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ANL-25/32

2024 LDRD Annual Report

Laboratory Directed Research and Development Program Activities

Laboratory Directed Research and Development

This material is based upon work supported by Laboratory Directed Research and Development (LDRD) funding from Argonne National Laboratory, provided by the Director, Office of Science, of the U.S. Department of Energy under Contract No. DE-AC02-06CH11357"

ANL-25/32

2024 LDRD Annual Report

Laboratory Directed Research and Development Program Activities

prepared by
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July 15, 2025

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 2024 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. Funded projects reflect the Laboratory 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.

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 2024 LDRD Summary Report

This summary report provides an overview of all LDRD projects at Argonne that concluded in Fiscal Year 2024. 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.

Table of FY24 LDRD Projects

LDRD Project Summary Report for FY 2024

LDRD # 2021-0044	Development of Time-Resolved Hard X-ray Bragg Ptychography
Investigators:	Haidan Wen Yue Cao, Mathew J. Cherukara, Burak Guzelturk, Martin V. Holt, Stephan O. Hruszkewycz

This project focused on the development of time-resolved hard x-ray Bragg ptychography, a novel technique leveraging x-ray coherence and timing structure at the Advanced Photon Source (APS) to achieve unprecedented spatiotemporal resolution in x-ray measurements. The project successfully developed the proposed instrument at Sector 26 of the Advanced Photon Source. The techniques, including instrumentation and data analysis tools based on machine learning and artificial intelligence, have proven beneficial not only for user programs at Sector 26 but also for studies of dynamic structural properties at other sectors, such as Sector 7 and Sector 34. The project enabled the study of nanoscale structural dynamics in quantum materials. In FeRh, ultrafast structural phase transitions were spatiotemporally. In $BiFeO_3$, the dynamical melting and reconfiguration of ferroelectric domains were observed. In $PbTiO_3/SrTiO_3$, the evolution of adaptive response of ferroelectric domains upon optical excitation was tracked, suggesting potential applications in artificial neuronal networks.

LDRD # 2021-0274	Artificial Intelligence-Emulator Assisted Data Assimilation (AIEADA)
Investigators:	Veerabhadra R. Kotamarthi Scott M. Collis, Emil M. Constantinescu, Ian T. Foster, Robert L. Jacob, Bethany A. Lusch, Romit Maulik, Vishwas Rao, Jiali Wang

The project successfully enhanced Argonne's capabilities in utilizing large-scale geophysical datasets and global weather/climate models by integrating Argonne-developed artificial intelligence and machine learning tools, such as Auto Machine Learning and DeepHyper, with new computational hardware. It delivered an open-source platform for conducting large-scale data assimilation studies using partial differential equation-based models combined with artificial intelligence/machine learning models. A significant achievement was the development of an emulator for the Rapid Radiative Transfer Model, which operates nearly 1000 times faster than existing models used in climate and weather forecasts. Additionally, a predictive model for the Madden-Julian Oscillation (MJO) was created, demonstrating notable success in forecasting its onset over the Southern Indian Ocean. These advancements are currently under journal review. The project also contributed to the development of the STORMER medium-range weather forecasting model, positioning Argonne's model among the top-performing artificial intelligence models globally for this purpose. Furthermore, the project fostered a collaborative community of researchers across multiple divisions, universities, and private industry, establishing a sustainable ecosystem for artificial intelligence model development and application in geophysics at Argonne. The work led to a publication and several presentations at national and international conferences, significantly advancing Argonne's presence in the artificial intelligence applications to weather research.

LDRD # 2022-0008	Development of 3-Dimensional Dichroic Ptychography at Advanced Photon Source
Investigators:	Joerg Strempfer Junjing Deng, Doga Gursoy

The project aimed to develop advanced microscopy techniques for the Polarization Modulation Spectroscopy Beamline (POLAR) at Argonne's upgraded Advanced Photon Source. POLAR will provide high coherent x-ray flux with enhanced control of x-ray polarization, enabling dichroic imaging methods to investigate magnetic domains and electronic orbital anisotropies at the nanoscale. The project successfully developed measurement procedures, reconstruction algorithms, and interferometry for these imaging techniques. Dichroic ptychography, scanning transmission X-ray microscopy (STXM), and fluorescence datasets were acquired from single crystal $Nd_2Fe_{14}B$ pillars at the Velociprobe instrument at Advanced Photon Source and the NanoMAX beamline at MAX IV. The 3-dimensional magnetization was reconstructed using 2-dimensional image registration and subsequent 3-dimensional tomographic reconstruction from dichroic STXM data. Further development is underway to extend this to dichroic ptychography and fluorescence data, integrating the software into the POLAR beamline data pipeline. Interferometry was designed and configured offline for measurement strategy development and integration into the POLAR experimental setup. The project has developed software tools and experimental configurations that are now available for users to conduct dichroic imaging experiments at the nanoscale. The project has laid the groundwork for measuring polarization-dependent two-dimensional and three-dimensional ptychographic images and reconstructing domain patterns at the POLAR beamline.

LDRD # 2022-0026	Enabling Nanoprobe-Based in situ Multimodal Studies on Energy Materials
Investigators:	Sarah Wieghold Mathew J. Cherukara, Jörg M. Maser, Dina Sheyfer, Hoydoo You

This project focused on performing in-situ multimodal correlation studies at Advanced Photon Source Upgrade (APS-U) feature beamlines to capture the evolution of chemical and structural properties of functional energy materials and devices under various environmental and operational stimuli. The aim was to enable the first in-situ and multimodal experiments at these beamlines by developing in-situ sample cells compatible with various material systems and beamlines, revealing the underlying mechanisms of chemical and structural correlation through multimodal and multiscale analysis. Additionally, the project sought to develop a data workflow for acquiring and analyzing multimodal data sets.

The project successfully developed in-situ cells capable of capturing major contrast mechanisms under operating conditions of energy devices simultaneously, including X-ray fluorescence, X-ray Laue microdiffraction, X-ray beam-induced current, and X-ray excited optical luminescence (XEOL). Furthermore, a visualization package for multimodal data sets was created using a web-based interface built with Python. The novel design of the in-situ cells and the development of XEOL signal collection have enabled a comprehensive understanding of the composition-performance relationship via in-situ multimodal characterization approaches at new feature beamlines.

The accomplishments of this project have paved the way for future advancements in multimodal in-situ characterization capabilities, allowing for the understanding of structural, electronic, optical, and elemental properties under device operating conditions for the first time. The integration of artificial intelligence-assisted data acquisition workflows and multimodal visualization software will further enhance the ability to conduct nanoscopic in-situ multimodal characterization of beam-sensitive materials, mitigating radiation damage and maximizing information gain.

LDRD # 2022-0037	Intermittent Dynamics in Hard and Soft Materials Enabled by Advanced Photon Source-Upgrade
Investigators:	Suresh Narayanan Yue Cao, Wei Chen, Mathew J. Cherukara, Eric Dufresne, Xiao-Min Lin, Subramanian Sankaranarayanan, Qingteng Zhang

This project aimed to advance the understanding of intermittent fluctuations, known as "rare events" or "avalanches," in soft and hard condensed matter across various length scales. These rare events are critical to the synthesis and co-design of materials and microelectronics. The X-ray Photon Correlation Spectroscopy (XPCS) feature beamline at the Advanced Photon Source Upgrade (APS-U) provided enhanced coherence, smaller beams, and higher energies, offering a unique opportunity to study these nonequilibrium dynamics. The project successfully established a framework connecting molecular dynamics (MD) simulations and XPCS experiments, utilizing artificial intelligence and machine learning (AI/ML) to link forward and reverse models. This connection enabled the application of artificial intelligence/machine learning to interpret speckle patterns and two-time correlations, facilitating the understanding of real-space dynamics. Significant progress was made in developing algorithms for higher-order spatiotemporal correlations and streaming XPCS analysis, advancing the study of aging dynamics in soft colloidal glasses and driven phase transitions in transition metal oxide heterostructures. The project also achieved artificial intelligence/model classification of dynamics models and molecular dynamics simulations of shear response in colloidal suspensions.

LDRD # 2022-0048	Low Power 3-Dimensional CMOS Circuits Enabled by Printable Redox-Gated Transistors
Investigators:	Wei Chen Mones A. Omari, Yuepeng Zhang, Hua Zhou

This project successfully demonstrated the potential of redox-gated three-dimensional printed hybrid electronics as an innovative approach for creating flexible, low-power microelectronic devices. The aim was to overcome key limitations of traditional microelectronics fabrication, specifically targeting challenges in gating stability and operational efficiency within printed hybrid electronics systems. The project focused on using vanadium dioxide nanoparticles, known for their phase-change properties, and employed a custom-designed aerosol jet printing process to achieve low-voltage, stable switching in redox-gated vanadium dioxide transistors.

Key achievements included the synthesis of redox-functional vanadium dioxide inks and their precise application using aerosol jet printing, leading to the creation of solid-state vanadium dioxide transistors that operate reliably at sub-volt gating voltages. These printed films exhibited consistent conductivity modulation over more than 6,000 switching cycles without degradation, validating redox gating as a robust method to control electronic properties without compromising material integrity. Furthermore, the project demonstrated that aerosol jet printing enables high-resolution printing of complex, multilayer structures suitable for both flexible and three-dimensional substrates, expanding the versatility of printed hybrid electronics.

The outcomes of this work laid a critical foundation for using redox-gated nanoparticles in advanced, low-power electronic applications, including neuromorphic computing and bioelectronics. Insights into ink formulation, colloidal processing, and aerosol jet printing techniques provide a roadmap for advancing flexible electronics manufacturing, establishing a platform for energy-efficient, durable devices across emerging fields in wearable and hybrid microelectronics.

LDRD # 2022-0057	Resolving Capacity Fade in Lithium-Ion Batteries with Single-Shot Chemical Mapping
Investigators:	George E. Sterbinsky Jason R. Croy, Steven M. Heald, Juanjuan Huang, Adam P. Tornheim

In this project, we successfully developed an innovative instrument for rapid spatially resolved determination of atomic and chemical structure using a novel method of obtaining x-ray absorption spectra in a single shot. This method utilizes bent crystals to disperse polychromatic x-rays in both space and energy after interacting with a submicron region of a sample, with high spatial resolution achieved through x-ray mirrors. This approach differs from conventional dispersive x-ray absorption spectroscopy, where bent crystals typically perform both focusing and dispersion, potentially limiting spatial resolution. The instrument's ability to spatially map the chemical state of materials at high temporal rates offers new opportunities to gain insights into chemical processes in-situ and in-operando. One application of this technology is investigating the charge capacity-fade observed in cobalt- and nickel-free lithium-ion battery cathodes, which are more sustainable due to the scarcity and cost of these elements. Using this instrument at Sector 25 of the Advanced Photon Source, we conducted proof-of-principle experiments on cobalt-free transition metal oxide cathodes. A manuscript detailing this work has been submitted for publication. The project has advanced our understanding of battery cathode technologies and contributed to the development of high-capacity, low-cost battery technologies essential for meeting future energy demands.

LDRD # 2022-0228	Accurate Simulations for Modern Cosmological Surveys
Investigators:	Nicholas J. Frontiere Jeffrey D. Emberson

This project aimed to enhance the Argonne cosmological simulation efforts by leveraging exascale computing to incorporate sophisticated numerical treatments of baryons, neutrinos, and non-Gaussianity. These advancements were crucial for interpreting data from upcoming cosmology surveys, such as the Department of Energy Dark Energy Spectroscopic Instrument and the Legacy Survey of Space and Time. Over the course of the project, significant progress was made in several areas. We developed and calibrated hydrodynamic sub-resolution methods, created new techniques for simulating neutrinos, and explored primordial non-Gaussianity. Additionally, we focused on cross-correlation calibrations of subgrid models and implemented higher-order density measurements of non-Gaussianity. We successfully integrated the neutrino extension of the Hardware/Hybrid Accelerated Cosmology Code into the hydrodynamic framework, enabling fully self-consistent simulations of baryons, neutrinos, and dark matter. These efforts have positioned Argonne to meet the simulation challenges required for next-generation cosmology surveys, providing more accurate theoretical predictions and a deeper understanding of the universe's matter distribution.

LDRD # 2022-0229	Artificial Intelligence-based Models of the Galaxy-Halo Connection
Investigators:	Matthew R. Becker Andrew P Hearin

We developed artificial intelligence-based models to explore the connection between galaxies and halos in cosmological structure formation. Our innovative methodology enabled these models to scale to exascale high-performance computers, including hardware accelerators, and we successfully ran them on up to 20 percent of the Department of Energy’s Aurora Exascale Supercomputer. The models support automatic differentiation, allowing integration with data using advanced Monte Carlo sampling and optimization algorithms. This approach facilitated rich, multiwavelength predictions for cosmological sky surveys of the 2020s, enhancing their ability to constrain dark energy. Throughout the project, we published three papers detailing physically motivated models of galaxy star formation histories, developed a JAX-native code for computing differentiable spectral energy distributions, and introduced techniques for computing differentiable summary statistics of galaxies. We also advanced differentiable models for galaxy orbits and population-level star formation histories. We implemented production kernels for differentiable summary statistics and extended our star formation pipeline to model burstiness, crucial for analyzing the bluest objects. Scaling tests of our summary statistics kernels were conducted on Nvidia hardware.

LDRD # 2022-0230	Productive Exascale Analysis Workflows for Numerical Cosmology
Investigators:	Michael Buehlmann Kyle I. Chard

This project successfully developed a framework for interoperability between cosmology simulation and analysis workflows, addressing the challenges posed by the disparate nature of current analysis tools. By defining common interfaces and representations for data exchange, the project harmonized the landscape of tools and democratized access to analysis capabilities. A catalog of common tools was created for community use, facilitating seamless orchestration and execution of workflows by managing data flows between tools. Additionally, a novel continuous integration environment was established, enabling real-world testing of the products and ensuring robust interoperability between components through complete end-to-end workflows. The project leveraged forthcoming exascale computing resources at Argonne and other national laboratories, enhancing the ability to exploit large-scale data generated by next-generation cosmological surveys.

