advanced hydrogen engines are not available. The effort leveraged computational fluid dynamics (CFD), exa-scale computing, and X-ray diagnostics to develop a predictive models for hydrogen injection and mixing. Direct Numerical Simulations (DNS) performed with the spectral element code Nek5000 and X-ray data were fused into the development of a computational fluid dynamics gaseous mixing model to improve the accuracy of Reynolds-Average Navier-Stokes (RANS) simulations using the commercial solver CONVERGE, which is routinely used by industry for engine simulations. The novel mixing model was implemented in CONVERGE using user defined functions (UDFs). The accuracy of the developed mixing model was initially assessed through benchmark against DNS and later validated against X-ray radiography from the Advanced Photon Source. The project was successfully completed. Among the several deliverables, the major ones: 1) the X-ray diagnostics was improved to visualize Helium gaseous jets and Helium is a very good surrogate for Hydrogen; 2) Nek5000 was developed to account for full compressibility effects and moving mesh boundaries; 3) RANS simulations of gaseous jet mixing were significantly improved through the development and implementation of this novel mixing model.

LDRD # 2022-0031	Clean Energy and Sustainability
Project Title:	Low-Temperature Membrane Reactor for Electrochemical Conversion of Carbon Dioxide
Investigators:	Tae H. Lee Uthamalingam Balachandran, Di-Jia Liu, Beihai Ma

Abstract

The purpose this project was to develop protonic solid oxide electrolytic cell (P-SOEC) to produce ethylene from carbon dioxide and water using renewable electricity at temperature 100-300°C. We synthesized and characterized several proton conducting electrolyte materials including cesium hydrogen phosphate, tin pyrophosphate, and their composites. For the electrocatalyst development, we designed and synthesized both carbon dioxide reduction-reaction (CO_2RR) cathode and oxygen evolution-reaction (OER) anode catalysts. Then, we synthesized and characterized more composites based on cesium hydrogen phosphate and silicon pyrophosphate. Among the composites, the composite with 70 percent cesium hydrogen phosphate and 30 percent silicon pyrophosphate showed even higher proton conductivity than that of the cesium hydrogen phosphate at temperatures between 135 and 240 degree Celsius. The composite showed high proton transfer number higher than 0.98,

however, the electrolyte deformed heavily during measurement due to compression of mechanical seal. The deformation of the electrolyte might be partly due to partial melting of the electrolyte at high water partial pressure. This issue needs to be resolved for further application of the electrolyte for the P-SOEC cell fabrication.

For the anode development, we prepared several cobalt metal organic framework (MOF) derived OER catalysts using an electrospin method followed by high temperature activation. We achieved promising performance with the cobalt-spinel catalyst derived from cobalt imidazolate framework. The measured current density of the cobalt-MOF was comparable to that of the iridium black catalyst. Our OER catalyst performance shows an overpotential within 30 millivolt to that of iridium, showing promise as a replacement for iridium. The current-voltage measurement of new catalyst was integrated into polymer electrolyte membrane (PEM) water electrolyzer and demonstrated excellent performance. Some platinum group metal (PGM)-free OER catalysts have been tested. An OER catalyst showed an overpotential of 354 millivolt at 10 milliampere per unit area (mA/cm^2), meeting the performance target.

For the cathode, we designed, synthesized, and characterized copper-X (X=tin, aluminum, zinc) based carbon dioxide reduction-reaction catalysts with Faradaic efficiency higher than 60 percent in the liquid electrolyte. Multiple carbon dioxide reduction-reaction catalysts were developed since the project inception, with new discovery on selective carbon dioxide to triatomic carbon chemical conversion. Catalysts have been integrated into membrane electrode assembly and demonstrated high current density and high selectivity towards diatomic carbon chemical conversion. Copper based catalysts tested in the electrolyzer cell achieved current density of 300 milliampere per unit area with near 80 percent of total diatomic carbon conversion Faradaic efficiency.

LDRD # 2022-0063	Clean Energy and Sustainability
Project Title:	Smart Control of Hydrogen-Compatible Combined Heat and Power Microturbines
Investigators:	Debolina Dasgupta Munidhar S. Biruduganti, Sibendu Som, Sarang Supekar

Abstract

Combined heat and power (CHP) includes a set of technologies that can produce electricity and heat simultaneously and offer advantages such as reduced costs, energy resilience, and higher overall energy efficiency. The electric grid is rapidly changing with the introduction of variable renewable generation. Combined heat and power systems will play a role by providing grid stability. Microturbines can provide both cooling and heating for a variety of commercial and industrial applications. Combined heat and power systems need to transition to high hydrogen content blends and pure hydrogen to meet 2050 decarbonization goals. Our project provides the knowledge, analyses and tools needed to control such microturbine systems with hydrogen fuel blends. A computational fluid dynamic driven digital twin for a typical microturbine combustor has been developed to define its operability map under variable load conditions and blends of natural gas and hydrogen. The operability map is developed as a surrogate model for the system using machine learning. Using this surrogate model, development of a smart

control framework to transition across loads while maximizing revenue, and ensuring high efficiency and low emissions of nitrogen oxides is underway.

LDRD # 2022-0067	Clean Energy and Sustainability
Project Title:	Electrochemical Upcycling Pathways of Plastic Wastes to Value-Added Monomers
Investigators:	Donghyeon Kang

Abstract

The objective of this project was to develop an innovative electrochemical depolymerization method for polyethylene terephthalate (PET), producing valuable monomers. We deciphered the fundamental chemistry behind PET's chemical breakdown and engineered a groundbreaking electrochemical depolymerization procedure. Our project exemplified a budget-friendly and energy-conscious approach to PET depolymerization, employing an innovative membrane-associated electrochemical method. Vital initial data corroborate the feasibility of this venture. Furthermore, we are actively engaging with multiple industrial partners to facilitate the seamless transfer and commercialization of this groundbreaking technology.

LDRD # 2022-0077	Clean Energy and Sustainability
Project Title:	Development of a Reinforcement Learning Environment to Advance Autonomous Control of Next-generation Nuclear Power Plants
Investigators:	Akshay J. Dave

Abstract

The operational and maintenance expenditures for nuclear power facilities significantly surpass those of natural gas plants, often by an order of magnitude. In the quest to mitigate these costs, the concept of implementing autonomous control has long been discussed but not actualized within the realm of advanced nuclear reactors. Recognizing this gap, our project hinged on a foundational tool—the System Analysis Module (SAM) developed by Argonne National Laboratory—as the computational cornerstone for modeling the complex dynamics of these power plants. We innovatively integrated SAM into a cutting-edge reinforcement learning (RL) framework. Our team's pioneering efforts led to creation of a sophisticated framework tailored for the control systems of advanced nuclear reactors, culminating in a comprehensive solution that harnesses RL to engineer physics-constrained, multi-objective agents. These agents have successfully demonstrated autonomous supervisory control during critical power transitions, marking a significant leap in optimizing the operations of such intricate systems. Our findings not only showcase the practical applications of RL in enhancing the efficiency of nuclear power plants but also set a precedent for its broader application in complex industrial systems.

LDRD # 2022-0078	Clean Energy and Sustainability
Project Title:	Ultra-High Temperature Materials and Associated Manufacturing for the Decarbonization of Process Industries
Investigators:	Dileep Singh

Abstract

Renewable energy sources such as solar are becoming key technologies that can aid in industrial decarbonization. Solar energy is not only used for carbon-free power generation. Solar heat can be used by process industries, especially those requiring very high temperatures. Those include steelmaking, cement calcination, mineral processing, and production of solar-energy-produced fuels like hydrogen, all mitigating carbon release. Many of those applications require very high temperatures (greater than 1000 degrees Celsius). Use of solar heat requires thermal systems that capture and deliver the heat and operate reliably at extreme conditions. Some such components are volumetric receivers (capturing heat), micro-channel reactors (solar fuel production), heat exchangers, and thermal storage systems. We have developed capability to fabricate ultra high temperature ceramic materials and composites using digital light processing technique additive manufacturing approach. We have demonstrated fabrication of complex structures of various ultra high temperature materials including silicon carbide, mullite and corderite using appropriate precursors and densification techniques.

LDRD # 2022-0083	Clean Energy and Sustainability
Project Title:	Sustainable Poly(Ionic Liquids) Towards a Circular Carbon Dioxide Economy
Investigators:	Wei Chen Matthew Tirrell, Jie Xu, Yuepeng Zhang

Abstract

In this initiative, we are at the forefront of polymer science, creatively utilizing carbon dioxide (CO_2) and bio-based by-products to forge carbon-negative, sustainable polymers. Our objective was the synthesis of poly(limonene carbonate) (PLimC) polymers, which stand out for their capability for chemical recycling to monomers while also enabling CO_2 capture and long-term storage. This dual functionality is a significant stride towards promoting a circular CO_2 economy.

We successfully produced PLimC-based gas separation membranes. Our initial testing phase revealed promising results, with both CO_2 permeability and CO_2/N_2 selectivity metrics aligning with industry benchmarks, akin to the renowned Pebax membranes by Arkema. In parallel, we have formulated an innovative PLimC-based concrete additive, remarkable for its water solubility and compatibility with conventional Portland cement. This eco-friendly substitute is poised to revolutionize traditional cement use, fulfilling the roles of a binder, superplasticizer, and water reducer without diminishing the mechanical integrity of the concrete. Through ongoing refinement, we foresee an increase in the mechanical strengths of polymer-modified concrete, thereby advancing the construction industry's move towards sustainable practices.

The implications of our research with PLimC polymers transcend academic pursuits, offering practical, scalable solutions to some of the most pressing environmental issues of our time. Our commitment to

innovation melds with pragmatic application, signaling the dawn of a new age in environmentally conscious materials engineering.

LDRD # 2022-0161	Clean Energy and Sustainability
Project Title:	Pathways for Low-Carbon Aviation
Investigators:	Dominik A. Karbowski Amgad A. Elgowainy, Nirmit Prabhakar

Abstract

This project has resulted in new capabilities that enable the evaluation and optimization of various technology pathways for low-carbon aviation, including electrification, using (1) Aeronomie, a recently developed aircraft/flight simulation tool, to design the propulsion systems, calculate the energy consumption and tank-to-wheel emissions for a given mission and (2) GREET for Life-cycle analysis of various electricity, hydrogen, and SAF pathways.

Aeronomie can now model various alternative energy propulsion for one aircraft, the commuter twin-engine plane Tecnam P2012. The Tecnam model in Aeronomie now comes in a baseline Internal combustion engine (ICE)-powered version, as well as battery-electric, hybrid-electric and fuel cell electric versions. The baseline version model energy consumption was validated against published manufacturer data. The modeling of the alternative energy propulsion systems also resulted in elemental blocks that will make similar modeling for other airplane much easier in the future. In addition, GHG emissions were added to the list of outputs generated by Aeronomie, using emission factors from GREET.

LDRD # 2022-0176	Clean Energy and Sustainability
Project Title:	Sustainable Conversion of Carbon Dioxide to C1+ and C2+ Fuels via Functional 2-dimensional Electrocatalysts
Investigators:	Zachary D. Hood Alex B. Martinson, Jianguo Wen

Abstract

Carbon dioxide reduction is recognized as a strategy to reduce atmospheric pollution associated with climate change. Although multiple strategies have shown carbon dioxide reduction to liquid fuels, two hurdles still exist for the most recent catalysts for this reaction; namely, the preparation of the catalysts are generally complicated and their selectivity towards specific products requires better control. Building from a report in the literature that could lead to breakthroughs in state-of the-art electrocatalysts for carbon dioxide reduction to liquid fuels, we developed a simple method to prepare copper-based catalysts for the carbon dioxide reduction reaction and evaluate their selectivity towards production of various fuels. The two-dimensional ultra-thin layered transition-metal-carbide or nitride exfoliated flakes known as MXenes were our choice for electrocatalyst substrate.

We synthesized greater than 25 grams of MXene and completed surface functionalization with copper using liquid-based approaches as well as atomic layer deposition (ALD). We performed basic characterization in the Materials Engineering Research Facility (MERF). We also evaluated the carbon dioxide reduction over copper-based MXene catalysts, assessing the products with the gas chromatography–mass spectrometry (GC/MS, and assessing the Faradaic Efficiency and partial current density of carbon dioxide reduction. Our team is leveraged X-ray photoelectron spectroscopy (XPS) to assess the surface chemistry of all electrocatalysts and Scanning/Transmission Electron Microscopy (S/TEM) images and energy dispersive x-ray spectroscopy (EDS) elemental mappings at the Center for Nanoscale Materials.

