ANL-20/43

2018 LDRD Annual Report

Laboratory Directed Research and Development Program

Laboratory Directed Research and Development

About Argonne National Laboratory

Argonne is a U.S. Department of Energy laboratory managed by UChicago Argonne, LLC under contract DE-AC02-06CH11357. The Laboratory's main facility is outside Chicago, at 9700 South Cass Avenue, Argonne, Illinois 60439. For information about Argonne and its pioneering science and technology programs, see www.anl.gov.

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2018 LDRD Annual Report

Laboratory Directed Research and Development Program

prepared by
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Laboratory Directed Research and Development , Argonne National Laboratory

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LDRD Program Overview

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

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

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

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

FY 2018 LDRD Program Components

LDRD Prime

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

Focus Areas with projects that ended in FY18 are:

Advanced Computing
Biological and Environmental Science Capability Development
Energy Manufacturing Science and Engineering
Hard X-ray Sciences
Materials and Chemistry
Securing Energy and Critical Resources
The Universe as Our Laboratory (ULab)

Director's Collaborations

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

Named Fellows

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

Innovate

The Innovate component of the LDRD program invests in a full spectrum of investigator-initiated proposals across the Laboratory in DOE-mission-related science and engineering areas. This provides an avenue for R&D staff to propose highly innovative projects in research areas outside the purview of the Prime Focus Areas.

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 2018 LDRD Summary Report

This summary report provides an overview of all LDRD projects at Argonne that concluded in Fiscal Year 2018. 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.

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ADVANCED COMPUTING

LDRD# 2017-034	Advanced Computing
Project Title:	The Perfect Thermodynamics of Imperfect Materials
Investigators:	Marius Stan Alejandro Lopez Bezanilla, Alvaro Vazquez Mayagoitia, Chris Benmore, John Low, Laura Ratcliff

Abstract

The goal of this project is to significantly improve our understanding and the prediction of thermodynamic stability/metastability of "imperfect" (e.g., highly defective, non-stoichiometric, or doped) oxide material phases via innovative theory, advanced experiments at the Advanced Photon Source (APS), and "intelligent" software (i.e., machine learning). Hafnium dioxide (HfO_2) has a high dielectric constant and is used in advanced metal-oxide-semiconductor (MOS) devices. Accurate free energies have been predicted from ab initio lattice dynamics up to 1300 K for monoclinic m-HfO_2. Using that approach, we predicted the heat capacity of HfO_2 and compared it to the experimental heat capacity. We applied a recently developed interatomic potential for hafnium oxide with which we reproduced the lattice parameters and energies of several crystal structures. All data resulting from theory, experiments and calculations were evaluated using Bayesian Analysis to determine the optimal free energy models of the non-stoichiometry oxide phases as function of temperature and oxygen-carrying defect concentration. Uncertainty evaluation is embedded in this multi-scale methodology via the Bayesian analysis. Our approach is based on calculating the free energy of each phase versus temperature and composition (oxygen and dopant content) by using atomistic, mesoscale, and continuum methods. We validated the models and computer simulations using high-temperature high-energy x-ray diffraction experiments on laser-heated and levitated samples at the Advanced Photon Source (APS) as well as performing neutron diffraction experiments on the amorphous form at the NOMAD instrument of the Spallation Neutron Source (SNS). The results on the main HfO_2-x phases demonstrated the viability of our approach.

LDRD# 2017-049	Advanced Computing
Project Title:	SLIK-D: Scalable Machine Learning Infrastructure for Knowledge Discovery
Investigators:	Venkatram Vishwanath Fangfang Xia, Kalyanasundaram Kumaran, Kevin Harms, Prasanna Balaprakash, Stefan Wild, Taylor Childers, Thomas Brettin, Thomas Uram, Vitali Morozov

Abstract

Machine learning (ML) and deep learning (DL) are having impact on applications ranging from self-driving cars to natural language processing. A key challenge in applying ML and DL is their adaptation to scientific datasets, which are diverse with respect to their data models and structures, size, complexity, and other parameters, and are increasing in complexity as datasets from several scientific domains are combined. Despite initial successes in executing on a set of a few nodes, effective ML and DL software that fully exploits the architectural features of supercomputers is lacking. We are developing scalable

ML and DL infrastructure to exploit data-driven and computational models in science and the architectures of supercomputers. We are collaborating with application scientists in genomics and high-energy physics (HEP). To achieve our goal, we are developing the Scalable Machine Learning Infrastructures for Knowledge Discovery framework, or SLIK-D, on leadership-class systems. Specifically, we are addressing scaling ML and DL algorithms to the Athena, Theta, and possibly Aurora supercomputers. We focused on (1) applications that apply DL and scalable data analytics to scientific application domains that require extreme scalability and (2) novel approaches to producing intelligent software systems for solving scientific problems without human intervention. We have completed work on evaluating the efficacy of DL for HEP and begun classification of features such as jets using HEP datasets. We are also developing a knowledgebase of machine learning toolkits, datasets, and models and are evaluating their efficacy on a wide-variety of systems.

LDRD# 2017-057	Advanced Computing
Project Title:	An Exascale Application for Simulating Urban Boundary Layers
Investigators:	Robert Jacob Aleksandr Obabko, Iulian Grindeanu, Rajeev Jain, Vijay Mahadevan

Abstract

An emerging challenge is the development of a model that assimilates data from urban sensor networks to provide urban weather forecasts for the next one to three hours. A community standard parallel high-performance computing model that combines 3D turbulent flow around and above buildings and in canyons, surface temperature models, and 3D radiative transfer does not yet exist. This modeling capability is necessary to fully realize the potential of urban observational networks and exascale computers for uses ranging from public health and safety to urban planning. The accurate description and resolution of complex spatio-temporal-scale interactions affecting the weather of a city will require a hierarchically coupled model approach, from regional climate models to urbanized mesoscale weather models and to detailed computational fluid dynamics. We have constructed an application and associated pre-processing tools for the highest-resolution component of that hierarchy: that is, an explicit physical model of the urban boundary layer that can be used to derive new parameterization schemes for regional climate and urbanized weather models. The development of such an application, focused on simulating the highly resolved turbulent airflow within a real city, requires tremendous computational resources. Current parameterization schemes for the urban atmospheric boundary layer are known to be deficient in representing momentum. To improve them, we created a model that explicitly simulates the winds in the boundary layer. The heart of this model is an existing fluid dynamics model developed at Argonne called Nek5000. We created a numerical mesh for Nek5000 in the urban domain of Chicago, specifically around the Lake Point Tower building in Chicago, to test the ability of Nek5000 to solve for the air motion around the structure. We verified its ability to do so, and now have a model ready for further enhancements that handle such effects as buoyance, etc.

LDRD# 2017-063	Advanced Computing
Project Title:	Enabling Multidimensional X-ray Nano-Tomography
Investigators:	Zichao Di Doga Gursoy, Francesco De Carlo, Si Chen, Stefan Vogt, Stefan Wild, Sven Leyffer

Abstract

In this project, we formulated new physical models to capture complex experimental setups; devised new mathematical optimization formulations and techniques for robust inversion of complex data; and provided scalable implementations of our models, formulations, and techniques that can run both on local machines and on clusters at the Advanced Photon Source (APS), as well as at the Argonne Leadership Computing Facility (ALCF). The results have been demonstrated on various datasets. This work anticipates the growth in amount and complexity of experimental data resulting from the APS upgrade. Full realization of the next generation x-ray source and its science output will rely on new analysis methods that leverage advanced computing, are capable of addressing massive volumes of data, and incorporate algorithms, such as those developed here, that mitigate sources of systematic and random errors like those due to probe drift – errors that can lead to reduced spatial resolution and even misinterpretation of underlying sample structures.

LDRD# 2017-066	Advanced Computing
Project Title:	End-to-End Genome Annotation and Phenotype Prediction with Deep Learning
Investigators:	Fangfang Xia Prasanna Balaprakash, Thomas Brettin, Venkatram Vishwanath

Abstract

To understand how biology really works, we need to bridge the divide between next-generation genome sequencing data and high-throughput phenotype screening data. Slow manual efforts cannot handle all the data, and shallow heuristics are not likely to grasp the complex connections. Machine deep learning can integrate all this information. Deep learning captures complex structures and nonlinear interactions in hierarchical abstraction layers. This is a promising approach, because genomes are full of hierarchical features interacting on multiple levels (nucleotides, motifs, genes, complexes, operons, pathways, communities). Our goal has been to build a scalable, extensible, interpretable model that bridges the phenotype-genotype divide. In this project, we developed a million-protein-scale genome annotation benchmark dataset leveraging decades of Argonne expertise in this area. In addition, we implemented deep, residual, convolutional neural networks that have been trained to classify 1,000 different protein functions with high validation accuracy. Large-scale and domain-specific benchmark data and reference implementations are critical to whether deep learning can make progress in a new domain. While we focused our initial effort on microbial genomics, the deep neural networks can be trained to recognize a much wider range of patterns.

LDRD# 2018-114	Advanced Computing
Project Title:	Computing Expedition 2018
Investigators:	Michael Papka Ian Foster, Paul Messina, Valerie Taylor

Abstract

The purpose of the project is to explore new computing technologies of interest to Argonne and to DOE. Investigators worked intensively for a three to four week period to develop algorithms, applications, and optimized codes using new computing technologies related to areas of quantum and neuromorphic computing and the Aurora 2021 (A21) exascale system at the Argonne Leadership Computing Facility (ALCF). Our effort included:

Quantum Expeditions:

Quantum Circuit Design; The most important stages in quantum circuit design flow are scheduling, placement, and routing processes of qubits and gates.

Scalable Quantum Optimal Control; Mapping logical quantum gates onto physical devices requires solving a quantum optimal control problem.

Neuromorphic Expeditions

Neuromorphic-Computing-Based Deep Learning for DOE Scientific Applications; To demonstrate the efficacy of neuromorphic computing (NMC) systems on DOE applications.

Real-Time Classification from Image Streams; Working on ways to automatically analyze images of objects in high throughput digitization pipelines.

Enabling Hyperparameters Optimization on Neuromorphic Computers; Hyperparameter optimization is an outstanding issue waiting for a solution in every aspect of science and engineering.

Neuromorphic Speed-Up of High Energy Physics Event Categorization; High energy physics machine learning (ML) algorithms, such as deep convolutional neural networks (CNNs), have been used to categorize events.

Neuromorphic Learning for Neuroimaging; We have built a deep learning model to predict clinical outcomes from 3D and 4D brain images.

Neuromorphic Event-Driven Dynamic Vision Sensing and Processing – Silicon Retina Imaging; We propose a responsive, high-spatial resolution imaging system, employing an event-based neuromorphic silicon retina imager.

Edge Processing on Neuromorphic Hardware: Modulatory Sparse Architectures Inspired on the Insect Brain; The ability to carry out online, fast, and dynamic processing and integration of multiple data streams is one of the hallmarks of biological systems.

A21 Expeditions

For Everyone A21: Distributed Electronic Structure Calculations Using a Globus-Enabled Programmable Cyberinfrastructure;

Mapping a 3D Reconstruction Algorithm to A21;

Tensor Contractions on A21;

Extreme Scale Systems for Machine Learning;

Reactor Simulation on A21;

Scale Up a New Argonne-Developed Calibration and Uncertainty Analysis Tool on A21;

Exploring A21 Programming Strategies for Bioinformatic Kernels;

Optimization of Light Source Data Analysis Tasks on A21 Exascale Supercomputer.

BIOLOGICAL AND ENVIRONMENTAL SCIENCE CAPABILITY DEVELOPMENT

LDRD# 2015-179	Biological and Environmental Science Capability Development
Project Title:	Illuminating Linkages Between Microbial Diversity and Biogeochemical Cycling in a Redox Dynamic Environment
Investigators:	Theodore Flynn Edward O'Loughlin, Kenneth Kemner, Pamela Weisenhorn

Abstract

Microbial communities are found in nearly every environment on Earth. Perhaps most importantly, they inhabit the “critical zone” (CZ) of air, water, soil, and sediment where they form the fragile layer that sustains Earth’s non-microbial residents. The elemental fluxes between different areas of the CZ drive most of Earth’s biogeochemical cycles, including the release or sequestration of greenhouse gases, the production or depletion of nutrients, and the fate and transport of anthropogenic contaminants. Most, if not all, of these fluxes are driven at least in part by microbial activity. By combining functional genomics approaches with detailed geochemical measurements (e.g., mass spectrometry, x-ray spectroscopy, gas/liquid chromatography, and microelectrode measurements) in field and lab-based experiments, we can shed new light on the intricate web of microbial processes that drive global biogeochemical cycles. In particular, the dynamic biogeochemical conditions of these environments are thought to cause large shifts in both the rates of greenhouse gas emissions and, concomitantly, the structure of microbial communities. The flux of methane from periodically flooded environments such as wetlands is a significant source of uncertainty in global climate. We uncovered basic organizing principles of biological systems in a freshwater wetland and saw how these relate to the emission of greenhouse gases such as carbon dioxide and methane in this critical environment.

LDRD# 2015-180	Biological and Environmental Science Capability Development
Project Title:	Functional Analysis of Proteins from a Key Signaling Network Involved in Plant Growth Promoting Bacteria
Investigators:	Marie-Francoise Gros

Abstract

Plant growth promoting (PGP) rhizobacteria exert their beneficial effects through direct and indirect interactions with plant roots, including mobilization of soil nutrients, production of antibiotics, and the elicitation of plant defense mechanisms. The underlying molecular mechanisms of the PGP activities of rhizobacteria remain poorly characterized. This limits our ability to design agricultural systems and cellulosic feedstock systems for biofuels that are both sustainable and highly productive. We therefore pursued mapping of the protein-protein interaction (PPI) networks in rhizobacteria that decipher environmental signals and initiate signal transduction pathways to trigger PGP effects. We focused on the PGP bacterial strains of Pseudomonas fluorescens. We succeeded in enhancing characterization capacity through using fluorescence microscopy, building a protein-protein network centered on a key signaling messenger in the environmental bacteria P. fluorescens, and developing a CRISPRi-based phenotyping approach to functionally explore the role of protein-protein complexes involved in biofilm formation at plant-roots.

LDRD# 2018-056	Biological and Environmental Science Capability Development
Project Title:	Linking Microbial Interactions to Biogeochemical Variability in Wetlands
Investigators:	Pamela Weisenhorn

Abstract

Microbiomes are important across many areas of our lives—health and disease, agricultural productivity and sustainability, industrial processes (such as oil refining and biofuel generation, as well as food production and processing), and maintenance of ecosystem processes in natural systems. Thus, an understanding that enables the management and manipulation of microbiomes is critical, and that requires an understanding of not only species composition, but also how those organisms interact to control the process of interest or to affect host biology. Critical knowledge gaps remain in our understanding of the interactions among community members, the stability and resilience of these interactions with respect to external conditions, and how changes in the frequency and strength of interactions influence the rate at which microbiomes affect their environment. We address these gaps by directly considering the effect of microbiome network structure, the pattern of interspecific interactions among microbes, on the stability of ecosystem processes, taking into account fluctuations in network structure through space and time. A method to combine mutual information criteria and random matrix theory to generate microbial interaction networks has been developed and tested on a dataset of weekly samples of sediment collected over one year. Data collection related to fine-scale temporal sampling of metabolic end-product emissions (carbon dioxide and methane) as well as continued sampling of soil, microbial, and water parameters was conducted. The analysis pipeline developed with this project was integrated into the KBase infrastructure. The analyses are used to examine the role of interspecific interactions in determining the impact of the microbiome on its environment and to develop a real-world relevant theory regarding application of these analyses.