LDRD # 2022-0231	Artificial Intelligence-Enabled Analytics for Cosmology
Investigators:	Nesar S. Ramachandra Patricia R. Larsen

Novel tools for cosmological analysis at the intersection of machine learning, high-performance computing, and physical modeling were developed as part of this project. The advancement of artificial intelligence capabilities, alongside the expansion of numerical simulations and exascale computing, allowed us to leverage extreme-scale simulations for efficient use within the broader cosmology community. Most notably, we created astrophysical emulators that are used in creating synthetic sky catalogs. The project aimed to advance the understanding of the dark Universe and its contribution to cosmological expansion by combining high-performance computing and physical modeling with artificial intelligence. This approach was crucial for improving the measurement of dark-energy-related parameters to percent-level precision, a prime goal identified in the Period-5 report for U.S. investments in particle physics. Throughout the project, we expanded the utility of Argonne simulations by developing a state-of-the-art, scalable inference suite based on machine learning methods. This significantly enhanced the accuracy and detail of simulation predictions and cosmological parameter inference, meeting key requirements for upcoming Department of Energy cosmology surveys, including the Roman Observatory Legacy Survey of Space and Time and the next-generation cosmic microwave background experiment. Our efforts included data pre-processing, building a framework to integrate artificial intelligence with simulation outputs, and incorporating digital twins. We focused on surrogate model building at scale and implemented extensive validations, culminating in the deployment of scalable Bayesian inference tools and ex-situ trained digital twins into numerical simulations. These accomplishments represent a novel approach in scientific machine learning, with the potential to play a central role in principled applications of artificial intelligence in physical sciences.

LDRD # 2022-0232	Novel Energy-Resolving Quantum Detectors
Investigators:	Tomas Polakovic

The project aimed to advance the capabilities of superconducting nanowire detectors by developing a novel type of detector capable of resolving the energy of incoming charged particles. This was achieved by integrating conventional superconducting nanowire single photon detectors into layered dielectric-superconductor heterostructures. The early stages of the project involved designing and developing superconducting nanowire detectors specifically tailored for particle detection. We successfully completed the designs of sensors and dielectric structures for these energy-resolving detectors. A significant achievement was the high-yield, large-scale fabrication of functioning single-layer detectors, with up to 512 individual pixels on a 4-inch silicon wafer, marking a crucial step towards the realization of the proposed devices. Additionally, we demonstrated the detection of alpha particles from sealed sources with energies around 5 million electron volts and protons from the Fermi National Laboratory test beam facility at 120 billion electron volts. These results indicate the technology's applicability to nuclear physics and high-energy physics projects supported by the Department of Energy. The project also involved testing at the Fermilab Test Beam Facility, where ultra-relativistic protons were detected with nominally 100% internal detection efficiency. Materials studies of superconducting thin films on various dielectric substrates were conducted, showing promising prospects for the fabrication of layered detector structures using silicon nitride as the dielectric spacing layer. This technology promises to provide new experimental capabilities for nuclear physics experiments and has potential applications in quantum information sciences and novel microelectronics development.

LDRD # 2022-0233	Optical Time Projection Chamber for the Realization of a Ton-Scale Neutrinoless Double Beta Decay Demonstrator
Investigators:	Leslie Rogers

This project aimed to advance the detection of neutrinoless double beta decay ($0\nu\beta\beta$), a rare nuclear process that could redefine the standard model of particle physics by demonstrating that lepton number is not conserved. Detecting this decay would provide insight into the universe's matter abundance. Current detectors are small, but a ton-scale or multi-ton-scale detector is necessary for clear observation. The project focused on developing scalable technology using xenon gas, specifically by employing a fast optical camera instead of silicon photomultipliers for time-over-threshold and time-of-arrival readout. A key challenge was achieving background-free detection, which requires distinguishing double beta decay events from radiogenic background events. One promising method under development involves direct detection of the barium(2+) daughter nucleus. The project successfully installed a small time projection chamber and electroluminescence region at Argonne National Laboratory, equipped with a TimePix camera, image intensifier, and optics for focusing, which observed tracks from cosmogenic rays. Additionally, a 100-kilogram scale detector was commissioned in Spain, utilizing Argonne's scalable field cage and high voltage feedthrough designs. The project resulted in a patent application and two papers in progress. Future work will focus on measuring the efficiency of barium tagging and utilizing image recognition techniques to discriminate between double and single escape peaks. The project's accomplishments lay the groundwork for a ton-scale detector, potentially revolutionizing the search for neutrinoless double beta decay.

LDRD # 2022-0236	Improving Data Movement Performance for Emerging Artificial Intelligence and Climate Science Workloads on Future Supercomputers
Investigators:	Kevin A. Brown

This project focused on advancing the design of next-generation supercomputers to accelerate discoveries in artificial intelligence and climate science. We addressed performance issues in current supercomputers and simulated new system designs to overcome these challenges. A key achievement was the development of a new performance measurement technique that simplifies the collection of comprehensive performance data from Argonne supercomputers. Additionally, our simulation framework was enhanced to more accurately predict the behaviors of real-world supercomputers. We demonstrated the impact of various parallelization techniques on the Weather Research and Forecasting model, providing insights into optimizing data movement for artificial Intelligence and climate science workloads. Our work identified data movement bottlenecks and proposed new strategies to optimize these processes, leveraging smarter routing and faster storage solutions. Collaborations with Argonne National Laboratory postdoctoral climate researchers allowed us to evaluate novel high-resolution simulations of North America. We also extended the integrated analysis workflow using tools like ReFrame and BenchPro to study complex data movement patterns. Furthermore, we identified low-level metrics to explain adaptive routing behaviors on dragonfly networks, leading to improvements in network throughput during congestion. This project has laid the groundwork for future architectures that promise a 10–20% performance improvement for emerging workloads, reducing the time domain scientists and administrators spend on optimizing data movement and communication infrastructure.

LDRD # 2022-0351	Surrogate Models for Science Applications
Investigators:	Eliu A. Huerta

This project aimed to develop innovative, physics-informed artificial intelligence (AI) surrogates to address computational challenges in multi-scale and multi-physics phenomena, which are typically intractable with traditional high-performance computing simulations or artificial Intelligence methodologies that do not utilize extreme scale computing. Throughout the project's three-year duration, significant advancements were achieved in various scientific domains. In the first year, a computational framework was established by integrating resources such as the Argonne Leadership Computing Facility (ALCF), the ALCF AI-Testbed, and the Materials Data Facility, among others. This framework facilitated the creation and dissemination of FAIR (findable, accessible, interoperable, and reusable) artificial Intelligence surrogates, which were applied to accelerate high energy diffraction microscopy and solve partial differential equations in applications like coastal dynamics and tsunami modeling.

In the second year, the focus shifted to developing artificial Intelligence surrogates for two-dimensional magnetohydrodynamics and high-energy black hole collision simulations. These surrogates demonstrated superior computational accuracy and speed compared to state-of-the-art Bayesian methods, enabling new insights in astrophysics. Additionally, an end-to-end artificial Intelligence framework was developed for modeling small molecules, inorganic crystals, and metal-organic frameworks, with results published in peer-reviewed journals.

The third year introduced novel generative artificial Intelligence approaches for discovering metal-organic frameworks on supercomputing platforms, published in Nature Communications Chemistry. Furthermore, innovative methods for gravitational wave detection were developed, combining hybrid dilated convolution neural networks and graph neural networks to improve signal modeling and detection accuracy. This work was published in Machine Learning: Science and Technology.

Overall, the project showcased the potential of artificial Intelligence and extreme scale computing to revolutionize scientific research across multiple fields, providing tools and methodologies that are now accessible to the broader scientific community. Key publications and awards, such as the HPCwire Readers' Choice Award, underscore the project's impact and contributions to advancing artificial Intelligence applications in science.

LDRD # 2022-0367	Autonomous Discovery for Science
Investigators:	Ian T. Foster Dionysios A. Antonopoulos, Ilke Arslan, Peter H. Beckman, Millicent A. Firestone, Arvind Ramanathan, Casey Stone

This project has brought together teams across Argonne National Laboratory to work together on demonstrating autonomous discovery on selected driving science problems. As a result of the need to integrate artificial Intelligence generated hypotheses with automated experimental setups, the Rapid Prototyping Laboratory (RPL) was established. A significant task was to elevate the competency of staff related to laboratory automation and artificial Intelligence. To address automation, equipment was purchased that enabled software integration of devices and robots. To address artificial Intelligence, pilot projects in polymer chemistry and microbiology were selected.

LDRD # 2022-0401	Nano X-ray to Image Bacterial Cells at the Intracellular Level and Grasp Mercury Transformations in the Environment
Investigators:	Si Chen Marie-Pierre Isaure

The project aimed to develop an advanced method for imaging mercury at the nanoscale within single bacterial cells in three dimensions, to better understand the metabolic pathways of mercury. Researchers evaluated the role of various genes using BerOc1 mutants, a strain of mercury-methylating bacteria. The Bionanoprobe at the Advanced Photon Source was utilized for its subcellular spatial resolution and trace elemental sensitivity, essential for preserving the native state of bacteria during metal imaging. Cryogenic sample preparation was optimized to maintain the integrity of the samples, allowing for study at liquid nitrogen temperatures. The team successfully refined x-ray fluorescence microscopy and ptychography techniques for imaging individual bacteria. Ptychography was initially performed in defocus mode to identify individual cells, followed by quantitative x-ray fluorescence mapping to measure elemental information, including mercury. This combination of techniques enabled researchers to contextualize elemental distribution within the ultrastructure of cells. Additionally, mercury L3-edge nano-scale x-ray absorption spectroscopy was conducted at the European Synchrotron Radiation Facility and National Synchrotron Light Source II to analyze mercury ligands within cells and extracellular clusters. The research outcomes are expected to be applicable to other bacteria and microorganisms, potentially opening new avenues in biological and environmental sciences.

LDRD # 2022-0407	Developing Next Generation Tools for Large-scale Computational Science
Investigators:	Jonathan Ozik

This project focused on advancing computational models, which are essential for developing scientific insights. As high-performance computing (HPC) resources become increasingly powerful and computational models grow in complexity, there is a need for sophisticated algorithms and HPC workflow technologies to efficiently characterize model behaviors. The project aimed to extend the development and application of HPC-oriented statistical optimization algorithms to lower barriers to large-scale computational approaches across various scientific domains. A significant achievement was the organization of a three-day workshop at Virginia Tech, which served to finalize project outputs and explore future funding opportunities. The workshop featured two experts in surrogate models for simulation modeling. Additionally, the project produced several manuscripts and software tools, contributing to the scientific community's understanding of model exploration.

LDRD # 2023-0003	Novel Polymer Binders for Direct Cathode Recycling
Investigators:	Ilya A. Shkrob Albert L. Lipson, Lu Zhang

This project addressed the critical need for recycling nickel and cobalt from spent lithium-ion batteries, driven by global shortages and supply chain uncertainties. Traditional recovery methods involve hydro- or pyro-metallurgical processing, which decomposes cathode materials. In contrast, direct recycling rejuvenates metal oxide particles in the cathode without decomposition, allowing them to be reused after heat treatment. However, the commonly used fluoropolymer binder releases hydrofluoric acid during this process, interfering with the metal oxide particles. To solve this end-of-life problem, the project focused on developing polymer/solvent systems specifically designed to facilitate direct recycling without compromising electrochemical cycling performance. As the investigation progressed, the project was re-scoped to demonstrate and scale up a fluoropolymer removal method discovered by the team. This method uses tetramethylurea in methyl isobutyl ketone, with the former acting as a polymer release agent and the latter as a solvent for the fluoropolymer. The process was successfully demonstrated five times on a kilogram scale, achieving 95-99% fluoropolymer removal. Additionally, the reuse of the mixed solvent by distillation was demonstrated, and the polymer could be reused in the liquor for new ink formulation or removed as a solid. This approach also led to methods for recycling fresh cathode laminate trimmings and increasing the efficiency of hydrometallurgical treatment.

LDRD # 2023-0014	Microarchitected Composites for Decarbonization of High Temperature Processes
Investigators:	Mark C. Messner

The project successfully developed a novel ceramic scaffold/aluminum matrix composite with a reduced coefficient of thermal expansion compared to pure aluminum, addressing the need for multifunctional high-temperature materials in decarbonizing industrial processes such as steel and ammonia production. This new category of Microarchitected Composites was designed with tailored microstructures to withstand extreme conditions. The structure of the material was characterized using microscale computed tomography imaging at the Advanced Photon Source (APS), providing a detailed view of the as-manufactured material structure. The project demonstrated the feasibility of the material system and manufacturing technique by fabricating a demonstration material with a silicon carbide/silicon oxide scaffold infilled by silicon metal. Iterations on base and scaffold material combinations led to the selection of an aluminum matrix and a silicon carbide scaffold. Two forward modeling and optimization techniques were developed to identify optimal scaffold topologies, resulting in preliminary optimal structures.

LDRD # 2023-0015	Integrated Electric Power: Communications Interdependency Tool
Investigators:	Stephen M. Folga Brian A. Craig, Edgar C. Portante, David Sehloff, Leah E. Talaber, Prakash R. Thimmapuram, Sang-il Yim

The project aimed to develop an integrated tool to address the interdependencies between the electric power and telecommunications sectors, which are critical for national security preparedness. The focus was on creating a deployable web-based tool for simulating these interdependencies. Key accomplishments included the development of a power system model for the New England area, tested with the EPfast tool, and the creation of a generalized framework to facilitate data exchange between EPfast and TelcoFast, a telecommunications model. A comprehensive database of electric utility control center locations across the U.S. was developed, along with mapping to electrical buses. The project also identified communication modes supporting electric control room operations and completed spatial analyses to identify electric and telecom assets at risk from outages. A web application was developed for end-to-end simulation of interdependencies between power system and telecommunication assets. This project has laid the groundwork for understanding and mitigating the risks associated with the interconnectedness of these critical infrastructures.

LDRD # 2023-0029	Intelligent Detector Design with Edge Computing
Investigators:	Jinlong Zhang Alexander A. Paramonov, Marco Trovato, Angel Yanguas-Gil, Kazutomo Yoshii

This project focused on developing digital circuit designs for artificial intelligence-based intelligent detection algorithms, aimed at significantly reducing bandwidth requirements without compromising performance. The project successfully demonstrated the functionality of these algorithms using simplified detector data and initiated their translation into hardware description language codes. The team identified and utilized spiking neural network software and tools for development, creating hardware design blocks as fundamental components for digital spiking neural network architecture. This work addressed the challenges faced by future detectors across various fields, proposing a multi-step plan to employ artificial intelligence and machine learning for intelligent detector designs that enable rapid data reduction and adaptation to harsh detector environments. The project laid the groundwork for building full-chain capability for intelligent detector design at Argonne National Laboratory.