LDRD # 2022-0212	Clean Energy and Sustainability
Project Title:	Artificial Intelligence-based Discovery for Carbon-Free Energy
Investigators:	Rajeev Assary Larry A. Curtiss, Ian T. Foster, Stephen J. Klippenstein

Abstract

We have developed and demonstrated an artificial intelligence guided computational platform to enable molecular discovery of liquid organic hydrogen carriers (LOHC) for hydrogen storage, and liquid organic hydrogen carriers for redox flow batteries. The established selection or scoring criteria scores the suitability of liquid organic hydrogen carrier candidates based on structural features of the molecules; we identified 11 descriptors based on descriptions of good liquid organic hydrogen carrier candidates. We developed an accurate benchmark database of of 100 molecules with desired physical and chemical properties for LOHC. Using HPC resources and preliminary selection criteria and cheminformatics-based similarity, we performed molecular screening of large chemical space of 177 Billion Molecules (GDB 17 dataset). We then performed 3000 DFT calculations for identifying dehydrogenation energetics and identified new molecular structures for LOHC and developed an active criteria with 21 descriptors for large scale screening including accurate dehydrogenation enthalpies (40-70 kJ/mol). Finally, we developed a kinetic scoring model to characterize the structure of the reaction sites and the types of forming and breaking bonds in the reaction pathway.

LDRD # 2022-0215	Clean Energy and Sustainability
Project Title:	Charting Synthetic Pathways of Sulfide Solid-State Electrolyte Materials Through Identifying Key Reaction Intermediates in the Solution-Phase Synthesis
Investigators:	Kamila M. Wiaderek Nigel H. Becknell, Tao Li, Lei Cheng

Abstract

In this study we aimed to explore the synthesis of sulfide solid-state materials through an integrated experimental and computational methodology. We employed Pair Distribution Function (PDF) and Raman analyses along with Molecular dynamics (MD) simulations and Density Functional Theory (DFT) calculations. We were focusing on early, solvothermal stages of the reactions during the synthesis process, where species are in the ionic solvated state. To enable such studies our team designed and

developed a multicell sample environment compatible with scattering data collection and solvothermal process. As a part of development we needed to overcome the data processing issues including proper data integration and corrections as a signal from these highly diluted samples was largely overwhelmed by solvent and container signals that changed with temperature. We were able to isolate signal from precursors and products *in situ*, with high enough quality that allowed further processing through modeling. The sample environment and data acquisition methodology will be available to structural science group users within Advanced Photon Source after the upgrade.

LDRD # 2022-0220	Clean Energy and Sustainability
Project Title:	Moderate Modulus, High Voltage Organic Cathodes for All-Solid-State Lithium-Ion Batteries
Investigators:	Chen Liao Hieu A. Doan, Lihong Gao, Zachary D. Hood, Noel J. Leon

Abstract

We examined organic electrodes with designed machine learning feedback loop for design of high energy high voltage lithium-ion batteries with a focus on the design of organic electrodes and the demonstration of their performance. We were able to illustrate new multielectron redox centers as well as materials demonstrating high voltage using anion insertion mechanisms. The organic materials suffered from degradation mechanisms other than dissolution, the EPR results showed the multielectron charges (as well as coulomb repulsion) caused the material to degrade rapidly.

LDRD # 2023-0360	Clean Energy and Sustainability
Project Title:	Development of Highly Efficient/Durable Water Electrolysis Catalysts by Powder Atomic Layer Deposition
Investigators:	Donghyeon Kang Jeffrey W. Elam

Abstract

The primary aim of this project was to demonstrate a powder coating process via atomic layer deposition (ALD) for the economic synthesis of Iridium oxide (IrO_2) oxygen evolution catalysts for use in water electrolyzers, to achieve future opportunities in hydrogen production research and development. Throughout the project duration, we successfully demonstrated the ALD coating processes for stable Iridium oxide (IrO_2) catalysts on conductive substrates.

LDRD # 2023-0362	Clean Energy and Sustainability
Project Title:	Novel High Performance Anion Exchange Membranes for Clean Hydrogen Production via Water Electrolysis
Investigators:	Meltem Urgun Demirtas

Abstract

Clean hydrogen production via anion exchange membrane water electrolysis has the unique advantages of using low-cost hydrocarbon membranes and non-noble metal catalysts. Unfortunately, current state-of-the-art anion exchange membranes still lack sufficient ion conductivity and durability to achieve the goals set by the Department of Energy with performance of more than 2.0 amperes per square centimeter membrane area at a cell voltage of 1.8 volts with less than 3 millivolts per kilohours degradation rate. We designed novel anion exchange membranes with rigid aromatic backbones and cross-linkable functional groups. Due to the very stable chemical structures, the anion exchange membranes were allowed to have much higher ion exchange capacity of more than 3 millimoles per gram of membrane than those reported in literature, leading to both high conductivity and high stability and durability. The team targeted the development of non-fluorinated anion exchange membranes and ionomers for water electrolysis with lower costs and higher performance and durability.

Climate and Energy Action

LDRD # 2022-0195	Climate and Energy Action
Project Title:	Climate Change Adaptation Strategies for Great Lakes Communities
Investigators:	Lawrence Lewis Elizabeth R. Bolton, Carmella A. Burdi, William Pringle, Vishwas Rao, Scott O. Schlueter, Jiali Wang

Abstract

The drive to reduce climate impacts on those most vulnerable to climate change worldwide creates an urgent need for world class climate adaptation decision support with associated high-quality tools and technical assistance. Communities along the Great Lakes are at risk from several climate-related hazards, which can result in economic instability and community development challenges. Adapting to climate change requires understanding the systemic risks, which combine extreme weather hazards, infrastructure characteristics, and communities' socio-economic status. This research enables a multidisciplinary approach combining environmental, infrastructure, and social sciences to develop a systemic risk analysis framework and climate change adaptation decision tool that will inform communities development strategies. These capabilities are needed by federal agencies (Department of Energy Office of Electricity for recovery investment, Housing and Urban Development for community development) and local governments for informing urban planning initiatives.

Initially, this project focused on the development of downscaled climate date at 4km^2 to inform infrastructure adaptation. This climate data was used to develop fragility curves defining the vulnerabilities of lifeline infrastructure to anticipated changes of the weather landscape. Finally, climate change data and fragility were combined to conduct GIS analysis to define potential infrastructure system cascading failures.

LDRD # 2022-0199	Climate and Energy Action
Project Title:	Towards Neighborhood Scale Climate Simulations using Artificial Intelligence and Accelerated Graphics Processing Units (GPUs)
Investigators:	Jiali Wang Piyush Garg, Haochen Tan, Xingqiu Yuan

Abstract

High spatial resolution climate information is needed to assess stakeholder-relevant local impacts of significant changes in climate processes. However, developing such high resolution data takes significant computational resources and wall-clock time. We have successfully implemented the state-of-the-art Graphics Processing Units (GPU) version of DOE's global scale high resolution (3km) atmosphere model, called SCREAM on all GPU high performance computers at the Argonne Leadership Computing Facility (ALCF), including ThetaGPU and Polaris. Such modeling capability allows us to generate sufficient training data for machine learning and artificial intelligence (ML/AI) development. We have also developed model calibration tools to calibrate the physics parameters in the SCREAM model. This allows the model to achieve better performance with higher confidence when used for future climate studies.

LDRD # 2022-0205	Climate and Energy Action
Project Title:	Impacts of Climate Change on Intensity, Duration, and Frequency of Drought Events at Different Geographic Scales
Investigators:	Vishwas Rao Mustafa S. Altinakar, Julie Bessac, Brandi L. Gamelin

Abstract

In this project we develop Machine Learning (ML) Algorithms to identify space-time extents of drought and investigate how they change under future climates. This is especially useful for resilience planning and mitigation strategies in relation to wildfire activities, agriculture management, hydropower management etc. We use ML Techniques to identify drought features and produce drought spatiotemporal clusters.

Additionally, we applied ML techniques to identify primary modes of large-scale drought variability in the United States. We also identified their northern hemisphere atmospheric and oceanic drivers.

Our approach isolated specific drought patterns in Argonne’s new drought index based on vapor pressure deficit (VPD): the Standardized VPD Drought Index (SVDI). This work identified oceanic and atmospheric drivers of drought variability to improve the predictability of future drought events to mitigate drought risks related to crop loss, reduced hydrological resources and wildfire issues.

LDRD # 2022-0207	Climate and Energy Action
Project Title:	Translating Climate Extreme Event Projections into Grid Planning Models: Role of Long Duration Energy Storage in Optimizing Cost-Reliability Tradeoffs Under Power System Uncertainty
Investigators:	Todd Levin Veerabhadra R. Kotamarthi

Abstract

The goal of this project was to incorporate projections of future climate data into grid planning models to ensure that we are planning our electricity systems to be reliable in the face of increasingly warm and extreme weather conditions.

We have (a) retrieved the nation-scale climate data for the early- mid- and end-century periods; (b) converted climate data from the Texas region to establish corresponding grid input data (hourly wind + solar availability and load projections) for both early- mid- and end of century; (c) utilized public HIFLD data to establish a nation-scale transmission network. We completed (a) data conversion modules to translate weather data to generate wind and solar generation availability, and augment electricity demand projections; (b) algorithms to identify periods of extreme operating conditions and capture these periods in representative scenario selection; (c) script to arbitrarily aggregate/scale transmission data into a regional DCOPF formulation or transport model representation.

LDRD # 2022-0210	Climate and Energy Action
Project Title:	Community and Infrastructure Adaptation to Climate Change: An AI-Driven Research Tool for Community Planners
Investigators:	Duane R. Verner Joshua D. Bergerson, Yan Feng, Leslie-Anne Levy, Tanwi Mallick, John T. Murphy

Abstract

Critical infrastructure systems throughout the U.S. are increasingly at risk due to systemic underinvestment and intensifying natural hazards related to climate change. Research on climate change, impacts on critical infrastructure, and infrastructure adaptation constantly evolves and is published at a blistering pace. This makes it nearly impossible to review and use the findings to inform climate adaptation decision-making.

Decision makers require actionable, understandable guidance on projected future location-specific climate conditions, the potential impacts of these future conditions on critical infrastructure systems, and remedial climate change adaptation strategies to enhance the resilience of these systems and reduce their future disruptions. But with the current resources, that is extremely difficult, if not impossible.

We have developed the Community and Infrastructure Adaptation to Climate Change (CIACC) tool to label many documents using a weak supervision-based natural language processing technique that uses semantic similarity between categories and documents. This method allows the labeling process to scale across multiple GPUs in a computing cluster at the Argonne Leadership Computing Facility.

The CIACC climate adaptation decision support tool leverages this unique combination of cutting-edge and interrelated capabilities. The tool can read millions of articles a day and track the evolution of topics over time, offering a solution to the challenge decision makers face in wrapping their arms around all the climate change and infrastructure impact research that exists. The Argonne tool analyzes research published on climate change, impacts on critical infrastructure, and adaptation and boils it down to manageable and understandable reports.

The tool offers decision makers cutting-edge and actionable information on climate hazards, threats to critical infrastructure, and climate adaptation best practices, helping them better safeguard systems and communities.

Energy Security and Sustainability

LDRD # 2022-0046	Energy Security and Sustainability
Project Title:	Advanced Reactor Design and Economics Analysis Framework
Investigators:	Nicolas E. Stauff William N. Mann, Paul K. Romano, Ling Zou

Abstract

Designing an advanced nuclear reactor involves close interactions between experts from many technical fields, including economics experts who provide information on component costs and on market analyses. During this project, a multidisciplinary team developed and demonstrated the use of the Workflow and Template Toolkit for Simulation (WATTS) framework. WATTS brings together reactor design tools with economics and energy market modeling tools, streamlining collaborations between different experts through the exchange of data within the different codes and facilitating the use of rigorous sensitivity and optimization approaches on complex calculation workflows. The WATTS platform has been released under an open-source license.

Energy Storage

LDRD # 2022-0020	Energy Storage
Project Title:	Accelerated Discovery of Entropy Stabilized Metal Fluorides as Conversion-type Cathode Materials via Combinatorial Synthesis and Computational Modeling
Investigators:	Jae Jin Kim Eungje Lee, Peter Zapol

Abstract

The concept of high-entropy materials, based on integrating multiple redox active cations into a specific crystal structure to enhance the stability of the crystal lattice, has opened an exciting opportunity to discover advanced materials with tailored properties. The objective of this project was to efficiently identify and synthesize novel entropy-stabilized, high-performing metal fluorides, as a conversion-type cathode material, by using data-guided prediction from advanced computational methods. Multi-cation metal fluorides, designed and synthesized in this work, showed distinct redox potential and high initial coulombic efficiency, different from the individual single-cation phases. The former feature could open a unique opportunity to tune working voltage, and the latter could imply that entropy-stabilization may benefit reversible conversion reaction. In addition, computationally guided material design enabled to increase materials stability and average voltage for non-equimolar earth abundant metal alloy compositions.