ENERGY MANUFACTURING SCIENCE AND ENGINEERING

LDRD# 2016-023	Energy Manufacturing Science & Engineering
Project Title:	Real-time Monitoring of Material Structure Evolution in Additive Manufacturing Processes
Investigators:	Tao Sun Haidan Wen, Kamel Fezzaa

Abstract

Additive manufacturing (AM, or 3D printing) can be more efficient than traditional manufacturing techniques, and it eliminates conventional tooling constraints, allowing fabrication of products with extreme complexities. Most metal AM products are made with direct metal laser sintering (DMLS) in which an intense laser beam melts and sinters a layer of metal particles and bonds them to the previously processed layer. However, the end products sometimes contain structural defects that compromise desired characteristics. During DMLS, the metal particles can experience within less than a millisecond a sequence of sintering, melting, chemical reaction, solidification, and phase transformation. To probe the fast dynamics of laser-particle interaction, we apply in situ high-speed x-ray imaging and diffraction techniques with 100-picosecond time resolution. We focus on in situ measurement of processing parameters that determine the amount of energy delivered to the surface, including laser power, beam spot size, scan rate, scan line spacing, and geometrical scanning strategy. Various powders of two commonly used metallic alloys were studied: $AlSi_{10}Mg$ and Ti_6Al_4V. The in-situ analyses were supplemented by ex situ x-ray micro- and nano-computed tomography, electron microscopy, and conventional measurements of mechanical properties. We demonstrated the feasibility of imaging the laser AM process with a rate of 10 million frames per second and developed computer codes for extracting quantitative structural information from X-ray data on melt pool dynamics, depression zone fluctuation, powder ejection velocity, solidification rate, and phase transformation rate. Direct observation of the formation of various defects, including powder-induced porosity, trapped-gas porosity, keyhole porosity, and hot cracks, was achieved.

LDRD# 2016-159	Energy Manufacturing Science & Engineering
Project Title:	Additive Manufacturing for Nuclear Energy Applications
Investigators:	Abdellatif Yacout Kent Wardle, Kun Mo, Michael Pellin, Peter Arron Kozak

Abstract

Additive manufacturing (AM) or three-dimensional (3D) printing is a tool to produce precise and complex forms and/or structures with the potential for application to large-scale manufacturing. AM involves manufacturing a part by depositing material layer by layer. Compared to injection molding or machining and/or subtractive technologies, AM has several advantages, some of which can be of particular benefit to nuclear energy applications. AM expands the options for complex structures; component materials (e.g., fluoropolymers for chemical resistance); and embedding of catalysts, reactants, and adsorbents, which could lead to hybrid devices that provide multiple separation pathways within a single device. We have demonstrated the possible role of AM in improving the

processes of material and components production. The main application considered here is the application to nuclear fuel (i.e., actinide) materials. We delivered several independent methods for low-cost, scalable 3D printing of metal for nuclear fuel applications: (1) use of metal-impregnated filament; (2) use of metallic paste and/or slurry; and (3) combining multiple advanced powder infiltration methods with AM techniques. All these techniques require post-processing either to remove the plastic matrix material or/and to sinter the part, and all the methods involve the challenges of dimensional changes during sintering and porosity mitigation. Our demonstrations of the feasibility of working with actinide metals included 3D printing of UO_2 fuel pellets and fabrication of U-Mo foils.

LDRD# 2017-042	Energy Manufacturing Science & Engineering
Project Title:	Metal Additive Manufacturing Modeling
Investigators:	Marius Stan, Michael North Angel Yanguas-Gil, Dileep Singh, Shashikant Aithal

Abstract

Additive manufacturing (AM) is the process of joining materials to make objects, usually layer upon layer, from three-dimensional model data. Use of AM for metal production components has been limited by a lack of fundamental scientific understanding of precursor materials and AM process parameters related to consistency and performance of the manufactured components. We have produced models of metal AM processes, by using both powder and wire feedstocks, that run at sufficient speeds to meet practical industrial and government needs. We used a two-stage approach. In the first, we constructed, calibrated, and validated fine-grai Machine learning (ML) and deep learning (DL) are having impact on applications ranging from self-driving cars to natural language processing. A key challenge in applying ML and DL is their adaptation to scientific datasets, which are diverse with respect to their data models and structures, size, complexity, and other parameters, and are increasing in complexity as datasets from several scientific domains are combined. Despite initial successes in executing on a set of a few nodes, effective ML and DL software that fully exploits the architectural features of supercomputers is lacking. We are developing scalable ML and DL infrastructure to exploit data-driven and computational models in science and the architectures of supercomputers. We are collaborating with application scientists in genomics and high-energy physics (HEP). To achieve our goal, we are developing the Scalable Machine Learning Infrastructures for Knowledge Discovery framework, or SLIK-D, on leadership-class systems. Specifically, we are addressing scaling ML and DL algorithms to the Athena, Theta, and possibly Aurora supercomputers. We focused on (1) applications that apply DL and scalable data analytics to scientific application domains that require extreme scalability and (2) novel approaches to producing intelligent software systems for solving scientific problems without human intervention. We have completed work on evaluating the efficacy of DL for HEP and begun classification of features such as jets using HEP datasets. We are also developing a knowledgebase of machine learning toolkits, datasets, and models and are evaluating their efficacy on a wide-variety of systems.ned physics- and chemistry-based models produced via multiphase continuum simulations and integrated computational materials engineering (ICME) methods, respectively. In the second stage, we used input from the first stage and several machine-learning techniques, to construct reduced-order models that can serve as useful tools for industry and government. Real-time thermal modeling of the electron beam additive manufacturing process was accomplished using an open-source spectral element

code, Nek5000. Efforts were directed towards model and code development and the study of modeling the e-beam energy deposition into a substrate. Code and model development was also required to deal with layer addition on a heated substrate, a more involved problem, since it requires superimposition of a new grid for the additional layer on the thermal field computed for the previous layer. The alloy Ti6Al4V was modeled and powders used for laser AM production of test samples, the microstructure of which were characterized with x-ray tomography and electron backscatter diffraction for successful model validation.

LDRD# 2017-084	Energy Manufacturing Science & Engineering
Project Title:	Advancing Additive Manufacturing of Metal Alloys; from Fundamental Principles to Durable Components
Investigators:	Aaron Greco Dileep Singh, Maria De La Cinta Lorenzo Martin

Abstract

We sought to develop a link from fundamental material characterization and modeling to applied engineering evaluation and optimization of additively manufactured (AM) metallic components. Our goal has been to establish a new approach relating microstructure/porosity to nano-mechanical properties, corrosion, and component level reliability. AM/3D printing enables not only unique macrostructural design of components, but also micro-/nano-structural material design. AM's layer-by-layer forming process permits localized material properties tailored for specific applications. However, a lack of fundamental understanding of precursor material and AM processing parameters as they relate to final component structural homogeneity and mechanical performance has been limiting. During AM, the material experiences thermal cycling and can see repeated solid-state and liquid-solid transformations. This leads to complex microstructural textures and grain orientations that are not typically found in conventionally manufactured materials. Components can exhibit increased residual stresses, porosity, and surface roughness. But AM can also be used to engineer material microstructure to enhance a component's performance beyond that attainable through conventional processes. We began by characterizing feedstock metal powders to be used to print experimental samples for model validation. We selected the titanium alloy Ti-6Al-4Al for our initial focus and added both Inconel nickel-chromium-based superalloy and the aluminum alloy Al-10Si-Mg to the study. The characteristics of the powders were used to identify the material properties for the integrated computational materials engineering (ICME) modeling and any discrepancies in the resulting builds. In-situ x-ray observation and post-build materials characterization were used to relate the precursor powders and AM process parameters to the final product's properties. Over 35 experimental runs were conducted and analyzed, including the addition of metal-ceramic composite precursors. The experience we gained and documented from precursor and process parameters to product outcomes will serve future development of the AM technology at Argonne well.

LDRD# 2017-159	Energy Manufacturing Science & Engineering
Project Title:	Production of Cellulose Nanocrystals from Miscanthus x. Giganteus
Investigators:	Gregory Krumdick

Abstract

Cellulose—the most abundant organic compound on earth—occurs naturally as a hierarchical architecture of fiber bundles that contain both highly crystalline and amorphous domains. Removal of the amorphous region allows access to highly crystalline, rod-like cellulose nanocrystals (CNCs), and the hydrolysis of natural cellulose with hydrochloric acid to yield CNCs is well known. Because of the inherent properties of CNCs, their surface structure is easily modified. The crystals are also very strong and exhibit high thermal stability. This strength and customizability allow CNCs to be used in many applications. They have applications for advanced battery and catalyst material, as well as additives to change the properties of other materials. This project sought to develop an advanced industrial process for the manufacturing CNCs from Miscanthus x Giganteus grass, a perennial, non-invasive grass hybrid. Small scale synthesis had been achieved, but the current lab scale processes for isolation and purification are not suitable for large scale production. We had to take the laboratory batch process from the gram to the kilogram scale. An industrial process must produce CNCs that match the properties from the small-scale process. It should produce crystals that are 5 nm wide and 200–300 nm long. The overall crystallinity of the product should be greater than 80%. The process must be optimized for efficiency to minimize waste production and energy use. A new industrial process was developed that significantly reduces the cost of producing cellulose nanocrystals. This was accomplished by the invention of an intensified unit operation that significantly reduces waste and allows for the manufacture of cellulose nanocrystals with 30% less equipment than would be required using prior art.

LDRD# 2018-003	Energy Manufacturing Science & Engineering
Project Title:	Atoms to Additive Manufacturing (A2AM) - Machine Learning Structure-Property-Process Triangle In Metals Additive Manufacturing
Investigators:	Olle Heinonen Badri Narayanan, Mathew Cherukara, Ross Harder, Subramanian Sankaranarayanan, Tao Sun, Thomas Peterka, Yuzi Liu

Abstract

Many materials properties depend on microstructure (e.g., grain sizes and microphases). Microstructure in turn has a complex dependence on composition, processing conditions, intrinsic materials properties, and kinetic properties. A long-standing challenge has been predicting a material's microstructure and, by extension, its microscopic and macroscopic properties. We pursued development of a predictive, data-driven framework that integrates machine learning and high-performance computing (HPC) by linking atomistic modeling and high-quality data sets with meso- and macroscale modeling. Machine learning is used to bridge different length and time scales associated with microstructural evolution. Our development work, as well as validation and verification, focused on two systems, namely precipitation-hardened aluminum (Al) and Co-based superalloys. This project made great advances in combining machine learning, molecular dynamics, continuum modeling, atop probe tomography, and X-ray studies of advance manufacturing processes. We generated high-quality interatomic potentials to study these

alloy systems, especially for nucleation of grains or precipitates and for grain growth. We analyzed samples of γ/γ' Co-Al-W superalloy using x-ray coherent diffraction imaging. We used several sets of 3D atom probe tomography data to build machine learning models in order to construct surrogate models for parameters needed in continuum modeling, and also to validate the molecular dynamics models. Lastly, we conducted experiments on laser melting and re-solidification using high-speed X-ray imaging.

LDRD# 2018-026	Energy Manufacturing Science & Engineering
Project Title:	Advanced Composite Materials Development for Nuclear Energy Applications
Investigators:	Abdellatif Yacout Michael Pellin

Abstract

Advanced nuclear reactor concepts require corresponding innovations in materials to meet the demand of their aggressive environment, which can include high-temperature corrosion challenges (e.g., molten salt and lead), enhanced accident tolerance, and high irradiation doses. Our work targets innovations in the manufacturing of material to support the development of such advanced nuclear energy systems. Specifically, we are developing a novel method for synthesis of composite materials by combining atomic layer deposition (ALD) with an advanced fast deposition technique (especially for ceramic materials), electrophoretic deposition (EPD). The combined method can be used to synthesize high-density composite material directly on a substrate to form a barrier coating, which can protect against extreme conditions in a nuclear reactor, or be combined with another material to form a complex composite with superior thermo-mechanical properties. We have deposited materials such as $ZrSiO_4$, SiO_2, and CeO_2 with EPD over Zircaloy and 316 stainless steel substrates, followed by ALD infiltration coating. We developed a composite within a 3D printed hexagonal architecture via infiltration of CeO_2 nano-powder with EPD followed by ALD alumina. Development of a ceramic cloth enveloped cladding tube, via off-the-shelf SiC cloth, EPD SiC and ALD-infiltrated $ZrSiO_4$, was successful. Studies were also made of infiltration depth of various ALD coatings in EPD coated SiC and its effect on coating adherence. Finally, coatings' tribological, mechanical, and corrosion resistance properties were also tested.

HARD X-RAY SCIENCES

LDRD# 2016-150	Hard X-Ray Sciences
Project Title:	A Conveyer Belt of Nanoliter to Picoliter Droplets for Hard X-ray Pump-probe Experiments
Investigators:	Anthony DiChiara Bin Hu, Chris Benmore, Kamleshkumar Suthar, Karl Unruh, Yuan Gao

Abstract

We are developing a sample management system (SMS) that provides a high degree of control for designer quantities of precious aqueous/liquid systems, such as nanoparticles (NPs), bio-molecules, fuels, and photocatalytic compounds. A containerless and contact-free sample environment of 10-µm

droplets will take full advantage of the increasingly smaller size coherent beams available at the Advanced Photon Source. These beams provide short x-ray pulses with long separation times, allowing isolation of single x-ray events with sub-nanosecond resolution using laser/x-ray "pump-probe" techniques. Pump-probe is stroboscopic and requires multiple identical events. We are combining existing acoustic levitation and optical trapping technologies to achieve a conveyer belt of droplets that can be advanced or held, depending on the needs of an experiment. Piezo-actuated nozzles are used to generate a variety of droplet sizes and the droplet array can be advanced on demand by phase shifting the acoustic wave in synchronization with the x-ray source. The studied systems include solid-state diffusion by ultrafast laser sintering of NPs; solution scattering with caged compounds; and advanced manipulation of objects within liquid environments to rotate or translate NPs within a single droplet. Focused sound waves were shown to reduce the droplet jitter by two orders of magnitude. We have developed a COMSOL simulation that models the mechanical geometry of our ultrasonic acoustic transducers to accurately simulate the resonator sound field. We have conducted container free SAXS/WAXS experiments on palladium NP formation and phase formation induced by super-saturation in several salt solutions. Self-assembly of silica particles in evaporative environments and growing conductive polymers in a container-free environment are additional experiments being pursued.

LDRD# 2017-028	Hard X-ray Sciences
Project Title:	Universal Superconducting Undulator
Investigators:	Yury Ivanyushenkov Joel Fuerst, Matthew Kasa

Abstract

The Advanced Photon Source (APS) Upgrade will make possible a round beam vacuum chamber as a result of a reduction in the horizontal dimension of the electron beam. An opportunity exists to develop an electromagnetic universal superconducting undulator (SCU) that employs four planar magnetic structures (jaws) around a cylindrical beam chamber. A higher magnetic field will be produced, generating higher photon fluxes at higher energies, through use of superconducting instead of normal conducting wire. Circular superconducting devices have been unworkable in current electron storage rings that have beam vacuum chambers of a wide rectangular shape. Our goal has been to enable fast (>10 Hz) polarization switching by using small current bumps to shift undulator harmonic energy in either one of two universal SCU devices collocated in a straight section of the storage ring, each preset to a well-defined polarization state. Realization of such fast polarization switching rates in the hard x-ray regime, while preserving x-ray flux, has not been possible with permanent magnet undulators or crystal optics. We have now successfully tested a prototype SuperConducting Arbitrarily Polarizing Emitter, or SCAPE undulator.