LDRD # 2023-0041	Discovering Biosignatures in the Transcriptional Response of Soil Microbes to Radiation
Investigators:	Daniel S. Schabacker Derek R. McLain, Justin C. Podowski

Microorganisms are ubiquitous in the environment and have the potential to serve as sensors for changes in any environment. In this project, we investigated the use of whole community ribonucleic acid (RNA) sequencing of soils to detect biosignatures for low dose ionizing radiation. We designed a method to expose soil to very low doses of radiation over long periods, modeling real-world scenarios for exposure. Through total community RNA sequencing, we observed changes in transcription related to metal uptake and metal cofactor use, with iron uptake and genes with iron cofactors downregulated, and genes with copper cofactors upregulated. These changes are likely related to reactive oxygen species generation and align with well-understood phenomena in pure culture studies. The biosignature genes associated with these changes allow for a limit of detection that is orders of magnitude lower than DNA-based methods for biosignature detection. Our accomplishments included the development of experimental designs and fabrication of exposure platforms for consistent and reproducible experiments. We refined methods for RNA extraction from soils, resulting in a reproducible protocol for extracting high-quality RNA. Preliminary experiments identified transcriptional changes in soil communities and the doses of ionizing radiation necessary to elicit these changes.

LDRD # 2023-0051	A Coating Strategy to Reduce Reliance of Nuclear Technologies on Critical Materials
Investigators:	Yinbin Miao Ahmed Abdelhameed, Sumit Bhattacharya, Edward A. Hoffman

This project aimed to establish a framework for evaluating coating solutions to accelerate the design, optimization, and qualification of advanced coatings, thereby reducing the nuclear industry's reliance on critical materials. The project successfully demonstrated Argonne National Laboratory's capabilities using coated stainless steel for molten salt reactors as a case study. In the first year, the team developed three key components of the framework: experimental approaches identified a promising multilayer coating solution, simulation approaches established a multiphysics model covering essential performance phenomena, and techno-economic analysis approaches developed a simplified cost model to assess coating solutions and their impact on reducing critical material utilization. In the second year, these components were further refined and integrated into a comprehensive evaluation framework. Enhancements included improving the multilayer coating design, upgrading the multiphysics model to 3-dimensional with additional physical phenomena, and establishing a nanoindentation model to connect measurement results with coating modeling parameters. The integrated framework was tested, and the principal investigators are now promoting these achievements to potential sponsors. This project has laid the groundwork for reducing reliance on critical materials in nuclear reactors.

LDRD # 2023-0054	Layered Transition Metal Dichalcogenide Barriers for Interconnects
Investigators:	Jeffrey W. Elam Charudatta M. Phatak

This project has successfully advanced the development of layered transition metal dichalcogenides (LTMDs) as diffusion barriers for copper interconnects in next-generation microelectronic circuits. Recognizing the limitations of conventional tantalum nitride barriers, we focused on ultra-thin, uniform, and conformal LTMD materials to address the challenges posed by high aspect ratio complex structures. Through innovative atomic layer deposition (ALD) techniques, we synthesized LTMDs such as MoS_2 and HfS_2, achieving precise control over their thickness and properties. Our comprehensive characterization using X-ray photoelectron spectroscopy, spectroscopic ellipsometry, and buffer oxide etch tests provided valuable insights into the suitability of LTMDs as diffusion barriers and seed layers for emerging metal interconnects. The project also leveraged in-situ transmission electron microscopy and the Advanced Photon Source for operando studies, enhancing our understanding of electromigration and material interactions. Overall, this project lays a strong foundation for the integration of LTMDs in future microelectronics, offering promising solutions for low line resistance and enhanced reliability in complex circuit designs.

LDRD # 2023-0073	Affordable Ultrafast Photomultiplier with 3-Dimensional Printed Microchannel Plates
Investigators:	Junqi Xie Dean Walters, Lei Xia

This project focused on the development of a novel photomultiplier tube (PMT) utilizing 3-dimensional-printed microchannel plates (MCPs). The aim was to create a cost-effective MCP-PMT device with performance comparable to existing commercial options. During the project, a dedicated fabrication facility for 10x10 cm air-sensitive devices was commissioned. Key advancements included the development of processes for photocathode growth and vacuum sealing essential for detector fabrication. Initial tests demonstrated successful pulse signal acquisition from the 3-dimensional-printed MCPs under high voltage conditions. The project holds significant promise for advancing photosensor technology, particularly in nuclear physics, by leveraging cutting-edge 3-dimensional printing techniques to produce affordable MCP-PMTs for large-area coverage.

LDRD # 2023-0086	Gas Turbine Combustor Performance Studies Using X-ray Diagnostics
Investigators:	Brandon A. Sforzo Christopher Powell

The Gas Turbine Combustor Performance Studies project successfully advanced experimental capabilities at our research facility, focusing on the atomization of gas turbine sprays using low- and zero-carbon fuels. Significant milestones were achieved, including the installation of critical power equipment and the fabrication of a low-pressure test rig, which facilitated the acquisition of x-ray imaging data under relevant conditions. These preliminary results demonstrated the diagnostic capabilities and provided valuable insights into the behavior of aviation fuels. The project also addressed potential noise and vibration impacts through successful characterization experiments.

LDRD # 2023-0090	Decarbonization of Magnet Recycling: Electrorefining of Rare Earths from Magnet Swarf and Magnet Scrap
Investigators:	Matthew R. Earlam Yuzi Liu

This project focused on the electrorefining of light rare earth metals, specifically neodymium, from magnetic scrap using a novel electrolysis process in a fluoride bath. The aim was to recycle neodymium from scrap magnets, avoiding the conventional solvent extraction method, which produces significant carbon dioxide emissions. The process operates in a molten fluoride salt and produces a liquid metal product. Initial experiments utilized commercial magnets, revealing that neodymium could be effectively removed from the magnets in a molten neodymium fluoride-lithium fluoride bath at 1000 degrees Celsius, with over 84% of the available neodymium extracted in some sections. The project successfully demonstrated the recovery of neodymium from magnet pieces and swarf provided by MP Materials. A ring magnet was electrorefined, and neodymium metal was collected at the cathode. The project also involved constructing a siphon to remove metal from the cell, avoiding manual ladling. The project has laid the groundwork for scaling up the process and comparing its economic impacts to conventional recycling methods.

LDRD # 2023-0097	Tuning Bacteriorhodopsin for De Novo Applications using Machine Learning and Automated Laboratory Approaches
Investigators:	Rosemarie Wilton Gyorgy Babnigg, Philip D. Laible, Jamie C. Overbeek, Elena A. Rozhkova, Richard D. Schaller

In this project, we developed methods to combine machine learning-directed protein engineering with autonomous laboratory approaches to design proteins and enzymes with optimized activities and biophysical characteristics. Our focus was on bacteriorhodopsin, a photoactive transmembrane protein pump from Halobacterium salinarum, which holds significant potential for bioelectronic and biohybrid energy capture devices. We successfully automated several molecular biology protocols using Opentrons robots, including protein purification via Nickel-affinity chromatography, plasmid preparations, Gibson assembly of plasmid vectors, and the setup of protein expression reactions using in-house produced cell-free expression reagents. These advancements allowed us to progress from machine learning-directed library design to the expression and characterization of a 48-member library within approximately one month. This achievement lays a solid foundation for the full automation of protein design, expression, and characterization processes.

Throughout the project, we constructed expression vectors for both cell-free and cell-based bacteriorhodopsin expression using automation-friendly designs and methods to facilitate planned library cloning. We established a cell-free expression system, producing all extracts and reagent solutions in-house, and demonstrated E. coli-based expression of model bacteriorhodopsin homologs. Automated purification of bacteriorhodopsin expressed in E. coli culture was successfully implemented using OT-2 robots. Additionally, we utilized SCHEMA software tools to design chimeric libraries and developed scripts to convert output to codon-optimized DNA sequences compatible with cloning methods. A 96-member bacteriorhodopsin chimeric library was designed and ordered, with plans for cloning into expression vectors. We also set up an apparatus for functional assays of bacteriorhodopsin's proton pumping capabilities and developed a high-throughput dye efflux assay to monitor proton pumping, which is currently undergoing optimization.

These accomplishments have positioned bacteriorhodopsin variants for follow-on funded research in bio-nano hybrid assembly, and the techniques developed are extensible to other enzyme systems for diverse applications in energy systems and synthetic biology.

LDRD # 2023-0101	Multiscale Modeling of Carbon Dioxide Mineralization Reactions in Porous Media
Investigators:	Peter Zapol Getnet D. Betrie, Paul A. Fenter, Zachary D. Hood, Sang Soo Lee

This project aimed to explore long-term carbon dioxide storage using basalt minerals and the conversion of carbon dioxide into useful chemicals. We focused on developing predictive models for carbon dioxide reactivity, integrating lab-scale experiments and multiscale computational modeling. Our efforts included the creation of novel electrocatalysts based on MBenes for synthesizing chemicals like urea, targeting circular economy development. We measured reaction outcomes in rock-carbon-dioxide-water systems using X-ray computed tomography and conducted initial network analysis. Quantum mechanics calculations provided reaction energies at mineral-water interfaces, informing continuum scale models. Additionally, digital holography experiments revealed variations in surface morphology under flow conditions using a single channel cell. Our work demonstrated the potential for large-scale carbon dioxide conversion into stable minerals, leveraging high-performance computing for multiscale modeling and providing quantitative comparisons with experimental data.

LDRD # 2023-0118	Economical Approach to Produce Large Scale Nuclear Grade Oxygen-Free Beta-Silicon Carbide Fiber
Investigators:	Sumit Bhattacharya Zhigang Mei

Our project successfully developed an alternative route for manufacturing β-phase silicon carbide (SiC) fibers, a critical material for high-temperature applications in aerospace, energy, and nuclear industries. We confirmed that our innovative high-temperature direct carbon-silicon interaction method outperforms conventional carbothermal reduction in generating the β a carbide phase. This method leverages the strong adhesion and high wettability of silicon melt on amorphous carbon surfaces, facilitating the production of homogeneous SiC fibers. Our experiments demonstrated that direct C-Si reactions at 1460°C lead to the formation of β-phase SiC, with significant grain coarsening observed after 24 hours, resulting in grain sizes between 300 and 500 nm. This advancement addresses the limitations of polymer-derived β-phase SiC fibers, which suffer from nanocrystalline structures that degrade performance under long-term exposure. The promising results have led to an intellectual property application. This breakthrough paves the way for an economical and scalable production process, reducing dependency on costly foreign-manufactured fibers and supporting the expansion of SiC applications in clean energy production.

LDRD # 2023-0131	Environmental Sensors Based on Solid-State Electrolyte Devices
Investigators:	Zachary D. Hood Shiba Adhikari, Jianguo Wen

Solid-state electrolyte-based sensors for detecting environmental pollutants like sulfur oxides are urgently needed for the sake of environmental protection. As of now, miniaturized sensors for the detection of sulfur oxides rely on high-temperature operation (greater than 500 degrees Celsius) because of the use of solid electrolytes that have relatively low room-temperature ionic conductivity, resulting in sluggish detection and regeneration of the sensor. This project has successfully advanced Argonne's expertise in the electrochemical detection of sulfur oxides at lower temperatures, leading to groundbreaking developments in compact sensor technology. By utilizing lithium borate ceramics as solid electrolytes, the project has created state-of-the-art sensors that offer a cost-effective alternative to traditional IR- or UV-based sensors. The successful synthesis of 100 grams of lithium borate and its transformation into pellets laid the foundation for solid-state gas sensors, demonstrating enhanced sensitivity to sulfur oxides.

LDRD # 2023-0134	Electrochemical Continuous Flow Reactor for Sustainable Aviation Fuel Generation
Investigators:	Zachary D. Hood Shiba Adhikari, Jianguo Wen

This project has achieved remarkable success, advancing the development of efficient and cost-effective electrocatalysts for SAF production. By simplifying catalyst synthesis methods, the project has streamlined the production process, significantly enhancing the selectivity of catalysts for valeric acid and n-octane during the electroproduction of levulinic acid. These advancements have improved the efficiency and performance of the electrochemical process at the lab scale. A pivotal collaboration with the Center for Nanoscale Materials (CNM) commenced in May 2023, providing critical S/TEM analysis that enriched the characterization of the electrocatalysts.

LDRD # 2023-0149	Mitigating the Corrosive Effects of Molten Salts Through Multi- layer ALD Coatings for Optimal Material Passivation
Investigators:	Elizabeth D. Laudadio

The project aimed to enhance corrosion resistance in highly corrosive environments, such as those found in molten salt reactors and concentrated solar power generators, by employing multiple layered materials. The research systematically investigated the effects of layer material composition, thickness, and the number and order of layers. Key technical achievements included the use of atomic layer deposition (ALD) with ozone generation to prepare molybdenum oxide thin films, and the acquisition and installation of advanced analytical tools such as an ellipsometer for thin film thickness analysis and a Fourier transform infrared (FTIR) spectrometer with a liquid-nitrogen filled detector for enhanced sensitivity. A protocol for x-ray photoelectron spectroscopy (XPS) was also established to analyze the thickness and composition of thin films after corrosion, with results showing agreement between ellipsometry and XPS analysis.

The research revealed that molybdenum oxide films were not effective corrosion blockers in chloride or nitrate salts, contrary to initial hypotheses. Instead, titanium dioxide films demonstrated superior corrosion resistance, maintaining strong surface integrity even after 10 days of exposure, as evidenced by XPS analysis. This suggests that titanium dioxide is a viable option for blocking salt corrosion. Future work should explore the use of gas diffusion blocking materials, such as silicon dioxide or zirconium oxide, in conjunction with titanium dioxide, and investigate various thicknesses and layer configurations to optimize corrosion protection. The project has laid the groundwork for expanding these model studies into real molten salt systems, with potential applications in enhancing the durability of materials used in manufacturing and infrastructure exposed to corrosive environments.