LDRD # 2022-0196	Energy Storage
Project Title:	Scalable Membrane Architecture with Robust Nanoscale Channels Advancing Low Cost Redox Flow Batteries
Investigators:	Lu Zhang Shrayesh Patel

Abstract

This project aimed to repurpose engineered plastics into cost-effective, high-performance membranes for redox flow batteries. Membranes are crucial for redox flow batteries, influencing both performance and system cost. Our focus was on establishing a scalable method to create low-cost, high-performance mesoporous and dense membranes from inexpensive plastic stock or waste. We demonstrated the fabrication of sulfonated mesoporous membranes from Noryl (a blend of poly[phenylene oxide] and high-impact polystyrene), suitable for extended cycling in alkaline quinone flow batteries. Then we concentrated on dense membranes, achieving significant progress using polystyrene (PS) plastic waste from utensils, coffee lids, and plates to create ion exchange membranes. These membranes exhibit excellent conductivity, permeability, and flow cell cycling performance. Our recent techno-economic analysis (TEA) indicates an attractive cost of $2-3 per m2 for both porous and dense membranes, representing a breakthrough in reducing redox flow battery costs.

Hard X-ray Sciences

LDRD # 2020-0071	Hard X-ray Sciences
Project Title:	Intelligent Ptychography Scan via Diffraction-Based Machine Learning
Investigators:	Zichao Di Junjing Deng, Yi Jiang

Abstract

In this project, we developed machine learning (ML) techniques to achieve intelligent data acquisition in X-ray ptychography experiments. First, we proposed a physics-informed unsupervised classification algorithm that is performed prior to reconstruction and removes data outside the region of interest based on the multimodal features present in the diffraction patterns.

This capability consequently reduces computational time dramatically while preserving reconstruction quality. Subsequently, we extended the application of the proposed unsupervised algorithm used for post-processing to data acquisition upfront. The newly developed algorithm introduces automatic guidance for data acquisition. Our algorithm first directs the scan point to actively search for the object of interest within the field of view. Subsequently, it intelligently scans along the perimeter of the sample, strategically acquiring measurements exclusively within the boundary of the region of interest. By employing this approach, we demonstrate the reduction in the number of measurements required to obtain high-resolution reconstruction images, as compared to conventional raster scanning methods. The capabilities achieved in this project to automatically identify and classify sample features will allow real-time “informative” feedback and is the key to studies of small features in a large sample volume.

LDRD # 2021-0005	Hard X-ray Sciences
Project Title:	High Pressure Material Characterization in 3-Dimensions Using X-ray Diffraction-Contrast Computed Tomography
Investigators:	Hemant Sharma Chihpin Chuang, Yue Meng, Hemant Sharma

Abstract

This project has led to the development of two flagship techniques at the Advanced Photon Source (APS) for studies of materials under high pressures: (a) High-Pressure X-ray Diffraction Computed Tomography (HP-XRD-CT) and (b) point-focus High Energy Diffraction Microscopy (pf-HEDM). Both these techniques push the limits of investigative techniques as follows:

(a) HP-XRD-CT goes further than powder-diffraction, which yields average information across specimens, by resolving local pressure and phase distributions in the specimens. Although the XRD-CT technique is not new, we have developed both hardware and software to successfully apply it to high-pressure experiments. On the hardware side, we developed two novel high-pressure devices, which allow for enough rotation for collecting high-quality diffraction data. In addition, we developed software routines employing Argonne's High-Performance Computing resources (including Laboratory Computing Resource Center) to obtain high-quality reconstructions, whilst keeping up with the projected Advanced Photon Source Upgrade (APS-U) data rates.

(b) pf-HEDM allows for even more detailed information: grain-resolved pressure and phase distributions. This is a first-of-its-kind technique that was developed to resolve the crystallographic properties of individual grains. Such detailed information about materials allows users to better understand and model the behavior of materials under dynamic conditions.

Both these techniques allow for studies of nanometer- to micrometer-sized grains, opening new possibilities for science. These techniques will be available for users after the APS-U at the High Pressure (16ID), and High Energy (1ID and 20ID) beamlines. (ID: Insertion Device)

LDRD # 2021-0012	Hard X-ray Sciences
Project Title:	Coherence-Enhanced Dark-Field Imaging for Structural Heterogeneity in Materials
Investigators:	Stephan O. Hruszkewycz Matthew J. Highland, Zahirul Islam, Xianbo Shi, Zhan Zhang

Abstract

The specific need we address in this project is rapid X-ray imaging of large fields-of-view with sensitivity to structural defects in materials that is not possible with current methods, which can take minutes and are entirely insensitive to lattice structures that manifest as complex-valued objects with respect to the Bragg diffraction condition. Our project developed proof-of-concepts for coherent X-ray lens-based full-field diffraction imaging that enables contrast enhancement of critical features in bulk materials via a wide range of image defocus between the back-focal plane and the image plane of the lens. This is a novel extension of dark-field X-ray microscopy that enhances signatures of structural heterogeneities that are encoded in the diffracted Bragg wave-front and made visible by contrast mechanisms of coherent wave propagation.

So far, the additional instrumentation at Sector 6 of the Advanced Photon Source has been acquired and set up, and initial measurements of carefully chosen materials have been made with the partially coherent beam as initial proofs of concept. In addition, a robust DFXM software package has been built to simulate the propagation of the Bragg-diffracted wave through an x-ray lens system through the fractional Fourier transform, customized to the newly installed DFXM infrastructure at Sector 6. This software has been used for initial demonstrations of the evolution of structural kinks in crystalline materials.

LDRD # 2021-0059	Hard X-ray Sciences
Project Title:	High-Energy X-ray Compton Imaging of Light Elements as Multimodal Technique for Li Battery Research and Beyond
Investigators:	Orlando Quaranta

Abstract

We designed and built a Compton imaging system, for *in situ* determination of lithium kinetics and plating during fast charging in a lithium-ion battery, and evaluated the feasibility of using a transition edge sensor detector for spectroscopic Compton imaging. Compton scattering, which is an inelastic scattering process of photons from electrons, becomes the dominant process for high-energy X-rays at scattering angles of 90 degrees. It has a high sensitivity to low atomic number elements such as lithium. Previously, neutron scattering has been the principal light element detection tool, but the low absorption of high-energy X-rays allows probing more deeply into complex devices. Diffraction of X-rays scattered into the forward direction allows for atomic-level structure analysis as well. Therefore, with both forward and side-scattered X-rays, unique simultaneous low Z element contrast imaging and structure analysis is possible. High spatial resolution Compton imaging was be enabled by advances in X-ray focusing and Transition Edge Sensor (TES) detector technologies. This technique is widely applicable to dynamic and static processes, particularly in battery research, enabling collaborations with energy storage consortiums.

A proof-of-principle experiment on multimodal technique (combining Compton scattering and high-energy X-ray diffraction) was carried out at 11-ID-C on lithium-ion battery related materials. A “donut-like” artifact made of lithium-foil was placed inside a coin-cell between graphite anode and nickel-manganese-cobalt-oxide (NMC)-cathode layers. Analysis of the S-parameter of Compton profile functions in the layer helped to identify Lithium-presence or absence at any point in the layer.

Additionally at 1-BM-C, high-resolution Compton spectroscopy was developed using a TES detector in comparison with existing beamline detectors. Proof-of-principle Compton scattering measurements and spectroscopic analysis were conducted on materials related to lithium-ion battery research demonstrating high-sensitivity of Compton profiles to low-Z elements, particularly lithium. Pouch cells, representative of real coin cell batteries, were prepared for experiments at 1-BM-C. Compton spectra collected at 1-BM-C both by the TES instrument and a Vortex detector allowed measurement of the relative quantity of lithium in the various positions of the pouch. This experiment proved the ability of the instrument to perform real-time scans of samples, a crucial ability for any modern X-ray sensor at beamlines. This new variant of the instrument is also able to measure well in the hard X-ray regime.

LDRD # 2021-0090	Hard X-ray Sciences
Project Title:	AutoPtycho: Autonomous, Sparse-sampled Ptychographic Imaging
Investigators:	Mathew J. Cherukara Junjing Deng, Zichao Di, Ross J. Harder, Yi Jiang, Charudatta M. Phatak

Abstract

AutoPtycho is an end-to-end AI-driven approach to data inversion, data abstraction and experimental control for Ptychography an advanced imaging technique used at multiple beamlines at the Advanced Photon Source (APS). For the first time, this project has enabled real-time ptychographic imaging at the APS by combining high performance computing (e.g. Polaris) + AI@Edge. This project has also resulted in the first autonomous steering of a scanning beamline experiment, a pre-trained AI model steered the experiment to maximize information gain. Both of these advances are being turned into production software for use at the APS.

Imaging and Detection of Signatures

LDRD # 2022-0030	Imaging and Detection of Signatures
Project Title:	High-Energy X-ray Imaging for Non-Destructive and Rapid Nuclear Forensics
Investigators:	Derek R. McLain Hemant Sharma

Abstract

This project aimed to use high energy x-ray diffraction microscopy (HEDM) and micro-computed tomography (u-CT) to characterize uranium oxide samples that were prepared under different conditions in order to quantify the particle size differences previously observed in such samples. This method will drastically reduce the time required to perform morphological analysis of samples of interest to the nuclear forensics mission. The first year of this two-year project was spent preparing the various oxide samples and performing conventional analyses on them for characterization. Conventional analyses included scanning electron microscopy, powder x-ray diffraction, infrared spectroscopy, and mass spectrometry. Year two was spent preparing samples for synchrotron analysis, executing beamtime at the Advanced Photon Source, and processing data. While high energy x-ray diffraction microscopy (HEDM) was not able to be carried out, a similar method (diffraction tomography, or DT) was used and was able to generate heat maps of crystal structures in three-dimensional.

As a separate part of this project, machine learning algorithms were developed to de-noise data gathered using tomographic techniques to streamline analysis and further reduce timelines. This portion of the project has resulted in the acquisition and integration of a new computing cluster at the 1-ID beamline and reduced the reconstruction timeline for diffraction tomography (DT) from hours for a single slice to less than 10 minutes per slice. This is a vast improvement for the beamline, which will lend to all future analyses.

LDRD # 2022-0087	Imaging and Detection of Signatures
Project Title:	Developing Metal-Organic Frameworks as Noble Gas Sample Collectors for Trace Isotope Analysis
Investigators:	Peter Mueller Massimiliano Delferro, Derek R. McLain

Abstract

Metal-organic frameworks (MOF) are a class of materials that promise to selectively adsorb and release noble gases above cryogenic temperatures. Such materials have the potential for applications in sampling and gas purification instrumentation used in radioactive noble gas trace analysis for environmental and national security applications with reduced cost, size, time, and energy requirements than current approaches. We have focused on synthesizing and characterizing MOF materials matched to the requirements of Argonne's leading technology applicable for radiokrypton and radioxenon detection, namely, Atom Trap Trace Analysis. For that technology, there are not yet available metal-organic frameworks for selective capture of krypton and xenon from air, which typically requires much smaller noble gas samples and less restrictive purity requirements than are needed by other trace isotope detection techniques. This work will now enable simple, small, unintrusive and portable in-the-field sampling/concentration instrumentation for national security applications. This new approach overcomes a major logistical hurdle experienced in environmental sampling campaign measurements in the past, namely the shipping of large pressurized air cylinders to an analysis facility.

LDRD # 2022-0164	Imaging and Detection of Signatures
Project Title:	Broadband Directional Detection of Electromagnetic Pulses Using Quantum Sensing
Investigators:	Angel Yanguas-Gil Marco Govoni, F. Joseph Heremans

Abstract

We explored the development of field-deployable quantum sensors for the for the detection of broadband electromagnetic (EM) pulses using semiconductor vacancies. We modeled compact approaches to these sensors that don't require the use of bulk nuclear magnetic resonance (NMR) magnets. From the magnetic field distributions we then modeled the interaction of electromagnetic radiation with semiconductor point defects, primarily NV centers in diamond, but also SiC divacancies. Finally, we explored scalable synthesis approaches to synthesize these defects in collaboration with Material Science Division researchers and the quantum foundry. We demonstrated that alternative approaches based on compact permanent magnets can lead to the magnetic fields required to achieve broadband detection in the gigahertz (GHz) range. One of the key advantages of quantum sensors is that they are optically coupled, which can help mitigate the impact of high energy electromagnetic (EM) radiation in the electronics. However, achieving both altitudinal and azimutal detection relies on the polarization state of the incident EM radiation.