LDRD# 2017-104	Hard X-ray Sciences
Project Title:	Integrated Imaging
Investigators:	Amanda Petford Long, Charudatta Phatak Doga Gursoy, Francesco De Carlo, Haidan Wen, Ian McNulty, Jeffrey Guest, Lynn Trahey, Maria Chan, Nicola Ferrier, Ross Harder, Subramanian Sankaranarayanan, Sven Leyffer, Thomas Peterka, Todd Munson, Xianghui Xiao, Yuzi Liu, Zhonghou Cai

Abstract

Most materials and systems are spatially complex and heterogeneous, and their behavior is typically linked to this heterogeneity. Imaging and microscopy offer a way to see a material in all of its complexity and to explore its local behavior; when combined with spectroscopy, diffraction, or other analytical methods, they allow one to understand what one sees. This combination of seeing and understanding is especially important when developing materials for efficient and sustainable energy generation, storage, and use. We sought an integrated, top-down approach to scientific discovery that integrates image-based information representations driven by pending hypotheses. This approach demands a perspective on the properties of a wide range of imaging modalities, tools, measurement protocols, and analytical techniques. We pursued four interacting subtasks: 1. Framework for integrated multimodal imaging of materials for energy storage; 2. Integrated imaging, modeling and analysis of ultrafast energy transport in nanomaterials; 3. Integrated imaging to understand and advance photocatalysis; and 4. Smart acquisition and sampling for simulation and imaging (SASSI). In pursuit of these goals, we have developed a dynamic sampling method based on a supervised learning algorithm and convolutional neural networks (CNNs) for data acquisition in a scanning electron microscope (SEM). We have demonstrated the results for (1) energy-dispersive X-ray spectroscopy (EDX) mapping and (2) secondary electron imaging. For EDX mapping, our method uses CNNs that use a dictionary for training and classification of the EDX spectra. We have implemented this on the desktop SEM for experimental acquisition of data. We have shown we can achieve high quality elemental maps with as low as 5% sampling. For secondary electron imaging, we used deep neural networks to predict optimal sampling locations based on a set of training images, and then reconstructed the final image. The network can be pre-trained using generic images available online, and was capable of reconstructing images with high signal to noise ratio with sampling as low as 20%. One impact of this project is that our methods apply to beam sensitive materials as well as to high-throughput materials characterization needed for applications such as in additive manufacturing.

MATERIALS AND CHEMISTRY

LDRD# 2016-120	Materials and Chemistry
Project Title:	New Thin Film Oxide, Chalcogenide and Oxy-chalcogenide Materials Discovery
Investigators:	Supratik Guha

Abstract

As computing workloads have changed and Moore's Law scaling ends, there is interest in new computing architectures such as neuromorphic computing, where the computing hardware elements

are akin to the functionality of neurons and synapses. The objective of this project is development of new families of electronic and optoelectronic materials from new oxide and chalcogenide thin films and nanostructures. We focus on the development of novel resistive switching materials and devices for application in solid-state memories and neuromorphic circuits. The emphasis is on new oxide materials with ultra-low power operation. Our research program involves materials synthesis using ultra-high vacuum deposition and atomic layer deposition, nanoscale device fabrication, materials characterization, and electrical measurements. Materials studied include new polymer-metal oxide nanocomposite films, VO_2, CeO_2, $Y2O_3$, HfO_2, and their heterostructures. We developed a novel resistive memory device with an ultra-porous metal-oxide/polymer nanocomposite material, polymethyl methacrylate (PMMA)-Al_2O_3. The nonvolatile switches operate at very low voltage (< 500 mV) and with high reliability. Materials having an electrically driven insulator-to-metal transition (E-IMT) are a promising candidate for neuromorphic circuit components. The current on/off ratio and voltage scalability were investigated in nanoscale E-IMT structures.

LDRD# 2016-123	Materials and Chemistry
Project Title:	New ^{6}Li Rich Semiconductors for Neutron Detection
Investigators:	Mercouri Kanatzidis Duck Young Chung

Abstract

There is a need to develop solid-state neutron detectors that are highly efficient, small, lightweight, portable, and low cost. These devices are important in preventing the proliferation of illicit nuclear materials. Unfortunately, the worldwide shortage of ^{3}He is a problem for the most widely used neutron detectors, ^{3}He proportional counters. A semiconducting material that contained a neutron absorber within its regular stoichiometry could be more efficient than a traditional p n junction device at recording the kinetic energy of a neutron-interaction-produced charged particle. If a voltage could be applied across such a bulk material, neutrons could be absorbed directly within the body of the bulk crystal. The resulting charged particle reaction products would deposit their energy directly within the bulk detector material, which would also serve as a charge transport medium. The idea of solid-state bulk materials in which the crystal lattice has ^{6}Li as a constituent to directly detect thermal neutrons is attractive. The materials to be investigated were semiconducting compounds that contain Li as a strong neutron-absorbing element and a good balance of properties needed for good, room-temperature thermal neutron detection. Through a wide range of chemical approaches and characterization and screening/evaluation of their sensitivity to thermal neutrons, we identified one new material, $LiInP_2Se_6$, suitable for thermal neutron detection. The compound we discovered can be grown at low cost as a large crystal and readily developed for the production of devices.

LDRD# 2016-133	Materials and Chemistry
Project Title:	Managing Emission and Thermal Absorption
Investigators:	Pierre Darancet Alex Martinson, Anand Bhattacharya, Gary Wiederrecht, Maria Chan, Richard Schaller, Stephen Gray, Subramanian Sankaranarayanan

Abstract

There is a need for better thermal management of devices in many fields. For example, it would affect the miniaturization of electronic components, durability of new-generation solar cells, and efficiency of thermoelectrics and thermophotovoltaics. Advanced nanoscale materials have properties that offer new degrees of freedom for controlling heat absorption, emission, and transport. Managing heat at the nanoscale has been challenging because the basic physical phenomena underlying heat absorption, emission, and transport require understanding and control of electron-phonon, photon-phonon, and phonon-phonon interactions. Through coordinated efforts in device/materials synthesis, characterization, and modeling, we are improving our understanding of the fundamentals of heat and thermal management at the nanoscale. We devised a novel computational approach capable of treating coupled electron-phonon dynamics, performed a set of seminal experiments in the field of the thermal properties of hybrid organic-inorganic perovskites, and optimized thermophotovoltaic emitters. For all materials, we found that the decay in electronic energy is concomitant with the appearance of a significant fraction of non-thermal vibrational modes. This result is in strong contrast to the current understanding of electron thermalization in metals and originates from the large heterogeneity in the time scales of electron–phonon and phonon–phonon interactions in these materials. We proposed a generalized two-temperature model accounting for these effects and used it to understand the nanosecond-long thermal equilibration processes we measured in hybrid perovskites.

LDRD# 2017-032	Materials and Chemistry
Project Title:	Catalysts Modeled After Nature's Enzymes
Investigators:	David Tiede Karen Mulfort, Lisa Utschig-Johnson, Massimiliano Delferro, Phani Pokkuluri, Rachel Klet, Yuping Yuan

Abstract

Our work fits into the rapidly expanding field of bio-inspired catalysis. The project combines typically non-overlapping expertise in synthetic chemistry, materials, and biology to create a coordinated research capability directed at the design of bio-inspired architectures for multistep catalysis modeled after Nature’s enzymes. We investigated the concept of coupling synthetic catalytic chemistry to biological redox chemistry, a new approach for bio-inspired catalytic chemistry, one targeted at directly linking primary light-induced redox reactions to energy-conserving hydrogenation catalytic chemistry. The project involved the development of catalysts and of host-guest chemistry for transfer hydrogenation chemistry, using both biological and synthetic host-guest frameworks. Success was demonstrated when we identified a light-excited state initiated transhydrogenation reaction pathway for an iridium complex, and developed a function redox protein host framework. Exploiting reaction landscapes using the excited states of visible light-absorbing catalysts represents a fundamentally new

contribution to the field of photoredox catalysis, and this work establishes approaches to developing bio-hybrids as host environments for synthetic catalysts and spatially organized reaction sequences.

LDRD# 2017-033	Materials and Chemistry
Project Title:	Advanced Materials for the Energy-Water Nexus
Investigators:	Seth Darling Alex Martinson, David Tiede, Di-Jia Liu, Jeffrey Elam, Juan De Pablo, Nestor Zaluzec, Paul Nealey

Abstract

True innovation in the energy-water nexus (EWN) space will hinge less on incremental improvements in traditional water technologies, such as reverse osmosis or chemical disinfection, and more on materials and engineering breakthroughs that create new sources of water at "pipe parity," i.e., where the costs of new supply are comparable to existing regional water sources. We applied energy efficiency, synthesis science, and biomimetic catalysis to develop advanced, integrated materials methodologies that offer flexible means of removing targeted species from water streams that have been challenging with traditional techniques. We target the most energy-intensive components of purification including low-concentration metals, dilute hydrocarbons, and steroids. Advanced materials, integrated into membranes and other porous media, have the potential to revolutionize water usage as we know it. Specifically, a variety of interface engineering strategies were developed to tune the properties of membranes and other components in water systems. Functionality ranging from passive anti-fouling to catalytic activity was demonstrated.

LDRD# 2017-035	Materials and Chemistry
Project Title:	Self-assembling Soft Nanostructures with Ultra-Slow Dissociation Kinetics
Investigators:	Jeffery Hubbell

Abstract

We focused on self-assembly of block copolymers of poly(ethylene glycol) (PEG) and oligo(ethylene sulfide) (OES). The OES domain is hydrophobic, which drives self-assembly in water; and the OES domain crystallizes, which drives very slow dissociation kinetics, yielding ultra-stable self-assemblies. Our objectives included assembling ultra-small, ultra-stable micelles for drug delivery, as well as self-assembling monolayers on gold that are highly stable and highly resistant to protein and cell adsorption for bioanalytical purposes. A combined computational and experimental approach was used to characterize these structures and guide design. Computationally, we found that micelles are about 7 nm in diameter, which is consistent with experimental measurements. We have also simulated the degree of crystallinity in the micelle core, which is approximately 40% and very difficult to measure experimentally. The computational model recapitulates very well what we see experimentally and sheds light on difficult-to-measure properties. We used ellipsometry and grazing-incidence wide-angle X-ray scattering (GIWAXS) to reveal an ordered structure for monolayers deposited on gold in agreement with computational predictions. Finally, we have coated quartz crystal microbalance chips to quantify these

films' resistance to protein adsorption and found a factor of ~100 reduction in protein coverage for the OES-PEG case studied. From a fundamental perspective, our work offers a new design approach that can be used by others. From an application perspective, our approach enables paths to (bio) analytical devices (e.g., spherical micelles, high surface area nanofibrils, ultrathin films), surface patterning (thin films), and separations (self-assembled membranes).

LDRD# 2017-050	Materials and Chemistry
Project Title:	Oxides for Novel Computational Approaches
Investigators:	Axel Hoffmann Anand Bhattacharya, David Awschalom, Dillon Fong, Giulia Galli, Supratik Guha

Abstract

Oxides offer a powerful platform to create, manipulate, and transfer quantum states within and across material heterostructures. Spin-based defects in this class of systems may be controlled optically, magnetically, electrically, and mechanically and may enable micrometer-scale optical patterning and broad tuning of the chemical potential. We explored quantum coherence in individual defect states of oxides, with the goal of harnessing these states for quantum computational information-processing schemes. We also used controlled manipulation of oxygen stoichiometry to explore development of biologically inspired neuromorphic computational schemes. Our approaches build on the realization of dynamic and reversible control of defects through local electric fields, which enables a reconfigurable energy landscape for various states of condensed matter. We demonstrated that optical excitation in strontium titanate ($SrTiO_3$) can be used for local gating of topological insulators. Using excitations at different wavelengths, we independently controlled magnetic properties and the chemical potential. Furthermore, we demonstrated that there is a more general scheme for local optical gating by combining other low-dimensional materials, such as graphene or molybdenum disulfide (MoS_2), with $SrTiO_3$. Toward the goal of electrically manipulating resistive states, we fabricated memristive devices based on strontium cobaltate ($SrCoO_x$) and directly imaged the electrochemical transformations using spatially resolved diffraction and chemical mapping with advanced X-ray characterization at the Advanced Photon Source. Finally, we developed a theoretical framework for predicting and guiding the search for quantum states in oxides, synthesized and characterized spin-defect states in oxides, and prepared and optimized SrFeO3-x-based analog state devices. We modeled electron-nuclear spin interactions and loss of quantum coherence in oxides, demonstrated coherent control and measurements of spin decoherence, and demonstrated basic neuromorphic functionalities with $SrFeO_{3-x}$-based memristors.

LDRD# 2017-061	Materials and Chemistry
Project Title:	Towards an Artificial Neuron - Non-Covalent Synaptic Assemblies
Investigators:	Harry Fry Boguslaw Nocek, Edward Barry, Elena Rozhkova, Philip Laible, Tijana Rajh

Abstract

Electrochemical energy moves across biological systems via ion cascades, triggered by neurotransmitters that depolarize a cell and cause a minute voltage spike. We are fabricating artificial bio-hybrid assemblies capable of mimicking this process. Demonstration of successful control over ion flow represents the first step toward the development of an artificial neuron. All transmembrane proteins involve the membrane as the scaffold and the protein as the functional unit. We developed functional peptide materials that served as both the scaffold and the functional component and were able to develop a series of peptides that self-assemble into bilayer assemblies with rich structural behavior and unusual transformation capabilities. Alternatively, spherical colloidal assemblies, also known as colloidosomes, can be formed surrounding water droplets in a hydrophobic solution that contains nanoparticles. These structures may function as surrogates to a lipid bilayer. Colloidosomes are of particular interest in this project as they respond to external stimuli, such as pH and temperature, and thus may be programmable. The generation of an artificial organelle was realized in the assembly of a colloidosome – gold/silver nanorods assembled into hollow spheres in a nanoemulsion. It was wrapped with “purple membrane” (a light activated proton pump bacteriorhodopsin). Upon illuminating the ensemble with white light, proton pumping of the purple membrane was enhanced by the plasmon resonance of the Au/Ag nanorods. We paired the colloidosome with an ATP synthase functionalized liposome and demonstrated communication between the colloidosome and liposome. That observation achieved a goal of this project which was to demonstrate cascading reactions much like in neuronal transmission.

LDRD# 2017-082	Materials and Chemistry
Project Title:	Developing Hierarchical Multi-functional Hybrid Polymer-Proteins Structures for Energy Applications
Investigators:	Tao Li Gyorgy Babnigg, Karolina Michalska, Lei Cheng, Randall Winans, Wei Chen, Yuzi Liu

Abstract

In order to enable design of truly integrated, multifunctional bio-hybrid materials, we proposed to develop new synthesis and assembly strategies for molecular-level control of the soft interface between polymers and proteins. A fundamental understanding of the molecular-level interaction during the formation of these hierarchical structures can guide transformative synthesis of hybrid polymer-protein nanoparticles. It is a challenge to develop a one-step control of structural features of proteins while preserving their activity and conformation. We have developed a method that allows enzyme conformation and functionality to be preserved by assembling the polymers and enzyme into a hierarchical structure. In addition, in situ X-ray scattering has been used to study the co-assembly process of Poly(4-vinylpyridine) (P4VP) and protein (apoferritein). Once protein and polymer were

mixed, the assembly occurred immediately, and the protein remained in its spherical shape. We also included tests of the effect of various water-soluble solvents on the hybrids, and we performed density functional theory (DFT) calculations comparing binding energies of several polymer-amino acid combinations. The methods developed in this project are broadly applicable for many enzymatic systems but we focused here on hierarchical polymer-cellulase structures for biomass conversion.

PHYSICAL SCIENCES AND ENGINEERING

LDRD# 2015-181	Physical Sciences & Engineering
Project Title:	Fine Resolution Reconstruction of Large Volumes of Brain
Investigators:	Narayanan Kasthuri

Abstract

The current challenge to map all the neurons and connections at the nanometer scale in a large volume of brain is addressed by this project in a unique manner. To date, most of the approaches elsewhere focus on serial section electron microscopy with variations in the way serial sections are produced and the type of electron microscopy used. We used serial section electron microscopy for the collection of data and determined how synchrotron-based high-energy X-ray microscopy of the same brains complement and extend such data. The Advanced Photon Source (APS) at Argonne is ideal for such experiments. Specifically, we used tape-based electron microscopy and have mapped an entire mouse brain. We developed the methods needed to employ high-energy X-rays from the synchrotron source to complement electron-based reconstruction approaches. Synchrotron based full-field nano-CT (computed tomography) holds the key to map the cellular composition of the entire brain cells. In addition, we pursued the implementation of automated algorithms based on machine learning to automatically trace neurons and their connections. These algorithms are now potentially the fastest and most powerful computational resources for brain mapping.