LDRD # 2023-0152	Synergistic Design of Sustainable Energy Materials: Merging Data Science, Synthesis, and Computation for Energy-Efficient Technologies
Investigators:	Gordon Peterson

The project aimed to develop sustainable solid-state materials for energy applications by integrating synthetic exploration, electronic structure calculation, and machine learning. The focus was on discovering novel thermoelectric materials, which are pivotal for energy recovery and serve as ideal systems to study the relationships between crystal structure and physical properties. Throughout the project, several thermoelectric families, including half-Heuslers, Zintl phases, and sulfosalt minerals, were investigated for their promising physical properties and potential for chemical tuning to reduce environmental impact. A significant achievement was the application of synchrotron X-ray diffuse scattering and three-dimensional pair distribution function (3-dimensional-ΔPDF) analysis to understand nanoscale ordering in structurally complex thermoelectric materials. Machine learning models were developed to predict thermoelectric properties, and large single crystals of various materials were synthesized for properties analysis. Collaborations were established to enhance crystal growth, structural analysis, and electronic structure calculations. The project successfully demonstrated the utility of a synergistic approach to materials design, identifying new materials with high thermoelectric figures of merit and contributing to the development of eco-friendly energy solutions.

LDRD # 2023-0153	Decarbonized Processes for use with Intermediate-Temperature Process Heat
Investigators:	Krista L. Hawthorne Nathaniel C. Hoyt

This project focused on developing novel intermediate-temperature industrial processes that can utilize novel process heat sources, such as concentrating solar power and advanced nuclear reactors, for use by the manufacturing sector. The project successfully identified and evaluated processes for ammonia and iron production. A liquid-metal mediated electrochemical route for ammonia synthesis was developed, combining aspects of existing synthesis routes to eliminate inefficiencies. An alternate flow-system arrangement using an upper storage vessel was designed to minimize back-reaction during the process. Chemical reaction products and yields for each reaction step were evaluated to understand inefficiencies and identify process conditions that minimize side reactions, resulting in efficient ammonia production. Additionally, the project expanded to include the development of a new chemical process for iron production and the investigation of using the electrochemical hydrogen evolution reaction in a molten salt electrolyte to produce hydrogen as a feedstock for testbed processes. The iron process involves dissolving iron oxide in a molten hydroxide salt and producing iron deposits at an inert electrode using electrolysis.

LDRD # 2023-0157	Polarized Ion Sources for Electron-Ion Collider (EIC) Science
Investigators:	Chao Peng

We successfully developed a prototype of polarized Lithium-6 and Lithium-7 sources to support the nuclear science program of the U.S.-based Electron-Ion Collider (EIC). This prototype produces a non-divergent and uniform beam of lithium atoms, which is essential for expanding the scientific reach of the EIC project to study the partonic structure of nucleons or light nuclei embedded in a nuclear medium. To achieve this, we finalized designs for the oven and nozzle, conducted Computational Fluid Dynamics (CFD) simulations to optimize the nozzle's inner contour for uniform beam generation, and constructed a platform for beam profile measurement. Two hot-wire scanners were developed and installed to determine the beam profile, and the measured profile data were used to benchmark the beam simulation. Additionally, we acquired a single-frequency tunable solid-state laser with a center wavelength of 671 nm to study the polarization of the beam.

LDRD # 2023-0159	Toward Scalable Superconducting Nanowire Detector Platform
Investigators:	Valentine Novosad Whitney R. Armstrong, Tomas Polakovic

The project successfully advanced the development of large-area superconducting detectors with single photon detection efficiency, addressing key challenges in scalability and readout efficiency. A state-of-the-art cryogenic testbed, capable of high-channel optical and electrical access for advanced superconducting device research, was procured and installed. This testbed operates at 1 Kelvin and is equipped with multi-channel superconducting cables and low noise amplifiers, enabling continuous operation and testing of large area samples. A novel binary branching readout scheme was developed, significantly reducing the number of required connections and demonstrating the potential to scale to millions of pixels with minimal wiring. This innovative multiplexing method has the potential to revolutionize superconducting detector applications by enabling faster, more efficient, and large-scale sensor networks. The project's success enhances Argonne National Laboratory's capabilities in superconducting detector research and development, contributing to key research areas such as Quantum Information Science, basic energy science, microelectronics in extreme environments, fundamental nuclear physics, and high-energy physics experiments. The project also laid the groundwork for developing fabrication processes for superconducting nanowires, demonstrating basic operational principles and assessing scalability potential.

LDRD # 2023-0161	Correlated Extreme Weather Risks to Energy Network Infrastructures
Investigators:	Zhi Zhou Akintomide A. Akinsanola, William N. Mann, David Sehloff, Eric R. Tatara

This project focused on developing a modeling workflow to assess the simultaneous risks posed by extreme weather to the interdependent electricity and natural gas transmission systems, which are increasingly interconnected. The project extended existing tools to model the probability of component outages on both gas and electric grids. The interdependency of these networks was modeled with consideration of risks from extreme weather-induced outages. In the first year, the team collected essential data, including climate, power system, and gas network operational data, and established a conceptual model and workflow. This work was presented at two conferences with a case study. In the second year, the team completed the workflow and conducted analysis using a stylized Independent System Operator-New England (ISO-NE) system as a test case. The results were presented at the Federal Energy Regulatory Commission (FERC) Technical Workshop 2024 and discussed with sponsors and collaborators from other national labs, universities, and industry organizations.

LDRD # 2023-0176	Development of National Future Flood Prediction Artificial Intelligence Tool and Improvement of Predictability of the Hydrologic System
Investigators:	Jeremy Feinstein Getnet D. Betrie, David Coe, Yuejun Eugene Yan

This project aimed to develop advanced artificial intelligence capabilities for national flood prediction, focusing on high-resolution mapping, computer vision technologies for flood observation, and assimilation techniques to enhance physics-informed modeling with observational data. During the first year of the project, we evaluated state-of-the-art hydrodynamic models for software selection and designed synthetic physical experiments for large data generation. It also developed and trained models for shallow water simulation using graph neural networks and advanced sampling techniques, and established workflows for collecting and generating flood observation data. Manual ground truthing of flood observation data from numerous storms in the southwestern United States was also completed. Building on these achievements, the second year of the project saw the development of a hydrodynamic model within artificial intelligence frameworks for portable model training, expansion of candidate neural networks for shallow water simulation, and completion of regional models for identifying flood pixels from various remote sensing inputs.

The project delivered significant scientific and technological advances, integrating artificial intelligence satellite data processing, high-resolution hydrodynamic modeling, and automated deep learning for flood prediction. These efforts addressed technical barriers such as poor resolution modeling, high computation costs, and scarcity of on-ground flood monitoring systems. Test cases demonstrated the tool's capabilities in distinct regions, including New York City and Chicago. The project showcased surrogate modeling capabilities in a manuscript and computer vision models for detecting flooded pixels using satellite technology. Additionally, a hydrodynamic assimilation graph neural network will be developed to correct flood predictions made by artificial intelligence surrogates using computer vision-detected floods.

LDRD # 2023-0177	Characterizing and Quantifying Dynamic Changes at Metal-Sulfide Electrolyte Interfaces for All-Solid-State Metal Batteries
Investigators:	Ying Shirley Meng

This project focused on the development of advanced cryogenic analytical electron microscopy (cryoAEM) tools, specifically the Analytical Electron Optical Beam line prototype, to perform metrological measurements at both the atomistic and macroscopic levels. The project aimed to apply these tools to study interfaces between reactive metal electrodes and sulfide-based or halide-based solid electrolytes for all-solid-state batteries.

The project had four key goals, three of which were related to instrumental and metrological developments, while the fourth involved applying these tools. The instrumental development milestones were successfully achieved and surpassed. These included: (1) creating workflows and tools for inert specimen preparation and transfer without environmental contamination under cryogenic conditions, along with new analytical characterization tools; (2) achieving ultra-high electron spectroscopy energy resolution of less than 20 millielectronvolts for nanoscale analytical characterization; and (3) developing ultra-low dose, high-resolution imaging at cryogenic temperatures.

The fourth goal, dynamic studies of lithium-ion transport across lithium-sulfur solid-state interfaces, faced limitations due to the absence of suitable inert/cryo-focused ion beam preparation facilities. Instead, the project pivoted to focus on preparing, transporting, and studying lithium and sodium reactive metals, their fluorides, oxides, and carbonates using the cryoAEM system. These materials are critical components of the solid electrolyte interphase in various battery. The successful completion of this substituted task demonstrated the effectiveness of the developed tools and methods.

LDRD # 2023-0179	Selective Electrochemical Reduction of Nitrate to Value-Added Products using a Reactive Electrochemical Membrane System
Investigators:	Jeffrey W. Elam

Nitrate contamination is a significant issue affecting groundwater worldwide, primarily due to fertilizer use in agriculture. In Illinois, the largest producer of soybeans and the second largest producer of corn in the United States, agricultural runoff has led to nitrate pollution in drinking water supplies, posing challenges for small communities that lack resources for effective treatment. Traditional nitrate removal methods, such as ion exchange, are costly and inefficient, as they merely transfer nitrate to a disposable adsorbed phase. This project aimed to develop reactive electrochemical membrane technology to selectively convert nitrate into nitrous oxide, a value-added product, offering a sustainable solution for local agricultural communities.Throughout the project, we successfully synthesized and characterized a series of palladium/indium (Pd/In) bimetallic catalysts on titanium dioxide (Ti_4O_7) membrane substrates. Additionally, we employed atomic layer deposition (ALD) processes to synthesize nickel oxide (NiO), iron oxide (Fe_2O_3), indium oxide (In_2O_3), and palladium (Pd), and calculated the structures and nitrous oxide binding energies for a series of palladium/copper (Pd/Cu) bimetallic catalysts using density functional theory (DFT). These advancements laid the groundwork for optimizing the Pd/In catalysts for enhanced electrochemical conversion of nitrate and increased selectivity towards nitrous oxide. The project has paved the way for further development of ALD catalysts with varied compositions, including palladium, nickel, cobalt, copper, and indium, to maximize efficiency and selectivity in nitrate conversion processes.

LDRD # 2023-0180	Predicting Hyperlocal Air Pollutant Concentrations Using Artificial Intelligence at the Edge
Investigators:	Nicola J. Ferrier Seongha Park

This project aimed to explore the use of proxy methods to estimate local air quality using cameras and artificial intelligence, potentially replacing the need for expensive air quality sensors. The focus was on developing artificial Intelligence methods to analyze camera data and predict traffic-related pollution concentrations, leveraging the Argonne National Laboratory's Waggle edge computing system and Sage node network. The project sought to complement the sparse network of regulatory pollution monitors maintained by the U.S. Environmental Protection Agency, offering a scalable system for hyperlocal pollution estimates. The team collaborated with the CROCUS project to deploy sensors co-located with cameras in urban settings, enhancing data collection efforts. artificial Intelligence methods for vehicle detection and classification were developed, although the first version required significant computing resources, making it impractical for deployment alongside air quality sensors. The project demonstrated potential for commercialization through a platform/application model similar to smartphones, with diverse applications available through an "app store."

LDRD # 2023-0181	High-Throughput Predictions of Polymer Fate and Effects
Investigators:	Kevin Hickey

Accumulation of petroleum-based waste plastics in the environment poses a significant global challenge, and there is a lack of data to inform the design of alternatives that are more environmentally benign at the end of life. This project aimed to develop and test quantum chemistry and machine learning techniques to predict properties influencing the fate and effects of plastics, guiding the screening of candidate polymers for more sustainable design. Key properties studied included mobility, bioaccumulation, persistence, and toxicity, which are critical drivers of polymer effects in the environment. Persistence models were developed by focusing on glass transition temperature and specific surface degradation rates to quantify the stability and degradation of plastics. Successful models were produced, with transition temperature modeled using both quantum chemistry and machine learning methods, and specific surface degradation rates modeled with machine learning. The transition temperature work resulted in a publication in Computational Materials Science. Toxicity models leveraged transfer learning frameworks with neural networks to predict toxic endpoints like median lethal dose values and hazardous concentrations, with promising results and a publication in preparation. Mobility and bioaccumulation were successfully modeled through quantum chemistry calculations of Abraham parameters to predict polymer hydrophobicity.

LDRD # 2023-0236	Augmenting Earth System Modeling Using Machine Learning
Investigators:	Troy Arcomano

Our project has made significant strides in addressing the challenge of uncertainty in earth system model projections by developing a hybrid modeling approach that integrates machine learning with the Department of Energy's Energy Exascale Earth System Model (E3SM). This innovative approach aims to reduce biases and enhance computational efficiency in climate simulations. In the initial phases, we successfully created a machine learning-based emulator of the atmosphere, utilizing high-resolution ERA5 data for training and evaluation. Our exploration of state-of-the-art machine learning architectures, such as graph neural networks and transformer-based models, demonstrated their scalability and forecast accuracy at the required resolutions. By incorporating methodologies from recent studies, we developed a hybrid atmospheric model capable of replicating nonstationary climate conditions. This model, trained on historical data and using sea surface temperatures as a control parameter, accurately reconstructed observed trends and extended these predictions into future periods.

LDRD # 2023-0363	A Collaborative Decarbonization Assessment Platform for Accelerating Energy Transitions in Line with Science-Based Targets
Investigators:	Bruce P. Hamilton

This research succeeded in developing enhanced capability within Argonne to apply the TIMES-VEDA Online analytical framework for collaborative energy system planning and evidence-based decision-support on pathways to manage the energy economy, enhance energy security, and spur economic growth. The project established a solid foundation for providing global energy planning technical assistance with a core team of Argonne staff skilled in applying the TIMES-VEDA Online energy system modeling tool, access to high-quality country-level data sources, and required software licenses procured to enable use by members of the Argonne global energy system analysis team.

LDRD # 2024-0073	Development of a Physics-informed Artificial Intelligence/Machine Learning Methodology for Gamma Source Characterization for Nuclear Material Accountancy
Investigators:	Lander Ibarra Patrick M. De Lurgio, Roberto Ponciroli, Patrick C. Shriwise

This project successfully developed a comprehensive framework for monitoring special nuclear materials and localizing gamma radiation sources using commercially available sensor technologies. A key outcome was the creation of a detailed database containing gamma source spectra, which includes measurements from various gamma sources at different incidence angles and with different detector types, such as Cadmium Zinc Telluride (CZT), High-Purity Germanium (HPGe), and Sodium Iodide (NaI). This database, stored in the Argonne GitLab repository, provides essential documentation and experimental data in XML-based N42 and CSV formats. Additionally, a digital twin database was assembled, containing models and scripts for validating OpenMC simulations against experimental data, which are crucial for training convolutional neural networks (CNNs).

The project also developed an initial CNN to reconstruct gamma fields from source to detector. The framework was designed to support nuclear material accountancy and monitoring in facilities like molten salt microreactors and nuclear reprocessing plants. It was developed through a series of tasks, including the creation of a digital twin and training data, the development of a physics-informed artificial intelligence/machine learning framework, and the execution of multi-source experimental measurements. The final task demonstrated the framework's applicability by validating its predictions against high-fidelity Monte Carlo simulations. This innovative approach offers a cost-effective monitoring method using a sparse network of inexpensive sensors, paving the way for enhanced non-proliferation applications.