LDRD # 2023-0019	Imaging and Detection of Signatures
Project Title:	Relating Uranium Oxide Morphological Differences to Calcination Temperature Using in situ Ultra-Small Angle X-ray Scattering
Investigators:	Anthony J. Krzysko Andrew T. Breshears, Jan Ilavsky, Ivan Kuzmenko, Derek R. McLain

Abstract

Uranium(V, VI) oxide (U_3O_8) is an important front-end intermediate in the nuclear fuel cycle. Accurately predicting the processing history of U_3O_8 samples can aid in determining the origin or intended use of unknown materials. Using the U_3O_8 calcination reaction as a model system, this research used *in situ* ultra-small angle X-ray scattering to demonstrate how the temperature used to produce U_3O_8 results in unique morphological features. These features can then be used as signatures to predict the processing history of U_3O_8 samples. The combination of scattering studies used in this research provided unique information that confirmed existing morphological nuclear forensics signatures and identified novel signatures. In addition, the results obtained using the X-ray scattering analysis assisted in validating and optimizing reaction mechanism models that seek to holistically understand chemical systems related to nuclear forensics.

Microelectronics

LDRD # 2022-0090	Microelectronics
Project Title:	Neuromorphic Computing based on Novel Electrochemical Transistors
Investigators:	Fangfang Xia Joseph W. Strzalka, Jie Xu, Yuepeng Zhang

Abstract

In this project, we proposed to advance the-state-of-the-art neuromorphic-computing design with an emerging type of device, organic electrochemical-transistor (OECT), to enable continuous on-device learning. Different from memristors as the conventional device basis for neuromorphic computing, OECT devices provide a large range of linear and symmetric conductance states with long retention time, which can support more effective and low-power learning. We set out to fill two gaps: (1) the knowledge gap regarding the relationships between the material/device properties and the learning performance at the system level; (2) the technology gap related to large-scale fabrication. To this end, we assembled a co-design team with expertise in materials design and synthesis, device fabrication and integration, as well as learning algorithms. We successfully completed our two-year project, making significant progress in both targeted gaps. Our co-design approach proved instrumental in integrating insights from diverse fields, leading to the development and fabrication of OECT arrays that demonstrated artificial intelligence capabilities in biomedical applications. Our work has also resulted in technological advancements in several areas, including the use of self-driving labs to explore materials and extract structure-property relationships, the development of photo-patterning and printing-based fabrication

for neuromorphic arrays, and the design and implementation of brain-inspired, local and continual learning algorithms.

Physical Sciences and Engineering

LDRD # 2020-0171	Physical Sciences and Engineering
Project Title:	Discovery of Novel Two-Dimensional Nanomaterials to Enable Optimum Gas and Water Sensing Performance in a Field-Effect Transistor Platform
Investigators:	Junhong Chen

Abstract

The project aimed to design and discover new 2D layered nanomaterials for field-effect transistor (FET) gas/water sensors by experimentally synthesizing new 2D nanomaterials and characterizing their sensing performance. We designed and set up a chemical vapor deposition (CVD) system that enables the synthesis of various 2D nanomaterials (MoS_2, WS_2, WSe_2) and their heterostructure (MoS_2/WS_2). Synthesis parameters of molybdenum disulfide (MoS_2), one of the most popular and promising 2D nanomaterials, have been optimized so that monolayer and multilayer MoS_2 layers can be successfully synthesized. Samples were characterized using Raman spectroscopy, scanning electron microscopy (SEM), and atomic force microscopy (AFM). The MoS_2 and WS_2 samples fabricated into FET sensors to detect gaseous and aqueous contaminants. The MoS_2 and WS_2 FETs showed promising n-type semiconductor behaviors with a high on-off ratio. In addition, FET water sensors were demonstrated using printed MoS_2 as the FET channel, and printed graphene as the electrodes and the sensing area. After functionalization of the sensing area, the printed water sensor showed a selective response to Pb^{2+} in water down to 2 ppb. Finally, we have demonstrated a scalable fabrication process and quality control procedure to realize graphene FET arrays able to simultaneously detect trace amounts of heavy metal ions and bacteria in flowing water.

LDRD # 2021-0184	Physical Sciences and Engineering
Project Title:	Accelerator-Based Radioisotope Discovery
Investigators:	Jerry A. Nolen, Jr. Matthew R. Dietrich

Abstract

This project focused on making important advances in several aspects of therapeutic medical isotopes. Some of the developments apply to improving the techniques for production and processing of all medical isotopes including isotopes optimized for imaging as well as cancer therapy. One such development is our concept of the “hot box” incorportating robotics in isotope production. Another important development conceived and demonstrated is a new approach for rapid on-line cross section measurements using light ion beams from our superconducting linac, ATLAS, combined with GammaSphere. Other research advanced production, processing, and delivery specifically of alpha- and

Auger-electron emitters, both of which have the untapped potential for greatly improved therapeutic benefits with greatly reduced toxic side effects. We have developed and patented a new method for the production of the highly important alpha emitter, Ac-225, at Argonne's electron linac using the photonuclear reaction mechanism which produces Ac-225 without the co-production of Ac-227. Our method has the additional advantage of high yields of Ac-225 using much less Ra-226 target material than alternate approaches.

Beyond this development, we have made excellent progress in the emerging field of precision medicine with the elusive Auger-emitters. We have demonstrated production of two promising new Auger electron emitters using proton, alpha, and lithium ion beams (Pt-191 & Tb-155). We have developed micro-dosimetry simulations demonstrating therapeutic promise of Auger emitters. This work has benefited from a collaboration with the UChicago Medical Physics/Cyclotron group.

LDRD # 2021-0193	Physical Sciences and Engineering
Project Title:	Development of Quantum-enabled Sensors for Fundamental Symmetries
Investigators:	David DeMille

Abstract

Quantum-enabled sensors are poised to open new frontiers in the study of fundamental symmetries in nuclear physics. This project was aimed at developing methods and apparatus needed for two planned experiments at the interface of nuclear physics (NP), quantum information science (QIS), and elementary particle physics. One, CeNTREX (Cold molecule Nuclear Time-Reversal EXperiment), is a search for a CP-violating (CPV) electric dipole moment (EDM) along the spin of the proton; this EDM would reveal new forces beyond the Standard Model of particle physics. The other, ZOMBIES (Zeeman-tuned, Optically detected Molecular Beam Investigation of Electroweak Symmetry breaking), aimed to measure nuclear spin-dependent parity-violation effects; this will provide new information on weak interactions between nucleons, which remain poorly understood. Both experiments used nuclei embedded in polar molecules, which serve as amplifying quantum sensors of virtual particles in the quantum vacuum. Molecules are prepared, manipulated, and probed using QIS methods.

Quantum Information Sciences

LDRD # 2021-0279	Quantum Information Sciences
Project Title:	Dissipation-aided Microwave-Optical Entanglement in Hybrid Superconducting Piezo-Optomechanics
Investigators:	Xu Han Aashish Clerk, Dafei Jin, Ivar Martin, Xufeng Zhang

Abstract

The overarching goals of the proposed work were to establish new protocols that leverage dissipation engineering to prepare and stabilize entangled microwave-optical quantum states, and experimentally demonstrate the microwave-optical entanglement for the first time in a superconducting piezo-optomechanical system. In particular, we developed novel mechanical resonators that can mediate efficient interaction between microwave and optics for generating strong entanglement of both the output traveling fields and the intra-cavity fields. The realization of microwave-optical entanglement will provide important quantum resources for quantum transduction and computation.

LDRD # 2021-0294	Quantum Information Sciences
Project Title:	On-chip Quantum Sensing Platform for Spectroscopy of Single Spin Defects in Silicon
Investigators:	Alan M. Dibos Valentine Novosad

Abstract

We performed spectroscopic measurements on individual optically-active spin defects in silicon (Si) that have promising coherence properties in ensembles but are too dim to resolve using conventional cryogenic microscopy. We merged nanophotonic cavity fabrication with ultra-sensitive on-chip superconducting nanowire single photon detectors (SNSPDs) to create an integrated quantum sensing platform to ensure high photon collection efficiency from individual spin defects in Si. In turn, we used these single defect centers as local quantum sensors to probe static and dynamic sources of optical and spin decoherence within the Si host, and explored methods of improving the coherence properties of individual qubits. We expect up to a 10,000-fold improvement in the photon rate collection rate, and this revolutionary approach will enable important but otherwise unachievable single-qubit sensing measurements, which could influence the development of future quantum network hardware. We have developed a successful integration process flow for Si photonic crystal cavity-coupled SNSPDs combining our existing superconductor growth expertise and the Si nanofabrication capabilities. We successfully measured cavity-modified optical lifetimes from ensemble erbium spin centers using our on-chip detectors. With the majority of technical challenges behind us, we then focused on systematically reducing the quantum emitter density to enable optical addressing of individual spin centers.

LDRD # 2021-0296	Quantum Information Sciences
Project Title:	Quantum-Computer-Based Simulation of Strongly Correlated Photocatalytic Materials
Investigators:	Stephen K. Gray Yuri A. Alexeev, Cristian L. Cortes, Dmitry Fedorov, Laura Gagliardi, Martin Suchara

Abstract

The simulation of chemical reactions, including the determination of the reaction path energetics and, ideally, potential energy surfaces, is envisioned to be a key application area for quantum computers. However, to date only very modest problems have been tackled. We developed quantum algorithms, i.e. procedures to run on quantum computers, that simulate the properties of chemical systems. Particular focus was on algorithms that can efficiently, relative to existing ones, simulate complex, correlated systems. A quantum algorithm analog of a recently proposed classical fragmentation method was developed that involved breaking a complex molecule up into parts and carrying out accurate quantum calculations on the individual parts and introducing coupling between the parts. Key outcomes included the development and refinement of such algorithms, and feasibility assessments and comparisons with other approaches.

LDRD # 2021-0300	Quantum Information Sciences
Project Title:	Quantum Simulation of Nonequilibrium Quantum Phases with Novel Single-Electron Qubits
Investigators:	Dafei Jin Xu Han, Ivar Martin, Xufeng Zhang,

Abstract

In this project, we targeted performing the initial but crucial tasks towards our long-term plan to build an unprecedented quantum simulator composed of 10-50 novel single-electron qubits. This all-electron quantum simulator will exhibit superior efficiency in emulating nonequilibrium quantum phases of many-electron systems. It will be constructed based upon our recent experimental breakthroughs in trapping single-electron qubits on solid neon surface, observing the vacuum Rabi splitting between the electron and single microwave photons, and dispersive reading out the electron's motional states, all in a state-of-the-art circuit quantum electrodynamics architecture. It will be further integrated with and boosted by the quantum charge-coupled-device architecture learned from the ion-trap systems, for deterministically trapping and clocked transferring single electrons.

We determined the qubit relaxation time T1 = 15µs and coherence time T2E = 220ns. By changing the solid neon growth condition, we have recently achieved a huge improvement to the relaxation time by one order to T1 = 100µs and the coherence time by three orders to T2E = 100µs. By optimizing the device design and adopting high-kinetic inductance superconducting resonators, we achieved a huge improvement with nearly one order of magnitude to g = 50µs. We have managed to perform single-shot readout with a readout fidelity of 98.1% without relying on a quantum-limited amplifier and a single-qubit gate fidelity 99.97%.

We have observed two-qubit strong coupling by exchanging microwave photons through the resonator. This lead to two-qubit entangled gates for universal quantum computing. These exciting new results suggest that our electron-on-solid-neon qubits have become the best electron charge qubits over any existing systems and are comparable to the superconducting transmon qubits.

Universe as Our Laboratory (ULab)

LDRD # 2023-0158	Universe as Our Lab (ULab)
Project Title:	Towards Prototyping the Design of a Generic Imaging Barrel Electromagnetic Calorimeter
Investigators:	Maria K. Zurek Manoj B. Jadhav

Abstract

The Electron-Ion Collider will address three fundamental questions about nucleons—neutrons and protons—and their assembly into atomic nuclei: How does the mass of the nucleon arise? How does the spin of the nucleon arise? What are the emergent properties of dense systems of gluons? To meet the stringent physics performance requirements for the Electromagnetic Calorimetry in the Central Barrel (Barrel ECal) region of the future Electron Ion Collider, the Yellow Report explores various technologies. However, none of them satisfy the stringent Barrel ECal requirements, particularly the electron-pion separation in the barrel, falling short by nearly two orders of magnitude. This poses a significant risk to critical elements of the Electron Ion Collider scientific program. In response to this challenge, the Argonne team proposed a unique Imaging Barrel Calorimeter, enhancing a state-of-the-art Pb/Scintillating-Fiber (Pb/ScFi) sampling calorimeter with low-cost silicon sensors (AstroPix) that provide a 3-dimensional shower profile.