LDRD# 2016-191	Physical Sciences & Engineering
Project Title:	Exploring Next Generation Coherent X-ray Science
Investigators:	Linda Young

Abstract

Pulses from x-ray free electron lasers (XFELs) provide enough fluence per pulse in a timescale sufficiently short to enable single-shot imaging and freezing the fastest molecular vibrations—and thus monitoring atomic and molecular motions on their natural timescales. This project aimed to define the science problems that make use of these revolutionary properties. We also wanted to identify and investigate potential scientific applications that require high pulse energy, temporally coherent, x-ray pulses in regimes that are not accessible with present XFELs, synchrotrons, or tabletop sources. Two questions to which we have applied computation and planned experimental observation are: (1) the birth and fate of an excess electron in liquid water created by ionization and (2) in pressure-induced phase transitions, what moves first, the atoms or the electrons.

SECURING ENERGY AND CRITICAL RESOURCES

LDRD# 2016-126	Securing Energy and Critical Resources
Project Title:	Advanced Control Algorithms For Improving Energy Consumption of Connected and Automated Vehicles
Investigators:	Jeffrey Larson Dominik Karbowski, Joshua Auld, Todd Munson, Vadim Sokolov

Abstract

Improving the highway system's capacity is a major benefit of vehicle automation, but there has been little research on how to make the system more efficient through automation and connectivity. Simulation tools for evaluating new connected/automated vehicle (CAV) technologies are being developed at Argonne, but to fully understand their implications, sophisticated mathematical and computational algorithms are needed to model control strategies that could improve traffic flow. We are developing such models and algorithms to control the speed and motion of CAVs. New algorithms to process large-scale traffic-flow data are being developed and analyzed for their sustainability and optimal energy benefits. Vehicle platooning, in which vehicles travel in a group with small inter-vehicle distances to reduce aerodynamic drag, has been a largely unaddressed approach to saving fuel. We are studying algorithms for maximizing fuel savings from CAVs throughout a large metropolitan area. Advanced control algorithms enabled by automation and connectivity could produce an energy-efficient roadway. In close collaboration with transportation system modelers, vehicle modelers, and computational scientists, we developed methods which showed that as vehicles increase their willingness to coordinate with other vehicles, the potential savings also increased. The proof-of-concept work in this project was interesting to DOE and is now directly funded by DOE through the SMART Mobility consortium.

LDRD# 2016-152	Securing Energy and Critical Resources
Project Title:	Integrated Water-Energy Systems Assessment Framework (IWESAF) for Water-Energy Sustainability and Resilience
Investigators:	Yuejun Eugene Yan Christopher Harto, Cong Liu, Corrie Clark, Feng Qiu, Getnet Betrie, Jenna Schroeder, Jiali Wang, Jianhui Wang, Josephine Wang, Leslie Poch, Matthew Mahalik, May Wu, Mi-Ae Ha, Thomas Veselka, Todd Levin, Vinod Mahat, Vladimir Koritarov, Zhi Zhou

Abstract

We are developing an Integrated Water-Energy Systems Assessment Framework (IWESAF) to evaluate and enhance the sustainability, reliability, and resilience of the energy-water systems. Existing models primarily focus on individual or a small set of sectors and processes and do not capture the complexity of the energy-water nexus (EWN). Water-energy systems are often characterized by multiple interactions, feedbacks, and tradeoffs that occur at multiple scales, across multiple sectors, and among various physical processes and human activities. To address this challenge, the IWESAF will be capable of (1) representing a full spectrum of relevant sectors and processes with temporal and spatial

scale-dependent interactions among them; (2) characterizing energy-water risk, uncertainty, and vulnerability; and (3) assessing different adaptation strategies and accounting for tradeoffs, co-benefits, and uncertainties associated with water-energy-related actions. This requires the simulation of natural, engineered, and human systems and their complex and dynamic interactions. The main IWESAF objectives were to (1) develop an energy-water framework that incorporates existing models (with modifications) to represent major energy sectors, hydrologic cycles, and human activities relevant to energy-water systems; (2) create a software library of utilities that process temporal and spatial coupling, allow for automatic aggregation and disaggregation of system components, and facilitate simulation of cross-domain feedbacks; and (3) validate IWESAF functionality and capability for various applications, such as evaluation of climate change, growth of demand, and adaptation and optimization strategies. We've built a database for energy and water systems and developed some key system components, including regional hydrologic and riverine thermal regime modeling, a climate and hydrologic extremes generator, a module for thermal power plant simulation, a risk-based reservoir adaptive management model, an energy-water economic model, and a power grid system model that includes thermo-, hydro-, solar, and wind power plants and linkages to other model layers.

LDRD# 2016-158	Securing Energy and Critical Resources
Project Title:	Production of Medical Isotopes using the Argonne Electron LINAC
Investigators:	Jerry Nolen, Michael Alex Brown Dave Rotsch, George Vandegrift, Jerry Nolen, Michael Alex Brown

Abstract

Relying on the photonuclear reaction mechanism, we researched medical isotope production possibilities at the 50-MeV/25-kW Argonne Electron Linear Accelerator (Linac) in Argonne's low-energy accelerator facility (LEAF). The LINAC is capable of producing large quantities of unique isotopes with bremsstrahlung photons. Two novel, tumor-destroying therapeutic radionuclides, 47Sc (scandium-47) and 225Ac (actinium-225), were chosen as appropriate for development. With titanium and radium targets, respectively, we explored the unique and challenging aspects of isotope production targetry. In each case, target preparation, accelerator tuning and preparation, radiometric analysis, chemical processing, and product evaluation were done. During the three years of this project, we demonstrated the potential viability of producing these two isotopes at LEAF. Continued development of isotope production methodology is being supported by the DOE isotope program. These isotopes will be added to the DOE Isotope Program Catalog available to the medical isotope community in the near future, supplementing the previously added 67Cu isotope for use in targeted radiotherapy. Several invention reports resulted from this project.

LDRD# 2017-040	Securing Energy and Critical Resources
Project Title:	Development of Molten-Salt Reactor Analysis Computation Tools to Support Emerging Markets
Investigators:	Florent Heidet Bo Feng, Mark Williamson, Rui Hu, Taek Kim

Abstract

Interest in advanced nuclear reactors has been rising. One reactor technology is the molten salt reactor (MSR), which is an attractive option, but is hindered by its relatively immaturity compared to others. Although salt chemistry has been studied for some time, few efforts have focused on modeling the reactor core itself, identifying viable system configurations and determining performance characteristics. Our objective has been to develop computational capabilities to model MSRs and to analyze their performance characteristics. These new computational capabilities will be useful for MSR design concepts. We made use of existing reactor physics codes where relevant, expanding their capabilities as needed, and developed new capabilities when no appropriate tool was available. We relied on the system analysis module (SAM) code to develop the new approaches and interface codes that enable modeling, analysis, and design of MSRs. In order to accurately model the fuel cycle of MSRs, new capabilities were developed from scratch, allowing us to model the complex transit of molten salt through the reactor system and to account for elemental removal, fuel make-up, and different cooling/residence times. A salt property database containing a few of the most commonly used salts was also constructed.

LDRD# 2017-055	Securing Energy and Critical Resources
Project Title:	Improving Cost and Energy Efficiency of Nontraditional Water Desalination through Innovative Material and Process Integration
Investigators:	YuPo Lin Di-Jia Liu, Zhi Zhou

Abstract

This project focused on materials R&D using electrochemical technologies to treat nontraditional water, initially that used for cooling in thermoelectric power plants. The salts in such water are in the ionic and fully dissolved forms. Therefore, an electrochemical process that can separate anions and cations through the static fields produced by highly porous and regenerable electrodes represents the most energy-efficient approach. The two electrochemical technologies evaluated in this project have been ion-exchange resin wafer electrodeionization (RW-EDI) and capacitive deionization (CDI). Innovative manufacturing techniques and novel electrode materials showed the path forward for highly energy-efficient water desalination technologies. Our deliverables are (1) to advance material development of a high-surface nanofibrous electrode in a membrane-electrode-assembly specifically designed for CDI; and (2) to demonstrate the electrochemical desalination performance of cooling water using RW-EDI and CDI for a power plant. Desalination of the cooling water, to prevent scaling and corrosion when it is recycled, is the most energy-intensive and expensive operation in these plants. If the energy efficiency of the process can be improved from 12% to 30% and implemented across the power plant industry, the energy savings will be equivalent to thirty 500-MW power plants. We

successfully applied an innovative nanomaterial made using an advanced manufacturing technique on the CDI process to purify saline water. We also evaluated the desalination performance of different types of nontraditional water using RW-EDI for cooling water applications. A level of more than 30% energy efficiency of impaired water was demonstrated using RW-EDI. The process performance was used to identify desirable properties for innovative material development.

LDRD# 2017-088	Securing Energy and Critical Resources
Project Title:	Understanding Resilient Infrastructure Dependencies and Interdependencies through Advanced Optimization and Simulation
Investigators:	Charles Macal Megan Clifford

Abstract

We worked to develop advanced mathematical formulations and computational solution approaches to modeling complex interdependent infrastructures using a "network of networks" approach. A community's ability to withstand all hazards depends on the security and resilience of physical and cyber infrastructure, which is increasingly challenged by population growth, aging and deterioration, and natural and manmade hazards, including severe weather, climate change, and deliberate attacks. Overcoming these challenges requires infrastructure systems to be adapted and designed for resilience, considering the dependencies and interdependencies of the lifeline infrastructure systems (i.e., energy, transportation, communications, water and wastewater). In addition, a key component of these infrastructure systems is the element of human interaction with the infrastructure, which has largely been ignored in infrastructure system modeling. The behaviors of decision makers who plan and operate the infrastructure, as well as consumers who utilize it, can be modeled in the same framework as the interconnected infrastructures themselves. We developed dynamic transient natural gas flow models and a telecommunications infrastructure model, both for subsequent connection to a power grid model. We demonstrated an n-K [total number of systems 'n' – number of failed systems 'K'] security-constrained analysis of the natural gas infrastructure system and documented an infrastructure model validation study. Development of logical data schema for interdependency connections among models of the electric power grid, natural gas, telecommunications, and water infrastructure elements has been pursued. We also began implementation of connected models of interdependent infrastructure systems and demonstrated an n-K security-constrained analysis of an interdependent natural gas infrastructure and the electric power grid. Real-time data assimilation capabilities were incorporated for dynamically recalibrating interdependent infrastructure models.

THE UNIVERSE AS OUR LABORATORY (ULAB)

LDRD# 2017-105	The Universe as Our Laboratory (ULab)
Project Title:	Superconducting Detectors for Future CMB Experiments
Investigators:	Stephen Padin Clarence Chang, Valentine Novosad

Abstract

This project will advance the science and technology of superconducting materials and detectors, with the goal of delivering detectors for the next generation of Cosmic Microwave Background (CMB) experiments. Superconducting detectors are the key technology for observations at millimeter and submillimeter wavelengths, where most of the extragalactic background light is emitted. They are essential for the upcoming CMB Stage-4 (CMB-S4) experiment to measure the energy scale of inflation, the number of neutrino species, the sum of the masses of neutrinos, and the equation of state of dark energy. They also have applications as x-ray detectors as well as in future particle physics experiments. Transition edge sensor (TES) bolometer arrays are currently the most successful superconducting detectors. Our focus is therefore on materials and devices for future TES arrays. The next generation CMB experiment will require ~500,000 detectors. This will require (1) more, narrower bands to remove foreground signals, which will require more sensitive detectors, and (2) fabrication based on excellent control of the superconducting material properties. A rigorous study of superconducting materials and detectors is crucial for addressing both issues. This project will build on experience from the South Pole Telescope detector program to develop novel materials and structures for the next generation experiments. We demonstrated detectors with performance appropriate for the upcoming (CMB-S4) experiment. Our main accomplishments have been: (1) the development of aluminum manganese (AlMn) transition-edge sensors with steep, smooth superconducting transitions in the temperature range 100-500 mK; (2) deployment of a prototype AlMn detector wafer in the South Pole Telescope (SPT)-3G camera; and (3) development of waveguide to planar transmission-line structures for horn-coupled detectors with small polarization errors and low leakage at the horn to detector wafer interface. These followed our successful fabrication and deployment of titanium-gold multilayer (Ti/AuTi/Au) TES detectors at the SPT as well as our determination as to how to improve thermal isolation structures required by TES detector assemblies.

LDRD# 2017-106	The Universe as Our Laboratory (ULab)
Project Title:	Exploring the Universe: Large Scale Structure to the First Stars
Investigators:	Katrin Heitmann Eve Kovacs, John Anderson, Lindsey Bleem, Melina Avila Coronado, Michael Carpenter, Ramesh Balakrishnan, Ravi Madduri, Salman Habib

Abstract

In the last three decades, researchers have established a precision cosmological model that describes the content and evolution of the Universe at the few percent level of accuracy. We have arrived at this point by understanding the physics of the cosmic microwave background (CMB) and by combining CMB observations with measurements from large-scale structure (LSS) surveys that map out the distribution

of galaxies in the Universe. In the next decade, new surveys will aim to push the limits of our current knowledge to the next level to unravel the secrets of the dark Universe—dark energy, dark matter, the origin of primordial fluctuations, and the physics of the neutrino sector. We have delivered a combined approach to address the different challenges we are facing in cosmology, from the handling of very large data sets (including containerization of analysis tools that provides for portability across multiple HPC systems); to sophisticated simulations (including detailed physics descriptions and new results concerning clusters of galaxies); to cross-correlations of different probes (including cross-correlating ground-based optical surveys with the cosmic microwave background); and experiments to help our understanding of the formation of the first stars (including increasing the beam rate capabilities of the Multi-Sampling Ionization Chamber (MUSIC) detector).

DIRECTOR'S COLLABORATIONS

LDRD# 2017-155	Director's Collaborations
Project Title:	Coherent X-ray Studies of Phase Transitions in the Complex Oxides
Investigators:	Dillon Fong

Abstract

We are conducting fundamental studies on the dynamics of phase transitions in complex oxide materials. These oxide materials are known to exhibit a wide variety of phases, ranging from superconducting to ferroelectric and antiferromagnetic to multiferroic. Such behaviors arise from close coupling between crystal structure and electronic structure: slight changes to either due to temperature, stress, chemistry, or an electromagnetic field can cause a substantial change in properties. Complex oxides are therefore candidate materials for "beyond Moore's Law" technologies, having the potential for advanced computation while expending minimal power. To achieve this, spatial and dynamical control of phase behavior in these materials is required. We conducted highly successful in-situ experiments exploring phase transitions and their dynamics in oxide heterostructures, using X-ray photon correlation spectroscopy (XPCS), a method only available at coherent light sources such as the Advanced Photon Source at Argonne. In particular, we focused on understanding the dynamics of electrochemically driven phase transitions in the cobaltites and nickelates, both correlated electron systems with properties that vary from insulating to metallic or antiferromagnetic to ferromagnetic, depending on the Co or Ni oxidation state. We discovered new phenomena associated with the phase behavior and phase transition behavior in the oxide heterostructures.