LDRD # 2024-0268	Characterizing Auger-Meitner Emitting Isotopes
Investigators:	Patrick R. Stollenwerk Jerry A. Nolen, Jr.

We successfully completed the design and testing for integrating a radioactive beam source of xenon-131 metastable isomer (Xe-131m) into the Argonne Auger-Meitner Radioisotope Microscope (AARM). This instrument is unique in its ability to simultaneously measure electron multiplicity and energy spectra during Auger cascades. The integration of a radioactive source marks a significant advancement in the application of AARM for measuring general radioisotopes, particularly those with medical relevance. Auger-Meitner emitters, known for releasing low-energy electrons in large quantities, hold promise for targeted radionuclide therapy in cancer treatment due to their localized damage to cancerous tissue DNA. However, predicting therapeutic effects based on emitter properties remains challenging, with few direct experimental measurements available to validate existing models. Our project addressed this gap by developing data analysis techniques for the new instrument. The cryogenic beam source, which entrains laser-ablated radioisotopes into a 4 Kelvin helium beam, was optimized for efficient isotope introduction into the microscope. In parallel, we characterized the electron multiplicity and energy spectrum of Xe-131m using a similar atomic beam source, demonstrating AARM's capacity to characterize radioactive emitters and accelerating the path toward medical isotope measurements.

LDRD # 2024-0271	Auger-Meitner Electron Specific Radiobiology Studies
Investigators:	Jerry A. Nolen, Jr. Harry C. Fry, Amy Renne

The project aimed to develop a peptide-based delivery system for an Auger electron-emitting radioisotope, specifically targeting prostate cancer tumors and localizing into the tumor cell nucleus.

Researchers at the Argonne Center for Molecular Engineering synthesized a nuclear localization sequence peptide derived from the simian virus 40 and attached it to a molecule known for delivering radioactive isotopes into prostate cancer cells. The promising Auger electron isotope, terbium-155, was successfully attached to the dual-function molecule. This initial success set the stage for in-vitro studies to evaluate the efficacy of the nuclear localization sequence peptide in delivering terbium-155 into the nucleus of prostate tumor cells. The project marked a significant step towards enhancing the therapeutic benefits of Auger-electron-emitting radioisotopes. The team developed methods to prepare various nuclear localization sequence peptides, enabling comparisons of their effectiveness in targeting the cancer cell nucleus.

LDRD # 2024-0273	Entanglement of Two Electrons on Solid Neon
Investigators:	Xinhao Li Qianfan Chen, Xu Han, Dafei Jin

We designed, fabricated, and tested novel superconducting devices to demonstrate qubit entanglement using electrons on neon charge qubits. Our project focused on characterizing the performance and noise properties of these electron qubits, achieving two-qubit coupling to the same resonator with independent tunability. In recent experiments, we characterized the noise spectrum of the electron-on-solid-neon (eNe) charge qubit using dynamical decoupling methods and long-term Ramsey measurements, both when qubits were biased on and off the charge sweet spot. We explored coherent interfaces between charge qubits on solid neon via near-range Coulomb interaction, observing strong exchange coupling between two charge qubits. Notably, one qubit (Qubit A) strongly coupled to a superconducting resonator, while the other (Qubit B) did not. These findings pave the way for scaling up electron-on-neon qubits and performing multi-qubit gates. Our work included the development of high-impedance titanium nitride superconducting resonators for enhanced eNe coupling strength and stability, and demonstrated dispersively strong-coupled two eNe charge qubits, achieving two-qubit gates such as iSWAP. We gained new insights into the coherence and noise properties of eNe charge qubits, contributing to a robust and scalable circuit quantum electrodynamics platform for developing eNe spin qubits for universal quantum computing.

LDRD # 2024-0274	Superconducting Radio-Frequency Switch Development
Investigators:	Amy N. Bender Thomas W. Cecil

The project aimed to develop a superconducting radio frequency switch utilizing nanowire technology, which holds significant potential for enhancing cryogenic characterization capabilities of radio-frequency detectors and accelerating their development process. The project progressed in three key areas. Firstly, a numerical model of the thermal behavior of the nanowire region was developed in collaboration with a summer Science Undergraduate Laboratory Internship (SULI) student. This model was designed to assess the impact of nanowire geometry on performance, and substantial progress was made, with a clear path to completion identified. Concurrently, an electromagnetic simulation of the junction region of the switch was created using commercial software, providing insights into the radio-frequency propagation of the design. These tools are essential for understanding experimental measurements and optimizing switch performance.

Secondly, multiple device designs and a fabrication layout were developed. The feasibility of using electron-beam lithography for fabrication was evaluated, and the layout was updated accordingly. Lastly, an experimental setup for cryogenic characterization of the switches was constructed. Fabrication of the initial design was planned to occur shortly, with subsequent characterization to follow.

The project aimed to enable cryogenic multiplexing of multiple microwave kinetic inductance detector (MKID) chips on a single coaxial cable, accelerating the characterization of kinetic inductance detector prototypes and advancing the technology for future cosmology experiments. This development also aimed to bolster Argonne's leadership in superconducting detector technologies. The objective deliverable was a packaged, standalone superconducting RF switch for integration into existing MKID test beds, with minimum deliverables including the switch design, simulations, and test data, along with identified pathways for performance improvement and future multiplexing scale-up.

LDRD # 2024-0276	In-Situ Transmission Electron Microscope Observation of Materials under Concurrent Straining and Irradiation
Investigators:	Wei-Ying Chen

The project aimed to develop the capability for conducting in-situ transmission electron microscope experiments with concurrent straining and irradiation at the Intermediate Voltage Electron Microscope user facility. Utilizing the recently acquired Bruker PI95 Picoindenter, a depth-sensing indenter capable of direct observation of nanomechanical testing inside a transmission electron microscope, the project performed various experiments on low melting point metals, multilayered materials, silicon carbide nanopillars, and molybdenum alloys. These experiments included direct and cyclic indentation, as well as testing before and after ion irradiation. The project successfully demonstrated the facility's ability to perform challenging in-situ experiments with high quality, providing more realistic insights into the deformation processes of materials inside nuclear reactors compared to traditional post-irradiation testing. As a result, the Intermediate Voltage Electron Microscope facility has gained extensive experience in in-situ nanomechanical testing with concurrent ion irradiation, positioning it as one of only two facilities worldwide with this capability. Additionally, the experiments on additive manufactured 316 stainless steels provided valuable insights into the impact of dislocation cell structures on deformation mechanisms under irradiation, potentially influencing future work scopes and funding opportunities.

LDRD # 2024-0278	Approaches to Molecular Beam Brightening for Tests of Fundamental Symmetries
Investigators:	David DeMille

The aim of this project was to enhance the intensity of single quantum states in a molecular beam for downstream experiments, particularly those employing Quantum Information Science protocols to explore fundamental physics questions. We focused on two methods: magnetic focusing and internal quantum state cooling using lasers, specifically applied to the ZOMBIES experiment, which investigates modifications of electroweak forces in the presence of nuclear strong forces. Through comprehensive simulations, we validated and predicted the performance of both methods, estimating an approximate eightfold improvement in signal size for magnetic focusing and a twelvefold enhancement for internal quantum state cooling. Both techniques can be implemented concurrently. This project has laid the groundwork for significant advancements in molecular beam applications in fundamental physics experiments.

LDRD # 2024-0280	Economic Model for Fusion Reactor
Investigators:	Benjamin A. Lindley Edward A. Hoffman

Our project successfully integrated a fusion reactor economic model into Argonne's open-source ACCERT tool, enhancing its capabilities for comprehensive cost analysis of fusion energy systems. This addition, based on the state-of-the-art PROCESS model from UKAEA, allows for flexible assessments and trade-off studies, addressing the unique economic challenges posed by fusion reactor design. The ACCERT model facilitates bottom-up cost estimates, considering factors such as high field magnets, thermodynamic efficiency, and material requirements, which are crucial for optimizing plant design and cost-effectiveness. Our efforts culminated in the successful commitment of this model to the ACCERT GitHub repository.

LDRD # 2024-0286	Hydrometallurgical Process for the Recovery of Market Standard Gallium and Indium from Discarded Lighting Emitting Diodes
Investigators:	Ayorinde E. Ajiboye

This project aimed to address the impending supply threat of gallium, a critical metal for the U.S. Department of Energy, by developing methods to recover gallium and indium from end-of-life light emitting diodes (LEDs). The focus was on recycling gallium nitride (GaN)-based LEDs due to their widespread use and potential impact. The project successfully identified two processing routes for gallium recovery: a two-step process involving calcination pre-treatment followed by hydrometallurgical leaching, and a pressurized gallium leaching method. The latter proved to be the most effective, achieving a leaching efficiency greater than 98%. The process involved sourcing discarded LEDs, calcinating the materials at high temperatures in an oxidative environment to convert GaN into easily leachable gallium oxide, and then subjecting it to a pressurized leaching process. The resulting products were characterized as market-standard gallium and indium salts. Additionally, the project explored the dismantling and digestion of LED components using hydrochloric and nitric acids, followed by characterization using X-ray diffraction. While copper was selectively extracted using a solvent membrane-based separator, further extraction of gallium was postponed due to safety concerns.

LDRD # 2024-0289	Critical Metals Recovery via Multistep Pyrolysis of Spent Lithium-ion Batteries
Investigators:	Bintang A. Nuraeni A.H.M. Golam Hyder, Albert L. Lipson

This project focused on developing a high-temperature recycling process that utilizes carbon dioxide to assist in the reduction of metal oxides found in the cathode of spent lithium-ion batteries. The process begins with the removal of the binder from the cathode, which generates carbon dioxide. This carbon dioxide is then used to pyrolyze the carbon-containing polymer in the battery's separator material. The resulting pyrolysis gas is employed to reduce the metal oxides in the cathode to their metallic forms, specifically cobalt and nickel. Preliminary results demonstrated that metallic cobalt and nickel could be partially recovered from the cathode oxides at 700 degrees Celsius. The project aimed to optimize the recovery yield of these metallic phases and explore the capture of lithium recovery from the gas byproduct. The approach sought to overcome the limitations of traditional pyrometallurgical battery recycling by developing a multistep pyrolysis process that enables the utilization of carbon dioxide, repurposing of spent battery separators, and full recovery of cobalt and nickel in metallic form. Additionally, the project explored the recovery of lithium as lithium hydroxide through electrodialysis, enhancing Argonne's capabilities in pyrometallurgical recycling practices.

LDRD # 2024-0291	Developing a Computational Fluid Dynamics Model of the Capacitive Deionization Process for Selective Separation of Rare Earth Elements
Investigators:	Debolina Dasgupta Lauren D. Valentino

This project aimed to develop a predictive computational fluid dynamics (CFD) model for the separation of critical materials from mineral processing feed streams using capacitive deionization (CDI). Capacitive deionization is an electrochemical method for extracting charged ions, such as rare earth elements, from aqueous solutions. The project successfully implemented the critical physics required to represent the flow of charged rare earth element ions through a CDI cell in the OpenFOAM software. Validation of the model was performed using existing experimental data, leading to a better fundamental understanding of the physics involved in electrochemical separations. The validated CFD model is now poised to accelerate the design process of CDI cells, aiding in the extraction of rare earth elements from aqueous mineral feeds. This work leveraged Argonne National Laboratory's expertise in CDI cells and computational modeling, contributing to the development of next-generation technologies for rare earth element extraction.

LDRD # 2024-0300	Enhancing the Capabilities of Power System Planning and Operation Tools by Incorporation of Hydrogen Production, Storage, and Electricity Generation
Investigators:	Carlos J. Lopez Zhi Zhou

This project focused on representing a hydrogen hub within a short-term power system optimization model. The research enhanced understanding of hydrogen production, particularly through Proton Exchange Membrane (PEM) electrolyzers, as well as hydrogen storage and electricity generation facilities in short-term optimization models for power systems. Using the proposed model, the project enabled quantitative assessment of the economic challenges associated with integrating hydrogen-based electric power generation and the potential benefits of incorporating hydrogen-responsive demand into joint electricity and gas markets. The project successfully extended Argonne's existing power system operation planning models to incorporate hydrogen as an emerging technology, enhancing the capabilities of the Argonne Low-carbon Electricity Analysis Framework.

LDRD # 2024-0307	In-situ Imaging of Thermal Runaway in Lithium-ion Batteries using X-ray Diagnostics
Investigators:	Ashwini Karmarkar Chi Young Moon

This project aimed to leverage x-ray tomography to explore structural changes in battery architecture and investigate potential causes of thermal runaway—a critical safety concern in lithium-ion batteries, especially for electric vehicles. The team developed an experimental plan and engaged with industry stakeholders to foster collaboration. Unfortunately, due to delays with the Advanced Photon Source Upgrade (APS-U), the planned x-ray measurements could not be conducted, and the project was ended early.

LDRD # 2024-0308	Integrated Bioelectrochemical Process for Carbon Waste Valorization: a Circular Economy Vased on Carboxylic Acid
Investigators:	Haoran Wu Lauren D. Valentino

This project developed a novel capability for converting organic waste streams and waste carbon dioxide into valuable platform chemicals, specifically carboxylic acids. These acids serve as crucial intermediates for synthesizing bioproducts like sustainable aviation fuel and bioplastics. The project team established a new laboratory capability for microbial electrosynthesis (MES) of carboxylic acids from waste carbon dioxide.

LDRD # 2024-0309	Addressing Deployment Challenges for In-Memory Computing: Modeling the Impact of Device-Level Imperfections and Degradation on Predictive Performance
Investigators:	Feng Qiu

This project aimed to address the critical energy consumption challenge posed by the von Neumann bottleneck in computer operations, which is projected to surpass energy production capacity by 2040. The focus was on Resistive Random Access Memory (RRAM) devices, which offer a promising solution through in-memory computing, providing highly-parallel and energy-efficient capabilities. However, performance degradation and reliability issues have hindered their widespread adoption. The team leveraged Argonne's existing expertise in degradation modeling from solar inverters, batteries, and hydropower generators to better understand and model the degradation process of RRAM devices. The project successfully developed an integrated framework that combines physical models and data-driven degradation modeling approaches to create generalizable prognostic models for RRAM devices. Dynamic transport equations of mobile defects, such as oxygen vacancies and silver ions, were solved to model degradation behavior under various operation modes. Characterization data was used to continuously update stochastic models, mimicking time-varying degradation behavior. These models were integrated through physics-informed machine learning, tested, and validated using real degradation data from RRAMs. Experiments analyzed the impact of degradation and device imperfections on machine learning tasks implemented on RRAM arrays, generating new metrics for degradation and improving understanding of failure processes on predictive task performance. This work significantly expanded Argonne's capability in asset health and degradation management, offering insights into the reliability characteristics of RRAM devices and paving the way for their broader adoption in energy-efficient computing.