The LDRD project focused on the silicon layers of monolithic AstroPix sensors—cost-effective, ultra-low-power silicon sensors developed for NASA—in the first half of the calorimeter to capture a 3-dimensional image of the developing shower. We assessed the feasibility of using an AstroPix-like sensor in an imaging electromagnetic calorimeter environment through beam and bench tests. We evaluated the optimal readout scheme for calorimetry and assessed the chip's radiation hardness. We concluded that the chip, with additional designed modifications in its next version, is well-suited for the Electron Ion Collider. After review, this calorimeter design has been unanimously selected for use in the baseline ePIC detector.

Director's Collaborations

LDRD # 2022-0234	Director's Collaborations
Project Title:	Liquid Inks for Near-Infrared Lasers
Investigators:	Benjamin T. Diroll

Abstract

This project fostered collaborative work under a France and Chicago Collaborating in the Sciences (FACCTS) research project between Argonne PI (Diroll, at CNM) and French partner (Ithurria, at ESPCI) with the goal of the creation of new liquid-dispensable materials which can serve as optical gain media for lasers in the near-infrared spectral region. The work used detailed spectroscopic investigations of samples to iteratively inform improved qualities of the subsequently synthesized optical materials. The first output from this collaboration was a work on the unusual violation of Kasha's rule and amplified emission in a bicolor emitter materials consisting of a type-II heterojunction with near-infrared emission. Additionally, stimulated emission was observed in semiconductor nanoplatelets with infrared band gaps spanning the near-infrared spectral window. This work will be summarized in one publication focusing on the properties of mercury telluride nanoplatelets and the mechanism of the observations. A second publication will focus on core/shell materials of similar composition and the synthesis which enables new spectral properties to be observed. Other contributions were made to the photophysical properties of near-infrared active semiconductor nanomaterials.

LDRD # 2023-0259	Director's Collaborations
Project Title:	Scalable Approaches to Nanostructured and On-Demand Single Photon Sources
Investigators:	Gary Wiederrecht Wei Wang

Abstract

This proposal described a France and Chicago Collaborating in the Sciences (FACCTS) research project. The goal of the work was to create “on-demand” single photon sources by adding efficient nanoscale emitters to photonic crystal (PC) structures that are created in a scalable manner, so as to impact the larger quantum optics community. The photonic crystal structures work in concert with the emitters to produce an increase of the photonic density of states near an optical emitter, which in turn increases the rate of optical emission and is known as the Purcell effect. An “on-demand” single photon source is produced when the rate of emission is significantly increased (e.g. dramatically shortening the timeframe for the production of an emitted photon). In this project we used photopolymerizable materials containing covalently bonded quantum dot optical emitters provided by the French collaborators. At the Center for Nanoscale Materials (CNM), these materials were photopolymerized with high spatial precision in an optical microscope onto one-dimensional photonic crystal structures made of silicon nitride (SiN). These silicon nitride structures themselves were fabricated in the Center for Nanoscale Materials (CNM) cleanroom. Optical emission spectra were collected from the photopolymer/one-dimensional nanostructures and analyzed for photonic effects.

Named Fellows

LDRD # 2021-0180	Named Fellows
Project Title:	High-Resolution Prediction of Nuclear Reactor Structural Expansion Feedback
Investigators:	April Novak

Abstract

The commercialization and deployment of Sodium Fast Reactors (SFRs) is expected to result in significant reductions in the amount and radioactive longevity of worldwide nuclear waste reserves. An important contribution to the safety and stability of SFRs is the thermal expansion of core structural materials - an inherent reactor stability mechanism that can cause reactor power surges to self-arrest due to the complex physics interactions. Many aspects of the design of SFRs depend on accurate computational prediction of this phenomenon, which is complicated by the large number of interacting physics domains with unique characteristic length and time scales. This project developed high-resolution computational algorithms and tools for modeling these structural expansion physics. Specifically, development of a multiphysics coupling between NekRS CFD and OpenMC Monte Carlo transport, with on-line mesh and geometry deformation from MOOSE. By coupling state-of-the-art tools for Monte Carlo neutron-photon transport, computational fluid dynamics, and structural mechanics, this research improves our understanding of fast reactor core physics and provides verification data and closures to inform lower-resolution models in use by industry. Key successes are the development of a generalized, high-fidelity multiphysics coupling framework with capabilities for general CAD geometry, multiscale analysis, and integration with digital engineering.

LDRD # 2021-0182	Named Fellows
Project Title:	Towards Heterostructure Superlattices for Enhanced Control of Quantum Defects
Investigators:	Katherine J. Harmon

Abstract

Multilayer heterostructures composed of alternating layers of silicon carbide (SiC) polytypes may provide a path toward enhanced room temperature coherent control and manipulation of optically active spin defects in SiC for quantum information science (QIS) applications. Heterostructure architectures are well-established in the semiconductor industry as a means to tailor the design of electronic band structures, owing to the disparity in band gap of the different component materials. This project applied this principle in SiC, one of the most promising and versatile quantum materials. SiC is reported to occur in over 200 polytypes, each of which has a different band gap and a variety of point defects whose optical properties are strongly influenced by the electronic structure (i.e., band structure) of the surrounding material. A detailed understanding of the SiC growth process and the synthesis phase space is essential for precisely selecting and then switching polytypes while ensuring ultrahigh quality growth of the individual layers. Until now, a few studies of SiC synthesis have been carried out in real time using electron diffraction techniques, providing a valuable but very limited view of the atomistic processes at play. We have developed and implemented an X-ray compatible *in situ* chemical vapor

deposition (CVD) instrument for the high-temperature synthesis of SiC, which is poised to enable the level of atomistic understanding needed to grow the desired SiC heterostructures. In our first *in situ* synthesis experiments under SiC growth conditions (elevated temperatures up to 1500 degrees celsius and with controlled flow of Si- and C-containing gas precursors) we successfully grew additional layers of the same polytype as the substrate material and observed the emergence of X-ray signals indicating the presence of a different polytype. These experiments demonstrate the practical implementation of our novel technology for *in situ* SiC synthesis characterization and set the stage for further in depth exploration of the growth parameters known to influence SiC polytype selection (i.e., temperature, sample pre-treatments, gas environment, etc.).

LDRD # 2021-0186	Named Fellows
Project Title:	Precise Characterization of the Cosmic Microwave Background with the Current and Next Generation of Experiments
Investigators:	Zhaodi Pan

Abstract

This project and fellowship has contributed to advancing cosmic microwave background (CMB) and millimeter-wavelength cosmological experiments, both through new detector development and novel analysis of data from the current South Pole Telescope SPT-3G experiment. The instrumentation component focused on optimizing microwave kinetic inductance detectors (MKIDs), a new detector technology crucial for next-generation millimeter-wavelength measurements. The performance of MKIDs has been directly enhanced as a result of this work, with notable results in probing material properties and detector geometries that influence the fundamental detector noise and optical performance. The results of this work are now providing a strong foundation for new efforts at Argonne for application specific MKID detectors.

In the SPT-3G data analysis, advanced statistical methods such as Fourier-space kernel convolution and joint posterior distributions were used to measure the gravitational lensing of the CMB and enable the reconstruction of the 2-dimensional matter distribution of the Universe. The measurement of SPT-3G gravitational lensing is in turn used to model the underlying cosmological physics such as the expansion rate of the Universe, the density of dark matter, and the sum of neutrino. These results achieve cosmological parameter constraints that are competitive in the field and set the stage for future high-precision CMB data analysis.

LDRD # 2021-0265	Named Fellows
Project Title:	Optimization and Control Techniques for the Electric Power Grid Using Specialized Machine Learning Methods
Investigators:	Alyssa A. Kody

Abstract

The electric power grid of the future faces many challenges including incorporating distributed generation, contending with power fluctuations from renewables, and aging infrastructure. Conventional grid control and optimization techniques may not be able to handle the resulting increase in system complexity and uncertainty. Machine learning offers promising tools that can aid in meeting these challenges. Instead of completely replacing conventional techniques with machine learning, in this project, we targeted the use of machine learning techniques to improve existing control and optimization algorithms. We combined model-based and data-driven methods, and focused the use of machine learning techniques where our existing tools perform poorly: dealing with nonlinearities and uncertainties. We explored these ideas in the context of operations problems under stress, i.e., where networks are forced to deviate from ordinary operating set-points, and use machine learning methods to (i) accelerate distributed optimization problems, (ii) identify lines to de-energize in the context of transmission switching problems, (iii) planning and operation problems accounting for extreme weather conditions, and (iv) increase computational efficiency and scalability. The merit of this research lies not only in solving urgent energy problems related to the safety, reliability, and resiliency of the grid, but also in contributions to incorporating data-driven methods into traditional model-based methods for large-scale engineering applications.

Innovate

LDRD # 2023-0246	Innovate
Project Title:	A Scalable Multi-Physics Optimization Framework for Particle Accelerator Design
Investigators:	Gongxiaohui Chen Tyler Chang

Abstract

The overarching goal of beamline design is to achieve a high brightness electron beam from the beamline. Traditional beamline design studies involved separate optimizations of radio-frequency cavities, magnets, and beam dynamics using different codes and pursuing various intermediate objectives. Given the ultimate goal of producing high brightness beam through the designed beamline, we developed a unified framework for global optimization that integrates multiple physics modules: 1) electromagnetic module (EMM) for solving the resonant cavity eigenmodes field (based on SUPERFISH), 2) magnetostatic module (MSM) for magnet studies (based on POISSON or parameter controlled extrapolated Bezier curve), 3) beam dynamics module (BDM) for particle tracking (based on ASTRA). To effectively handle a large number of tuning variables during the global optimization process, a localized model-based optimization method was employed, which requires fewer simulation evaluations than other comparable methods.

To demonstrate the global optimization workflow, we optimized an S-band (2.856 GHz) BNL type 1.5-cell photogun with standard beam and photogun operation parameters. The emittance generated by the optimized beamline was found to converge to approximately 0.3 um, which is comparable to state-of-the-art results. This framework offers great flexibility in accelerator design, making it easier to explore various physics concepts and obtain more statistical results through systematic geometry tuning (for example, changing cell lengths or adding concave features to introduce a focusing field on the cathode).

LDRD # 2023-0248	Innovate
Project Title:	Synthesis of Uniform, Morphologically Controlled Particle Reference Materials for Nuclear Safeguards using Microfluidic Flow Lithography
Investigators:	Anna Servis Anthony J. Krzysko

Abstract

Exposing the misuse of nuclear facilities and materials requires the development of advanced analytical methods capable of detecting and characterizing individual actinide particles in environmental samples. Development of these methods has recently been limited by a shortage of particle reference materials needed for instrument calibration and method validation. We have demonstrated the feasibility of preparing shape-controllable inorganic microparticles using a photopolymerizable hydrogel template and cerium, a widely used actinide surrogate. Using this approach, enabled us to control the nanoscale morphology, and chemical and isotopic composition of actinide-bearing microparticles. This innovative

new technology for actinide microparticle synthesis promises to facilitate the development of increasingly sensitive particle detection methods and analyses.

LDRD # 2023-0250	Innovate
Project Title:	Improving Gamma Ray Spectroscopy with Natural Language Processing
Investigators:	David Lenz Tamas Budner

Abstract

The goal of this project was to investigate how principles from natural language processing (NLP) could be used to identify gamma rays from data produced in large gamma-ray spectrometer experiments funded by the U.S. Department of Energy. Gamma-ray spectroscopy is an experimental method in nuclear physics for identifying the underlying structure of atomic nuclei by measuring patterns in the gamma rays they emit. Our guiding hypothesis was that subtle patterns connecting different gamma rays could be inferred in the same way that NLP infers semantic information from the probability that two words appear close to each other in written language. Over the course of this project, we successfully adapted simple language models to learn from gamma ray spectroscopy data instead of textual sources. However, we found that our method failed to discern meaningful patterns in the data, missing even obvious patterns that could be gleaned without advanced techniques. We posit a reason for this failure: in gamma ray spectroscopy, scientists seek to detect subtle patterns buried in noisy, random data, but in NLP all of the text fed into a model is assumed to be a valid representation of human language. As such, we believe these NLP models may not be well suited for applications where a large fraction of the data under consideration is noisy, random, or otherwise “patternless.” Nevertheless, discussions from this project resulted in a new idea which could provide a breakthrough in “spectrum background subtraction,” an unrelated area of gamma-ray spectroscopy. This idea is now being tested in another project involving the Principal Investigators. This project also produced open source code that adapts NLP models to gamma ray data, which may allow other researchers to extend or modify our ideas to achieve success in the future.