NAMED FELLOWS

LDRD# 2016-180	Named Fellow
Project Title:	In situ Polarized Spectroscopy of Optically Transparent TRGO-Polymer Solar Cells
Investigators:	Muge Acik Seth Darling

Abstract

The initial objective of this project was to examine surfaces and interfaces of thermally reduced graphene oxide (TRGO)/polymer nanohybrids for significant chemical and thermal stability, high electrical conductivity, and optical visible/infrared transparency, thereby overcoming interfacial imperfections (e.g., degradation, insufficient charge transfer, recombination/resistance). We sought to understand the interfacial chemical reactions that alter device power efficiencies in graphene-based perovskite solar cells by examining the interfaces between thin films of methylammonium (CH_3NH_3) lead halide (MAPbTx, T = I, Br, Cl) perovskite (the light-harvesting layer) and graphene oxide (GO) (the hole-transporting layer). The two main goals of the work were (1) to derive the mechanisms of degradation of reduced GO (RGO) during perovskite growth and (2) to analyze the impact of the halide anions of MAPbTx crystals, which modify the graphene/perovskite interfaces. This research helped to determine key factors in poor solar power conversion efficiencies in some perovskite photovoltaics. We employed in situ polarized spectroscopic characterization -- infrared, Raman, and ultraviolet-visible light (UV-Vis) techniques. Initial experiments involved the fabrication of single-layered GO thin films to study interfacial chemical interactions upon perovskite growth. We found that the type of halide functioning as the anion in these organo-lead halide perovskites determined the chemical modifications at the graphene interface. Whereas the bromine and chlorine halides resulted in acceptable interfaces on GO, the iodide did not. We therefore moved to an alcohol based substitutional growth (SG) approach to the formation and deposition of the $MAPbI_3$ that avoids the annealing process and the use of high-boiling-point solvents. The alcohols not only lowered the reaction temperature by catalyzing the growth but also significantly improved the reaction yield. High quality, low defect interfaces resulted. This work has reached a key milestone in the introduction of a novel, environmentally benign approach. We have applied for a patent for the development of lead halide perovskites using the SG method.

LDRD# 2016-184	Named Fellow
Project Title:	Investigation of Solid-Liquid Interfaces in Energy Materials Interfacing Multi-scale Modeling with Experimental Characterization
Investigators:	Kendra Letchworth-Weaver Maria Chan, Paul Fenter

Abstract

The project has led to capability development in first principles modeling of surfaces and solid-liquid interfaces, especially in comparison with x-ray reflectivity experiments. The capabilities have been applied to mineral-water interface, battery-electrolyte interface, and the growth process of solid oxides. Electrochemical energy storage and conversion devices, such as batteries and solar-to-fuel technology,

must become more efficient and less costly. However, the complexity of the interface between the charged electrode surface and the fluid electrolyte presents a challenge. Studying these devices under operating conditions requires bridging length-scales from the atomic scale to the macroscopic scale of the devices themselves. The highly scalable yet accurate joint density functional theory (JDFT) approach complements existing molecular dynamics techniques to offer a predictive multiscale description of the solid-liquid interface that can be integrated with experimental measurements. This pioneering work broadens basic understanding of fundamental processes at the electrochemical interface, including ion solvation, catalysis, and electrode stability to dissolution, offering crucial insights into a wide range of energy materials. As just one example, our simulations capture experimentally observed trends in solubility and cyclic voltammetry of the phosphonate-coated $LiMn_2O_4$ surface in a Li-ion battery, demonstrating how adjusting the length and functionalization of the phosphonates can maximize Li-ion conductivity but minimize cathode dissolution.

DRD# 2016-185	Named Fellow
Project Title:	Charge Transport in Nanostructured Materials from ab initio Simulations
Investigators:	Marton Andras Voeroes Larry Curtiss

Abstract

Charge transport processes are ubiquitous in materials-relevant energy conversion and storage. Our goals of understanding and engineering charge transfer and charge transport processes in complex nanostructured materials are therefore relevant to the energy utilization landscape. We developed a unified platform, based on first principles methods, to investigate electron and hole transport in nanocomposite materials with compPulses from x-ray free electron lasers (XFELs) provide enough fluence per pulse in a timescale sufficiently short to enable single-shot imaging and freezing the fastest molecular vibrations—and thus monitoring atomic and molecular motions on their natural timescales. This project aimed to define the science problems that make use of these revolutionary properties. We also wanted to identify and investigate potential scientific applications that require high pulse energy, temporally coherent, x-ray pulses in regimes that are not accessible with present XFELs, synchrotrons, or tabletop sources. Two questions to which we have applied computation and planned experimental observation are: (1) the birth and fate of an excess electron in liquid water created by ionization and (2) in pressure-induced phase transitions, what moves first, the atoms or the electrons.lex interfaces, i.e., in heterogeneous media. It focuses on nanostructured systems for solar energy conversion. The approach creates a unified scheme to investigate charge transport both in the band and hopping regimes, with the intermediate regime treated in an approximate, although non-empirical, hierarchical manner. In the case of nanoparticle arrays, we develop ab initio codes to optimize and tune material properties to achieve high carrier mobilities, while maintaining the Coulomb interaction-enhanced carrier multiplication effect. Based on constrained density functional theory (CDFT) as well as a semi-empirical kinetic Monte Carlo method, we have described hopping charge transport in nanoparticle arrays and published the results.

LDRD# 2017-029	Named Fellow
Project Title:	Novel Oxide Materials for Energy-Efficient Neuromorphic Computing
Investigators:	Jianqiang Lin Supratik Guha

Abstract

As conventional von Neumann computers based on digital silicon CMOS technology reach their performance limits, a paradigm shift in computational systems is becoming necessary to sustain the growth of information technology. Neuro-inspired computation is an attractive alternative to conventional computation, because of its massively parallel approach, reconfigurability, and energy efficiency. For this to be realized, novel material systems must be developed. Materials with an electrically driven insulator-to-metal transition (E-IMT) are promising candidates for neuromorphic circuit components such as integrate-and-fire artificial neurons. Recent advances in fabrication technology and probabilistic computation algorithms open up new options to realize a novel neuromorphic computer. We explored new materials such as those with the E-IMT property; resistive switching materials; new devices, including semiconducting and native devices; and new system architectures, including non-Boolean architectures and probabilistic algorithms. Specifically, we studied the E-IMT material vanadium oxide (VO_2) grown on a sapphire substrate. We demonstrated a nanoscale E-IMT structure of dimensions 40 nm by 200 nm. We realized an E-IMT with a record low operating voltage of 0.2 V and an on/off current ratio above 3,000, which is ten times better than the existing technology.

LDRD# 2017-095	Named Fellow
Project Title:	Combining Electrochemistry and Ultrafast Spectroscopies: Real Time Characterization of Multi-Electron/Proton Intermediates in Hydrogen and Oxygen Evolving Catalysts
Investigators:	Ryan Hadt Lin Chen

Abstract

Sunlight is a promising resource; the photon energy delivered to the earth's surface in one day is enough to power our planet for three years at the current level of energy consumption. However, sunlight is diffuse and intermittent. To fully harness solar energy, we need to view sunlight as a chemical reagent and store its energy in the form of high-energy chemical bonds (e.g., H_2, CH_3OH, etc.), i.e., solar fuels. Development for solar fuels generation revolves around the fundamental understanding of photo--driven reactions. We are combining electrochemistry and photochemistry to synchronize specific reaction steps and to trigger formation of intermediates. Ultrafast optical and x-ray spectroscopies were used to characterize the structures of intermediates in real time. In particular, we have provided a detailed spectroscopic characterization of high-valent centers embedded into a lattice and have demonstrated that the presence of the centers tracks the enhancement of oxygen evolution reaction (OER) activity. We have also developed a mechanism for the acid dissolution of template-stabilized, earth-abundant oxygen evolution catalysts (OECs) and have provided a framework for the development

of acid stable OECs. These studies will allow for the further characterization of highly reactive, transient intermediates that carry out the conversion of solar energy to fuels.

LDRD# 2017-096	Named Fellow
Project Title:	On the Colloidal Suspension of Lithium Clusters in Molten Lithium Chloride
Investigators:	Augustus Merwin Chris Benmore, Mark Williamson

Abstract

Much research is devoted to developing next-generation pyroprocessing technologies that will enable the transition from a once-through to a sustainable nuclear fuel cycle. A key technology for the transition is an electrolytic reduction process that converts oxide nuclear fuel to its metallic form. The formation and dissolution of lithium during this process is a noted source of inefficiency, yet little has been known regarding the mechanism of lithium solvation in the molten lithium chloride electrolyte. Our goal has been to elucidate the nature of lithium bonding in molten solutions of lithium chloride and elemental lithium to advance the development of next-generation reprocessing technologies. We have employed high-energy x-ray diffraction at the Advanced Photon Source to characterize the nature of lithium bonding in the molten lithium chloride-lithium system as metallic lithium is electrochemically introduced into the melt. The objective was to confirm the presence and elucidate the structure of lithium nanoclusters. Such nanoclusters may explain previously unattributed physical and chemical phenomena exhibited by these solutions. The time-resolved scattering data suggest the formation of metastable lithium nanoclusters that are predominantly Li8, along with smaller clusters and dissolved Li ions with electrons. Confirmation of the existence of lithium nanoclusters in molten lithium chloride is an advancement in the field of lithium chemistry. The electrochemical behavior of the interface between Li metal and molten LiCl was characterized for the first time. This behavior has been utilized to develop an electrochemical method for quantitatively determining the endpoint of the oxide reduction process. This advancement to process monitoring has significant engineering implications and the potential to increase the efficiency of advanced fuel cycles. It could be used to mitigate current process inefficiencies in the pyrochemical treatment of actinide oxides, and also may have impacts on battery electrolyte chemistry, nanomolecular science, and molten salt physical chemistry. Analogous molten salts of sodium (Na), cesium (Cs), calcium (Ca), and magnesium (Mg) were studied to see if electron solvation observed in molten LiCl-Li solutions is a general phenomenon.

INNOVATE

LDRD# 2016-001	Innovate
Project Title:	Structure and Dynamics of Chiral Molecules and Radicals
Investigators:	Stephen Pratt Ananya Sen

Abstract

Many pharmaceutical molecules are chiral. Stereo-specific synthesis of specific enantiomers is often difficult or expensive, so many of these molecules are synthesized, sold, and administered as racemic (i.e., 50:50) mixtures. The DNA nucleosides and many of the amino acids that form proteins are chiral molecules with nonsuperimposable mirror images, or handedness. The chemical and physical properties of the two forms, or enantiomers, are usually the same, but they do interact differently with polarized light. Most amino acids in living systems are of a specific (L) form, and most sugars in DNA nucleosides are of the specific (D) form. The different enantiomers of many chiral molecules can react very differently with biological systems. We investigated circular dichroism in molecular photoionization induced by circular polarized light. We also studied the photoionization and photodissociation of chiral molecules by using circular polarized vacuum ultraviolet light. Improved signal to noise ratios will be required to fully explore this approach. Techniques to detect and distinguish specific enantiomeric forms, and to determine the fate of potentially harmful metabolites, can benefit both fundamental science and industry. Reduced cost and waste avoidance, defeat of possible harmful side effects of the non-effective isomer, and understanding possible effects in the body of metabolites of the two isomers, are just a few reasons to develop this capability.

LDRD# 2016-010	Innovate
Project Title:	A Theory of Out-of-Equilibrium Phase Transitions
Investigators:	Valerii Vinokour Alexey Galda

Abstract

We are developing theoretical and numerical tools for a quantitative description of nonequilibrium physics in a broad class of systems, focusing in particular on dynamic phase transitions and spintronics. We lack a good understanding of physical processes that occur in thermodynamically open dissipative systems under the influence of external fields. A rigorous theoretical formalism can be developed and successfully applied to describe a wide range of nonequilibrium quantum systems by means of a non-Hermitian extension of the standard Hamiltonian approach. Previous work has indicated that this can be achieved by introducing a complex “Hamiltonian” – the mathematical non-Hermitian operator corresponding to the total energy of the system. All dissipation mechanisms and nonequilibrium forces in the system can then be incorporated in its imaginary part. The Hamiltonian extension is endowed with parity-time (PT) symmetry, which is broken when the external field exceeds a critical value. The loss of the PT-symmetry and subsequent transition from stationary to nonstationary dynamics creates a bifurcation point amenable to quantitative description. We apply this technique to the dynamic Mott metal-insulator transition in small superconducting islands and to dissipative dynamics of quantum

electron spins in micromagnetic devices. Our goal was fully accomplished: we constructed the non-Hermitian approach to quantitative non-perturbative description in open dissipative driven system. The developed theory provides a quantitative description of far-from-equilibrium quantum systems enabling the design of novel devices for quantum information science, including quantum computing and communications.

LDRD# 2016-054	Innovate
Project Title:	Perovskite Halide-based Intermediate-Band Solar Cells
Investigators:	Alex Martinson Maria Chan

Abstract

Perovskite halides are the most recent addition to the list of outstanding optoelectronic materials and are prime candidates for application in intermediate-band (IB) photovoltaic (PV) technology. Their optical extinction, mobility, and excited state lifetimes are comparable to single crystal gallium arsenide (GaAs). These photophysical properties are responsible for the conversion efficiencies of 20% that have been achieved since development of this PV material began. Intermediate-band solar cells are designed to retain the high-output voltages of large bandgap semiconductors, while harvesting significantly more of the solar spectrum. By using the IB as a stepping-stone, electron-hole pairs may be generated from photons with insufficient energy to pump electrons directly from the valence band (VB) to the conduction band (CB). It seems that an absorber base with the necessary photophysical properties and substitutional control is required to achieve excited state lifetimes needed for successful charge extraction. Recent advances in perovskite halide materials computation screening methods offered an opportunity for advances in this emerging field. We developed an experimental and computational foundation for tailoring discrete energy levels in substituted and mixed metal halide perovskites. The substitution of several transition metals for lead was achieved and, in some cases, new absorption features were observed. in particular, Co-substituted methylammonium lead bromide ($MAPbBr_3$) was studied using ultra-fast transient absorption (TA) and time-correlated single photon counting (TCSPC) of luminescence. The charge dynamics of variable mixtures of the relatively narrow band gap (1.57 eV) $MAPbI_3$ and wide band gap (3.02 eV) $MAPbCl_3$ perovskites were probed to map charge and energy flow direction and kinetics. A chemical formulation and perhaps new physical form are still sought in order to achieve the charge extraction goal, and the methods shown to be valuable in this project are now well suited to that task.

LDRD# 2016-063	Innovate
Project Title:	Efficient Droplet-Based Environmental Mechanical Energy Harvesting Through Reverse Electrowetting
Investigators:	Alexey Snezhko Gasper Kokot

Abstract

Energy harvesting from mechanical movements offers the possibility to extract energy from vibrating car engines, buildings, bridges, or land next to a highway. It can potentially provide a valuable portable energy source for mobile devices and applications such as cell phones, laptops compuiters, pacemakers, and other devices. There is a need for innovations that would combine high-efficiency energy harvesting with the possibility of widespread implementation. We pursued development and optimization of a microfluidic-based environmental mechanical energy harvester with enhanced efficiency compared to currently available realizations. The method is based on electric energy generation by changing the overlap between the droplet and electrode interface. It is caused by a mechanical actuation coming from a vibration, shear, or pressure. Microfluidic droplets exploiting the reverse electrowetting process promise a high-energy harvesting efficiency. To maximize the efficiency of the portable droplet-based energy harvesters, an optimal droplet surface-area-to-volume ratio for electrodes, electrode patterning schemes, and energy harvesting layer stacking needed development. Sizes of the droplets were scaled down to microscale. We achieved stable droplet generation with microfluidic channel widths down to 30 µm. The prototype cells were used to generate electric current under constant droplet flow conditions over the patterned electrodes of the harvester.

LDRD# 2016-069	Innovate
Project Title:	GO-IN-EM - Genetic algorithm Optimization of INterface structure from Electron Microscopy
Investigators:	Maria Chan Fatih Sen, Jianguo Wen

Abstract

Optoelectronic, mechanical, electrochemical, and piezoelectric properties, to name a few, are significantly influenced by the presence and characteristics of solid-solid interfaces, yet characterizing these interfaces via both experiments and computational modeling remains challenging. In particular, the interfacial region is often poorly imaged by electron microscopy, even if atomic resolution is achieved away from the interface. Our goal is to combine experimental input in the form of transmission electron microscopy (TEM) and scanning transmission electron microscopy (STEM) images, atomistic modeling, the use of empirical potentials and density functional theory (DFT), S/TEM image simulation, and multi-objective genetic algorithm optimization to determine the full three-dimensional structure of materials at solid-solid interfaces. Grain boundaries (GBs) are far more influential than even surfaces. We first built initial grain boundary/interface atomistic models, then computed S/TEM images from atomistic models. We used a multi-objective genetic algorithm optimization loop to link the two. At the initial stage of the project, we chose CdTe grain boundaries as a prototype for algorithm and code testing. The image matching, TEM/STEM simulation, and optimization codes have been used in concert

to solve for the 3D structure of a CdTe grain boundary. With a straightforward addition to the modular code developed in this project, the combined energy-experiment optimization approach has also been applied to other experimental characterization techniques, most notably, X-ray pair distribution functions.