LDRD # 2024-0314	Initial Forensic Examination of Chicago Pile 1 Fuel
Investigators:	Michael A. Brown John J. Arnish, Derek R. McLain, Shayan Shahbazi

The Chicago Pile 1 (CP-1) project successfully conducted a forensic examination of a graphite block containing uranium fuel from the historic CP-1 reactor, led by Enrico Fermi and Arthur Compton in 1942. This analysis, carried out by Argonne's Chemical and Fuel Cycle Technologies, Nuclear Science and Engineering, and Strategic Security Sciences divisions, revealed significant insights into the material's origin and irradiation history. Key findings include the identification of over 5 kilograms of natural uranium, likely sourced from Czechoslovakian mines and chemically purified in 1937, with fabrication at Ames Laboratory in Iowa. The uranium exhibited intense irradiation, evidenced by detectable anthropogenic radionuclides such as ^{90}Sr, ^{137}Cs, ^{236}U, and ^{239}Pu, suggesting subsequent irradiation in CP-2. The graphite was identified as T-10 grade AGOT, the purest at the time. This project provides valuable historical context tied to World War II and the Manhattan Project.

LDRD # 2024-0315	Actinide Electrochemistry and Transport Properties in Deep Eutectic Solvents
Investigators:	William Dean

Deep eutectic solvents (DESs) emerged as a promising class of tunable solvents, gaining attention for their potential in electrochemical and separation applications. These solvents are typically formed by combining a hydrogen bond donor and acceptor, creating a hydrogen bonding network that imparts unique properties such as large electrochemical windows, high thermal stability, and chemical resilience. Despite their potential, DESs have seen limited use in radiochemistry. This project aimed to explore their applicability in this field by establishing ambient-temperature electrochemical testing capabilities for small volumes (100 microliters) of radionuclides at Argonne National Laboratory. We conducted comprehensive measurements of redox potentials for select f-elements, including cerium, neodymium, erbium, and terbium, in various DESs. Cyclic voltammetry was performed at both macro and micro scales across a range of DESs with diverse functionalities, enabling the investigation of interactions between DESs and f-elements. This work assessed the potential of DESs in f-element electrochemical separations and explored their applicability within the nuclear fuel cycle. The project successfully demonstrated microliter-scale voltammetry using macro- and microelectrodes, and steady-state microelectrode measurements determined diffusion coefficients for redox-active species in DESs.

LDRD # 2024-0317	Graph Physics-Informed State-Space Neural Networks: a Digital Twin for Autonomous Operation of Nuclear Power Plants
Investigators:	Akshay J. Dave

The project successfully addressed the scalability and generalization limitations in traditional Digital Twins for nuclear power plants by developing a Graph Physics-informed State-space Model framework. This innovative approach embedded physics-based models into Graph Neural Networks, significantly enhancing the capability of Digital Twins for autonomous operation, flexibility, and real-time fault tolerance. The project involved defining the mathematical structure of the model, developing a functional framework using PyTorch, and generating a training dataset from an existing pebble-bed molten salt reactor model. A preliminary capability demonstration was conducted through the simulation of load-follow transients, resulting in a validated prototype.

LDRD # 2024-0318	Support for Prioritizing Interventions for Resilience to Extreme Temperatures (SPIRET)
Investigators:	Jordan R. Branham Michael R. Alexander

In response to risks posed by extreme heat, our project developed a comprehensive decision support tool aimed at enhancing community resilience across key U.S. metropolitan areas. By integrating high-resolution climate projections with detailed building attributes, landscape characteristics, and demographic data, we evaluated the multifaceted drivers of heat vulnerability in Chicago, Houston, New

York City, Phoenix, Seattle, and Washington D.C. This innovative approach allowed us to visualize these vulnerabilities at the Census tract and block levels through an online GIS interface, empowering users to explore and prioritize resilience interventions. Our findings underscore the importance of targeted actions, such as building weatherization, HVAC investments, and strategic land use planning, to mitigate heat-related risks. This project not only advances our understanding of urban heat vulnerability but also provides a crucial tool for policymakers and stakeholders to implement effective heat resilience strategies.

LDRD # 2024-0322	Mechanistic Understanding of Hydrogen Container Failures used in Clean Energy Applications
Investigators:	Vineeth Kumar Gattu J. Ernesto Indacochea, Sarah A. Stariha

This project aimed to investigate the effects of hydrogen exposure on high-strength steels used in hydrogen clean energy applications, focusing on understanding material degradation and failure mechanisms. The study identified Invar 36 and 304 stainless steel as potential candidate materials for hydrogen storage and transportation, based on discussions with industrial manufacturers. Flat and cylindrical coupons of these alloys were prepared to simulate cold-working conditions with varying residual stress levels. Some coupons underwent hydrogen charging in an inert atmosphere furnace at 180°C for 12 and 24 hours to simulate different levels of hydrogen-induced damage. Corrosion tests were conducted on both as-received and hydrogen-charged coupons in artificial seawater using standard electrochemical methods and protocols from the Argonne ElectroCorrosion Toolkit™. X-ray diffraction (XRD) was employed to measure changes in diffraction patterns and assess the effects of hydrogen content on the surface, revealing peak displacement with hydrogen charging duration. Scanning electron microscopy (SEM) characterized the effects of individual and combined test conditions on surface microstructures. Electrochemical corrosion test results for Invar 36 showed a correlation between cold work, hydrogen damage, and corrosion behavior, with the highest corrosion currents measured in long-term potentiostatic tests for the 24-hour charged rod sample. For 304 stainless steel, the tests indicated effects of hydrogen damage and cold work on corrosion behavior, although no significant difference in corrosion currents was observed between uncharged samples and those charged for 24 hours. This research contributes to understanding the susceptibility of steels to hydrogen attack, which is crucial for ensuring the reliability and safety of hydrogen storage and transportation applications.

LDRD # 2024-0323	Development of a Novel Microfluidic Lithography Platform to Synthesize Inorganic Microparticles for Nuclear Forensics
Investigators:	Anthony J. Krzysko Derek R. McLain, Anna Servis

This project aimed to develop a novel microfluidic lithography platform for synthesizing inorganic microparticles, specifically targeting applications in nuclear forensics. Building upon an existing microfluidic system, the team optimized a microfluidic droplet generator coupled with in-line ultraviolet polymerization for flow lithography. The focus was on demonstrating that the sizes and shapes of cerium oxide microparticles, which serve as analogs for uranium, could be precisely controlled by adjusting both physical parameters, such as flow conditions, and chemical parameters, like precursor constituent concentrations. The successful completion of this project showcased a fast, scalable, and robust method for synthesizing controlled distributions of microparticles, which holds significant promise for advancing nuclear forensic techniques.

LDRD # 2024-0338	Foundational Artificial Intelligence Model for Seasonal to Sub-Seasonal Weather/Climate Forecasting
Investigators:	Veerabhadra R. Kotamarthi Troy Arcomano, Samuel A. Foreman, Sandeep R. Madireddy

The project successfully developed HR-Stormer, a high-resolution version of the Stormer Foundation Model, designed for medium-range weather forecasting. HR-Stormer is a cutting-edge, data-driven global weather forecasting model that significantly enhances the accuracy of predictions compared to existing numerical-based and data-driven models. This innovative model employs a weather-specific transformer architecture with a randomized forecasting objective, allowing it to forecast weather dynamics over varying time intervals. This approach enables the generation of multiple forecasts for a target lead time, which are then combined to improve forecast accuracy. HR-Stormer demonstrated favorable scaling properties, as evidenced by its ability to train on the Polaris supercomputer in just 8.1 hours using 1024 graphics processing units (GPUs). For predictions, a 15-day forecast can be generated in 10 seconds using a single GPU. This efficiency facilitated the creation of a pioneering 5,120-member ensemble system for hurricane forecasting, which operates in only 10 minutes using 512 GPUs. The project highlighted the potential of artificial intelligence-based foundational climate models to address climate resilience at community scales, offering a promising solution to the limitations of traditional climate models in resolving extremes and their impacts.

LDRD # 2024-0343	Development of Magnetohydrodynamics Capability in NekRS for Fusion Blanket Applications
Investigators:	Jun Fang Thanh Q. Hua, Mi Sun Min, Dillon R. Shaver

We successfully advanced Magnetohydrodynamics (MHD) modeling capabilities within the nekRS code. This project focused on integrating existing nekRS modules related to MHD and establishing a demonstration case to simulate liquid metal flow under strong magnetic fields, typical in fusion blanket components of Magnetic Confinement Fusion systems. The successful demonstration of nekRS's ability to handle complex MHD flows marks a pivotal step in enhancing fusion energy research. By providing a high-fidelity tool for simulating fusion reactor physics, particularly the intricate fluid dynamics and MHD effects within fusion blankets hold the potential to accelerate fusion energy research, paving the way for breakthroughs in fusion.

LDRD # 2024-0349	Developing Reduced-Order Models with Machine Learning for Nuclear Technology Applications
Investigators:	Dezhi Dai Haoyu Wang, Haomin Yuan

The successful completion of this project resulted in the development of two machine learning models designed to predict pressure drop and pebble-to-helium convective heat transfer in Pebble Bed Reactors. These models were trained using data from a 1568-pebble packed bed and tested against a 500-pebble packed bed dataset, demonstrating significant improvements over conventional empirical correlations. The project aimed to revolutionize nuclear technology by integrating machine learning-enhanced Reduced-Order Models with traditional simulations to improve the accuracy and reliability of safety analyses in Pebble Bed Reactors. The versatile framework developed is poised for wide adoption across various nuclear sectors, including Light Water Reactors, Fast Breeders, and Molten Salt Reactors, promising to enhance safety and efficiency standards.

LDRD # 2024-0351	Modeling and Evaluation of Direct Air Capture (DAC) with Various Carbon Dioxide (CO2) Capture Sorbent Materials
Investigators:	Pingping Sun Amgad A. Elgowainy, Lili Sun

This project focused on advancing the understanding and development of moisture-driven sorbent materials for direct air capture (DAC) of carbon dioxide. Through comprehensive Techno-Economic Analysis (TEA) and Life-Cycle Assessment (LCA), we evaluated various sorbent materials, examining key parameters such as capacity, lifetime, and cost. Our analysis revealed insights into the energy consumption and impacts associated with these materials, highlighting opportunities for high-performance and cost-effective solutions. The integration of TEA and LCA findings identified potential pathways to optimize sorbent materials.

LDRD # 2024-0354	Ultramicroelectrode Sensors for Molten Salt Applications
Investigators:	Timothy T. Lichtenstein

This project focused on the development of a novel ultramicroelectrode using liquid metals to detect and quantify dissolved species, such as corrosion and fission products, in molten salts. The aim was to enhance the detection limits beyond those achievable with conventional macroelectrodes, which is crucial for the deployment of next-generation green technologies like molten salt reactors. The project successfully demonstrated a proof-of-concept ultramicroelectrode capable of detecting surrogate fission products at concentrations below the detection limit of traditional electrochemical sensors. The ultramicroelectrode was constructed using boron nitride as an inert housing for the liquid metal electrode, with gallium and bismuth as target liquid metals. The electrode's geometry was verified using scanning electron microscopy, and its operation was validated through cyclic voltammetry in well-characterized molten salt systems, such as LiCl-KCl-$LaCl_3$. The project also demonstrated the ability to discriminate between electroactive species with overlapping electrochemical signals, such as $LaCl_3$ and

$GdCl_3$, using the ultramicroelectrode. Additionally, the project explored the use of a piezoelectric step motor for controlled vertical translation of the ultramicroelectrode, enabling the investigation of localized electrochemical phenomena at elevated temperatures. These advancements hold promise for improving the understanding of reaction kinetics in electrochemical processes like corrosion and electrodeposition, as well as benefiting existing green technologies by providing insights into localized parasitic reactions and electrochemical kinetics. The successful outcomes of this project pave the way for further development of this promising technology to quantify a variety of soluble products below the detection limit of conventional electrochemical sensors.

LDRD # 2024-0356	Mapping the Spinning Gluons in the Nucleon
Investigators:	Yong Zhao Rui Zhang

In this project, we conducted theoretical and numerical studies on lattice quantum chromodynamics to calculate the gluon helicity parton distributions within the nucleon. Our efforts led to significant advancements in two key areas. First, we successfully calculated the one-loop perturbative matching coefficient for specific operators that define the gluon transverse-momentum-dependent parton distribution function. This achievement is crucial for enabling lattice quantum chromodynamics calculations of the gluon helicity transverse-momentum-dependent distribution, thereby contributing to a comprehensive three-dimensional image of the nucleon. Second, we discovered a novel interpolation operator for boosted hadrons, which enhances the signal-to-noise ratio in lattice simulations by an order of magnitude. This discovery is pivotal, as the gluon parton distribution functions and transverse-momentum-dependent matrix elements are derived by inserting gluonic operators into correlations of these interpolators, averaged over configurations. Our findings promise to significantly improve statistical precision and reduce computational costs, marking a breakthrough for challenging lattice computations. These accomplishments lay the groundwork for further simulations of gluonic operators with the new interpolators, aiming to identify the optimal choice for gluon helicity distributions.

LDRD # 2024-0357	Bias Correction of Dynamically Downscaled Climate Models
Investigators:	Veerabhadra R. Kotamarthi Thomas A. Wall

The project successfully developed a new methodology for bias correction in regional-scale climate model projections, addressing inherent errors in the models' physics and systematic biases transmitted through model boundaries. This innovative approach utilized Empirical Model Decomposition (EMD), an empirical, nonlinear analysis tool for complex, non-stationary time series. EMD adaptively and locally decomposes any non-stationary time series into a sum of Intrinsic Mode Functions (IMFs), which represent zero-mean amplitude and frequency-modulated components. This fully data-driven and unsupervised signal decomposition method does not require any predefined basis system and satisfies the perfect reconstruction property, allowing for the reconstruction of the original signal without information loss or distortion. While EMD offers greater flexibility and generalizability compared to traditional Fourier or wavelet decompositions, interpreting IMFs remains challenging, often relying on binary decisions for partial reconstruction. The Argonne Dynamically Downscaled Data Archive (ADDA) has become a valuable resource for stakeholders, including public and private sector utilities and communities, providing physically consistent multivariable outputs of present and future climate states. The project aimed to develop a multivariate bias correction methodology for select variables in ADDA, preserving the shape, variability, and time relationships over the entire historical sequence for three climate models.