LDRD # 2023-0255	Innovate
Project Title:	A Numerical Framework for Simulating Direct Air Capture of Carbon Dioxide
Investigators:	Akash V. Dhruv

Abstract

This project introduced new capabilities within Flash-X (https://flash-x.org), an open-source simulation software, to model multiscale chemical transport dynamics that occur at solid-liquid-gas interface in various engineering applications. The model was extensively verified for performance and accuracy, and applied to high-fidelity simulations of direct air capture process in industrial scale engineering components. An important objective of the project was to build foundation for a computational workflow that can integrate chemical process design, with development and optimization of engineering

components to accelerate real-world implementation of decarbonization solutions. This goal was accomplished through this seed proposal and will serve as a motivation for future research work.

LDRD # 2023-0263	Innovate
Project Title:	Three-Dimensional Computational Fluid Dynamics Framework for Predicting Fracture Closure Rates in Enhanced Geothermal Systems
Investigators:	Diego B. Bestel

Abstract

According to the U.S. Department of Energy (DOE) Geothermal Technologies Office, Enhanced Geothermal Systems (EGS) have the potential to power over 40 million American homes and are the next frontier for renewable energy development alongside solar and wind. Unlike intermittent renewable energy sources, Enhanced Geothermal Systems (EGS) offer continuous (always-on), dispatchable, and reliable renewable energy for heating, cooling, and electric power production. Although several large-scale Enhanced Geothermal Systems (EGS) field projects have succeeded in recent decades, Enhanced Geothermal Systems (EGS) has yet to be validated as an optimized technology for commercial-scale deployment. Several hurdles, from capital cost to the long-term durability of energy production, have prevented commercial deployment. In the context of long-term durability, a detailed understanding of fracture opening and closing rates could allow the prediction of Enhanced Geothermal Systems (EGS) performance and durability over time. Current literature on the modeling of fracture closing rates relies on simplified models and extensive calibration against measured data and lacks a detailed description of the physical and chemical mechanisms in the porous media. On the other hand, Computational Fluid Dynamics (CFD) modeling has proved to be a powerful tool for reacting flow coupled with surface reactions and fluid-solid interactions, providing unprecedented details on the underlying physics and chemistry of various systems. Thus, this project aimed to develop a three-dimensional Computational Fluid Dynamics (CFD) modeling framework to predict the fracture closure rates in Enhanced Geothermal Systems (EGS) due to fluid flow and surface/precipitation reactions using computational methods and codes amenable to high-performance computing. As a first step, well-characterized, laboratory-scale experiments were identified from data survey. Although this step seemed trivial, the size of the Enhanced Geothermal Systems (EGS) posed a challenge to Computational Fluid Dynamics (CFD) models regarding computational cost, boundary, and initial conditions. Correctly identifying conditions and a suitable computational domain was key to developing Computational Fluid Dynamics (CFD) models for Enhanced Geothermal Systems (EGS) since most available data is too large to be modeled using Computational Fluid Dynamics (CFD) methods. Having identified the conditions and computational domain to be modeled, Computational Fluid Dynamics (CFD) codes were surveyed regarding their capabilities and suitability to simulate moving boundaries due to surface reactions. CONVERGE was selected as the Computational Fluid Dynamics (CFD) code to be used since it is capable of integrating surface reactions and deforming the boundaries due to silica deposition, effectively closing the fractures in an Enhanced Geothermal Systems (EGS). Finally, a silica deposition mechanism was identified and implemented in CONVERGE as a proof-of-concept framework to predict the fracture closure rates in Enhanced Geothermal Systems (EGS). The laboratory-scale experimental conditions and computational domain (geometry) were used for this testbed.

LDRD # 2023-0273	Innovate
Project Title:	Towards Exascale Chemical Dynamics Via Graphics Processing Unit-Accelerated Polynomial Expansions
Investigators:	Yeonjun Jeong

Abstract

This project investigated methods to enable semiclassical nonadiabatic chemical dynamics in larger chemical systems (~100s of atoms rather than the usual ~10s of atoms) utilizing the forthcoming graphical processing unit (GPU) resources such as Polaris and Aurora at Argonne. We realized this by using permutationally-invarant polynomials (PIPs) to construct the potential energy surfaces (PESs) of molecules on which their dynamics propagates, since GPUs can efficiently perform PIP evaluations. Our initial demonstration of CH4+H trajectory propagation comparing 1 GPU with a single CPU core showed a 8x higher throughput. This speedup was also observed in a production-like setup using a full Polaris node computing multiple trajectories per GPU (1 CPU thread each). The speedup is expected to be greater for larger systems involving excited states, and GPU acceleration of the PES construction using our in-house code (as well as evaluation) supports the ongoing efforts for large-scale production runs.

LDRD # 2023-0280	Innovate
Project Title:	Enhancing Argonne's On-Road Connected and Automated Vehicle Testbed with an On-Vehicle Weather-Prediction AI Model
Investigators:	Nicholas A. Goberville Joseph O'Brien, Matthew E. Tuftedal

Abstract

This project focused on enhancing the current testbed for Connected and Automated Vehicle (CAV) features by incorporating real-time weather data. Weather data (gathered from a Vaisala FD-70) and automotive LiDAR (Ouster OS2-128) were recorded during a 1.5-month duration from March 1st to April 15th, 2023. A data pipeline was developed to synchronize the two sources and filter the dataset to use for AI/ML applications. A model was developed for the task of weather prediction. The primary insight from this work was that weather prediction using LiDAR as a singular source results in inaccurate predictions. However, LiDAR can be used for estimating sunlight intensity and/or cloud coverage estimates, which can produce useful information for vehicles about the present environmental conditions.

LDRD # 2023-0284	Innovate
Project Title:	Enhanced Reinforcement Learning Based Algorithms for Autonomous Materials Synthesis
Investigators:	Anirban Chandra Subramanian Sankaranarayanan

Abstract

Synthesis of complex materials with tailored mechanical/electrical/thermal properties has been an overarching goal for material scientists. Traditionally, the synthesis of a material in a laboratory necessitates rigorous experimentation involving considerable, repetitive human effort. Autonomous robotic systems have significantly reduced the laborious human component, but most autonomous algorithms do not take into consideration that materials synthesis is inherently a time-dependent process. Moreover, certain metastable materials with unique properties are only formed when processing conditions are varied temporally in a specific manner. In this project, we developed a reinforcement learning algorithm for autonomous experimentation. Currently, we have efforts to build on this algorithm and implement it with the polybot system in the Center for Nanomaterials at Argonne National Laboratory.

LDRD # 2023-0287	Innovate
Project Title:	Enabling Accurate and Computationally Efficient Modeling of Supercritical and Supersonic Combustion in Advanced Aerospace Propulsion Systems
Investigators:	V. M. Surya Kaundinya Oruganti Youngjun Lee

Abstract

The main objective of this LDRD project was to evaluate the capabilities of FLASH-X solver for modeling the physics of high-speed multiphase flows for clean energy generation applications. To this end, as a first step we were able to successfully model the canonical test case of underexpanded jets in supersonic crossflows using the diffuse interface methodology in FLASH-X. The multispecies solver was able to qualitatively predict the physics of complex shock structures and jet mixing patterns for different gaseous injectants. In the second step, we implemented the Nobel-Abel Stiffened Gas (NASG) equation of state model with simplified mixing laws to simulate the physics of liquid jet in supersonic crossflow. While the equation of state model was able to capture the physics of simplified 1-D and 2-D test cases, there were numerical instabilities for the complex jet in crossflow problem which were not fully addressed in the six months duration of this project.

LDRD # 2023-0294	Innovate
Project Title:	Machine Learning Data Fusion for Online Estimation of Traffic State in Urban Road Networks
Investigators:	Arindam Fadikar Natalia Zuniga Garcia

Abstract

Accurate and frequent traffic state estimates, including traffic volume, density, and speed, are critical for urban traffic control and management strategies. The deployment of several intelligent transportation systems, such as loop detectors and telematics sensors, provides access to multiple data sources for traffic state estimation. However, the data granularity and spatio-temporal heterogeneity make it difficult to estimate system-level and online or real-time metrics. Through this project, we explored the availability and scope of using auxiliary data to improve the spatio-temporal model for predicting traffic volume in the Chicago area. We partnered with Smarking and acquired historical parking data for the Millennium Park parking garage. Preliminary exploratory data analysis was performed and presented to the industry collaborators.

LDRD # 2023-0313	Innovate
Project Title:	Energy-Efficient Lithium Hydroxide Production From Spodumene Via Microwave-Assisted Solid-State Lithium Salt Production And Electrodialysis
Investigators:	A. H. M. Golam Hyder

Abstract

Spodumene is one of the domestic lithium mineral sources. As domestic spodumene mines have been under development, it is important to develop an energy-efficient and robust technology to produce lithium hydroxide from spodumene for the forthcoming domestic lithium production. Current processes to extract lithium from spodumene and produce lithium hydroxide are energy-intensive. We developed a new microwave-assisted technology to extract lithium from Spodumene by integrating three process steps: (i) solid-state reaction, (ii) water leaching, and (iii) electrodialysis. Results showed that our process steps lead to a significant yield of lithium with a purity of ~93 %.

LDRD # 2023-0318	Innovate
Project Title:	Science Preserving Data Approximation and Input Output Optimization Using Artificial Intelligence for Real-time High-resolution Tomography on Integrated Research Infrastructure
Investigators:	Robert Underwood Viktor Nikitin

Abstract

In this project, we conducted an end-to-end evaluation of how lossy compression affects the tomographic datasets collected at the imaging beamline 2-BM of the Advanced Photon Source. These

datasets typically have huge data sizes (30-50GB) for static samples, and more than 1TB for dynamically changing samples. During the project, we have demonstrated a compression ratio of 95.6x with sufficiently accurate reconstructions. This represents a substantial improvement from both 1.15x with lossless compression and our initial goal to achieve a 10x compression ratio. We believe additional improvements are possible. This work substantially improves the storage and data transmission capabilities for tomographic codes to address the growing needs of the Advanced Photon Source beamlines with APS-U.

Additionally, we improved the ability to stream the extremely large datasets used in tomography through the development of four new open-source software capabilities. On the compression side: a) we completely rewrote the data management in LibPressio to handle out-of-core data to work with extremely large datasets used in tomography that do not always fit in system RAM or more limited GPU RAM, especially under contention from analysis codes; b) we created LibPressio-Predict, a new library to provide applications a portable way to predict compression performance. LibPressio-Predict can be used to predict streaming performance for performance tuning of streaming and new modules in LibPressio to support for additional compressors relevant for light sources (e.g. TEZip, cuSZp, SZx) for a full evaluation of appropriate methods. On the tomography side, theTomocuPy package used at the Imaging group for reconstruction was enhanced to better support in-memory streams of data to improve throughput and integration with compression codes.

LDRD # 2023-0320	Innovate
Project Title:	Direct Physics Extraction from X-ray Scattering Data using Interpretable Artificial Intelligence
Investigators:	James Horwath Qingteng Zhang

Abstract

Macroscopically measurable properties of a material represent an average over the entire system, however local instabilities or heterogeneity can drive significant irreversible changes to the material properties in an unpredictable manner. Therefore, the development a precise, quantitative link between macroscopic observations and microscopic dynamics is an important challenge in physics and materials science. X-ray Photon Correlation Spectroscopy (XPCS) is uniquely capable of capturing these local fluctuations by tracking non-equilibrium dynamics in materials across wide ranges of length and time scales, however the empirical nature of traditional XPCS analysis makes it difficult to explicitly tie observed dynamics to fundamental properties or mechanisms in a material. The goal of this project was to use symbolic machine learning, which learns simple, interpretable equations directly from raw data, to link empirical parameters derived from XPCS fitting to analytical physical models. We developed this capability in the context of understanding rheological relaxation in a jammed glassy colloid, where a vast literature provides models to describe the evolution of shear stress as a function of time during relaxation under a variety of conditions and stimuli, yet the underlying microstructural mechanisms which enable relaxation are still unknown. Our method started by using conventional XPCS analysis to extract characteristic time constants describing microscopic dynamics. In parallel, we used a symbolic machine learning algorithm called SINDy (Sparse Identification of Non-linear Dynamics) to learn a

physical equation which describes the macroscopic relaxation process and yields a fundamental relaxation time for the system. Finally, we identify relationships between the microscopic trends from XPCS with the physical model produce by SINDy to understand how local dynamics impact the larger system. Our results showed that there are two distinct relationships between these quantities. The first regime showed a simple linear relationship between macroscopic and microscopic time scales clearly identifying viscous flow in the colloid as the microscopic driver of the relaxation process. However, the second regime found macroscopic relaxation rates which were entirely independent of the microscopic dynamics. This trend occured at points in the experiment which saw a sudden drop in measured shear stress, indicating that we were observing avalanche dynamics where local changes appeared minimal while dramatic changes in the material were seen over a larger scale. This research provided an intuitive explanation of microscopic relaxation processes and provided a new interpretation of avalanche dynamics, which are commonly observed in a wide range of physical systems.