LDRD# 2016-092	Innovate
Project Title:	Spin Vortex-based Non-volatile Superconducting Memory
Investigators:	Valentine Novosad

Abstract

We developed a hybrid memory architecture that marries the best features of magnetic memory with high fidelity superconducting electronics. This memory concept is energy efficient, non-volatile, with fast operation time and compatible with superconducting microwave circuitry. Our motivation: the opportunity to reduce energy consumption by exploring possibilities of computing architectures that go beyond complementary metal-oxide semiconductor (CMOS). The replacement technology is superconducting (SC) computing, which offers the prospect of moving information without loss over zero-resistance channels. In our design, the hybrid memory architecture marries the best feature of magnetic memory (i.e., low power resonant excitation of spin waves) with that of high-fidelity superconducting electronics (i.e., high Q-factor superconducting quantum interference device [SQUID]-based readout system). This promises to revolutionize exascale computing. Our concept has significant advantages over existing superconducting memory systems: reliance on resonant spin excitations requires much lower power to alter the state of a memory element and the dynamic switching is in the range of microwave frequencies, so pulses with these frequencies are readily available in the logic boards. And, the memory is non-volatile up to the Curie temperature of the magnetic structure while the multiplexed SQUID readout scheme makes the integration density scalable with the Q-factor of the cells.

LDRD# 2016-094	Innovate
Project Title:	Ordered Core-shell Nanostructure for Transverse Thermoelectric Applications
Investigators:	Xing Chen, Kaizhong Gao Ana Aragon Sanchez, Dileep Singh, Moinuddin Ahmed, Ziyao Zhou

Abstract

For conventional thermoelectric (TE) materials, where heat flow and electric current flow in the same direction, there is always a trade-off between a material's thermal conductivity (which needs to be low) and electrical conductivity (which needs to be high). The transverse thermoelectric effect (TTE) has attracted attention because of its unique mechanism in which the electrical and thermal flows are perpendicular to each other. We are making nanostructure transverse thermoelectric (TTE) materials. TTE has several key advantages compared to conventional longitudinal thermoelectric (LTE) effects that could significantly improve device performance in certain applications. We prepared metallic nanowires using an electrospinning system followed by a heat-treatment. We produced aligned 25-30 nm FeCo

alloy metallic nanowires with high thermal/electrical conductivity for the planned core-shell structure. Using atomic layer deposition (ALD), seven nanometers of zinc oxide (ZnO) thermoelectric (TE) with low thermal and electrical conductivity were coated onto the high-aspect-ratio metal nanowires with high uniformity. As the core metal fibers are ferromagnetic, they were aligned through an external magnetic field. These TE oxides are expected to have high ZT value, high stability at high temperature, be capable of operating over a wider temperature range, be made up of non-supply-critical and non-toxic elements, be lightweight, and have small thermal expansion. Once consolidated and properly characterized, devices such as compact, high-efficiency TTE coolers can be integrated into integrated circuits, aircraft, and satellites where the volume and mass are critical for electronics cooling. This work continues under external sponsor support.

LDRD# 2016-098	Innovate
Project Title:	Images from Inner Space: Exposing Quantum Mechanics within Nucleons and Nuclei
Investigators:	Ian Cloet John Arrington

Abstract

Over 50% of allocated running time at the upgraded Jefferson Lab is dedicated to exposing the quark and gluon tomography of nucleons and nuclei. To develop these 3D images will require significant theoretical guidance. We have developed a theory framework to calculate the quark and gluon tomography of mesons using non-perturbative methods in quantum chromodynamics (QCD). This technique first determines the light front wave functions of mesons which can then be directly used to determine a partonic tomography. Our project is at the interface of large-scale computing, theory, and experiment. We employ continuum quantum field theory to compute Wigner distributions and transform these results into predictions for properties of nucleons and nuclei. We use these methods to bridge the gap between real-world observables and contemporary formulations of lattice-quantum chromodynamics (QCD) to produce a real impact in interpreting experimental projects. We test the physical assumptions used in the analysis of experimental data and define the experimental phase space where resources can be focused on the best discovery potential. The combined experimental and theoretical understanding can then reliably be used to define the goals and needs of future international hadron physics facilities. We will see vastly improved kernels for the hadron bound-state problem, with the expression of numerous effects driven by dynamical chiral symmetry breaking. Generalized parton distributions (GPDs) and transverse momentum-dependent parton distributions (TMDs) will be inferred from calculation of the actual observables measured in our experiments. This direct computation will guide experiments toward optimal kinematic conditions for the measured initial and final states -- namely, to the domains that are most sensitive to the GPDs and TMDs of interest.

LDRD# 2017-002	Innovate
Project Title:	Developing Superconducting MgB_2 films on Copper Radio-Frequency Accelerating Structures
Investigators:	Alireza Nassiri Angel Yanguas-Gil, Robert Kustom

Abstract

Magnesium diboride (MgB_2) is one of the promising "intermediate-Tc" binary intermetallic superconducting materials that could provide low surface resistivity at relatively high temperatures. MgB_2 is reported to be a two-gap material, in which the higher-energy gap (6–7 meV) is dominant in the superconductivity, with a stronger coupling to phonons, while the lower-energy gap (1–4 meV) exhibits a much weaker coupling. MgB_2 coatings potentially offer a way to operate superconducting radio-frequency (SRF) cavities at higher temperatures compared to other conventional superconductors, such as niobium tin (Nb_{3Sn}), with a fairly negligible loss in surface resistance. That would decrease the operating costs of particle accelerators by enabling the use of low-temperature helium gas instead of liquid helium for cooling. Assuming an energy gap of about 6 meV (critical temperature Tc of about 39.4 K), a niobium (Nb) SRF cavity coated with an MgB_2 film would have the same surface resistivity at about 8 K that an uncoated Nb cavity would have at 2 K. This research is directed at developing and demonstrating deposition of MgB_2 thin films on bare copper (Cu) or Nb SRF cavities by employing plasma-enhanced atomic layer deposition (PE-ALD), as well as hybrid-physical chemical vapor deposition (HPCVD) with improved coating processes for better surface quality and lower RF surface resistivity. Our work will extend our deposition methods to complex 3D structures. We successfully demonstrated that a uniform MgB_2 coating with good superconducting properties can be achieved on the inner walls of commercially available copper tubes utilizing HPCVD. Tests have also been performed using the PE-ALD method.

LDRD# 2017-004	Innovate
Project Title:	The Missing Link in XTIP: Synergy of Experiments and Theory for Argonne's Global Leadership
Investigators:	Volker Rose Anh Tuan Ngo, Larry Curtiss, Nozomi Shirato, Saw Wai Hla

Abstract

The synchrotron x-ray scanning tunneling microscopy (SX-STM) technique developed at Argonne combines the atomic-scale spatial resolution of STM with the superior chemical and magnetic contrast offered by synchrotron x-rays. In SX-STM, monochromatic x-rays excite core-level electrons while a specialized tip tunnels over a sample surface of interest. Some excited electrons are promoted to unoccupied levels above the Fermi energy, but below the work function. Whereas in a conventional x-ray measurement these electrons would just de-excite, in SX-STM they can tunnel into the STM tip. This allows the direct study of excited molecular orbitals, which are at the core of any chemical reaction. Initial measurements conducted at room temperature have already led to record-breaking resolution for chemical contrast, and we recently built the world's first cryogenic SX-STM. The construction of a dedicated beamline, XTIP, for further development and application of SX-STM at the Advanced Photon

Source (APS) is now supported. We are working to generate a synergy between experimental and theoretical SX-STM research that leads to a new capability to use x-ray standing waves (XSWs) for local 3D molecular imaging. This cannot be achieved by any other technique available today. We are integrating a rotation stage into the existing SX-STM setup, preparing samples for molecular 2D imaging, generating XSWs via x-ray/crystal interactions and using them for molecular 3D imaging. We are also using polarized x-rays to achieve magnetic contrast. Our concurrent theoretical work includes ab initio calculations of molecular geometry on surfaces, comparisons of theoretical predictions with experimental data, development of a framework for 3D imaging using the ab initio calculations, and development of theoretical tools for magnetic mapping. Initial tests included examining cobalt (Co) atoms adsorbed to a 111-oriented crystalline gold (Au) surface. The theoretical calculations agree very well with the experimental data.

LDRD# 2017-007	Innovate
Project Title:	A Novel Method of Longitudinal Bunch Shaping by Double Emittance Exchange
Investigators:	John Power Yine Sun

Abstract

Methods to control the transverse phase space of electron bunches have long been available to accelerator designers; however, methods for controlling the longitudinal phase space are scarce but essential for a variety of innovative accelerator applications. Direct control over the longitudinal position coordinate is extremely difficult because of the femtosecond-to-picosecond range of a typical electron bunch. Double emittance exchange is a concept that allows precision manipulation of a relativistic electron beam's longitudinal structure with the ease of transverse beam manipulation. By facilitating longitudinal bunch shaping, a double emittance exchange (EEX) beamline will have applications in bunch compression, bunch train frequency conversion for generating x-ray free electron lasers or coherent terahertz radiation, ultrahigh-frequency microbunch generation, and other technological advances that will extend the reach of accelerator-based science. We completed the final design of the double-EEX beamline, fabricated necessary equipment, and are installing the beamline at the Argonne Wakefield Accelerator bunker.

LDRD# 2017-012	Innovate
Project Title:	Integrating High Throughput Computation and Wet-chemistry Synthesis for Functional Supercrystals
Investigators:	Badri Narayanan Elena Shevchenko, Subramanian Sankaranarayanan, Wei Jiang, Xiao-Min Lin, Yuzi Liu

Abstract

The self-organization of nanoparticle building blocks into periodic arrays holds tremendous promise in energy, electronics, and devices. Ligand-decorated nanoparticles in various two-dimensional and three dimensional superlattices exhibit a variety of exotic properties that make them suitable for widespread

applications. An in-depth knowledge of the factors governing self-assembly of nanoparticles could provide design principles for engineering novel ordered nanoparticle architectures with prescribed mechanical, electronic, and physical properties. We are unraveling mechanisms underlying the self-assembly process, elucidating the role played by ligands in the self-organization of nanoparticles, and identifying the impact of ligand dynamics on the properties of assembled supercrystals. We have gained significant (experimental and theoretical) molecular insights into thermal and mechanical properties of Au-based nanoparticle superlattices, both at two and three-dimensions. We have had success in understanding the mechanical properties such as fatigue response, crack propagation and fracture in these hierarchical structures. By integrating structural search evolutionary algorithms and popular molecular dynamics (MD) simulation packages, we can now sample thousands of possible configurations to obtain a set of configurations that are most thermodynamically favorable in a certain environment such as temperature and pressure.

LDRD# 2017-013	Innovate
Project Title:	Atomic Layer Deposition of Silicon Carbide for Nuclear Applications
Investigators:	Zhi-Gang Mei Aaron Oaks, Abdellatif Yacout, Angel Yanguas-Gil, Laura Jamison, Michael Pellin, Sumit Bhattacharya

Abstract

The goal of this project has been to develop low-temperature atomic layer deposition (ALD) of silicon carbide (SiC) with film morphology and properties appropriate for nuclear applications such as the coating of Zircaloy fuel cladding in a light water reactor. Typical SiC ALD film deposition requires temperatures that a Zircaloy substate cannot tolerate without leaving local defects and pores that can impair a coating's adhesion strength. To adequately reduce the deposition temperature, new precursors are needed, which in turn require a better understanding of the film growth process. First, we constructed a large chemical reaction database of silicon- and carbon-containing molecules with chemical properties calculated using density function theory (DFT). With the chemical reaction database, we applied machine learning to develop a chemical reaction prediction model using a novel fingerprint for molecules. Using high throughput density functional theory (DFT) calculations and kinetic Monte Carlo (KMC) simulation, we were able to screen thousands of potential chemical precursors for SiC formation at lower temperatures, and the identified precursors were validated through designed ALD experiments.

LDRD# 2017-016	Innovate
Project Title:	BOLT: OpenMP over Lightweight Threads
Investigators:	Pavan Balaji, Sangmin Seo Abdelhalim Amer, Pavan Balaji

Abstract

OpenMP is a widely used directive-based parallel programming model for shared-memory programming for next-machine architectures, and future exascale machines. The core idea that we are proposing is centered on a new, very lightweight and fine-grained OpenMP implementation called BOLT. BOLT will have the unique ability to be specialized for massive and fine-grained parallelism, such as that expected on many supercomputers coming online in the next decade, while providing efficient support for both legacy and future computational science domains. BOLT will enable a lower-overhead and more-scalable OpenMP runtime, which will in turn enable exposure of more parallelism in scientific computing applications, allowing faster science across a variety of domains. We designed and implemented a BOLT prototype and performed initial correctness and performance testing. BOLT shows promising preliminary results in terms of robustness and performance. Our evaluation of a popular and fast multiple-method implementation, which generates nested parallelism by offloading linear algebra operations to a math kernel library, performs roughly three times better when using BOLT instead of the original version of the OpenMP runtime. From a robustness perspective, BOLT can be considered production quality. BOLT has been deployed on large-scale production systems such as Theta at Argonne National Laboratory.

LDRD# 2017-017	Innovate
Project Title:	A Continuously Refinable Mesh, Limited Area Atmospheric Model
Investigators:	Jiali Wang Emil Constantinescu, Iulian Grindeanu, Rao Kotamarthi, Robert Jacob, Vijay Mahadevan

Abstract

Current atmospheric models that attempt to capture mesoscale-to-microscale interactions commonly use a nesting approach in which higher-resolution domains are placed inside a lower-resolution domain. However, each nest introduces a new numerical boundary that loses information. At smaller scales, all of the relevant turbulence data can be lost. Thus, new code is needed to bridge the scales from kilometers to meters without nesting. Several existing mesh geometries and dynamical cores allow continuously refined meshes—for example, the Model for Prediction Across Scales (MPAS). However, MPAS is a global-scale model, and using it for small-scale modeling has a prohibitive computational cost. If one is only interested in resolving flow and atmospheric dynamics over a limited area, then remote regions are not computationally relevant. The key is the development of an MPAS-based, exascale-capable limited area model (LAM). We developed a new capability to model the atmosphere using a continuously refined mesh based on a dynamical core that operates over a limited area. We managed definition of boundary conditions that are consistent with the LAM, generation of conforming meshes over the prescribed boundary, and computing the solution to the atmospheric flow along the boundary. We adapted an existing mesh refinement algorithm to create grid refinements for the

MPAS-Atmosphere over the continental United States (CONUS). We worked with the Energy Exascale Earth System Model (E3SM), focusing on variable-mesh methodologies to assist E3SM in reducing model bias over the refined area by smoothing the boundary conditions of the refined mesh. And, we set up and built three E3SM cases to investigate the value added by high-spatial resolution and data assimilation over the refined region.