LDRD # 2024-0374	One Gigahertz Niobium-tin Quarter Wave Cavity for Future Ion Accelerators
Investigators:	Yang Zhou Berardino M. Guilfoyle, Troy B. Petersen

This project demonstrated the feasibility of a high-frequency, 1 gigahertz, compact triniobium tin-coated quarter-wave cavity for future ion accelerators. Triniobium tin cavities offer 1-2 orders of magnitude lower resistance compared to standard niobium, such as that used in the ATLAS superconducting linear accelerator. This allows for efficient operation at much higher frequencies, presenting opportunities for transformative size reductions in accelerating cavities. We designed, built, and tested a 1 gigahertz quarter-wave cavity, approximately the size of a soda can, using niobium left over from previous projects. Electromagnetic simulations were conducted to optimize its performance. The triniobium tin coating was applied through vapor diffusion, and two cryogenic cold tests were performed. The measured superconducting transition temperature was 17.7 Kelvin ± 0.1 Kelvin, nearly twice that of niobium and close to the expected value for triniobium tin. Notably, we observed a quality factor exceeding 10^9 at low accelerating electric fields, surpassing the theoretical limit for pure niobium at 1 gigahertz. This performance is attributed to the significantly lower losses achievable with triniobium tin. The development of this high-frequency compact cavity has the potential to transform low-beta ion accelerators by enabling dramatic size reductions and replacing large helium cryoplants with distributed small plug-in cryocoolers, thereby lowering overall costs and eliminating major single-point failures in modern superconducting linear accelerators.

LDRD # 2024-0383	Scalable Quantum Computing Error Characterization Method
Investigators:	Alvin Gonzales

In this project, I addressed the challenges associated with error-prone quantum computers by developing a novel correction method that does not rely on quantum error correcting codes. The approach focused on correcting the output distribution of quantum computers under a biased noise model, which can be achieved using a technique known as twirling. I analytically demonstrated that the ideal output distribution and the noisy distribution of a quantum circuit are connected through a stochastic error matrix. By characterizing and inverting this matrix on the noisy distribution, I was able to recover the ideal output distribution. This method proved effective in simulations and is set to be tested in hardware experiments. Although the approach requires additional executions on the quantum computer, it offers a scalable and accurate characterization of errors specific to the executed quantum circuit, potentially enhancing quantum computing accuracy.

LDRD # 2024-0387	Intelligent Lossy Compression of Deep Underground Neutrino Experiment Data
Investigators:	Amit Bashyal

In an effort to enhance data management for the Deep Underground Neutrino Experiment (DUNE), our project successfully developed a comprehensive framework for testing intelligent lossy compression algorithms on both synthetic waveforms and actual Proto-DUNE raw data. The results demonstrated that algorithms like SZ3 can effectively compress and decompress experimental data while maintaining sufficient fidelity, paving the way for efficient data storage and transmission. The framework supports the persistence of compressed raw data in ROOT, with flexibility to incorporate other algorithms such as MGARD and IDEALEM. Extensive testing explored various compression parameters and synthetic data variations. This work positions Argonne for future DUNE research, facilitating the integration of these advancements into production workflows and reinforcing our leadership in neutrino physics data management.

LDRD # 2024-0388	Machine Learning-Driven Insights for Monitoring Community Climate Resilience
Investigators:	Michael R. Alexander

This project successfully leveraged machine learning techniques to explore the effects of drought and weather variability on urban vegetation health. By applying three distinct machine learning methods, researchers identified minimum daily temperatures and the daily standardized precipitation index as the most influential factors affecting urban greenness. Although the project faced challenges in developing a predictive model due to data format issues and lack of model convergence, it yielded significant insights and established a standardized approach for analyzing satellite data. The project's outcomes underscore the potential of machine learning to integrate diverse data streams, ultimately aiding planners in quantifying the cooling effects of urban tree canopies and informing robust plans that incorporate both nature-based and technological solutions.

LDRD # 2024-0390	Development Toward Full Quantum Molecular Cascade Protocol
Investigators:	Yeonsig Nam

In this project, we developed a computational framework to model the absorption of X-rays by carbon monoxide (CO) molecules, followed by electron loss through photoionization and further electron emission via Auger decay. This work laid the foundation for accurately predicting molecular behavior under high-energy radiation, which is crucial for applications in astrophysics, plasma physics, and radiation chemistry. Utilizing advanced quantum chemistry and nuclear dynamics methods, we successfully simulated key molecular breakup pathways, enhancing the accuracy and efficiency of future computational models. Our efforts addressed the challenge of predicting radiation damage in molecules, a significant issue in large facilities like the Advanced Photon Source. The project's success has motivated the development of workflows to leverage the computational power of the Argonne Leadership Computing Facility (ALCF) in future research.

LDRD # 2024-0392	Dual-Area Selective Deposition (Dual-ASD) for 3-Dimensional Patterning
Investigators:	Chi Thang Nguyen Angel Yanguas-Gil

The primary aim of this project was to demonstrate the feasibility of the dual-area selective deposition concept for three-dimensional patterning applications in semiconductor fabrication. The project investigated ruthenium atomic layer deposition using a novel ruthenium precursor, tricarbonyl (η4-2-methylene-1,3-propanediyl)Ruthenium(II) (TRuST), and oxygen as the reactant. Additionally, bis(N,N-dimethylamino)dimethylsilane (DMADMS) was employed as an inhibitor in the ruthenium area selective deposition process. This inhibitor selectively adsorbs on silicon dioxide surfaces, preventing unwanted reactions with the ruthenium precursor. By using ozone as a reactant, the inhibitor was converted into a silicon dioxide atomic layer deposition thin film, avoiding contamination. The project successfully demonstrated the dual-area selective deposition concept through the selective and simultaneous growth of ruthenium and silicon dioxide films. This achievement paves the way for future three-dimensional patterning applications, offering a promising solution to the challenges faced by conventional patterning techniques in microelectronics due to shrinking device sizes and alignment issues.

LDRD # 2024-0393	Elemental and Chemical Species Quantification of Rare-Earth Feedstock and Recovered Magnets Using X-ray Emission Spectroscopy
Investigators:	Khagesh Kumar Eva Allen, Mikhail A. Solovyev

The project aimed to utilize the combination of X-ray emission and absorption spectroscopy to quantify the chemical speciation and composition of neodymium and iron in the precursor and extracted product of end-of-life neodymium iron boron magnets. We conducted X-ray absorption spectroscopy measurements at the National Synchrotron Light Source II, where the proximity of energy between neodymium and iron absorption edges enabled single-scan data collection. This approach allowed us to estimate the oxidation states of neodymium and iron in both pristine and extracted materials. Additionally, the relative intensity of neodymium and iron absorption lines provided insights into the efficiency of the extraction process. The project addressed the limitations of traditional lab-based characterization tools, which are slow and often require downstream refining due to feature overlap among rare-earth elements. By employing synchrotron-based X-ray emission spectroscopy, we focused on specific emission features using analyzer crystals in Bragg geometry, making it well-suited for probing rare-earth elements. This method enabled the investigation of multiple elements in a single experiment, facilitating the quantification of composition and chemical speciation. The project successfully procured standard compounds and fabricated analyzer crystals for neodymium and iron, modifying an existing spectrometer to accommodate these new crystals. The quantification was achieved by comparing data against calibration curves from known samples, while chemical speciation was determined using reference compounds to identify key spectral features. A Python code was developed to conduct the analysis, and the results were anticipated to be within error from expected values. The findings laid out design principles for real-time analysis of the recovery process and were disseminated through a manuscript and report, contributing to renewed focus and further development in the field of rare-earth element recovery.

LDRD # 2024-0395	Ultrafast Joule Heating Synthesis of Cathode Materials for High Performance Lithium-Ion Batteries
Investigators:	Boyu Shi Tianyi Li

The project successfully utilized Joule heating as an ultrafast and energy-efficient technique for synthesizing cathode materials for lithium-ion batteries. Joule heating, known for its ultra-high heating and cooling rates, significantly reduced processing times and energy consumption compared to conventional synthesis methods. Despite challenges such as temperature fluctuations and limited synthesis durations, the project developed an advanced Joule heating system capable of achieving a wide temperature range (240°C–3600°C) with ramping rates of up to ~1000°C/s. This system enabled precise temperature control up to 2800°C for sustained periods of up to 4 minutes and could operate under various gas atmospheres. The versatility of this Joule heating system opened new avenues for synthesizing cathode materials. The project demonstrated the synthesis of cation-disordered rock-salt (DRX) cathode materials and conventional cathode materials, such as $LiCoO_2$, NMC, and $LiMn_2O_4$, verifying the method's versatility. The synthesized materials exhibited enhanced energy density, cycling stability, and rate capability. The project also explored the scalability of the Joule heating synthesis process, aiming to synthesize large quantities of cathode materials for commercial applications. This research has the potential to revolutionize the production of lithium-ion battery cathode materials, making them more cost-effective and environmentally friendly, and could lead to new commercial opportunities and collaborations with industry partners.

LDRD # 2024-0398	Electrochemical Ion Separation Using Birnessite Nanosheets for Manufacturing and Water Treatment
Investigators:	Evelyna Tsi-Hsin Wang Baris Key

In this project, we successfully exfoliated birnessite nanosheets from bulk lithium manganese dioxide cathode materials, aiming to utilize these two-dimensional materials for aqueous ion adsorption. The primary focus was on testing exfoliation methods and preparing electrodes. Characterization confirmed the successful exfoliation into nanosheets; however, challenges with binder compatibility led to poor electrode performance and suboptimal electrochemical results. Although the desired electrochemical ion adsorption performance was not achieved, the project underscored the necessity for further chemical processing development to utilize these materials effectively. This work has the potential to contribute to the development of new electrochemical separation materials, with applications in desalination, wastewater treatment, and lithium-ion sieving.

LDRD # 2024-0401	Sodium-Ion Batteries for Space Exploration
Investigators:	Marco F. Rodrigues Nicolas Folastre

Economic exploration of celestial bodies is anticipated to be a significant aspect of humanity's future, necessitating reliable energy storage solutions for long-term missions on planets, moons, and asteroids. This project aimed to evaluate the feasibility of sodium-ion batteries as an alternative to lithium-ion batteries for space applications, given sodium's abundance across the Solar system. We investigated whether sodium-ion cells could withstand the high radiation environments encountered in space. State-of-the-art sodium-ion batteries were subjected to gamma radiation at doses of 5, 10, and 20 Mrad and analyzed using various characterization tools. Our findings revealed negligible damage to the polyethylene separator and hard carbon anodes, with no clear evidence of damage to the layered oxide cathode. However, radiation-induced decomposition of the liquid electrolyte led to cell dilation and increased resistance due to gassing. These results suggest that developing radiation-resistant electrolyte components could eliminate the need for shielding sodium-ion batteries during space deployment, thereby reducing volume and weight requirements for battery packs. Despite these promising insights, current sodium-ion battery technology still necessitates shielding in missions with high radiation exposure.

LDRD # 2024-0405	Adaptive Continuous Scans for Scanning X-ray Microscopy
Investigators:	Ming Du

This project focused on enhancing scanning x-ray microscopy by developing an adaptive fly-scan algorithm that combines continuous data acquisition with smart adaptive sampling. Traditional step-scan methods, which acquire images by scanning one location per pixel, are limited by inactive time required for probe movement. The recent upgrade to the Advanced Photon Source aimed to reduce individual scan times, but probe movement remained a bottleneck. Our innovative approach integrated the continuous scanning of fly-scans with adaptive sampling to optimize scan paths, thereby reducing scan times significantly and improving scientific throughput.

The adaptive fly-scan algorithm we developed successfully suggested optimal scan paths that maximize measurement in regions with uncertainty or significant local variations. Compared to random sampling, our method improved the peak signal-to-noise ratio of acquired images by at least 30%, while requiring only 20% of the samples or experiment time of traditional dense scanning. We demonstrated the effectiveness of our method on four images with diverse visual characteristics, consistently achieving superior results.

Additionally, the project facilitated the recruitment of an intern, whose contributions were instrumental in the development of the algorithm. His involvement has sparked a keen interest in advancing this domain further, potentially enhancing beamline operations at the Advanced Photon Source. This project not only advanced the field of x-ray microscopy but also nurtured the career of a promising young scientist.

Overall, the project laid the groundwork for future research by implementing fly-scan pathing in software and evaluating adaptive path creation and updates. The optimization of probe paths involved minimizing x-ray exposure and uncertainty in reconstructed samples, using constraints such as probe path, x-ray exposure limits, and probe maneuverability. While fast optimization methods were explored, the foundational work in simulation and optimization remains valuable for ongoing and future advancements in x-ray microscopy.

LDRD # 2024-0413	Novel Room Temperature Electrowinning of Sodium
Investigators:	Michael J. Dziekan

In this project we successfully adapted our innovative chlorine-free, room-temperature lithium electrowinning technology to produce sodium metal, marking a significant advancement in the alkali electrowinning portfolio. Traditionally, sodium metal production involves electrowinning molten sodium chloride, which is energy-intensive and generates chlorine gas. By contrast, Argonne's new process operates at room temperature and produces oxygen gas instead of chlorine, offering a more energy-efficient alternative. The proof of concept for sodium metal electro-refining and electrowinning was achieved, demonstrating the potential for this method to be used in metallothermic reduction of metals such as titanium and rare earth elements.

LDRD # 2024-0415	Crystal Graph-based Generative Model for X-ray Crystallography
Investigators:	Xiangyu Yin

This project focused on developing a novel method for X-ray diffraction data analysis using crystal graph-based generative models. Retrieving crystal structures from X-ray diffraction data has been a long-standing challenge in materials science, traditionally requiring a multi-step "fitting" approach and significant expert effort. Our approach aimed to revolutionize this process by employing unsupervised machine learning techniques to directly predict crystal structures from X-ray diffraction data. This method diverged from the conventional supervised learning paradigm, which relies on large amounts of labeled training data and often lacks generalizability. Instead, we utilized generative models to create a "generation" approach, offering a more streamlined and potentially more accurate analysis. Our strategy involved three key steps: preparing an advanced crystal graph generative model, developing structure retrieval algorithms, and implementing these methodologies into a software toolkit compatible with data formats and analysis frameworks used at the Advanced Photon Source (APS). The successful application of these methodologies across multiple Advanced Photon Source beamlines is expected to have significant scientific and practical impacts in the fields of materials science, microscopy, and optimization.