Swift

LDRD # 2022-0409	Swift
Project Title:	Computational Analysis of High Resolution X-ray Data for Brain Imaging
Investigators:	Nicola J. Ferrier

Abstract

The goal of this project was to develop scalable algorithms for accurately analyzing large volumes of high-resolution x-ray data to segment nanometer scale structures. Specifically, we sought to extract the individual trajectories of myelinated axons (MA) from high resolution x-ray data of cm^3 brain volumes (multi-petabyte datasets expected from Advanced Photon Source Upgrade (APS-U)). We identified the key technical and algorithmic challenges and plausible solutions for tracing axons across the gap in the three dimensional data that occurs due to the cutting of samples.

LDRD # 2023-0185	Swift
Project Title:	Evaluating the Utility of Fluorine Analysis Methods for Nuclear Forensics
Investigators:	Derek R. McLain John J. Arnish, Jennifer L. Steeb

Abstract

Fluorine is an integral part of the nuclear fuel cycle and its presence in a sample of nuclear material is a signature of prior chemical processing. This project examined various methods of conventional fluorine analysis and established the feasibility of differentiating fluorine-containing uranium compounds from other common uranium compounds using a synchrotron light source.

Methods for the quantitation of fluorine in inorganic substances date back over 200 years. They tended to have poor detection limits but were still employed commonly as late as the 1950s. Since then, a variety of analysis methods have been employed to determine total fluorine, including combustion ion chromatography (CIC), ion selective electrodes, and inductively coupled plasma mass spectrometry (ICP-MS), either directly or through the generation of polyatomic barium monofluoride (BaF+) after an initial ICP-MS screening step (ICP-MS/MS). Generally, these methods have much better limits of detection but also require the decomposition of samples. Non-destructive methods have also been developed including instrument neutron activation analysis (INAA), proton induced gamma emission (PIGE), and x-ray photoelectron spectroscopy (XPS), which have comparable limits of detection but are limited to surface investigation or suffer from interference effects.

In more recent years (since 2015), synchrotron techniques have also been used to identify fundamental parameters of solid uranium fluoride compounds. Typically, these use a combination of extended x-ray absorption fine structure (EXAFS) and x-ray absorption near edge structure (XANES) measurements to discern things like bond distance, oxidation state, and orientation. There has also been at least one effort to map fluoride in a uranium sample using scanning transmission x-ray microscopy (STXM) with soft x-rays sensitive to fluorine (F) fluorescence. Through simulation, this project determined that EXAFS/XANES could be used to differentiate uranium-fluorine compounds from other uranium

compounds of similar structure without having to use x-rays in the soft/tender region that require vacuum environments to successfully detect. Future work will involve the experimental validation of these simulations.

LDRD # 2023-0189	Swift
Project Title:	Feasibility Study on Transportation of Spent Microreactors
Investigators:	Tingzhou Fei

Abstract

Microreactors are small nuclear reactors envisioned for remote deployment. Most plans involve combined transportation of the microreactor and its spent fuel. United States regulations require that spent fuel must be transported in a "Type B" package, which has strict performance requirements on radiation shielding and criticality safety, among other characteristics. However, the package must also meet size and weight requirements to be transportable. This project investigated the feasibility of transporting spent microreactors in Type B packages. Through parametric analyses, the study found that shields are likely the dominant contributor to the total weight of the spent microreactor to be transported. The transportation of a spent microreactor by road is feasible though challenging. The transportation requirement would affect the performance (burnup) the reactor can achieve, so it is recommended to consider transportation requirement in the early design phase of a microreactor.

LDRD # 2023-0193	Swift
Project Title:	High-speed X-ray Imaging for Lithium-Ion Battery Safety
Investigators:	Chi Young Moon Alan L. Kastengren, Christopher Powell

Abstract

This project was aimed at investigating thermal runaway of lithium-ion batteries using high-speed X-ray imaging. The thermal runaway process is of great interest for safe deployment of lithium-ion batteries, especially in automotive applications. Thermal runaway can be caused by excessive temperature and mechanical damage, and can result in cascading failure of neighboring cells, fires, explosions, and release of flammable and toxic gases. This project demonstrated that X-ray imaging can achieve microsecond time scales and micrometer spatial resolution using a cylindrical battery cell, as well as quantifying structural features including manufacturing defects using X-ray microtomography.

LDRD # 2023-0194	Swift
Project Title:	Potential of 2-Dimensional Materials in Electronic Device for Nuclear Energy System
Investigators:	Wei-Ying Chen Xuedan Ma

Abstract

Two-dimensional materials offer opportunities in advanced electronic devices for their unique electronic and photonic properties. As the technology is maturing, more two-dimensional materials will see their applications in nuclear reactors and other nuclear facilities. The irradiation effect on the performance of two-dimensional materials will need to be considered. We initiated a new research area by performing *in-situ* and *ex-situ* irradiation on two important two-dimensional materials, graphene and hexagonal boron nitride, and using the subsequent electrical conductivity measurements and high-resolution single particle spectroscopy to evaluate their electrical and photonic property degradation in the environment of nuclear reactors.

We investigated several ion irradiation approaches aiming to enhance the efficiency of producing single photon emitters (SPE) in hexagonal boron nitride. The first approach was implanting a very low amount of carbon based on the hypothesis overlapping single photon emitter signals would diminish the detection rate. The second approach was implanting a high amount of carbon to determine if we did not have enough single photon emitters in the samples. The third approach was heavy ion irradiation on manganese and carbon-coated hexagonal boron nitride (h-BN) with the idea to implant manganese and carbon into hexagonal-boron nitride through ion-mixing effect. This assumed that implanted carbon and manganese were a source of a single photon emitter. The fourth approach was to place a carbon thin film in front of the hexagonal boron nitride (h-BN) to reduce the carbon ion energy so more carbon would be implanted.

LDRD # 2023-0196	Swift
Project Title:	Improve Electronic Data Exchange for Natural Gas Data Collection
Investigators:	Dariusz Blachowicz

Abstract

This project researched characteristics for data availability for capacity data for natural gas pipelines and conducted experiments to adjust the schedules for sending data requests to align our systems with the correct cycle postings processed by various natural gas pipelines. The consolidated dataset provided the synchronized information for all natural gas pipelines, providing a solid view of the state of the natural gas pipeline system at certain stages of each Gas Day. Data collected by our Electronic Data Interchange (EDI) system is used to analyze the effects of man-made or natural disasters on the energy system in the United States in real-time.

LDRD # 2023-0197	Swift
Project Title:	Building a Framework to Connect GCMat with GREET
Investigators:	Jarod C. Kelly Allison C. Bennett, Matthew E. Riddle, Michael Wang

Abstract

Carbonates and hydroxides of lithium are key battery materials extracted from natural resources, namely brines and ores (spodumene). These natural resources are spread unevenly across the planet, and their extraction and processing occur in different countries, creating complex supply chains. The resulting environmental footprint of these supply chains can vary considerably due to the different electricity mixes in each country and the varying emissions from transportation between stages. The focus of this work is on the carbon footprint of these processes. This carbon footprint can change with supply chain disruptions, eventually affecting a single battery's carbon footprint. However, there are limited tools to determine the carbon footprint of the global lithium supply chain and its effect on the battery carbon footprint. To fill this knowledge gap, a novel model (G-Bridge) was developed that bridges the capabilities of two important models from Argonne National Laboratory: GREET® (Greenhouse gases, Regulated Emissions, and Energy use in Technologies) and GCMat (Global Critical Materials). The G-Bridge model was subsequently used to investigate the variation in lithium-ion batteries' carbon footprint under various supply chain disruption scenarios simulated using agent-based modeling. Overall, the results of this study can help determine the impacts of the battery supply chain, and inform stakeholders of how their choices may alter those results.

LDRD # 2023-0209	Swift
Project Title:	Incorporating Equity into Electricity Planning Model Decisions
Investigators:	William N. Mann

Abstract

The goal of this project was to develop a novel capacity expansion module in the Argonne Low-carbon Electricity Analysis Framework (A-LEAF) that incorporates societal objectives, such as energy justice and equity considerations, when determining optimal electricity infrastructure investment decisions. Since there is no best approach to evaluating equity, we implemented several different features in A-LEAF to demonstrate the trade-offs of these approaches. We successfully improved the equity of outage distributions in a case study of the ERCOT grid. For the equity of electricity reliability, we found that higher network resolution was essential to evaluating equity impacts, and further work is needed to increase the network resolution and connect with demographic data at the census tract level, which is far more detailed than typical grid capacity expansion models. The outcomes from this project were used as part of a successful, Argonne-led Grid Modernization Initiative proposal on incorporating equity into grid long-term planning models.

LDRD # 2023-0214	Swift
Project Title:	Quantifying the Effect of Porosity on the Environmental Degradation of Additively Manufactured 316L in Light Water Reactor Coolant
Investigators:	Bogdan Alexandreanu Xuan Zhang

Abstract

Our research was directed at quantifying the effect of porosity on the onset of environmental degradation/crack initiation in a relevant light water reactor (LWR) environment. Under the program, compact tension specimens were fabricated and produced using the Renishaw 400 printer and were then characterized. Based on the outcomes of this initial research, a new program was developed and awarded under the DOE Light Water Reactor Sustainability Program (LWRS). Under this new program, these specimens will be tested. The additively manufactured alloy's response to mechanical testing will be correlated with its microstructural features, and, overall, the performance of the additively manufactured alloy surface will be compared with that of the "traditionally" produced alloys that are currently in use.

LDRD # 2023-0218	Swift
Project Title:	Embedded Watermarks in 3-Dimensional-Printed Metallic Structures
Investigators:	Bogdan Alexandreanu Alexander Heifetz, Xuan Zhang

Abstract

Research is being directed at creating embedded watermarks in laser additive manufactured components "certified" for service in nuclear power plants. Given the vast number of printers in various settings worldwide – private and academic – it is envisioned that only a small number of facilities will undergo the certification process for a specific set of parts to be used in the nuclear industry. To distinguish such parts, a watermark – ideally internal, thus not readily reproducible – will be needed, allowing for the part's provenance to be ascertained unambiguously. This project evaluated two ways of producing a watermark using the two types of available printers, a laser power bed fusion and a directed energy deposition. A non-destructive examination based on the Pulsed Infrared Thermography method was adapted and optimized for the task.

Two stainless steel blocks were 3D-printed with several embedded watermarks features such as the Argonne logo, laboratory initials ANL, and bars of different thicknesses resembling "barcodes" underneath the surface at 0.5 mm and 0.25 mm depths. The embedded features were then imaged by the Pulsed Infrared Thermography (PIT) method.

LDRD # 2023-0222	Swift
Project Title:	Sustainable Aviation Fuels and Hydrogen Production from Biomass and Plastic through Two-Step Process
Investigators:	Donghyeon Kang David Kaphan

Abstract

The objective of this project was to pioneer an electrochemical production of sustainable aviation fuels (SAFs) from bio-acids and plastic wastes. We demonstrated a groundbreaking electrochemical procedure. An advantage of our two-step process is that it does not require extreme conditions such as high temperature and high pressure, which are conventional in chemical upcycling of polyethylene and polypropylene. Additionally, pure hydrogen fuel is co-produced in the electrochemical system, resulting in reduction of the cost of production for sustainable aviation fuel. Our process is cost-effective and energy-saving compared to conventional processes to produce sustainable aviation fuels from biomass wastes. Important initial data corroborated the feasibility of this method.