LDRD# 2017-022	Innovate
Project Title:	Engineered Interfaces for Gallium Oxide Power Semiconductor Devices
Investigators:	Angel Yanguas-Gil

Abstract

Recent studies have shown that gallium oxide (Ga_2O_3) could replace silicon carbide (SiC) and gallium nitride (GaN) as the material of choice for advanced power devices. Gallium oxide is a wide-bandgap semiconductor with a crystal growth process offering much lower defect density. Single-crystal gallium oxide has been synthesized by several melt-growth methods. Before it can become an alternative to GaN and SiC, high-quality interfaces between it and either oxides or metal contacts and that remain stable at high operating temperatures must be formed. We pursued a rational approach to the design of high-quality metal/dielectric/gallium-oxide and metal/gallium-oxide interfaces. By a combination of in situ and electrical characterizations of test structures, we studied the electronic properties of the different interfaces. We then used this knowledge to develop high-quality interfaces fabricated with in situ plasma processes to control the interface chemistry, density of states, and stability. We developed a fundamental understanding of the correlation between microstructure and the electronic properties of the dielectric/gallium oxide interface, including density of interfacial defects and the band alignment between gallium oxide semiconductor samples and aluminum oxide, hafnium oxide, and nanolaminate materials. Through this project we also explored the impact of doping on the properties of halide vapor phase epitaxy (HVPE) gallium oxide samples and their interfaces. We employed photoluminescence and spectroscopic ellipsometry to study surface states under varying conditions. And we have carried out extended X-ray absorption fine structure (EXAFS) and Fourier transform infrared spectroscopy (FTIR) studies of gallium oxide laminates with aluminum and hafnium oxides.

LDRD# 2017-023	Innovate
Project Title:	New Techniques to Manipulate Rare Isotopes using Adaptive Optics
Investigators:	Michael Bishof Matthew Dietrich, Peter Mueller

Abstract

We are developing new techniques for quickly and efficiently transporting ultracold atoms of rare radioactive isotopes in vacuum for use in investigations such as precision probes of nuclear structure and tests of fundamental symmetries. We are employing adaptive optics to extend the use of optical dipole traps (ODTs) and to achieve reliable transfer from initial atom collection in a magneto-optical trap (MOT) to the final measurement region. Currently, transporting captured atoms requires physical

translation of the focusing optic which is slow and causes atom heating and loss. Focus-tunable lenses (i.e., adaptive optics) translate the focus position of the ODT, eliminating mechanical movement of optical elements and dramatically reducing the time required to prepare atoms for precision measurements. Transporting atoms with greater efficiency should improve the sensitivity of a broad spectrum of experiments that rely on rare isotopes such as 225Ra, 6He, 38mK, and various isotopes of francium. In particular, atom transport efficiency will be improved for the radium EDM (electric dipole moment) apparatus in the Physics Division at Argonne National Laboratory.

SWIFT

LDRD# 2017-108	Swift
Project Title:	Validating Replicability of Waggle Urban Deployments
Investigators:	Charles Catlett Peter Beckman

Abstract

The Array of Things project in progress at Argonne is working to build a smart city with urban sensors in Chicago. We conducted a controlled trial to deploy an Array of Things network in Detroit, focusing on replicating the technical, logistical, policy, and science support processes that had been developed earlier in Chicago. The Detroit trial is a partnership with the City of Detroit Health Department. The project uses Waggle, Argonne's novel wireless sensor platform designed to enable a new breed of sensor-driven environmental science and smart city research. Logistical aspects include coordination with installation personnel from Detroit, for instance, related to power, mounting, and scheduling. Policy encompasses public engagement, governance, privacy, and data stewardship. Science process includes, for example, selection of installation locations and suitability of measurements to address science (and policy) goals. The Waggle architecture is scalable, designed to enable turnkey installation whereby all nodes, irrespective of location, are centrally managed and all data are centrally managed. The Detroit project provides valuable information on the scalability of the support and data management systems and replicability such that the policies and agreements developed with Chicago can enable other labs and universities to rapidly reach similar agreements with their campuses and cities. We learned that successful deployment requires a contact within the city who has the authority to support the research and can champion the project from the top-down. We also determined that a local research partner (a national laboratory or university) is critical as the main partner rather than the city directly, providing that the partner has the wherewithal to engage their local city effectively. The Waggle system could become the platform of choice for field measurements in cities and non-urban settings worldwide.

LDRD# 2017-110	Swift
Project Title:	Argonne Extended Range Friction Tribometer
Investigators:	Robert Erck George Fenske, Nicholaos Demas

Abstract

It is critical to test engine lubricants at temperatures above 70–100°C at high speeds to simulate real engine conditions. High speeds and temperatures are necessary to ensure that the surface chemistries are identical to those experienced in actual use. This project developed an Extended Range Friction Tribometer that operates at high speeds (up to 4 meters per second), high temperatures (up to 150°C), and at the low-to-high contact loads required to cover the entire range of lubrication regimes. In addition to all lubrication regimes, the system enables simulation of friction environments at speeds up to 10 times faster at prototypic temperatures in which surface chemical reactions are active. For rheological studies of existing and experimental fluids, it is necessary to determine the fluid properties in the elastohydrodynamic regime, i.e., rolling-sliding contacts where the fluid confined between them is at high pressure, causing them to be separated. We studied a variety of fluids (more than thirteen) to understand which would be most suitable in industrial machinery. This work found that two fluids in particular showed distinctly low stiffness and limiting shear stress, a polyalkylene glycol monobutyl ether and a fatty acid methyl ester poly-di-n-butyl phosphonate based on palm oil. In contrast, two fluids showed large stiffness and limiting shear stress, the polypropylene glycol monobutyl ether and an adipate diester. The tribometer developed in this project will be used to simulate high-speed tribological environments commonly found in many applications and to evaluate and optimize new tribological technologies (e.g., diamond-like carbon coatings, other coatings, materials, and additives).

LDRD# 2017-113	Swift
Project Title:	Ultra-High Efficiency Fuel Cell-Heat Engine (FC-HE) Hybrids
Investigators:	Sreenath Gupta Brian Ingram, Theodore Krause

Abstract

The objective of our work is to develop hybrid systems that combine a fuel cell with either an internal combustion engine or a microturbine. A multipronged approach was followed that investigated newer architectures, advanced manufacturing methods, and advanced materials for improved durability. A preliminary literature survey showed that candidate fuel cells are polymer electrolyte membrane, alkaline, phosphoric acid, molten carbonate, and solid oxide. Similar candidates for heat engines are internal combustion engines with low-temperature combustion strategies, microturbines, and Stirling engines. Various configurations of FC-HE hybrids were modeled for performance and cost projections. The most promising configurations were refined to size individual components.

LDRD# 2017-131	Swift
Project Title:	Argonne's UAS Capability: Application Framework Development and Benchmarking
Investigators:	Yuki Hamada

Abstract

A number of scientific and technological challenges need to be addressed for making unmanned aircraft systems (UAS) a suitable platform for environmental observations. The goals of our project are to (1) develop operational workflows that provide templates for various UAS application types and (2) obtain a set of fundamental specifications and baseline information associated with UAS observation applications. Initially, we collected and processed multi-temporal multispectral images using a true-color or color-infrared camera over land, first over a variety of natural environments, then over man-made environments. This tested three-dimensional information retrieval using UAS while also providing baseline information for evaluating light detection and ranging technology. For the topographic survey, we collected stereoscopic images of objects of known dimensions to test the accuracy and precision of UAS imaging using a photogrammetric technique. A third experiment used an infrared camera system to observe variables that are invisible to the naked eye. For vegetation-change analysis, test plots simulating an arid landscape were established, and time series images were collected using a UAS-mounted camera. We evaluated and determined trade-offs among the specifications (such as spatial resolution, areal coverage, and data collection time) and constraints (such as cost, timeline, sensor and platform availability). We also calculated operational cost and time for each task and developed scenarios for cost and time estimates for integrated tasks.

LDRD# 2017-137	Swift
Project Title:	Accurate, Real-time Categorization of Unmanned Aircraft Systems (UAS) in an Urban Environment Through Application of Deep Learning Strategies Leveraging Distributed Computing Technology
Investigators:	Adam Szymanski

Abstract

Technological advances of unmanned aerial system (UAS) platforms pose new threats to urban landscapes, requiring mitigation of growing risks through advanced, real-time detection capabilities. Our project develops a distributed image-based detection system utilizing upward facing video cameras on large-scale urban sensor networks by using advanced machine learning (ML) techniques. The basis of a good image recognition algorithm is quality training data. To obtain the needed data, we deploy data collection nodes where we can collect video and imagery of flying objects in urban and semi-urban environments. The data are collected, analyzed, and properly tagged for use as training data for ML recognition algorithms. We have purchased and built custom data collection nodes based on the Waggle platform; collected a significant amount of skyward-facing video data representing birds, airplanes, drones, and other objects; and processed and assembled a tagged data set of ~3,000 training images. We refined and continued development of a deep learning (DL) classification algorithm and used several DL libraries as well as several network structures to compare model results and to create the most effective classifier. As detection information is correlated from multiple distributed sensors with time

synchronization, known locations can be used to determine takeoff location, flight paths, and intended trajectory. Our research also provides findings from an investigation into node density and placement for appropriate coverage, camera requirements, and hardware computing requirements.

LDRD# 2017-141	Swift
Project Title:	Applying Community Network Analysis to Generalize Microbial Assembly Rules
Investigators:	Pamela Weisenhorn

Abstract

This project examines whether stressful environmental conditions result in more tightly knit, co-dependent microbial communities when compared to nearby but less stressful environments. We generate a rich dataset of wetland microbiome data and associated meta-data (such as salinity, flooding depth, flooding duration, sample location, plant community composition), which are mined and serve as preliminary results for future research. The project's network-based approach to microbial ecology allows generalization and scaling of biological processes from organisms to complex communities, aids in the development of fundamental understanding and design rules governing microbial communities, and has the potential to use network parameters to improve modeling capabilities and advance predictive understanding and manipulation of microbiomes. Our research focused on three hypotheses. In the first, methanogenic taxa will have greater relative abundance at the more flooded (anoxic) end of all transects, regardless of salinity. Sites with greater salinity will have lower relative abundance of methanogenic taxa overall. In the second, increased variety of thermodynamically feasible redox-pairs under more oxic conditions will result in increasing diffusivity of the community network. This effect on network diffusivity will be most pronounced under low-salinity conditions. In the third, limited energy environments under more anoxic conditions will have greater across-site consistency in the parameters of community composition and hydrophobic organic compounds. This effect on consistency will be most pronounced under high-salinity conditions.

LDRD# 2017-161	Swift
Project Title:	A Mechanically-based Antenna for RF Incompatible Environments
Investigators:	Kaizhong Gao Changyao Chen, Yuepeng Zhang

Abstract

Our project followed two related paths. We focused on delivering and demonstrating technology that is able to produce electromagnetic fields by mechanically moving trapped charges at low or ultra-low frequencies. This is particularly important for applications in typical radio frequency-denied environments, including underwater and underground conditions. We sought a device-level design solution to provide a large-amplitude mechanical oscillation at the targeted frequency with a very low power consumption. The research includes the need to build a charge carrier with extremely high surface area, which involves developing a charge-trapping layer that can hold a large electric charge to achieve a large field magnitude. This can be done using magnetic nanowire composite materials

developed recently as the base material for the electrode. For an antenna, we also needed to demonstrate a mechanical oscillator with a near resonance condition, which involved building an oscillator and matching its resonant frequency to the target value. The high electric charge trapping layer is also applicable to our second goal, i.e., supercapacitor design. We prepared iron-cobalt (FeCo) nanofibers with diameters of 25–30 nm using electrospinning and a two-step, post-spinning heat treatment. The nanofibers at this dimension have extremely high surface area to volume ratio. As magnetic fibers are attracted to each other, they provide good contact and can serve as an ideal electrode. We coated the nanofibers with a platinum-hafnium oxide-platinum ($Pt/HfO_2/Pt$) trilayer using atomic layer deposition (ALD) to fulfill their function as capacitor media. We characterized the coated fibers using transmission electron microscopy (TEM) and energy dispersive x-ray (EDX) spectroscopy.

LDRD# 2017-163	Swift
Project Title:	Biofilms and Human-made Surfaces: How Microorganisms Attach to and Influence the World Around Us
Investigators:	Dionysios Antonopoulos Kenneth Kemner, Marie-Francoise Gros

Abstract

Biofilms are a prevalent form of microbial life on the planet. What environmental components specifically trigger biofilm formation and subsequently what combination of biotic-abiotic interactions ultimately maintain and keep biofilms in check is not understood. The effect of these processes on the degradation of surfaces to which biofilms are attached is also poorly understood. We pursued creation of an experimental platform and a set of probes to visually track members of a microbial community during biofilm formation. To better simulate naturally occurring multi-member biofilm-producing microbial communities, we used a simple and defined microbial community derived from the environment that (1) naturally produces a biofilm in a dynamic flowing setting (e.g., chemostat) and (2) has members of that community genetically engineered so that they can be visually tracked by real-time imaging. We screened previously enriched communities of low complexity derived from the environment (temperate and tropical forest soils) for their ability to form biofilms in the laboratory. Many of these “minimized communities” form biofilms. We used confocal microscopy to evaluate the main parameters (densities, thickness, surface area) of biofilm architecture on abiotic surfaces by tracking bacteria (Pseudomonas species) that have been engineered to emit light (fluorescence). And, we began work using a flow cell coupled to fluorescence microscopy to observe component members of the enriched environmental community. Lastly, we designed experiments to investigate the colonization of concrete by bacteria and subsequent modifications to the concrete resulting from the surface colonization.

LDRD# 2017-164	Swift
Project Title:	Simulating The Dynamics of Gene Drive Propagation Through Populations
Investigators:	Charles Macal Andrew Castiglioni, Jessica Brooke Trail, Margaret MacDonell

Abstract

Targeted genome editing techniques (such as CRISPR-Cas9 [Clustered Regularly Interspaced Short Palindromic Repeats, protein-9 nuclease]) allow deoxyribonucleic acid (DNA) to be modified with unprecedented precision and speed, adding desired traits and eliminating undesirable traits of an organism. In genetics, gene drive is a technique that promotes the inheritance of a particular gene to increase its prevalence in a population. Applications of gene drive include preventing the spread of insects that carry pathogens (in particular, mosquitoes of the Aedes variety that transmit malaria, Dengue Fever, Zika Virus Disease, and Chikungunya Virus Disease), controlling invasive species, or eliminating herbicide or pesticide resistance. The potential impact of releasing gene drives in the wild raises major concerns regarding their development and management. We used our advanced agent-based modeling (ABM) technology to develop the capability to model gene drives propagating through arbitrary populations of humans, animals, or plants. We modeled both the individuals in the population and their interactions, as well as the genome of the individuals and how the genomes change and evolve. Each agent's characteristics include the representation of its genome. Population processes that affect the genome are modeled explicitly, including effects of targeted genome editing application, mating and crossover, and spontaneous mutation, among other variables. The initial work focused on mosquito population dynamics. To that end, we developed population models of mosquito maturation through their lifecycle stages, including egg, larval, pupal and adult stages. And, we created environment and habitat models to reflect location-specific environmental factors that drive mosquito population dynamics.

LDRD# 2017-165	Swift
Project Title:	Global Survey of CRISPR-CAS Systems in Archeal and Bacterial Species
Investigators:	James Davis Maulik Shukla

Abstract

Bacteria and archaea have evolved molecular mechanisms that confer immunity to invading genetic elements. One important mechanism for this is CRISPR (Clustered Regularly Interspaced Short Palindromic Repeats)-Cas (CRISPR associated system), which enables the incorporation of short foreign DNA fragments into the host genome, essentially enabling the host to memorize the genetic composition of viruses that it has encountered and providing immunity. The goal of this project is to understand the global scope of naturally occurring CRISPR-Cas systems; provide the necessary background for identifying new, naturally and non-naturally occurring systems; and provide insights into the history of virus infection across the bacterial and archaeal kingdoms. Variants of the CRISPR-Cas system will be uncovered through extensive bioinformatic analysis of all available bacterial sequence data. We identified CRISPR-Cas systems by searching for CRISPR elements and Cas proteins. Since CRISPR elements must be differentiated from common genomic repeats, we evaluated bespoke

algorithms for finding CRISPRs on a set of well characterized genomes. The evaluation of state of the art CRISPR finding software led us to incorporate updates in our RAST (Rapid Annotation using Subsystem Technology) CRISPR finding software. We also searched the literature for characterized Cas proteins to seed searches for homologs in PATRIC (PAThosystem Resource Integration Center), a collection of over 120,000 genomes. We found 2,035 protein Cas-related protein families, representing a total collection of 7,725,067 nonredundant Cas-related proteins, and their annotations were updated for projection to all newly annotated genomes.