LDRD # 2024-0422	Coupling Argonne's DRE4M and A-LEAF Models for Grid and Industry Cross-Sectoral Decarbonization Analysis
Investigators:	David P. Thierry

We explored the integration of two Argonne-developed models—A-LEAF for grid generation and transmission, and DRE4M for industrial manufacturing—to better understand the interdependence between the industrial and power sectors. Our efforts successfully established a methodology for parametrizing DRE4M with data from A-LEAF, considering the different time resolutions of each model. While DRE4M generates results using grid information from A-LEAF's planning, the integration remains incomplete, as it does not fully account for increased electrical demand at each node from A-LEAF. Despite this, the project laid the groundwork for future simplifications of DRE4M to enable less intrusive linking with A-LEAF, potentially achieving feedback from increased electricity demand. This research represents a significant step toward creating a powerful next-generation analysis capability, with the potential to elucidate the interplay between the industry and energy supply sectors.

LDRD # 2024-0424	All-Sky Cloud Classification for Cosmic Microwave Background Observations
Investigators:	Kathleen Harrington

Observations of the cosmic microwave background (CMB) at millimeter wavelengths provide crucial insights into the fundamental nature of the universe. However, ground-based CMB observations are often disrupted by atmospheric conditions, particularly clouds, which introduce linearly polarized anomalies into the data. This project successfully developed an all-sky cloud monitoring system using a consumer-level digital single-lens reflex (DSLR) camera equipped with a fish-eye lens. The camera system was installed on the roof of a building at Argonne National Laboratory, where it collected test data to refine image quality and develop cloud detection algorithms. The image data generated was used to evaluate machine learning algorithms for cloud detection and classification, aiming to enhance data cleaning processes for CMB observations. In collaboration with the Simons Observatory, plans were made to deploy the camera system at Cerro Toco, the future Chilean site for the CMB-S4 project. The data collected from this site is expected to provide essential information for optimizing observation efficiency and understanding the impact of cloud signals on data from small aperture telescopes. This project has the potential to significantly improve the observation efficiency and simplify data cleaning for ground-based CMB observatories.

LDRD # 2024-0441	Towards Fast Quantitative Regression Analysis with Vision Transformers and Bayesian Neural Networks
Investigators:	Weijian Zheng

This project investigated the integration of Vision Transformers and Bayesian Neural Networks to enhance regression analysis for predicting diffraction peak locations in High Energy Diffraction Microscopy data. By combining Vision Transformers' advanced image segmentation and self-attention capabilities with Bayesian Neural Networks' uncertainty quantification, the project aimed to achieve fast, accurate peak predictions accompanied by confidence scores—an essential feature for applications where prediction reliability is paramount. This novel approach addressed limitations in traditional Convolutional Neural Network-based methods, such as BraggNN, which lacked confidence scoring. The project began by comparing the convergence rates of a simple Vision Transformer model and the traditional Convolutional Neural Network-based BraggNN model on different graphics processing units, finding that the Vision Transformer model converged faster. Next, Bayesian Neural Network layers were introduced to enable confidence scoring in predictions. By replacing specific layers in both the Vision Transformer and Convolutional Neural Network-based models, the enhanced model accurately predicted peak locations with reliable confidence scores.

LDRD # 2024-0444	Improving the Electrochemical Stability and Efficiency of Sodium-ion Battery Electrolytes via Mixed-Anion Ion Pair Formation
Investigators:	Stefan Ilic

Despite growing interest and progress in sodium-ion batteries, electrolyte development is still in its early stages. In this project, we advanced the development of next-generation electrolytes for sodium-ion batteries by leveraging a mixed contact-ion pair strategy, previously successful in magnesium and zinc systems. Recognizing the unique solvation structure of sodium compared to lithium, we conducted density functional theory calculations to screen six commercially available sodium salts, identifying anions that form either weakly or strongly coordinating pairs with sodium ions in propylene carbonate solvent. Our findings revealed that strongly coordinating anions, such as triflate and trifluoroacetate, significantly lower stripping potentials, correlating with their higher sodium binding tendencies. This insight into mixed-anion contact-ion pair formation establishes new design principles for stable sodium battery electrolytes. Our work not only enhances understanding of electrolyte behavior but also sets the stage for future implementation in battery cell testing.

LDRD # 2024-0449	Data-Driven Reduced-Order Digital Twin Framework Development for Smart Control of Dynamical Systems
Investigators:	Chandrachur Bhattacharya

This project focused on developing a versatile data-driven reduced-order digital twin framework aimed at enhancing smart control of dynamical systems within the Advanced Energy and Transportation portfolio, including gas turbines, vehicles, and heat exchangers. Utilizing deep learning techniques, specifically deep state-space modeling with recurrent neural networks, the framework was designed to model complex transient behaviors by focusing on key input and output variables, thus eliminating the need for full-system simulations. This reduced-order approach was primarily intended to facilitate real-time control and optimization, which are crucial for decarbonization efforts and improving operational efficiency. Validation of the framework on a simple dynamical system, such as the spring mass damper system, demonstrated its applicability and potential for integration into existing AET tools like Autonomie and X-in-the-loop. Although the incorporation of Bayesian methods for uncertainty quantification is planned for future work, the current results confirmed the framework's effectiveness and applicability to real-world systems.

LDRD # 2024-0458	Empowering Artificial Intelligence at the Edge with Federated Learning
Investigators:	Charikleia Iakovidou Seongha Park

Artificial intelligence-driven distributed sensor networks are available within the development of edge computing platforms, including those supporting GPUs. Federated learning is a recent machine learning paradigm which permits generating collaborative, general machine learning models from private and decentralized data, and could serve as an alternative to processing large-scale and sensitive data at the edge and transmitting only the processed results to the cloud for the creation of artificial intelligence models. Modern advanced sensor networks, however, may consist of hundreds of nodes varying widely with respect to computation, communication and memory capacities, power usage, and collected data modalities, prohibiting the straightforward application of existing federated learning algorithms. To tackle this issue, we developed a custom federated learning algorithm with a flexible structure, which allows for adjusting the amounts of computation and communication performed at each node by appropriate selection of algorithm parameters. Moreover, the algorithm is equipped with a probabilistic error correction mechanism, that alleviates the distributional bias caused by non-iid local data distributions and can be adapted to the local resources available to each node. We implemented this algorithm on the Advanced Privacy-Preserving Federated Learning (APPFL) software platform, to leverage its existing differential privacy infrastructure. Finally, we streamlined the packaging and deployment of APPFL to an arbitrary number of sensors on the Waggle edge computing platform, and verified the APPFL-Waggle integration by successfully training a small convolutional neural network for an image classification problem with decentralized data stored on 18 distinct Waggle sensors.

LDRD # 2024-0465	Development of Signatures
Investigators:	John J. Arnish

We were successful in our Development of Signatures project. We identified 16 unique signatures associated with the project.

LDRD # 2024-0466	Enhancing High-Precision Inductively Coupled Plasma-Quadrupole Mass Spectrometry (HP-ICP-QMS) Method Capability
Investigators:	Nicholas J. Condon Jennifer L. Steeb

Our team successfully developed a unique, high-precision inductively coupled plasma-quadrupole mass spectrometry (ICP-QMS) method that enables relatively inexpensive and easy-to-operate instruments to provide isotope ratio measurements with precision comparable to more expensive and complex techniques. This project focused on further advancing this method by creating and testing a new, user-friendly software package in Python. The existing Excel Visual Basic for Applications (VBA) code for the correction method was translated into Python, with the assistance of large language model-based artificial intelligence tools. The resulting Python code was thoroughly validated against real data to ensure its functionality and accuracy.

The project has resulted in a mature method for measuring isotope ratios with high precision using commonly available ICP-QMS instruments. This capability significantly impacts the range of work that can be conducted at Argonne National Laboratory, providing a local source of high-precision data previously accessible only at facilities with dedicated high-precision mass spectrometry equipment and staff. It opens new opportunities for projects where isotope ratios are crucial, such as radioisotope production, nuclear safeguards, and nuclear and chemical forensics.

LDRD # 2024-0474	Liquid Metal Converter Simulation
Investigators:	Sergey D. Chemerisov Roman Gromov, Andrei S. Patapenka

The project focused on optimizing the design of a liquid metal converter for high-power applications, specifically targeting the production of medical isotopes and neutron generation. Using the Monte Carlo particle tracing code FLUCA, we analyzed 432 simulation cases for two target materials: a lead-bismuth (PbBi) stream encapsulated in a pipe and a PbBi jet. The optimization process considered various beam sizes, ranging from 1 mm to 6 mm full width at half maximum (FWHM), different target sizes, and converter thicknesses. We determined that the optimal converter thicknesses, aimed at maximizing specific isotope production, varied between 1.4 millimeters and 3.6 millimeters, depending on the target thickness. Further optimization of the converter thickness was conducted for subsequent radiation energy deposition calculations and computational fluid dynamics studies. These studies demonstrated the superior performance of the liquid metal converter compared to conventional solid water-cooled converters. Computational fluid dynamics simulations revealed a localized temperature rise of 400 degrees Celsius at the center of the beam impact. With realistic flow rates, the heated volume of the converter was completely replaced by cold liquid metal, ensuring continuous operation at the accelerator's maximum repetition rate. This project has highlighted the advantages of using liquid metal as a coolant or heat transfer medium, offering a wide temperature range and high thermal conductivity, making it attractive for cooling applications in compact nuclear reactors and high-power accelerator applications.

LDRD # 2024-0476	Direct Electrochemical Transformation of Biomass into Valuable Dicarboxylic Acids
Investigators:	Donghyeon Kang Magali S. Ferrandon, Pingping Sun

The objective of this project was to develop an innovative electrochemical method for depolymerizing lignin and to establish robust protocols for product characterization, with a focus on producing valuable bio-acids, particularly oxalic acid. We successfully demonstrated a novel electrochemical lignin depolymerization process using nickel-form electrodes under mild conditions. In a 1M sodium hydroxide solution, the process yielded 12% oxalic acid after 7 hours of electrolysis. Additionally, we established effective methods for lignin separation and product characterization, utilizing acidification, precipitation, high-performance liquid chromatography, and nuclear magnetic resonance spectroscopy. This project highlights a cost-effective, energy-efficient approach to lignin depolymerization, advancing the sustainable production of bio-acids. Furthermore, we developed a techno-economic analysis model to evaluate the economic impacts of the electrochemical processes.

LDRD # 2024-0482	Bio-Based Material Strategies for the Circular Blue Economy
Investigators:	Edward F. Barry Pahola Thathiana Benavides Gallego, Lily A. Robertson, Casey Stone, Abraham Stroka, Rebecca B. Weinberg

This project successfully developed innovative mycelia-based materials cultivated from organic wastewater, demonstrating their potential as effective membranes and adsorbents for selectively removing minerals, nutrients, and PFAS from low-concentration aqueous sources. Through rigorous testing, we quantified key performance metrics related to both synthesis pathways and adsorption capacities, providing valuable data for techno-economic assessments. This project was pivotal in establishing new synthetic capabilities for these materials.

LDRD # 2024-0483	Measurement for Prediction
Investigators:	Jonathan Ozik Arindam Fadikar, Abby Stevens

This project successfully demonstrated a proof-of-concept for developing advanced predictive artificial intelligence capabilities that enhance "measurement for prediction" (MfP) through the integration of artificial intelligence models and detailed simulations. We utilized the CityCOVID agent-based model to generate training data, enabling us to explore the relationship between wastewater viral load measurements and hospital utilization across Chicago. By employing a Recurrent Graph Neural Network, our artificial intelligence model effectively learned the impact of population mobility on health outcomes, showcasing the potential of granular, real-time predictive models. This work lays the foundation for creating "always on" smart sensor networks that can support public health and emergency management by optimizing the types of empirical data needed for high-quality forecasts.

LDRD # 2024-0486	Using Large-Language Models in the Preparation of National Environmental Policy Act Documentation
Investigators:	Ellen Moret White Troy Arcomano, Nicholas Lee-Ping Chia, Heidi M. Hartmann, Kurt C. Picel, Arvind Ramanathan, Leroy J. Walston, Konstance L. Wescott

In this project, we investigated the potential of large-language models to enhance the efficiency of preparing National Environmental Policy Act (NEPA) documentation, which involves comprehensive reviews of environmental, social, and economic impacts from proposed federal actions. Our analysis revealed that large-language models could significantly reduce the time required for tasks such as collating sources and baseline data, summarizing public comments, developing reference lists, and managing documents. Although the initial output from these models requires further refinement by users, it generally saves considerable time. However, the complexity of tasks influences the degree of human involvement needed, with more complex tasks necessitating increased editing and oversight. The implementation of these models may be limited by the programming skills required to manipulate scripts, create digital repositories, or develop task-specific tools, which could be a barrier for staff lacking these skills. Key variables affecting the results include data access, the type of large-language model used, and file size. Future research could focus on developing tools to manage tasks behind the scenes to avoid token limits and standardize tasks with varying variables.

LDRD # 2024-0487	Mobile Robotics for Medical Isotope Production and Processing
Investigators:	Ravishanker V. Gampa Jerry A. Nolen, Jr., Doga Y. Ozgulbas

During this project, we made significant strides in the development of dual-arm mobile robots capable of operating in high radiation areas, which could revolutionize medical isotope production. We successfully demonstrated key technologies that bring these robots closer to achieving the ambitious goal of automated radioactive material retrieval. Our achievements included reliable mobile robot movement, precision docking, and the ability to disconnect coolant tube connections to the target block using coordinated movements of two robotic arms. These advancements suggest the potential for a novel robot that can enter irradiated areas much earlier than human agents, thereby reducing process time, increasing the yield of medical isotopes, and substantially reducing radiation exposure for technical staff. Our work involved integrating sensor data, reducing system latency, and utilizing augmented reality-based software to enable simultaneous yet independent cooperative movement of both robotic arms. This project has laid the groundwork for the automation of medical isotope processing, marking a significant step forward in the field.

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