LDRD # 2023-0224	Swift
Project Title:	Probabilistic Assessment of Damage Status, Risk and Resilience due to Tropical Cyclones (PADSAR-TC)
Investigators:	Mustafa S. Altinakar Iain R. Hyde

Abstract

This project created a tool to assess the impacts of tropical cyclones (TC) on the built environment in a probabilistic framework in order to aid risk and resilience assessments. It builds upon previous work generating synthetic tropical cyclone tracks and probability grid creation. The project focused on creating annual exceedance probability grids (extreme wind and extreme rainfall total) for current and future climate scenarios using synthetic tropical cyclone tracks developed using our own Argonne model and tracks from literature. Other project goals included also acquiring a geolocated asset data with attributes allowing fragility curve selection and building a knowledge base of fragility curves for different types of assets (buildings and infrastructure). These would then be used with probability grids for extreme wind and extreme rainfall totals to obtain damage status probabilities for the assets. During the course of the study, the objectives had to be modified due to the difficulties in getting geolocated asset data. Therefore it was decided to focus the project on generating the probability grids. It was necessary to create a new set of grids with a higher resolution that brings the cell size to 4 kilometers. The grid covers the entire continental United States. The standard output is a GridData object and most of the math occurs at the GridPoint level. During the grid filling process, some basic global statistics were performed at the cell level. Since GridData object contains complete time histories of wind and rainfall, it is also possible go back anytime to perform all sorts of additional analysis for various other types of variables (such as event durations, wind direction, etc.).

LDRD # 2023-0231	Swift
Project Title:	Direct Lithium Extraction from Geothermal Brine by Ion-Selective Electrode Combined Electrodialysis Cell
Investigators:	Jeffrey W. Elam Donghyeon Kang

Abstract

This project validated a novel direct lithium extraction technology to advance the domestic critical materials supply chain, and to promote commercialization in collaboration with our industry partners. This project enabled us to acquire essential tools vital for the progress of direct lithium extraction research and development. We successfully demonstrated the enhanced selectivity of lithium extracted from geothermal brine, by the implementation of the Argonne-developed coating process. We have accumulated crucial preliminary data that will allow future research with industrial partners.

LDRD # 2023-0232	Swift
Project Title:	Federated Learning for Battery Prognostics
Investigators:	Feng Qiu Susan J. Babinec, Alinson Santos Xavier

Abstract

Batteries, in energy storage and electrical vehicles, have become a center piece for renewable integration, transportation electrification and decarbonization. Battery degradation and health management is a key issue for lowering leveraged costs and maintenance costs and reducing unexpected failures. However, two major barriers to battery storage health management are 1) lack of advanced battery degradation modeling that can accurately predict remaining useful life at any time point in the battery's lifetime given various environment conditions and degradation behaviors, and 2) the data privacy issues, which could prevent storage operators and manufacturers from accessing the monitoring data at the customer's site to perform diagnostics or prognostics. In this work, we investigated advanced machine-learning-based battery degradation modeling and proposed a federated learning framework for prognostics-based battery storage asset management. The proposed framework allows operators, developers, and manufacturers to perform machine learning-based storage asset management without collecting private battery monitoring and operational data from the customers. The developed distributed and privacy-protecting asset management approach could also be extremely useful in many other large-scale distributed energy resource integration and management. In this project, we focused on lithium-ion batteries.

LDRD # 2023-0235	Swift
Project Title:	Testing and Characterization of the Wakefield Accelerator Prototype Module
Investigators:	Alexander A. Zholents

Abstract

We proposed to develop a sustainable Extremely High Frequency-band collinear two-beam wakefield accelerator possessing the record performance characteristics, including a) an accelerating gradient up to 100 megavolt per meter limited by the drive bunch beam break-up instability, b) a high bunch repetition rate up to 20 kilohertz limited by a material stress caused by a heat load on the accelerator structure deposited by the electron beam, c) a nanosecond long pulse of the accelerating filed perfectly synchronized with the arrival time of the witness bunch resulting with ability to withstand a high surface field without a breakdown. In this project we relied on a recently developed technology at the Advanced Photon Source for a fabrication of miniature accelerator parts commensurable with using the Extremely High Frequency-band. We also used a new millimeterWave laboratory equipped with instruments for precision measurements in the Extremely High Frequency-band. This project provided an opportunity to advance a state-of-the-art of modern accelerator technology and pave the way for building a compact X-ray Free Electron Laser at Argonne National Laboratory. A shorter version of the compact wakefield accelerator module (0.3 m versus 0.5 m) was fabricated, assembled and tested with the electron beam at the Accelerator Test Facility at Brookhaven National Laboratory. Preliminary results indicate that it performs as designed within a margin of acceptable errors.

LDRD # 2023-0342	Swift
Project Title:	Automate Electronic Data Exchange for Natural Gas Data Collection
Investigators:	Dariusz Blachowicz

Abstract

This project involved developing software that allows the automated control of the incoming Electronic Data Interchange (EDI) data with real-time alarms of any inconsistencies from the required timely posted cycles processed by natural gas pipelines. This is an improvement in collecting Electronic Data Interchange data that refers to a flow of natural gas by interstate pipelines. The data includes operational capacity (the total scheduled gas quantity and operationally available capacity), unsubscribed capacity (what gas is available beyond the Contracted Firm Capacity), and operational notices (which are often informational, but can also include critical information on planned and unplanned disruptions). The automation allows screening for missing data and the generation of e-mails to the system operator in the case of interruptions in data collection.

LDRD # 2023-0353	Swift
Project Title:	Knowledge Extraction
Investigators:	Scott F. Parent Benjamin A. Blakely, Douglas E. Johnson, Heather A. Seger

Abstract

International events impact the global nuclear landscape at an increasingly rapid pace, and the ability to detect and understand these events in a timely manner can strengthen nonproliferation efforts. To monitor these global events in real-time, nonproliferation analysts must discover and process huge volumes of information in their efforts to detect, verify, and predict potential proliferation activities. Assessing proliferation-relevance of fresh content with precision requires connection and comparison to domain expertise. Prior efforts to facilitate this kind of assessment with Information Retrieval and Natural Language Processing techniques and technologies have been relatively limited both in terms of precision and of recall. This project successfully capitalized on recent large language model developments to prototype a knowledge extraction capability that significantly outperforms both prior Argonne efforts and the team's most ambitious expectations. Demonstrations of strategy components have successfully identified activities, agents, impacts, locations, relationships, and properties from text in a novel way that support the application of nonproliferation expertise in the form of ontology rules and accepted facts—and provide a basis for additional post-processing to further improve on the accuracy of these results. In short, this prototype outperformed expectations in its ability to extract knowledge from open source texts in a form conducive to supporting nonproliferation analyses.

LDRD # 2023-0354	Swift
Project Title:	Corrosion Performance Assessment of Additive Manufactured Materials for Nuclear Energy Systems
Investigators:	Vineeth Kumar Gattu J. Ernesto Indacochea

Abstract

The U.S. Department of Energy is actively supporting the advancement of manufacturing methods to expedite innovations that reduce costs and timelines in constructing new nuclear plants. Streamlining the fabrication of nuclear power plant components to make it faster, more cost-effective, and reliable is a key focus. Next-generation molten salt reactors present a challenge as structural materials must withstand harsh operating conditions, emphasizing the crucial need for corrosion-resistant materials. Additive manufacturing (AM) is being used to produce large, complex alloy parts crucial for various industrial applications in corrosive environments. However, ensuring the long-term performance of these materials necessitates a comprehensive understanding of the relationships between processing methods, resulting microstructures, and corrosion resistance. The Argonne ElectroCorrosion Toolkit^{TM} technology was utilized to assess the microstructures and electrochemical corrosion behaviors of 316L, and Inconel 718 alloys produced through additive manufacturing and traditional methods. Changes in electrical properties of corroded surfaces were measured using electrochemical impedance spectroscopy, and surface microstructures were examined through scanning electron microscopy.

Differences in test responses and corrosion behaviors were quantified and correlated to the microstructural characteristics and AM processing techniques.

LDRD # 2023-0355	Swift
Project Title:	Collaborative Cyber Analysis for Energy (CCafe)
Investigators:	Daniel J. Harkness

Abstract

Collaborative Cyber Analysis for Energy (CCafe) is a new approach to connect cyber defenders to the data they need to 1) understand an anomaly, 2) identify potential responses, and 3) better understand adversaries. This allows for more complete response, improved defense, and higher quality cyber threat intelligence. The CCafe project took this from a conceptual idea to a tangible proof-of-concept with the following capabilities:

1) A guided tool helping analysts create requests for information
2) Capabilities to share the request across a trusted community of peers
3) Capabilities for recipients to review incoming requests, query common internal systems, and respond to the original request with their relevant data
4) Capabilities for the original analyst to collect and review the data sent from their peers

In addition to the proof-of-concept, a scenario and demonstration was developed. In the demonstration, four fictional energy companies collaborated to analyze suspicious activity that turned out to be a bad actor hijacking energy infrastructure to do crypto mining. The scenario demonstrated the development and sharing of a query by one of the companies and responses from the other three. The responses included automated queries using two different data sources commonly used internally as well as one manual response. The proof-of-concept together with the demonstration provides something tangible that can be used when discussing the concept with potential sponsors, collaborators, and users.

LDRD # 2023-0357	Swift
Project Title:	Applying Nuclear Energy to Accelerate Large-Scale Atmospheric Carbon Removal Through Engineered Enhanced Weathering
Investigators:	Nicolas E. Stauff

Abstract

Enhanced weathering is one of six emerging technologies for capturing and sequestering carbon dioxide (CO_2) from the atmosphere and is considered crucial for achieving net-zero emissions by 2050. Enhanced weathering could potentially capture 4.5 Gt CO_2 annually, and engineered enhanced weathering with heat and electricity generated by nuclear power could further accelerate capture and achieve high-efficiency removal with co-benefits of additional revenue for the power plants. This project enabled initial development of a model that quantifies with increased fidelity the performance of coupling nuclear power and engineered enhanced weathering.

LDRD # 2023-0361	Swift
Project Title:	Advanced Rare Earth Magnets
Investigators:	Matthew R. Earlam Michael J. Dziekan, John N. Hryn

Abstract

We constructed a light rare earth electrowinning cell. The cell is operated with a Neodymium Praseodymium fluoride-Lithium Fluoride Electrolyte, with Neodymium-Praseodymium Oxide as the feed material. The cell operates at 1010-1060 Celsius. Direct current is applied across the electrodes to produce Neodymium-Praseodymium metal and carbon dioxide. This is the largest cell of this type in operation in the United States. Operation of this cell in future projects will produce data that could be used to construct and operate a commercial size cell.

LDRD # 2023-0364	Swift
Project Title:	Enhancing Nonlinear X-ray Spectroscopy in Transition Metal Complexes with Entangled Photons
Investigators:	Phay J. Ho Anouar Benali, Gilles Doumy, Kevin E. Gasperich, Linda Young

Abstract

The objective of this project was to pioneer the exploration of quantum sensing within the x-ray domain. We achieved this by developing a novel theoretical framework and precise quantum chemistry algorithms to simulate entangled two-photon x-ray absorption in transition metals. The insights gained from this endeavor held promise for inspiring innovative x-ray spectroscopy methods applicable to various materials, such as solar cells, catalytic centers in transition metal complexes, and quantum materials. Furthermore, this research stimulated interest in the development of quantum x-ray light sources and high-resolution imaging using correlated photons at Advanced Photon Source Upgrade. Over the course of the four-month funding period, our focus centered on expanding our accurate first-principles theoretical method, employing the selected Configuration Interaction (sCI) method to investigate x-ray spectroscopy in earth-abundant transition metal complexes. Implementation of the sCI within the Quantum Package (QP2) code, showcased its capability to predict ground and core-excited states of iron with high accuracy. Notably, our approach surpasses existing quantum chemistry methods, eliminating the reliance on chemical intuition.

LDRD # 2023-0369	Swift
Project Title:	Distributed Quantum Sensing Using Entangled Photons in a Campus-Scale Quantum Network
Investigators:	Xu Han Rajkumar Kettimuthu, Ivar Martin, Joaquin F. Chung Miranda

Abstract

The overall goal of this project was to demonstrate distributed quantum sensing (DQS) by harnessing the fiber-optical quantum network at the Argonne campus. We aimed to distribute high-fidelity entangled photons and maximize the potential benefits of multipartite entanglement in our quantum network, and probe a global RF signal using the entangled photons distributed in separate labs to achieve quantum-enhanced sensitivity.

During the project, we finished building a campus-scale fiber-optical quantum network at Argonne, which interconnects several quantum labs. These fibers serve as the low-loss quantum communication channels of our quantum network. We demonstrated bipartite polarization-entangled photon distribution between two buildings with a high fidelity and did preliminary work on the photon entanglement distribution between the new fiber link connecting two buildings as an initial step for realizing DQS. We also successfully installed a pair of superconducting nanowire single-photon detectors (SNSPDs) in a dilution refrigerator. These efforts paved the way for future cross-lab experiments in our quantum network.

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