LDRD# 2018-057	Swift
Project Title:	High Dimensional Single Cell Transcriptomic and Proteomic Analysis and Comparison at High Throughput
Investigators:	Oni Basu

Abstract

Accepted convention assumes a correspondence between the amount of ribonucleic acid (RNA) in a cell and its protein content. However, for some biological instances, this hypothesis may not necessarily hold true. We investigated if and when gene expression correlates with protein content at the single-cell level. We compared messenger RNA (mRNA) profiles with protein expression levels from thousands of single cells by combining two recently developed high-throughput single-cell techniques—Drop-Seq for single-cell RNA expression analysis and CyTOF (cytometry by time-of-flight) for single-cell protein content measurements. Analyzing tens of thousands to hundreds of thousands of single cells will provide the superior statistical resolution needed to compare the gene and protein expression patterns in complex immune cells as T cells.

LDRD# 2018-063	Swift
Project Title:	Development of Non-PGM Filter Regeneration Catalysts for Diesel-powered Equipment
Investigators:	Hee Je Seong

Abstract

As emissions regulations require ever-greater reductions in hazardous soot and gaseous emissions from power generators that use carbon-containing fuels, technologies that trap and oxidize soot in porous filters have proven to be reliable soot emissions mitigators. However, typical exhaust temperatures (in particular, those in diesel engines) are too low to enable filter regeneration through soot oxidation, and post fuel injection or electric heaters are typically required to elevate these temperatures. Catalyst coatings on filters enable significantly lower light-off temperatures so that passive or active regeneration is achieved in a relatively low temperature range. However, this strategy depends on costly platinum-group metals (PGMs). Our project seeks improve on an existing process for producing multilayered, several-micrometers-long nanosheets of Co_3O_4, chained with ~20-nm spherical nanoparticles. These would be a cost-effective non-PGM catalytic material that significantly lowers light-off temperatures for filter regeneration. It is already known this material can lower soot oxidation

temperatures, shows good catalytic activity for oxidizing CO and total hydrocarbons (THCs), and removes gaseous emissions. The goal of this project is to evaluate these catalytic materials under realistic simulated exhaust and accelerated aging conditions to further advance the technology and achieve efficient soot oxidation with no or reduced NO2 in a temperature range of 300–400°C under normal engine operating conditions. The multi-chained Co_3O_4-based materials were tested for soot oxidation in a flow reactor. They showed super active performance, even at 350°C under simulated engine exhaust conditions, implying that they can be utilized for passive regeneration in diesel engines without additional post fuel injection to elevate exhaust temperatures. SO_2 poisoning of the catalyst remains a problem to be addressed, possibly with thin coating, multi-metal alloys and support materials.

LDRD# 2018-064	Swift
Project Title:	Real Time Nucleation and Particle Growth Microscale X-ray Analysis
Investigators:	Janine Lichtenberger Cari Launiere, Jeffrey Fortner, Jody Cananday

Abstract

Understanding the chemical engineering design factors that impact particle size and morphology during plutonium oxalate precipitation from nitric acid media is a crucial requirement for better understanding the forensic signature of PuO_2. We investigated the kinetics of precipitation in situ by combining high-speed x-ray microanalysis techniques at Argonne's Advanced Photon Source (APS) with the controlled conditions enabled by microfluidic chemical reactions. Microfluidics provides a means for high-throughput analysis under precisely controlled conditions, while keeping reagent usage to a minimum. The precision and intensity of the beamline at the APS makes it possible to isolate and study nucleation phenomena. The detailed experimental data resulting from this project will validate the computational predictions of plutonium precipitation and growth phenomena, including both rates and morphologies. The combined modeling and experimental work form a basis for determining forensic signatures for interdicted plutonium oxide of unknown origin. Based on our successful application of the combined x-ray microanalysis-microfluidics technique, a variety of reactive precipitation processes can be investigated. For example, the technique could be expanded to study protein crystallization or nanoparticle synthesis and could have far reaching implications in the nanotechnology field and in the biotechnology, pharmaceutical, and chemical industries.

LDRD# 2018-067	Swift
Project Title:	Cooperative Multitasking for Stochastic Particle Transport Simulations
Investigators:	Paul Romano

Abstract

Simulation of neutral- and charged-particle transport phenomena will pose significant challenges in the march toward exascale computing. Stochastic methods are unique in their ability to accurately simulate complex phenomena without resorting to approximations. However, their use in many applications is severely limited by long integration times. The objective of this project is to assess a new class of parallel

algorithms for stochastic particle transport that rely on cooperative multitasking. Monte Carlo (MC) methods are typically formulated using the so-called history method where each thread is responsible for simulating a single particle at any given time, following it from birth to death. Successive events may be unrelated, but are simulated in sequence. In our plan, each thread has a scheduler that simulates many particles concurrently. With this approach, events that are likely to request data from the same region of memory can be scheduled successively, thereby improving the number of cache hits. In principle, our algorithm has the potential to improve the performance of stochastic particle transport methods by orders of magnitude, since the performance for latency-limited applications is tied directly to the average memory access time. Two frameworks were evaluated for implementing the scheduler: a runtime system such as Argobots, which already provides lightweight threading/tasking constructs, and a dataflow-based execution framework such as PaRSEC, which offers a higher level of abstraction. The goals of the project were met in the sense that we were able to evaluate whether the Argobots framework could be used as the scheduler in our cooperative multitasking algorithm. We found that such a framework would indeed provide the flexibility necessary to implement physics-informed task scheduling for the proposed algorithm. However, because the scheduling decisions would be tied closely to the Monte Carlo transport solver, it was not clear how one would generalize an abstraction layer on top of Argobots that could be utilized in a more generic fashion. For architectures with significantly more data-level parallelism, the cooperative multitasking algorithm may be beneficial in that it exposes a greater degree of concurrency.

LDRD# 2018-068	Swift
Project Title:	Nuclear Data Applications of Atom Trap Trace Analysis (ATTA)
Investigators:	Jake Zappala Michael Bishof, Peter Mueller

Abstract

Our aim was to apply the atom trap trace analysis (ATTA) technique, which measures the relative abundances of rare radioisotopes, to nuclear data research. Detection of product isotopes from nuclear reactions in nuclear data studies can be hindered by low detection efficiency and interference from other isotopes, isobars, and/or molecular species. ATTA is a laser-based detection method with extremely high sensitivity and near-perfect selectivity and no interference from other species. It can provide a tool for accessing nuclear data previously inaccessible by other methods. This project had three goals: (1) determine what nuclear reactions can be studied in a unique way using the ATTA method; (2) produce a calibration for measuring absolute radioisotope abundances of ^{81}Kr, which serves as a proof-of-principle for future application to other ultra-rare isotopes; and (3) demonstrate the extraction, purification, and processing of rare isotopes from sample materials for measurement via ATTA. We were able to produce the necessary calibration of our ATTA instrument for ^{81}Kr, as well as create and validate a system for degassing and trapping radioisotopes directly from natural ores. We have conducted measurements that, combined with the absolute calibration made possible through this project, will produce neutron cross-sections for the ^{80}Kr(n,γ)^{81}Kr and ^{84}Kr(n,γ)^{85}Kr reactions that are of interest for *s*-process studies in nuclear astrophysics.

LDRD# 2018-069	Swift
Project Title:	Performance-based Design, Testing, and Modeling of Biomaterials
Investigators:	William Ebert Vineeth Kumar Gattu

Abstract

We are conducting fundamental studies on the dynamics of phase transitions in complex oxide materials. These oxide materials are known to exhibit a wide variety of phases, ranging from superconducting to ferroelectric and antiferromagnetic to multiferroic. Such behaviors arise from close coupling between crystal structure and electronic structure: slight changes to either due to temperature, stress, chemistry, or an electromagnetic field can cause a substantial change in properties. Complex oxides are therefore candidate materials for "beyond Moore's Law" technologies, having the potential for advanced computation while expending minimal power. To achieve this, spatial and dynamical control of phase behavior in these materials is required. We conducted highly successful in-situ experiments exploring phase transitions and their dynamics in oxide heterostructures, using X-ray photon correlation spectroscopy (XPCS), a method only available at coherent light sources such as the Advanced Photon Source at Argonne. In particular, we focused on understanding the dynamics of electrochemically driven phase transitions in the cobaltites and nickelates, both correlated electron systems with properties that vary from insulating to metallic or antiferromagnetic to ferromagnetic, depending on the Co or Ni oxidation state. We discovered new phenomena associated with the phase behavior and phase transition behavior in the oxide heterostructures.

LDRD# 2018-071	Swift
Project Title:	Machine Learning for Large-Scale Power System Optimization
Investigators:	Feng Qiu Alinson Santos Xavier

Abstract

Understanding, characterizing, and optimizing application data flows on next generation supercomputer architectures is difficult. The difficulty lies in effectively abstracting the system architecture; mapping the complex organization of the system architecture with the data movement requirements in both space and time, such that the information can be used to explore trade-offs (such as power consumption versus execution time); and implementing transformations and mapping that may result in better performance. Moreover, these supercomputing infrastructures are being shared by diverse, concurrent applications, including those that require data-intensive flows. New approaches are required for taking us to the next level in understanding interactions between system infrastructure and application data flows at extreme scales. Our project will help accelerate the transformation of raw data into insights and discoveries and improve our understanding of the data-centric infrastructures needed at exascale and future facilities. We have defined resource abstraction models, taking into account complex supercomputing architectures expected to be deployed at Argonne. Our initial focus was on network topology. We have designed algorithms for data movement optimizations accounting for system topology. In one case, we were able to improve the data movement for a data-intensive cosmology application kernel by 45% on the Mira Blue Gene/Q supercomputer.

LDRD# 2018-073	Swift
Project Title:	An Additive Manufactured All-flexible Wireless 2D Strain Sensor
Investigators:	Yunsong Xie

Abstract

Cellulose—the most abundant organic compound on earth—occurs naturally as a hierarchical architecture of fiber bundles that contain both highly crystalline and amorphous domains. Removal of the amorphous region allows access to highly crystalline, rod-like cellulose nanocrystals (CNCs), and the hydrolysis of natural cellulose with hydrochloric acid to yield CNCs is well known. Because of the inherent properties of CNCs, their surface structure is easily modified. The crystals are also very strong and exhibit high thermal stability. This strength and customizability allow CNCs to be used in many applications. They have applications for advanced battery and catalyst material, as well as additives to change the properties of other materials. This project sought to develop an advanced industrial process for the manufacturing CNCs from Miscanthus x Giganteus grass, a perennial, non-invasive grass hybrid. Small scale synthesis had been achieved, but the current lab scale processes for isolation and purification are not suitable for large scale production. We had to take the laboratory batch process from the gram to the kilogram scale. An industrial process must produce CNCs that match the properties from the small-scale process. It should produce crystals that are 5 nm wide and 200–300 nm long. The overall crystallinity of the product should be greater than 80%. The process must be optimized for efficiency to minimize waste production and energy use. A new industrial process was developed that significantly reduces the cost of producing cellulose nanocrystals. This was accomplished by the invention of an intensified unit operation that significantly reduces waste and allows for the manufacture of cellulose nanocrystals with 30% less equipment than would be required using prior art.

LDRD# 2018-076	Swift
Project Title:	Magnetic-Enabled Self-Heating Membrane for Efficient Water Desalination
Investigators:	Li Tang Valentine Novosad

Abstract

Our project followed two related paths. We focused on delivering and demonstrating technology that is able to produce electromagnetic fields by mechanically moving trapped charges at low or ultra-low frequencies. This is particularly important for applications in typical radio frequency-denied environments, including underwater and underground conditions. We sought a device-level design solution to provide a large-amplitude mechanical oscillation at the targeted frequency with a very low power consumption. The research includes the need to build a charge carrier with extremely high surface area, which involves developing a charge-trapping layer that can hold a large electric charge to achieve a large field magnitude. This can be done using magnetic nanowire composite materials developed recently as the base material for the electrode. For an antenna, we also needed to demonstrate a mechanical oscillator with a near resonance condition, which involved building an oscillator and matching its resonant frequency to the target value. The high electric charge trapping

layer is also applicable to our second goal, i.e., supercapacitor design. We prepared iron-cobalt (FeCo) nanofibers with diameters of 25–30 nm using electrospinning and a two-step, post-spinning heat treatment. The nanofibers at this dimension have extremely high surface area to volume ratio. As magnetic fibers are attracted to each other, they provide good contact and can serve as an ideal electrode. We coated the nanofibers with a platinum-hafnium oxide-platinum ($Pt/HfO_2/Pt$) trilayer using atomic layer deposition (ALD) to fulfill their function as capacitor media. We characterized the coated fibers using transmission electron microscopy (TEM) and energy dispersive x-ray (EDX) spectroscopy.

LDRD# 2018-081	Swift
Project Title:	High Temperature Reactor for Methane Conversion on Clusters
Investigators:	Stefan Vajda Avik Halder

Abstract

Biofilms are a prevalent form of microbial life on the planet. What environmental components specifically trigger biofilm formation and subsequently what combination of biotic-abiotic interactions ultimately maintain and keep biofilms in check is not understood. The effect of these processes on the degradation of surfaces to which biofilms are attached is also poorly understood. We pursued creation of an experimental platform and a set of probes to visually track members of a microbial community during biofilm formation. To better simulate naturally occurring multi-member biofilm-producing microbial communities, we used a simple and defined microbial community derived from the environment that (1) naturally produces a biofilm in a dynamic flowing setting (e.g., chemostat) and (2) has members of that community genetically engineered so that they can be visually tracked by real-time imaging. We screened previously enriched communities of low complexity derived from the environment (temperate and tropical forest soils) for their ability to form biofilms in the laboratory. Many of these “minimized communities” form biofilms. We used confocal microscopy to evaluate the main parameters (densities, thickness, surface area) of biofilm architecture on abiotic surfaces by tracking bacteria (Pseudomonas species) that have been engineered to emit light (fluorescence). And, we began work using a flow cell coupled to fluorescence microscopy to observe component members of the enriched environmental community. Lastly, we designed experiments to investigate the colonization of concrete by bacteria and subsequent modifications to the concrete resulting from the surface colonization.

LDRD# 2018-108	Swift
Project Title:	Towards Predictive Simulations of Rotating Detonation Engines Using Supercomputing
Investigators:	Sibendu Som Pinaki Pal

Abstract

Targeted genome editing techniques (such as CRISPR-Cas9 [Clustered Regularly Interspaced Short Palindromic Repeats, protein-9 nuclease]) allow deoxyribonucleic acid (DNA) to be modified with unprecedented precision and speed, adding desired traits and eliminating undesirable traits of an organism. In genetics, gene drive is a technique that promotes the inheritance of a particular gene to increase its prevalence in a population. Applications of gene drive include preventing the spread of insects that carry pathogens (in particular, mosquitoes of the Aedes variety that transmit malaria, Dengue Fever, Zika Virus Disease, and Chikungunya Virus Disease), controlling invasive species, or eliminating herbicide or pesticide resistance. The potential impact of releasing gene drives in the wild raises major concerns regarding their development and management. We used our advanced agent-based modeling (ABM) technology to develop the capability to model gene drives propagating through arbitrary populations of humans, animals, or plants. We modeled both the individuals in the population and their interactions, as well as the genome of the individuals and how the genomes change and evolve. Each agent's characteristics include the representation of its genome. Population processes that affect the genome are modeled explicitly, including effects of targeted genome editing application, mating and crossover, and spontaneous mutation, among other variables. The initial work focused on mosquito population dynamics. To that end, we developed population models of mosquito maturation through their lifecycle stages, including egg, larval, pupal and adult stages. And, we created environment and habitat models to reflect location-specific environmental factors that drive mosquito population dynamics.

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